

**Mercury Contamination in Indonesia: A Critical Review and
Case Study of Communities Near Gold Mining Areas.**

インドネシアの水銀汚染：金採掘地域のレビューおよび事例研究



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September 2023**

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ABSTRACT

Mercury (Hg) is a prevalent pollutant in Indonesia, primarily as a result of its extensive use in gold mining, especially artisanal and small-scale gold mining (ASGM). This activity is responsible for 67% (244 tons) of the total mercury emissions in the country. Approximately 32 percent is released into the atmosphere. ASGM amalgamation techniques release large amounts of mercury into water bodies, polluting aquatic ecosystems and bioaccumulating mercury in fish, a staple meal for many Indonesians. Indonesian mercury contamination has far-reaching effects. Mercury exposure can harm miners and neighbors. This study aims to evaluate the current status of mercury pollution in Indonesia, analyze the key sources and pathways of contamination, assess the impacts on human health and the environment, and analyze the spatial distribution and risk to the community near ASGM.

The review included 54 studies and found considerable variations in mercury levels across different regions in the country. The arithmetic means of mercury concentrations in air, soil, water, and sediment were $1.97 \mu\text{g}/\text{m}^3$ ($n=3$), $52.26 \text{ mg}/\text{kg}$ ($n=9$), $76.33 \mu\text{g}/\text{L}$ ($n=18$), and $14.06 \text{ mg}/\text{kg}$ ($n=7$), respectively, far exceeding the Indonesian standard values of $1 \mu\text{g}/\text{m}^3$, $0.3 \text{ mg}/\text{kg}$, $1 \mu\text{g}/\text{L}$, and $0.3 \text{ mg}/\text{kg}$. The study also found that vegetables and aquatic food samples had high mercury concentrations, with arithmetic mean values of $3.52 \text{ mg}/\text{kg}$ dry weight ($n=4$) and $0.61 \text{ mg}/\text{kg}$ wet weight ($n=19$), respectively. Approximately 50% of vegetable samples and 21% of aquatic food samples exceeded the Indonesian standard values of $0.03 \text{ mg}/\text{kg}$ and $0.5 \text{ mg}/\text{kg}$, respectively. Root vegetables and mollusks had the highest mercury concentration.

A case study in Mandailing Natal District showed results of Hg concentrations in the rice and vegetables were $50 \pm 33 \mu\text{g}/\text{kg dw}$ ($n=20$) and $2,100 \pm 2,500 \mu\text{g}/\text{kg dw}$ ($n=12$), respectively, and that in the paddy soil and farm soil were $5,600 \pm 12,000 \mu\text{g}/\text{kg dw}$ ($n=20$) and $19,000 \pm 33,000 \mu\text{g}/\text{kg dw}$ ($n=12$), respectively. Hg concentrations in the food, soil and drinking water samples decreased statistically significantly with increasing distance from the amalgam burning facility to the sampling site, suggesting that the burning facility is a major source of mercury in this area. All drinking water samples were below the WHO safe value of mercury, whereas 96% of the vegetable and 82% of rice samples exceeded the safe value from the Indonesian National Standard or the FAO/WHO. The

non-cancer risk calculated from the hazard quotients for the rice and vegetables exceeded 1 for children and adults. These results suggest potential health risks for residents who rely primarily on locally produced vegetables and rice.

The study highlights the urgency of addressing mercury contamination in Indonesia, especially in ASGM areas, and provides a reference for future research and policy development to reduce mercury exposure. Furthermore, a case study in Mandailing Natal District indicates a possible health risk for people who mostly consume locally grown vegetables and grains. Mercury contamination in vegetables and rice cultivated in the regions of this study will certainly rise over time due to the Hg deposition to the soil by gold mining activities. Further monitoring against Hg contamination is required to reduce the health risks for residents.

Keywords: *mercury, environmental media, foodstuff, rice, vegetable, Indonesia*

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CHAPTER I
INTRODUCTION

1.1 Background

Mercury, recognized as one of the top ten chemicals of public health concern by the World Health Organization (WHO, 2021b), poses a substantial threat to human well-being globally. This concern is exacerbated by the prevalence of mercury pollution originating from artisanal and small-scale gold mining (ASGM), which has emerged as a prominent source of mercury contamination both on a global scale and within Indonesia. ASGM operations, often informal and lacking in environmental safeguards, significantly contribute to global mercury emissions, accounting for up to 37 percent of the total emissions (UNEP, 2019). Notably, Indonesia stands as a noteworthy contributor to this issue, responsible for a staggering 64 percent of the country's total mercury emissions (IPEN, 2018). While ASGM constitutes a primary source, other sectors such as industry, fuel consumption, intentional use, and waste also play roles in mercury pollution (UNEP, 2019). The gravity of mercury contamination necessitates comprehensive research efforts and interventions to mitigate its adverse effects on the environment and public health.

Mercury contamination poses a grave global concern, with up to 2200 tonnes of mercury emissions annually originating from over 70 countries worldwide (UNEP, 2013a). Among these countries, Indonesia stands out as a significant contributor, representing approximately 29 percent of global mercury emissions associated with artisanal and small-scale gold mining (ASGM) (Telmer & Veiga, 2008).

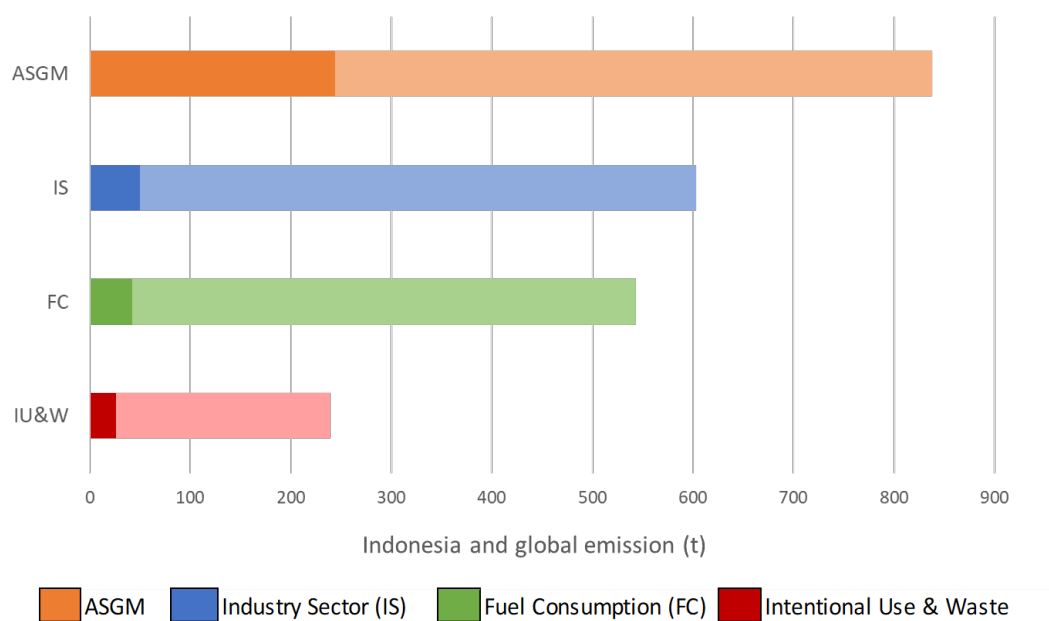


Figure 1.1 Anthropogenic mercury emissions by sector. Indonesia's contribution (darker bars) to global emissions (lighter bars). (IPEN, 2018; UNEP, 2019)

This notable contribution ranks Indonesia as the third-largest emitter globally, following China and Colombia (UNEP, 2013b). This statistic underscores the substantial environmental impact of ASGM activities within Indonesia and emphasizes the urgency of addressing mercury pollution in this region.

Indonesia faces a complex challenge when it comes to artisanal and small-scale gold mining (ASGM), with more than 2,741 illegal ASGM sites spread across 30 provinces. Some of these operations have been established for over a decade, revealing the persistence and scale of this informal mining sector (Kambey et al., 2001). The extensive reach of ASGM in Indonesia is further underscored by the fact that over 1 million individuals are directly employed in ASGM activities (BaliFokus, 2018). Beyond the immediate workforce, the livelihoods of more than 5 million people depend on ASGM in various capacities, highlighting the socioeconomic significance of this sector (BaliFokus, 2018). The prevalence of illegal ASGM operations and the substantial number of individuals reliant on this sector emphasize the pressing need for sustainable and responsible mining practices to mitigate the associated environmental and health risks.

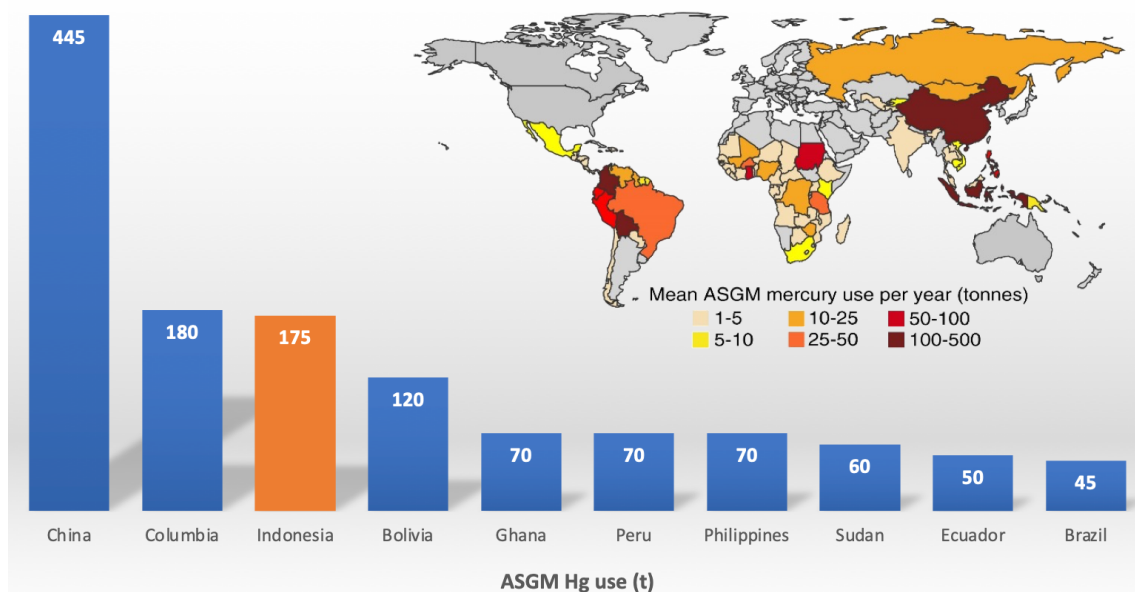


Figure 1.2 Mercury consumption in ASGM (UNEP, 2013)

In response to the multifaceted challenges posed by mercury pollution, Indonesia has undertaken significant policy measures and initiatives. In 2013, the country ratified the Minamata Convention on Mercury, signaling its commitment to addressing and mitigating mercury-related issues on a national scale (Djatzmiko et al., 2019). Furthermore,

Indonesia has initiated a National Action Plan for Reduction and Elimination in 2021, reflecting a proactive stance in combating mercury pollution and safeguarding the environment and public health (Presidential Regulation of Indonesia, 2019). Among the pivotal targets set within this framework, the ASGM sector is slated to achieve Mercury-Free status by 2025, highlighting the government's dedication to curbing the adverse impacts of mercury contamination within this sector and ensuring a sustainable and responsible approach to gold mining (Presidential Regulation of Indonesia, 2019). These policy endeavors exemplify Indonesia's commitment to addressing the pressing issue of mercury pollution and its proactive efforts to safeguard environmental and public well-being.

1.2 Problem of the Study

Drawing from the contextual background provided, several critical issues have come to light. Firstly, there exists a significant gap in the availability of a comprehensive overview of the mercury situation in Indonesia. This gap hampers our understanding of the sources, pathways, and impacts of mercury pollution within the country. Secondly, the persistence of small-scale gold mining activities in the Hutabargot District of Mandailing Natal Regency is noteworthy. These operations continue to flourish, raising concerns about their environmental and health implications. Thirdly, small-scale gold miners in this region employ mercury (Hg) in their operations, specifically in the process of amalgam burning, which has significant implications for both local ecosystems and human health.

