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ARTICLE

Presence of toxic metals in rice with human health hazards in Tangail district of Bangladesh

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ABSTRACT

Rice is the staple food of Bangladeshi people and is consumed at least twice a day. Thus, the presence of toxic metals in rice grains has become a major public health concern in Bangladesh. The present research was conducted to investigate the concentrations of toxic metals in rice grains and their possible human health risks in the Tangail district of Bangladesh. Toxic metals were measured by using an inductively coupled plasma mass spectrometer (ICP-MS), and the mean concentrations of toxic metals in rice samples were found in order of Cr > Pb > Ni > As > Cu > Cd. The concentrations of Cr, Pb, As, and Cd in the studied rice grain samples exceeded the FAO/WHO standard values for food samples by 100%, whereas the Ni concentrations by 10%. The principal component analysis (PCA) revealed significant anthropogenic contributions of Cr, Ni, As, and Pb concentrations in rice grains. The metal concentrations in rice grain samples showed strong significant correlations by forming primary clusters with each other. The estimated daily intake (EDI) values of Cr, Ni, As, Cd, and Pb from all samples were higher than the maximum tolerable daily intake (MTDI) allowed. The total targeted hazard quotient (TTHQ) values of Cu, Ni, As, Cd, and Pb also exceeded the threshold value of 1.00, indicating a potential non-carcinogenic risk. The estimated target carcinogenic risk of As was higher than the USEPA threshold level 10^{-4} (0.0001) indicating increased risk of cancer for adults and children in the study area.

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Heavy metals; rice; transfer factor; cancer risk; Bangladesh

Introduction

Rice is considered the world’s third largest crop, which plays a significant role in human nutrition (Proshad et al. 2019a). It is an important source of energy, vitamins, minerals and amino acids for its consumers around the world (Zeng et al. 2009). Some researchers estimated that rice is supplying almost half of the daily calories of the world population (Jaisut et al. 2008; Zhang 2009; Abbas et al.

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2011; Pishgar-Komleh et al. 2011). Gbabo et al. (2009) found that rice supplies more than half of all the calories humans consume comparing with wheat and maize. According to Wailes et al. (2005), calories obtained from rice is 20.5% globally, 29.2% in low-income countries, and 31.6% in Asian countries. In Bangladesh, over 80 different rice varieties are cultivated in three seasons (aus, aman and boro) and 34.7 million metric ton of rice grain is produced per year (BBS 2016). Four-fifths of the total population of Bangladesh consumes rice three times a day as their main meal which provides nutrients to consume (Islam et al. 2015).

At present, there is increasing concern of toxic metal contamination in the arable lands and their transfer to agricultural crops, especially to rice (Zhao et al. 2010). Industrial discharge, application of agrochemicals, and mining are the major anthropogenic sources of toxic metal contamination in the environment (Granero and Domingo 2002; Lee et al. 2005; Han et al. 2006; Kamani et al. 2018), which subsequently contaminate the total environmental components including soil, water and plants (Bundschuh et al. 2012; Ahmed et al. 2015). Depending on prevailing conditions, toxic metals present in soils could be readily available to plants (Chandrajith et al. 2012). Agricultural crops grown in toxic metal contaminated soils could accumulate greater amount of metals than those grown in uncontaminated sites (Sharma et al. 2006, 2007; Marshall et al. 2007). Considering the food safety issues and potential health risks, accumulation of toxic metals in arable land is now a growing concern worldwide, especially for agro-economic countries like Bangladesh (McLaughlin and Singh 1999; Bishwajit et al. 2014).

Toxic metals can be accumulated and transferred to rice plants and grains when it is grown in metal-contaminated soils. A variety of toxic metals including arsenic (As), cadmium (Cd), copper (Cu), lead (Pb) and mercury (Hg) are of primary concern in rice cropping systems (McLaughlin and Singh 1999). Fu et al. (2008) reported that the average level of lead (Pb) was 0.69 mg/kg in polished rice in an E-waste recycling area in southeast China, while Zhao et al. (2010) found that Cd had a a maximum value of 0.467 mg/kg in rice. A high level of Pb (0.957 mg/kg) in rice was also observed by Hang et al. (2009). These studies denote the possible contamination of rice plants and grains by toxic heavy metals.

Although there are many routes of heavy metals to be absorbed into the human body, ingestion of food is one of the principal pathways. Continuous exposure to such toxic metals through the consumption of food, even contaminated with low concentrations, can cause serious problems to human health throughout their lifetime (Mohammadi et al. 2019). For instance, a long time exposure to Cd can damage to the kidneys and liver as well as hamper the nervous systems (Wang and Du 2013), whereas neurological, immunological, cardiovascular, renal, and reproductive problems are associated with low levels of Pb exposure (NTP 2012). Abdul et al. (2015) reported that As exposure in the human body can cause different types of integumentary, nervous, respiratory, cardiovascular, immune, endocrine, hepatic and renal complications. Because of its toxicity at low exposures, aresenic is known as a toxic heavy metal though it is a metalloid irrespective of its atomic mass (Duffus 2002; Tchounwou et al. 2012).

Since rice is dominantly produced and consumed in Bangladesh, there is a high probability of carcinogenic and non-carcinogenic health risks associated with the consumption of metal-contaminated rice grains. Though several research studies have been performed in the past by several researchers to assess potential health risks for population living near mine and smelter sites because of consumption of toxic metal contaminated farm crops (Cui et al. 2004; Zheng et al. 2007; Sipter et al. 2008; Zhuang et al. 2009), detailed research about metal toxicity in rice is still in its infancy in Bangladesh. Therefore, toxic metals present in rice grown on agricultural lands in the vicinity of the industrial areas in Tangail district, Bangladesh was assessed along with possible human health risks in this study.

Materials and methods

Study area

A total of 100 rice samples were collected from ten selected upazila of Tangail district, Bangladesh (Figure 1), while one upazila was considered as the sampling site. Tangail district covers an area of 3375 km^2 and is situated at the central part of Bangladesh. Tangail district is a densely populated area in Bangladesh and the population density is 975/km^2 (Banglapedia 2014). Agriculture is the principal land-use type in Tangail district and about 2800 km^2 of cultivatable lands are available in the district. About 50% of the populations of Tangail district are involved with agricultural activities, and paddy is the main agricultural product (Rahman and Mian 2015). In Tangail, there are about 1200 industries (BBS 2013) including textile and garments industries, dyeing industries, battery manufacturing industries, packaging industry, glass industries, tanneries, metal workshops, pesticide and fertilizer industries, and food processing industries which collectively produce large volumes of effluents containing toxic metals (Tusher et al. 2017; Proshad et al. 2019a). These industries discharge their untreated effluents randomly onto the surrounding agricultural lands (Tusher et al. 2017), rivers and/or canals for waste dumping (Proshad et al. 2019a). Those wastewaters, containing different toxic heavy metals, get mixed with soils and thus the soil of the area is continuously being polluted by toxic elements. From the soil, toxic metals may transfer to crop plants resulting in serious health problems to both humans and animals alike.

