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Original Article

The health risk assessment of mercury in rice from paddy fields around Nam Son landfill, Hanoi, Vietnam

Nguyen Thi Quynh[1], Huiho Jeong[1], Ahmed Elwaleed[1], Yasuhiro Ishibashi[2] and Koji Arizono[2,3]

[1]*Graduate School of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto, 3-1-100 Tsukide, Higashi-ku, Kumamoto 862-8502, Japan*
[2]*Faculty of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto, 3-1-100 Tsukide, Higashi-ku, Kumamoto 862-8502, Japan*
[3]*Graduate School of Pharmaceutical Sciences, Kumamoto University, 5-1 Oe-honmachi, Chuo-Ku, Kumamoto 862-0973, Japan*

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ABSTRACT — Mercury (Hg) accumulation in rice is a health concern due to the consumption of rice as the staple food. This study evaluated the mercury contamination in rice plants, which are typical foods cultivated in the Red River Delta. During the harvest season, rice samples were collected and separated into husk and brown rice, together with polished white rice and bran rice from mill shop. For brown rice, the Hg concentration ranges from 7.18 ± 0.73 to 16.32 ± 2.57 µg/kg. Additionally, brown rice samples near landfill or highway tend to have higher Hg concentrations than sites farther away. Hazard quotient (HQ) was used to measure the health risk of Hg in this study. HQ values of male and female all were less than one, indicating that consuming rice from Nam Son and Bac Son might not cause potential human health risk of Hg exposure.

Key words: Mercury, Nam Son landfill, Rice, Paddy field, HQ

INTRODUCTION

Mercury (Hg) is one of the ten most toxic elements that affect on human health, in the natural, Hg exist in the three form: elemental mercury or quicksilver (metallic mercury and vapor mercury, Hg^0), inorganic mercury (Hg^+ and Hg^{2+}), and organic mercury (methylmercury and ethylmercury), from natural sources (volcanic emission, cinnabar) and anthropogenic sources (coal-fired electric utilities, waste combustion, and hazardous-waste incinerators (Cariccio *et al*., 2019). Hg compounds can cause kidney failure, nervous system disorders, fetal toxicity, and death (Cariccio *et al*., 2019; Fernandes *et al*., 2012). One of the most serious Hg poisoning cases in history was the Minamata disaster in Japan, which caused the deaths of 1043 people, the reason is consumption contaminated fish and shellfish by the discharge activities of factory manufacturing the chemical acetaldehyde (Harada, 1995). Hg is unique heavy metal due to its toxicity, long-distance transport, persistence and bio-accumulation (Wang *et al*., 2016).

Rice consumption has recently been considered by researchers as one of the main routes of mercury exposure to the human body through the food chain besides fish (Horvat *et al*., 2003; Du *et al*., 2021). Paddy fields are a very special environment, not only the amount of Hg accumulated from the air, but rice soil also accumulates mercury from cultivation such as irrigating farmland

Correspondence: Koji Arizono (E-mail: arizono@kumamoto-u.ac.jp)

with Hg-contaminated water, extensive use of fertilizers, and pesticides containing Hg (Huang *et al*., 2022). In particular, paddy fields located near anthropogenic sources such as artisanal and small-scale gold mining (ASGM) (Qiu *et al*., 2008), industrial areas, and landfills (Tang *et al*., 2015) are increased risk potential for Hg contamination (Tang *et al*., 2020). Numerous studies have shown that rice, fish, and fruits harvested from area located near sources of mercury pollution is contaminated with significant amounts of Hg (Peng *et al*., 2015; Wu *et al*., 2013).

Rice (Oryza sativa) is a staple food crop, and Vietnam is one of five major rice exporters (India, Pakistan, Thailand, the US and Vietnam) (WEF, 2022). The Red River Delta (RRD) located in the North of Vietnam is the second largest granary in the country, behind Mekong Delta located in the South. The average annual rice cultivated area of RRD is roughly 1.07 million hectares (Vietnam General Statistics Office - GSO), accounting for about 14% of Vietnam's total rice growing area (USDA, 2020), also is the second largest granary in the country. With the advantage of favorable terrain, RRD is a large plain, supplied with annual alluvium from the Red River, creating fertile land and creating favorable conditions for cultivating crops. RRD has two main crops a year: the summer rice crop and the Summer-Autumn crop.