Considering these issues, the research problems can be formulated as follows: Mercury Condition in Indonesia: How can we comprehensively assess the state of mercury in Indonesia, encompassing its sources, pathways, and impacts on foodstuffs and human populations? Mercury Assessment in ASGM Areas: What is the current status of mercury contamination in the environment and food sources within a specific case study area, namely the small-scale gold mining (ASGM) region of Hutabargot District, Mandailing Natal Regency, North Sumatra? These research questions serve as the guiding framework for our study, facilitating an in-depth exploration of mercury pollution in Indonesia and a localized assessment of its impact in the specified ASGM area.

1.3 Objectives of the Study

The main purpose of this study was to be understanding the mercury contamination of environment and food, and the implication to human health and environment.

The specific objectives were to:

- a. Summarize and review the mercury contamination in the environmental media (air, soil, water and sediment) and foods consumed or produced in Indonesia and human hair to improve the understanding at regional, national, and global levels.
- b. Assessment of the mercury contamination in the environment and food in residents near the gold mining area, and risk characterization of mercury consumption patterns from environmental and food in the general population

These three specific objectives constituted chapters 2 and 3 of this study, respectively.

1.4 Scope/Limitation of the Study

This study adopts a research framework established by the 1986 National Risk Council, emphasizing the identification of environmental hazards and subsequent risk categorization. Our research scope, as detailed in the specific objectives, encompasses various environmental media, including air, soil, water, and sediment, as well as food sources, specifically plants and fish, with a particular emphasis on their influence on mercury levels in human hair. In the case of review studies, we adopt a focused approach, limiting our analysis to systematic reviews due to the substantial heterogeneity of available data. It is essential to acknowledge that variations in sample size and geographic locations within Indonesia may introduce potential limitations, which could impede broad generalization. Nevertheless, despite these constraints, this research endeavor aims to provide a comprehensive overview of mercury pollution in Indonesia, yielding valuable insights into the extent and implications of this pervasive issue.

Furthermore, in the assessment of health risks within the Mandailing Natal case study, located in Indonesia, our investigation pertains to potential health hazards stemming from combined exposure routes, namely oral ingestion, inhalation, and dermal contact. The case study in Mandailing Natal is deliberately delimited to artisanal small-scale gold mining (ASGM) due to its status as the largest source of mercury emissions in Indonesia. Additionally, rice and vegetables are selected as primary food samples in this study, aligning with their significance as staple foods for many Indonesians, thus

rendering them suitable indicators for assessing mercury-related health risks.

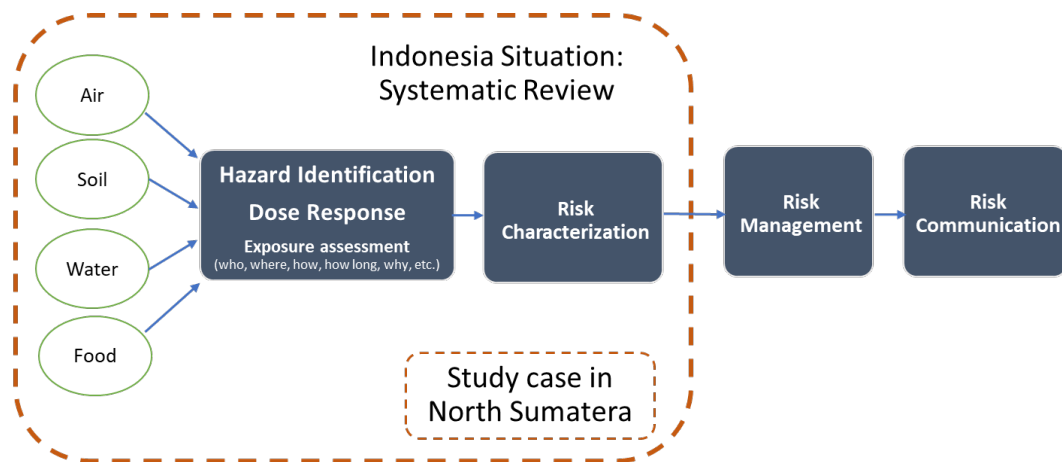


Figure 1.3 Research Framework / Risk Analysis Process
(Council of the European Communities, 1986)

CHAPTER II
Critical Review of Mercury Contamination in Indonesia

2.1 Introduction

Mercury is one of the most toxic heavy metals that is released into the environment through various anthropogenic activities such as artisanal and small-scale gold mining (ASGM), coal combustion, industrial processes, and waste incineration. Only about 10% of mercury emissions are estimated to come from natural sources, while the remaining 90% come from human activities (UNEP, 2013a, 2022). These activities have led to the accumulation of mercury in different environmental compartments, including air, water, soil, and sediment. Consequently, the release and dispersion of mercury have caused adverse effects on both the environment and human populations.

In Indonesia and around the world, ASGM is a prominent source of mercury pollution (IPEN, 2018; UNEP, 2013a, 2019). ASGM involving amalgamation contributes to the release of substantial amounts of mercury into water bodies, resulting in the contamination of aquatic ecosystems and the bioaccumulation of mercury in fish which is a dietary staple for many Indonesians. Additionally, industrial activities, such as coal-fired power plants and waste incineration, further contribute to the emission of mercury into the atmosphere, exacerbating the pollution issue, and approximately 32 percent is released into the atmosphere (UNEP, 2013a).

Based on the 2019 report by the United Nations Environment Programme (UNEP, 2019), worldwide mercury emissions resulting from artisanal and small-scale gold mining (ASGM) amount to 838 tons, accounting for 38 percent of global emissions. Additionally, findings from the 2022 report by The International Pollutants Elimination Network (IPEN, 2018) indicate that ASGM activities in Indonesia alone contribute to about 244 tons of mercury emissions, making up 67 percent of the country's total emissions. Notably, Indonesia represents 29 percent of global ASGM-related mercury emissions, ranking as the third largest contributor globally after China and Colombia (UNEP, 2013a). Indonesia also confronts grave threats from the global illegal trade in mercury and the environmental degradation caused by mercury emissions and releases (Drwiega, 2018).

In Indonesia, mercury contamination has far-reaching effects. Both mine employees and residents of affected areas may experience severe health effects from mercury exposure (Bose-O'Reilly et al., 2016a). Mercury toxicity can cause neurological disorders, developmental delays in children, and other systemic health problems (WHO, 2021a). To address these issues, Indonesia has implemented various policies and initiatives to

mitigate mercury pollution, including ratifying the Minamata Convention on Mercury 2017 (WHO, 2021a), and initiating National Action Plan for Reduction and Elimination in 2021, with the goal of achieving mercury-free status for ASGM sector by 2025 (Presidential Regulation No. 21. National Action Plan for the Reduction and Abolishment of Mercury, 2019). This critical review aims to analyze the current state of mercury pollution in Indonesia, investigate the key sources, and pathways of contamination, and assess the effects on human health and the environment.

2.2 Materials and Methods

2.2.1 Literature search strategy

In this study, the search was conducted in various international databases, namely PubMed, Scopus, Embase, and EBSCO, as well as local databases such as Neliti and OneSearch. The literature search focused on six variables and sources of Hg pollution: air, soil, water, sediment, food, and hair. A systematic review employed the phrases "Mercury," "Hg," "Air," "Soil," "Water," "Sediment," "Food," and "Hair" alone or in combination with "OR" and/or "AND". The literature search was conducted in accordance with the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guideline, as depicted in Figure 2.1 (Moher et al., 2009).

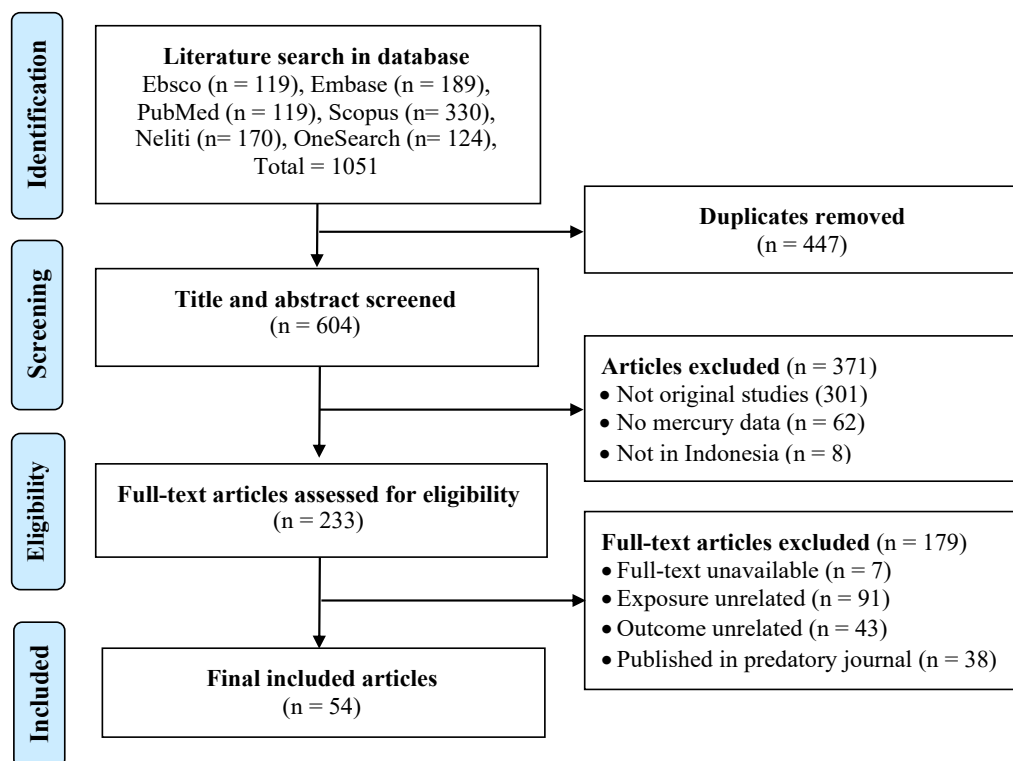


Figure 2.1 Flowchart of the study selection process.

2.2.2 Study selection

The reference citations were exported to EndNote (version X9.3.3), and the deduplication feature was used to identify references listed in the replicate. Two independent reviewers screened the titles and abstracts of the identified studies based on pre-defined inclusion and exclusion criteria. The inclusion criteria were adopted in the original studies reporting the levels of mercury in environmental media (air, soil, water, and sediment), foodstuffs (vegetables and fish), and human hair in Indonesia. The exclusion criteria included research that was not conducted in Indonesia, was not available in full text, was not available in English or Indonesian, and was published in a predatory publication. In situations where reviewers disagreed, a third reviewer was consulted.

2.2.3 Data extraction and quality assessment

Data on mercury concentrations in environmental media were extracted from the selected studies. The following information was extracted: first author, year of data collection, study location/site description, sample type, sample size, analytical method, and average or range of Hg concentrations. The data were organized and analyzed using Microsoft Excel for Microsoft 365.

2.2.4 Data analysis

The data extracted from the included studies were synthesized and presented in a narrative synthesis format. The findings were summarized in tables and graphs to illustrate the variation in mercury levels across different regions in Indonesia. The articles included in the study comprised a highly diverse population. As a result, the authors chose to describe the levels in each of the several populations rather than combining the data in a meta-analysis.

2.3 Results and Discussion

2.3.1 Literature search results and discussion

The systematic literature search yielded 1,051 studies, of which 54 met the review's inclusion criteria. On the islands of Sumatra and Kalimantan, relatively little research has been conducted, while research in this systematic review is mainly found on the islands of Java and Sulawesi. The most studied research variable is environmental media 53% ($n=37$), followed by foodstuff 33% ($n=23$), and human hair 14% ($n=10$).

Table 2.1. Mercury concentration ($\mu\text{g}/\text{m}^3$) in the outdoor air in Indonesia.