Rice sample collection and preparation for laboratory analysis

Rice grain (*Oryza sativa* L.; rice variety: BRRI dhan28) samples were collected from ten selected sampling sites of agricultural fields in the vicinity of the industrial areas. BRRI dhan28, one of the

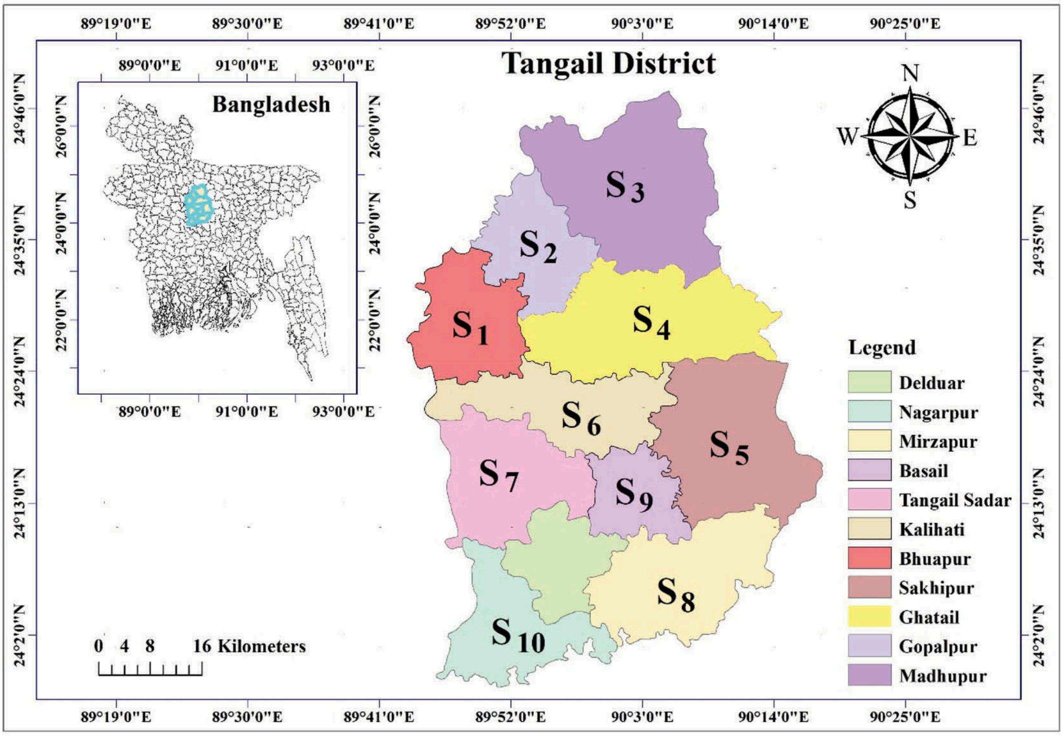

Figure 1. Map showing the study area of Tangail district, Bangladesh.

modern and high-yielding rice varieties developed by Bangladesh Rice Research Institute (BRRI), was selected for the present study since this rice variety is highly popular and predominantly cultivated during all growing seasons in Bangladesh. One hundred rice samples were collected by hand from the selected agricultural fields at the beginning of May 2018. Ten samples were collected from each sampling site. For each sample, rice grains were collected from three places in the same field as sub-samples and were thoroughly mixed to form a composite sample. The rice samples collected for chemical analysis were kept in polythene zip-bags with appropriate markings and labeling and brought to the laboratory on the same day of sampling. Samples were then washed with distilled water and were kept in an oven at 70–80°C to attain a constant weight (Tiwari et al. 2011).

Toxic metal analysis

All the chemicals used were of analytical grade reagents, while the Milli-Q water (Elix UV5 and Milli-Q, Millipore, Boston, MA, USA) was used for the preparation of solutions. The digestion and analysis of the collected samples were performed following the procedures described by Proshad et al. (2019b). Briefly, about 0.3–0.5 g of the rice sample was treated with 6 mL of 69% HNO_3 (Kanto Chemical Co., Inc., Tokyo, Japan) and 2 mL of 30% H_2O_2 (Wako Pure Chemical Industries, Ltd., Osaka, Japan) in a closed Teflon vessel and was digested in a Microwave Digestion System (Berghof speedwave, Eningen, Germany). The digested samples were then transferred to a Teflon beaker, and the total volume was increased to 50 mL with Milli-Q water. The digested solution was then filtered by using a syringe filter (DISMIC1–25HP PTFE, pore size = 0.45 mm; Toyo Roshi Kaisha, Ltd., Tokyo, Japan) and stored in 50 mL polypropylene tubes (Nalgene, New York, NY, USA). Prior to its use, the Teflon vessel and polypropylene containers were properly cleaned, soaked in 5% HNO_3 for more than 24 hours, then rinsed with Milli-Q water and dried. The digestion tubes were then cleaned using a blank digestion procedure following similar procedure of samples. Afterwards, the samples were analyzed using an inductively coupled plasma mass spectrometer (ICP-MS, Agilent 7700 series, Santa Clara, CA, USA) to measure the concentrations of toxic metals. The detection limits of ICP-MS were 0.7, 0.6, 0.8, 0.4, 0.06, and 0.09 ng/L for Cr, Ni, Cu, As, Cd, and Pb, respectively. Multi-element Standard XSTC-13 (Spex Certi Prep®, Metuchen, NJ, USA) solutions were used to prepare the calibration curves. Internal calibration standard solutions containing 1.0 mg/L of indium, yttrium, beryllium, tellurium, cobalt and thallium were purchased from Spex Certi Prep® (Metuchen, NJ, USA). During the procedure, a 10 mg/L internal standard solution was prepared from the primary standard and added to the digested samples. A multi-element solution (Agilent Technologies, Japan) was used as the tuning solution covering a wide range of masses of elements. All test batches were evaluated using an internal quality approach and validated to see if they satisfied the defined Internal Quality Controls (IQCs). Before starting the analysis, the relative standard deviation (RSD, <5%) was checked by using tuning solution purchased from Agilent Technologies. The certified reference materials INCT-CF-3 (corn flour) bought from the National Research Council (Canada), were analyzed to confirm analytical performance and good precision (relative standard deviation bellow 20%) of the applied method.

Transfer factor (TF) of heavy metals

The transfer factor (TF) of metals from soil to plant parts was defined as the ratio of the metal concentration in the plant's tissues to the metal concentration in soil. The transfer factor was calculated for each plant sample separately. A transfer factor can be used to evaluate the potential capability of plants to transfer metals from soil to plant tissues. The transfer factor was calculated as follows:

$$TF = C_{plant}/C_{soil} \tag{1}$$

Where, C_{plant} and C_{soil} represent the total metal concentration in the plant part (mg/kg) and total metal concentration in soil (mg/kg) on a dry weight basis (Li et al. 2012).

Estimated daily intakes (EDIs)

Estimated daily intakes (EDIs) of toxic metals (mg/day) were calculated using their respective average concentration of heavy metals in rice by the weight of rice consumed by an individual (body weight 60 kg for an adult in Bangladesh) (FAO/WHO 2011), which was obtained from the household income and expenditure survey (HIES 2011). They are calculated by the following formula:

$$EDI = (FIR \times C)/BW \tag{2}$$

FIR is the food ingestion rate (g/person/day), C is the metal concentration in rice samples (mg/kg), and BW is the body weight assuming 60 kg for adult residents. Food ingestion rate was addressed by a questionnaire survey in the study area. A total of 100 households, 10 from each upazila/sampling site, were surveyed and finally the data were used to calculate the average rice ingestion rate. This rate was found to be 445 g per adult and 200 g for children within the study area of Bangladesh.

