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Research article

Heavy metals in commonly consumed rice grains in Bangladesh and associated probabilistic human health risks

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ABSTRACT

Food contamination by heavy metals is a concerning issue worldwide. The presence of elevated levels of heavy metals in commonly consumed rice has emerged as a critical issue in ensuring food safety. This research encompassed the collection of 44 rice samples, representing seven distinct varieties, through a randomized sampling approach across various retail markets within Dhaka city. The investigation of heavy metal content (including Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb) within the rice samples was carried out employing Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The findings disclosed substantial disparities in heavy metal concentrations among the different rice varieties. Notably, the concentrations of Cr (0.99 mg/kg), Fe (8.35 mg/kg), Pb (0.49 mg/kg), and Co (0.02 mg/kg) were observed to exceed the established maximum permissible limits. This contamination of the rice varieties may stem from either natural processes or human activities. Utilizing these metal concentrations, this study employed Monte-Carlo simulation to calculate health risk and probabilistic health risk. The Target Hazard Quotients (THQs) for Cr, Fe, and As exceeded the threshold of 1 in each rice variety, particularly in Amon and Lal Biroi in Mymensingh, Najirshail in Bogra, 28 in Sherpur and Kushtia, Miniket in Bogra and Rajshahi, and lastly, paijam in Rangpur and Tangail. Furthermore, the Hazard Index (HI) exceeded 1 in all rice varieties, implying that the consumption of these selected rice grains may have a substantial impact on overall food quality and its potential health consequences. Both carcinogenic risk and probabilistic risk for As, Ni, Cd, Cr, and Pb were found to surpass the threshold levels and safe limits, respectively. This suggests that individuals who have regularly consumed these rice varieties may face a heightened probability of developing cancer in the future, as predicted. According to sensitivity analysis, metal concentration and food ingestion rate (FIR) are the most relevant components that contribute to the significant impact of carcinogenic health hazards. Finally, the study concluded that heavy metal intake from food poses a risk to

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human health, necessitating the effective monitoring based on each geographic location and identify the source of this heavy metal to limit its exposure.

1. Introduction

Agricultural crops represent the primary and most substantial source of essential nutrients in our diet, encompassing proteins, carbohydrates, vitamins, dietary fibers, minerals, crucial metals, antioxidants, and other elements. These nutritional components also play a pivotal role in mitigating the adverse impact of harmful substances on the human body [1–3]. Despite their fundamental importance for sustaining life, food crops are susceptible to various forms of contamination [4]. This contamination can arise from multiple sources, including water, air, dust, insects, human contact, microbiological agents, chemical pollutants, and human activities such as the use of pesticides and industrial waste discharge [5]. Among the foods derived from plants that can acquire heavy metals via the earth's surface as well as the irrigation waters [6].

Bangladesh is a developing country where rice is the staple food. Approximately 80 % of the country's population consumes rice as their primary meal, often multiple times a day, providing them with vital nutrients [7,8]. Bangladesh boasts the cultivation of approximately 80 different rice varieties across three distinct seasons (Aus, Aman, and Boro), resulting in an annual production of 37.8 million metric tons of rice grains [9]. Remarkably, Bangladesh holds the world record for the highest per capita rice consumption, with individuals consuming an average of 470.79 g of rice per day [10]. Rice also constitutes a substantial portion, accounting for 66.7 % of the daily caloric intake of the Bangladeshi population [11]. According to the Rice Consumption worldwide statistics for the 2021/22 season, China leads as the world's largest rice producer, producing 154,890 thousand metric tons, while Bangladesh secures the fourth position with a production of 36,700 thousand metric tons [12]. In the past decade, rice consumption in Bangladesh has experienced a significant surge, with a notable proportion of consumers shifting from other staples to rice. However, even in contemporary times, rice remains vulnerable to metal contamination due to its efficient uptake mechanisms and the prevalence of heavy metals in the environment from both natural sources and human activities [13]. Heavy metals are particularly concerning due to their toxic nature, non-biodegradability, and persistence in the environment [14].

Food safety has emerged as a pressing global public health concern driven by the escalating prevalence of metal pollution in the environment [15]. The widespread contamination of heavy metals has extended its reach across the world, causing substantial disruptions to the environment and posing significant health hazards to human populations [5]. The spectrum of environmental pollutants has expanded exponentially since the onset of the Industrial Revolution and economic globalization, giving rise to an array of anthropogenic sources [5]. Moreover, food grains may also become polluted throughout the manufacturing, storage, and marketing processes [16].

Recent research findings have indicated a global increase in the levels of heavy metal content in rice [17]. Another study specifically assessed the presence of heavy metals in Taiwanese rice samples, revealing concentrations of 0.08 mg/kg for As, 0.01 mg/kg for Cd, 0.10 mg/kg for Cr, 2.22 mg/kg for Cu, 0.001 mg/kg for Hg, 0.29 mg/kg for Ni, and 0.01 mg/kg for Pb. Notably, one sample exhibited a significantly higher level of 14.7 mg/kg for Zn. To assess the associated dietary risks, the average weekly consumption of heavy metals from rice by the Taiwanese population was analyzed. Encouragingly, the study findings indicated that heavy metal intake from rice remained below acceptable weekly intake limits [18]. In China, there has been a steady increase in the levels of heavy metals found in agricultural soils. This trend not only hampers rice cultivation and growth, leading to production losses, but it also poses a significant threat to food safety. Heavy metal contamination in paddy soil has the potential to be absorbed and stored by rice plants, posing a risk to human health and jeopardizing China's food safety as these contaminants move up the food chain [19].

Dhaka, the capital of Bangladesh, is renowned for its dense population, extensive traffic congestion, and vibrant industrial activities. The city's bustling marketplaces serve as central hubs for the distribution of rice sourced from various regions across the country, catering to the needs of its millions of residents. Situated at the heart of these communities, these marketplaces expose food items to a multitude of environmental factors, including heavy metal contaminants originating from vehicular emissions and atmospheric deposition, in addition to potential chemical and microbial agents. Reports indicate that air deposition and automobile emissions can significantly elevate the levels of heavy metals present in food during the marketing process [16]. Consequently, it is reasonable to assume that food sold in open markets may exhibit elevated concentrations of heavy metals, giving rise to legitimate health concerns.

A study has drawn attention to the escalating threat to food safety posed by heavy metal contamination, particularly in developing nations such as Bangladesh [20]. Extensive research was carried out to assess the extent of metal contamination in both paddy field soil and subsoil, as well as in rice crops within the Barapukuria coalfield zone of Bangladesh. The study revealed that the mean levels As, Cr, Cu, Mn, Ni, Pb, and Zn exceeded global averages, with mining activities exerting a notable adverse impact on the surveyed soils, ranging from moderate to severe. Intriguingly, the absorption of As, Cd, and Pb by rice grains was approximately 6.87, 1.58, and 5.26 times higher than the maximum allowable concentration, respectively. This underscores the concerning tendency for these elements to migrate from contaminated ground into the rice crop [21].

Therefore, heavy metal pollution in food has become a major environmental and global public health problem, especially in Bangladesh [22]. Meanwhile, both manufacturers and consumers have recognized that the presence of heavy metals in meals is harmful to human health [22]. The World Health Organization (WHO) and the International Program on Chemical Safety (IPCS) have categorized heavy metals as chemicals of public concern [23]. These metals, owing to their enduring nature and resistance to degradation, persist in the ecosystem, where they accumulate in food, including plant and animal tissues. Subsequently, they traverse the food chain, posing a significant threat to human health [24]. Toxic metals, contaminating agricultural crops result in their accumulation in humans via food-chain interaction. Even at low levels over a lifetime, it may result in a variety of carcinogenic and non-carcinogenic health issues, including neurological, immunological, cardiovascular, nephrotoxicity, and reproductive abnormalities [25]. Toxic metals, according to a study by Liang et al., 2019 [26], disrupt innumerable biochemical processes in the human body, pose a significant health risk to humans, and ultimately lead to an increase in the prevalence of chronic diseases such as neurological disorders, central nervous system destruction, malformation, and cancer. As a result, the accumulation of heavy metals in the human body from these typical foods is currently a prominent concern.