Province	Location/Site Description	Samples (n)	Concentration ($\mu\text{g}/\text{m}^3$)	Ref.
Bengkulu	Muara Aman, ASGM area	3	1.30	(Nakazawa et al., 2021)
	Muara Aman, resident area	6	0.01 ± 0.00	
Central Kalimantan	Palangkaraya, river area	4	0.01 ± 0.01	
	Palangkaraya, city area	4	0.04 ± 0.07	
Central Sulawesi	Palu, ASGM area	2	4.08	
	Palu, resident area	18	0.52 ± 0.42	
Central Sulawesi	Palu, ASGM area	11	9.17 ± 16.42	(Nakazawa et al., 2016)
	Palu, resident area	60	0.20 ± 0.18	
South Sulawesi	Pangkep, cement power plant	10	2.43 ± 0.98	(Mallongi et al., 2020)
Air standard	ACGIH – United States (8 hr)		25	(NJDH, 2009)
	SCOEL – European Union (8 hr)		20	(SCOEL, 2007)
	WHO (Annual)		1	(WHO, 2000)
	Japan (Annual)		0.04	(Takiguchi et al., 2018)

Abbreviations: ACGIH, American Conference of Governmental Industrial Hygienists; SCOEL, Scientific Committee on Occupational Exposure Limits; WHO, World Health Organization

2.3.2 Mercury concentrations in environmental media

Table 2.1 presents Hg concentrations ($\mu\text{g}/\text{m}^3$) in outdoor air in Indonesia. Mercury exists in three major forms in the atmosphere: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particle-bound mercury (PBM). GEM has a residence time in the atmosphere ranging from 3 to 12 months, while GOM and PBM have shorter residence times, typically lasting from a few hours to weeks (Horowitz et al., 2017). Nakazawa et al., (2021; 2016) analyzed GEM concentrations, while Mallongi et al., (2020) examined total mercury in dust, which includes both gaseous and particulate mercury. Nakazawa et al., (2016) found the highest Hg concentration of $9.17 \mu\text{g}/\text{m}^3$ in the ASGM area in Central Sulawesi. The concentration decreased to $4.08 \mu\text{g}/\text{m}^3$ in 2021, with nearby residential areas showing $0.20 \mu\text{g}/\text{m}^3$ and $0.52 \mu\text{g}/\text{m}^3$ in 2016 and 2021, respectively Nakazawa et al., (2021; 2016). A study conducted in Bengkulu province also reported higher concentrations of Hg in the air in ASGM areas compared to residential areas (Nakazawa et al., 2021). Mallongi et al., (2020) determined that Hg concentration of $2.43 \mu\text{g}/\text{m}^3$ in South Sulawesi from the cement power plant area. This study found that both the ASGM process and cement power plants had higher mercury levels compared to residential regions. The concentration of mercury in the air of Indonesia is relatively low compared to ACGIH (2009) and SCOEL (2007) for 8 hours but above WHO's (2000) recommended levels for the annual averaging period. This study reports the average mercury concentration in Indonesia's air at $1.97 \mu\text{g}/\text{m}^3$, higher above the WHO (2000)

and Japanese (Takiguchi & Tamura, 2018) guidelines of 1 and 0.04 $\mu\text{g}/\text{m}^3$, respectively.

Table 2.2. Mercury concentration in soil (mg/kg) in Indonesia

Province	Location/Site Description	Samples (<i>n</i>)	Concentration (mg/kg)	Ref.
Aceh	Nagan Raya, ASGM area	3	0.28 ± 0.04	(Nisah et al., 2022)
Banten	Lebaksitu, ASGM area	15	1.24 ± 0.89	(Novirsa et al., 2019)
West Java	Bogor, ASGM area	9	9.00 ± 8.40	(Tomiyasu et al., 2017)
West Java	Depok-Bekasi, e-waste recycling area	5	15.20 ± 28.5	(Soetrisno et al., 2020)
East Java	Batu, Agriculture	4	<LOD	(Agil et al., 2021)
South Sulawesi	Pangkajene, agriculture coal, and cement industry area	13	61.90 ± 51.70	(Astuti et al., 2021)
	Pangkajene, non-agriculture coal, and cement industry area	9	61.80 ± 42.5	
South Sulawesi	Maros, agriculture cement plant area	10	77.30 ± 43.59	(Rauf et al., 2020)
	Maros, non-agriculture cement plant	10	42.20 ± 25.11	
South Sulawesi	Pangkep, cement power plant	10	0.02 ± 0.01	(Mallongi et al., 2020)
Southeast Sulawesi	Bombana, ASGM area	8	390 ± 860	(Basri et al., 2020a)
	Bombana, mining commercial area	12	13.00 ± 17.00	
	Bombana, control area	6	7.40 ± 9.90	
Soil standard	Indonesia		0.30	(IMEF, 2021)
	Canada		0.16	(CME, 2011)
	South Africa		0.93	(DEA, 2010a)
	Australia		1.00	(CEC, 1986)
	European Union		1.50	(EPAA, 2018)

Nine (9) investigations found Hg contamination in Indonesian soil samples from various districts. The primary sources of contamination include atmospheric deposition from ASGM, coal, cement, and additional industrial activities. Table 2.2 provides an overview of the soil Hg analysis findings. The highest Hg concentration in soil was found in the ASGM area of Southeast Sulawesi (390 mg/kg) by Basri et al., (2020a). This was followed by the cement power plant area in South Sulawesi (77.3 mg/kg) (Rauf et al., 2020) and the coal and cement industrial area in South Sulawesi (61.9 mg/kg) (Astuti et al., 2021). Similar results were observed in other research areas, such as the ASGM area in Banten (1.24 mg/kg) (Novirsa, Dinh, et al., 2019), and in West Java (9.00 mg/kg) (Tomiyasu et al., 2017). However, disappointing results were obtained from the ASGM area in Aceh (0.28 mg/kg) (Nisah et al., 2022) and cement power plant in South Sulawesi (0.02 mg/kg) (Mallongi et al., 2020). The lower Hg concentrations in Aceh can be attributed to the early stages of ASGM in the region, with few hotspots present. Several variables, including total organic carbon (TOC) and distance from the contamination source, influence Hg concentrations in soil (Cheng et al., 2020). The extremely high Hg

result of Bombana ASGM area is possibly due to the naturally occurring cinnabar in the soil (Idrus et al., 2017). Cinnabar is the most prevalent natural source of mercury and has been mined for thousands of years. According to UNEP, between 2010 and 2015, Indonesia and Mexico opened new mercury mines, both before ratifying the Minamata Convention (UNEP, 2017). The average mercury concentration in the soil of Indonesia is 52.26 mg/kg, surpassing the Indonesian safe limit (0.3 mg/kg) (IMEF, 2021). In areas affected by gold mining and the cement industry, the average mercury concentration in soil was found to be 174 times higher than the recommended level, highlighting the severity of the issue and the urgent need for comprehensive measures.

Table 2.3. Mercury concentration ($\mu\text{g/L}$) in water in Indonesia

Province	Location/Site Description	Samples (n)	Concentration ($\mu\text{g/L}$)	Ref.
Aceh	Nagan Raya, river ASGM area	3	<0.05	(Nisah et al., 2022)
Aceh	Aceh Jaya, 3 river ASGM area	18	0.09 ± 0.09	(Suhud et al., 2020)
Jakarta	Jakarta bay, seawater in coastal area	82	13.47 ± 16.06	(Siregar et al., 2016)
Jakarta	Jakarta bay, seawater in coastal area	6	0.13 ± 0.05	(Riani et al., 2018)
Central Java	Banyumas, Tajum river ASGM area	7	1003	(Budianta et al., 2019)
West Java	Bogor, Cikaniki river ASGM area	5	3.46 ± 4.45	(Tomiyasu et al., 2013a)
West Java	Bogor, Cikini river resident area	2	0.34 ± 0.13	(Yasuda et al., 2011)
West Java	Bogor, Ciliwung river, resident area	6	<0.10	
West Java	Depok-Bekasi, e-waste recycling sites	5	0.12 ± 0.11	(Soetrisno et al., 2020)
South Sulawesi	Parepare, seawater in coastal area	3	5.33 ± 2.08	(Rusman et al., 2020)
South Sulawesi	Pangkep, cement power plant area	10	0.48 ± 0.70	(Mallongi et al., 2020)
Southeast Sulawesi	Bombana, drinking water in ASGM area	32	0.33 ± 0.07	(Suramas et al., 2019)
Gorontalo	Bone Bolango, Bone river ASGM area	11	0.34 ± 0.02	(Gafur et al., 2018)
Gorontalo	North Gorontalo, Hulawa river ASGM	5	8.26 ± 10.60	(Hiola, 2017)
Gorontalo	Boalemo, Buladu contaminated river	8	122.25 ± 36.7	(Mallongi et al., 2015)
Gorontalo	Boalemo, Buladu uncontaminated river	10	21.97 ± 11.14	
Maluku	Buru, drinking water near ASGM area	3	0.50 ± 0.00	(Pridianata et al., 2019)
Papua	Jayapura, 4 river ASGM area	4	41.25 ± 10.00	(Tanjung et al., 2022)
Papua	Depapre, seawater in coastal area	5	<0.80	(Hamuna et al., 2021)
Papua	Mimika, seawater in coastal area	6	<0.70	(Tanjung et al., 2019)
Water standard	Indonesia		1	(IMH, 2017a)
	WHO		6	(WHO, 2017)

Table 2.4. Mercury concentration in sediment (mg/kg) in Indonesia

Province	Location/Site Description	Samples (n)	Concentration (mg/kg)	Ref.
Aceh	Aceh Jaya, 3 river ASGM area	18	0.001 ± 0.001	(Suhud et al., 2020)
West Java	Bogor, Cikani river ASGM area	8	17.90 ± 29.21	(Tomiyasu et al., 2013a)
Central Java	Banyumas, Tajum river ASGM area	7	9.75	(Budianta et al., 2019)
Gorontalo	Boalemo, Buladu contaminated river	10	5.11 ± 1.43	(Mallongi et al., 2015)
Gorontalo	Boalemo, Buladu river uncontaminated	8	3.32 ± 1.74	
Gorontalo	Bone Bolango, Bone river ASGM area	11	0.08 ± 0.03	(Gafur et al., 2018)

Province	Location/Site Description	Samples (<i>n</i>)	Concentration (mg/kg)	Ref.
Maluku	Buru island, river sediment	2	54.26 ± 39.78	(Reichelt et al., 2017)
	Buru island, coastal sediment	7	22.05 ± 16.46	
Papua	Mimika, sediment in coastal area	6	<0.001	(Tanjung et al., 2019)
Sediment	Indonesia		0.3	(IMEF, 2021)
standard	Canada		0.2	(CME, 2011)

Similarly, mercury contamination in water and sediment is a significant concern in Indonesia. The results of the analysis of Hg concentration in surface water ($\mu\text{g/L}$) and sediment (mg/kg) in Indonesia are reported in Table 2.3 and Table 2.4, respectively. The sources area of water and sediment samples varied across different areas, including ASGM areas, coastal areas, cement power plant areas, e-waste sites, and residential areas. In Central Java, Budianta et al., (2019) recorded the highest Hg concentration, reaching up to 1,003 $\mu\text{g/L}$. Similarly, river water in various ASGM areas such as West Java (3.46 $\mu\text{g/L}$) (Tomiyasu et al., 2013a), Gorontalo (8.26; 122.25 $\mu\text{g/L}$) (Hiola, 2017) (Mallongi et al., 2015), and Papua (41.25 $\mu\text{g/L}$) (Tanjung et al., 2022) and coastal area in Jakarta (13.47 $\mu\text{g/L}$) (Siregar et al., 2016), as well as South Sulawesi (5.33 $\mu\text{g/L}$) (Rusman et al., 2020), have all exceeded the guidelines standard. On the other hand, river water in ASGM areas such as Aceh (<LOD; 0.09 $\mu\text{g/L}$) (Nisah et al., 2022) (Suhud et al., 2020), Southeast Sulawesi (0.33 $\mu\text{g/L}$) (Suramas et al., 2019), and Maluku (0.50 $\mu\text{g/L}$) (Pridianata et al., 2019) exhibited relatively lower concentrations.

