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Heavy Metal Uptake Pattern and Potential Human Health Risk through Consumption of Tomato Grown in Industrial Contaminated Soils

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Authors' contributions

This work was carried out in collaboration with all authors. Authors RH and MIJA performed the pot experiment, extract preparation and data recording. Author HMZ designed the study, managed the literatures and wrote the manuscript. Authors SM and MRS helped in manuscript preparation. All authors read and approved the final manuscript.

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ABSTRACT

An experiment was conducted in pots to study heavy metal uptake pattern and to assess health risk for adult male and female through consumption of tomato grown in industrial contaminated soils. The experiment was conducted at the net house of the Department of Agricultural Chemistry, Bangladesh Agricultural University, Mymensingh-2202 followed by completely randomized design (CRD) with four replications. Tomato fruits were grown in two types of agricultural soils, one was industrial contaminated and the other was normal farm soil. Edible parts of tomato fruits were harvested at maturity. The amount of Fe, Mn, Cu, Zn, Cr and Pb present in dried fruits, leaves, shoots and roots of tomato were extracted using di-acid mixture and the concentrations of these metals in aqueous extracts were determined by an atomic absorption spectrophotometer (AAS). Health risk was measured by calculating target hazard quotients (THQ) as established by the US EPA. Heavy metals uptake pattern was in the sequence of Cr > Fe > Mn > Cu > Zn = Pb; Fe > Cr > Mn > Cu > Zn > Pb; Fe > Cr > Mn > Zn > Cu > Pb and Cr > Fe > Mn > Zn > Cu > Pb in fruits,

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leaves, roots and shoots of tomato, respectively. The present study revealed that tomato fruits didn't accumulate Zn although there was a significant amount of available Zn in the soils. The order of Zn, Cr and Cu accumulation by tomato plants was root ≥ shoot > leaf > fruit. In case of Fe and Mn the sequence were root > leaf > shoot > fruit and leaf > shoot > root > fruit, respectively. Among the metals, available concentration of Cr in soils collected from both sites exceeded the soil quality standards, indicating a high risk to the surrounding ecosystems. The calculated THQ values for the metals showed that only Cr had individual value that surpassed 1, and the values for male were 6.15 & 13.26 and for female were 10.63 & 22.93 due to consumption of tomato grown in farm and industrial contaminated soils, respectively. The overall results showed that industrial contaminated sites were more susceptible than normal agricultural farm sites. The study results inferred that Cr health risk through consumption of tomato is unsafe; and in both places female is more vulnerable than male. Finally, the study recommended to investigate the levels of heavy metals in other vegetables and cereals, and also on the occurrence of the diseases linked to heavy metals in the study area.

Keywords: Heavy metal uptake; health risk; tomato; industrial contaminated soil.

1. INTRODUCTION

Heavy metal contamination in cereals and vegetables is a burning question in Bangladesh agriculture, which has become a challenge for both producers and consumers. There are several reports that discharge of untreated industrial waste water is polluting soil and water in the country [1-8]. Crops and vegetables grown in contaminated soils can be a dietary source of heavy metal for human beings [9-12]. Vegetables grown in heavy metal contaminated soils usually showed an increased metal uptake trend in all over the world. Thus, crops and vegetables cultivated in contaminated soils acquire heavy metals in huge quantities to cause potential health risks to the consumers [13]. It has been reported that only 15 ppm available arsenic is enough to create severe health risk through the consumption of vegetables grown in contaminated soils [9]. Accumulation of heavy metals in human body through consumption of cereals and vegetables created a growing concern in the recent days. A number of serious health problems such as kidney problem, anaemia and blood disorders, stomach irritation, vomiting etc. can develop due to excessive dietary intake of heavy metals [14-15].