The contamination of rice grains by toxic metals during the cultivation process, harvesting, transportation, storage, and food processing stages represents a critical concern for ensuring food quality and safety in Bangladesh. This study aimed to address the research question of whether commonly consumed rice varieties in Bangladesh are burdened with a high metal load that could potentially pose health risks to humans. To address this question, our study had three main objectives: first, to determine the levels of hazardous metals in rice grains frequently consumed in Bangladesh; second, to assess the associated health hazards; and third, to elucidate probabilistic carcinogenic health risks through a sensitivity analysis using the Monte-Carlo simulation method. This research investigated the prevalence of heavy metals in commonly consumed rice grains obtained from markets in Dhaka city and evaluated the potential health risks associated with their consumption. By shedding light on the impact of increased anthropogenic activities, this study aims to provide valuable insights for the development of effective control measures and regulatory limits for hazardous metals in various staple food products produced and marketed in Bangladesh.

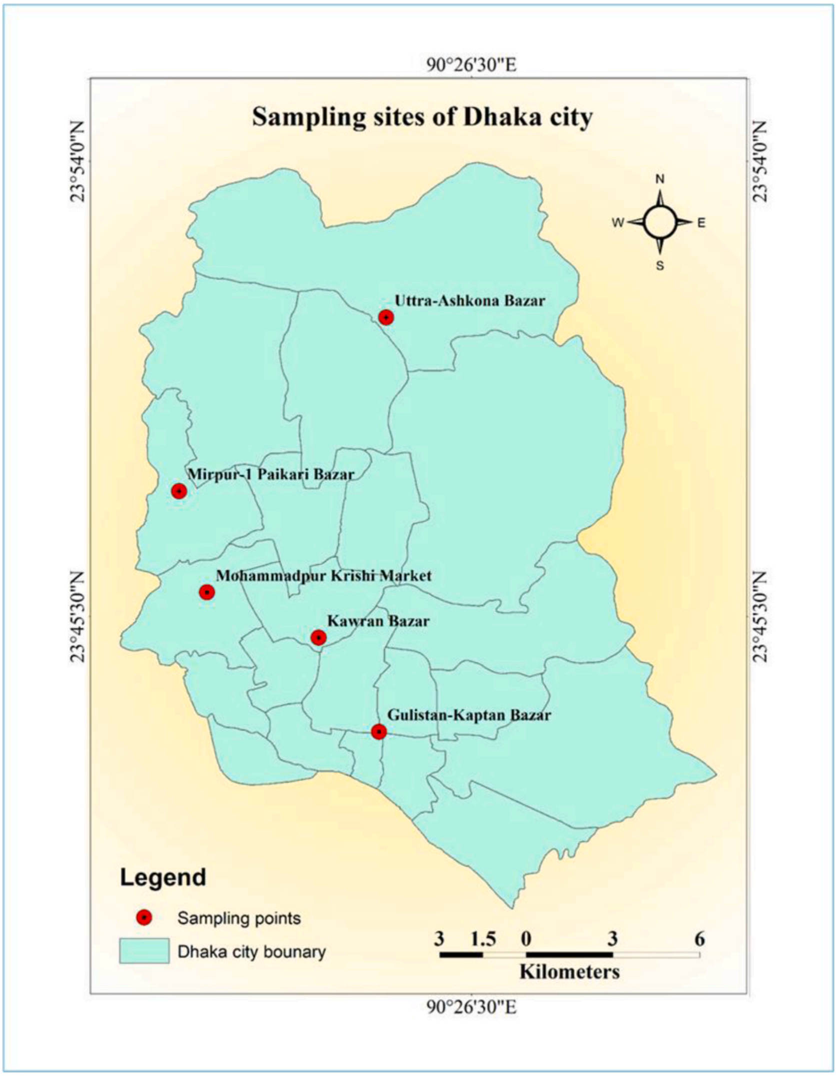

Fig. 1. Sample Collection locations in the study area of Dhaka City.

2. Materials & method

2.1. Study area and sample collection

In order to conduct this study, 44 rice samples, a total of 7 different varieties, were collected during the month of January of the year 2022. Five famous and most frequented big markets (Kawran Bazar, Mirpur Paikari Market, Mohammadpur Krishi Market, Gulistan-Kaptan Bazar, and Uttara Ashkona Bazar) were randomly selected in Dhaka city for sample collection (Fig. 1). About 250 g samples of each variety were purchased from three shops in a single market, which was later mixed uniformly to make one and stored in individual zipper bags. Then, it went through the labeling process. After the proper labeling, They were then transported to the Institute of National Analytical Research and Service (INARS), Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka, Bangladesh, which is an ISO/IEC 17025:2017 accredited laboratory, for further analysis.

The rice samples represent the major brands available and most frequently consumed in the local markets of Bangladesh, as discovered from our literature review, market analysis, and observation. Details of rice sample varieties and origin from five different markets in Dhaka city are described in the supplementary information file (Table S1).

2.2. Analytical method and quality control

Rice grain samples were crushed to a fine powder with the help of a grinding machine, weighed using an analytical balance (EK 300H), and oven-dried (about 10g at 110 °C) for 24 h. Desiccators and crucible tongs were used to carry empty beakers and samples for cooling the heated objects and to absorb moisture in humid conditions. After drying, samples were placed inside the muffle furnace (model: WISD) at a temperature of 150 °C, which was gradually, step-by-step, increased to 600 °C within 4 h, and then left at this temperature for the next 10 h. The residual ash content was digested with 10 ml of HNO_3, 2.5 ml of H_2O_2, and deionized water in a hotplate at a temperature of 150 °C, filtered into a 25 ml volumetric flask using Whatman filter paper made up to the mark with DI water, and stored until analytical determination. Elemental analysis (Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb) was performed by Inductively coupled plasma mass spectrometry (ICP-MS). Appropriate quality assurance methods and safety measures were taken to ensure the accuracy of the outcomes. The entire study was carried out with deionized water. The reagents were of analytical grade, and the glassware used to quantify metals was immersed in 20 % HNO_3 for an entire night. To assess for contamination, 10 samples were combined with a blank sample. To calibrate the instrument, standards from each metal's stock solution were created. Repeated analyses were used to verify the precision and accuracy of the analysis. The level of accuracy was between 1 and 2%. Certified reference materials for rice grains were provided by the INARS, BCSIR, Dhaka, Bangladesh, to ensure quality.

2.3. Human health risk assessment

2.3.1. Estimation of Daily Metal Intakes (DMI)

The estimated daily intake of heavy metals (EDI) from rice was calculated by following equation (1) [27] [28].

$$\mathrm{DMI} = \frac{FR \times C}{BW} \tag{1}$$

In the above equation, the FR indicates the food ingestion rate (mg person^{-1} day^{-1}) (Household Income and Expenditure Survey of Bangladesh HIES, 2017). The food ingestion rate (g/person/day) is 367.19 (for adults) and 150 (for children). C here represents the concentration (mg kg^{-1}, fresh weight), and BW denotes the average body weight (adult = 60 kg, children = 16 kg) [8].

2.3.2. Non-carcinogenic risk

The target hazard quotient (THQ) [29] was used to quantify the non-carcinogenic risk of each specific metal through food consumption. This is defined as 'the ratio of a single drug exposure level over a particular time period (e.g., sub-chronic) to a reference dose (RfD) for that substance determined from a similar exposure period. The Hazard Index (HI) has been developed based on the health risk assessment of chemical combinations of USEPA standards [29] to quantify the complete prospective non-cancer-related impacts from numerous heavy metals [30]. THQ's relationship with total target hazard quotient (TTHQ) and HI was assessed using Eqs. (2) and (3) accordingly [31].

$$\mathrm{THQ} = \frac{EFr \times FIR \times C}{RfD \times BW \times AT} \times 10^{-3} \tag{2}$$

$$\mathrm{HI} = \mathrm{THQ1} + \mathrm{THQ2} + \mathrm{THQ3} \ldots \mathrm{THQ\ n} \tag{3}$$

Here, EFr is the exposure frequency (365 days/year) [32,33]. FIR, C, BW, RfD is the food ingestion rate (g/person/day), metal concentration (mg/kg), average body weight (Adult = 60 kg, Children = 16 kg) [33] and oral reference dose (mg/kg/day), respectively. AT is the averaging time for non-carcinogens (365 day/year* number of exposure years, assuming 70 years (25,550 days) and 6 years (2190 days) were adopted for carcinogenic risk calculation of adults and children, respectively [33].

If the THQ$<$1, the exposed population is unlikely to suffer evident negative consequences. If the THQ $\geq$1, a substantial health threat occurs, then appropriate actions and protective measures should be implemented [34–36].