ASGM activities are predominantly located near streams and rivers, as water is required for these operations. The higher Hg concentrations in river water from ASGM areas can be attributed to factors such as the direct discharge of untreated wastewater into rivers (Tomiyasu et al., 2013a) (Tanjung et al., 2022), particularly evident in Central Java due to the proximity to ASGM activities and poor sewage systems (Budianta et al., 2019). Studies by Mallongi et al., (2015) and Tanjung et al. (2022) have shown that upstream areas exhibit higher Hg concentrations, likely due to the transportation and accumulation of Hg-containing sediments and contaminants from upstream sources (Hiola, 2017). Siregar et al., (2016) found that high concentrations of mercury in seawater did not originate from industrial wastewater and gold-mining activities but rather from near-shore regions such as municipal incinerator ash used as a substitute for sand in artificial reefs or waste-dumping by shipping vessels. The relatively low Hg concentration in Aceh is attributed to the early stages of ASGM in the region. Similarly, the lower Hg

concentrations in South Sulawesi and Maluku are due to the processes involved in treating drinking water samples, which have the potential to reduce Hg concentrations. It is worth noting that the average mercury concentration in the water exceeded the guideline standard by 76 times, while the national standard is set at 1 µg/L (IMH, 2017a).

In sediment, Reichelt-Brushett et al., (2017) recorded the highest Hg concentration in the ASGM area in Maluku (54.26 mg/kg), followed by ASGM in West Java (17.9 mg/kg) (Tomiyasu et al., 2013a) and Gorontalo (5.1 mg/kg) (Mallongi et al., 2015). The factors influencing sediment Hg concentrations are similar to those observed in river water, including water treatment and upstream areas. It is worth noting that sediments exhibited higher concentrations compared to water samples from the same study area, highlighting the significance of addressing sedimentary mercury pollution. The average mercury concentration in sediments was 14.06 mg/kg, which is 47 times higher than the Indonesian standard of 0.3 mg/kg (IMEF, 2021).

This study revealed significant contamination of air, soil, water, and sediment with mercury (Hg) in various regions of Indonesia. Elevated concentrations of Hg were observed in water and sediment samples, particularly in areas associated with artisanal and small-scale gold mining (ASGM), coastal regions, cement power plants, and industrial activities. The findings emphasize the urgent need for comprehensive measures and effective mitigation strategies to address mercury pollution in these environmental compartments.

Table 2.5. Mercury concentration in vegetable samples (mg/kg dry weight) in Indonesia

Province	Location/Food Description	Samples (n)	Concentration (mg/kg dw)	Ref.
Banten	Lebak, rice grain (<i>Oryza sativa</i>)	7	0.10 ± 0.08	(Novirsa et al., 2019)
West Java	Cisitu, rice grain (<i>Oryza sativa</i>)	11	0.30 ± 0.33	(Bose et al., 2016b)
West Java	Sukabumi, cassava leaf (<i>Manihot esculenta</i>)	2	4.61 ± 1.51	(Saragih et al., 2021a)
	Sukabumi, cassava root (<i>Manihot esculenta</i>)	2	31.07 ± 17.25	
	Sukabumi, papaya leaf (<i>Carica papaya</i>)	2	1.53 ± 0.89	
	Sukabumi, papaya fruit (<i>Carica papaya</i>)	1	0.10	
	Sukabumi, rice grain (<i>Oryza sativa</i>)	1	0.09	
	Central Sulawesi	Palu, maize cropping (<i>Zea mays L</i>)	4	
Palu, rice grain (<i>Oryza sativa</i>)		4	0.20 ± 0.06	
Palu, cassava cropping (<i>Manihot esculenta</i>)		4	0.33 ± 0.31	
Palu, onion cropping (<i>Allium cepa L.</i>)		4	0.20 ± 0.16	
Food other than fish standard	Indonesia		0.03	(IFDA, 2022)
	WHO		0.01	(WHO, 2006a)

2.3.3 Mercury concentrations in foodstuffs

The analysis of Hg concentration in vegetables (mg/kg dw) in Indonesia is reported in Table 2.5. All vegetable samples in this study were collected from the ASGM area and exhibited concentrations that exceeded the Indonesian guidelines (0.03 mg/kg) (IFDA, 2022). Cassava root demonstrated the highest mercury concentration, averaging 31.07 mg/kg (dw), followed by leaf vegetables with an average concentration of 4.61 mg/kg (dw) (Saragih et al., 2020) (Saragih et al., 2021a). This observation aligns with the findings of Addai-Arhin et al. (2022), who reported that root samples tend to have higher concentrations due to adsorption from the contaminated soil in closer proximity compared to leaf or fruit samples (Addai-Arhin et al., 2022). Ramlan et al. (2022) found a correlation between the accumulation of Hg and the amount of mercury in plant tissues (Ramlan et al., 2022), indicating that adsorption varies for each vegetable species (Rea et al., 2001). The average mercury concentration in vegetables was 3.52 mg/kg (dw), surpassing the Indonesian Food and Drug Administration guidelines by 117 times (0.03 mg/kg) (IFDA, 2022). These findings underscore the potential risk of high mercury exposure through the consumption of certain foodstuffs in Indonesia, particularly for individuals consuming large amounts of cassava root and leaf vegetables in mining areas. Effective measures should be implemented to minimize mercury contamination in vegetables and ensure the safety of food consumption in affected regions.

Table 2.6. Mercury concentration in aquatic food (mg/kg wet weight) in Indonesia

Province	Location/Food Description	Samples (n)	Concentration (mg/kg ww)	Ref.
Mollusk				
Jakarta	Jakarta Bay, Green mussel (<i>Perna viridis</i>)	10	7.39 ± 1.33	(Riani et al., 2018)
Central Java	Demak Coast, Blood mussel (<i>Anadara granosa</i>)	100	<LOD	(Yulianto et al., 2020)
East Java	East Java Coast, Clam (<i>Meretrix lyrata</i>)	36	0.05 ± 0.01	(Soegianto et al., 2021)
East Java	East Java Coast, Blood mussel (<i>Anadara granosa</i>)	48	0.27 ± 0.05	(Soegianto et al., 2020)
Gorontalo	Boalemo,			(Mallongi et al., 2015)
	Shelfish (<i>Bellamnya javanica</i>) contaminated track	120	1.24 ± 0.11	
	Shelfish (<i>Bellamnya javanica</i>) uncontaminated track	150	0.13 ± 0.06	
	Shelfish (<i>Mya arenaria</i>) in contaminated track	120	1.66 ± 0.19	
	Shelfish (<i>Mya arenaria</i>) in uncontaminated track	150	0.19 ± 0.08	
Crustacean				
East Java	East Java Coast, Mud crab (<i>Scylla serrata</i>)	45	0.011 ± 0.0002	(Soegianto et al., 2022)
East Java	East Java Coast, blue swimming crab (<i>P. Pelagicus</i>)	45	0.004 ± 0.0002	(Soegianto et al., 2022)
Fish				
Banten	Banten bay, greenback mullets (<i>Planiliza subviridis</i>)	5	<LOD	(Afiyatillah et al., 2022)

Province	Location/Food Description	Samples (n)	Concentration (mg/kg ww)	Ref.
Banten	Binuangen, Torpedo scad (<i>Megalaspis cordyla</i>)	30	0.06 ± 0.01	(Suratno et al., 2021)
	Binuangen, Indian scad (<i>Decapterus russelli</i>)	15	0.24 ± 0.10	
Jakarta	Muara Angke, 10 fish species in estuary	10	0.11 ± 0.12	(Rahayu et al., 2014)
Jakarta-Bali	Fish processing plants,			(Handayani et al., 2019)
	Yellowfin tuna (<i>Thunnus albacares</i>)	167	0.27 ± 0.11	
	Bigeye tuna (<i>Thunnus obesus</i>)	126	0.22 ± 0.08	
	Oilfish (<i>Ruvettus pretiosus</i>)	150	0.41 ± 0.06	
	Swordfish (<i>Xiphias gladius</i>)	150	0.87 ± 0.14	
	Marlin (<i>Macaira indica</i>)	165	0.85 ± 0.08	
West Java	Cisitu, freshwater fishponds near ASGM	6	0.91 ± 1.12	(Bose et al., 2016b)
West Java	Saguling reservoir, residents, and tourism area	12	0.03 ± 0.02	(Marselina et al., 2020)
East Java	East Java Coast, ponyfish (<i>Leiognathus equulus</i>)	75	<LOD	(Chudaifah et al., 2014)
East java	East Java Coast, Halfbeaks (<i>Hyporhamphus affinis</i>)	75	0.05 ± 0.02	(Asmysari et al., 2013)
Central Kalimantan	Galangan, 25 species of fish near ASGM	263	0.20 ± 0.36	(Castilhos et al., 2006a)
North Sulawesi	Tatelu, 11 species of fish near ASGM	131	0.58 ± 0.45	(Castilhos et al., 2006a)
North Sulawesi	Buyat Bay, 91 fish species, metal mining tailings	327	0.17 ± 0.21	(Shepherd et al., 2018)
Gorontalo	Limboto lake, Nila fish (<i>Oreochromis niloticus</i>)	6	0.06 ± 0.03	(Nakoe et al., 2014)
Aquatic food standard	WHO, Indonesia		0.50	(IFDA, 2022)
	WHO, Indonesia (predator fish)		1.00	

The analysis of Hg concentration in aquatic food (mg/kg wet weight) in Indonesia is reported in Table 2.6. Mollusks exhibited the highest mercury concentration, averaging 1.56 mg/kg (ww), followed by fish (0.27 mg/kg ww) and crustaceans (0.01 mg/kg ww). In Jakarta Bay, mollusks showed the highest Hg concentration with values of 7.39 mg/kg (ww) (Riani et al., 2018), while mollusks at the ASGM area in Gorontalo recorded values of 1.66 and 1.24 mg/kg (ww) (Mallongi et al., 2015). The pollution in Jakarta Bay is attributed to hazardous and toxic waste from Jakarta Province, intensified by various activities such as residential and industrial operations, transportation, seaports, hospitals, fishing, commerce, and services (Riani et al., 2018). In Gorontalo, the accumulation of mercury in shellfish showed a linear relationship with Hg concentration in water and a significant correlation with Hg concentrations in sediment (Mallongi et al., 2015). On the other hand, mollusks and crustacean collected in Central Java (Yulianto et al., 2020), and East Java (Soegianto et al., 2020, 2021; Soegianto, Nurfiyanti, et al., 2022; Soegianto, Wahyuni, et al., 2022) likely due to sampling from areas without mining or industrial influence.

On average, fish in this study displayed relatively low Hg concentrations, with an average of 0.27 mg/kg ww. However, high Hg concentrations were found in fish collected from the ASGM area, such as West Java (0.13-1.3 mg/kg ww) (Bose-O'Reilly et al.,

2016b), Central Kalimantan (0.05-1.24 mg/kg ww)(Castilhos et al., 2006a) and North Sulawesi (0.04-0.68 mg/kg ww) (Castilhos et al., 2006a). Hg concentrations were found to be higher downstream and in close proximity to gold mining regions. Despite the relatively small size of the fish, fish from water basins contained the maximum levels of mercury (Castilhos et al., 2006a). In contrast, freshwater fish from residential areas in West Java (0.03 mg/kg ww) (Marselina & Suhada, 2020) and Gorontalo (0.06 mg/kg ww) (Nakoe et al., 2014) exhibited lower concentrations. The highest concentration among sea fish was observed by Handayani et al. (2019) for tuna and tuna-like species from the Southern Indian Ocean (Indonesia), where tuna fish ranged from 0.05-0.65 mg/kg ww (Handayani et al., 2019), while tuna-like ranged from 0.01 to 1.91 mg/kg ww (Handayani et al., 2019) exceeding the predator fish standard of 1 mg/kg (IFDA, 2022). Although the mercury levels in these species are relatively lower compared to swordfish and marlin, the higher weight of the fish contributes to higher mercury levels (Handayani et al., 2019). On the other hand, fish from the sea in Banten showed relatively low concentrations (<LOD; 0.06 mg/kg ww) (Afiyatillah et al., 2022) (Suratno et al., 2021), as did fish from Jakarta (0.24 mg/kg ww) (Rahayu et al., 2014), and East Java (<LOD. 0.05 mg/kg ww) (Chudaifah et al., 2014)(Asmysari et al., 2013).