In Vietnam, rice products are always fully utilized. Besides rice, other products including bran, rice husk, and straw are also used. Specifically, rice bran is the outer brown layer, including the rice germ that is removed during milling of brown rice to produce milled rice (Rosniyana *et al*., 2009), which is used as food for livestock (pig, cow, chicken, duck, etc.). Rice husk are used as fertilizer, barn bedding, fuel, and mushroom growing material. Straw is used as food for cattle, fertilizer, and fuel. Therefore, Hg pollution in rice not only affects the quality of rice but also affects the quality of livestock and the environment, hence Vietnam is the third country had the highest Hg production density (behind Bangladesh and India) (Liu *et al*., 2019).

In this study, we collected rice samples from paddy fields surrounding the Nam Son landfill area to evaluate the level of Hg contamination in rice around landfill area. The influence of manual drying method on Hg level in rice was discussed. Besides, for the first time, potential health risk assessment based on local resident's consumption of rice harvested from the study area.

MATERIALS AND METHODS

Sampling site

The study site is in paddy field around Nam Son landfill site in Soc Son district of Hanoi, Vietnam, located in Red River Delta (RRD), where is the primary region for rice cropping in northern Vietnam (Nguyen *et al*., 2016).

Nam Son waste treatment complex has a total area of 83.5 hectares, including a wastewater treatment plant, a waste power plant and closed and operating landfills (Fig. 1). The process of transporting, burying and treating waste can release toxic substances including organic compounds (Andrews *et al*., 2011) and heavy metals (Hoai *et al*., 2021; Giang *et al*., 2018) into the environment around the landfill. This study focuses on assessing the level of mercury contamination in rice from paddy fields surrounding Nam Son landfill. Due to terrain characteristics and incomplete planning, some rice fields are only 200 m from Nam Son landfill. In previous research we have demonstrated that rice paddy soil surrounding the landfill area is likely to be contaminated with Hg from the landfill (Thi Quynh *et al*., 2024), therefore a study to evaluate the absorption and transport of Hg from the soil into rice plants in this area is really necessary.

Sample collection

Rice samples collection

Sampling campaign was conducted in the paddy fields around Nam Son landfill on September 15th 2021, during the Summer-Autumn harvesting season. A total of 15 rice samples (R01 – R15) were collected in Nam Son and Bac Son commune, using a sickle. After collection, all rice samples were washed with tap water 3 times and rinsed with milli-Q water. They were dried in the oven at 40°C for two weeks. Then, husks were separated from grains by a huller. Brown rice and husk samples were ground using a porcelain mortar and pestle and sifted through a 200 µm mesh size sieve to obtain finer particles. The dried samples were labeled and stored in a polyethylene ziplocked bags and kept at 4°C.

In addition, due to the farming habits rice is often dried on the ground of garden, road or highway under sunlight, leading to the rice being contaminated with Hg. Therefore, ten polished white rice and its rice bran (n = 10, and n = 10) were collected at mill shops located in Bac Son commune and Nam Son commune. At the mill shops, rice was milled removing the husk, bran layers and germ. Polished white rice, and rice bran were then ground using a porcelain mortar and pestle and sifted through a 200 µm mesh size sieve to obtain finer particles. The dried samples were labeled and stored in a polyethylene ziplocked bags and kept at 4°C in the dark.