The daily vegetable consumption by an adult of Bangladesh is 130 g [16]. Different kinds of vegetables are grown during the year in tropical Bangladesh, but very little is known about the metal contents of vegetables [17]. According to naser et al. [18], sporadic information regarding the accumulation of heavy metals in vegetables grown in industrially polluted soils of the country is available. Nowadays, crops and vegetables grown in contaminated soil or use of untreated industrial wastewater for irrigation is a common scenario in Bangladesh. But intake of heavy metals contaminated vegetables may pose a risk to the human health. So, heavy metal contamination of the food items is one of the most important assessment parameters of food quality assurance [13,19], and international and national regulations on food quality have lowered the maximum permissible levels of toxic metals in food items due to an increased awareness of the risk [20]. Considering the fact, this study was planned to study uptake patterns of different heavy metals and to assess their potential health risk for human through consumption of tomato grown in industrial contaminated soils of Bangladesh.

2. MATERIALS AND METHODS

2.1 Experimental Site

The pot experiment was carried out at the Net House, Department of Agricultural Chemistry, Faculty of Agriculture, Bangladesh Agricultural University (BAU), Mymensingh-2202, Bangladesh during the period from October 2015 to November 2016.

2.2 Collection of Soils for Experiment

Two types of agricultural soil (farm soil and industrial contaminated soil) were used for the pot experiment. Among these, farm soil was collected from the field of Genetics and Plant Breeding Farm of BAU, Mymensingh-2202, Bangladesh. Industrial contaminated soil was collected from the site near to Noman Composite Textile Ltd., Habirbari, Bhaluka of Mymensingh.

Requisite amount of both the soils were brought to the Department of Agricultural Chemistry, BAU, Mymensingh and processed for pot experiment.

2.3 Design Used for the Experiment

After collection, both soil samples were analyzed for an available fraction of heavy metals (Fe, Mn, Cu, Zn, Pb and Cr) as described by Tessier *et al.* [21] and analytical results are presented in Table 1. The experiment was laid out, followed by completely randomized design (CRD) with four replications and thus total number of pots were 8 (2×4).

2.4 Test Crops and Intercultural Operations

The experiment was conducted with the seedlings of tomato (*Solanum lycopersicum L.*) var. *Udayon* collected from Horticultural Farm of BAU, Mymensingh-2202, Bangladesh.

Fertilizers applied in the pots as recommended for high yield goal and medium soil fertility status as described in Fertilizer Recommendation Guide [22]. Intercultural operations viz. weeding, irrigation, disease and pest management were done as and when necessary.

2.5 Harvesting and Processing of Samples

Tomato fruits were harvested during early ripening stage when they attained red colour. Harvesting was started on March 14 and completed by March 28, 2016. Different plant parts viz. leaf, shoot and root were also collected after completion of tomato fruit harvesting and samples were tagged and taken to the laboratory. Then the samples were air dried for four days followed by oven drying for 72 hours until the moisture content attained at desirable state. Then dried samples were ground and stored at room temperature for chemical analyses.

2.6 Chemical Analysis of Plant Samples

Powdered samples of different parts of tomato plant (viz. leaf, shoot, root and fruit) were used to prepare aqueous extract by a wet oxidation method using di-acid mixture as described by Singh et al. [23]. The concentrations of different heavy metals in aqueous extracts were measured by atomic absorption spectrophotometer (AAS) (AA-7000, Shimadzu, Japan). Mono element hollow cathode lamp was employed for the determination of each heavy metal of interest. At first the AAS was calibrated followed by the manufacturer's recommendation. Then the aqueous extract was diluted and/or run directly in AAS for the determination of metal in the sample.

2.7 Estimation of Daily Metal Intakes (DMI)

To appraise the health risk associated with heavy metal contamination in edible parts of tomato, the daily intake of metal was calculated with the following formula-

$$DMI = (VIR \times C)/ BW$$

Where, VIR is the vegetable ingestion rate (mg person^{-1} day^{-1}), C is the individual metal concentration in edible parts of tomato samples (mg kg^{-1}, fresh weight), BW is the body weight assuming 70 kg for adult male and 50 kg for adult females in the present study [24].

