RESEARCH ARTICLE

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HEAVY METALS CONCENTRATION IN VEGETABLES, FRUITS AND CEREALS AND ASSOCIATED HEALTH RISK OF HUMAN IN KHULNA, BANGLADESH

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SUMMARY

Vegetables, fruits, and cereals are the main sources of nutrients, playing a pivotal role in the prevention of several diseases. Unfortunately, these essential foods are polluted by heavy metals through natural and anthropogenic activities. Therefore, this research attempts to assess the human health risk by measuring the concentration of seven heavy metals (Fe, Mn, Ni, Cu, Zn, As and Pb) in some popular vegetables, fruits and cereals in Khulna, Bangladesh. United States Environmental Protection Agency (US-EPA) deterministic approaches were used to assess the carcinogenic and non-carcinogenic health risks. The concentration of Fe, Mn, Cu, Zn, and Pb were below the maximum tolerable limit proposed by the Joint FAO/WHO Expert Committee on Food Additives. Assessment of non-carcinogenic health hazards by Target Hazard Quotient (THQ) indicated health risk for As, Ni, and Pb. But the combined impacts of all metals, Hazard Index (HI), were higher than the acceptable limit (HI=1.0) for vegetables (4.0257) and cereals (12.6269). So, the consumption of vegetables and cereals are of the biggest concern for non-carcinogenic health effects. Target Cancer Risk (TCR) values of Pb and As exceeded the US-EPA threshold risk limit (10^{-6}) that revealed moderate to high cancer risks. The overall analysis of THQ, HI and TCR indicated that long term consumption of food species from this study area might pose health risks and therefore regular monitoring of heavy metals is strongly recommended.

***Keywords**:* Heavy metal, Health risk, Carcinogenic, Hazard Index, Target cancer risk

1. INTRODUCTION

Heavy metals are non-biodegradable, persistent and known to exert deleterious effects on human health (Javed and Usmani, 2011). Dietary intake of contaminated foods is the main route of heavy metal exposure (Calderon et al., 2003). Both acute and prolonged exposure of heavy metals is assumed to be responsible for damaging vital organs like the brain, kidney, and liver (Burger et al., 2007; Thomas et al., 2009; Jeftic et al., 2010; Johri et al., 2010, Dhar et al., 2019). An elevated amount of heavy metals can alter the gene expression (Jomova et al., 2011), affect the central nervous functions, (Bouchard et al., 2011).

Fresh vegetables, fruits, and cereals are of significance in the daily diet because they contain vitamins and minerals. Besides, they play a vital role in the maintenance of sound health and the prevention of various chronic diseases (Ara et al., 2018a). However, these foods contain different concentrations of essential and toxic elements. Among them, heavy metals are the major contaminants that affect the nutritive values of vegetables, fruits, and cereals. When food species are cultivated in a polluted environment, they could readily absorb heavy metals through the leaves or roots, leading to the accumulation of toxic metals in plant tissues (Singh et al, 2010). The entry of heavy metals into the food chain not only inhibits the normal physiological functions of the human body, but it also affects the growth, nutrient uptake, nitrogen fixation, and metabolism of plants (Ara et al., 2018a).

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As alertness about the risk of heavy metal pollution, national and international regulations on food quality have lowered the maximum acceptable levels of toxic heavy metals in food items (Radwan and Salama, 2006). The objectives of these regulations are to save natural resources and human health from the toxicity of these heavy metals. Assessment and estimation of heavy metals concentrations in the food items have been carried out in some developed and developing countries. Still now, there is insufficient data available about the commination status of heavy metals in vegetables, fruits, and cereals cultivated in different areas of Bangladesh.

Considering the detrimental impacts of heavy metals in foodstuff this research was carried out. This study, therefore, aimed to examine the concentrations of heavy metals (Fe, Mn, Ni, Cu, Zn, As and Pb) in some selected vegetables, fruits, and cereals; to assess the health risks (non-carcinogenic and carcinogenic) of inhabitants in Khulna, Bangladesh.

2. MATERIALS AND METHODS

2.1. Study area

The study area Khulna district is located at 22.35° North latitudes and 89.30° east longitude in south-western region of Bangladesh (Figure 1). It has an area of 4394.45 km² and is bordered on the north by the Jashore District and the Narail District, on the south by the Bay of Bengal, on the east by the Bagerhat District, and on the west by the Satkhira District (Mallik, 2012). Because of geographical location, huge cultivation, and household works, vegetables, fruits and cereals in this study area might be contaminated.

