




Trace element contamination in rice and its potential health risks to consumers in North-Central Vietnam

Thao Thanh Le · Kyoung-Woong Kim ·
Dinh Quoc Nguyen · Huong Thi Thuy Ngo 

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Abstract Lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), and nickel (Ni) are poisonous, widely distributed, persistent, and transferable to crops, posing potential health risks. This study aims to assess the potential health risks of those elements in rice collected from North-Central Vietnam: Thanh Hoa, Nghe An, and Ha Tinh provinces. Element analysis was performed on rice harvested in November 2020 by ICP-MS. The estimated daily intake (EDI), target hazard quotient (THQ), non-carcinogenic hazard index (HI), and target carcinogenic risk (TR) were

used to assess potential health risks for different population groups. The highest element levels (mg kg^{-1} dry weight) were observed for Cr (0.30 ± 0.11), As (0.17 ± 0.025) and for Pb (0.24 ± 0.013) in Thanh Hoa, and for Cd (0.088 ± 0.015) in Ha Tinh. Strong links were observed between geological formations, mining activities and Cr in rice (Thanh Hoa), or industrial activities and Ni accumulation in rice (Hung Nguyen and Ky Anh districts). Children had greater EDIs than adults, with As having a higher EDI than RfD. Rice THQs indicated a risk trend: Thanh Hoa > Ha Tinh > Nghe An, with As being a significant contributor to HIs. Cr and Cd were significant risk factors and HIs in female children were 1.5 times higher than in other groups. Based on TR values for Ni and Pb, a potential carcinogenic risk to rice eaters was observed, particularly Ni. The data revealed a significant human health risk (both non-carcinogenic and carcinogenic) connected with rice consumption. Therefore, crops and foods from North-Central Vietnam should be strictly regulated.

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T. T. Le · H. T. T. Ngo (✉)
Faculty of Biotechnology, Chemistry and Environmental Engineering, Phenikaa University, Hanoi 12116, Vietnam
e-mail: huong.ngothithuy@phenikaa-uni.edu.vn

T. T. Le · H. T. T. Ngo
Bioresource Center, Phenikaa University, Hanoi 12116, Vietnam

K.-W. Kim
School of Earth Sciences and Environmental Engineering,
Gwangju Institute of Science and Technology (GIST),
123 Cheomdan-Gwagi-ro, Buk-Gu, Gwangju 61005,
South Korea

D. Q. Nguyen
Economic Geology and Geomatics Department, Vietnam
Institute of Geosciences and Mineral Resources,
Hanoi 12109, Vietnam

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Introduction

Lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), and nickel (Ni) are all trace elements found in nature. Unfortunately, the increased mining and

industrial processing caused by humans has made these trace elements a worldwide environmental threat (Fan et al., 2017; Zhuang et al., 2009). Contamination of soil by trace elements has been one of the most challenging pollution concerns because of the elements' high toxicity, widespread distribution, persistence, and transferability to plants, which can lead to different diseases (Huang et al., 2007; Qu et al., 2015; Rodriguez et al., 2007). While some trace elements in the soil are essential for plant and crop growth, others are hazardous to humans. Plants can absorb trace elements from the soil and store them in their tissues, eventually making their way into the human body via the food chain (Qu et al., 2015; Xiao et al., 2017).

As the economy has developed rapidly, many trace elements from industrial or mining activities have seeped into crop tissues via contaminated soils, wastewater irrigation, and polluted air (Huang et al., 2007; Sridhara Chary et al., 2008). Numerous metal-liferous mines have been set up across Vietnam due to the country's rich mineral deposits. These deposits have yielded a wide variety of metals, including antimony, chromite, copper, tin, and tungsten. Kien et al., (2010) found that lower stream and farmland areas were contaminated with trace elements despite the fact that mining and industrial zones were planned to prevent the release of tailings, wastewater, and solid waste. Excessive concentrations of Cd, Pb, and As were found in soil and water samples from the provinces of Thai Nguyen and Bac Kan, where Pb–Zn mining had taken place (Bui et al., 2016; Nguyen et al., 2011). The provinces of Thanh Hoa, Nghe An, and Ha Tinh, located in North-Central Vietnam, have a diverse range of mineral resources, mining activities, and industrial zones. Multiple mining industries, including those for chromite, tungsten, quart-gold, industrial minerals, graphite, and construction materials, existed in Thanh Hoa, and bauxite was mined in Nghe An, as stated by Khoi (Khoi, 2014).

Trace element contamination in rice has attracted the attention of scientists all over the world because it is a staple food in many countries, especially in Asia (Shimbo et al., 2001; Zarcinas et al., 2004). It is well-known that exposure to As, Pb, Cd, Cr, and Ni at a certain level can have carcinogenic and non-carcinogenic consequences. Several types of cancer have been linked to As poisoning: skin cancer, lung cancer, kidney cancer, and bladder cancer

(Bui et al., 2016; Fan et al., 2017); lung cancer, prostate cancer, and kidney cancer from exposure to Cd and Pb (Bui et al., 2020; Chen et al., 2019). Therefore, numerous international consortiums have been formed, and efforts have been made to establish various standards and limits for trace elements in food in order to protect human health from intoxication and poisoning resulting from food consumption. One of these is the Codex Alimentarius Commission (CAC, 2019), the Joint FAO/WHO Expert Committee on Contaminants in Food; this committee established the maximum limits of trace elements in rice grains for human consumption, with As and Pb at 0.2, Cd at 0.04, Cr at 1.3 and Ni at 10 mgkg⁻¹, etc. (CAC, 2019). Although soil contamination with trace elements has been previously reported, the assessment of human exposure to these elements through rice consumption in Vietnam, in particular, and in Southeast Asia, in general, has only recently been considered (Zulkafflee et al., 2022; Chu et al., 2021; Hengsawang et al., 2020). However, the health risk associated with these metals through rice consumption in the local populations has not been addressed systematically. Hence, the question has emerged as to whether the rice farmed in the North-central region of Vietnam is safe for consumption. In order to answer this question, this study aims to investigate (1) bioaccumulation of trace elements (Pb, Cd, As, Ni, and Cr) in rice grown in the North-Central provinces of Vietnam; (2) the potential health risk of consuming rice for local residents, using potential non-carcinogenic (HQ, HI) and carcinogenic risk (TR) indices; and (3) a possible link between mining/industrial activities and trace element levels in rice. These five trace elements were chosen for research due to their high toxicity and relevance to the region's mining and industrial operations.

Furthermore, to the best of our knowledge, this is the first study to assess the dietary exposure and potential health hazards associated with these five trace elements in the North-Central Vietnam population through rice intake. The findings help to build a database of potential health concerns associated with trace elements in rice in Vietnam, offering essential information to governments, policymakers, and researchers. It also assists in controlling and minimizing the hazards connected with consuming rice contaminated with trace elements for populations.