Table 2.7. Mercury concentration in human hair (mg/kg) in Indonesia

Province	Location/Sample Description	Samples (n)	Concentration (mg/kg)	Ref.
Banten	Lebak, resident near ASGM area	41	3.20 ± 1.97	(Novirsa et al., 2020a)
Banten	Sukabumi, males in ASGM area	48	3.27 ± 2.89	(Harianja et al., 2020a)
	Sukabumi, females in ASGM area	33	5.91 ± 4.69	
	Sukabumi, children in ASGM area	4	5.34 ± 0.00	
	Depok, landfill area	85	0.56 ± 0.43	
West Java	Depok-Bekasi, children in e-waste sites	22	8.00 ± 4.2	(Soetrisno et al., 2020)
Yogyakarta	Depok-Bekasi, children control area	22	2.00 ± 1.10	(Ernawati et al., 2021)
	Kulon Progo, maternal in ASGM area	16	0.79 ± 2.02	
	Kulon Progo, maternal postpartum in ASGM	16	10.59 ± 33.38	
	Kulon Progo, infants in ASGM area	16	129.90 ± 0.98	
	Kulon Progo, maternal II in ASGM area	16	0.19 ± 0.15	
West Nusa Tenggara	Kulon Progo, infants II in ASGM area	16	17.33 ± 0.20	(Ekawanti et al., 2015)
	West Lombok, miner in ASGM area	100	2.60 ± 1.70	
	West Nusa Tenggara	West Sumbawa, direct-exposed miner	90	
South Sulawesi	West Sumbawa, indirect-exposed miner	30	1.30	(Krisnayanti et al., 2016)
	West Sumbawa, non-exposed miner	30	0.56	
	Makassar, direct exposed miner in ASGM	52	10.80 ± 9.50	
	Makassar, indirect exposed miner in ASGM	43	6.50 ± 3.50	
	Makassar, control group	80	2.80 ± 1.90	(Abbas et al., 2017)

Province	Location/Sample Description	Samples (n)	Concentration (mg/kg)	Ref.
Southeast Sulawesi	Bombana, direct exposed miner in ASGM	45	15.71 ± 13.49	(Sakakibara et al., 2017)
	Bombana, indirect exposed miner in ASGM	25	9.25 ± 5.65	
	Bombana, control area	11	5.70 ± 2.28	
Gorontalo	North Gorontalo, resident near ASGM area	95	7.10 ± 2.00	(Arifin et al., 2015)
Human hair standard	US EPA		1	(USEPA, 1997)
	WHO		1	(WHO, 2008)

2.3.4 Mercury concentrations in human hair

The concentrations of Hg in human hair (mg/kg) in Indonesia are presented in Table 2.7. The majority of studies were conducted in ASGM areas, with 90% of the studies showing concentrations above the World Health Organization (WHO) standard of 1 mg/kg (WHO, 2008). The highest Hg concentration of 129 mg/kg was reported in hair samples from infants residing in mining areas in Yogyakarta (Ernawati et al., 2021). Mercury accumulates in both mothers and neonates, with prenatal exposure through the placenta and postnatal exposure through breast milk. Thus, infants may be at risk of mercury exposure through maternal transfer. Miners showed the highest Hg concentrations, with an average concentration of 15.71 mg/kg in Southeast Sulawesi (Sakakibara & Sera, 2017), followed by West Nusa Tenggara (13.00 mg/kg) (Krisnayanti et al., 2016), and South Sulawesi (10.80 mg/kg) (Abbas et al., 2017). Residents living near the ASGM area in Gorontalo (7.10 mg/kg) (Arifin et al., 2015), and Banten (3.20 mg/kg) (Novirsa et al., 2020a), as well as those near e-waste sites in West Java (8.00 mg/kg) (Soetrisno & Delgado-Saborit, 2020), also showed elevated concentrations exceeding the standard. Direct exposure to mercury during mining activities was associated with higher mercury concentrations in hair. The duration of time spent in proximity to gold mining was found to be positively correlated with Hg accumulation in hair, as observed by Sakakibara et al., 2017; Ernawati et al., 2021. Furthermore, a correlation was found between hair mercury levels and the consumption of substances containing mercury, as observed by Arifin et al. (2015). Localization effects were also observed, where groups consumed mercury from the same contaminated source, such as food and fish from the same area. In contrast, hair samples from a landfill area displayed the lowest concentration of 0.56 mg/kg (Hardiyanti et al., 2020). The analysis of human hair samples in Indonesia revealed an average mercury concentration of 11.41 mg/kg, exceeding guidelines by 11 times. Targeted interventions are urgently needed to mitigate mercury exposure, particularly in ASGM areas. Addressing maternal-fetal transfer of

mercury is crucial in combating mercury contamination and protecting the health of vulnerable populations.

Table 2.8. Percentage of the studies exceeding the standard

Samples	Total study	Standard guideline	Ref.	Exceeded standard	% of exceeded the standard
Air	3	1 µg/m ³	(WHO, 2000)	2	67%
Soil	9	0.3 mg/kg	(IMEF, 2021)	6	67%
Water	18	1 µg/L	(IMH, 2017a)	7	39%
Sediment	7	0.3 mg/kg	(IMEF, 2021)	4	57%
Vegetable	4	0.03 mg/kg	(IFDA, 2022)	4	100%
Aquatic food	19	0.5 mg/kg	(IFDA, 2022)	4	21%
Human hair	10	1 mg/kg	(USEPA, 1997)	9	90%

The identification of the most polluted samples was accomplished through the utilization of a frequency table, as presented in Table 2.8. The results of the study indicate that vegetables exhibited the highest degree of contamination, with all studies above the Hg guidelines. This was followed by human hair, where 90% of the samples were found to be contaminated, and air and soil, where 67% of the samples were found to be contaminated. The results of the study indicate that the aquatic food sample obtained from Indonesia exhibited the lowest level of contamination. The researchers discovered a significant correlation between the concentration of Hg in soil and that in vegetables or foodstuffs (Li et al., 2017; Yu et al., 2018), as well as a connection between sediment and aquatic food (Leady & Gottgens, 2001), even though the ability of different species to accumulate Hg varies.

2.3.5 Study limitations

The study acknowledges several limitations that should be considered. A scarcity of available data on mercury concentrations in different regions of Indonesia may have restricted the comprehensiveness of the review. The included studies exhibited heterogeneity in methodologies, sample sizes, and geographic locations, which may have introduced variability and hindered generalizability. The exclusion of non-English and non-Indonesian language publications may have resulted in the omission of valuable sources. Additionally, the focus on online accessibility may have excluded relevant papers not available electronically. Despite these limitations, the study provides a comprehensive overview of mercury pollution in Indonesia, offering valuable insights into the extent and implications of this issue.

2.4 Conclusion

The findings of this study highlight the widespread presence of mercury contamination in multiple environmental media and food sources in Indonesia. The exceeding of national and international guidelines highlights the urgent need for comprehensive and targeted interventions to reduce mercury exposure and protect public health. Particularly, the areas of artisanal and small-scale gold mining (ASGM) were identified as mercury contamination regions, with elevated levels observed in proximity to mining activities. Vegetables and mollusk in the ASGM area are not suitable for consumption. The accumulation of mercury in environmental media and food sources poses significant risks to human health, especially for populations living in or in proximity to ASGM areas.

Future research should focus on long-term monitoring and trend analysis to track mercury levels, evaluation of health impacts on vulnerable populations, exploration of remediation techniques, and strategies, effective risk communication, and regulatory improvements. A collaborative effort is required to reduce the risk of exposure to mercury and to protect the health and well-being of the population and the environment in Indonesia.

CHAPTER III

Spatial Distribution and Human Health Risks of Mercury in the Gold Mining Area of Mandailing Natal District, Indonesia

3.1 Introduction

Mercury is highly toxic to human health and is one of the top 10 chemicals of major public health concerns (WHO, 2021b). Most of the Hg in the environment results from human activity, including coal-fired power plants (UNEP, 2013b). Artisanal and small-scale gold mining (ASGM) is the single largest source (37%) of airborne global anthropogenic Hg emissions (Gibb & O’Leary, 2014; UNEP, 2013c; WHO, 2021b). Hg is used in gold mining to extract gold from ore by forming amalgam in ASGM, and estimated Hg emissions from ASGM doubled between 2005 and 2010 (UNEP, 2013b).

Indonesia has more than 1200 ASGM hotspots (BaliFokus, 2018). The ASGM sector accounts for about 69.7% of the total national Hg released into the environment (about 244 tonnes) in Indonesia (BaliFokus, 2018). Hg concentrations in air, river water, soil and fish around ASGM sites have been investigated in several areas; including Central Kalimantan (Elvince et al., 2008), Gorontalo (Mallongi et al., 2014), North Sulawesi (Limbong et al., 2003), Java (IQBAL Rofiq & INOUE Takanobu, 2011) and West Java (Harianja et al., 2020b; Yustiawati et al., 2006) in Indonesia. Hg pollution has been investigated in detail in West Java and high Hg concentrations in river water, soil and air near ASGM sites have been reported (Kono et al., 2012; Kono & Tomiyasu, 2013; Tomiyasu et al., 2019; Tomiyasu, Kono, Kodamatani, Hidayati, et al., 2013b; Yasuda, Yustiawati, Suhaemi Syawal, et al., 2011; Yustiawati et al., 2006). The health risks to miners and residents, including children, have also been reported (Castilhos et al., 2006; Bose-O’Reilly et al., 2008, 2010, 2016; Gibb and O’Leary, 2014; Nakazawa et al., 2016). Mandailing Natal, North Sumatra, Indonesia is a region with abundant gold-containing geological resources. Since 2005, gold rock exploration has been conducted in the rugged region of Mount Hutabargot using conventional, primitive technology (Agrawal & Susilorini, 2020). However, more than a decade has passed since ASGM began in Mandailing Natal, yet information on Hg contamination is still scarce.

Several studies have suggested that rice has been identified as a possible source of methylmercury (MeHg) exposure from food (Feng et al., 2008; Novirsa, Quang, et al., 2019). Globally, Indonesia ranks as the third largest consumer of rice (OECD/FAO, 2021a), and each year, rice consumption amounts to roughly 37.7 million tonnes, most of which is produced domestically (OECD/FAO, 2021b). Indonesia’s per capita rice consumption per year is higher than that of China (76.5 kg/year) and India (69.9 kg/year),

at about 126 kg/year (OECD/FAO, 2021b). In addition, Addai-Arhin et al. suggested that vegetables, such as cassava, cultivated around ASGM sites are potential source of Hg (Addai-Arhin et al., 2022), but little is known about Hg concentration in vegetables in Indonesia. Thus, investigating Hg concentration in rice and vegetables is necessary to evaluate the health risk to residents around ASGM sites.

In the present study, the Hg contamination in rice, vegetables, soil and water was investigated near two gold mining areas in Mandailing Natal, North Sumatra; the Hg intake of local residents was estimated, and potential health risks associated with Hg exposure were discussed.

3.2 Materials and Methods

3.2.1 Study area and sampling sites

The study area is in Mandailing Natal District, North Sumatra Province, Indonesia (Fig. 3.1). Nauli and Simalagi villages, which are affected by Hg pollution from gold mining and processing activities, were selected. ASGM has been active in this location since 2005 and is becoming one of the most promising sources of income in addition to farming for most local people. The total populations of Nauli and Simalagi villages were 1599 and 460, respectively (BPS, 2021).

Agricultural soil ($n = 32$), rice ($n = 20$), vegetables namely cassava (*Manihot esculenta*) leaves, katuk sweet (*Sauropus androgynus*) leaves, papaya (*Carica papaya*) flower, bilimbi (*Averrhoa bilimbi*) fruit, water spinach (*Ipomoea aquatica*) and susumber (*Solanum torvum*) ($n = 12$), drinking water ($n = 16$) and groundwater ($n = 16$) were collected in late October 2021 in the rainy season from the villages (Fig. 3.1). Soil samples were collected from the same site as the rice and vegetable samples. The communities consume drinking water from groundwater, so we collected samples of drinking water and groundwater from the same source. We also collected reference samples of rice ($n = 3$), vegetables consisting of green onion (*Allium cepa*), cabbage (*Brassica oleracea*) and baby spinach (*Spinacia oleracea*) ($n = 3$), drinking water ($n = 3$) and groundwater ($n = 3$) from non-contaminated areas with no industry and mining activities 10 km from the sampling area to compare with the results from the study area and we assumed that the Hg level is negligible.

