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Translocation of heavy metals from industry into vegetables and crops through water and soil of Mokesh Beel in Bangladesh and their impact on human body

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Abstract:*The contaminations of toxic heavy metals such as, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Hg and Pb in irrigation water, soil and highly consumed seven vegetables and crops at Mokesh Beel, Gazipur in Bangladesh, were determined by inductively coupled plasma-mass spectroscopy (ICP-MS). Most of the metal's concentrations exceeded the maximum permissible limit set by Pescod, (1992); EU, (2002); Awashthi, (2000); FAO/WHO, (1994, 2001) and others international standard for irrigation water, soil, crops and vegetables. The mean concentrations of metals were observed decreasing in the following order: Fe > Zn > Mn > Cr > Pb > Cu > Hg > As > Cd > Ni in irrigation water; Fe > Mn > Cr > Zn > Ni > Pb > Cu > As > Cd > Hg in soil and Zn > Fe > Pb > Mn > Cu > Ni > Cr > As > Cd > Hg in vegetables and crops. Contamination factor (CF) showed that the crops and vegetables were moderate to highly contaminated by Cu, Zn, Cd, Hg and Pb. The pollution load index (PLI) and the geo-accumulation index (I_{geo}) values of analysed samples revealed that the crops and vegetables wereuncontaminated to highly contaminated. The target qutioned hazards (THQs) of most of the metals were >1, suggesting remarkable non-carcinogenic health hazards for adult population. Total THQs of all metals were >1 through consumption of all crops and vegetables, indicating significant health risks. The total carcinogenic risk (TR) of arsenic was above the safe standard (1E-06 to 1E-04) as well as Pb in most samples, suggesting remarkable carcinogenic risk (TR) from their consumption. The present study revealed that the health risks associated with the consumption of heavy metals through the intake of vegetables and crops in adult population of Mokesh Beel area in Bangladesh.*

Keywords: *Heavy metals; Target hazard; Carcinogenic; Pollution load; Mokesh Beel.*

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I. Introduction

Contamination of heavy metals in the aquatic environment has attracted considerable global attention owing to its abundance, persistence and environmental toxicity(Ahmed et al., 2015a,b). Both natural and anthropogenic activities are responsible for the abundant of heavy metals in the environment (Wilson & Pyatt, 2007; Khan et al., 2008). However, anthropogenic activities can effortlessly generate heavy metals in sediment and water that pollute the aquatic environment (Sánchez-Chardi et al., 2007). Heavy metal pollution of agricultural soil and vegetables through waste water is one of the most severe ecological problems on a world scale as well as in Bangladesh. The food chain contamination is the major pathway of heavy metal exposure for humans (Khan et al., 2008). Industrial or municipal wastewater irrigation is a common reality in three fourth of the cities in Asia, Africa, and Latin America (Gupta et al., 2009). Rapid and unorganized urban and industrial developments have contributed to the elevated levels of heavy metals in the urban environment of developing countries such as Egypt (Radwan and Salama, 2006), Iran (Maleki & Zarasvand, 2008), China (Wong et al., 2003), India (Sharma et al., 2008a,b) and Bangladesh (Ahmad & Goni, 2010).

Long-term use of industrial or municipal wastewater in irrigation is known to have significant contribution to trace elements such as Cd, Cu, Zn, Cr, Ni, Pb, As, Hg and Mn in surface soil (Mapanda et al., 2005). Excessive accumulation of trace elements in agricultural soils through wastewater irrigation may not only result in soil contamination but also affect food quality and safety (Muchuweti et al., 2006; Sharma et al., 2007). Therefore, the Food and Agricultural Organization (FAO), World Health Organization (WHO), US Environmental Protection Agency (USEPA), and other regulatory bodies of various countries have established the maximum permitted concentrations of heavy metals in foodstuffs (Xue et al., 2012). Vegetables cultivated in soils polluted with toxic metals due to industrial activities take up heavy metals and accumulate them in their edible and non-edible parts in quantities high enough to cause clinical problems both to animals and human beings consuming these metal-rich plants because there is no good mechanism for their elimination from the human body (Arora et al., 2008; Alam et al., 2003). Industries located in different areas of Bangladesh discharge their effluents without any prior treatment into nearby water bodies and agricultural land, thus contaminating the crops and vegetables grown in and around the surrounding areas (Ahmad and Goni, 2010). A number of studies have reported the deposition of metals in soil, crops and vegetables grown in the vicinity of industrial areas around the world (Mottalib et al., 2016; Grytsyuk et al., 2006; Khan et al., 2008; Gupta et al., 2009; Smith, 2009; Goni et al., 2014).

There is, however, limited published information on the contamination of wastewater, soil, crops and vegetables in the vicinity of industrial areas of Bangladesh. So, a complete study was badly needed at Mokesh Beel which is situated at Gazipur industrial area in Bangladesh. This beel usually deluges during summer season. People of this area, irrigate crops and vegetables only during winter season. The objective of this study was to determine the concentration of Mn, Cu, Zn, As, Hg, Pb, Cr, Cd, Fe and Ni in water, soil, vegetables and crops of the Mokesh Beel Industrial area of Bangladesh and to identify the interaction of metals between soil, vegetables and crops. Human health risk estimation was also investigated.

II. Materials and methods

Study area

The study site covered the agriculture area around Mokesh Beel, near Gazipur industrial area of Bangladesh. Gazipur has an area of 1741.53 km^2, located in between 23°53' and 24°21' north latitudes and in between 90°09' and 92°39' east longitudes. Mokesh is a perennial beel of Gazipur, located approximately fifty-five kilometers north of Dhaka and lies in the Turag River basin surrounded by sal (*Shorea robusta*) forests, numerous canals (*khals*) and Juran Beel. This area is an integral part of local livelihoods and culture. The wetland of Mokesh Beel plays an important role in ground water recharge and discharge, storage of food water, shorelines stabilization. This is one of many similar industrial clusters that has developed as part of the rapid economic growth of Bangladesh, as is the case with all, it is also an area of serious water pollution. Although poultry farms, pharmaceuticals industries, cement industries, glass factories and battery industries has been established there, textile manufacturing industries including dyeing and printing units, still dominating the area (Ullah et al., 2006). These industries are providing employment, increasing local incomes and earning foreign exchange for the country. However, these industries discharge their waste directly into the water body, adversely affecting the livelihoods and the day to day life of the whole community of the area. The people around the Mokesh Beel, cultivate vegetables and crops during winter season only. Current study was conducted to investigate the pollution load from industry to food chain through water, soil and vegetables and finally to human being.