Materials and methods

Sampling sites

Ten districts in three provinces, Thanh Hoa, Ha Tinh, and Nghe An, were selected as sampling locations (Fig. 1, Table S1). In each district, five rice samples were randomly collected.

- (1) Thanh Hoa province: samples were collected from three districts: (a) in Nghi Son district, samples were taken in Tinh Hai commune, just adjacent to the Nghi Son refinery and petrochemical LLC, and Nghi Son iron and steel complex, which were put into operation in 2018 and 2019, respectively; (b) in Quang Xuong districts, samples were collected from Quang Giao commune, which is close to the ilmenite mine and located in the catchment area of the active and largest chromite mine (Co Dinh, 18.6 million tons) in Vietnam; (c) in Bim Son district, samples were collected from Dong Son, which is close to the Bim Son A&B industrial zones.
- (2) Nghe An province: the sampling locations were along with the Lam River upstream: (a) in Hung Nguyen district, Hung Tay commune was chosen for sampling, which is about 7–10 km away from the industrial zones of Bac Vinh and Nam Cam; whereas (b) Thanh Chuong and (c) Nghia Dan districts are located in the highlands and far from industrial zones, as well as a plain district (d) Do Luong, were also included for rice sampling.
- (3) Ha Tinh province: the sampling sites in (a) Nghi Xuan district, samples were taken in Xuan Hai, Dan Truong, and Xuan Hoi communes, at the Lam River mouth; while samples in (b) Ky Anh district were collected right adjacent to the Vung Ang industrial park-seaport complex; and (c) in Duc Tho district, samples were collected from Tung Anh and Bui La Nhan communes, and Duc Tho town, close to the La River and quite distant from the new industrial zone of the district.

Sampling and sample analysis

The rice grown at the studied locations, which accounted for regional consumption, was harvested in November 2020. All samples were removed husks

and polished by the rice milling machine before sampling from the household and transferring to the laboratory for further processing. Rice samples were dried in a vacuum oven at 70 °C for at least 72 h. Afterward, the samples were reweighed, pulverized by the grinder, and stored in food-grade zip bags for further preparation and analysis.

The samples were prepared following Phan et al., (2013) with some modifications. Shortly, an aliquot of 0.5 g of each ground rice sample was digested with 4 ml of HNO₃ (70%, Electronic Grade, Duksan, Korea) using a microwave digestion system (Multiwave 7000, Anton Paar GmbH, Graz, Austria). The digestion program for high organic compounds was listed as follows: (1) stage 1: 250 °C for 20 min, (2) stage 2: 250 °C for 15 min. After that, the digested samples were filtered and diluted with distilled water (> 18 MΩ cm⁻¹). The trace elements in rice samples were analyzed using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7900, USA).

Quality assurance and quality control

Appropriate quality assurance procedures and precautions were carried out to ensure the reliability of the results. The glassware had been soaked with 5% HNO₃ for 24 h and thoroughly rinsed with double-distilled water before use. The reagents used for analytical work were of analytical grade. An internal standard of ICP multi-element standard solution (supplied by Merck, Germany) and an analytical blank were used for each batch of 20 samples to ensure the quality assurance and precision of the elemental analysis. Each sample was analyzed in triplicate, with the recovery rates ranging from 84.6 to 101.3%, with average recoveries of 91.3, 84.6, 87.3, 101.3, and 94.5% for As, Cd, Cr, Pb, and Ni of internal standards, respectively.

Assessment of potential human health risk

Estimated daily intakes (EDIs) of trace elements

The EDIs of trace elements (Cr, Ni, As, Cd, and Pb) varied depending on rice's metal concentration and the amount consumed. The following equation was used to calculate the EDIs of trace elements for the locals:

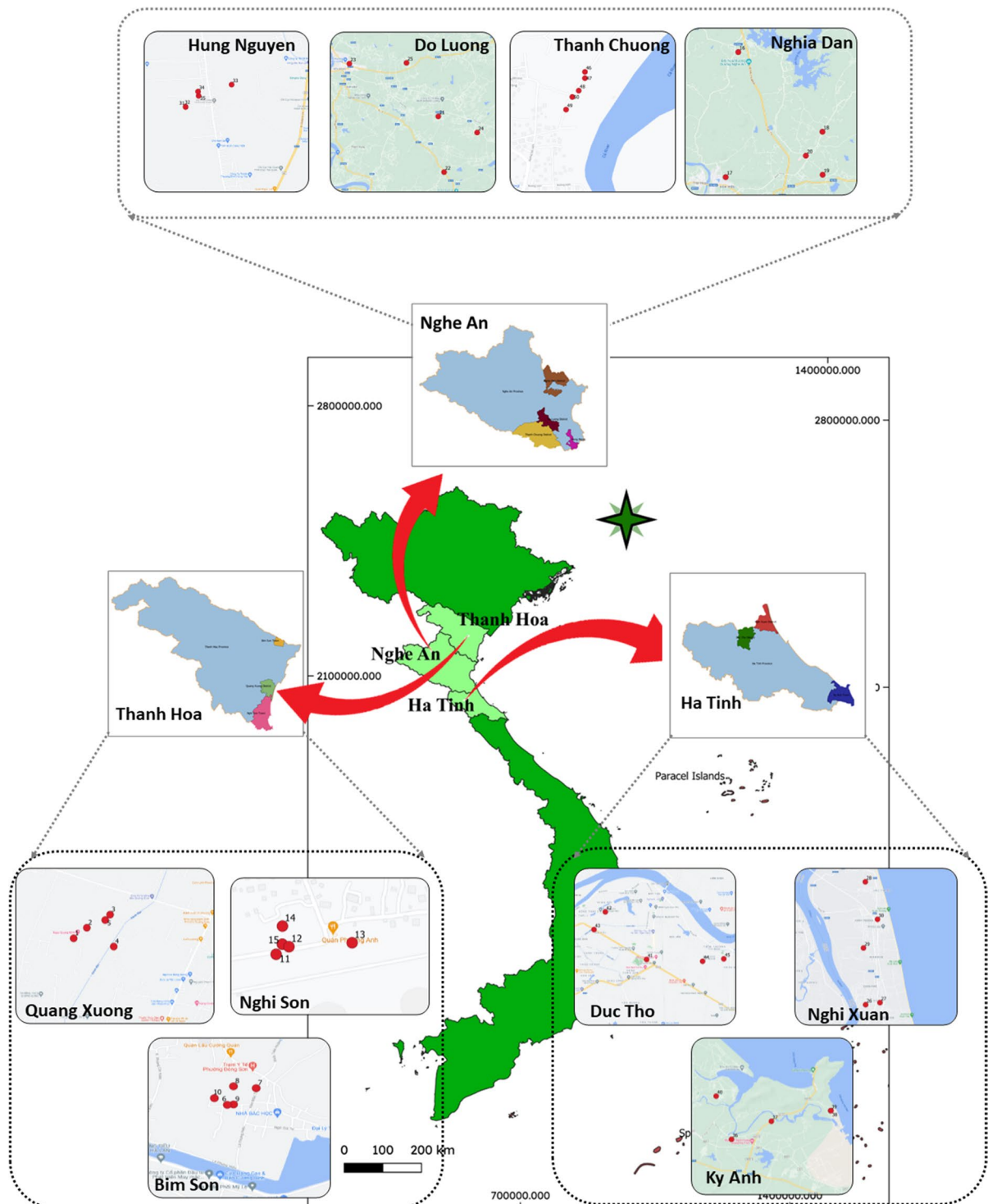


Fig. 1 Sampling sites in Thanh Hoa, Ha Tinh and Nghe An provinces. (1) Industrial zones: Bim Son, Nghi Son, Ky Anh and Hung Nguyen; (2) Mining region: Quang Xuong, Bim Son

and Nghi Xuan; and (3) Low industry activity region: Duc Tho and Do Luong; (4) Regions without mining and low industrial activities: Thanh Chuong and Nghia Dan

$$EDI = \frac{M_c \times IR}{B_w} \quad (1)$$

where M_c ($\mu\text{g g}^{-1}$) is the concentration of trace elements in contaminated rice; IR represents the average daily consumption of rice; B_w is the body weight. Based on the data survey from FAOSTAT (FAO, 2021), the adults in Vietnam had an average consumption per person (59.4 kg and 50.3 kg in body weights of males and females, respectively) of 325.5 g day^{-1} (Nutrition, 2012). In contrast, the investigated one in children under 5 years old (16.4 kg and 15.2 kg in body weights of male and female children, respectively) was 205 g day^{-1} (Nguyen et al., 2021).

Target hazard quotients (THQ)

The health risks associated with rice consumption by local people were assessed using the THQ. If the ratio is less than one, the exposed population is unlikely to experience apparent adverse health effects. In contrast, rice consumers may face a potential health risk if the ratio is higher than one. The method for estimating potential health risk with THQ was described in Regional Screening Levels (RSLs) – Equations (US EPA, 2021) and Chien's study (Chien et al., 2002) and is based on the equation as follows:

$$THQ = \frac{M_c \times IR \times 10^{-3} \times Ef \times Ed}{RfD \times B_w \times ATn} \quad (2)$$

where THQ is the target hazard quotient; Ef is exposure frequency ($365 \text{ days year}^{-1}$); Ed is total exposure duration (70 years); IR is the daily ingestion rate for locals ($\text{g person}^{-1} \text{ d}^{-1}$); M_c is metal concentration in food ($\mu\text{g g}^{-1}$); RfD is the oral reference dose ($\text{mg kg}^{-1} \text{ d}^{-1}$); B_w is the average body weight; Atn is the average time for non-carcinogens ($365 \text{ days year}^{-1} \times \text{number of exposure years}$, assuming 70 years in this study). Oral reference doses were based on 3×10^{-4} , 10^{-3} , 1.1×10^{-2} , and $3 \times 10^{-3} \text{ mg kg}^{-1} \text{ d}^{-1}$ for As, Cd, Ni, and Cr, respectively (US EPA, 2014).

There is no RfD for Pb, so the THQ for Pb was calculated as follows:

$$THQ = \frac{M_c}{MRL} \quad (3)$$

where the MRL is the maximum limit of Pb in rice grains acquired by the Codex Alimentarius Commission Standard (CAC, 2019).

Hazard index (HI)

The HI is the sum of the individual target hazard quotients for the tested elements from rice. According to the HI, eating rice may simultaneously expose to several potentially toxic elements. Hence, the cumulative effects of multiple elements from rice consumption may result in adverse health effects, specifically when HI exceeds one. The HI equation is as follows:

$$HI = \sum_{n=1}^i THQ_n \quad (4)$$

Target cancer risks

The target cancer risk (TR) is used to assess the potential risk of lifetime exposure to a carcinogenic agent. An oral slope factor is used instead of an oral reference dose to determine THQ. This factor, along with the carcinogen dose, determines the likelihood of cancer risk over the lifetime of the exposed individual. TR can be calculated using the following formula:

$$TR = \frac{Ef \times Ed \times E \times M_c \times CPS_0}{BW_a \times AT_c} \times 10^{-3} \quad (5)$$

where: Ef is the exposure frequency; Ed is the exposure duration (70 yrs); IR is the food ingestion rate in grams per day for the rice; M_c is concentration of trace element in the given rice ($\mu\text{g g}^{-1}$); CPS₀ is the oral carcinogenic slope factor for Pb, As, Ni and Cr of 8.5×10^{-3} , 1.5, 0.91, and $0.5 \text{ mg kg}^{-1} \text{ day}^{-1}$, respectively; B_w is the reference body weight; Atc is the averaged exposure time to the carcinogen ($365 \text{ days} \times 70 \text{ yrs}$) and 10^{-3} is the unit conversion factor.

Data processing and analysis

Microsoft Excel was used to process and analyze all the data collected. ANOVA was used to detect differences in metal concentrations in rice across sampling sites. The Tukey–Kramer test was used if statistically

significant differences ($p < 0.05$) were discovered (GraphPad InStat, San Diego, CA, USA).

Results and discussion

Bioaccumulation of trace elements in rice grown in study sites

The average trace element concentrations in rice samples were highest for Ni, followed by Cr, As, then Cd, and lowest for Pb (Table 1; Fig. 2). Thanh Hoa had the highest levels of trace element contamination in rice, particularly Cr, As, and Pb, followed by Ha Tinh and Nghe An (Table 1). High concentrations of Ni (mg kg^{-1} dw) were found in the Hung Nguyen (0.36 ± 0.092) district (Nghe An province), the Ky Anh (0.35 ± 0.071) and Nghi Xuan (0.23 ± 0.065) districts (Ha Tinh province), and in the Bim Son (0.29 ± 0.045) and Quang Xuong (0.27 ± 0.071) districts (Thanh Hoa province) ($p < 0.05$). Thanh Hoa had the highest concentrations of Cr (mg kg^{-1} dw), with an average concentration of 0.30 ± 0.11 ($p < 0.05$), about ten times higher than the other provinces (Nghe An: 0.036 ± 0.004 ; and Ha Tinh: 0.029 ± 0.03 ; Table 1). Cr concentrations in the districts of Quang Xuong, Bim Son, and Nghi Son were 0.43 ± 0.17 , 0.39 ± 0.20 , and 0.091 ± 0.058 , respectively (Fig. 2,