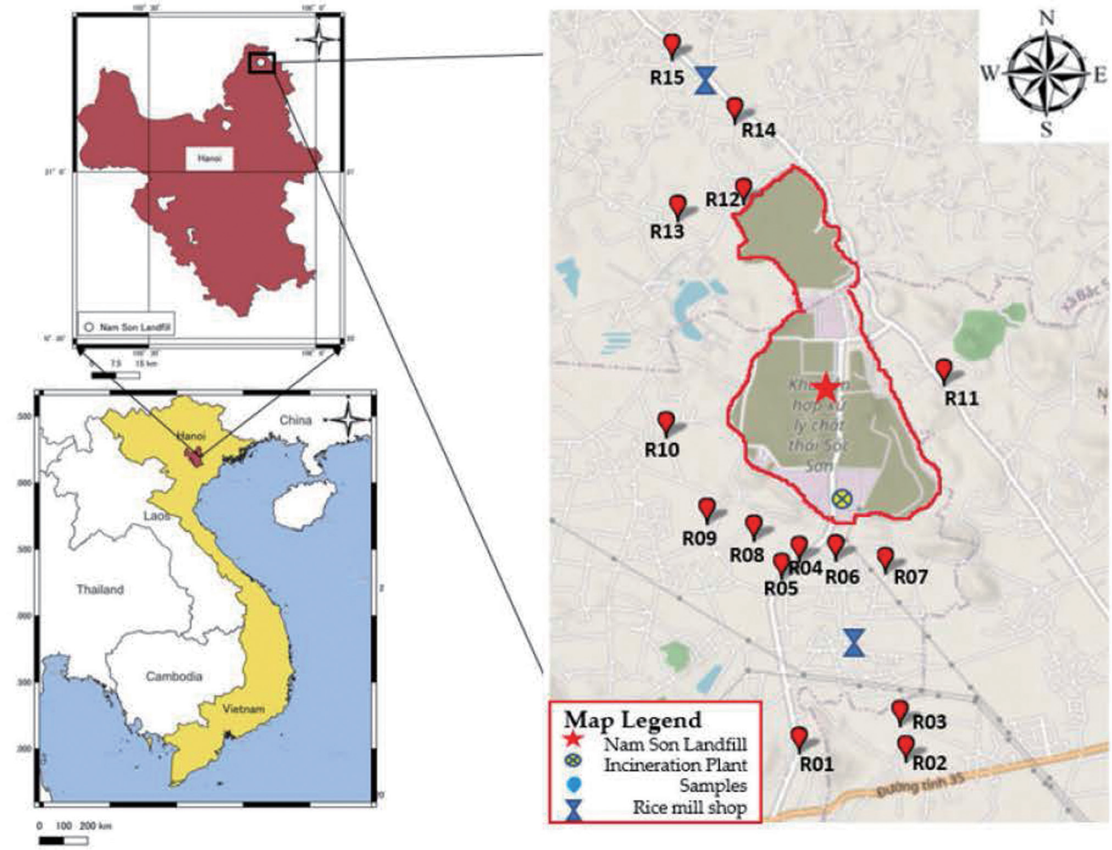

Fig. 1. Map of sampling site, Nam Son, Soc Son, Hanoi

Sample analysis

Hg analysis

The Hg concentration in all samples were determined using the direct thermal decomposition mercury analyzer (MA-3000, Nippon Instruments Corporation, Tokyo, Japan). Approximately 50 mg of each sample was weighed into sample boats in triplicates and placed in MA-3000. Calibration curves were generated using standard solutions of Hg^{2+} at concentrations of 10 µg/kg, 100 µg/kg and 1000 µg/kg for both low and high calibration curves.

Ecological Risk Assessment

Potential ecological risk index (PER) was employed using the Hakanson, 1980 model. This index considers the sensitivity of biological communities, toxicity, measured concentration and background concentrations. Hg contamination coefficient (C) was calculated using Equation 1. Then potential ecological risk index (PER) of Hg was calculated using Equation 2. Input parameters for these equations are detailed in Table 1.

$$\mathbf{C = C_{Hg}/B_{Hg}} \quad \text{(Equation 1)}$$

C < 1: low contamination factor (indicating low sediment contamination of the substance in question); 1 < C < 3: moderate contamination factor; 3 < C < 6: considerable contamination factor; C > 6: very high contamination factor.

$$\mathbf{PER = C \times T} \quad \text{(Equation 2)}$$

PER < 40: low risk, 40 ≤ PER < 80: moderate risk, 80 ≤ PER < 160: considerable risk, 160 ≤ PER < 320: High risk, PER ≥ 320: very high risk.