Table 1. Morphological characteristics and available heavy metal contents present in the soils used for the study

Name of soil	Agro-ecological zone (AEZ)	Land type	Soil colour	Conc. of available heavy metal (µg g^{-1})					
				Cu	Zn	Pb	Cr	Fe	Mn
Industrial contaminated soils	AEZ-9 (Old Brahmaputra Floodplain)	Medium high land	Light brown	9.05	66.34	Trace	79.43	14.99	25.50
Farm soils	AEZ-9 (Old Brahmaputra Floodplain)	Medium high land	Dark grey	8.87	13.23	Trace	57.90	16.56	10.96

2.8 Target Hazard Quotients (THQ)

THQ is calculated by the general formula established by the US EPA as follows-

$$THQ = (E_F \times F_D \times DMI) / (RfD \times W \times T)$$

Where, E_F is exposure frequency; F_D is the exposure duration; DMI is the daily metal ingestion (mg person^{-1} day^{-1}); RfD is the oral reference dose (mg kg^{-1} day^{-1}); W is the average body weight (kg) and T is the average exposure time for noncarcinogens (365 days year^{-1} × number of exposure years).

3. RESULTS AND DISCUSSION

3.1 Uptake Pattern of Heavy Metals in Different Plant Parts of Tomato

Tomato fruits uptake heavy metals in the sequence of Cr > Fe > Mn > Cu > Zn = Pb and concentration of different heavy metals in tomato leaves was obtained in the order of Fe > Cr > Mn > Cu > Zn > Pb (Figs. 1 and 2). Tomato roots uptake different heavy metal in the sequence of Fe > Cr > Zn > Mn > Cu > Pb whereas, shoots uptake in the order of Cr > Fe > Zn > Mn > Cu > Pb (Figs. 3 and 4). The study results inferred that tomato plants uptake very little amount of Pb than other heavy metals, which might be due to trace amount of available Pb content in both the soils used for this study. On other hand, the concentrations of available Zn in both the farm and industrial contaminated soils were 13.23 and 66.34 µg g^{-1}, respectively (Table 1). Present study results revealed that tomato fruits didn't accumulate Zn although there was a significant amount of available Zn in the soils. The sequence of Zn accumulation in tomato plants was root ≥ shoot > leaf > fruit. Furthermore, it was reported that characteristically Pb and Zn interact with each other negatively if present in mixture form. In case of *Solanum lycopersicum* L. seedlings both of Pb and Zn antagonistically affected the uptake rates of each other. This finding is consistent with the result reported by MacFarlane and Burchett [25], who observed that the accumulation of Zn reduced the accumulation of Pb in leaves and vice versa. The uptake pattern of Zn and Pb in Zn/Pb amended soil showed that both Zn and Pb affect the uptake of each other in an antagonistic way [26]. The uptake pattern of different heavy metals by the tomato plants was at par with the results observed by Ngayila et al. [27] for growth of *Brassica juncea.* However, the sequences of Cr, Cu, Fe and Mn accumulation by different parts of tomato plants were shoot ≥ root > leaf > fruit, root > shoot > leaf > fruit, root > leaf > shoot > fruit and leaf > shoot > root > fruit, respectively.

3.2 Copper Contents in Different Plant Parts of Tomato

Copper is an essential micronutrient for normal plant growth and metabolism. It plays an important role in a large number of metalloenzymes, photosynthesis-related plastocyanin and membrane structure [28]. On the other hand, it has also been reported to be among the most toxic of the heavy metals [29]. The mean Cu content in tomato fruits was 31.77±1.84 µg g^{-1} in the sample grown in farm soil while it was 43.10±0.51 µg g^{-1} for industrial contaminated soil (Fig. 1). The average concentrations of Cu in tomato leaves, shoots and roots were 39.21±1.40, 47.93±3.96 and 63.06±2.21 µg g^{-1}, respectively for tomato grown in farm soils (Figs. 2-4). On the other hand, in case of industrial contaminated soils, the mean concentrations of Cu in tomato leaves, shoots and roots were 57.54±1.77, 60.12±4.24 and 72.09±5.32 µg g^{-1}, respectively (Figs. 2-4). The Cu content in fruits, leaves, shoots and roots of tomato grown in both farm and industrial contaminated soils exceeded the critical limit (5.10-30.00 µg g^{-1}) as reported by Kabata-Pendias and Pendias [30]. Alam et al. [17] analyzed vegetable samples from Samta village of Jessore, Bangladesh and reported average Cu concentrations in leafy and non-leafy vegetables were 15.50 and 8.51 µg g^{-1}, respectively.