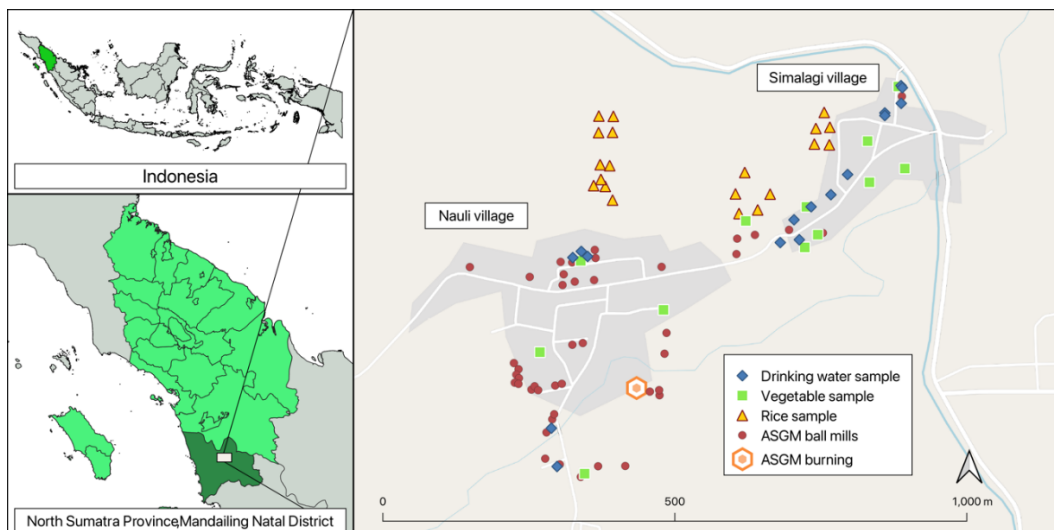


Figure 3.1. Map of sampling sites

3.2.2 Sample preparation and pre-treatment

Rice and vegetable samples (about 200 g) were packed in zipped plastic bags, washed with distilled water and oven dried at 40°C (DX601, Yamato Scientific co., ltd., Tokyo, Japan) until the weight was stable. Soil samples (about 500 g) were taken from the surface to a depth of 10 cm with a plastic scoop and then placed in zipped plastic bags. Roots, rocks, wood and extraneous matter were removed and the samples were oven dried at 40°C for until the weight was stable. The dried rice samples were hulled to separate the husks and grains before the grains were ground with a porcelain mortar and pestle and sifted through a 150 µm sieve. The samples were homogenised. The water samples (around 100 mL) were placed in polyethylene bottles. Clean handling of all samples was performed to prevent cross-contamination. All the samples were stored at 4°C and kept in light-dark conditions until the analysis.

3.3.3 Analytical methods

Total mercury concentration in every sample was determined with a mercury analyser (MA-3000, Nippon Instrument Corporation, Tokyo, Japan). Hg concentration in rice, vegetables and soil was determined by thermal decomposition using method number 7473 published by the U.S. Environmental Protection Agency (USEPA). Hg in samples was analysed without chemical pre-treatment. Three sample boats were used to weigh three aliquots (50 mg) from each sample to obtain triplicate data.

Hg concentration in water was determined by reduction vaporization using method number 245.1 published by the USEPA. The unfiltered water samples (100 mL) were poured into Teflon bottles and 0.5 N sulfuric acid (5 mL) and nitric acid (2.5 mL) were added. Then, 5% potassium permanganate (15 mL) was added, and the mixture was left for 10 min to ensure that it remained purple, before 5% potassium persulfate (8 mL) was added. The samples were heated in a water bath at 95°C for a period of 2 hours with the bottle lids tightened so that the samples would not leak. After cooling the samples to a room temperature, a solution of sodium chloride and hydroxylammonium chloride (6 mL) was applied to eliminate excess permanganate, and then 10% tin (II) chloride (0.3 mL) was added to the samples for immediate analysis. The analysis was conducted using a total of 5 mL of the treated water sample for each measurement. All of the chemical reagents, with the exception of tin (II) chloride, were of analytical grade and procured from FUJIFILM Wako Pure Chemical Corporation. Tin (II) chloride was purchased from Kanto Chemical co. (Tokyo, Japan).

3.3.4 Quality assurance and quality control

Each instrumental analysis was performed on three replicates, and a blank analysis was performed for every three samples. Three certified reference materials (CRM) were used to determine the accuracy of the results for rice, vegetables and soil. The CRMs used were 7302-a from the National Metrology Institute of Japan for trace elements in marine sediment, European Reference Material ERM-CC580 for estuarine sediment and 7402-a for trace elements in cod fish tissue with the value obtained was 0.51 ± 0.02 mg/kg, 132.4 ± 6.06 mg/kg and 0.61 ± 0.01 mg/kg respectively ($n = 3$, recovery rates of 94%–102%, 96%–105% and 99%–101% respectively). The recovery analysis for Hg in water involved the addition of a known amount of Hg to both blank and sample water to evaluate the accuracy of the analysis. The coefficient of variation was 3.3%.

3.3.5 Risk assessment methodology

To conduct this research, several assumptions were made. It was assumed that body weight refers to the guidelines for environmental health risk analysis by the Indonesian Ministry of Health of 55 kg for adults and 15 kg for children. Adults are defined as being older than 19 years, and children as being 6 to 12 years old. We also assumed that local people consume food and drink from the area where they live. Data on the consumption

of vegetables, rice and drinking water were taken from a similar study in Indonesia, namely the risk assessment of the population in Bogor (Rahman et al., 2019), and the parameters are listed in Table 3.1.

Hg can enter the human body by the following pathways: oral ingestion of food (e.g. fish, meat, rice, vegetables and other agricultural products), drinking water and soil particles; skin contact with water and soil particles; and inhalation of soil particles, gaseous elemental Hg and particulate Hg. This study did not measure airborne Hg; thus, it was not included.

Table 3.1. Parameters used to estimate average daily intake (ADI)

Parameters	Unit	Adult	Child	Reference
Hg concentration, C	$\mu\text{g}/\text{kg dw}$; $\mu\text{g}/\text{L}$	–	–	This study
Ingestion rate of rice, IR_{rice}	$\text{kg ww}/\text{day}$	0.4	0.2	(Rahman et al., 2019)
Ingestion rate of vegetables, IR_{veg}	$\text{kg ww}/\text{day}$	0.029	0.014	(Rahman et al., 2019)
Ingestion rate of water, IR_{dw}	L/day	1	0.5	(Rahman et al., 2019)
Ingestion rate of soil, IR_{soil}	mg/day	100	200	(US EPA, 2011)
Rice dw to ww conversion factor, WCF	$\text{kg ww}/\text{kg dw}$	0.91	0.91	(Staven et al., 2003)
Vegetable dw to ww conversion factor, WCF	$\text{kg ww}/\text{kg dw}$	0.23	0.23	(Staven et al., 2003)
Exposure frequency, EF	day/year	350	350	(IMH, 2012; US EPA, 2011)
Exposure duration, ED	Years	30	6	(IMH, 2012; US EPA, 2011)
Body weight, BW	kg	55	15	(IMH, 2012)
Averaging time, AT	days	10,950	2190	(IMH, 2012; US EPA, 2011)
Skin surface, SA	cm^2	5700	2800	(US EPA, 2011)
Skin adherence factor, AF	$\text{mg}/\text{cm}^2/\text{day}$	0.07	0.2	(US EPA, 2011)
Dermal absorption factor, ABS	–	0.1	0.1	(DEA, 2010b)
Conversion factor, CF	kg/mg	10^{-6}	10^{-6}	(US EPA, 2011)
Inhalation rate, IR_{inh}	m^3/day	20	12	(IMH, 2012)
Particulate emission factor, PEF	m^3/kg	1.36×10^9	1.36×10^9	(US EPA, 2011)
Oral reference dose, RfD_{Oral}	$\mu\text{g}/\text{kg}/\text{day}$	0.3	0.3	(DEA, 2010b; US EPA, 2011)
Dermal reference dose, RfD_{Dermal}	$\mu\text{g}/\text{kg}/\text{day}$	0.3	0.3	(DEA, 2010b; US EPA, 2011)
Inhalation reference dose, $RfD_{\text{Inhalation}}$	$\mu\text{g}/\text{kg}/\text{day}$	0.0857	0.0857	(DEA, 2010b; US EPA, 2011)

To estimate the potential Hg exposure of the local population through consumption from several exposure sources, the following calculation was used. Average daily intake (ADI) by oral ingestion, dermal and inhalation exposure was estimated using equations 1–3 provided by the US EPA (US EPA, 1989, 2011; Yeganeh et al., 2013). The estimation

of ADI-Inhalation in this study is solely based on the Hg content in the soil and does not incorporate measurements of atmospheric Hg levels.

$$\text{ADI – Ingestion} = \frac{C \times IR \times EF \times ED}{BW \times AT} \times WCF \text{ — (1)}$$

$$\text{ADI – Dermal} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF \text{ — (2)}$$

$$\text{ADI – Inhalation} = \frac{C \times IR - inh \times EF \times ED}{BW \times AT \times PEF} \text{ — (3)}$$

Here, ADI is the average daily intake exposure dose via the subscript pathway; C is the pollutant concentration in environmental media ($\mu\text{g}/\text{kg}$ or $\mu\text{g}/\text{L}$); IR is the daily intake rate (kg/d); EF is exposure frequency (day/year); ED is exposure duration (years); BW is body weight (kg); AT is the exposure time ($ED \times 365$) (days); SA is body surface area (cm^2); AF is skin adherence factor ($\text{mg}/\text{cm}^2/\text{day}$); ABS is dermal absorption factor (unitless); CF is conversion factor (kg/mg); and PEF is the particle emission factor (m^3/kg). Detailed information on these parameters is listed in Table 3.1.

Assessments for non-cancer risks were estimated by dividing the estimated daily intake by the reference dose (RfD), following USEPA guidelines.

$$\text{HQ} = \frac{\text{ADI}}{\text{RfD}} \text{ — (4)}$$

where HQ is the hazard quotient (HQ). An HQ value of less than 1 indicates that there is no potential risk of adverse health effects from exposure to Hg over a lifetime. However, if the frequency and severity of exposure exceed the RfD, the probability of side effects also increases.

3.3.6 Statistical analysis

Statistical analysis was performed using SPSS 26.0 software (IBM Corp., Armonk, NY, USA) and R version 3.6.3 (R Core Team, 2022, Vienna, Austria). The difference of log Hg concentrations between two groups was tested with Student's *t*-test. The significance of the correlation coefficients (*r*) was confirmed by the test for non-correlation between two variables. The difference of slopes between two regressions was tested by an analysis of covariance (ANCOVA). Differences were declared as significant when $p < 0.05$. The bootstrap method was used to assess the uncertainty of the sample estimates (Kulesa et al., 2015). The non-parametric bootstrap method was applied to the log Hg concentration in each medium. The bootstrap was repeated 10,000 times.

3.3 Results and Discussion

3.3.1 Hg concentration in rice and vegetable samples

The Hg concentrations in the samples are shown in Table 3.2. The Hg concentrations in the rice samples ranged from 26 to 180 $\mu\text{g}/\text{kg dw}$ with an average standard deviation (SD) of $50 \pm 33 \mu\text{g}/\text{kg dw}$ ($n = 20$). These Hg concentrations were much higher than those in the rice samples collected from the reference sites in Indonesia ($8.3 \pm 2.0 \mu\text{g}/\text{kg dw}$, $n = 3$) and in the rice samples bought in markets around the world ($1.1\text{--}5.3 \mu\text{g}/\text{kg dw}$) (Wang et al., 2020). However, the Hg concentration in this study area was lower than that in the rice samples collected from other gold mining areas in Cisit, Gorontalo, Lebaksitu and Sukabumi, Indonesia (mean 90–560 $\mu\text{g}/\text{kg dw}$) (Bose-O'Reilly et al., 2016a; Mallongi et al., 2014; Novirsa et al., 2019; Saragih et al., 2021b) and Camarines Norte, Philippines (600 $\mu\text{g}/\text{kg dw}$) (Murao et al., 2019). Variations in the Hg concentrations among the study areas may result from the scale and years of operation of each ASGM, and the distance from the hotspot (Novirsa et al., 2019).