Table 1. Input parameters for evaluating Hg concentration coefficient (C), and Potential ecological risk index (PER).

Parameter	Meaning	Value (unit)	Reference
C_{Hg}	Hg concentration in rice	(µg /kg)	This study*
B_{Hg}	the local Hg concentration background for soil	7.47 (µg /kg)	Thi Quynh *et al*., 2024
T	The toxic factor of Hg	40	Hakanson *et al*., 1980

Table 2. Input parameters for evaluating Estimated Weekly Intake (EWIhm) and Hazard Quotient (HQ).

Parameter	Meaning	Value (unit)	Reference
EF	The exposure frequency	365 (days/year)	USEPA, 1989
ED	The exposure duration	70 (years)	USEPA, 1989
IR_{hm}	The rice grain ingestion rate	0.589 (kg/day)	Chu *et al*., 2021
C_{Hg}	The concentration of Hg in rice grain	(µg /kg)	This study*
φ	The bio-accessibility of Hg	45 (%)	Lin *et al*., 2019
BW_{hm}	The average body weight	58 for male and 45 for female (kg)	Chu *et al*., 2021
AT	The average exposure time for non-carcinogens	For non-carcinogens, AT = ED × EF	USEPA, 1989
RfD	The oral reference dose of Hg via the oral exposure route	0.0003 (mg/kg/d)	Jin *et al*., 2023

Tolerable intake of Hg and risk characterization

a) For human

Health risks associated with Hg calculated using the model in the FAO/WHO Joint Expert Committee on Food Additives (JECFA) withdrew the former Provisional Tolerable Weekly Intake (PTWI), it indicates the noncarcinogenic risk of oral exposure. The JECFA established a PTWI for inorganic Hg is 4 µg /kg b.w. The Estimated Weekly Intake (EWI_{hm}) of Hg from consumption of rice can be calculated by Hg concentration in rice with the consumption levels per week dividing by the average body weight of the population by sex, using the Equation 3 (Ahmad *et al*., 2022).

Health risks associated with Hg calculated using the Hazard quotient (HQ) was used to estimate the probability of health effects due to the exposure through consumption of rice. The following Equation 4 was used to calculate HQ.

Input parameters for these equations are detailed in Table 2.

$$\mathbf{EWI_{hm} = (IR_{hm} \times 7 \times C_{Hg} \times \varphi) / BW_{hm}} \qquad \text{(Equation 3)}$$

$$\mathbf{HQ = (EF \times ED \times IR_{hm} \times C_{Hg} \times \varphi) / (BW_{hm} \times RfD \times AT)} \qquad \text{(Equation 4)}$$

HQ < 1: no negative noncarcinogenic health effects for the exposed human population, 1 ≤ HQ: the pollutant is likely to have some noncarcinogenic deleterious effects on human health.

b) For Chicken

Similar to humans, the Estimated Weekly Intake (EWI) of Hg from consumption of rice bran by chickens can be calculated based on Hg concentration in rice bran. The hypothetical chickens considerd in this study were commercial broiler chickens (Hybro strain), using the Equation 5 (Ahmad *et al*., 2022).

$$\mathbf{EWI_{ck} = (IR_{ck} \times 7 \times C_{HgRB}) / BW_{ck}} \qquad \text{(Equation 5)}$$

Where:

IR_{ck} : the rice bran ingestion rate (0.15 kg/kg/day (Zhang *et al*., 2021))
C_{HgBR} : the concentration of Hg in rice bran (µg /kg)
BW_{ck} : The average chicken weight (0.22 kg (Atuahene *et al*., 2000))

Quality assurance (QA) and quality control (QC)

In the laboratory analysis, strict measures were taken to ensure a contamination-free environment. An ultra-clean bench was used, and all water used had a high purity level of 18.2 MΩ·cm @ 25°C (Millipore, USA). Reagents of guaranteed purity (GR) were utilized to avoid any contamination or ultra-low blanks.

During the analysis of ultrapure water was employed as a blank to establish baseline measurements. To ensure the accuracy and reliability of the Hg analyses, a comprehensive quality assurance and quality control program was implemented. Method blanks, certified reference

Table 3. The C and PER values of brown rice in Nam Son and Bac Son commune.