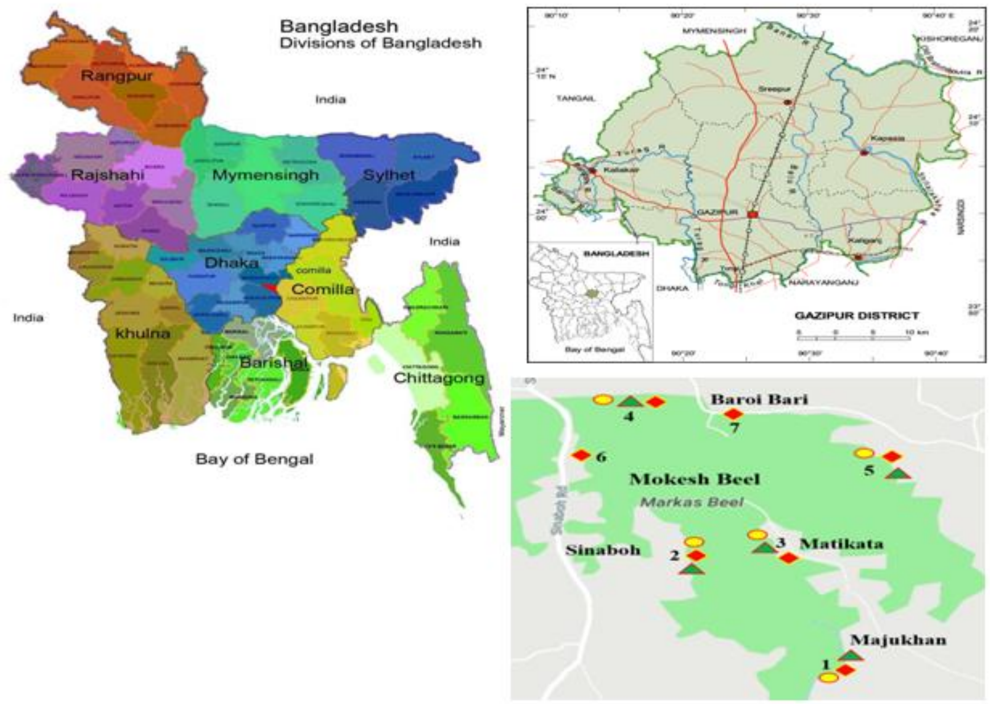

Fig. 1: Map of the study area of Mokesh Beel, Gazipur, Bangladesh

Sampling and pretreatment

All the samples were collected during post-monsoon season, because the Beel area usually deluges in pre-monsoon season. The water that was used to measure the concentration of heavy metal was randomly collected from different areas of Mokesh Beel. Five water samples i.e. S1, S2, S3, S4 and S5 were collected from five different locations like Majukhan, Sinaboh Bazar, Matikata and Baroi Bari (2 sites) respectively. Water samples (up to 500 mL) were collected from the five sites and 1.0 mL conc. HNO_3 was added to each water sample to prevent microbial activity. The bottles were brought back to the laboratory and digestion was completed within a week.

Five soil samples like S1 (Bottle Gourd soil), S2 (Pumpkin soil), S3 (Tomato soil), S4 (Red Spinach soil) and S5 (Paddy soil) were collected from five different locations on the beel and its tributaries i.e. Majukhan, Sinaboh, Matikata and Baroi Bari (2 sites) respectively during winter season (Nov-Dec) in 2017. Soil samples were air and oven dried, crushed, and passed through 2-mm-mesh sieve and stored at 4°C temperature before analysis of soils for determining of heavy metals.

Best quality fresh samples of vegetables were collected from the located Beel during winter season (Nov-Dec) in 2017. The vegetable samples which were collected from different locations of the Beel i.e. Majukhan (S2), Matikata (S1), Sinahboh Bazar (S3, S4, S5), Baroi Bari (S6, S7). Where, S1- Tomato (*Solanum lycopersicum*), S2- Red Spinach (*Amaranthus dubius*), S3- Paddy (*Oryza sativa*), S4- Bottle Gourd (*Lagenaria siceraria*), S5- Gourd Spinach (*Lagenaria siceraria*), S6- Pumpkin (*Cucurbita maxima*), S7- Pumpkin Spinach (*Cucurbita maxima*). All samples were washed with distilled water and were cut into small pieces which dried at 105°C in oven till constant weight was achieved. The samples were then crushed separately through a mortar and re-weighted.

Digestion and analysis

Fifty-milliliter of mixed effluent-contaminated water sample was digested with 5 mL of concentrated HNO_3 at 80°C until the solution became transparent (APHA, 1985). The solution was filtered through Whatman no. 42 filter paper and the filtrate was diluted to 50 mL with distilled and deionized water. Each soil and vegetable sample (0.5 g) was digested with a mixture of 14 mL concentrated HNO_3, H_2SO_4, and $HClO_4$ in 5:1:1(v/v) ratio at 85°C until a transparent solution was obtained (Allen et al., 1986). Digested soil and crop samples were filtered through Whatman no. 42 filter paper, and the filtrates were diluted to 50 mL with deionized water. Digested samples were then stored at 4°C temperature until metal analysis. All chemicals used were Merck Germany analytical grade including standard stock solutions of known concentrations of different metals. The concentrations of heavy metals in filtrate of water, soil, and vegetable samples were analyzed using ICP-MS (Inductively Coupled Plasma-Mass Spectroscopy) (Optima-7000 DV, USA). Detection limits and wavelength used in analysis of the metals were 3.60, 0.07, 0.007, 0.90, 0.40, 1.00, 1.40, 1.20, 1.00 and 1.00 ppb and 193.696, 214.440, 283.553, 224.700, 231.604, 213.857, 220.353, 253.718, 260.568 and 259.940 nm respectably for As, Cd, Cr, Cu, Ni, Zn, Pb, Hg, Mn and Fe.

Statistical analysis

All statistical analyses were performed using the Microsoft Excel (version 2016). Analysis of variance (Two-way ANOVA) and correlation matrix were employed to examine statistical significance of differences in the mean concentration of metals between (or among) group of families of water, soil and vegetable. A probability level of $P < 0.05$ was considered statistically significant.

Assessment of heavy metals in soil to vegetable

In the interpretation of geochemical data, choice of background values plays a significant contribution. Several researchers have used the average shale values or the average crustal abundance data as reference baselines (Loska & Wiechuła, 2003; Singh et al., 2005; Islam et al., 2015a). The degree of contamination from heavy metals could be evaluated by determining the contamination factor (CF), pollution load index (*PLI*) and geo-accumulation index (I_{geo}).