2.3.3. Carcinogenic risk

The carcinogenic risk is characterized as the potential of an individual's developing cancer as a result of being exposed to carcinogenic exposures over their lifetime [30].

$$CR = \frac{EFr \times ED \times FIR \times CSFo \times C}{BW \times AT} \times 10^{-3} \quad (4)$$

Where CR is the carcinogenic risk. ED is the exposure duration adult = 70 years [32,33] and children = 6 years, [30]. CSF_0 is the Cancer Slope Factor (mg/kg/day). The oral carcinogenic slope factor from the Integrated Risk Information System (USEPA, 2010) database is presented in Table 1. According to USEPA (1989) [29], CR and TCR (Total Cancer Risk) values less than $1*10^{-6}$ are considered insignificant; however, CR and TCR values greater than $1*10^{-4}$ are likely to be harmful to human health.

The following formulae (5) and (6) can be used to compute the toxicity scores of each pollutant in a polluted medium [33].

Score of non-carcinogenic toxicity; $TS = C_{max}/ RfD$ (5)

Score of carcinogenic toxicity; $TS= SF * C_{max}$ (6)

Where TS is the Toxicity Score, C_{max} is the maximum pollutant concentration at the exposure point (mg/kg). SF is the slope factor (mg/kg/day). The oral carcinogenic slope factors from the Integrated Risk Information System [37] database are presented in Table 1.

2.3.4. Uncertainty and sensitivity analyses

In this study, we employed the Monte Carlo Simulation method to estimate the probability of carcinogenic risk associated with exposure to carcinogenic metals through rice consumption. This approach is widely recognized for its effectiveness in assessing risks and uncertainties in risk-based evaluations [34]. Our research considered several input variables, including the concentrations of metals (As, Pb, Ni, Cr, Cd), Exposure Frequency (EF), Exposure Duration (ED), Food Ingestion Rate (FIR), Body Weight (BW), Average Time (AT), and Cancer Slope Factor (Csfo). These input variables were treated as lognormal distribution functions, while AT and Csfo were considered as point estimates for the simulation. To ensure the accuracy of our results, each simulation involved 10,000 random trials for each input variable. We extracted the mean, median, and the 5th and 95th percentiles of the cancer risks associated with As, Pb, Ni, Cr, and Cd from the probability distribution of the TR model in this investigation. Additionally, we conducted a sensitivity analysis to identify the input variables that could significantly impact risk estimation under specific assumptions [34]. To execute the risk probability and sensitivity analysis, we utilized Oracle Co.'s Crystal Ball software version 11.1.2.4, ensuring a comprehensive and robust evaluation of the potential carcinogenic risks linked to metal exposure through rice consumption.

3. Results and discussion

3.1. Heavy metal concentration in rice grains

The heavy metal concentrations were analyzed and summarized using descriptive statistics, and the results are presented in Table 2. Detailed heavy metal concentration data for various rice varieties, including As, Pb, Cd, Cu, Zn, Cr, Mn, Fe, Co, Hg, and Ni, analyzed via ICP-MS, are provided in Supplementary Information 2 (Table S2), with concentrations reported in milligrams per kilogram (mg/kg). It's worth noting that heavy metal concentrations in rice exhibited significant variations both among and within different rice species. These variations in heavy metal concentrations within rice can be attributed to a range of factors, including climatic conditions, growth rates, and the duration of cultivation [38,39]. Furthermore, the metal concentrations in the cultivated soil and irrigation water also play a crucial role in influencing the variability of heavy metal concentrations observed in rice. In this study, the metal concentrations in rice varieties were arranged in descending order as follows: Zn > Fe > Mn > Cu > Ni > Cr > Pb > Cd > As > Co > Hg, where all results found were referred to fresh weight basis. The mean concentrations of Cr, Pb, Fe, and Co were found to be higher than other trace metals. The observed variations in metal concentrations among the analyzed food products can be attributed to the differing abilities of crops to absorb and accumulate metals [39]. These differences in heavy metal content can arise from various sources or factors, including variations in the growth environment (such as the presence of heavy metals in soil and the use of artificial fertilizers or pesticides), losses during machine-based production, or the use of food additives [40]. For a comprehensive overview of the metal concentrations in rice grains, please refer to the descriptive statistical summary presented in Table 2.

The mean concentration of Cr was observed to be 0.99 mg/kg in our study, which is notably higher than the average concentrations

Table 1
Oral reference dose (RfD) and Slope Factors for Carcinogenic Metals.

	As	Pb	Cd	Ni	Cr	Zn	Cu	Mn	Fe	Co	Hg
Reference Dose (RfD) (mg/kg/day)	0.0003 (Real et al., 2017)	0.004 (WHO 1993)	0.001 (US DOE (2011)	0.02 (USDOE 2011)	0.003 (USDOE 2011)	0.3 (US DOE 2011)	0.04	0.14 (US DOE 2011)	0.007	0.0003	0.00016 (Wei et al., 2015)
Carcinogenic Slope Factor (mg/kg/day)	1.5 US EPA (1991)	8.5×10^{-3}	6.1	1.7(Onuoha et al. (2015)	0.41	–	–	–	–	–	–

Table 2
Descriptive statistical summary of metals in rice grains.

	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd	Hg	Pb
Mean (mg/kg)	0.99	3.45	8.35	0.02	1.02	1.79	8.78	0.09	0.10	0.0007	0.49
SD (mg/kg)	0.009	0.029	0.11	0.0002	0.014	0.019	0.074	0.0011	0.0012	0.00005	0.005
Coefficient of Variance (%)	0.009	0.008	0.013	0.010	0.014	0.011	0.008	0.012	0.012	0.071	0.01
Min (mg/kg)	0.41	0.08	0.50	0.01	0.48	0.01	2.54	0.00008	0.01	0.0001	0.14
Max (mg/kg)	2.45	11.19	25.67	0.08	1.81	4.29	22.91	0.19	0.32	0.01	1.36
Permissible limit	0.1[a]	5[b]	5[b]	0.01[d]	1.5[b]	40[b]	60[b]	0.2[c]	0.4[c]	0.02[e]	0.2[c]

[a] CODEX-2011.
[b] WHO.
[c] CODEX) (FAO/WHO 2015.
[d] FAO.
[e] Zeng et al., 2015).

reported in previous studies, which were 0.183 mg/kg [30] and 0.01 mg/kg [33]. The highest concentration of Cr (2.45 mg/kg) was found in Lal Biroi rice, originating from Mymensingh. Cadmium is a metallic element that naturally occurs in low concentrations in the environment [30]. In our study, the mean and highest Cd concentrations were determined to be 0.1 mg/kg and 0.32 mg/kg, respectively, in Chinigura rice (originating from Chapai-Nawabganj). These values were higher than those reported in previous studies by Rahman and Islam, (2019) [8] and Ahmed et al. (2015) [30], where Cd concentrations were 0.04 mg/kg and 0.08 mg/kg, respectively. Lead is a non-essential element, and it is well established that Pb exposure can lead to neurotoxicity, nephrotoxicity, and a range of other adverse health effects [41]. The mean concentration of Pb was determined to be 0.49 mg/kg in our study, which is higher than the values reported by Rahman and Islam, (2019) [8] and Real et al. (2017) [33], who found concentrations of 0.44 mg/kg and 0.08 mg/kg, respectively, in rice samples. Notably, the highest Pb concentration (1.36 mg/kg) was detected in Lal Biroi rice, originating from Mymensingh. Nickel is typically present at low environmental levels and can have various detrimental effects on lung health, including inflammation, fibrosis, emphysema, and the development of tumors [42]. In this study, the mean concentration of Ni was found 1.02 mg/kg whereas Yasmin et al. (2019) [43] studied rice grains from local markets of Dhaka and observed a Ni concentration of 0.36 mg/kg which is also minor compared to the present study. The rice variety BR-28 from Sherpur had the highest Ni concentration (1.81 mg/kg), followed by Chinigura (1.70 mg/kg) from Chapai, Rajshahi. The concentration of Fe exceeded permissible limits in our study, with mean and maximum concentrations of 8.35 mg/kg and 25.67 mg/kg, respectively, in Amon rice from the Mymensingh region. These values were notably higher than those reported by Yasmin et al. (2019) [43]. In this study, the mean concentration of Co exceeded permissible limits, with the highest concentration (0.08 mg/kg) found in Amon rice from Mymensingh. According to WHO guidelines, the mean concentration of Mn remained within permissible limits. However, rice varieties such as Lal Biroi (11.19 mg/kg) and Amon (7.72 mg/kg) from the Mymensingh region exceeded these limits. On the other hand, essential nutrients Zn, As, Cu, and Hg had mean concentrations within international standard limits in the rice varieties studied. The mean concentrations were 8.78 mg/kg for Zn, 0.09 mg/kg for As, 1.79 mg/kg for Cu, and 0.0007 mg/kg for Hg. On the other hand, essential nutrients, Zn, As, and Cu are essential nutrients where the mean concentration of Zn (8.78 mg/kg), As (0.09 mg/kg), Cu (1.79 mg/kg), and Hg (0.0007) in rice varieties are within the International standard limit.