Non-carcinogenic risk

Hazard quotients (HQs)

The methodology for the estimation of non-carcinogenic risks was applied in accordance with that provided by the U.S. Environmental Protection Agency (USEPA) Region III's risk-based concentration Table (USEPA 2010). The non-carcinogenic risk for each individual metal through rice consumption was assessed using the target hazard quotient (THQ)(USEPA 1989), which is 'the ratio of a single substance exposure level over a specified time period (e.g. sub-chronic) to a reference dose (RfD) for that substance derived from a similar exposure period'. The equation used for estimating the target hazard quotient is as follows:

$$THQ = (EFr \times ED \times FIR \times C)/(RfD \times BW \times AT) \times 10^{-3} \tag{3}$$

THQ is the target hazard quotient, EFr is the exposure frequency (365 days/year), ED is the exposure duration (70 years), FIR is the rice ingestion rate (g/day) (daily rice consumption rate for adult residents was an average of 445 g fresh weight basis, questionnaire survey of this study), C is the metal concentration in rice (mg/kg fw), RfD is the oral reference dose (mg/kg/day), and AT is the averaging time for non-carcinogens (365 days/year × number of exposure years). The oral reference doses were based on 1.5, 0.02, 0.04, 0.0003, 0.0005, and 0.0035 mg/kg/day for Cr, Ni, Cu, As, Cd, and Pb, respectively (USEPA 2007, 2010, 2015). If the THQ is equal to or higher than 1.00, there is a potential health risk and related interventions and protective measurements should be taken (Wang et al. 2005).

Hazard index (HI)

In order to assess the overall potential for non-carcinogenic effects from more than one heavy metal, a hazard index (HI) was formulated based on the guidelines for health risk assessment of chemical mixtures of USEPA (USEPA 1999). The hazard index (HI) from THQs is expressed as the sum of the hazard quotients (USEPA 2010). If the hazard index is less than one, there is no significant risk of the non-cancer effect. If the hazard index is more than one, then there is a chance that non-cancer effects may occur (MohseniBandpi et al. 2018; Jafari et al. 2018). The equation used for estimating the hazard index is as follows:

$$HI = \Sigma TTHQ \tag{4}$$

$$= \text{TTHQ food}_1 + \text{TTHQ food}_2 + \ldots\ldots\ldots + \text{TTHQ food}_n \quad (5)$$

$$\text{TTHQ(Individual food)} = \text{THQ toxicant}_1 + \text{THQ toxicant}_2 + \ldots\ldots + \text{THQ toxicant}_n \quad (6)$$

Carcinogenic risks

The target carcinogenic risks derived from the intake of As and Pb were calculated using the equation provided in USEPA Region III Risk-Based Concentration Table (USEPA 2006).

$$\text{TR} = \{(\text{EFr} \times \text{ED} \times \text{FIR} \times \text{C} \times \text{CSFo})/(\text{BW} \times \text{AT})\} \times 10^{-3} \quad (7)$$

EFr is the exposure frequency (350 days/year), ED is the exposure duration (30 years) (USEPA 2006) and AT is the averaging time for carcinogens (365 days/year × 70 years). CSFo is the oral carcinogenic slope factor from the Integrated Risk Information System (USEPA 2010) database were 1.5 and 8.5×10^{-3} $(\text{mg/kg/day})^{-1}$ for As and Pb, respectively.

Statistical analysis

The data were statistically analyzed using the statistical package SPSS 20.0 (International Business Machines Corporation [IBM] Armonk, NY, USA), and calculations of means, standard deviations and health risk indices were done using Microsoft Excel 2013. Multivariate methods in terms of Pearson's bivariate correlation matrix and principal component analysis (PCA) were used to evaluate the inter-element relationship and also to interpret the potential sources of toxic metals in rice (Islam et al. 2016; Proshad et al. 2019a). The extraction method was performed to find out the principal components (PC) in PCA analysis that was Eigen values. For dividing the toxic metals into several groups, cluster analysis (CA) with dendrogram using Ward's method was adopted by using the overall metals concentrations in rice samples (Islam et al. 2016; Proshad et al. 2019a). The CA was also used to obtain the detailed information of the dataset and to gain insight into the distribution of toxic metals by detecting similarities or differences in toxic metals in rice samples (Proshad et al. 2019a).

Results and discussion

Toxic metal concentrations in rice grain samples

The concentrations of Cr, Ni, Cu, As, Cd, and Pb (mg/kg) were determined in rice grain samples of Tangail district, Bangladesh and presented in Table 1. In rice grain samples, Cr, Ni, Cu, As, Cd, and Pb concentrations (mg/kg fw) varied considerably and the studied samples showed a decreasing trend of Cr > Pb > Ni > As > Cu > Cd. It is clear from the data presented in Table 1 that the toxic metal contaminations in rice samples from ten locations of the study area were not uniform. According to several researchers (Alam et al. 2003; Santos et al. 2004; Pandey and Pandey 2009; Saha and Zaman 2013; Garg et al. 2014), variability in metal concentrations was found among the different sampling sites because of natural characteristics, extent of anthropogenic activities, and levels of contamination. The highest concentration of metals was found in S7 site which might be due to the presence of bed rock in that site (Shah et al. 2010) with high levels of heavy metals content as compared to rocks of other sites. The use of different agricultural applications such as manure, fertilizer and pesticide; and extensive use of wastewater for irrigation also brings changes in the level of heavy metals in soils which ultimately transfer and affect the metal levels of cultivated rice samples (Islam et al. 2016). Following the standard values set by FAO/WHO for food samples (FAO/WHO 2011), metal concentration studied from rice grain samples in this study were compared (Table 1). Samples collected from 10 study sites indicated Cr, As, Cd, and Pb for all samples and Ni in samples from one study site exceeded the standard permissible limit with the exception of Cu which was lower than the standard value. In Table 1, six metal concentrations in

Table 1. Heavy metals (mg/kg) in rice collected from rice fields around industrial areas of Tangail district, Bangladesh.

Sites		Cr	Ni	Cu	As	Cd	Pb
S1	Range	8.46–13.0	0.88–8.12	9.33–31.76	0.78–1.19	0.05–0.2	0.42–6.39
	Mean±SD[a]	11.47 ±1.79	3.81 ±3.38	17.77 ±10.9	0.96 ±0.21	0.11 ±0.07	2.8 ±2.52
S2	Range	2.48–20.47	1.62–20.75	2.51–25.68	1.09–2.95	0.11–0.19	1.68–10.18
	Mean±SD[a]	14.38 ±7.33	13.23 ±8.31	16.84 ±8.71	1.72 ±0.75	0.15 ±.63	5.56 ±3.41
S3	Range	9.11–9.84	3.0–4.99	18.89–41.04	0.72–2.26	0.06–0.16	2.45–6.39
	Mean±SD[a]	9.29 ±0.31	3.9 ±0.91	28.75 ±9.46	1.28 ±0.74	0.13 ±0.05	5.51 ±1.7
S4	Range	2.77–19.65	3.27–8.58	4.72–35.15	1.21–2.46	0.11–0.18	1.59–6.15
	Mean±SD[a]	12.87 ±6.86	6.36 ±1.95	26.67 ±12.53	1.82 ±0.56	0.15 ±.05	3.41 ±1.67
S5	Range	7.82–12.3	2.24–7.89	18.74–33.25	0.99–1.85	0.07–0.37	1.18–8.71
	Mean±SD[a]	9.44 ±1.72	5.75 ±2.47	26.01 ±7.02	1.34 ±0.46	0.24 ±0.12	4.04 ±3.68
S6	Range	0.47–15.82	1.77–16.05	1.83–30.93	1.51–2.04	0.05–0.24	0.35–8.08
	Mean±SD[a]	8.49 ±5.85	8.75 ±5.28	15.54 ±10.82	1.72 ±0.19	0.19 ±0.1	4.85 ±3.07
S7	Range	20.06–26.39	5.94–6.85	34.72–47.69	1.62–2.49	0.05–0.15	8.15–10.22
	Mean±SD[a]	22.46 ±2.72	6.30 ±0.34	38.47 ±5.46	2.05 ±0.31	0.11 ±0.04	9.11 ±0.91
S8	Range	2.56–32.49	2.86–7.73	1.4–36.38	1.68–2.29	0.11–0.18	0.79–18.05
	Mean±SD[a]	21.22 ±13.36	6.27 ±1.99	23.86 ±13.42	1.9 ±0.28	0.15 ±0.23	11.92 ±7.1
S9	Range	3.43–21.37	3.03–5.67	1.46–16.46	3.23–6.05	0.02–0.24	0.63–11.29
	Mean±SD[a]	10.73 ±7.48	3.92 ±1.06	7.6 ±5.85	4.66 ±1.01	0.12 ±0.01	8.7 ±0.26
S10	Range	1.65–12.98	2.72–6.25	1.54–6.21	3.23–5.56	0.04–0.34	0.75–9.7
	Mean±SD[a]	7.12 ±4.03	4.28 ±1.55	3.92 ±168	4.09 ±1.03	0.14±0.11	5.08 ±0.38
Permissible levels as per (FAO/WHO 2011)		2.3	10	40	0.1	0.05	0.1
Maximum allowable concentration (MAC) (mg/kg fw)		1.0[b]	0.5 [c]	20[b]	0.1 [c]	0.4 [c]	0.2 [c]