Pollution load index (PLI) and contamination factor (CF)

To evaluate the sediment quality, combined approaches of pollution load index of the four metals were calculated according to the study by Islam et al., (2015b). The Pollution Load Index (*PLI*) and geo-accumulation Index (I_{geo}) were employed. To determine the magnitude of heavy metal contamination in the vegetables, Pollution Load Index for each site was evaluated using the procedure of Tomlinson et al., (1980)as follows,

$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n)^{1/n}$

Where, n = number of metals and *CF* = contamination factor, which is the ratio between the metal concentration in vegetable to the background values of metals ($CF_n = C_{vegetable}/C_{background}$). The ratio of the measured concentration to natural abundance of a given metal had been proposed as the contamination factor (*CF*) being classified into four grades for monitoring the pollution of one single metal over a period of time (Islam et al., 2015b): low degree (*CF* < *1*), moderate degree (*1* ≤ *CF* < *3*), considerable degree (*3* ≤ *CF* < *6*), and very high degree (*CF* ≥ *6*). Thus, the *CF* values can monitor the enrichment of one given metal in soil to vegetable over a period of time. The *PLI* value > 1 is polluted while *PLI* value < 1 indicates no pollution. *2.6.2. Geo-accumulation index (I_{geo})*

The degree of contamination from the heavy metals could be determined by measuring the geo-accumulation index (I_{geo}) and it was calculated using the equation(Saleem et al., 2015),

$$Igeo = Log2[\frac{C_n}{1.5B_n}]$$

where C_n is the measured concentration of metal n in the vegetables and B_n is the geochemical background value of element n in the background sample (soil in present study) (Yu et al., 2011; Rahman and Ishiga, 2012; Islam et al., 2015a). The factor 1.5 is introduced to minimize the possible variations in the background values which may be qualified to lithogenic effects. Geo-accumulation index (I_{geo}) values were interpreted as: $I_{geo} \le 0$ – practically uncontaminated; $0 \le I_{geo} \le 1$ - uncontaminated to moderately contaminated; $1 \le I_{geo} \le 2$ - moderately contaminated; $2 \le I_{geo} \le 3$ - moderately to heavily contaminated; $3 \le I_{geo} \le 4$ - heavily contaminated; $4 \le I_{geo} \le 5$ - heavily to extremely contaminated; and I_{geo}>5 - extremely contaminated.

Health risk estimation

Estimated daily intake of heavy metals

Estimated daily intakes (*EDIs*) of heavy metals were calculated using their respective average concentration in food samples by the weight of food items consumed by an individual (body weight 60 kg for an adult in Bangladesh) (FAO, 2006), which was obtained from the household income and expenditure survey (HIES, 2011) and calculated by the following formula:

$$EDI = \frac{E_f \times E_D \times F_{IR} \times C_f \times C_m}{W_{AB} \times T_A} \times 10^{-3}$$

Where E_f is the exposure frequency (365 days/year); E_D is the exposure duration, equivalent to average lifetime (60 years for Bangladeshi population; Uddin et al., 2010); F_{IR} is the fresh food ingestion rate (g/person/day), which was considered to be 126, 29, and 48 g/person/day for vegetables, fruits, and fishes, respectively (Ali & Hau, 2001); $C_f$is the conversion factor (00.208) to convert fresh weight (f.w.) to dry weight (d.w.) considering 79 % of moisture content; $C_m$is the heavy metal concentration in foodstuffs (mg/kg d.w.); W_{AB} is the average body weight (b.w.) (average adult body weight was considered to be 60 kg); and T_A is the average exposure time for noncarcinogens (it is equal to $E_f \times E_D$) as used in many previous studies (Wang et al., 2005).

Noncarcinogenic risk

Non-carcinogenic risk assessments are typically conducted to estimate the potential health risks of pollutants using the THQ, which is the ratio between the estimated exposure and the oral reference dose (RfD, mg/kg bw/day). The oral reference doses are 1.5, 0.02, 0.04, 0.3, 0.0003, 0.001, 0.0003 0.0035, 0.7 and 0.14 mg/kg/day for Cr, Ni, Cu, Zn, As, Cd, Hg, Pb, Fe and Mn, respectively (USEPA, 2015). The target hazard quotient (*THQ*) and total target hazard quotient (*TTHQ*) can be calculated as (FAO/WHO, 2011),

$$THQ = \frac{EDI}{RfD}$$

TTHQ (individual food) = THQ (metal 1) + THQ (metal 2) + ……… + THQ (metal n)

A *THQ*< 1 means the exposed population is unlikely to experience obvious adverse effects, whereas a *THQ* ≥ 1 means that there is a chance of noncarcinogenic effects, with an increasing probability as the value increases. In order to assess the overall potential for noncarcinogenic effects from more than one heavy metal, a hazard index (*HI*) has been formulated based on the Guidelines for Health Risk Assessment of Chemical Mixtures of USEPA as follows (USEPA, 1989):

HI = ∑ TTHQ

= TTHQ (food)1 + TTHQ (food 2) + ……… + TTHQ (food n)

Carcinogenic risk

The target carcinogenic risk (*CR*) factor (lifetime cancer risk) (USEPA, 1989) can be calculated as,

TR = Csfo × EDI

where *TR* is the probability of excess lifetime cancer or simply target cancer risk, *Csfo* is the oral carcinogenic slope factor obtained from the integrated risk information system (USEPA, 2015) database, which was 1.5 and 8.5×10^{-3} $(mg/kg/day)^{-1}$ for As and Pb, respectively and *EDI* is the estimated daily intake of heavy metals. In 2010, the USEPA proposed a *Csfo* value of 25.7 $(mg/kg/day)^{-1}$ for bladder and lung cancer based on epidemiologic literature (USEPA, 2010).According to the USEPA, cancer risk is considered “acceptable” when the ILCR is below 1×10^{-6}, while it becomes dangerous when the value exceeds 1×10^{-4} (USPEA, 2011).

III. Results and discussion

Metal concentration in water

The results of heavy metal concentrations in effluent contaminated irrigation waste waters of Mokesh Beel were shown in Table 1.

Table 1: Heavy metal concentration (mg/L) in wastewater used for irrigation in Mokesh Beel with maximum permitted concentration and other relevant studies in Bangladesh.