Fig. 2(a–k) illustrates the concentrations of heavy metals in various rice samples. It is evident that distinct rice varieties originating from different regions exhibit varying levels of metal concentrations. Among the 11 metals studied, six heavy metals (Cr, Fe, Pb, Co, Mn, and Ni) exceeded the permissible limits in a majority of the samples. Specifically, the concentration of Cr was notably high in Amon rice, Lal Biroi rice, Najirshail rice (originating from Mymensingh), Miniket rice (from Bogura), Chinigura rice (Chapai, Rajshahi), and Paijam rice (Naogaon). Similarly, the highest Fe concentration was detected in rice from Mymensingh and Barishal regions, including Amon, Lalbiroi, and Najirshail rice varieties. Elevated levels of Pb were found in the Barishal and Mymensigh regions, particularly in Amon and Lal Biroi rice. Additionally, Najirshail (Bogura), Miniket (Dinajpur), Chinigura (Chapai-Nawabganj), 28 (Sherpur), and Paijam (Naogaon) rice varieties exceeded international standard limits for Co concentration. Co concentration was found to be similar to Cr, Fe, and Pb in Amon and Lalbiroi rice varieties, both originating from the Mymensingh region. Notably, the highest levels of Co were observed in Najirshail (Sherpur), 28 (Sherpur), Miniket (Kushtia), Chinigura (Chapai-Nawabganj), and Paijam (Tangail). Mn concentration exhibited a similar pattern in Amon rice, Lal biroi, and Najirshail rice. Furthermore, Nickel was detected in four regions of Bangladesh, associated with four rice varieties: Lal Biroi (Mymensingh), 28 (Sherpur), Chinigura (Chapai, Rajshahi), and Paijam (Rangpur).

3.2. Health risk assessment

3.2.1. Daily intake of metals

Human exposure to metals typically occurs through ingestion, inhalation, and skin contact. Among these pathways, the most significant is ingestion, primarily through the food chain. In this study, rice consumption serves as the medium for the ingestion of metals, including As, Pb, Cd, Cu, Zn, Cr, Mn, Fe, Co, Hg, and Ni. To calculate the Daily Metal Intake (DMI) values for specific metals, we considered both the metal concentrations in rice and an individual's daily consumption rate. Different food intake rates were obtained from a preliminary report on Bangladesh's Household Income and Expenditure Survey (HIES) to estimate DMI. According to this data, the per capita daily rice intake is 367.19 g [44]. Our study revealed the following total mean daily intake values for these metals in

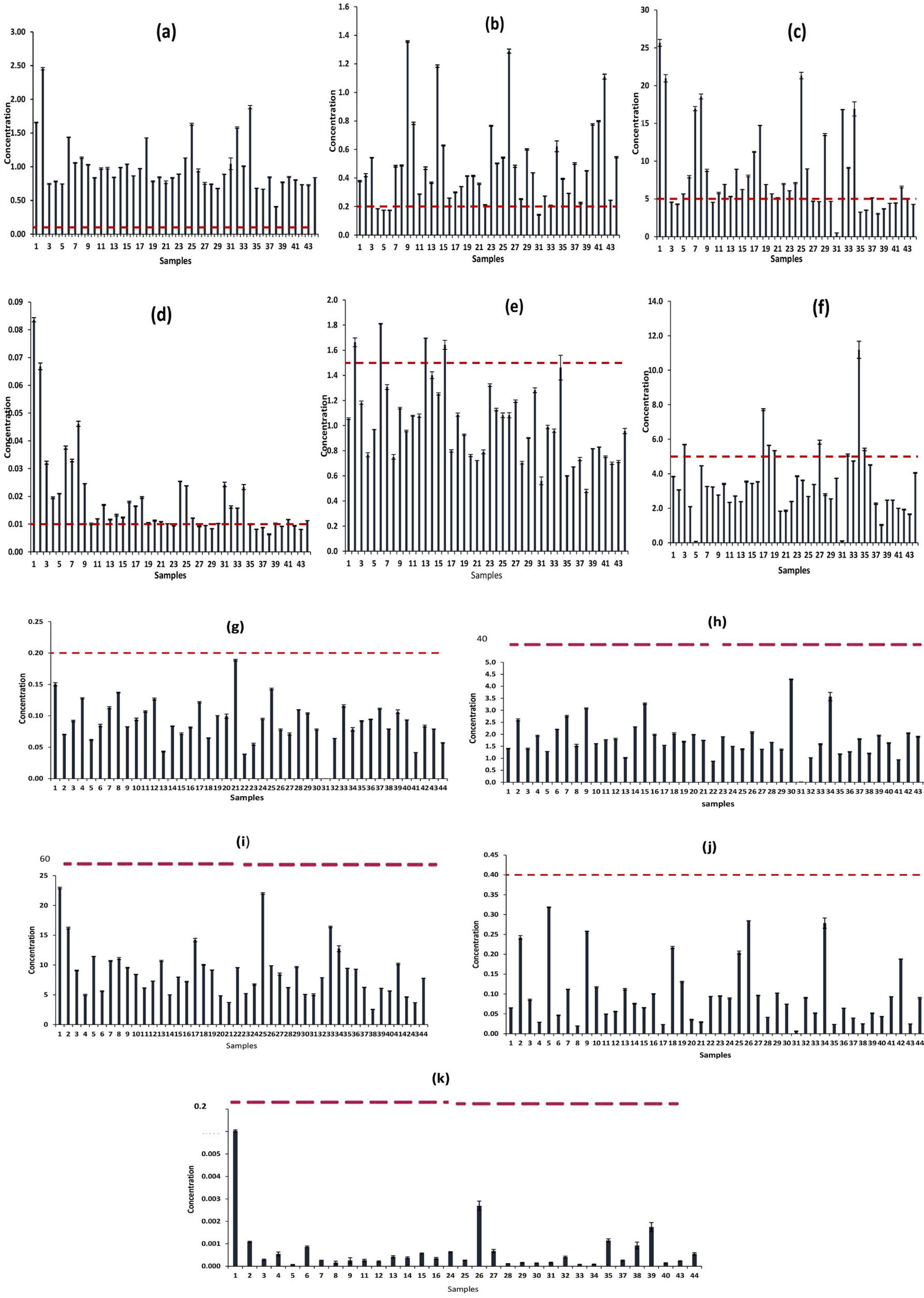

Fig. 2. Heavy metal concentration of (a) Cr (b) Pb (c) Fe (d) Co (e) Ni (f) Mn (g) As (h)Cu (i) Zn (j) Cd (k) Hg.

milligrams per day (mg/day) for both adults and children: Cr (0.00607, 0.0093), Mn (0.0211, 0.0324), Fe (0.0511, 0.0783), Co (0.000114, 0.174), Ni (0.00622, 0.00954), Cu (0.011, 0.0168), Zn (0.0537, 0.0823), As (0.000552, 0.000846), Cd (0.000603, 0.000923), Hg (0.00000411, 0.00000630), and Pb (0.00301, 0.00462) for adults and children, respectively. These results indicate that, for both adults and children, the order of total mean DMI values for these eleven individual trace metals from rice grains is as follows: Hg < Co < As < Cd < Pb < Cr < Ni < Cu < Mn < Fe < Zn. Importantly, the DMI values for these metals from rice grains were found to be below the Maximum Tolerable Daily Intake (MTDI) values recommended by international regulatory bodies, indicating that the metal intake from rice consumption does not exceed safe limits.

3.2.2. *Non-carcinogenic health risk*

3.2.2.1. Target hazard quotients (THQ). In our study, we utilized the THQ to assess the potential human health risks associated with the consumption of rice grains contaminated with metals by both adult residents and children. The THQ values for adults and children, concerning the eleven studied metals, are presented in a summarized table (Supplementary Information 3 and 4, Tables S3 and S4). In the context of heavy metal exposure, THQ values below 1.0 are considered “safe,” values between 1.0 and 5.0 indicate “a potential risk of adverse effects,” and values exceeding 5.0 signify “an unsafe level of exposure” [22]. Our study results reveal that the THQ values for Cr, Fe, and As across all rice varieties exceeded 1, indicating a potential health risk associated with exposure to these metals through rice consumption. Cu, on the other hand, exceeded a THQ value of 1 in only one rice variety, suggesting a potential health risk associated with the ingestion of this specific metal in that rice variety. Moreover, Co, Cd, and Pb exceeded THQ values of 1 for certain rice varieties, further highlighting the potential health risks linked to the consumption of these metals through contaminated rice. However, it’s important to note that other analyzed heavy metals, namely Mn, Ni, Zn, and Hg, did not exhibit THQ values indicative of any health hazards.