[a]SD = standard deviation. [b](FAO/WHO 2002b) [c](JECFA 2005)

collected samples which exceeded permissible limits are presented. Cr, Ni, Cu, As, Cd, and Pb exceeded at 100%, 10%, 0%, 100%, 100%, and 100%, respectively, posing significant threat to human health. Jafari et al. (2018) reported the average concentration of Cd, Pb, As, Cu, Cr and Ni for Iranian grown rice samples as 0.16, 0.196, 0.046, 0.29, 0.22 and 16 mg/kg, respectively, whereas 0.13, 0.55, 0.057, 0.61, 0.76 and 2.08 mg/kg, respectively, where reported for imported rice.

The mean concentration of Cr ranged from 7.12 (S10) to 22.46 mg/kg (S7) among all samples collected from ten sampling sites (Table 1). The mean concentration of Cr in rice grain samples collected from the sampling sites in descending order was S7 > S8 > S2 > S4 > S1 > S9 > S5 > S3 > S6 > S10. The Cr concentration for all studied samples exceeded the standard value (2.3 mg/kg for food) (FAO/WHO 2011). This denotes that wide chromium contamination of rice grain might be due to the effects of different industrial effluents discharged into the study area without proper treatment (Bhuiyan et al. 2011; Rahman et al. 2013). The mean Cr concentration measured from each study site in this study was higher than the Cr concentrations in rice samples collected from Mymensingh, Gazipur and Faridpur districts (Jahiruddin et al. 2017) and Bogra district (Islam et al. 2015) of Bangladesh. The Cr concentrations in the present study were compared with other studies conducted by several researchers (e.g. Ahmad and Goni 2010; Li et al. 2012; Rahman et al. 2013; Islam et al. 2014) and also found different concentrations of Cr in food samples though the values were comparatively less than the reported value in this study. Rahman et al. (2014) reported that the mean Cr concentration in collected rice samples was 0.19, 0.14, 0.079, 0.41 and 0.07 mg/kg in Indian rice, Australian rice, Pakistani rice, Thailand rice and Vietnam rice, respectively. All of these values found by Rahman et al. (2014) were less than the reported Cr concentrations in rice grain of the present study. The main sources of increased Cr concentration in Bangladesh soils are the application of agro-chemical, untreated or poorly treated industrial effluents, and open dumping of industrial wastes (Islam et al. 2009; Bhuiyan et al. 2011). The WHO has listed Cr as a carcinogenic toxic metal in human beings (WHO 1996a). Higher exposure to Cr might pose a threat to human health through its toxic, genotoxic and carcinogenic effects (Stanin and Pirnie 2004).

Nickel (Ni) is also considered as toxic heavy metal which can cause cancer and many other health complications in humans (Denkhaus and Salnikow 2002). In this study, the mean concentration of Ni in rice samples ranged from 3.81 (S1) to 13.23 mg/kg (S2) (Table 1). The Ni concentration in all samples collected from the sampling sites followed this descending order of S2 > S6 > S4 > S7 > S8 > S5 > S10 > S9 > S3 > S1. Among all sampling sites, only Ni concentration in the sample collected from one site (S2) exceeded the FAO/WHO standard value of 10 mg/kg (FAO/WHO 2011), indicating possible contamination of rice grains. It might be due to Ni containing poorly treated or untreated industrial effluents to surrounding soils (Proshad et al. 2019a). The Ni concentration in rice grain samples of the present study was higher than the other studies conducted to assess the metal concentrations in foodstuffs (Kachenko and Singh 2006; Ahmad and Goni 2010; Li et al. 2012; Rahman et al. 2013; Islam et al. 2014, 2016). In this study, Ni concentration was comparatively higher than the study of Shraim (2017) who reported that 93% contained no detectable Ni among 70 samples collected from KSA, while another five samples contained low Ni concentrations with a mean of 0.064 mg/kg. On the other hand, relatively higher Ni levels were detected in Australian samples (n = 12) with a mean of 1.83 mg/kg, while no Ni was observed in almost all the rice samples collected from Bangladesh (Shraim 2017). The Ni and its compounds are widely used as catalysts and pigments in various metallurgical, chemical and food processing industries as well as for the production of Ni alloys with high corrosion and temperature resistance (Cempel and Nikel 2006). Higher levels of Ni found in this study might be attributed to the presence and operation of such industries near the sampling sites of Tangail district of Bangladesh.

On the other hand, copper (Cu) may cause liver and kidney damage in case of excess consumption (Tuzen 2009). The present study found that the mean concentration of Cu ranged from 3.92 (S10) to 38.47 mg/kg (S7) (see Table 1). The mean Cu concentration of all samples followed the descending order of S7 > S3 > S4 > S5 > S8 > S1 > S2 > S6 > S9 > S10 in the study area. Average Cu concentration in rice grain samples in the present study was higher than the values reported by Islam et al. (2015) (mean: 1.985 mg/kg) and Rahman et al. (2014) (mean: 1.6 mg/kg). Hang et al. (2009) reported the mean Cu concentration in rice grain samples as 3.84 mg/kg at Changshu City, China. Cao et al. (2010) found the average Cu concentration in rice grain as 2.64 mg/kg at the Southern Jiangsu, China which is less than the present study. In a recent study at Kolkata, India by Avijit and Anindya (2018) it was found that the mean Cu concentration in rice was 2.78 mg/kg which is also lower than in this study. According to the FAO/WHO (2011) standard value (40.0 mg/kg for rice), Cu concentration was under the permissible limit for all samples collected from ten sampling sites; indicating no probable risk of contamination of rice by Cu. However, according to the FAO/WHO, the maximum allowable concentration (MAC) of Cu (20.0 mg/kg), in 50% of the collected samples had exceeded the maximum allowable limit (FAO/WHO 2002a).