Sites	Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb
S1	0.25	0.32	3.58	0.02	0.07	0.37	0.002	0.02	0.21	0.04
S2	0.21	0.38	3.79	0.01	0.06	0.65	0.002	0.02	0.003	0.05
S3	0.04	0.40	4.16	0.01	0.07	0.49	0.003	0.02	0.008	0.13
S4	0.05	0.39	4.31	0.01	0.07	0.47	0.003	0.007	0.007	0.11
S5	0.05	0.43	4.55	0.01	0.08	0.66	0.003	0.01	0.006	0.11
Average	0.12	0.38	4.08	0.01	0.07	0.53	0.0026	0.02	0.05	0.09
± SD	± 0.10	± 0.04	± 0.39	± 0.00	± 0.01	± 0.12	± 0.00	± 0.01	± 0.09	± 0.04
Pescod, (1992)	0.1	0.2	-	0.2	0.2	2	-	0.01	-	0.5
WHO, (2004)	0.005				2		0.01	0.003	-	0.01
FAO, (1975)	-	0.2	5	0.2	0.2	-	-	-	-	-
WHO, (1971)	-	-	-	-	-	-	-	-	0.01	-
TRV[a]	0.011	-	-	0.052	0.009	-	0.15	0.002	-	0.003
EPA PR China, (2002)	0.01	-	0.3			0.05	-	0.001	-	0.01
Ahmad & Goni, (2010)	0.43	-	4.94	0.19	2.17	0.95	-	0.06	-	0.21
Ahmad et al., (2010)	0.58	-	-	0.009	0.16	-	-	0.009	-	0.065
Islam et al., (2015b)	0.083	-	-	0.039	0.073	-	0.046	0.011	-	0.035
Rahman et al., (2014)	0.093	0.088	-	0.035	1.053	3.318	0.024	0.007	-	0.108

The average concentration of studied metals in waste waters followed the descending order of Fe > Zn > Mn > Cr > Pb > Cu > Hg > As > Cd > Ni. The mean concentration of Cr in waters was observed 0.12 mg/L (0.04-0.25 mg/L) which was much higher than the WHO, (2004) and Pescod, (1992) (only for S1 and S2 sites) safe standard level, for drinking water 0.005 mg/L and for irrigation water 0.1 mg/L respectively. The average concentration of Hg was observed 0.05 mg/L (0.003-0.21 mg/L) during winter in the beel area. Interestingly, the highest concentration of Hg was observed at S1 site (0.21 mg/L) which might be attributed to the effluent and runoff of industries surrounding the beel area (Islam et al., 2015a/b; Wu et al., 2008). Arsenic forms a variety of inorganic and organic compounds of different toxicity reflecting the physicochemical properties of arsenic at different valences. The average concentration of As was 0.0026 mg/L (0.002-0.003 mg/L) which was below the WHO, (2004) safe limit 0.01 mg/L. The average concentration of Cd was obtained 0.02 mg/L (0.007-0.02 mg/L) which exceeded the WHO, (2004) drinking water standard (0.003 mg/L) as well as Pescod, (1992) irrigation water standard (0.01 mg/L). The mean concentration of Pb and Mn were observed 0.09 mg/L (0.04-0.13 mg/L) and 0.38 mg/L (0.31-0.43 mg/L) respectively where only Pb exceeded the irrigation water quality standard (Pescod, 1992). Considering the toxicity reference values (TRV) proposed by USEPA, (1999) almost all the heavy metals especially Cr, Cd and Pb greatly exceeded the limit which indicated that water from this beel is not safe for drinking and/or cooking as well as for irrigation. The average concentration of Fe, Ni, Zn and Cu were obtained 4.08 mg/L (3.58-4.55 mg/L), 0.01 mg/L (0.01-0.02 mg/L) and 0.07 mg/L (0.06-0.08 mg/L) respectively, where all of these metals were below the safe standard limit of irrigation water (Pescod, 1992). The highest level of heavy metals found in the Mokesh Beel area during winter, might be due to lower dilution effect of water (Islam et al., 2015a/b; Mohiuddin et al., 2012).

Metal concentration in soil

Heavy metal concentrations of wastewater irrigated soils were presented in Table 2.

Table 2: Heavy metal concentrations (mg/kg) in wastewater irritated soil of Mokesh Beel with international guidelines and other relevant studies in Bangladesh

Samples	Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb
S1	38.71	229.87	18818.7	19.37	12.17	5.17	3.89	0.09	-	21.25
S2	115.06	145.2	18712.3	39.69	28.69	65.69	2.61	0.17	-	39.08
S3	62.84	86.28	18100.3	35.69	21.89	36.65	2.67	0.15	-	31.36
S4	57.02	272.16	17567.5	18.83	25.81	154.06	2.64	0.21	-	14.27
S5	55.95	134.66	20406.5	34.25	19.94	37.15	4.22	0.2	-	32.12
Average	65.92	173.63	18721.1	29.57	21.70	59.74	3.21	0.16	-	27.62

± SD	± 28.91	± 75.54	± 1000	± 9.76	± 6.32	± 56.91	± 0.78	± 0.05	-	± 9.80
CoŞKun et al., (2006)	70	-	1000	50	30	90	-	0.35	-	35
Awashthi, (2000)	-	-	-	75-150	135-270	300-600	-	3-6	-	250-500
CNEMC, (1990)	-	-	-	-	-	-	-	0.3	0.3	-
EU, (2002)	150	-	-	75	140	300	-	3	-	300
Ahmad & Goni, (2010)	53.7	-	1715.8	58.16	39.14	115.43	-	11.42	-	49.71
Ahmad et al., (2010)	177.53	-	-	200.45	27.85	-	-	3.33	-	69.75
Islam et al., (2015a)	45	-	-	34	30	-	12	0.72	-	25
Islam et al., (2015b)	118	-	-	103	82	-	27	1.5	-	63
Rahman et al., (2014)	98	483.5	-	26	31	117	1.93	0.61	-	60
Goni et al., (2014)	51.22	-	2146.75	38.79	40.97	116.1	-	10.67	-	46