3.3 Zinc Contents in Different Plant Parts of Tomato

The concentration of Zn in tomato fruit was found as trace for both the studied soils (Fig. 1). The mean concentrations of Zn in tomato leaves were traced and 53.10±2.94 µg g^{-1} for farm and industrial contaminated soils, respectively (Fig. 2). On the other hand, the mean concentrations of Zn in tomato roots grown in farm and industrial contaminated soils were 41.87±6.16 and 337.75±9.81 µg g^{-1}, respectively (Fig. 4). The average concentration of Zn in tomato shoot was 293.27±19.52 µg g^{-1} in industrial contaminated soil and it was 47.01±4.95 µg g^{-1} in farm soil (Fig. 3). The study results inferred that leaves, roots and shoots of tomato plant contained higher amount of Zn, which was grown in industrial contaminated soil. This is due to presence of >5 times of higher amount of available Zn (66.34 µg g^{-1}) in industrial contaminated soil than farm soils of BAU (13.23 µg g^{-1}) (Table 1). According to Codex Alimentarius Commission [31], maximum permissible level of Zn in vegetables is 100 µg g^{-1}. Considering this result, Zn content in leaves of both industrial contaminated and farm soils was below this limit but industrial contaminated root and shoot crossed this limit. According to Islam et al. [16] Zn concentrations of leafy vegetables, fruiting vegetables, and root and tuber vegetables ranged from 5.81-25.40 µg g^{-1}, 9.61-30.48 µg g^{-1} and 1.98-18.50 µg g^{-1}, respectively.

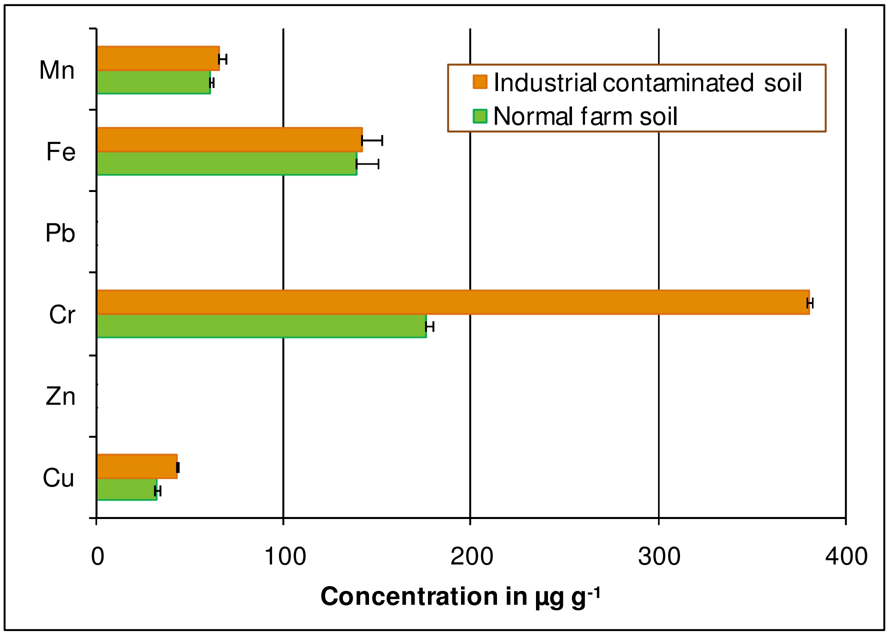

Fig. 1. Heavy metal concentrations (µg g^{-1}) in tomato fruits grown in both industrial contaminated and normal farm soils