Fig. 3a and b specifically highlight the heavy metals contributing significantly to non-carcinogenic health risks (THQ>1) and their association with the origin of each rice variety. In Fig. 3a, the Amon rice variety stands out, with Fe levels posing a higher THQ than other metals, particularly impacting the vulnerability of children over adults. The regions of Mymensingh and Manikganj exhibit varying orderings of target hazard quotients, with high levels of Fe, Cr, As, and Cd found in Mymensingh, Co in both Mymensingh and Manikganj, and Pb in Mymensingh, but specifically affecting children. Fig. 3b demonstrates the Lal Biroi rice from the Mymensingh region, with metal concentrations in the order of Fe > Cr > As > Cd > Pb > Co. THQ for Najirshail rice, sourced from Gulistan Kaptan Bazar in Bogura, shows the highest levels of Fe, As, Cd, and Cu, while Uttra-Ashkona Bazar contains higher levels of Cr and Pb compared to other metals. The common source for all these metals is traced back to the Bogura region. Notably, the THQ value for Miniket rice is notably higher in the children’s group. Fe and Cr levels are elevated in Uttra-Ashkona Bazar (Shantahar, Bogra), As is prominent in Gulistan Kaptan Bazar (originating from Chapai, Rajshahi), and Pb is elevated in Krishi Market (originating from

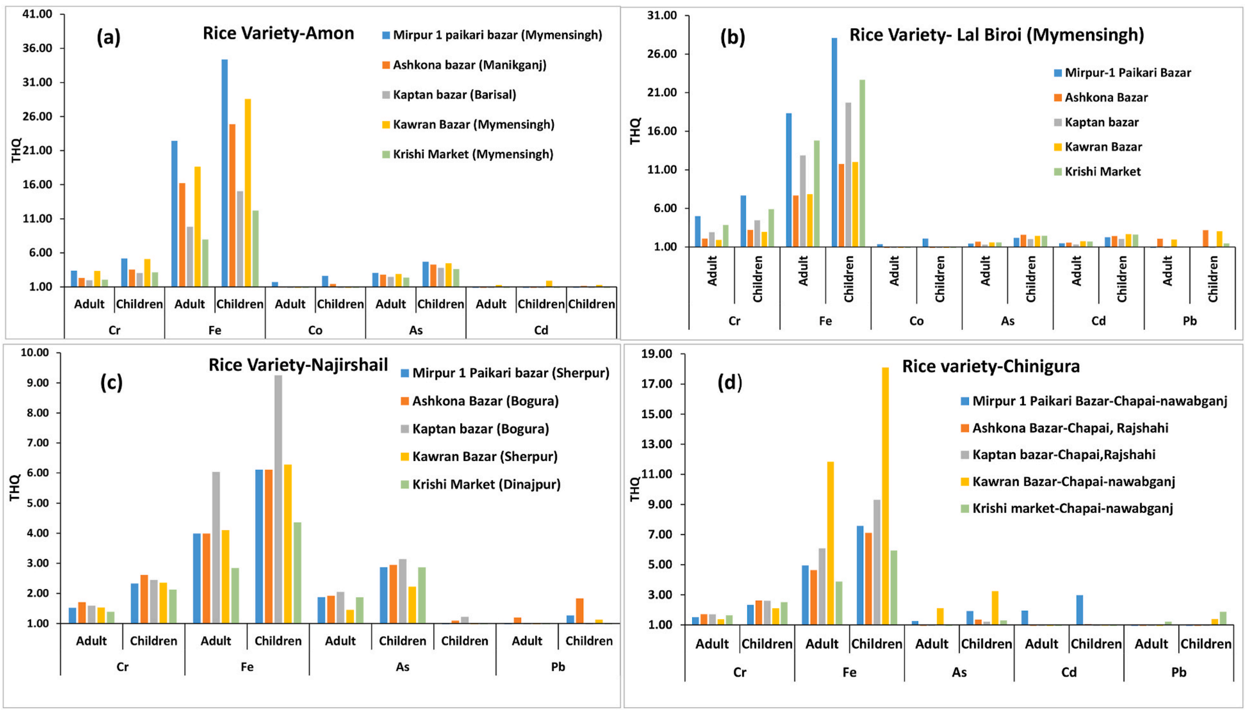

Fig. 3a. Target Hazard Quotient (THQ) for metals along with rice variety and Origin (a) Amon Rice (b) Lal Biroi Rice (c) Najirshail Rice (d) Chinigura Rice.

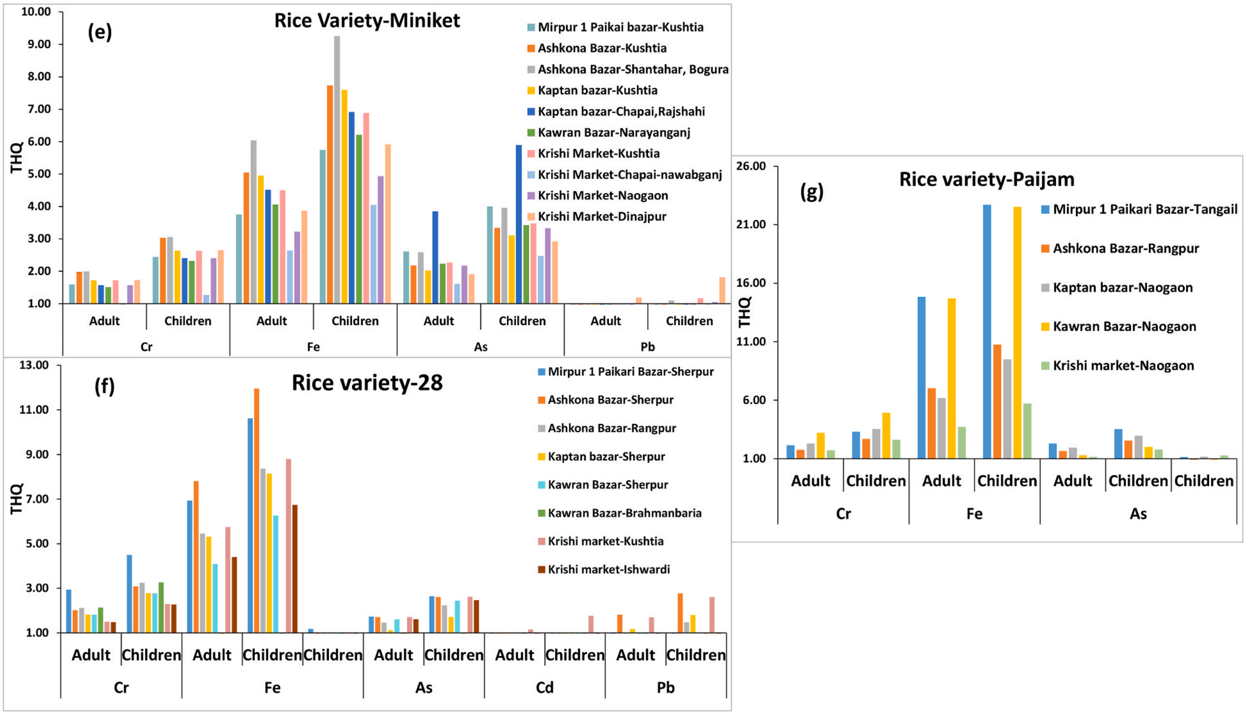

Fig. 3b. Target Hazard Quotient (THQ) for metals along with rice variety and Origin (e) Miniket Rice (f) BR-28 Rice (g) Paijam Rice.