Arsenic (As) is known as a globally important environmental toxicant (Das et al. 2004). It can be found everywhere in the environment and chronic arsenic poisoning might lead to keratosis, hypertension, cardiovascular diseases and diabetes (Ng et al. 2003). Among all sampling sites, the average concentration of As in collected samples was in the order of S9 (4.66) > S10 (4.09) > S7 (2.05) > S8 (1.9) >S4 (1.82) >S6 (1.72) > S2 (1.71) > S5 (1.34) > S3 (1.28) > S1 (0.96) (Table 1). In Bangladesh, the occurrence of As in rice and other foods depends on the growing area because water/soil content widely varies among different parts of Bangladesh. Presently, in Bangladesh, there is no clear regulation about As levels in foodstuffs. The permissible level of As is only available for drinking water (50 μg/l) in Bangladesh. Aziz et al. (2015) conducted a research at Sonargaon, Bangladesh and found 2.55 mg/kg total As in brown rice grain samples. It means that 0.85–1.276 mg of inorganic As was present per kg rice (assuming about 33–50% of total As is inorganic in rice grain sample (Meharg et al. 2008). It shows an agreement with the present study except two sampling sites that had higher mean concentrations of As (4.66 and 4.09 mg/kg at S9 at S10, respectively). Compared with the Chinese permissible limit set by USDA (0.15 mg/kg inorganic As in foods) and FAO/WHO standard value (0.1 mg/kg As in rice), rice grain samples in the present study exceeded the limit which can lead to elevated exposure to the chronic carcinogen in human beings (USDA 2006; FAO/WHO 2011). The present study showed a strong disagreement with the previous studies conducted by Das et al. (2004) and Williams et al. (2005) where they found that mean arsenic values in rice grain samples were 0.14 and 0.13mg/kg, respectively. In Bangladeshi rice, Islam et al. (2015) found average As value of 0.321 mg/kg which is also very low compared to the present study. Islam et al. (2015) mentioned that increased accumulation of As in grain in the anaerobic paddy-soil systems from soil or uncontrolled use of As-enriched agrochemicals may contribute to this variation. The transfer of As from soil to plant during crop production by excessive use of agrochemicals and/or As contaminated groundwater as irrigation for rice production (Alam et al. 2003; Neumann et al. 2010, 2011; Bhuiyan et al. 2011; Polizzotto et al. 2013) might also be the causal factor behind this variation in this study.

Like As, Cadmium (Cd) occurs naturally and exists at low levels in the environment. It is highly toxic even at extremely low level of exposure. Rahman et al. (2014) reported air, water, and food are the major sources of cadmium exposure. Long-term Cd exposure may be associated with bone defects, high blood pressure, and myocardial dysfunctions (Duruibe et al. 2007). The present study found that the mean Cd concentration (mg/kg) was in order of 0.24 (S5) > 0.19 (S6) > 0.16 (S2) > 0.15 (S8) > 0.14 (S4) > 0.13 (S3) > 0.13 (S10) > 0.12 (S9) > 0.11 (S7) > 0.1(S1). According to the standard value of Cd concentration in rice (0.05 mg/kg) set by FAO/WHO (2011), this study exceeded the permissible level for 100% of the samples indicating higher contamination by Cd. The transfer of higher concentration of Cd from soil to rice might be due to small scale industries such as dyeing, electroplating, fabrics printing, batteries and paints, which discharge their effluents directly into the surface water used for irrigation purposes (Khan et al. 2015, 2017). Cadmium has easy availability and solubility in soils and hence accumulates in the edible parts of the plants (Luo et al. 2011). Previous studies found that the Cd concentration in rice was 0.088 mg/kg (Rahman et al. 2014) and 0.073mg/kg (Ahmed et al. 2015) in Bangladeshi rice. The average concentration of Cd in the present study was noted to be higher than the studies previously conducted by Rahman et al. (2014) and Ahmed et al. (2015). Zhang (2009) conducted a study at four mine sites in South China and found 0.5, 0.7, 1.2, 1.6 mg/kg mean Cd concentration which was higher than this study. Singh et al. (2011) conducted a research at Gorakhpur, UP, India and found that Cd concentration was 0.014 mg/kg at rice grains which was lower than the present study and the FAO/WHO standard value (WHO 1996a; Singh et al. 2011).

Lead (Pb) is absorbed in inorganic forms and is treated as the most significant toxic metal. Direct ingestion of food and water and inhalation are the ways of Pb exposure. Lead poisoning causes kidney dysfunction, reproductive system disorder, acute and chronic damage to the central nervous system, peripheral nervous system, and damage to the urinary tract. Lead also affects sound brain development of children and it leads to poor intelligence quotient (IQ) (Duruibe et al. 2007). Mean lead concentrations in rice grain samples at ten study sites were in the decreasing order of S8 > S7 > S9 > S2 > S3 > S10 > S6 > S5 > S4 > S1. According to FAO/WHO (2011) the standard value is 0.1 mg/kg in rice and 100% of the samples exceeded the allowable limit. Mean Pb concentration in the present study was much higher than the values of 0.713 and 0.019 mg/kg found by Islam et al. (2015) and Rahman et al. (2014), respectively. This study showed a wide similarity with the study findings conducted by Avijit and Anindya (2018) regarding Pb concentration (5.67 mg/kg). At Ismailia City, Egypt, Loutfy et al. (2012) found lower mean Pb concentration (0.094 mg/kg) than the present study. Singh et al. (2011) estimated 0.54 mg/kg of average Pb concentration in rice which was also lower than this study. The reason behind the elevated Pb concentrations in samples of this study might be due to the Pb smelting industry or related activity in the study area (Proshad et al. 2019a).

Source analysis of toxic metals in rice grains

Inter-relationships among the measured metal concentrations in the rice samples were investigated in terms of Pearson's correlation coefficient matrix which is shown in Table 2. Inter-metal interactions may illustrate the sources and pathways of the metals present in rice grains. A clear pattern of strong association was found among metal pairs in rice grains. In rice samples, Cr showed strong

Table 2. Correlation coefficient matrix of heavy metals in rice collected from industrial vicinity of Tangail district, Bangladesh.

Metals	Cr	Ni	Cu	As	Cd	Pb
Cr	1					
Ni	0.419**	1				
Cu	0.494**	0.271	1			
As	−0.115	−0.045	−0.371**	1		
Cd	−0.025	0.099	0.327*	−0.245	1	
Pb	0.734**	0.408**	0.619**	−0.265	.106	1

* = Correlation is significant at the 0.05 level (two-tailed)
** = Correlation is significant at the 0.01 level (two-tailed)

significant positive correlations with Ni (r = 0.419**), Cu (r = 0.494**), and Pb (r = 0.734**) whereas Ni showed significant associations with Pb (r = 0.408**). The value of Cu showed significant positive correlation with Pb (r = 0.619**) and negative correlation with As (r = −0.371**). Arsenic, cadmium, and lead did not show any significant positive or negative correlations. Higher correlation coefficients between the metals indicated common sources, mutual dependence, and similar or nearly identical metal accumulation properties in rice (Abbasi et al. 2013; Xu et al. 2013; Proshad et al. 2019b).

A principal component analysis (PCA) was performed following the standard procedure stated in the literature (Franco-Uría et al. 2009; Kikuchi et al. 2009), to identify the hypothetical sources of heavy metals (natural or anthropogenic) in rice for this present study. The PCA was performed on the tabular and dimensionless standardized form of the data set and presented in Table 3 and Figure 2. Two principal components (PC) were obtained and the variances explained by them were 74.99% and 16.84% for present study. Overall, the PCA revealed two major groups of the studied six metals in rice. PC1 was strongly correlated with Cr, Ni, As, Pb, and PC2 was also strongly correlated with

Table 3. Total variance explained and component matrix for the heavy metals in rice collected from Tangail district, Bangladesh.