Concentrations of heavy metals at samples S2, S4 and S5 were much higher because these samples sites are located at very near the industries which discharge extensive amount of untreated effluents to the beel. Metal concentrations in soil were higher in winter because lower water flow of the Mokesh Beel. The average concentration of heavy metals in soil were in the decreasing order of Fe > Mn > Cr > Zn > Ni > Pb > Cu > As > Cd > Hg. The concentration of Cr in soil was moderate than other metals as a result of direct discharging untreated wastes from different industries and textile mills (Islam et al., 2015a). The highest concentration of Cr was found in S2 samples (115.06 mg/kg) indicates its higher input, which might be originated from the urban and industrial waste (Mohiuddin et al., 2012). The average concentration of As in soil samples was 3.21 mg/kg (2.61-4.22 mg/kg) which was much higher than that of Bangshai river 1.93 mg/kg (Rahman et al., 2014). High As concentration in sediments might be attributed to the anthropogenic activities such as treatment from the fertilizers and arsenical pesticides industries (Fu et al., 2014; Ahmed et al., 2016). The mean concentration of Cd was 0.16 mg/kg (0.09-0.21 mg/kg) which was lower than the international safe standard (Table 2). Average concentration of Fe was 18721.07 mg/kg (17567.5-20406.5 mg/kg) which was too much higher than the previous study CoŞKun et al., 2006 (Table 2). Moderate level of Fe was found in winter which might be due to the differences in water capacity of the beel where low water flow in winter resulted the precipitation of Fe in soil; there by rising its concentration (Islam et al., 2015b). The average concentrations of Ni, Cu and Zn were 29.57 mg/kg (18.83-39.69 mg/kg), 21.70 mg/kg (12.17-28.69 mg/kg) and 59.74 mg/kg (5.17-154.06 mg/kg) respectively, where all of these concentrations were below the EU, (2002) and Awashthi, (2000) international safe limit (Table 2). The average value of Mn was 173.63 mg.kg where samples S4 and S1 showed maximum concentration at 272.16 and 229.87 mg/kg respectively. All the Mn values in soil samples were much lower than the previous study (483.5mg/kg) done by Rahman et al., 2014. The mean concentration of Pb was observed 27.62 mg/kg (14.27-39.08 mg/kg) which was very lower than EU, (2002) and Awashthi, (2000) standard. Slightly higher level of Pb content was obtained from soil samples sites which could be due to the effect from point and non-point sources; such as leaded gasoline, petroleum, municipal runoffs and atmospheric deposition (Mohiuddin et al., 2012; Shikazono et al., 2012), chemicals and electronics manufacturing, cables, oils, tire and textile and garments factory nearby the study beel of Gazipur district. Interestingly, Hg was found below detection limit at the study area. As a whole, the present study was compared with some previous literatures which studied in different areas of Bangladesh (Table 2).

Metal concentration in vegetables

The highest amount of Cr was observed in Paddy (38.02 mg/kg) and the lowest amount in Pumpkin (4.3 mg/kg) (Table 3) in the vegetable samples.

Table 3: Heavy metal concentrations (mg/kg) in vegetables grown in and around Mokesh Beel area with safety guidelines and some relevant studies in Bangladesh

Samples	Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb
Gourd Spinach	9.04	208.81	948.45	12.14	20.83	710.36	1.43	0.83	0.77	37.62
Bottle Gourd	16.1	58.81	619.29	38.47	21.22	3170.42	1.16	1.69	0.76	48.18
Pumpkin	4.3	25.25	245.96	11.31	11.42	882.72	1.06	1.08	1.21	40.29
Pumpkin Spinach	3.8	92.53	552.73	11.27	22.86	305.01	1.07	0.54	0.33	14.62
Tomato	8.7	28.57	738.17	4.34	18.42	3184.55	1.16	1.29	0.65	19.87
Red Spinach	20.04	57.91	1615.71	9.29	23.27	818.35	1.33	0.83	0.47	497.99
Paddy	38.02	44.23	520.39	98.9	95.36	79.07	0.96	0.4	1.09	623.17
Average	14.29	73.73	748.67	26.53	30.48	1307.21	1.17	0.95	0.75	183.11
± SD	± 12.03	± 63.68	± 438	± 33.77	± 28.89	± 1309	± 0.16	± 0.44	± 0.31	± 260.6
FAO/WHO, (1984)	5	-	450	20	40	60	-	0.3	-	5
Awashthi, (2000)	20	-	-	1.5	30	50	-	1.5	-	2.5
FAO/WHO, (2001)	2.3	-	-	-		-	0.1	0.2	0.03	0.3
SEPA, (2005)	0.5	-	-	10	20	100	-	0.2	-	9
Islam et al., (2015b)	1.05	-	-	2.15	3.25	-	0.42	0.165	-	0.7
Rahman et al., (2013)	0.98	124	-	2.1	18	55	0.113	0.134	-	3.1

Ahmad & Goni, (2010)	6.65	-	263.79	11.88	38.52	29.61	-	2.47	-	15.55
Goni et al., (2014)	8.24	-	745.62	16.81	24.2	51.86	-	3.4	-	17

The mean concentration of Cr was observed 14.29 mg/kg which exceeded all the safe limit (Table 3) except Awashthi (2000). In the literature, Cr contents were reported as 0.6 mg/kg (Rahman et al., 2013) and 7.90 mg/kg in vegetable samples (Maleki and Zarasvand, 2008). The lowest and highest amounts of Mn were found in Pumpkin (25.25 mg/kg) and Gourd Spinach (208.81 mg/kg), respectively among the vegetable samples (Table 3). In previous studies, mean Mn concentration was found 65 mg/kg (Rahman et al., 2013) and ranging from 0.18 to 2.8 mg/kg (Singh and Taneja, 2010). Mean concentration of Fe was 748.67 mg/kg (245.96-1615.71 mg/kg) which exceeded the FAO/WHO, (1984) standard (Table 3). The present study showed higher range of Fe concentrations than those of vegetables observed (111–378 mg/kg)by Arora et al., (2008). The minimum and maximum Ni levels were found as 4.34 mg/kg in Tomato and 98.9 mg/kg in Paddy, respectively (Table 3) among the studied vegetables and crop samples. The average concentration of Ni was observed 26.53 mg/kg which exceeded all the international standard (Table 3). In a Nigerian study, Sobukola et al., (2010) reported that mean concentration of Ni was 0.24 ± 0.01 mg/kg in selected different leafy vegetables. From the vegetable samples, Cu concentration varied in the range of 11.42-95.36 mg/kg, and decreased in the following order: Paddy > Red Spinach > Pumpkin Spinach > Bottle Gourd > Gourd Spinach > Tomato > Pumpkin (Table 3). The mean concentration of Cu found 30.48 mg/kg which was below the international safe limit (Table 3) except Paddy (95.36 mg/kg).