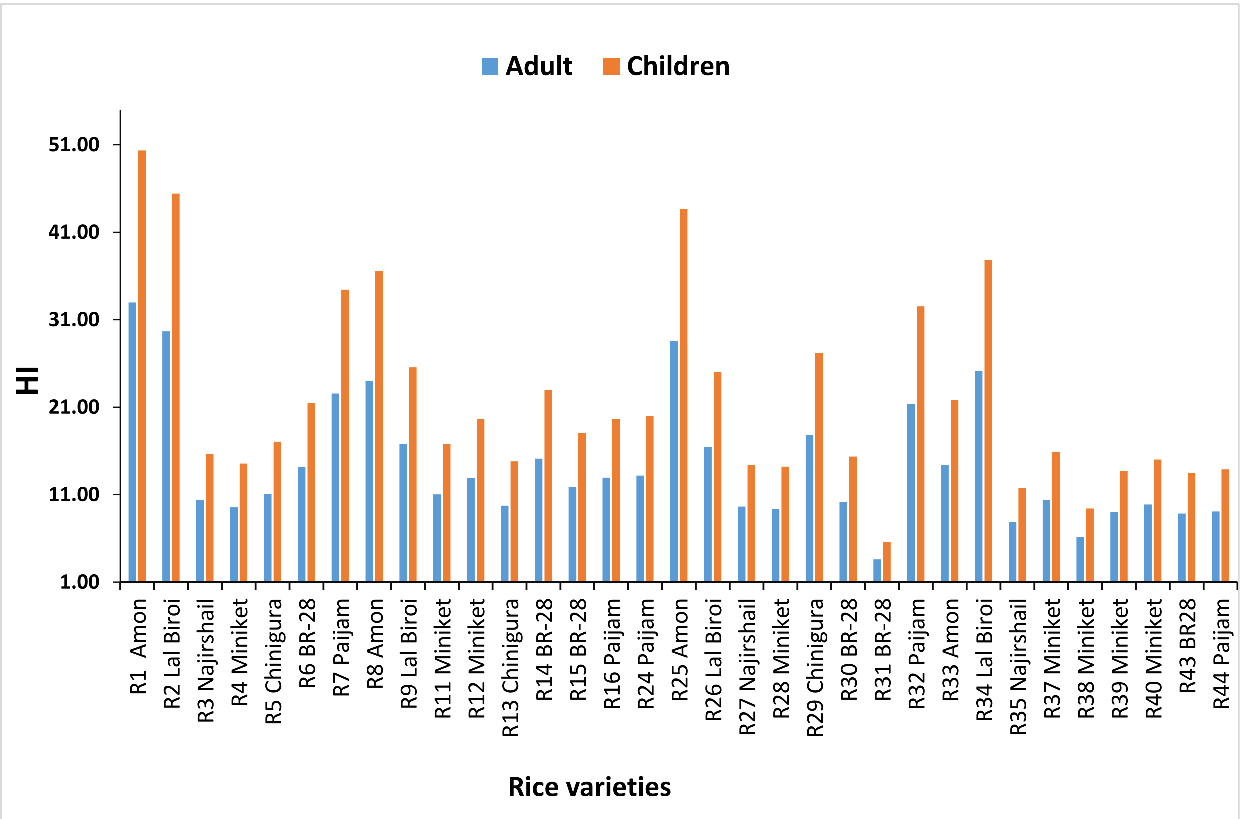

Fig. 4. Hazard Index of rice varieties considering multiple metals addressing Adult and Children groups.

Dinajpur, Kushtia), all signifying health risks. The Chinigura rice variety demonstrates significant THQ values for the metals depicted in the figure. Kawran Bazar (Chapai Nawabganj) records the highest values for Fe and As. Conversely, rice from Chapai Nawabganj (Mirpur-1 Paikari Market) shows the highest health risk for Cd. Metals in BR-28 rice exceed permissible limits for Cr, Fe, As, Cd, and Pb, posing significant health risks. BR-28 exceeds limits only in Mirpur-1 Paikari Market (originating from Sherpur) and Mohammadpur Krishi Market (originating from Kushtia) in terms of Co and Cd. Cu exceeds limits only in rice originating from Sherpur, with children being particularly at risk. Pb levels are notably high in Mohammadpur Krishi Market (originating from Kushtia), Gulistan Kaptan Bazar, Kawran Bazar, and Uttra-Ashkona Bazar (originating from Sherpur), with Kawran Bazar posing a health risk exclusively for children. Lastly, the Tangail and Naogaon regions exhibit the highest levels of Fe and Cr for Paijam rice, as depicted in Fig. 3b. All varieties of Paijam rice exceed permissible limits for Fe, Cr, and As, with only Pb and Cd presenting non-carcinogenic risks for children in the Tangail region, while the others show no significant health risks.

3.2.2.2. Hazard Index (HI). The HI is a measure that quantifies the combined non-carcinogenic effects of multiple metals. As illustrated in Fig. 4, the HI values resulting from rice consumption were consistently found to exceed 1 for all the rice varieties. Notably, almost all the rice varieties exceeded the permissible limits, with the vulnerability of the children's group being notably higher than that of adults.

3.2.2.3. Carcinogenic health risk. In this study, we focused on assessing the carcinogenic risk to humans exposed to specific metals, namely As, Cd, Cr, Pb, and Ni. According to the USEPA (1989) [29], cancer risk below 1×10^{-6} (1 chance in 1,000,000 lifetime exposure) is minimal, but cancer risk beyond 1×10^{-4} (1 chance in 10,000-lifetime exposure) has a significant adverse consequence, and risk management judgments are taken between these values [37]. Our findings regarding the carcinogenic risk (CR) indicate that Ni (1.88E-02 for adults and 2.89E-02 for children) presents the highest level of cancer risk. As depicted in Fig. 5, the order of cancer risk, from highest to lowest, is Ni > Cd > Cr > As > Pb. It is evident that each of these metals poses a significant health risk for both adults and children across various rice varieties, with Pb showing a notably high average risk. This suggests the presence of sources of Ni, Cd, Cr, and As near agricultural land, as all rice varieties exceeded permissible limits. Notably, children are more vulnerable than adults in every aspect of these risks. Consequently, it is imperative to implement proper risk management strategies, particularly with a focus on protecting children's health.

In our assessment of heavy metal toxicity scores, prioritized by their carcinogenic effects, the ranking is as follows: Ni > Cd > Cr > As > Pb. For non-carcinogenic effects, the heavy metal toxicity score sequence is Fe > Cr > As > Pb > Cd > Co > Cu > Ni > Mn > Zn > Hg. According to the toxicity scale employed in this study, heavy metals Ni and Fe emerge as the most hazardous. This underscores the importance of vigilant monitoring of potential sources of heavy metals in various food products. Utilizing toxicity ratings can provide valuable guidance not only to heavy metal users, such as industrialists selecting environmentally friendly raw materials, but also to regulatory authorities investigating safety and standard compliance issues.

3.2.2.4. Probabilistic risk assessment and sensitivity analysis. The probability of cancer risk through ingestion of rice was assessed using CR, and As, Pb, Ni, Cr, and Cd were studied using the Monte-Carlo Simulation method in crystal ball software demonstrated in Fig. 6a and b considering both adult and children groups. Details of the results are described in Supplementary Information 5, Table S5. The results reveal that the mean CR probabilities for As, Pb, Ni, Cd, and Cr were 8.3×10^{-4}, 2.57×10^{-5}, 6.61×10^{-3}, 2.5×10^{-4}, 3.74×10^{-2} and 1.28×10^{-3}, 3.93×10^{-5}, 1.01×10^{-2},3.89×10^{-4}, 5.70×10^{-2} for adult and children respectively. At the same time, the median values of CR for As, Pb, Ni, Cd, and Cr were 8.1×10^{-4}, 2.51×10^{-5}, 6.45×10^{-3}, 2.5×10^{-4}, 3.63×10^{-2} and 1.25×10^{-3}, 3.84×10^{-5}, 9.86×10^{-3}, 3.80×10^{-4} and 5.57×10^{-2} respectively for adult and children group with 100 % certainty. The 5th and

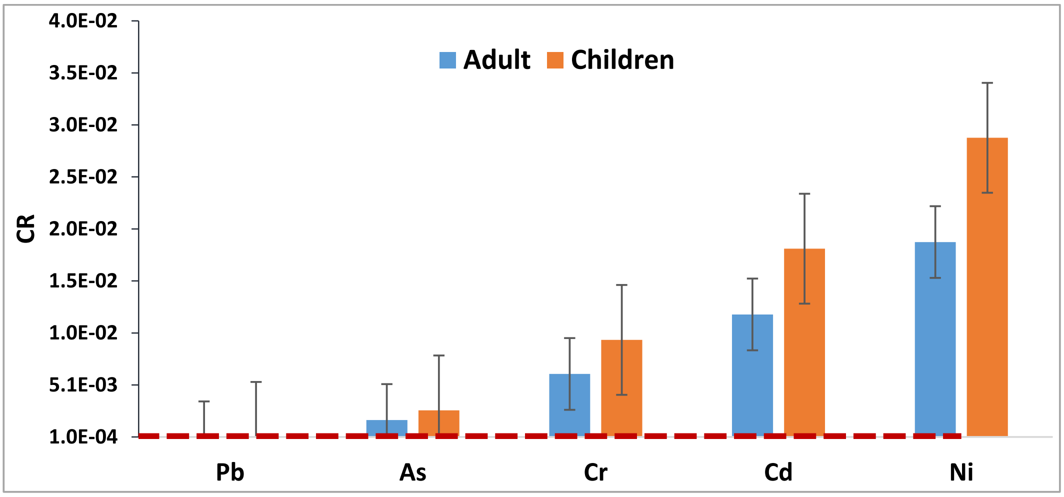

Fig. 5. Carcinogenic health risk between Adult and Children group.