	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
Component	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %
1	198.05	74.99	74.99	198.05	74.99	74.99	103.44	39.17	39.17
2	44.49	16.84	91.84	44.49	16.84	91.84	139.09	52.67	91.84
3	13.93	5.27	97.11						
4	6.19	2.34	99.46						
5	1.41	0.535	99.99						
6	0.007	0.003	100						

	Component matrix				Rotated Component Matrix			
Elements	Raw component		Rescaled component		Raw component		Rescaled component	
	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2
Cr	12.61		0.97		7.45		0.981	
Ni	3.21	1.78	0.73	0.41	3.39	1.41	0.781	0.32
Cu	−0.45		−0.35		2.31		0.543	
As					5.56	11.68	0.430	0.90
Cd	5.08	5.48	0.66	0.72		0.03		0.38
Pb	1.56	1.70	0.36	0.40		−0.48		−0.37
KMO and Bartlett's Test of Sphericity								
Kaiser-Meyer-Olkin Measure of Sampling Adequacy.								0.716
Bartlett's Test of Sphericity				Approx. Chi-Square				85.49
				df				15
				Significance				0.00

Extraction Method: Principal Component Analysis.

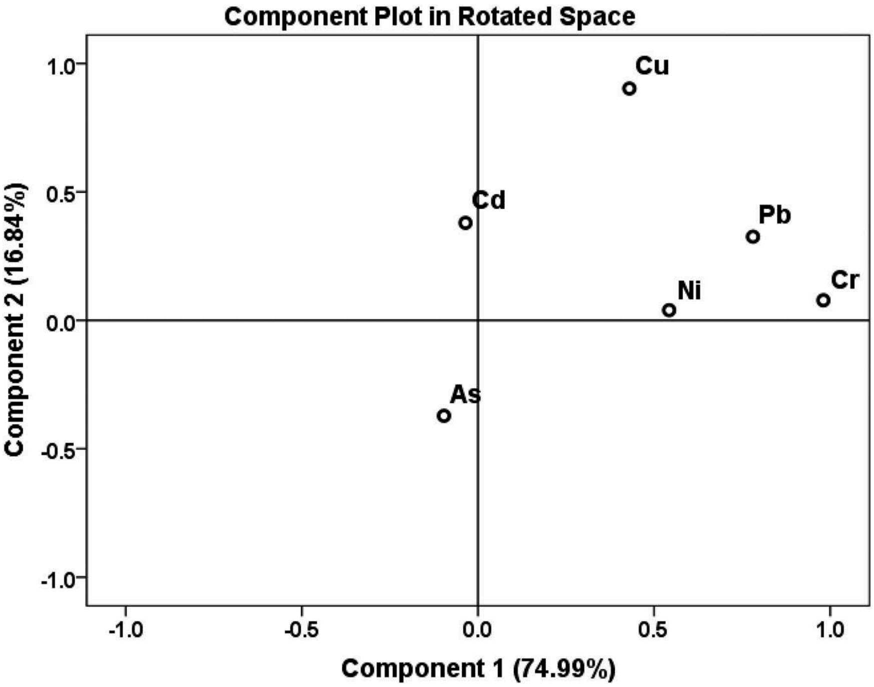

Figure 2. Principal component analysis (PCA) of heavy metals in rice samples collected from different agricultural fields of Tangail district, Bangladesh.

Cu and Cd. The source of PC1 and PC2 can be considered as a mixed source of anthropogenic inputs, particularly from industrial effluents and agricultural activities in the study area. The depositions of atmospheric particulates released by automobile emissions as well as coal and fuel combustion, and municipal waste disposal were believed to contribute these metals in the urban areas, from where the rice samples were collected (Islam et al. 2017). Due to emissions of heavy metals in the environment and accumulation by the plants, PCA analysis revealed that the apportionment of the same kind of heavy metals in rice were not similar.

Furthermore, using heavy metals concentration in rice samples, cluster analysis (CA) with dendrogram using Ward’s Method was adopted to divide the heavy metals into several groups as shown in Figure 3. Several cluster shapes were found between the studied metals and the metals which were in same cluster result for similar resemblance in nature. Again, toxic metal concentrations in rice showed strong significant correlations by forming primary clusters with each other (Figure 3). The primary clusters of As, Cd, Ni, and Pb were formed within a distance of five points on the scale (Figure 3).

Transfer factor (TF) of toxic metals from soil to rice

The transfer factor (TF) indicates the ability of the plant to translocate ionic metals from the roots to the aerial parts (Olguín and Sánchez-Galván 2012). Calculating TF is an appropriate method to assess the heavy metal level in plant tissues as a fraction of the metal concentrations in soils. TF was found to be different from one metal to another and from one plant species to another (Proshad et al. 2019a). Metals with high TF are more easily transferred from soil to the edible parts of plants than the ones with low TF. Heavy metal transfer from soil to rice is presented in Table 4 and Figure 4. Variations in the TF values were observed for different metals. The variations found might result from the plant physiological conditions in which absorption

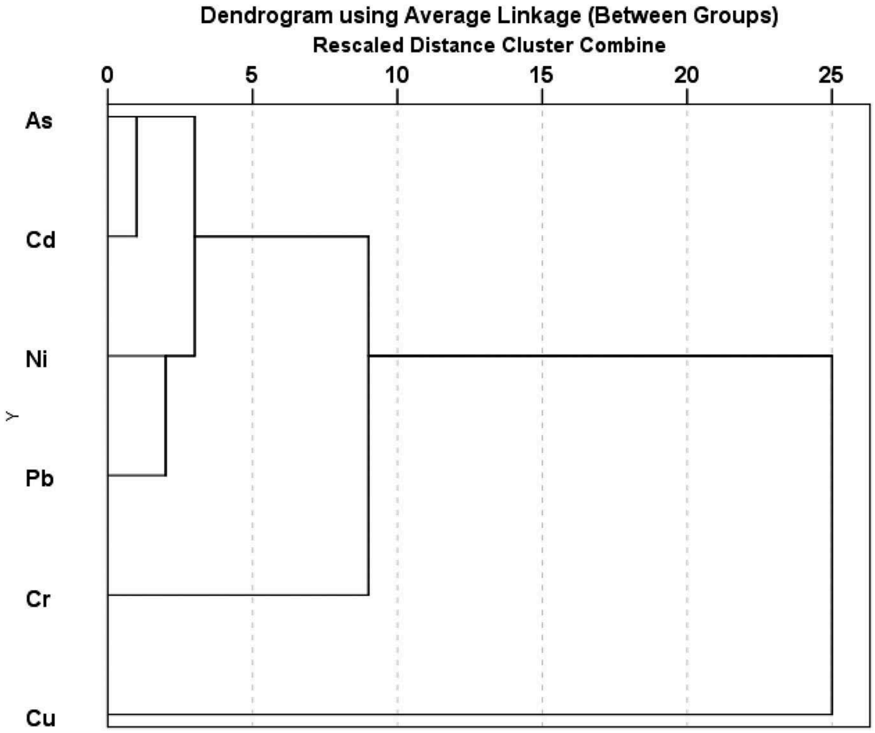

Figure 3. Cluster analysis of heavy metals in rice samples collected from Tangail district, Bangladesh.

Table 4. Transfer factor (TF) of heavy metals collected from industrial areas of Tangail district, Bangladesh.