Zn concentration varied in the range of 79.07-3184.55 mg/kg with the lowest value in Paddy and highest in Tomato (Table 3). The average concentration of Zn was observed as 1307.21 mg/kg which exceeded all the international safe guideline (Table 3). In the literatures, median Zn concentration in the vegetable samples was found to be 50 mg/kg in a severely arsenic contaminated area of Bangladesh (Rahman et al., 2013) and in the range of 19.54-42.06 mg/kg in another study found (Ahmad and Goni, 2010). Arsenic content in vegetable samples was found lower in Paddy (0.96 mg/kg) and higher in Gourd Spinach (1.43 mg/kg) (Table 3). Mean concentration of As was found in vegetable samples 1.17 mg/kg which was higher than the safe limit (0.1 mg/kg) (FAO/WHO, 2001; Table 3). Previous studies have reported that As concentration in all vegetables imported from Bangladesh were found to be 0.0545 mg/kg (Rmalli et al., 2005). The lowest and highest Cd content in vegetable and crop samples were found in Paddy (0.4 mg/kg) and Bottle Gourd (1.69 mg/kg), respectively (Table 3). The mean value of Cd in vegetable samples was 0.95 mg/kg which exceeded the permissible limit of FAO/WHO, (1984), FAO/WHO, (2001) and SEPA, (2005) (Table 3). Rmalli et al., (2012) reported that Cd levels in nonleafy vegetables was (0.008 mg/kg) and Sobukola et al., (2010) observed that Cd level as 0.21 mg/kg in selected vegetables grown in Nigeria. Cadmium is apparent not essential element which is absent at birth but accumulate at gizzard and that occur with increasing of age (Khalafallah et al., 2011). The average value of Hg was observed 0.75 mg/kg (lowest 0.33 mg/kg in Pumpkin Spinach and highest 1.21 mg/kg in Pumpkin) which exceeded too much from the FAO/WHO, (2001) safe standard. In vegetables, Pb was found to be the highest in Paddy (623.17 mg/kg) and lowest in Pumpkin Spinach (14.62 mg/ kg) (Table 3). The mean concentration of Pb in vegetables was 183.11 mg/kg which exceeded all the international safe standard (Table 3). Pb concentration of the vegetable samples ranged from 3E-05 to 7E-04 mg/kg in a study on 24 different types of vegetables grown in the Chapai Nawabganj district of Bangladesh (Islam et al., 2015c). The present study was compared with three recent studies of vegetables in Bangladesh (Islam et al., 2015c, Rahman et al., 2013, Ahmad and Goni, 2010) at Table 3.

Assessment of metal pollution

To evaluate the metal pollution in soil and vegetables, contamination factor (CF), pollution load index (PLI) and geo-accumulation index (I_{geo}) were calculated which showed in Table 4 and Table 5 respectively.

Contamination factor (CF) and pollution load index (PLI)

The average contamination factor (*CF*) for all metals were in the descending order of: Zn > Hg > Pb > Cd > Cu > Ni > Mn > As > Cr > Fe. The present study showed, Cr, Mn, Fe, Ni, Cu and As were a CF < 1 which indicate lower degree of pollution in vegetable samples except Ni in Bottle Gourd and Paddy as well as Cu in Gourd Spinach and Bottle Gourd ($1 \leq CF \leq. 3$, moderate degree). Pb in Bottle Gourd, Gourd Spinach and Pumpkin showed moderate degree of pollution where Red Spinach and Paddy observed a very higher degree of pollution ($CF \geq 6$). Unfortunately, Zn, Cd and Hg in all vegetables were observed very higher degree of pollution.

The analyzed pollution load index (*PLI*) values of metals in vegetables were presented in Table 4.

Table 4: Contamination factor (*CF*) and pollution load index (*PLI*) of heavy metals from irrigated soil to vegetables in Mokesh Beel

	Contamination factor (*CF*)										*PLI*
	Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb	
Gourd Spinach	0.23	0.91	0.05	0.63	1.71	137.40	0.37	9.22	77.00	1.77	1.93
Bottle Gourd	0.42	0.26	0.03	1.99	1.74	613.23	0.30	18.78	76.00	2.27	2.43
Pumpkin	0.04	0.17	0.01	0.28	0.40	13.44	0.41	6.35	121.00	1.03	0.73
Pumpkin Spinach	0.03	0.64	0.03	0.28	0.80	4.64	0.41	3.18	33.00	0.37	0.63
Tomato	0.14	0.33	0.04	0.12	0.84	86.89	0.43	8.60	65.00	0.63	1.10
Red Spinach	0.35	0.21	0.09	0.49	0.90	5.31	0.50	3.95	47.00	34.90	1.49
Paddy	0.68	0.33	0.03	2.89	4.78	2.13	0.23	2.00	109.00	19.40	1.66
Average	0.27	0.41	0.04	0.95	1.60	123.29	0.38	7.44	75.43	8.63	-

Pumpkin and Pumpkin Spinach were observed no pollution (*PLI*< 1) where other vegetables showed high pollution index (*PLI*> 1). Bottle Gourd had showed most pollution load index (Fig. 2).

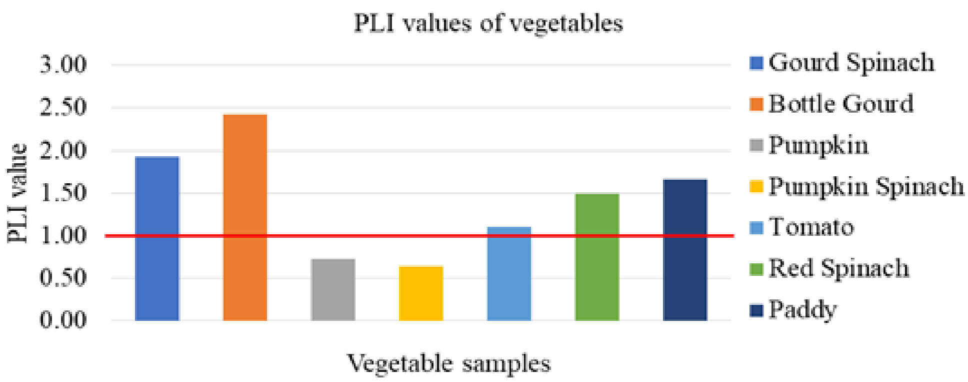

Fig. 2: Pollution load index (PLI) value of heavy metals in vegetables of Mokesh Beel, Gazipur, Bangladesh. Horizontal red line indicates the baseline of pollutants.

The *PLI* can provide some understanding to the inhabitants about the quality of the environment. In addition, it also provides valuable information to the decision makers on the pollution status of the area (Suresh et al., 2012).

Geo-accumulation index (I_{geo})

The geo-accumulation factor (I_{geo}) values of the studied metals were shown in Table 5.