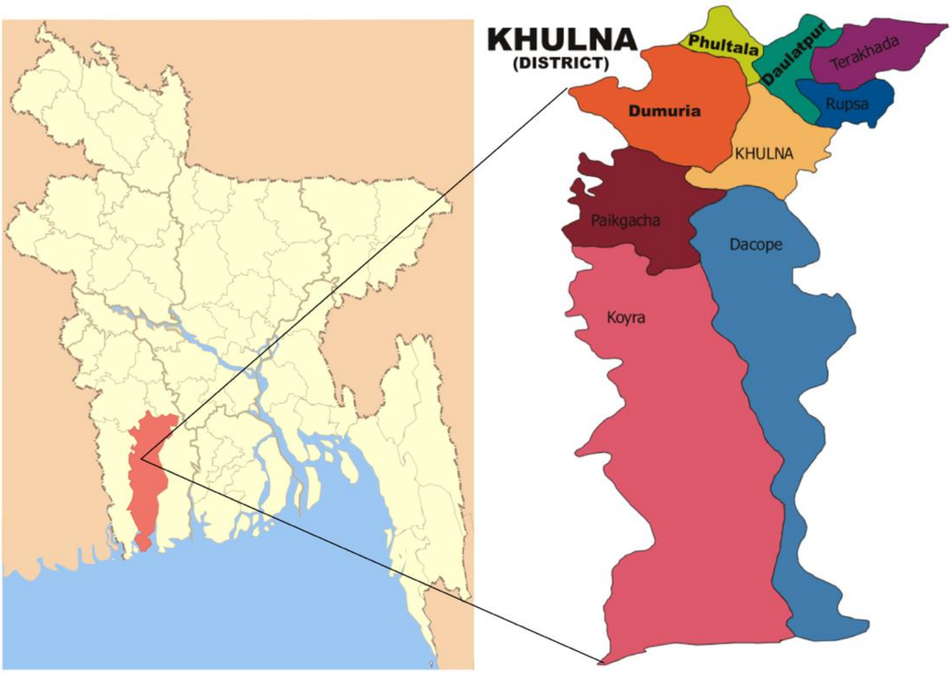

Fig.1. Map of the study area (Khulna, Bangladesh).

2.2. Sample collection and analysis

The selected vegetables, fruits and cereals samples (Table 1) were collected from Khulna, Bangladesh in 2017. To avoid any intermediate pollution, samples were collected from the study area field. During sampling, the study area was split into many sampling units, selected by planting mode and pollution background. The studied samples were collected along the same gradients. At each sample location, vegetable samples were collected from three different locations to provide replicate samples of each plant. Vegetable samples were divided into leaf, stem, and root. Only the edible parts were collected and all samples were stored in clean polythene bags and brought to the laboratory for analyses. The collected samples were washed with distilled water to remove the dust particles. Then samples were cut to small pieces using a clean knife and dried in an oven at 100°C. After drying the samples were ground into a fine powder using a commercial blender and stored in polyethylene bags until used for acid digestion.

About 15 ml of 5:1:1 tri-acid mixture (70% high purity HNO_3, 65% $HClO_4$ and 70% H_2SO_4) was added to the beaker containing 1 g dry sample (Allen et al., 1986). The mixture was subjected to digest at 80°C until the transparent solution appeared. After cooling, the digested samples were filtered using Whatman no. 42 filter paper and poured into the Teflon bottle. Finally, the filtrate was diluted to 50 ml with deionized water. Determination of Fe, Mn, Ni, Cu, Zn, As and Pb concentration in different digested samples were made directly using Shimadzu AA-7000 Atomic Absorption Spectrophotometer (AAS).

Table 1. General description of studied vegetables, fruits, and cereals.

