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Energy dispersive X-ray fluorescence detection of heavy metals in Bangladesh cows' milk

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Abstract

It is considered that cow's milk is almost complete food for human as it provides most of the micronutrients and macronutrients. The cow's milks are essential for the growth and development especially for children. The main compositions of cow's milk are protein, fat, carbohydrates, vitamins and minerals which are well defined. Presently, the study of micronutrients and toxic elements in cow's milk has been widely carried out particularly in the industrialized and polluted regions because of its possibility of contamination, and thereby health risk of the consumers. The elemental composition in local cow's milk samples in Bangladesh is not well studied yet. The present study was therefore aimed to determine the level of heavy metals (Cr, Ni, As, Cd, Hg, Pb, Mn, Cu, Zn and Fe) in cow's milk using EDXRF technique. Subsequently, the experimental data was used to calculate the human health risk through the intake of both powder and liquid cows' milk available in Bangladesh. The results showed that powder milk contains significantly higher concentration of heavy metals than liquid milk samples. The HRI (health risk index) and HI (hazard index) values for most of the elements in all milk samples were within the safe limit (<1.0) or close to safe limit (≤ 1.0) with an

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exception of Hg. However, HRI value for Hg in powder milk samples for both children and adult showed a value higher than one (>1). MPI (metal pollution index) value for powder milk samples are very high compared to other type of milk samples analyzed in this study. Therefore, it has been suggested that heavy metal contamination through local powder milk samples might have significant negative impact (threat) on human health.

Keywords: Food science, Analytical chemistry

1. Introduction

Agriculture, industry and transportation produce huge amount of wastes and different types of pollution every day due to civilization and modernization as well (Rahman et al., 2014). Nowadays, pollution becomes increasingly prevalent in our daily life and it has a fairly negative impact on our health (Rahman et al., 2012). Among all the pollutions, these days heavy metals contamination are a universal problem as they are not bio-degradable and when exceeded permissible level (Addo et al., 2012) most of them create toxicity to living organisms.

Soil, air, water are traditionally using as sites for disposal of industrial waste, agricultural chemicals and transportation waste, and thus they have got polluted by heavy metals. Polluted environment supplies heavy metal into food chain. Plants as essential components of natural ecosystems and agro systems represent the first compartment of the terrestrial food chain. Due to their capacity to accumulate toxic elements, they grow and survive on contaminated soil (Ana-Irina et al., 2008). When these plants are taken by any animal as food, heavy metal can enter into the animal body and ultimately into the human body. Heavy metals like Cu, Fe, Zn and Mn are essential to maintain proper metabolic activities in living organisms; whereas Pb, Cd, Cr, Hg, Ni and As are non-essential, have no biological impact and hence at high concentrations, all they can cause toxicity in human body (Abdulkhaliq et al., 2012).

Milk is a complex food and a bioactive substance which enhances growth and development of mammalian infants. FAO estimates that 85% of milk is commercially available worldwide were produced from cows (FAO, 2008). It is a good source of energy, water, carbohydrate, fat, protein, sugars, vitamins, minerals, minor biological proteins and enzyme (Milk Fact, 2015) and hence considered as a nearly complete food. It contents macro-elements (Ca, Mg, Mn, K and P) and microelements (Cu, Fe, Zn, Na, Se) but sometimes additional amounts of contaminant metals might enter milk, increase the levels that are harmful to human (Milk Fact, 2015). In Bangladesh usually people get cow's milk from direct farm house in liquid form or as packaging products of different brands (both liquid and powder form) from local market. It should be noted that maximum commercial farm houses are established near to industrial area or bank of rivers, which might

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be receiving industrial effluents. On the other hand, the farm owner feed cows mainly green fodder (grass, leaves, water hyacinth etc.), rice straw etc. grown near the field of river or lake, which are contaminated with heavy metals and thus cow's milk gets contaminated by heavy metals.

In Bangladesh per capita need of milk is assumed to be 250 ml/day but the availability of milk is only 32.6 ml/day. Consequently, consumers face an acute shortage of milk for which supply fails to meet the requirements of 85% of the population (FAO, 2008). According to FAO, average annual milk production in Bangladesh is 2,264,000 tones and annually only 13 kg of milk is available for every person (FAO, 2008). Total milk produced is mostly consumed by farming households or sold on the informal market, and less than 20% is transferred to the formal milk processing sector (pasteurized and ultrahigh temperature) (Saadullah, 2000).Thus low production results in the import of bulk amount of powdered milk. On the other hand, due to less supply of milk and sometimes for more business profit some businessmen mix water with raw milk. In most cases they use contaminated water collected from nearby water body and thus milk gets contaminated especially with heavy metals. Even in transportation or manufacturing or packaging process, due to unsafe handling, milk also gets contaminated by heavy metals. As milk is a novel food and it provides more nutrients, it should not be contaminated. This work was sketched to assess the status of heavy metals (Cr, Ni, As, Cd, Hg, Pb, Mn, Cu, Zn and Fe) in various type of milk samples and to evaluate the pollution load and health risk that may be associated by consuming the milk samples.

2. Materials and methods

2.1. Selection of study area

Commercially available milks (both powder and liquid) were collected mainly from different shops of Dhaka City. Farm-house milks were collected from the farms near the bank of the Buriganga River, industrial disposal areas of Keraniganj Upazilla and Dhaka Metropolitan area of Dhaka District in Bangladesh. The areas of sample collection are shown in Fig. 1 and details of which are given in Table 1.

2.2. Sample preparation

2.2.1. Preparation of powder sample

All powdered milk samples were oven dried at around 70 °C for overnight and continued until a constant weight was obtained. The dried mass of each sample was ground to fine powder using a carbide mortar and pestle, preserved in a plastic vial with the identification mark inside desiccators until irradiation.