Location		Concentration (µg/kg) (min – max)	C	PER
Rice paddy field	Nam Son (South of the landfill)	11.31 (7.68 - 16.32)	1.51	60.6
	Bac Son (North of the landfill)	8.81 (7.23 – 10.94)	1.18	47.2

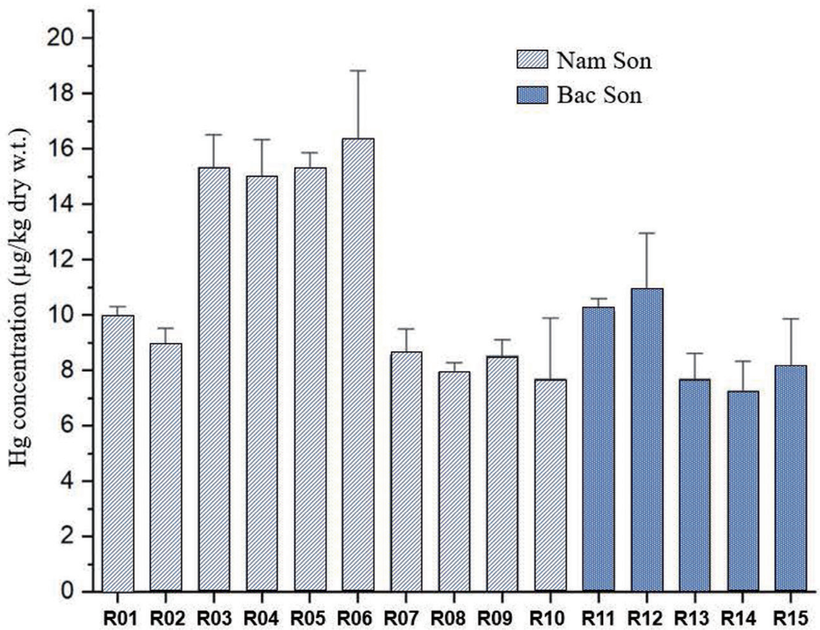

Fig. 2. Hg concentration in brown rice samples.

materials (CRM), specifically ERM-CC580 (Institute of Reference Materials Measurements, Belgium) for Hg, and duplicate samples were used. The obtained results from this method demonstrated excellent agreement with the reference values, with a mean value of 132 ± 3 mg/kg. The recovery rates for the certified reference materials ranged from 96.9% to 101.7% for the Hg analysis, and the relative standard deviations were consistently < 5% for Hg.

Statistical analysis

The statistical analysis was conducted using IBM SPSS Statistics SPSS 26.0 software from IBM Corporations, New York, USA. The Shapiro-Wilk was used to assess whether data might be well-described by a Normal distribution. The dietary exposure levels for Hg by sex and residential area categories were compared by one-way analysis of variance (ANOVA). Graphs were crafted utilizing OriginPro and Microsoft Excel.

RESULTS AND DISCUSSION

Hg concentration in rice

Hg concentration in brown rice was shown in Fig. 2. The mean concentrations of Hg in brown rice measured at Nam Son and Bac Son were 11.3 ± 1.08 µg/kg and 8.81 ± 1.22 µg/kg, respectively.

Specifically, sampling points with higher Hg concentrations include R03, R04, R05, R06 with Hg concentration ranging from 15.0 ± 1.39 µg/kg to 16.3 ± 2.57 µg/kg. It can be seen that these are all locations in Nam Son located near the main gate and close to the landfill. A similar trend was also observed in Bac Son, R11 and R12 (10.2 ± 0.47 µg/kg and 10.9 ± 1.81 µg/kg) located near roads and the landfill had higher Hg concentrations than other sites. This may be explained by the fact that during the time these rice samples were collected, the Thien Y waste power plant located near the south gate of the landfill was under construction (September 2021). Therefore,