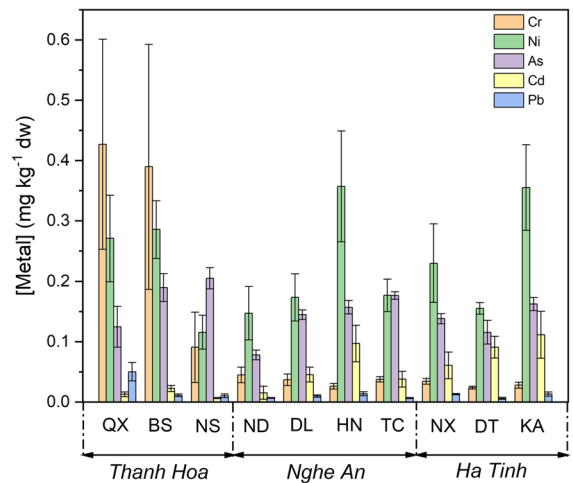


Fig. 2 Average concentrations of Cr, Ni, Cd, Pb, and As in rice (mg kg^{-1} dried weight -dw) collected from ten districts in three north-center provinces of Vietnam. QX: Quang Xuong, BS: Bim Son, NS: Nghi Son ND: Nghia Dan, DL: Đô Lương, HN: Hung Nguyen, TC: Thanh Chuong, NX: Nghi Xuan, DT: Duc Tho, KA: Ky Anh

Table 1). Even though these trace element concentrations are still lower than the WHO allowable limit (CAC, 2019), anthropogenic activities such as mining or industrialization may have a long-term impact on trace metal contamination in soils and rice. According to Kien (Kien et al., 2010), the Co Dinh chromite mine (Thanh Hoa) contaminated

Table 1 The concentrations of Cr, Ni, Cd, Pb, and total As in rice samples collected from the provinces of Thanh Hoa, Ha Tinh, and Nghe An. The values are expressed as the average \pm standard deviation

Provinces	Districts	Cr	Ni	Total As	Cd	Pb
Thanh Hoa	Quang Xuong	0.43 ± 0.17	0.27 ± 0.071	0.13 ± 0.034	0.013 ± 0.004	0.05 ± 0.02
	Bim Son	0.39 ± 0.20	0.29 ± 0.048	0.19 ± 0.023	0.022 ± 0.005	0.011 ± 0.002
	Nghi Son	0.091 ± 0.058	0.12 ± 0.028	0.21 ± 0.018	0.007 ± 0.001	0.010 ± 0.003
Average		0.30 ± 0.11	0.22 ± 0.054	0.17 ± 0.025	0.014 ± 0.005	0.024 ± 0.013
Ha Tinh	Nghi Xuan	0.034 ± 0.005	0.23 ± 0.065	0.14 ± 0.008	0.061 ± 0.022	0.013 ± 0.001
	Duc Tho	0.024 ± 0.002	0.16 ± 0.010	0.12 ± 0.020	0.091 ± 0.018	0.006 ± 0.002
	Ky Anh	0.028 ± 0.005	0.36 ± 0.071	0.16 ± 0.011	0.11 ± 0.039	0.013 ± 0.003
Average		0.029 ± 0.003	0.25 ± 0.058	0.14 ± 0.014	0.088 ± 0.015	0.011 ± 0.002
Nghe An	Nghia Dan	0.045 ± 0.013	0.15 ± 0.044	0.078 ± 0.008	0.015 ± 0.011	0.007 ± 0.001
	Do Luong	0.037 ± 0.010	0.17 ± 0.039	0.15 ± 0.008	0.045 ± 0.012	0.010 ± 0.002
	Hung Nguyen	0.026 ± 0.004	0.36 ± 0.092	0.16 ± 0.011	0.097 ± 0.030	0.014 ± 0.003
	Thanh Chuong	0.038 ± 0.004	0.18 ± 0.027	0.18 ± 0.006	0.038 ± 0.013	0.007 ± 0.001
Average		0.036 ± 0.004	0.21 ± 0.048	0.14 ± 0.021	0.049 ± 0.017	0.009 ± 0.002
WHO permissible limits (CAC, 2019)		1.30	10	0.2	0.04	0.2

lowland paddy fields with Cr, Ni, and Co, posing significant health risks through agricultural products. He also stated that water contaminated with Cr and Ni in the mining area could pollute nearby rivers. Cr and Ni levels were found to be highly correlated with industrial and mining activities in the studied areas. In Thanh Hoa, in particular, ultramafic outcrops are rarely exposed and severely weathered; as a result, trace elements are diluted and sedimented in the Ma River delta plain, where Quang Xuong and Bim Son are situated. In addition, the distances between these two districts and the chromite mine (Co Dinh) are approximately 16 km and 50 km, respectively, and they are located in the same catchment.

The previous study (Hang et al., 2009) discovered Cr in rice at relatively high concentrations of 0.22 ± 0.14 (in Changshu) and 0.107 (Taizhou). According to the findings, industrial activities may have influenced Cr concentrations in Changshu's rice. In the current study, industrial activities appear to be a minor factor contributing to the Cr concentration in rice. However, there is a strong correlation between industrial activities and Ni accumulation in rice grown in the region. Specifically, Ni levels in samples from Hung Nguyen district (0.36 ± 0.092), Nghe An province (about 12–15 km to the industrial zones of Bac Vinh and Nam Cam), or Ky Anh district (0.36 ± 0.071), Ha Tinh province (next to the industrial zones of Vung Ang), and Bim Son district (0.29 ± 0.048), Thanh Hoa province (less than 10 km from Bim Son industrial zone) were about double that of other districts ($p < 0.05$; Table 1). This finding is consistent with the findings of Proshad et al., (2019) in Tangail, Bangladesh, where untreated nickel-containing wastewater from industries correlated well with very high levels of this metal in rice.

The average concentrations of As (mg kg^{-1} dw) in rice samples collected from Thanh Hoa, Ha Tinh, and Nghe An were 0.17 ± 0.025 , 0.14 ± 0.014 , 0.14 ± 0.021 , respectively. There have been several studies that have investigated the As contents in rice. Our findings were comparable to those reported in other studies conducted in the Red River Basin (from 0.053 to 0.47 mg kg^{-1} dw) (Tran et al., 2020) and near the mining sites (0.052 mg kg^{-1} – 0.33 mg kg^{-1} dw) (Chu et al., 2021). However, the average As concentration in the current study was less than 0.2 mg kg^{-1} , less than the permissible level from the FAO-WHO

Codex Alimentarius Commission (CAC, 2019), with only samples from Nghi Son were exceeding the FAO-WHO standard (Table 1).