Types of foods	English name	Scientific name	Local name
Vegetables	Cabbage	*Brassica capitata*	Bandhacopi
	Coriander leaf	*Coriandrum sativum*	Dhona pata
	Basella alba	*Basella alba*	Pui shak
	Carrot	*Daucus sativus*	Gajor
	Potato	*Citrullus lanatus*	Alu
	Radish	*Raphanus sativus*	Mula
	Cauliflower	*Brassica botrytis*	Fulcopi
Fruits	Mango	*Magnifera indica*	Amm
	Guava	*Psidium guajava*	Payara
	Jackfruit	*Artocarpus heterophyllus*	Kathal
	Banana	*Musa sapientum*	Kala
	Tamarind fruit	*Tamarindus indica*	Tetul
Cereals	Rice	*Oryza sativa*	Chaal
	Wheat	*Triticum aestivum*	Moyda

2.4. Health risk assessment

The values of heavy metals concentrations in vegetables, fruits, and cereals were used to calculate the estimated daily intake of metals (EDI), target hazard quotients (THQ), hazard index (HI) and target cancer risk (TCR) for study area peoples.

2.4.1. Estimated daily intake (EDI)

Studies have shown that the ingestion of contaminated foods is the main exposure pathway of heavy metals to humans (Chary and Kamala, 2008). Estimated daily intake (EDI) of heavy metals was calculated using the following equation:

$$\mathrm{EDI} = \frac{\mathrm{C_m} \times \mathrm{I_g}}{\mathrm{W_b}}$$

Where, C_m is the metal concentration in vegetables, fruits, and cereals ($mg.kg^{-1}$.dry weight). I_g is the ingestion rate, which is taken as 0.126 $kg.day^{-1}$ for vegetables, 0.029 $kg.day^{-1}$ for fruits and 0.400 $kg.day^{-1}$ for cereals from previous studies (Ali and Hau, 2001; Saha et al., 2012). W_b is an average body weight of Bangladeshi peoples which is taken as 49.5 kg (Ara et al., 2018a; Uddin et al., 2019).

2.4.2. Target hazard quotient (THQ):

THQ value was determined using the equation:

$$\mathrm{THQ} = \frac{\mathrm{C_m} \times \mathrm{I_g} \times \mathrm{E_f} \times \mathrm{D_e}}{\mathrm{D_f} \times \mathrm{W_b} \times \mathrm{T_{avncar}}}$$

Where, THQ is non-carcinogenic risk. E_f is the exposure frequency (365 days $year^{-1}$). D_e is the exposure duration (71.8 years). D_f is the reference dose of Fe, Mn, Cu, Zn, As, Ni, Pb are 0.7, 0.14, 0.04, 0.30, 0.0003, 0.02 and 0.0035 ($mg.kg^{-1}$ day^{-1}) respectively (USEPA, 1989; USEPA, 2011; USEPA, 2012). T_{avncar} is the average time for non-carcinogens (365 $days.year^{-1} \times D_e$) (USEPA, 2011).

2.4.3. Hazardous index (HI)

Hazard index (HI) is an estimation of the health risk posed by more than one heavy metal, which is the sum of the hazard quotients for all metals and calculated by the following equation (USEPA, 1989; Guerra et al., 2010).

$$HI = \sum HQ = THQ_{Fe} + THQ_{Mn} + THQ_{Cu} + THQ_{Zn} + THQ_{As} + THQ_{Ni} + THQ_{Pb}$$

2.4.4. Target cancer risk (TCR)

The Target cancer risk (TCR) was estimated using the given formula (USEPA, 2011):

$$TR = \frac{\mathrm{C_m} \times \mathrm{I_g} \times \mathrm{E_f} \times \mathrm{D_e} \times \mathrm{S_{cpo}}}{\mathrm{D_f} \times \mathrm{W_b} \times \mathrm{T_{avcar}}}$$

Where, S_{cpo} is the carcinogenic potency slope As and Pb (1.5 and 0.0085) in $mg.kg^{-1}$ body $weight.day^{-1}$. T_{avcar} is the averaging time for carcinogens (365 $days.year^{-1}$ × 71.8 years).