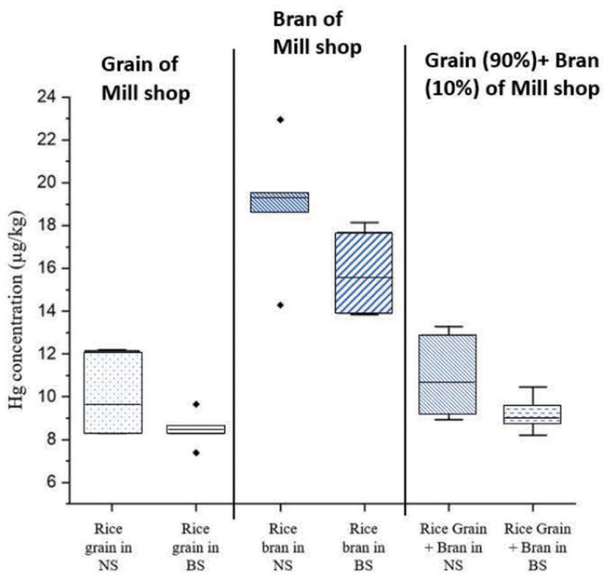

Fig. 3. Hg concentration in ten polished white rice (grain rice) and bran rice from mill shops in Nam Son commune (NS) and Bac Son commune (BS).

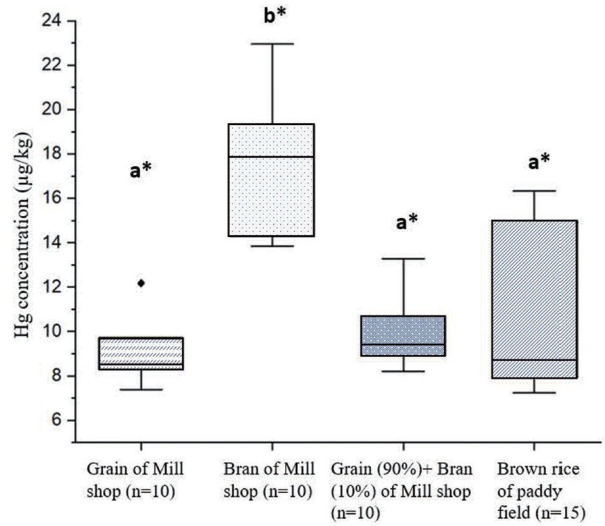

Fig. 4. Comparison between Hg concentration in polished white rice (grain rice) and bran rice from mill shop and brown rice from paddy field.* The same letters represent no significant difference ($p > 0.05$) and the different letters represent a significant difference ($p < 0.05$) by the one-way ANOVA test.

besides garbage trucks, a large number of trucks transporting construction materials pass through Nam Son Street to the south gate of the landfill. This contributes to increased mercury concentrations in the environment in the southern region, leading to higher Hg concentrations in rice grains.

Similar mean ± SD Hg concentration have been reported from Hg-contaminated site in Pakistan in the study of Aslam *et al.*, 2020 (12.20 ± 16.97 ng/g). Chu *et al.*, 2021 reported Hg concentration in rice in not Hg-contaminated and mining activity area was 7 (nd – 11) µg/kg. Compared with the study of Du *et al.*, 2018 conducted with rice samples collected in areas not contaminated with Hg, with an average value of Hg concentration in rice samples of 4.6 ± 3.0 µg/kg in Hubei and 3.9 ± 1.6 µg/kg in Anhui, China (Du *et al.*, 2018), brown rice samples from the area around Nam Son landfill had higher Hg concentrations. This indicated that rice grown in this area is contaminated with a certain amount of Hg from Nam Son landfill.

However, compared with the reported data of Hg concentration in rice of ASGM area in Lebaksitu, Indonesia was 32.2 (9.1 – 115) µg/kg (Novirsa *et al.*, 2020), Hg mining area of 10.3 – 1120 µg/kg of dry w.t in Guizhou, China (Qiu *et al.*, 2008), and 643 µg/kg at the Xunyang Hg mine located in Shaanxi Province, China (Ao *et al.*, 2020), the data obtained from this study was much lower. Because the brown rice collected in this study area is contaminated with Hg from Nam Son landfill, although the degree of impact is there, to compare with areas Hg-contaminated by Hg mining, and ASGM, its concentration is extremely small.