According to Tran's study (Tran et al., 2020), grain As content increases gradually from mountainous and hilly regions to lowland (coastal) regions, but soil As content does not. In other words, the high As concentration in rice could be attributed to factors other than natural sources, such as anthropogenic activities, and depends on bioavailable forms of this element. Furthermore, mining activity may play a role in rice's high As content (Chu et al., 2021). Irrigating rice field with arsenic-contaminated water raises the concentration of As in the topsoil and its bioavailability to rice crops (Azam et al., 2016). It has been reported that As accumulation in rice grains increases due to As high bioavailability and mobility in paddy rice (Xu et al., 2008). In this study, the lowest As content was found in samples from Nghia Dan, which is located in the highlands and far from industrial zones, whereas high As levels were found in samples from Bim Son (near the Bim Son industrial zone), Nghi Son (close to the Nghi Son refinery and petrochemical LLC, and Nghi Son iron and steel complex), and Ky Anh (right next to the Vung Ang industrial park-seaport complex). These high As levels could be attributed to industrial activities in these areas. It is worth noting that the Nghi Son industrial zones have only recently begun operations; however, they have already negatively impacted the surrounding environment and, as a result, agricultural products. Therefore, appropriate monitoring and regulation should be maintained to prevent the release of toxic substances into the surrounding environment and potential adverse impacts on human health.

The highest Cd levels (mg kg^{-1} dw) were found in Ha Tinh (0.088 ± 0.015), followed by Nghe An (0.049 ± 0.017) and Thanh Hoa (0.014 ± 0.005) (Table 1, Fig. 2). Specifically, 60% and 40% of the rice samples collected from Ha Tinh and Nghe An, respectively, contained Cd levels above CODEX permissible limits (CAC, 2019); this appeared in districts with high industrial activity, namely Ky Anh, Duc Tho and Hung Nguyen. In comparison to the current findings, some studies have found lower Cd concentrations in rice, e.g., in the Red River basin (0.033 mg kg^{-1} dw) (Bui et al., 2016) and Changshu city (0.019 mg kg^{-1} dw) (Hang et al., 2009). However, others reported that the Cd concentrations

in rice grown near mining sites in Hunan province ($0.103 \text{ mg kg}^{-1} \text{ dw}$) (Fan et al., 2017), the Yang Zhong district ($0.224 \text{ mg kg}^{-1} \text{ dw}$) (Hang et al., 2009), and the Jin-Qu basin ($0.163 \text{ mg kg}^{-1} \text{ dw}$) (Guo et al., 2020) were higher than those in the present data.

Pb concentrations ($\text{mg kg}^{-1} \text{ dw}$) in the rice samples ranged from 0.006 ± 0.002 (Duc Tho) to 0.050 ± 0.015 (Quang Xuong). The average Pb content in Thanh Hoa was more than double that of Ha Tinh and Nghe An ($p < 0.05$; Table 1). In comparison to previous studies on the Red River Delta ($0.075 \text{ mg kg}^{-1} \text{ dw}$) (Chu et al., 2021), the Yangtze River Delta ($0.957 \text{ mg kg}^{-1} \text{ dw}$) (Hang et al., 2009), and the Jin-Qu Basin ($0.148 \text{ mg kg}^{-1} \text{ dw}$) (Guo et al., 2020), the current results revealed significantly lower Pb levels.

Mining and industrial activities could be the sources of Pb and Cd in rice grain. Another report (Hang et al., 2009) found that the erosion of these metals from e-waste recycling activities could be discharged into the surrounding environment, indicating contamination by these elements in the studied areas. Guo et al., (2020) also reported that Cd contamination in rice grains was higher in the Jin-Qu Basin than in other cultivation areas, which nonferrous metal plants could influence. This similar condition could explain the high Cd content in rice from Ha Tinh province. Ky Anh district (Ha Tinh province) is home to the Vung Ang industrial zone, which includes a thermal power plant and a steel factory, and the industrial activities in that area may result in high Cd levels in rice. On the other hand, Pb could be introduced into rice grains through atmospheric deposition during metal-mining activities via dust and particulate material deposition, as observed in the Quang Xuong district (Thanh Hoa province). Other studies have found that Cd and Pb quickly transfer from soils, particularly in mining areas, and accumulate in vegetables and rice (Bui et al., 2016; Mao et al., 2019; Zhuang et al., 2009).

Daily intake of trace elements via rice consumption and the potential risk to human health

The non-carcinogenic and carcinogenic health risk assessments of trace elements can be calculated and evaluated using the method of health risk assessment provided by the US EPA (US EPA, 2021).

Estimated dietary intake of trace elements

Although there are numerous pathways by which humans can be exposed to trace elements, rice consumption has been identified as one of the major routes. This means as rice consumption increases in Vietnam, so will human exposure to these contaminants. The dietary intake of trace elements from rice consumption for adults (both male and female) and children (male and female children under the age of five) was calculated (Table 2), assuming that the local population primarily consumes local rice.

In general, EDI trends for trace elements in rice were in the order of $\text{Cr} > \text{Ni} > \text{As} > \text{Cd} > \text{Pb}$. The highest EDIs were found in Thanh Hoa for Cr ($4.1 \pm 1.43 \text{ } \mu\text{g d}^{-1}$), As ($2.3 \pm 0.33 \text{ } \mu\text{g d}^{-1}$) and Pb ($0.32 \pm 0.18 \text{ } \mu\text{g d}^{-1}$), and in Ha Tinh for Ni ($3.3 \pm 0.79 \text{ } \mu\text{g d}^{-1}$) and Cd ($1.2 \pm 0.20 \text{ } \mu\text{g d}^{-1}$). Among the investigated population groups, EDIs of all five trace elements from the children's groups were significantly higher than in adult groups ($p < 0.05$) and showed higher risk ($p < 0.05$) in all investigated sites and with all studied elements (Table 2). Data also revealed that the residents faced the potential risks with As due to higher EDI value than RfD (Table 2). The same observation was made in Northern Thailand reported by Chanpiwat et al. (2019). This could be because people in Vietnam and Thailand consume more rice than in other Asian countries. Because of the bodyweight effect, males (59.4 kg) with rice consumption of 325 g day^{-1} had the lowest EDI values. The EDIs for Cr, As, and Cd from rice consumption, both adults and children, in this study are higher than those in the Yangtze River Delta, China (Hang et al., 2009; Mao et al., 2019) but lower than those in the mining areas of the Jin-Qu Basin (Guo et al., 2020) and Dabaoshan mine (Zhuang et al., 2009). In addition, the present data revealed that the average EDI values for As, Cr, Ni, and Pb in the adult group from three studied provinces were higher than those found in the Red River Delta (Chu et al., 2021).