Sampling sites	Cr	Ni	Cu	As	Cd	Pb
S1	0.79	0.44	1.27	0.08	0.04	0.46
S2	2.33	0.71	1.15	0.16	0.04	0.48
S3	0.92	0.25	1.10	0.10	0.04	0.77
S4	2.84	0.53	1.67	0.43	0.12	0.36
S5	0.28	0.57	3.05	0.72	0.48	0.86
S6	1.00	0.28	0.52	0.33	0.03	0.15
S7	1.65	0.10	0.65	0.36	0.08	0.32
S8	1.78	0.25	0.48	0.22	0.04	0.83
S9	1.13	0.33	0.28	0.82	0.09	0.56
S10	1.26	1.10	0.36	1.04	0.12	0.32
Mean±SD	1.40 ±0.76	0.46 ±0.28	1.05 ±0.83	0.42 ±0.32	0.11 ±0.13	0.51 ±0.23

depends on the concentration of metals in the soil and the plant physiological demand (Millaleo et al. 2010). It was observed in the present study that transfer of Cr and Cu from soils was higher in rice than among the other metals. Chromium and Cu accumulate preferentially over other metal ions in rice, so they can be regarded as a hyper accumulators (Liu et al. 2016). This may be the result of the high mobility of Cr and Cu from soil to rice plants. The highest average TF value for Cr and Cu was found to be 1.40 and 1.05, respectively, in the present study. Cadmium showed the lowest TF as Cd in soil does not have any direct influence on concentrations in plants, but it may instigate its concentration in the roots (Cannata et al. 2013). Other studied metals like Ni, As and Pb transferred lower amounts of metals from soil to plant due to the formation of stable complexes with amino acids (Mengel et al. 2001). Again, transfer factor depends on soil physical-chemical properties, especially on soil pH (Li et al. 2011). The TF values of these studied heavy

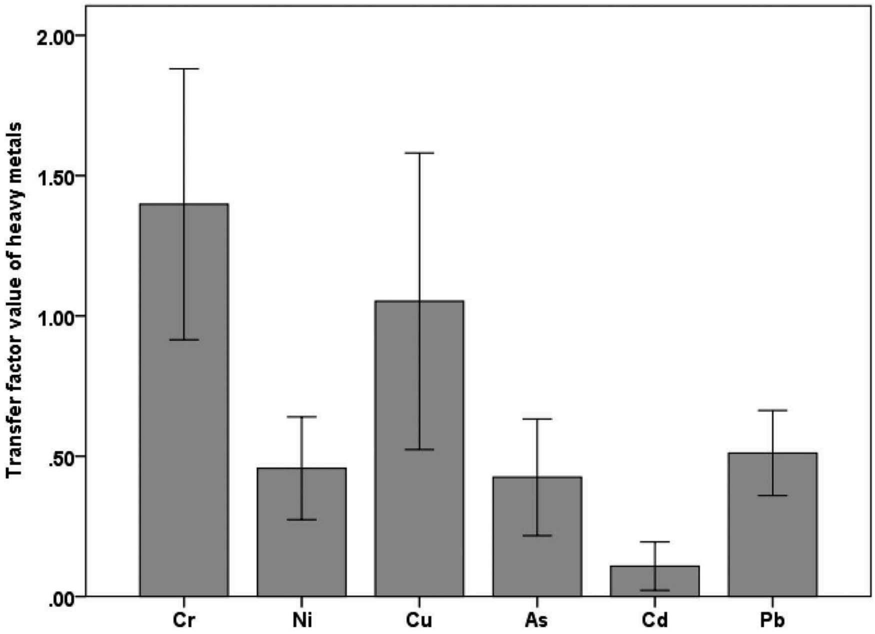

Figure 4. Transfer factor (TF) of heavy metals from soil to rice in present study area of Tangail district, Bangladesh.

metals were found different in the present study. This might be the result of the mobility and transfer behavior of metals from soil to rice grains as well as soil physico-chemical properties (Proshad et al. 2019a). Islam et al. (2016) also reported that the bioaccumulation and mobility of heavy metals from soil to rice is restrained by soil pH.

Assessment of health risk

Estimated daily intakes (EDI) of toxic metals from rice

According to the FAO/WHO (1985), the dietary intake of food is an effective tool to investigate nutrients, bioactive compounds, and contaminants in a population's diet and provide important

Table 5. Estimated daily intake (mg/kg bw/day) of heavy metals from rice for adult and children of Tangail district, Bangladesh with the corresponding maximum tolerable daily intake (MTDI).

	Estimated daily intake (EDI) (mg/day)											
	Cr		Ni		Cu		As		Cd		Pb	
tab	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children
S1	0.89	0.38	1.13	0.13	0.73	0.56	0.27	0.13	0.18	0.37	0.21	0.13
S2	0.33	0.25	1.2	0.44	0.82	0.46	0.31	0.16	0.37	0.25	0.32	0.22
S3	0.47	0.31	0.53	0.13	0.52	0.7	0.49	0.24	0.22	0.14	0.41	0.18
S4	0.3	0.14	0.65	0.21	0.4	0.39	0.11	0.26	0.18	0.28	0.53	0.11
S5	0.37	0.31	0.24	0.19	0.59	0.29	0.4	0.14	0.71	0.26	0.23	0.13
S6	0.56	0.28	0.65	0.23	0.42	0.45	0.21	0.57	0.18	0.37	0.36	0.16
S7	0.27	0.27	0.15	0.12	0.29	0.23	0.15	0.37	0.17	0.24	0.48	0.3
S8	0.42	0.17	0.47	0.22	0.38	0.38	0.14	0.32	0.11	0.25	0.27	0.4
S9	0.18	0.36	0.91	0.13	0.36	0.25	0.53	0.16	0.19	0.14	0.65	0.29
S10	0.25	0.24	0.74	0.24	0.29	0.41	0.3	0.14	0.51	0.18	0.38	0.17
Daily intake via food consumption	4.03	2.72	6.66	2.05	4.79	4.11	2.94	2.49	2.82	2.48	3.83	2.1
MTDI	0.2[a]		0.3[b]		30[c]		0.126[c]		0.046[c]		0.21[c]	

[a](RDA 1989), [b](WHO 1996b), [c](JECFA 2003), MTDI =Maximum tolerable daily intake

information about nutritional deficiencies or exposure of contaminants in food. The EDIs of six heavy metals (Cr, Ni, Cu, As, Cd, and Pb) were estimated according to mean concentrations of each metal found in rice samples and the respective consumption rates of adults and children are presented in Table 5. Total daily intakes of Cr, Ni, Cu, As, Cd, and Pb for adult inhabitants were 4.03, 6.66, 4.79, 2.94, 2.82, and 3.83 mg/day, respectively (Table 5). For children, the total daily intake of Cr, Ni, Cu, As, Cd, and Pb were 2.72, 2.05, 4.11, 2.49, 2.48, and 2.1, respectively (Table 5). Total EDIs of Cr, Ni, As, Cd, and Pb from all samples were higher than the maximum tolerable daily intake (MTDI) (Table 5). Jolly et al. (2014) studied the probable health risk of heavy metals for adults via consumption of rice grown in eight districts of Bangladesh including Joypurhat, Naogaon, Satkhira, Khagrachori, Rangamati, Bandarban, Jessore and Dhaka (Savar). It was found that the EDIs of Cr, Ni, Cu, As, Cd, and Pb ranged from 0.008–0.109, 0.009–0.037, 0.025–0.052, 0–0.004, 0–0.007, and 0–0.016, respectively. Islam et al. (2016) studied the health risks through the consumption of contaminated vegetables in Bogra district and observed that the total EDIs of Cr, Ni, Cu, Zn, As, Cd, and Pb for adults were 1.36, 1.01, 3.72, 6.30, 0.52, 0.12, and 1.09, respectively. The findings of these previous studies indicate that the consumption of rice grown in Tangail district, as compared to the rice in other districts of Bangladesh, would increase the daily intake of toxic metals which may pose severe health hazards to the consumers. Jafari et al. (2019) revealed that the daily intake of toxic metals via oral ingestion was comparatively higher than other intake pathways like dermal contact and inhalation. This indicates that ingestion of contaminated foods is the primary pathway of toxic metal intake which therefore discourages to consume contaminated foods, especially rice, on the regular basis. It showed that these heavy metals present in rice samples had a major contribution to the potential health risk to consumers if they consume rice in the study area. Based on these data obtained from this study, we conclude that Cr, Ni, As, Cd, and Pb were the major elements contributing to the potential health risks via consumption of rice around the industrial areas at Tangail district in Bangladesh.