Table 5: Geo-accumulation (I_{geo}) factor of heavy metals in vegetables of Mokesh Beel

	Geo-accumulation factor (I_{geo})									
	Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb
Gourd Spinach	0.71	1.03	0.74	0.95	1.28	3.54	0.60	-0.25	-0.10	1.25
Bottle Gourd	0.85	0.82	0.69	1.29	1.29	4.27	0.48	-0.61	-0.10	1.32
Pumpkin	0.42	0.73	0.61	0.76	0.83	1.63	0.55	-0.56	-0.21	1.08
Pumpkin Spinach	0.39	0.97	0.68	0.76	1.02	1.40	0.56	-0.06	0.10	0.83
Tomato	0.63	0.83	0.71	0.54	1.03	2.19	0.61	-0.64	-0.06	0.96
Red Spinach	0.83	0.79	0.79	0.87	1.05	1.36	0.71	-0.44	0.01	2.25
Paddy	0.98	0.84	0.67	1.34	1.55	1.26	0.35	0.19	-0.19	1.84

Among the studied heavy metals, the I_{geo} values of Cr, Mn, Fe, Ni and As were observed between $0 \leq I_{geo} \leq 1$ indicating uncontaminated to moderately contaminated status of the vegetables. On the other hand, the range of I_{geo} values for Cu, Zn and Pb were 0.83-1.55, 1.26-4.27 and 0.83-2.25 respectively indicate moderately to heavily contaminated status. Interestingly, the I_{geo} values for Cd and Hg were obtained below zero (≤ 0) which indicate no contamination of vegetables by these two metals. However, the highest I_{geo} value was observed for Zn in Bottle Gourd (4.27) which might be due to the higher concentration in vegetable and lower concentration in background sample (soil).

Human health risk estimation

To estimate the human health risks from the studied vegetable samples, estimated daily intake (*EDI*), target hazard quotient (*THQ*), total target hazard quotient (*TTHQ*) and target cancer risk (*TR*) were calculated which were presented in Table 6, Table 7 and Table 8.

Estimated daily intake (*EDI) of heavy metals*

The *EDI* of heavy metals (Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Hg and Pb) was calculated according to concentration of each metal in vegetables and the respective consumption rates. The *EDI* and maximum tolerable daily intake (MTDI) of studied metals from consumption of vegetables were shown in Table 6.

Total daily intake of Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Hg and Pb were 0.04, 0.23, 2.29, 0.08, 0.09, 4, 0, 0, 0 and 0.56 mg/day, respectively. Daily intakes of all the metals were less than the MTDI. In vegetable samples, mean values of *EDI* decreased in the following order: Zn > Fe > Pb > Mn > Cu > Ni > Cr > As > Cd > Hg.

Table 6: Comparison of the estimated daily intake (*EDI*) of heavy metals from highly consumed vegetables with corresponding maximum tolerable daily intake (*MTDI*) in the Mokesh Beel area population.

	Consumption rate (g/day/person)	Estimated daily intake (*EDI*) toxic heavy metals (mg/kg)									
		Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb
Gourd Spinach	130	3.95E-03	9.12E-02	4.14E-01	5.30E-03	9.10E-03	3.10E-01	6.25E-04	3.63E-04	3.36E-04	1.64E-02
Bottle Gourd	130	7.03E-03	2.57E-02	2.71E-01	1.68E-02	9.27E-03	1.38E+00	5.07E-04	7.38E-04	3.32E-04	2.10E-02
Pumpkin	130	1.88E-03	1.10E-02	1.07E-01	4.94E-03	4.99E-03	3.86E-01	4.63E-04	4.72E-04	5.29E-04	1.76E-02
Pumpkin Spinach	130	1.66E-03	4.04E-02	2.41E-01	4.92E-03	9.99E-03	1.33E-01	4.67E-04	2.36E-04	1.44E-04	6.39E-03
Tomato	130	3.80E-03	1.25E-02	3.22E-01	1.90E-03	8.05E-03	1.39E+00	5.07E-04	5.63E-04	2.84E-04	8.68E-03
Red Spinach	130	8.75E-03	2.53E-02	7.06E-01	4.06E-03	1.02E-02	3.57E-01	5.81E-04	3.63E-04	2.05E-04	2.18E-01
Paddy	400	1.66E-02	1.93E-02	2.27E-01	4.32E-02	4.17E-02	3.45E-02	4.19E-04	1.75E-04	4.76E-04	2.72E-01
Total daily intake from vegetables	-	0.04	0.23	2.29	0.08	0.09	4.00	0.00	0.00	0.00	0.56
MTDI	-	0.2	2-5	8	0.3	30	60	0.13	0.021	0.011	0.21

Non-carcinogenic risk

The non-carcinogenic risk was assessed by target hazard quotient (*THQ*) and total target hazard quotient (*TTHQ*) of ten heavy metals in vegetables showed in Table 7.

Table 7: Target hazard quotient (*THQ*) and total target hazard quotient (*TTHQ*) of heavy metals from consuming vegetables in Mokesh Beel.

	Target hazard quotient (*THQ*)										*TTHQ*
	Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb	
Gourd Spinach	2.63E-03	6.51E-01	5.92E-01	2.65E-01	2.27E-01	1.03	2.08	3.63E-01	1.12	4.69	11.03
Bottle Gourd	4.69E-03	1.83E-01	3.86E-01	8.40E-01	2.32E-01	4.62	1.69	7.38E-01	1.11	6.01	15.81
Pumpkin	1.25E-03	7.88E-02	1.53E-01	2.47E-01	1.25E-01	1.29	1.54	4.72E-01	1.76	5.03	10.70
Pumpkin Spinach	1.11E-03	2.89E-01	3.45E-01	2.46E-01	2.50E-01	4.44E-01	1.56	2.36E-01	4.80E-01	1.82	5.67
Tomato	2.53E-03	8.91E-02	4.61E-01	9.48E-02	2.01E-01	4.64	1.69	5.63E-01	9.46E-01	2.48	11.16
Red Spinach	5.84E-03	1.81E-01	1.01	2.03E-01	2.54E-01	1.19	1.94	3.63E-01	6.84E-01	62.15	67.98
Paddy	1.11E-02	1.38E-01	3.25E-01	2.16	1.04	1.15E-01	1.40	1.75E-01	1.59	77.77	84.72

THQs of Fe in Red Spinach; Ni and Cu in Paddy; Zn in Bottle Gourd, Pumpkin and Red Spinach; As in all vegetables; Hg in all vegetables except Pumpkin Spinach, Tomato and Red Spinach; and Pb in all vegetables exceeded the safe standard 1 (USEPA, 1989, 2011, 2015). Moreover, *TTHQs* were found to be >1 in all vegetables (Table 7), indicating the potential non-carcinogenic health risk of its consumption (Fig. 3).