Rice consumption on a daily base in Vietnam was comparable to that of other Asian countries. In China, daily rice consumption ranges from 238 to $389 \text{ g person}^{-1} \text{ day}^{-1}$ (Guo et al., 2020; Hang et al., 2009; Mao et al., 2019) for adults, with children consuming $198 \text{ g person}^{-1} \text{ day}^{-1}$ (Hang et al., 2009; Mao et al., 2019), whereas Vietnamese adults and children under 5 years old consume 325

Table 2 Estimated daily intakes (EDI) of trace elements from rice ($\mu\text{g d}^{-1}$) for adults and children (under 5 years old) from three locations: Thanh Hoa, Ha Tinh, and Nghe An provinces. The values are expressed as the average \pm standard deviation

Elements/Locations		Thanh Hoa	Ha Tinh	Nghe An	RfD ($\mu\text{g d}^{-1}$)
Cr	Male	1.7 ± 0.58	0.16 ± 0.03	0.20 ± 0.02	3
	Female	2.0 ± 0.67	0.18 ± 0.03	0.24 ± 0.02	
	Male children	3.8 ± 1.33	0.34 ± 0.05	0.45 ± 0.05	
	Female children	4.1 ± 1.43	0.37 ± 0.06	0.49 ± 0.05	
Ni	Male	1.2 ± 0.30	1.4 ± 0.32	1.2 ± 0.26	11
	Female	1.5 ± 0.35	1.6 ± 0.38	1.3 ± 0.33	
	Male children	2.8 ± 0.68	3.1 ± 0.73	2.7 ± 0.60	
	Female children	2.9 ± 0.84	3.3 ± 0.79	2.9 ± 0.65	
As	Male	0.95 ± 0.14	0.76 ± 0.07	0.76 ± 0.12	0.3
	Female	1.1 ± 0.16	0.90 ± 0.09	0.87 ± 0.14	
	Male children	2.2 ± 0.31	1.7 ± 0.17	1.7 ± 0.27	
	Female children	2.3 ± 0.33	1.9 ± 0.18	1.9 ± 0.29	
Cd	Male	0.08 ± 0.03	0.48 ± 0.08	0.27 ± 0.10	1
	Female	0.09 ± 0.03	0.57 ± 0.10	0.31 ± 0.11	
	Male children	0.17 ± 0.06	1.1 ± 0.18	0.61 ± 0.22	
	Female children	0.19 ± 0.06	1.2 ± 0.20	0.66 ± 0.23	
Pb	Male	0.13 ± 0.07	0.06 ± 0.01	0.05 ± 0.01	0.2 (MRL)
	Female	0.15 ± 0.09	0.07 ± 0.01	0.06 ± 0.01	
	Male children	0.30 ± 0.16	0.13 ± 0.03	0.11 ± 0.02	
	Female children	0.32 ± 0.18	0.14 ± 0.03	0.12 ± 0.02	

and $205 \text{ g person}^{-1} \text{ day}^{-1}$, respectively. Rice consumption in Vietnam (325 g day^{-1}) is higher than in some parts of China, Japan, Taiwan, and Thailand ($238, 119, 132$, and 272.6 g day^{-1} , respectively). It can be explained that the Vietnamese primarily consume rice as a carbohydrate source, whereas other Asian countries consume wheat, buckwheat, and other grains.

Furthermore, because Cr and Ni are essential micronutrients for human health, moderate levels of Cr and Ni found in rice samples may pose a significant health risk (Uriu-Adams & Keen, 2005). Many studies have found that people are exposed to metals through other foods such as wheat, vegetables, fruit, fish, meat, eggs, water, and milk (Antoine et al., 2017; Bui et al., 2016; Ngo et al., 2021; Zheng et al., 2007). However, because Asians consume much rice daily, rice is a significant pathway for local people's dietary exposure to toxic elements through food. As a result, residents must reduce their rice consumption and diversify their diets to reduce the health risks associated with dietary trace elements intake.

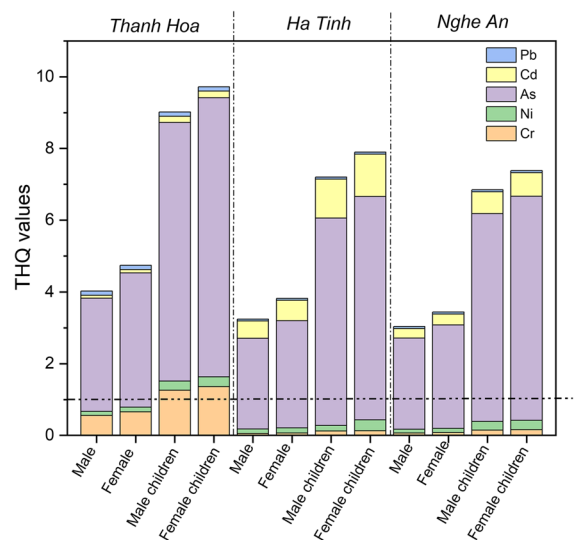


Fig. 3 The spatial target hazard quotients (THQs) of five trace elements derived from rice consumption for adults and children (under 5-year-old)

Potential health risk of individual trace elements

Although adults consume more rice than children, the THQ values of trace elements for adults were lower than those for children (Fig. 3). Consistent with the EDI results, THQ values for As were the highest, exceeding one, implying that people were exposed to health risks from consuming As contaminated rice. Furthermore, the high THQs indicating that Cd and Cr in Thanh Ha and Ha Tinh, respectively, could pose health risks to rice consumers. In contrast, THQs for Ni and Pb were less than 1 at all sites, indicating that there was no potential health risk to consumers. For adults, the THQs from rice consumption an all groups are in the following order $As > Cd > Cr > Ni > Pb$, except in Thanh Hoa, where THQ for Cr is higher than 1. Among the three provinces, Thanh Hoa had the highest THQs for studied elements, followed by Ha Tinh and Nghe An (Fig. 3). Thanh Hoa also has significantly higher THQ for Cr than other provinces, exceeding 1 for both male and female children groups, indicating a potential health risk for rice consumers. This result indicated that residents of Thanh Hoa suffered more adverse health effects from rice consumption than residents of other provinces. THQ values of As for adults in Thanh Hoa, Nghe An, and Ha Tinh were 3.1 ± 0.45 – 7.8 ± 1.1 , 2.5 ± 0.25 – 6.2 ± 0.68 , and 2.5 ± 0.39 – 6.3 ± 0.96 , respectively (Fig. 3).