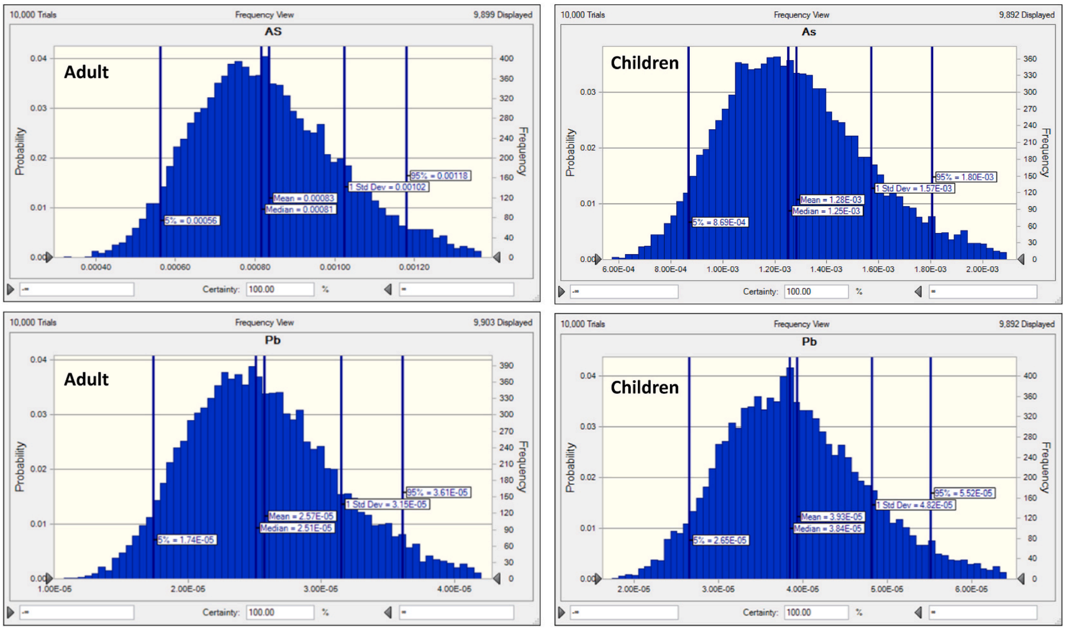

Fig. 6a. Predicted probability results of the target carcinogenic risk (CR) for As & Pb for adult and Children Group.

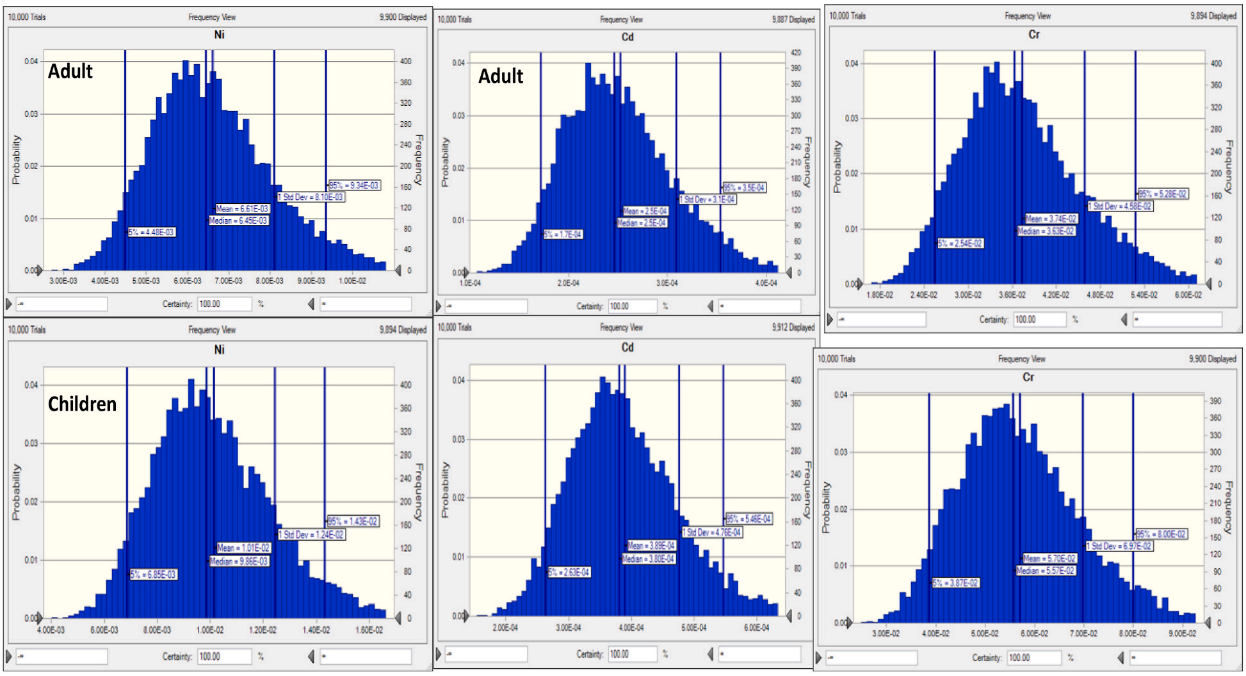

Fig. 6b. Predicted probability results of the target carcinogenic risk (CR) for NI, Cd, and Cr for adult and Children Group.

95th percentile values for adult group were found 5.6×10^{-4} and 1.18×10^{-3} for As, 1.74×10^{-5} and 3.61×10^{-5} for Pb, 4.48×10^{-3} and 9.34×10^{-3} for Ni, 1.7×10^{-4} and 3.5×10^{-4} for Cd, 2.54×10^{-2} and 5.28×10^{-2} for Cr and the children group the percentile values found 8.69×10^{-4} and 1.80×10^{-3} for As, 2.65×10^{-5} and 5.52×10^{-5} for Pb, 6.85×10^{-3} and 1.43×10^{-2} for Ni, 2.63×10^{-4} and 5.46×10^{-4} for Cd, 3.87×10^{-2} and 8×10^{-2} for Cr. Following the USEPA (2010) [37] guideline, it was observed that the mean and 95th percentile values of As, Ni, Cd, and Cr exceeded the threshold value (>10-4), which indicates that 95 % of people would experience high potential cancer risk from rice consumption gradually. Additionally, the median and 5th percentile values of As, Ni, Cd, and Cr exceeded the safe limit ($<10^{-6}$). In contrast, the mean, median, and 95th percentile values for Pb were all higher than

(10^{-6}), implying that 95 % of people in the study area have crossed the safe limit boundary and may confront a lifetime risk of cancer from eating Pb-contaminated rice, even though the value was within an acceptable range (10^{-4} to 10^{-6}). However, approximately 5 % of the population would be free of the danger of Pb-induced cancer through rice consumption. Additionally, Ni, As, Pb, Cd, and Cr can be regarded as the priority heavy metals due to their carcinogenic risks.

The importance of the input variables involved in the CR calculation was assessed by sensitivity analysis Bodrud-Doza et al., 2019 [45] where the result revealed that the concentration of Pb, Ni, and Cr is the most important factors in the CR values for metals illustrated in Fig. 7. For As induced CR calculation, Concentration, Food Ingestion Rate (FIR), Exposure Duration (ED), and Exposure Frequency (EF) revealed the positive influences with the percentage of 20.6 %, 20.7 %, 19.9 %, and 19 % for adult group and 19.4 %, 19.9 %, 19.4 % and 19.6 % respectively for children group. Only Body Weight (BW) had a negative impact on the Carcinogenic Risk (CR) estimate, with a percentage of −19.8 percent and −21.7 % respectively. For the Pb-induced CR calculation, concentration (20.5 & 19.8 %), FIR (20.4 & 19.3 %), EF (19.9 & 20.4 %), and ED (19.6 & 21.1 %) all showed positive influences, whereas only BW (−19.6 & −19.4 %) showed a negative impact for adult and children.