Non-carcinogenic and carcinogenic risk

The health risk assessment of each chemical or contaminant is normally based on determining the risk level and is categorized in terms of carcinogenic or non-carcinogenic health risks (Keramati et al. 2018; Kamarehie et al. 2019). Based on the target hazard quotients (THQs) and the target carcinogenic risk (TR), the non-carcinogenic and carcinogenic risks from consumption of studied rice grain samples by the adults and children were assessed. The estimation procedure of THQs offers an indication of the risk level due to contaminant exposure but it does not allow a quantitative estimation on the probability of an exposed population experiencing a reverse health effect. The THQ values of six heavy metals are presented in Table 6. Among all the studied heavy metals, the THQ value for individual metals from each sample were less than 1.00 and are considered safe for human consumption. Total THQ values for Cu, Ni, As, Cd, and Pb were greater than 1.00 (Table 6); thus the consumption of these rice samples were considered to be unsafe but their daily consumption was not recommended. The results concluded that the consumers are at a higher risk with respect to Cu, Ni, As, Cd, and Pb which can cause non-carcinogenic risks; while Cr will not cause a non-carcinogenic risk. Similar results were also found by Proshad et al. (2019a) who reported that Cr concentrations in rice and vegetable samples collected from Tangail Sadar upazila, Bangladesh did not pose a non-carcinogenic health risk though other toxic metals like Cu, Cd, As, Pb and Ni were found to pose potential health hazards. Islam et al. (2015) also reported that the THQ value for individual metals excluding Cu, As and Pb was less than 1.00; indicating that foods grown in Bogra district was considered to be unsafe and regular consumption might cause severe non-carcinogenic health hazards. However, the extent of metallic contamination was found to be less when compared with the values found in Tangail district. On the other hand, Jafari et al. (2019) observed that the HQ values of all heavy metals in all studied sampling points of Iran (Lorestan province) were less than 1.00 which means that agricultural crops produced in Bangladesh, especially in Tangail district, are considered to be unsafe as compared to other countries. The hazard index (HI) value

Table 6. Target hazard quotient (non-carcinogenic risk) of heavy metals from rice for adult and children of Tangail district, Bangladesh.

	Target hazard quotient (THQ)													
	Cr		Ni		Cu		As		Cd		Pb			
Sampling sites	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	TTHQ (Adult)	TTHQ (Children)
S1	0.02	0.038	0.61	0.953	1.43	2.221	10.29	16.000	0.71	1.100	2.57	4.000	15.64	24.312
S2	0.03	0.048	2.13	3.308	1.35	2.105	18.44	28.667	0.97	1.500	5.11	7.943	28.03	43.570
S3	0.02	0.031	0.63	0.975	2.31	3.594	13.72	21.333	0.84	1.300	5.06	7.871	22.58	35.104
S4	0.03	0.043	1.02	1.590	2.14	3.334	19.51	30.333	1.54	2.400	3.13	4.871	27.39	42.571
S5	0.02	0.031	0.92	1.438	2.09	3.251	14.37	22.333	1.22	1.900	3.71	5.771	22.34	34.725
S6	0.02	0.028	1.41	2.188	1.25	1.943	18.44	28.667	0.71	1.100	4.46	6.929	26.28	40.854
S7	0.05	0.075	1.01	1.575	3.09	4.809	21.98	34.167	0.71	1.100	8.37	13.01	35.22	54.740
S8	0.05	0.071	1.01	1.568	1.92	2.983	20.37	31.667	0.97	1.500	10.96	17.02	35.26	54.816
S9	0.02	0.036	0.63	0.980	0.61	0.950	49.97	77.667	0.77	1.200	8.00	12.49	60.00	93.261
S10	0.02	0.024	0.69	1.070	0.32	0.490	43.85	68.167	0.90	1.400	4.67	7.25	50.44	78.408
TTHQ	0.27	0.425	10.06	15.64	16.52	25.67	230.96	359.00	9.33	14.50	56.04	87.11		

Note. Bold indicates HQ > 1.

Table 7. Carcinogenic risks of As and Pb due to consumption of rice in Tangail district, Bangladesh.

	Target carcinogenic risk (TR)			
	[a]As		Pb	
Sampling sites	Adult	Children	Adult	Children
S1	8.8E-04	3.07E-03	1.45E-06	5.08E-06
S2	1.51E-03	5.50E-03	2.88E-06	1.01E-05
S3	1.17E-03	4.10E-03	2.86E-06	9.99E-06
S4	1.66E-03	5.82E-03	1.77E-06	6.18E-06
S5	1.23E-03	4.29E-03	2.09E-06	7.33E-06
S6	1.57E-03	5.50E-03	2.51E-06	8.80E-06
S7	1.87E-03	6.56E-03	4.72E-06	1.65E-05
S8	1.74E-03	6.08E-03	6.18E-06	2.16E-05
S9	4.26E-03	1.49E-02	4.51E-06	1.58E-05
S10	3.74E-03	1.30E-02	2.63E-06	9.21E-06
Total	1.97E-02	6.89E-02	3.16E-05	1.11E-04

[a]Assuming 50% inorganic As in foods (Saha and Zaman 2013)

denotes the summed up non-carcinogenic effects of multiple elements (Table 6). It was found that the maximum hazard index was at the S9 study site (60.00) and the lowest hazard index was at the S1 study site (15.64), for adults. The hazard index for adults were the following in decreasing order: S9 > S10 > S8 > S7 > S2 > S4 > S6 > S3 > S5 > S1. The highest hazard index for children was investigated at the S9 study site (93.261) and the lowest hazard index was at the S1 site (24.31).

The target carcinogenic risk (TR) values of studied heavy metals are presented in Table 7. The TR values for As in the studied rice grain samples were 8.8E-04 to 4.26E-03 for adults and 3.07E-03 to 1.49E-02 for children (see Table 7). The TR values for Pb in the studied food items were 1.45E-06 to 6.18E-06 and 5.08E-06 to 2.16E-05 for adult and children respectively (Table 7). The target carcinogenic risk (TR) of Pb from samples in the present study was more than the negligible value (10^{-6}). On the other hand, the risk for As from most of the rice grain samples was higher than the acceptable level of 10^{-4} (USEPA 1989, 2015). The present study clearly showed that consumption of rice definitely poses cancer risks to the population. Therefore, the potential carcinogenic risk, poses a threat to urban residents through food ingestion and should be considered. Several studies (e.g. Islam et al. 2014, 2017; Proshad et al. 2019a) also concluded that the toxic metals would definitely pose carcinogenic health risks to the inhabitants of Bangladesh via consumption of contaminated food.

Conclusions

In conclusion, the present study revealed that the concentrations of Cr, Ni, As, Cd, and Pb in rice samples exceeded the WHO and FAO permissible limits which could pose a potential health concern to local residents. The multivariate analysis showed that Cr, Ni, As, and Pb in rice samples were mostly contributed through industrial activities. The total estimated daily intake (EDI) values of all the metals except Cu were higher than the maximum tolerable daily intake (MTDI), indicating a considerable health risk to consumers. The total targeted hazard quotient (TTHQ) values of Cu, Ni, As, Cd, and Pb exceeded the threshold value of 1.00 which can lead to non-carcinogenic health risks. The estimated target carcinogenic risk of As was higher than the USEPA acceptable level of 0.001 (10^{-4}); exhibiting the higher probability of lifetime carcinogenic health risk to consumers. This study suggested that more attention should be directed to the the prevention and control of toxic metals contamination in rice production to mitigate their associated health risks in Bangladesh.

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Disclosure statement

The authors declare that there are no conflicts of interest.

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