In addition to rice consumption, which was the most significant dietary factor contributing to dietary trace metal exposure, other indirect factors such as age and body weights could be investigated. Previous studies (Nguyen et al., 2021; Chanpiwat et al., 2019) discovered that the As exposure dose from rice consumption varied across age groups and gender due to rice intake and body weight differences in Vietnam and Thailand. According to the authors, females consumed more As than males, and children consumed more As than adults per day due to a higher rice intake/body weight ratio. The current study's As-THQs for adults and children were significantly higher than those in the Yangtze River Delta, which were 0.028 and 2.3 (Hang et al., 2009), and 0.024 and 2.7 (Mao et al., 2019), respectively. The disparity in THQ values could be attributed to THQ's effective parameters and their variations across geographic regions. Furthermore, based on EDI and THQ results, children under five in Thanh Hoa and Ha Tinh may face greater health risks due to Cr and

Cd, respectively. In fact, this risk assessment was based not only on trace element levels in rice, but also on rice consumption rates.

Potential health risks of combined trace elements and Target cancer risks

The HI was calculated by considering the cumulative effect of consuming five potentially hazardous elements from rice (Fig. 4). The calculated HI values range from approximately 1.7 ± 0.64 for males in Nghia Dan (Nghe An) to the highest value of about 11 ± 4.7 for female children in Bim Son (Thanh Hoa). All calculated HI values surpassed 1, suggesting potential health risks for local people due to daily rice consumption (Fig. 4). The current HIs were significantly higher than those in previous studies (Antoine et al., 2017; Guo et al., 2020; Hang et al., 2009; Zheng et al., 2007), which could be attributed to differences in consumption rates and average body weight. The HI trends were as follows: Bim Son > Nghi Son > Ky Anh > Hung Nguyen ~ Thanh Chuong > Quang Xuong > Nghi Xuan ~ Do Luong > Duc Tho > Nghia Dan. The results showed that As is a critical component contributing to the potential health risk of non-carcinogenic effects in all groups, with Cr and Cd serving as secondary components.

In this study, only Ni and Pb were considered for assessing target cancer risk (TR) (Table 3, Fig. 5). TR values for Ni and Pb ranged from 1.1×10^{-3} to

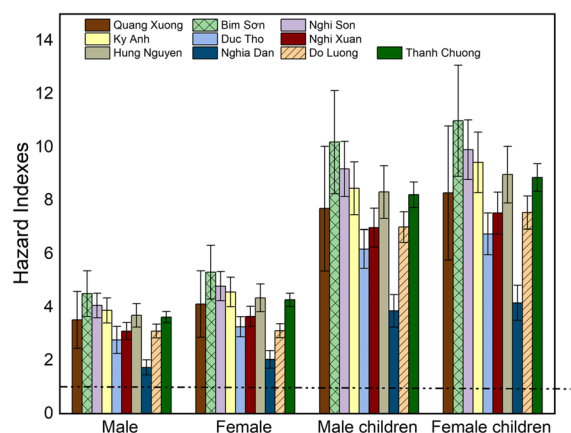


Fig. 4 The total hazard index (HI) of five elements posed to human health due to rice consumption in different population groups from Thanh Hoa, Nghe An and Ha Tinh provinces. $HI < 1$, acceptable risk; $HI > 1$, potential risk

Table 3 Target cancer risk (TR) of trace elements from rice consumption in adults and children (under 5 years old) from three studied provinces: Thanh Hoa, Ha Tinh, and Nghe An. $TR < 1 \times 10^{-6}$, insignificant risk; $1 \times 10^{-6} < TR < 1 \times 10^{-4}$, low or acceptable risk range; and $1 \times 10^{-3} < TR < 1 \times 10^{-1}$, high cancer risk levels

	Groups	Ni	Pb
Thanh Hoa	Male	1.1E-03	1.1E-06
	Female	1.3E-03	1.3E-06
	Male children	2.6E-03	2.5E-06
	Female children	2.8E-03	2.7E-06
Ha Tinh	Male	1.2E-03	5.0E-07
	Female	1.5E-03	5.9E-06
	Male children	2.8E-03	1.1E-06
	Female children	3.0E-03	1.2E-06
Nghe An	Male	1.1E-03	4.3E-07
	Female	1.2E-03	4.9E-07
	Male children	2.4E-03	9.8E-07
	Female children	2.6E-03	1.1E-06

3.0×10^{-3} and 4.3×10^{-7} to 5.9×10^{-6} , respectively, indicating a potential carcinogenic risk to rice consumers in most investigated areas, particularly Ha Tinh and Thanh Hoa (Table 3), with Ni showing the highest cancer risk ($TR > 1 \times 10^{-3}$). For example, in Ha Tinh, the risks come from Ni for adults and children under 5 years old were 1.45×10^{-3} (female) and 3.03×10^{-3} (female children), implying that 145 per 100,000 adults and 303 per 100,000 female children under 5 years old are at the highest risk of cancer caused by Ni intake from rice. Following publications that used 10^{-6} – 10^{-4} as the range for acceptable risk of developing cancer (Antoine et al., 2017; Shaheen et al., 2016), 10^{-4} was accepted as the upper limit. In this study, high cancer risk would be expected to be associated with Ni (Fig. 5; Table 3). The cancer risk calculated for Pb was within the low or acceptable range of 10^{-7} – 10^{-5} , indicating that the cancer risk from Pb-contaminated rice consumption in the three provinces studied was tolerable. However, the total cancer risk (TCR) analysis revealed the potential adverse cancer risk induced by Ni and Pb from rice consumption, as the TCR values were significantly higher than the threshold level ($> 10^{-6}$) at all sampling sites.

It should be noted that this study did consider special groups, such as women and children under the age of five, who are presumably more vulnerable to

pollutants that have both non-carcinogenic and carcinogenic effects. THQs of As for children under 5 years old, particularly female children, were 2–2.5 times higher than adults, implying that rice consumption may pose serious health risks depending on gender and age group. Furthermore, the average HI values for female children were 1.5 times higher than for other groups, indicating that the investigated trace elements had an apparent combined effect on the general health of rice consumers in these areas.

The results showed the current state of some trace elements accumulated in rice from North-Central Vietnam. However, as previously stated, because this area is experiencing rapid industrial development and has only recently begun operations, it has already had a negative impact on the environment. As a result, future research should include more elements of concern and concentrate on the effects of human activity on environmental matrices like soils and rice plants, as well as the transformation pathway of those impacts on human health risk. As a result, vegetables and foods from North-Central Vietnam should be strictly regulated for safety. Furthermore, rice is a significant source of toxic element exposure, so residents must reduce rice consumption and diversify their diets to reduce associated risks.