Table 3.2. Hg concentrations in rice, vegetable, soil, and water samples

	All study area		Nauli Village		Simalagi Village	
	Mean \pm SD	<i>n</i>	Mean \pm SD	<i>n</i>	Mean \pm SD	<i>n</i>
Rice, <i>Oryza sativa</i> ($\mu\text{g}/\text{kg dw}$)	50 ± 33	20	54 ± 43	10	47 ± 18	10
Paddy soil ($\mu\text{g}/\text{kg dw}$)	$5,600 \pm 12,000$	20	$9,000 \pm 16,000$	10	$2,200 \pm 1,800$	10
Vegetables ($\mu\text{g}/\text{kg dw}$)	$2,100 \pm 2,500$	12	$4,500 \pm 2,800$	4	870 ± 920	8
Cassava (<i>Manihot esculenta</i>)	$2,000 \pm 1,600$	6	$3,000 \pm 1,100$	3	880 ± 740	3
Katuk sweet (<i>Sauropus androgynus</i>)	4,800	2	9,000	1	590	1
Papaya (<i>Carica papaya</i>)	2,800	1	–	–	2800	1
Bilimbi (<i>Averrhoa bilimbi</i>)	220	1	–	–	220	1
Water spinach (<i>Ipomoea aquatica</i>)	550	1	–	–	550	1
Susumber (<i>Solanum torvum</i>)	100	1	–	–	100	1
Farm soil ($\mu\text{g}/\text{kg dw}$)	$19,000 \pm 33,000$	12	$55,000 \pm 35,000$	4	$1,400 \pm 1,200$	8
Drinking water ($\mu\text{g}/\text{L}$)	0.59 ± 0.09	16	0.67 ± 0.06	5	0.55 ± 0.08	11
Groundwater ($\mu\text{g}/\text{L}$)	0.62 ± 0.18	16	0.71 ± 0.27	5	0.58 ± 0.09	11
Reference sites						
Rice, <i>Oryza sativa</i> ($\mu\text{g}/\text{kg dw}$)	8.3 ± 2.0	3				
Vegetables ($\mu\text{g}/\text{kg dw}$)	82 ± 23	3				
Drinking water ($\mu\text{g}/\text{L}$)	0.040 ± 0.0058	3				
Groundwater ($\mu\text{g}/\text{L}$)	0.060 ± 0.0058	3				

The Hg concentrations in the vegetable samples ranged from 100 to 9000 µg/kg dw with an average SD of $2,100 \pm 2,500$ µg/kg dw ($n = 12$). The average Hg concentration in the study area was about 25 times higher than that in the vegetable samples collected from the reference sites in Indonesia (82 ± 23 µg/kg dw for green onion, cabbage and baby spinach, $n = 3$). In contrast, the Hg concentrations in vegetables in this study area were equivalent to those in vegetables, such as cassava leaf, papaya leaf and neem leaves collected in gold mining areas in Sukabumi and Bombana, Indonesia (1,780–9,900 µg/kg dw) (Basri et al., 2020b; Saragih et al., 2021b), and were higher in concentration than that in the cassava's edible part (115–3,331 µg/kg dw) and peel (370–991 µg/kg dw) collected near an ASGM site in Ghana (Addai-Arhin et al., 2022).

The Hg concentration in the vegetables was highest in katuk sweet leaf (mean 4,800 µg/kg dw, $n = 2$), followed by *Carica papaya* flower (2,800 µg/kg dw, $n = 1$) and cassava leaves (mean 2,000 µg/kg dw, $n = 6$). The mean Hg concentration in the vegetables was about 40 times higher than that in the rice. The difference in Hg concentration between the vegetables and rice samples can be attributed to the variation in plant parts analysed. While no parts were removed from the vegetable samples, the husks were excluded from the rice samples. Removing the husks could result in a lower Hg concentration since the grains generally have lower Hg levels compared to the roots and husks (Enamorado-Montes et al., 2021).

3.3.2 Hg concentration in soil samples

The Hg concentrations in the paddy soil ranged from 260 to 58,000 µg/kg dw with an average SD of $5,600 \pm 12,000$ µg/kg dw ($n = 20$) (Table 3.2). In contrast, the Hg concentrations in the farm soil ranged from 180 to 100,000 µg/kg dw with an average SD of $19,000 \pm 33,000$ µg/kg dw ($n = 12$). The mean Hg concentration in the farm soil was about 3.4 times higher than that in the paddy soil due to shorter distances to the burning sites, for Hg farm soil (average length of 390 metres) and paddy soil (average length of 430 metres), emphasising the importance of spatial factors in Hg contamination. The Hg concentrations in the paddy field soil were similar to those found in previous studies. For example, research conducted in ASGM areas of Buladu, Gorontalo, Indonesia, showed that the Hg concentrations in the paddy field soil ranged from 484 to 4,244 µg/kg (Mallongi et al., 2014). Several studies (Krisnayanti et al., 2012; Tomiyasu, Kono, Kodamatani, Hidayati, et al., 2013b) have reported significantly elevated levels of Hg in

ASGM areas in Indonesia, with the highest concentrations observed in samples collected near mining operations.

3.3.3 Hg concentration in water samples

The Hg concentrations in the drinking water ranged from 0.45 to 0.75 $\mu\text{g/L}$ with an average SD of $0.59 \pm 0.090 \mu\text{g/L}$ ($n = 16$) (Table 3.2). The average concentration of Hg in drinking water analysed in this study was comparable to that collected from a gold mining area in Pakistan (0.91 $\mu\text{g/L}$) (Riaz et al., 2019) and other areas (sampling site unknown) in Iran (0.31 $\mu\text{g/L}$) (Yeganeh et al., 2013) and Nepal (0.5 $\mu\text{g/L}$) (Sarkar et al., 2022). These Hg concentrations around gold mining areas have higher order of magnitude than that collected from the reference site in Indonesia ($0.050 \pm 0.0082 \mu\text{g/L}$).

The Hg concentrations in the groundwater ranged from 0.45 to 1.3 $\mu\text{g/L}$ with an average and SD of $0.62 \pm 0.18 \mu\text{g/L}$ ($n = 16$). No difference in Hg concentration between the groundwater and the drinking water was observed ($p = 0.59$).

3.3.4 Relationship of Hg concentration between food and soil samples

The relationship of log Hg concentration between the food (rice and vegetables) and soil samples is shown in Fig. 3.2. The log Hg concentration in the food was significantly correlated with that in the soil ($p < 0.05$), indicating that the Hg concentration in the food reflected that in the soil. Furthermore, the slope in the regression equation between the vegetables and the farm soil differed significantly from the slope between the rice and the paddy soil (ANCOVA, $p < 0.05$) (Fig. 3.2). The larger slope for the vegetables suggests that the vegetables may absorb more Hg than the grain rice from the soil and that they may have received some of the load from atmospheric deposition.

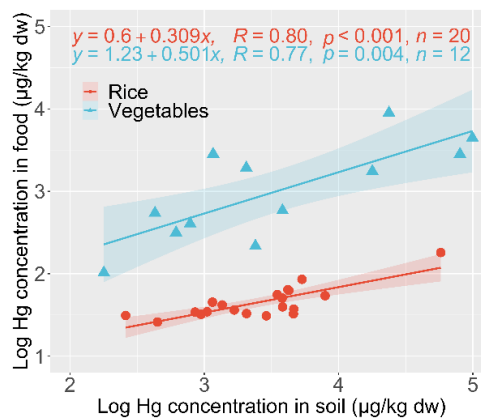


Figure 3.2. Relationship of log Hg concentration between the food and soil samples.

The wider 95% confidence interval in the regression equation for the vegetables than for the rice may be explained by the variation of the Hg concentration in the vegetables. The mean ratio of the Hg concentration between the rice and soil was about 0.0090, but that for the vegetables varied from 0.06 (cassava leaves, $n = 6$) to 2.4 (*Carica papaya* flower, $n = 1$) depending on species (Fig. S2). This variation may be caused by differences in the part analysed (leaf, flower and fruit), species-specific physiological factors, such as the absorption mechanism, and the cultivation environment, such as the distance from the burning site. The ability of plants to accumulate Hg in their tissues differs between species. Hg can be accumulated by plant roots through the process of transpiration or dry deposition, which occurs through foliar absorption mediated by stomata (Rea et al., 2001). For the rice samples, the soil Hg pool was confirmed to be the primary source of inorganic Hg in the roots and a unique bioaccumulation pathway of MeHg for rice plant tissues was identified (B. Meng et al., 2010, 2011; M. Meng et al., 2014).

3.3.5 Spatial distribution of Hg concentration in the study area

The Hg concentrations in the soil, vegetable and drinking water samples collected in Nauli village were statistically higher than those in Simalagi village (Table 3.2 and Fig. 3.3). The higher Hg concentration in Nauli village probably resulted from the facilities and activities relating to ASGM (Fig. 3.1). Nauli village has a burning facility, many ball mills and more frequent activities than Simalagi village.

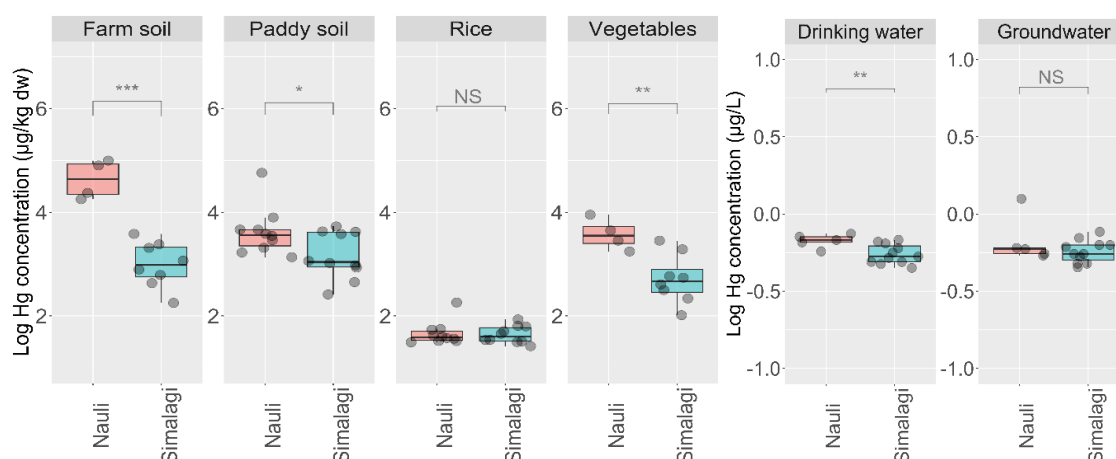


Figure 3.3. Comparison of log Hg concentration in the samples between the villages. The t-test was performed on the log concentrations. ***, **, * and NS in the figure indicate $p < 0.001$, $p < 0.01$, $p < 0.05$, and not significant, respectively.

The relationship between the Hg concentration and the distance from the burning facility to the sampling site was examined (Fig. 3.4). The log Hg concentrations in samples other than groundwater decreased significantly with increasing distance from the burning facility to the sampling site ($p < 0.05$). This result suggested that the Hg concentrations in the samples were mainly explained by the distance from the burning facility to the sampling site. The slopes and intercepts in the regression equations were similar between the farm and paddy soil samples (Fig. 3.4 (a)). Furthermore, although the mean Hg concentration differed between the farm and paddy soil samples, the farm soil corresponded well with the paddy soil at 300 to 600 m from the burning facility. The Hg concentration in the soil around the burning facility was estimated to be on the order of about $10^5 \mu\text{g/kg dw}$ from the intercept of the regression equation in Fig. 3.4 (a).