Concentration, Food Ingestion Rate (FIR), Exposure Duration (ED), and Exposure Frequency (EF) all had a positive effect on Ni-induced CR calculation for adults and children, with percentages of 19.1 % & 20.5 %, 20 % & 19.5 %, 20.1 % & 19.7%and 20.1 % respectively. Only Body Weight (BW) had a negative impact on Carcinogenic Risk (CR) estimation, with percentages of −20.6 and −19.9 severally. Sensitivity analysis for Cr considering both groups where Cr concentration level is responsible for adult cancer risk estimation. All input variables revealed positive influences in Cd, where FIR had more impact than others. However, this study indicates that metal concentration and food ingestion rate are significantly responsible for cancer risk estimation. The overall findings of the study are summarized in Fig. 8.

4. Conclusions

This research addressed the presence of heavy metals in commonly consumed ready-to-cook rice grains in Bangladesh and their associated health risks, with a specific focus on distinguishing risks for adults and children. Toxicity scores aided in identifying key elements warranting further investigation. The study revealed that mean metal concentrations (Cr, Fe, Co, Pb) in rice exceeded permissible limits, prompting a detailed health risk assessment employing carcinogenic and non-carcinogenic health indices. Children exhibited higher vulnerability. Unacceptable health risks were identified for Cr, Fe, and As, along with significant cancer risks from Ni, Cd, Cr, and As, affecting 95 % of both groups if contamination sources are not controlled. This research highlighted specific regions in Bangladesh—Mymensingh, Bogura, Sherpur, Kushtia, Chapainawabganj, Naogaon, and Tangail—as major contributors to heavy metal-contaminated rice. Further studies considering water and soil quality, as well as the rice production process, are warranted to identify contamination sources. In conclusion, consistent monitoring, quality assurance and a proper management plan are required to

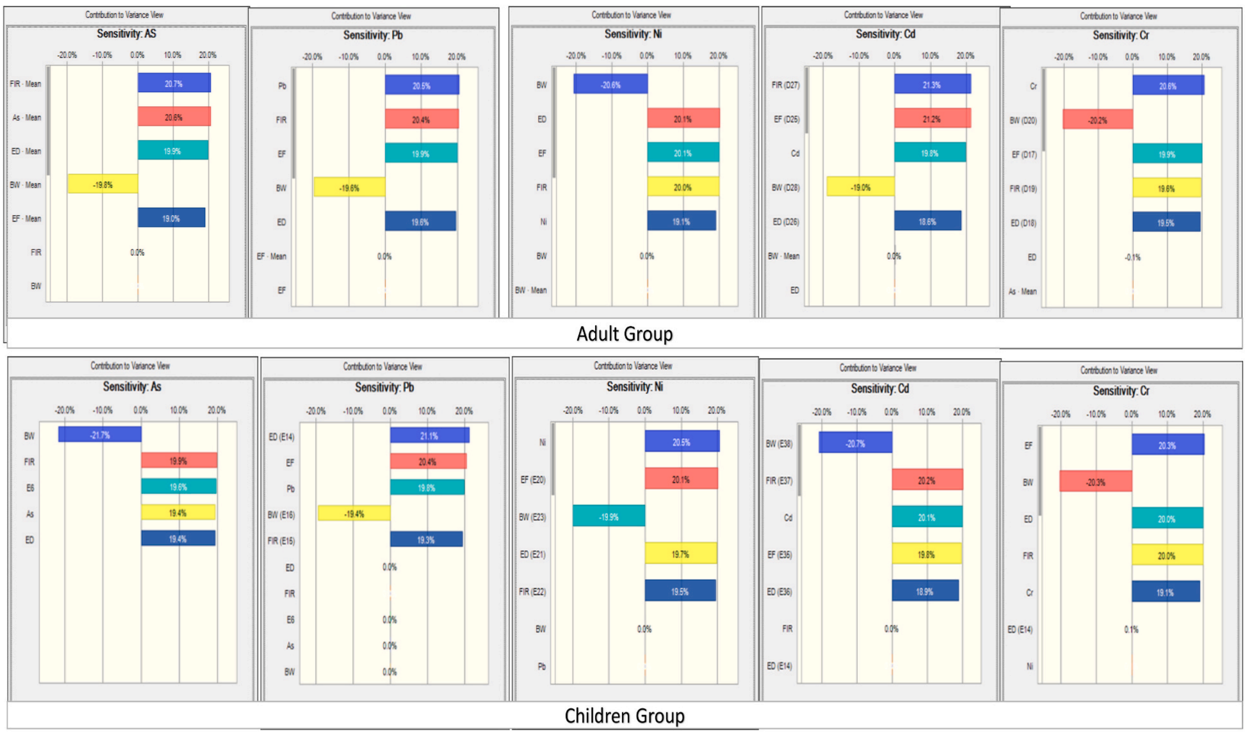

Fig. 7. Sensitivity analysis on the Target Carcinogenic Risk for As, Pb, Ni, Cd, and Cr.

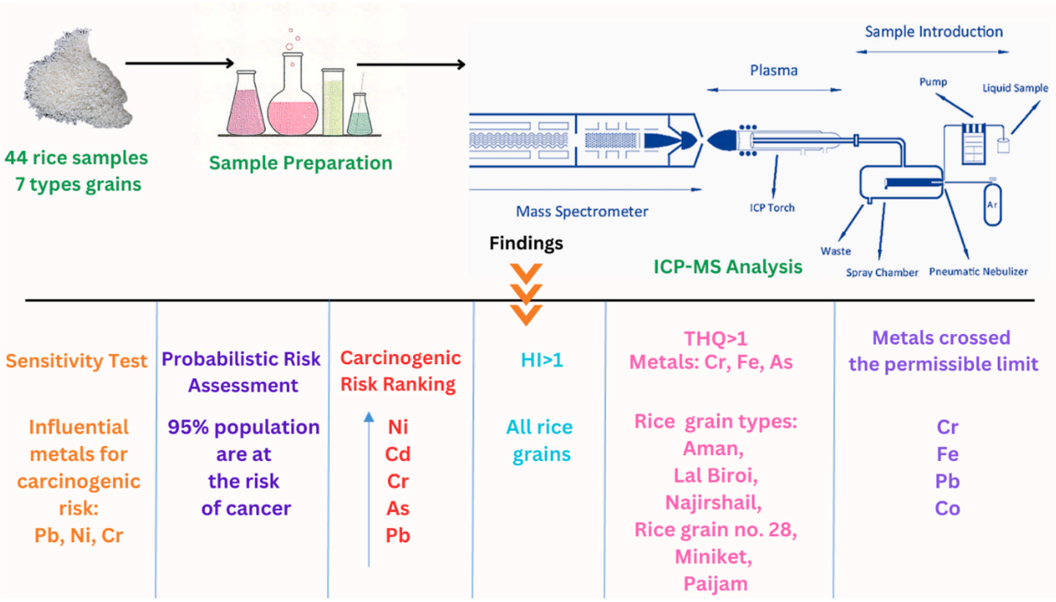

Fig. 8. Overall findings of the study.

ensure food security in Bangladesh.

CRediT authorship contribution statement

Shamaila Islam: Writing – original draft, Methodology, Formal analysis, Conceptualization. **Md Ahedul Akbor:** Writing – review & editing, Visualization, Investigation, Formal analysis, Data curation, Writing – review & editing, Validation, Supervision, Resources, Project administration, Investigation, Data curation. **Farah Noshin Chowdhury:** Visualization, Software, Resources, Project administration. **Mehedi Hasan:** Visualization, Validation, Resources, Project administration, Investigation. **Aynun Nahar:** Visualization, Validation, Supervision, Software, Resources, Methodology. **Md Abu Bakar Siddique:** Visualization, Resources, Methodology, Investigation, Data curation. **Md Moniruzzaman:** Visualization, Software, Project administration, Formal analysis, Data curation. **Md Selim Reza:** Writing – review & editing, Visualization, Methodology, Formal analysis, Data curation. **Md Iftakharul Muhib:** Writing – review & editing, Validation, Supervision, Resources, Methodology, Data curation.

Data availability

The datasets will be available from the corresponding author on reasonable request.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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