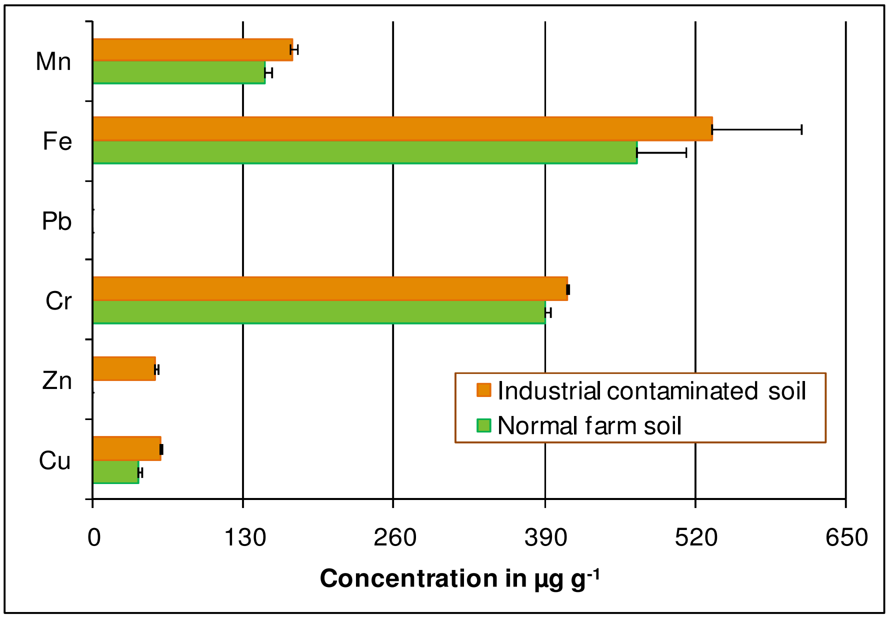

Fig. 2. Heavy metal concentrations (µg g^{-1}) in tomato leaves grown in both industrial contaminated and normal farm soils

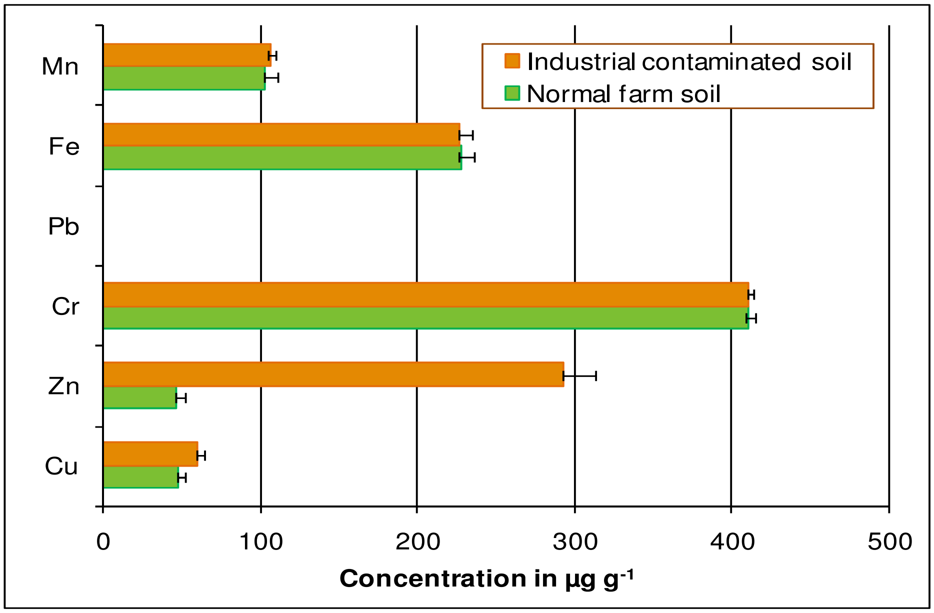

Fig. 3. Heavy metal concentrations (µg g^{-1}) in tomato shoots grown in both industrial contaminated and normal farm soils