The Hg concentration on the rice and vegetables also decreased with increasing distance from the burning facility to the sampling site (Fig. 3.4 (b)). These food samples were affected by the Hg concentrations in the soil and the potential for atmospheric deposition of Hg, as described in Section 3.4.4. The Hg concentration in the drinking water decreased significantly with increasing distance from the burning facility to the sampling site, but that in the groundwater did not (Fig. 3.4 (c)).

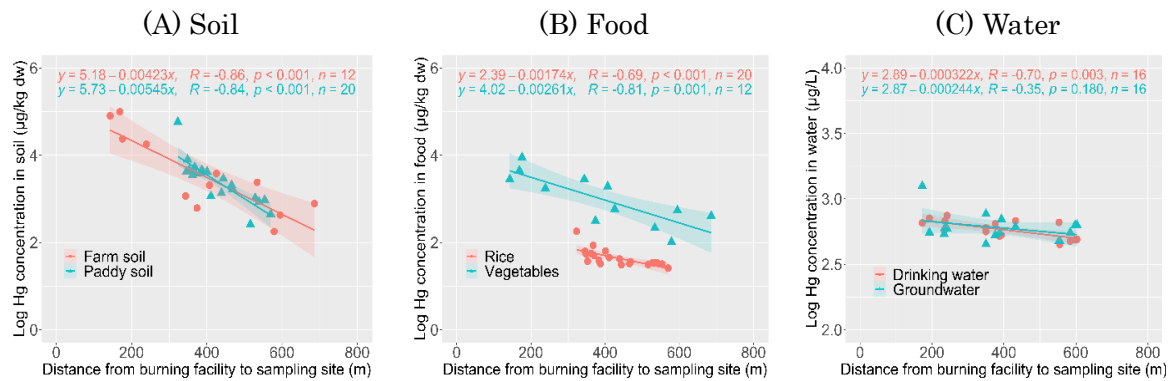


Figure 3.4. Relationship between the log Hg concentration in the samples and the distance from the burning facility to the sampling site. The site of the burning facility is 0 m on the x-axis.

3.3.6 Evaluation of Hg contamination in the study area

To obtain a more precise assessment, the non-parametric bootstrap method was applied to the log Hg concentration in the food, soil and water samples (Fig. S1 and Table S1). The proportion of samples exceeding the safe value proposed by Indonesia Food and

Drugs Administration (30 µg/kg) (BPOM, 2017) was estimated to be 82% for rice. The proportion exceeding the standard value given by the FAO/WHO (100 µg/kg) (WHO, 2006b) was estimated to be 96% for vegetables. The Indonesian Ministry of Environment and Forestry has set a permissible tolerable value of 300 µg/kg for agricultural soil (Minister of Environment and Forestry, 2021) while the Council of the European Communities standard for agriculture soil is 1000 µg/kg (Council of the European Communities, 1986). The proportions of paddy soil and farm soil samples exceeding the Indonesian values were estimated to be 97% and 89%, respectively. Thus, almost all the food and soil samples in this area exceeded the safe value or the permissible tolerable value and these results indicate that severe Hg contamination is occurring in the target area. The Indonesian standard is 1 µg/L for drinking water (IMH, 2017b), and the proportions exceeding this value in the drinking water and the groundwater were estimated to be 0.03% and 1.4%, respectively. Thus, the proportions of water samples exceeding the standard value were smaller than those of the food and soil samples.

Table 3.3. Average daily intake (ADI) and hazard quotient (HQ) of Hg for the resident

Sample	Nauli village				Simalagi village			
	ADI (µg/kg/day)		HQ		ADI (µg/kg/day)		HQ	
	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Oral (<i>RfD</i> = 0.3 µg/kg/day)								
Rice	0.34	0.63	1.1	2.1	0.30	0.54	1.0	1.8
Vegetables	0.52	0.93	1.7	3.1	0.10	0.18	0.3	0.6
Drinking water	0.01	0.02	0.04	0.07	0.01	0.02	0.03	0.06
Soil	0.04	0.28	0.13	0.95	0.00	0.02	0.01	0.08
Dermal (<i>RfD</i> = 0.3 µg/kg/day)								
Groundwater	4.9×10^{-7}	2.5×10^{-6}	1.6×10^{-6}	8.5×10^{-6}	4.5×10^{-7}	2.3×10^{-6}	1.5×10^{-6}	7.7×10^{-6}
Soil	1.6×10^{-2}	8.0×10^{-2}	5.2×10^{-2}	2.7×10^{-1}	1.4×10^{-3}	7.0×10^{-3}	4.5×10^{-3}	2.3×10^{-2}
Inhalation (<i>RfD</i> = 0.086 µg/kg/day)								
Soil	5.7×10^{-6}	1.3×10^{-5}	6.7×10^{-5}	1.5×10^{-4}	5.0×10^{-7}	1.1×10^{-6}	5.9×10^{-6}	1.3×10^{-5}

3.3.7 Resident health risk assessment

The ADIs and HQs of food, drinking water, soil and groundwater samples are shown in Table 3.3. The ADIs of the residents from rice for children and adults in all the study areas were far higher than the USEPA recommended guideline values. However,

vegetable samples in Simalagi village were indeed lower than the USEPA recommended guideline values. For oral exposure, the consumption limit is 0.3 $\mu\text{g}/\text{kg}/\text{day}$ for adult and children (DEA, 2010b; US EPA, 2011). The HQ of rice and vegetables was the highest of the consumption sources in Nauli village. The HQ of rice was 2.1 and 1.1 in Nauli village and 1.8 and 1.0 in Simalagi village for children and adults, respectively. The HQ of the vegetables from Nauli village exceeded the safe value but was below the safe value in Simalagi village for both adults and children. The high HQ for rice may be caused by the high daily consumption of rice in the community because it is the staple food. Likewise, the HQ for vegetables from Nauli village was high because of the high Hg concentration in these plants. However, the ADI of drinking water and soil was far below the safe limit and the HQ value was below 1 for all study areas for children and adults. The soil ingestion pathway exposure has a value that is near the threshold for having an impact on human health, especially Nauli village.

Even though the ADI of dermal and Inhalation exposure was very low, it has had an impact on society. According to USEPA, the absorption limit for dermal exposure is 0.3 $\mu\text{g}/\text{kg}/\text{day}$ for adults and children, and the absorption limit for inhalation exposure is 0.086 $\mu\text{g}/\text{kg}/\text{day}$ for adults and children (DEA, 2010b; US EPA, 2011). The relatively low ADI and HQ values arise from the low potential for entry into the body owing to various factors, such as dermal absorption and particulate emission. Several similar studies have also shown a low HQ value for oral ingestion, inhalation and dermal exposure from soil in both children and adults. Sources of exposure vary between ASGM and agriculture (Table S2).

The health risk assessment of Hg via oral ingestion exposure showed that rice and vegetables have an HQ value above 1, which means that rice and vegetables have the potential to affect health in children and adults. A similar study by Novirsa et al. (2020) in Indonesia shows that rice consumption and oral exposure have a high HQ value and can affect health (Novirsa et al., 2020b). The average ADI of people living in the mining area in this study was 0.116 $\mu\text{g}/\text{kg}/\text{day}$ (range 0.040–0.240 $\mu\text{g}/\text{kg}/\text{day}$). Natasha et al. (2020) calculated the possible health risks associated with the consumption of Hg-contaminated food both in adults and children (Natasha et al., 2020). The study concluded that the accumulation of Hg in edible plant parts can pose health hazards to both adults and children. As a result, it emphasised the need for effective measures to control the

transfer of Hg from soil to plants and ultimately to humans to minimise health risks.

3.4 Conclusions

Mercury contamination in the gold mining area, Mandailing Natal District, Indonesia, was investigated. The study found high concentrations of Hg in the food, soil and water samples from the study area, despite the gold mining activity only being conducted for about 15 years. The Hg concentrations in the samples collected in Nauli village were statistically higher than those in Simalagi village, and the horizontal distribution of Hg concentrations in the study area was strongly related to the distance from the burning facility. This result suggested that a distance of more than 500 m from the burning site is needed to reduce Hg exposure. According to the safe value from the Indonesian National Standard or the permissible tolerable value from the FAO/WHO, the level of Hg contamination in vegetables and rice was above these values, making them unfit for human consumption. With an HQ greater than 1, this study indicates a possible health risk for people who mostly consume locally grown vegetables and grains. Hg contamination in vegetables and rice cultivated in the regions of this study will certainly rise over time due to the Hg deposition to the soil by gold mining activities. Further monitoring of and countermeasures against Hg contamination are required to reduce the health risks for residents.

**CHAPTER IV
CONCLUSIONS**

4.1 Conclusions

In conclusion, the two studies collectively present a disconcerting panorama of mercury contamination that pervades various environmental components. These contaminants span across the spectrum, affecting air, soil, water, sediment, and even permeating into the food chain, including vegetables and grains. In the first study, the pronounced emphasis on soil as a primary contributor to mercury contamination underscores its pivotal role, particularly in influencing the safety of agricultural produce, specifically vegetables. This highlights the imperative need to devise strategies to curtail and mitigate soil pollution as a fundamental step in ensuring the integrity of food sources in these regions.

Furthermore, the study identifies a particularly vulnerable group: areas characterized by artisanal small-scale gold mining (ASGM). Vegetables originating from such regions are notably unsafe for consumption due to heightened mercury contamination, posing a considerable health risk to the local population. The implications are profound, necessitating urgent intervention and comprehensive regulatory measures in ASGM areas to safeguard both human health and the environment.

The presence of significant mercury concentrations in human hair, as indicated in the first study, raises concerns about direct human exposure. This observation underscores the pressing need for health monitoring and risk assessment in affected communities, emphasizing the urgent necessity for public health intervention and education to mitigate potential adverse health outcomes.

Additionally, the study hints at a possible correlation between soil-vegetable and sediment-aquatic food contaminations, suggesting that controlling mercury contamination at its source, such as soil and sediment, may have a cascading effect in reducing contamination in food sources. This notion underscores the interconnectedness of various environmental compartments and underscores the importance of holistic environmental management strategies.

Shifting focus to study case in the second study, it sheds light on the enduring legacy of mercury contamination in the Mandailing Natal district, even after 15 years of gold mining activity cessation. This highlights the persistence of the problem and the long-term ecological impact of such mining operations. The study stresses that mercury contamination remains prevalent in environmental components like soil, water, and food,

with a notable emphasis on local vegetables and grains. The mention of a "Hazard Quotient >1" underscores the immediate health risks posed by elevated mercury levels in these food sources.

Moreover, the study offers a stark warning regarding the future trajectory of mercury contamination, particularly in crops. Ongoing mining activities are predicted to exacerbate mercury contamination in food sources, necessitating ongoing monitoring and intervention efforts to mitigate long-term health and environmental risks.

In conclusion, these two studies present a sobering account of widespread mercury contamination across multiple environmental media and food sources. They emphasize the urgent need for comprehensive intervention strategies, including soil remediation, health monitoring, public education, and regulatory measures, especially in ASGM areas. Furthermore, they underscore the enduring impact of past mining activities and the imperative of proactive measures to protect both the environment and the health of local communities from the continued threat of mercury contamination.

4.2 Recommendation

Considering the significantly elevated levels of mercury (Hg), particularly in the food chain within the vicinity of the Artisanal and Small-Scale Gold Mining (ASGM) area, it is strongly advised that the local community refrains from consuming such food items. Furthermore, it is recommended that the government disseminate this information to the affected community.

There is a distinct possibility that mercury contamination in the environment, specifically in soil, sediment, vegetables, and rice within the ASGM area, will progressively escalate over time due to the deposition of mercury resulting from gold mining activities.

Further studies on risk management strategies in environmental media are crucial to prevent new cases of mercury contamination. Comprehensive evaluations of mitigation approaches are needed to address the complex challenges posed by mercury pollution. By exploring innovative and context-specific techniques, researchers can develop targeted interventions to minimize mercury emissions and enhance containment measures at pollution sources.

A collaborative effort involving government bodies, research institutions, and local communities is essential to effectively reduce the risk of mercury exposure and safeguard the health and well-being of the Indonesian population and the environment. Implementing and enforcing countermeasures to mitigate mercury contamination is vital to prevent further escalation of health risks for residents in affected regions.

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