Conclusion

In summary, Thanh Hoa had the highest concentrations of Cr, As, and Pb in rice, followed by Ha Tinh and Nghe An. The results revealed a strong link between geological formations, mining, and industrial activities with trace element bioaccumulation in rice. Outcrops of ultramafic rock and mining activities correlated with Cr accumulation in rice from Quang Xuong and Bim Son districts (near the Co Dinh chromite mine); and industrial activities correlated with Ni accumulation in rice from Hung Nguyen district (about 12–15 km to the Bac Vinh and Nam Cam industrial zones) or Ky Anh district (next to the Vung Ang industrial zones (Table 1).

Even though these trace element concentrations are lower than the WHO allowable limit (CAC, 2019), mining or industrialization may have long-term effects on trace metal contamination in soils and rice. The calculated risk indices revealed that most of the investigated rice posed a significant human health

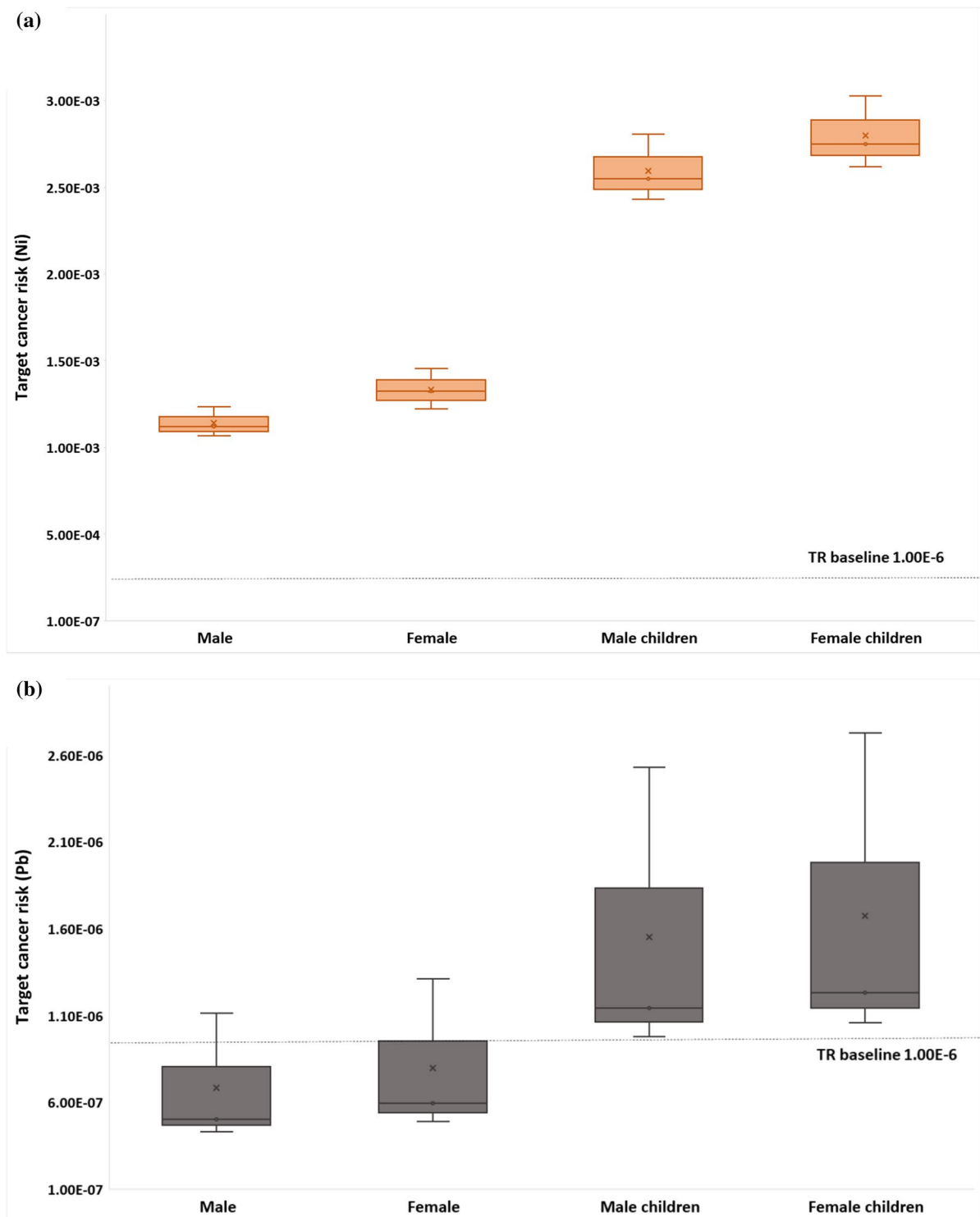


Fig. 5 The average values for the target cancer risk (TR) posed by **a** Ni and **b** Pb due to rice consumption. $TR < 1 \times 10^{-6}$, insignificant risk; $1 \times 10^{-6} < TR < 1 \times 10^{-4}$, low or acceptable risk range; and $1 \times 10^{-3} < TR < 1 \times 10^{-1}$, high cancer risk level

risk (both non-carcinogenic and carcinogenic). EDIs for all five trace elements were higher in the children group than in adult ones in all investigated sites (Table 2). Although EDI trends for trace elements in rice were $\text{Cr} > \text{Ni} > \text{As} > \text{Cd} > \text{Pb}$, data revealed that residents faced potential risks from As due to higher EDI than RfD (Table 2).

By using various health risk indices such as THQ (for As, Cr, and Cd), HI (for As, Cr, and Cd), and TCR (for Ni and Pb), it is shown that those trace elements were found to have potential impacts on all studied groups in three provinces. THQs were highest in Thanh Hoa, followed by Ha Tinh and Nghe An (Fig. 3), particularly As and Cr, which exceeded 1 for children's groups, indicating a health risk for rice consumers. The HI trends were as follows: Thanh Chuong > Quang Xuong > Nghi Xuan; Do Luong > Duc Tho > Nghia Dan; Bim Son > Nghi Son > Ky Anh > Hung Nguyen; As was the main contributor to non-carcinogenic health risks in all groups. TR values for Ni and Pb indicated a potential carcinogenic risk to rice consumers in the most studied areas (Table 3), with Ni showing the highest cancer risk. Total cancer risk (TCR) revealed that Ni and Pb in rice might pose a cancer risk at all sampling sites. Results from this study will help build a database of rice trace element contamination in North Central Vietnam, which will help with industrial effluent and food safety management.

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Author contributions TLT: Investigation, Methodology, Writing original draft—Review and Editing. KKW.: Resource, Validation. DNQ: Validation, Conceptualization. HNTT: Conceptualization, Methodology, Writing—Review and Editing, Supervision. All authors have read and agreed to the published version of the manuscript.

Data availability The datasets used in this study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors declare no conflict of interest.

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

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