3.4 Chromium Contents in Different Plant Parts of Tomato

Chemically, trivalent Cr is non-toxic and necessary for humans, while the hexavalent form is toxic. Vegetables and fruits that contain higher amount of chromium are tomato, spinach and broccoli, and a half cup of these vegetables contained 11.00 µg g^{-1} Cr [32]. The average concentration of Cr in tomato fruits was 176.28±3.23 µg g^{-1} for farm soil while it was 380.20±1.40 µg g^{-1} for industrial contaminated soils (Fig. 1). The mean concentrations of Cr in tomato leaves were 390.78±2.62 and 408.65±1.93 µg g^{-1} for farm and industrial contaminated soils, respectively (Fig. 2). Average concentrations of Cr in tomato roots were 402.61±8.37 and 410.75±6.10 µg g^{-1} for farm and industrial contaminated soils, respectively (Fig. 4). The mean concentrations of Cr in tomato shoots grown in farm and industrial contaminated soils were 410.03±4.53 and 410.16±2.89 µg g^{-1}, respectively (Fig. 3). The study results inferred that the concentrations of Cr in all the samples crossed the critical limit as mentioned by Kabata-Pendias and Pendias [30], which is due to the presence of higher amount of DTPA (Diethylene triamine penta acetic acid) extractable i.e. available Cr in both farm and industrial contaminated soils (57.90 and 79.43 µg g^{-1}, respectively).

3.5 Lead Contents in Different Plant Parts of Tomato

The concentration of Pb in tomato fruits, leaves, roots and shoots sample was found as a trace. This might be due to the presence of trace amount of available Pb content in both the soils used in the study. Furthermore, it was reported by MacFarlane and Burchett [25], that the accumulation of Zn reduced the accumulation of Pb in leaves and vice versa. Lima et al. [33] reported that the highest Pb concentration was found in carrot roots, while green collards and cabbage showed the lowest level of Pb allocated in the root system. On the other hand, according to Manecki et al. [34], there was a very low bioaccumulation rate of the Pb present in the soil, due to not only the toxicity of this metal to the plants, but also to its inherent low solubility in the soil.

3.6 Iron Contents in Different Plant Parts of Tomato

Iron is an essential nutrient for plant growth and development, but after certain limit it is regarded as toxic element for the plants. The safe limit of Fe in plants is 140 µg g^{-1} [35]. The average concentration of Fe in tomato fruits was 138.90±11.24 µg g^{-1} for farm soil while it was 142.03±9.34 µg g^{-1} for industrial contaminated

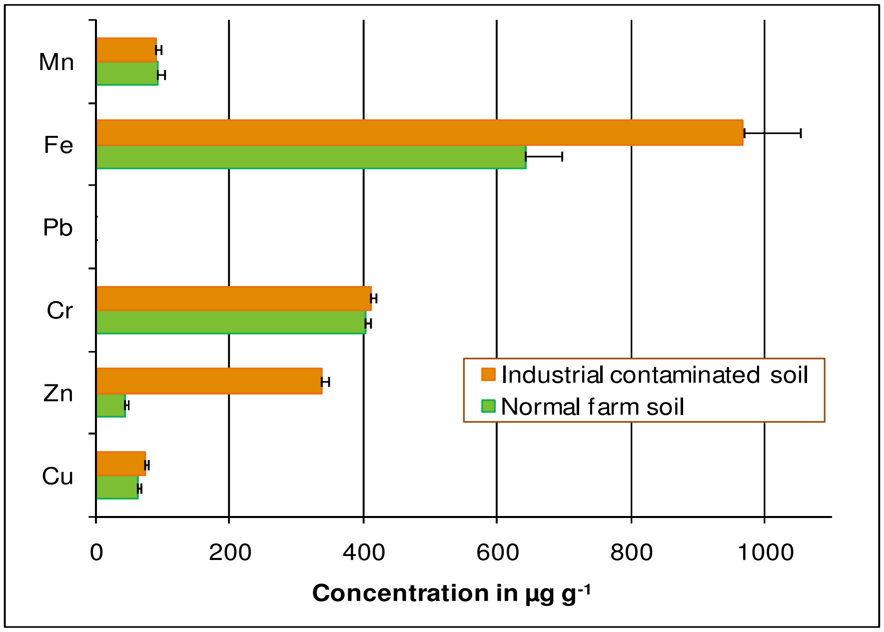

Fig. 4. Heavy metal concentrations (µg g^{-1}) in tomato roots grown in both industrial contaminated and normal farm soils