3. RESULTS AND DISCUSSION

The concentrations of heavy metals (Fe, Cu, Mn, Zn, As, Ni and Pb) in vegetables, fruits, and cereals from the different locations of Khulna are presented in Table 2. The decreasing order of the average concentration of heavy metals in different kinds of vegetables, fruits and cereals were Fe> Mn> Zn> Cu> Pb> Ni> As, Fe> Zn> Mn> Cu> Ni> Pb> As, and Fe> Mn> Zn> Cu> Ni> Pb> As respectively. The highest average concentration of Fe (97.05 $mg.kg^{-1}$), Cu (4.19 $mg.kg^{-1}$), Pb (2.69 $mg.kg^{-1}$) were observed in vegetable species whereas elevated amount of Mn (9.03 $mg.kg^{-1}$), Zn (8.67 $mg.kg^{-1}$), As (0.13 $mg.kg^{-1}$) and Ni (2.94 $mg.kg^{-1}$) were found in fruits species. Furthermore, the highest concentration of Fe was estimated in all types of studied samples. Fe often participates in photosynthesis and chlorophyll synthesis as a result; vegetables contain higher levels of Fe compared to fruits and cereals (Mahfuza et al., 2014). The observed concentrations of metals were compared with the maximum tolerable limit established by the joint FAO/WHO (FAO/WHO, 1989; FAO/WHO, 1993). The concentrations of Fe, Mn, Cu, Zn, and Pb were within the permissible limits while the concentrations of Ni and As were greater than the recommended values. Ni is the 22nd abundant element on earth crust (Hussain et al., 2013), and availability of Ni in the soil can be found as 0.2-450 $mg.kg^{-1}$ (Chen et al., 2009). So the plant can readily absorb Ni from the soil. Besides, the accumulation of higher Ni levels might be due to the excessive use of inorganic fertilizers like $NiSO_4$. Furthermore, Ni enters the environment through natural and anthropogenic activities and after circulating within the biosphere, just a part of Ni is accumulated by plants (Orts et al., 2007). "As" is another toxic metal that generally targets the pulmonary nervous system and responsible for chronic diseases (Dhar et al., 2019). The deposition of an excessive amount of As might be due to repetitive use of As contaminated water during irrigation.

Concentrations of heavy metals in cultivated vegetables vary depending on geographical location and environmental conditions. The obtained results of the study were compared with similar studies, and the concentrations of almost all heavy metals were lower than others (Table 3).

Table 2. Average concentrations of heavy metals ($mg.kg^{-1}$ dry weight) in vegetables, fruits, and cereals.

Sample ID	Fe	Mn	Cu	Zn	As	Ni	Pb
Vegetables	97.05 (8.59-165.06)	7.19 (1.13-20.02)	4.19 (0.56-9.13)	7.06 (2.19-22.11)	0.11 (0.005-0.17)	2.56 (0.21-6.24)	2.69 (0.15-4.24)
Fruits	52.13 (1.78-89.03)	5.25 (1.12-10.12)	2.93 (0.12-5.14)	6.14 (0.89-11.18)	0.09 (0.00-0.14)	1.78 (0.34-4.21)	1.56 (0.09-3.01)
Cereals	78.12 (3.34-112.87)	9.03 (3.13-16.13)	3.89 (2.12-6.19)	8.67 (1.29-19.13)	0.13 (0.06-0.16)	2.94 (0.70-6.73)	2.38 (0.89-5.99
Reference value	450.0^{a}	500.0^{a}	40.0^{a}	60.0^{a}	0.10^{a}	1.0^{a}	5.0^{b}

^{a}FAO/WHO, 1989; ^{b}FAO/WHO, 1993.

Table 3. Comparison of heavy metals ($mg.kg^{-1}$ dry weight) in studied samples with similar findings.

Location	Fe	Mn	Cu	Zn	As	Ni	Pb	References
Jashore, Bangladesh	190.32	33.297	9.31	28.23	-	-	5.45	Ara et al., 2018b
Satkhira, Bangladesh	356.71	33.91	10.27	33.59	-	-	9.76	Uddin et al., 2019
Mongla, Bangladesh	489.47	35.24	9.65	33.59	-	-	8.15	Ara et al., 2018a
Dhaka, Bangladesh	1160.0	--	17.63	-	-		11.4	Tasrina et al., 2015
Rajshahi, Bangladesh	-	4.54	-	-	-	-	5.1	Saha et al., 2012
Narayanganj, Bangladesh	-	-	9.37	19.76	-		3.69	Ratul et al., 2018
Misurata, Libya	-	-	3.36	8.15	-	0.24	0.25	Elbagermi et al., 2012
Varanasi, India	-	-	22.38	48.62	-	-	1.15	Sharma et al., 2009
Southeast, China	-	-	-	-	0.03	-	0.43	Liu et al., 2013
Khulna, Bangladesh	**97.05**	**7.19**	**4.19**	**7.06**	**0.11**	**2.56**	**2.69**	**Present Study**