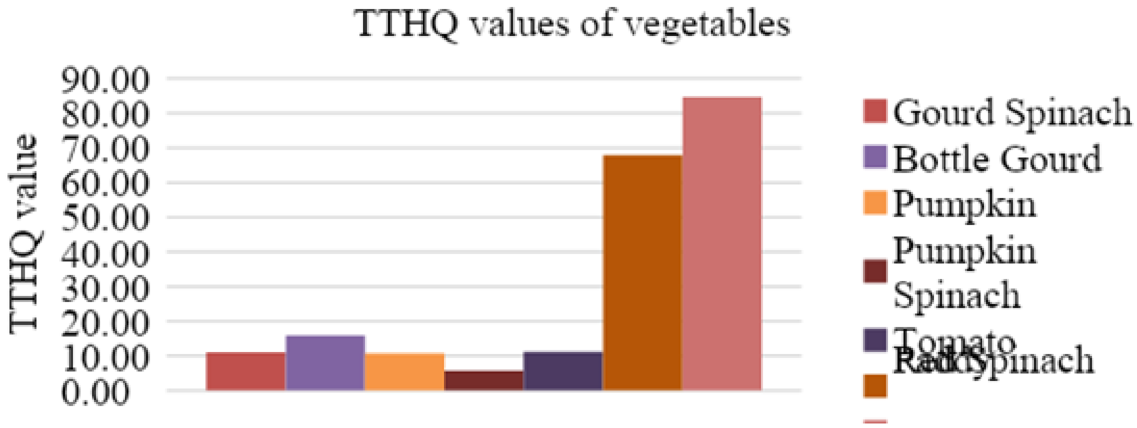

Fig. 3: Total target hazard quotient (TTHQ) of heavy metals from consuming vegetables. Horizontal red line indicates the safe standard of hazard quotient.

Consumption of these vegetables, people in Mokesh Beel area, would be experienced significant non-carcinogenic health risks of ingesting over a 60 years lifetime.

Carcinogenic risk

The target cancer risks (*TRs*), derived from the intake of As and Pb, were calculated in Table 8, since other heavy metals usually don't show carcinogenicity.

Table 8: Target cancer risk (*TR*) of heavy metals due to consumption of vegetables in Mokesh Beel

	TR	
Samples	As	Pb
Gourd Spinach	9.37E-04	1.40E-04
Bottle Gourd	7.60E-04	1.79E-04
Pumpkin	6.95E-04	1.50E-04
Pumpkin Spinach	7.01E-04	5.43E-05
Tomato	7.60E-04	7.38E-05
Red Spinach	8.71E-04	1.85E-03
Paddy	6.29E-04	2.31E-03
Total TR	5.35E-03	4.76E-03
USEPA, (1989, 2011, 2015)	1×10^{-6} to 1×10^{-4}	

All the values of *TR* for As and Pb exceeded the safe standard value 1E-06 to 1E-04 (USEPA, 1989, 2011, 2015) except Pb in Gourd Spinach, Bottle Gourd, Pumpkin, Pumpkin Spinach and Tomato. Comparing the *TR* values with guideline values in the analyzed samples with standard guideline values, indicates that vegetables from Mokesh Beel studied area are not safe for the people.

Anova test and correlation co-efficient (r)

There were no significant ($p < 0.05$) difference in metal concentrations among vegetable samples. On the contrary, concentration of all heavy metals with different vegetable samples were significantly ($p > 0.05$) different.

The correlation co-efficient (r) measures the strength and direction of a linear relationship between two variables on scatter plot. The Pearson coefficient for vegetables parameters was used to calculate the correlation between trace metals concentrations and to establish the mutual influence and the results are presented in Table 9.

Table 9: Correlation co-efficient (*r*) between the heavy metals in vegetables of Mokesh Beel

	Cr	Mn	Fe	Ni	Cu	Zn	As	Cd	Hg	Pb
Cr	1									
Mn	-0.21	1								
Fe	0.17	0.26	1							
Ni	0.88*	-0.20	-0.28	1						
Cu	0.90*	-0.15	-0.14	0.94*	1					
Zn	-0.22	-0.29	-0.03	-0.26	-0.42	1				
As	-0.28	0.69*	0.73*	-0.56*	-0.50	0.10	1			
Cd	-0.33	-0.23	-0.07	-0.34	-0.57*	0.91*	0.19	1		
Hg	0.30	-0.24	-0.56*	0.49	0.36	-0.10	-0.39	0.06	1	
Pb	0.90*	-0.25	0.39	0.67*	0.77*	-0.46	-0.17	-0.53*	0.16	1

* Values > 0.5 or < -0.5 are significantly correlated

Some metals in vegetables showed significant correlation with each other suggesting similar sources of input (human or natural) for these metals in the beel water (Bastami et al., 2012) where Mn and Pb didn't showed any significant correlation. In the present study, the correlations between Ni-Cr, Cu-Cr, Cu-Ni, Pb-Cr, Cd-Zn, As-Fe and Pb-Cu were observed highly correlated. High correlations between specific heavy metals in water may reflect similar levels of contamination and/or release from the same sources of pollution, mutual dependence and identical behavior during their transport in the river or beel system into the soil (Li et al., 2009; Chen et al., 2012; Suresh et al., 2012; Jiang et al., 2014).

IV. Conclusions

Heavy metal is a major problem for the industrial area like Mokesh Beel at Gazipur. The level of heavy metals found in different sources of the present study were compared with the prescribed safe limit provided by WHO, (1971, 2004); SEPA, (2005); EU, (2002); CNEMC, (1990); Pescod, (1992); FAO, (1975); EPA-PR China, (2002); FAO/WHO (1994, 2001) and other studies. In the present study, Cr, Mn, Cd and Pb were found higher concentration than the international safe standard (WHO, 2004; Pescod, 1992). The concentration of Fe and As in soil exceeded the previous studies (Rahman et al., 2014; Ahmad et al., 2010). Most of the metals in collected vegetables were too much higher than the international safe limit (FAO/WHO, 1984, 2001; Awashthi, 2000). Moreover, Pb was found maximum concentration among vegetables. The contamination factor (*CF*), pollution load index (*PLI*) and geo-accumulation factor (I_{geo}) revealed that vegetables were uncontaminated to extremely contaminated by heavy metal in that area. In addition, As, Hg and Zn showed a higher *THQ* value than the safe limits where *TTHQ* values were too much higher than the safe standard in all vegetables. Target cancer risks (*TRs*) values of As in all vegetables and Pb in Red Spinach and Paddy showed carcinogenic possibility. Finally, it is cleared that water, soil and vegetables in Mokesh Beel are highly contaminated by heavy metals. Thus, long-term consumption of vegetables from this area can cause different carcinogenic and non-carcinogenic diseases among the people.

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