soils (Fig. 1). Minimum and maximum concentrations of Fe in tomato fruits were 130.28 and 152.74 µg g^{-1}, respectively. The mean concentrations of Fe in tomato leaves were 469.76±41.84 and 534.25±75.85 µg g^{-1} for farm and industrial contaminated soils, respectively (Fig. 2). In case of root, the mean concentrations of Fe were 641.72±54.43 and 968.24±84.13 µg g^{-1} in tomato grown in farm and industrial contaminated soils, respectively (Fig. 4). On the other hand, average concentrations of Fe in tomato shoots were 227.63±8.17 and 226.80±7.34 µg g^{-1} in farm and industrial contaminated soils, respectively (Fig. 3). So that it can be said that Fe concentrations in all parts of tomato exceeded the safe limit (140 µg g^{-1}) as stated by Misra and Mani [35]. Arora et al. [36] reported that Fe concentration in wastewater-irrigated plants as 116.0 to 378.0 µg g^{-1} and the highest mean levels of Fe was detected in mint and spinach.

3.7 Manganese Contents in Different Plant Parts of Tomato

Naturally, Mn occurs in many food sources, such as leafy vegetables, nuts, grains and animal products [37]. The concentrations of Mn in different plant parts of tomato samples were within the normal concentration (20-300 µg g^{-1}) for plant as described by Kabata-Pendias and Pendias [30]. The highest amount of Mn (68.62 µg g^{-1}) in tomato fruits was recorded in industrial contaminated soil and the lowest (59.76 µg g^{-1}) was obtained in tomato harvested from farm soils. The average concentration of Mn in tomato fruits was 60.64±0.94 µg g^{-1} for farm soil while it was 65.50±2.73 µg g^{-1} for industrial contaminated soils (Fig. 1). The mean concentrations of Mn in tomato leaves were 148.41±4.58 and 170.96±5.06 µg g^{-1} for farm and industrial contaminated soils, respectively (Fig. 2). In case of shoot, the mean concentrations of Mn were 103.25±7.48 and 106.26±3.00 µg g^{-1} in tomato grown in farm and industrial contaminated soils, respectively (Fig. 3). On the other hand, average concentrations of Mn in tomato roots grown in farm and industrial contaminated soils were 92.98±8.57 and 90.00±5.13 µg g^{-1}, respectively (Fig. 4).

3.8 Estimation of Daily Metal Intakes (DMI)

To evaluate the health risk associated with heavy metal contamination of tomato, the daily intake of metals was calculated. There are several possible pathways of exposure of metals to humans, but the food chain is the most important. The daily intake of different heavy metals was calculated according to the average vegetable consumption for both adults male and female. A survey was conducted in March 2016 by using a prepared questionnaire of 30 family heads at industrial contaminated sites in the Habirbari area of Bhaluka Upazila and 50 family heads at Sutiakhali area of Mymensingh Sadar Upazila. Thus a total of 80 families faced the interview and in total 270 persons were effectively interviewed from two study areas. This survey data were used to calculate an average consumption rate of tomato per person per day. The survey results revealed 0.017 kg of tomato as typical serving for a day for male and 0.015 kg for female [9]. The daily metals intakes estimate of Fe, Mn, Zn, Cu, Cr and Pb from tomato were calculated by multiplying the daily intake (from survey results) by the mean metal concentrations determined in this study. The DMI was compared with the upper tolerable daily intakes for metals. It is evident from Table 2 that daily metal intake for Cr, Mn and Cu were several time higher than that of oral reference doses.

3.9 Target Hazard Quotients (THQ)

Target hazard quotients (THQ) are a complex parameter used for the estimation of potential health risks associated with long term exposure to chemical pollutants [11,42-43]. The THQ <1 means the exposed population is assumed to be safe, 1 < THQ < 5 means that the exposed population is in a level of concern interval and THQ > 5 indicates the exposed population is in health risk. THQ parameter is a dimension less index and THQ values are additive, but not multiplicative.