The values of non-carcinogenic (THQ, HI) and carcinogenic (TCR) health risk parameters are shown in Table 4. These health risk assessment parameters were introduced by EPA in the United States to estimate the potential health risk caused by any chemical contaminant over prolonged exposure (US-EPA, 1989). According to the New York State Department of Health (NYSDOH, 2007), if the ratio of EDI/D_f is less than or equal to the D_f value of the corresponding heavy metal, the risk will be no risk. But if this ratio is 1–5 times of D_f then risk will be low, if >5–10 times the D_f then risk will be moderate, however, if >10 times the D_f then the risk will be high. In this study, the ratio EDI/D_f of As and Pb were observed several hundred times greater than their corresponding D_f values, which indicated potential health risk to the people.

To assess the non-carcinogenic health risk, the target hazard quotient (THQ) of individual heavy metals was estimated (Table 4) and the acceptable guideline value for THQ is 1.0 (US-EPA, 2011). According to Ambedkar and Maniyan if the THQ values exceed this limit then the metal might pose a non-carcinogenic health risk among the population (Ambedkar and Muniyan, 2011). In this study, the THQ values of Fe, Mn, Cu, and Zn were less than 1.0. On the other hand, the THQ of As (3.5017 in cereals), Ni (1.1879 in cereals) and Pb (1.9563 in vegetables and 5.4950 in cereals) exceeded this tolerable limit, which revealed potential non-carcinogenic health risk. Among all types of food species Pb should be paid more attention owing to the high non-carcinogenic health risks. Moreover, the combined impacts of all metals (HI) excessed the acceptable limit of 1.0 for vegetables and cereals. So, the consumption of these vegetables and cereals together is the biggest concern for non-carcinogenic health effects.

In this study, the probability of developing cancer by the ingestion of these food species was estimated (Table 4). According to New York State Department of Health, the TCR categories are described as, if TCR ≤ 10^{-6}, the risk is low; if TCR is 10^{-5} to 10^{-3}, it is associated with moderate health risk; if TCR is 10^{-3} to ≥10^{-1} then the risk is high (NYSDOH, 2007). In this study, As showed high cancer risk while Pb showed moderate cancer risk among the population.

Table 4. EDI, THQ, HI and TCR values of metals via consumption of selected vegetables, fruits, and cereals.

Parameters	Food species	Fe	Mn	Cu	Zn	As	Ni	Pb
EDI ($mg.kg^{-1}.day^{-1}$)	Vegetables	0.2470	0.0183	0.0107	0.0180	0.0003	0.0065	0.0069
	Fruits	0.0305	0.0031	0.0017	0.0036	0.0001	0.0010	0.0009
	Cereals	0.6313	0.0730	0.0314	0.0701	0.0011	0.0238	0.0192
THQ	Vegetables	0.3529	0.1307	0.2666	0.0599	0.9333	0.3258	1.9563
	Fruits	0.0436	0.0220	0.0429	0.0120	0.1758	0.0521	0.2611
	Cereals	0.9018	0.5212	0.7859	0.2335	3.5017	1.1879	5.4950
HI	Vegetables				4.0257			
	Fruits				0.6095			
	Cereals				12.6269			
TCR	Vegetables	--	--	--	--	$1.4E^{01}$	--	$1.7E^{-02}$
	Fruits	--	--	--	--	$6.5E^{-02}$	--	$3.6E^{-04}$
	Cereals	--	--	--	--	$1.4E^{01}$	--	$6.7E^{-03}$

4. CONCLUSION

According to this study, it can be concluded that the commonly consumed vegetables, fruits, and cereals collected from the Khulna district contain different concentrations of heavy metals and the degree of accumulation vary among different species. The average concentrations of Ni and As were greater than the recommended value proposed by FAO/WHO. Analysis of health risk indices (THQ, HI) revealed long term consumption of these foods might pose non-carcinogenic health risks and Pb should be paid more attention. Besides, the TCR values of Pd showed moderate carcinogenic risk while As indicated high risks. Therefore, awareness should be raised among the people and regular monitoring requires controlling the contamination burden of heavy metals in food species.

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