Hg concentration in ten polished white rice and bran rice from mill shops in Nam Son and Bac Son commune are shown in Fig. 3. Both the Hg concentration in polished white rice and rice bran in Nam Son and Bac Son were far below the recommended Vietnamese guideline values of 50 µg/kg for coffee, tea and its products (QCVN 8-2:2011/BYT, 2011). All the samples, Hg concentration in rice brans are higher than its polished white rice, Hg concentration in rice bran are almost equal twice Hg concentration in polished white rice.

Rice bran is a by-product of rice industry containing about 87% dry matter, 11–15% crude protein, 15–20% lipids and 6.5–10% ash (de Campos *et al.*, 2007). Rice bran are feed livestock, hence higher concentrations of Hg in rice bran will increase the potential risk of Hg contamination. Hg pass through the food chain causing a

Table 4. EWI_{hm} and HQ level of polished white rice and brown rice, and EWI_{ck} level of rice bran from Nam Son and Bac Son commune

Location			Concentration (µg/kg) (min – max)	Male		Female		Chicken
				EWI_{hm} (µg/kg b.w)	HQ	EWI_{hm} (µg/kg b.w)	HQ	EWI_{ck} (µg/kg b.w)
Rice mill shop	Nam Son (South of the landfill)	Polished white rice	10.4 (8.47 – 12.2)	0.333	0.158	0.429	0.204	-
		Bran rice	19.5 (15.9 – 22.9)	-	-	-	-	20.4
	Bac Son (North of the landfill)	Polished white rice	8.47 (7.39 – 9.65)	0.271	0.129	0.349	0.166	-
		Bran rice	15.8 (13.8 – 18.2)	-	-	-	-	16.6
Rice paddy field	Nam Son (South of the landfill)	Brown rice	11.3 (7.68 - 16.3)	0.364	0.172	0.469	0.222	-
	Bac Son (North of the landfill)	Brown rice	8.81 (7.23 – 10.94)	0.280	0.134	0.364	0.173	-

-: no applied

potential risk of Hg exposure to humans. An investigation into the amount of rice bran used for livestock production and food consumed by people every day is urgently needed in the next study.

On the other hand, according to Nurul Husna Shafie and Norhaizan Mohd Esa, 2017 researched on the structure of rice grain, rice husk accounts for 20% and brown rice accounts for 80% (of which is rice bran 8%, white rice is 70%, and rice germ is 2%). Based on the structure of rice, we estimated the amount of Hg present in brown rice from the mill shop equivalent to 90% of the Hg concentration in polished white rice (grain rice) plus 10% of the Hg concentration in bran rice (Fig. 4). The result indicating no signification difference between brown rice from mill shops and paddy fields. However, Hg concentration in all bran samples were signification higher than white rice and brown rice from mill shops and paddy fields. Probably suggesting that rice grains absorb Hg from atmosphere such as dry and wet deposition, vehicles, fertilizers, and pesticides containing Hg (Huang *et al*., 2022). Previous studies have confirmed that Hg uptake from rice root hardly transported into rice aerial parts, and Hg absorbed rice grain is mainly in the gaseous elemental Hg (Hg^0_g) entering through stoma (Tang *et al*., 2020). Recently, research by Phan Dinh *et al*., 2022 also detected a relatively high amount of Hg in street dust in Hanoi city. Therefore, the amount of Hg accumulated from the air environment in the bran covering rice grains during maturation has led to the amount of Hg in rice bran being higher than rice grain.

Comparing the Hg concentration between brown rice from paddy fields in Nam Son and Bac Son commune, we observed that Hg concentration in brown rice of Nam Son was higher than its of Bac Son, however, the previous study showed that Hg concentration in soil of Nam Son was lower than its of Bac Son (Thi Quynh *et al*., 2024). This indicated that Hg concentration in soil has no significant effect on Hg concentration in rice, while Hg soil contamination is related to the cannel, drain water of the wastewater plant (the north gate of the landfill), but air contamination is one of essential factors of the rice samples. Ao *et al*., 2020 also demonstrated that significant Hg accumulation in rice grains is a result of atmospheric mercury deposition. Some previous studies have shown that the Hg content accumulated in street dust is higher in soil (Dat *et al*., 2023; Sahakyan *et al*., 2019).