In this study, THQ was calculated considering DMI of people, average body weight for male: 70 kg and female: 50 kg as mentioned by Guyton and Hall [44], and average life expectancy (male: 70.6 and female: 73.1) [24]. Values of this parameter (THQ) due to edible part of tomato for investigated metals are presented in Table 3, and there was only one individual THQ value that surpassed 1 i.e. Cr; and the values for male were 6.15 & 13.26 and for female were 10.63 & 22.93 for farm and industrial contaminated soils, respectively. So it can be inferred from the present study that Cr health risk through the food chain via consumption of tomato was unsafe; and in both places female is more vulnerable than male. The contributions from all metals bring the combined target hazard quotients (CTHQ) value was also surpassed 1 and

Table 2. Daily intakes of heavy metals (DMI) from tomato fruits for both male and female at farm and industrial contaminated soils of the study areas

Heavy metals	DMI from tomato grown in farm soils (mg day^{-1} $person^{-1}$)		DMI from tomato grown in industrial contaminated soils (mg day^{-1} $person^{-1}$)		Oral reference doses (RfD) (mg kg^{-1} day^{-1})	Tolerable upper intake level (UL) (mg day^{-1} $person^{-1}$)
	Male	Female	Male	Female		
Cu	0.463	0.572	0.628	0.776	0.040[a]	10.00[d]
Zn	0	0	0	0	0.300[a]	40.00[d]
Cr	2.568	3.172	5.539	6.842	0.003[b]	NE[d]
Pb	0	0	0	0	0.004[c]	0.24[e]
Fe	2.024	2.500	2.069	2.556	0.700[a]	45.00[d]
Mn	0.883	1.091	0.954	1.179	0.014[a]	11.00[d]

NE= Not established; [a] = US EPA [38]; [b] = IRIS [39]; [c] = Khan et al. [13]; [d] = FDA [40] and [e] = Garcia-Rico et al. [41]

Table 3. Target hazard quotients (THQ) and combined target hazard quotient (CTHQ) of heavy metals for both male and female due to consumption of tomato fruits

		Target Hazard Quotients (THQ)						CTHQ
		Cu	Zn	Cr	Pb	Fe	Mn	
Industrial contaminated soils of the study area	Male	0.113	0	13.260	0	0.021	0.490	13.884
	Female	0.195	0	22.932	0	0.037	0.847	24.011
Farm soils of the study area	Male	0.083	0	6.148	0	0.021	0.453	6.705
	Female	0.144	0	10.633	0	0.036	0.784	11.596

the value for male ranged from 6.71 to 13.88 and for female 11.60 to 24.01, which is also an indication of potential health risks for population of the study areas.

4. CONCLUSION

Tomato fruits, leaves, roots and shoots contained metals in the order of Cr > Fe > Mn > Cu > Zn = Pb; Fe > Cr > Mn > Cu > Zn > Pb; Fe > Cr > Mn > Zn > Cu > Pb and Cr > Fe > Mn > Zn > Cu > Pb, respectively. The present study revealed that tomato fruits didn't accumulate Zn although there was a significant amount of available Zn in the soils. But roots and shoots of tomato plants contained higher amount of Zn for both types of soils and the sequence of Zn accumulation was root ≥ shoot > leaf > fruit. On the other hand, the order of Cr, Cu, Fe and Mn accumulation by different parts of tomato plants were shoot ≥ root > leaf > fruit, root > shoot > leaf > fruit, root > leaf > shoot > fruit and leaf > shoot > root > fruit, respectively. The present study also revealed that available concentration of Cr in soils collected from both sites exceeded the soil quality standards, indicating high risk to the surrounding ecosystems. Tomato grown in those soils was also contaminated by the relevant metal, which could pose a potential health concern to the local residents.

Among the heavy metals present in edible parts of tomato, only Cr had individual THQ value that surpassed 1 (6.15 to 13.26 for male, and 10.63 to 22.93 for female). Thus the study results inferred that Cr health risk through the food chain via consumption of tomato was unsafe; and in both places female was more vulnerable than male. In conclusion, as the estimated intake of heavy metals in the present study does not include the contribution of other vegetables and foods that may represent further contamination sources to the population subjected. So further investigation should be focused on the levels of heavy metals in other vegetables and cereals along with water and air, also on the occurrence of the diseases linked to heavy metals in the study area.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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