Ecological risk assessment

The results for the contamination coefficient (C), and potential ecological risk (PER) index are shown in Table 3. Base on the assessment, the brown rice from both Nam Son and Bac Son had $1 < C < 3$, indicating moderate contamination factor. The brown rice from both Nam Son and Bac Son had $40 \leq PER < 80$, indicating moderate risk. Hence, all the rice from paddy field around Nam Son landfill may pose potential ecological risks to human, livestock, and the entire paddy field ecosystem. However, continuous release of Hg through landfill activities into the surrounding area may result in higher contamination of the paddy fields and their produce in future.

Health risk assessment

To evaluate the dietary exposure risk from rice grains, we analyzed the estimated weekly intake (EWI) of Hg through polished white rice from mill shop and brown rice for human and rice bran for chickens. The bio-accessibility of Hg in this study used a value of 45% (Lin *et al*., 2019), applied to cooked rice. Considering the worst-case scenario, with the highest Hg bio-accessibility values. Compared to the bio-accessibility for fish, this ratio was lower, Bradley *et al*., 2017 reported up to in raw tuna 75%, and 75 – 92% in raw shrimp (Siedlikowski *et al*., 2016), however the bio-accessibility is reduced if the food is cooked (Abdullah, 2020).

The average EWI_{hm} value of Hg through consumption of polished white rice from mill shop is 0.333 µg/kg b.w and 0.271 µg/kg b.w for male, and 0.429 µg/kg b.w and 0.349 µg/kg b.w for female in Nam Son and Bac Son commune, respectively (Table 4). This value with consumption of brown rice from paddy fields is 0.364 µg/kg b.w and 0.280 µg/kg b.w for male, and 0.469 µg/kg b.w and 0.364 µg/kg b.w for female in Nam Son and Bac Son commune, respectively. Generally, EWI_{hm} of brown rice was higher the polished white rice. All values were lower than the PTWI of JECFA established for inorganic Hg is 4 µg/kg b.w. Along with that HQ values of male and female in Nam Son and Bac Son all were lower than one, indicating that consuming rice from Nam Son and Bac Son might not cause potential human health risk of Hg exposure.

The average EWI_{ck} value of Hg through consumption of rice bran from mill shop is 20.4 µg/kg b.w and 16.6 µg/kg b.w for chicken in Nam Son and Bac Son commune, respectively (Table 4). EWI_{ck} values in chickens are several times greater than EWI_{hm} in humans, suggesting that a large amount of Hg is absorbed by chickens. That amount of Hg can accumulate in chicken parts and affect human health through the food chain. In this study, the Hg concentration in chicken parts such as meat, skin, stomach, liver and bones were not evaluated. Therefore, a future study evaluating the Hg content in chicken parts such as muscle, skin, bond and egg is necessary.

In conclusions, based on the analysis results of rice samples from paddy fields around Nam Son landfill, Hanoi, Vietnam, this study showed the condition of Hg in rice planted in this area. The Hg concentrations in brown rice samples from paddy fields of this study ranges 7.18 ± 0.73 to 16.32 ± 2.57 µg/kg dry w.t. Among them, brown rice samples near landfill or highway tend to have higher Hg concentrations than sites farther away. This result indicates that activities related to traffic and construction sites may play an important role in the dispersion of Hg in the study area.

The brown rice from both Nam Son and Bac Son had 40 ≤ PER < 80, indicating rice from paddy field around Nam Son landfill pose potential ecological risks to human, livestock, and the entire paddy field ecosystem.

HQ was calculated to assess the potential health risk of Hg in this study. HQ values of male and female all were less than one, it demonstrated a relatively low risk of Hg ingestion cause by rice consumption planted in paddy fields around Nam Son landfill. However, the EWI of chickens is many times greater than that of humans, suggesting that a large amount of mercury is absorbed by chickens through the consumption of rice bran, thereby causing a potential risk to human health through the food chain.

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

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