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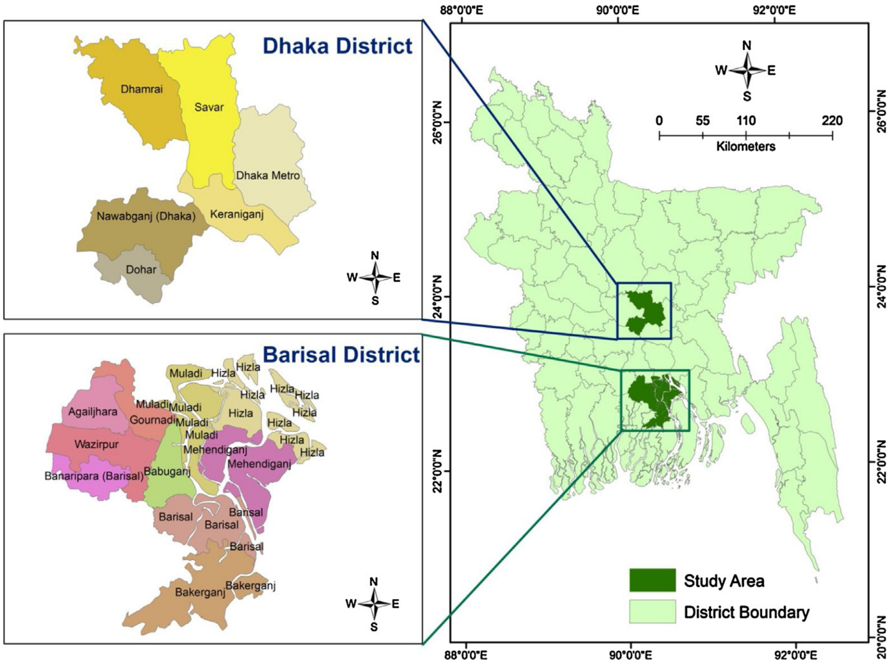

Fig. 1. Location of milk sampling in Dhaka District and Barishal District of Bangladesh.

2.2.2. Preparation of liquid sample

100 ml liquid cow's milk samples were centrifuged by cooling centrifuge machine maintaining the speed 400 rpm for 15–20 minutes to remove fat from the sample. All fat free liquid milk samples, each with a particular identification number were first dried in water bath at 100 °C and finally in an oven at around 70 °C for overnight until a constant weight were obtained. Each dried sample was then ground to fine powder using carbide mortar and pestle and preserved in a plastic vial with the identification mark inside desiccators up to analyses.

2.3. Sample analysis

Energy Dispersive X-ray Fluorescence (EDXRF) Spectrometer (Model: Epsilon 5, PANalytical, The Netherlands) was the major analytical technique used to determine the concentration of elements in various milk samples.

2.3.1. Irradiation of milk samples with X-ray beam

2 gm of each powdered milk samples was pressed into a pellet of 25 mm diameter with a pellet maker (Automatic Hydraulic Presses, model: 3889-4NEI) using 10 tons of pressure. The sample pellets were loaded into the X-ray excitation chamber for irradiation with the help of automatic sample changer system. A time-based program, controlled by a software package provided with the systems was used to irradiate the real samples and the standard materials as well for the construction of

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Table 1. Identification of all samples.

Samples	Sample ID	Description of brand/Farm House	Milk form
Pran Milk Powder	PP	Pran dairy Ltd. (production & packaging)	Powder
Marks Milk Powder	MP	Mare golbarn co-operative Co. Ltd. (production) Abulkhyar milk products Ltd. (Packaging)	Powder
Fresh milk powder	FP	Production in Australia ThanvirfoodLtd.Megna group (Packaging)	Powder
Danish milk Powder	DsP	Danish milk Bangladesh, Partex group (Production & Packaging)	Powder
Dano milk powder	DnP	Arolla foods, Amba, Denmark (Production) Arolla foods, Bangladesh (Packaging)	Powder
Nido milk powder	NP	Fonter Ltd. Newziland (Production) Nesle Bangladesh (Packaging)	Powder
Diploma milk powder	DiP	Production in Australia Newzealand dairy products Bangladesh Ltd. Rupgonj, Narayangonj (Packaging)	Powder
Arong Pasteurized milk	APL	BRACK food Products	Liquid
Pran Pasteurized milk	PPL	Pran dairy Ltd (production & packaging)	Liquid
Milk vita Pasteurized milk	MPL	Bangladesh DugdhoUtpadonkariSomobay Union Ltd.	Liquid
Fresh Pasteurized milk	FPL	Akij Food and Beverage Ltd.	Liquid
Arong UHT milk	AUL	BRACK dairy & food products	Liquid
Fresh UHT milk	FUL	Akij Food and Beverage Ltd.	Liquid
Pran UHT milk	PUL	Pran dairy ltd (production & packaging)	Liquid
Dairy farm 1	F_1L	Jaker dairy farm, Mohammadpur, Dhaka	Liquid
Dairy farm 2	F_2L	K-1, Keranigong, Dhaka	Liquid
Dairy farm 3	F_3L	K-2, keranigong, Dhaka	Liquid
Dairy Farm 4	F_4L	K-3. Keranigong, Dhaka	Liquid
Dairy farm 5	F_5L	K-4, keranigong. Dhaka	Liquid
Dairy farm 6	F_6L	Polasnogor dairy farm, Mirpur, Dhaka	Liquid
Dairy farm 7	F_7L	Chain dairy farm, Mirpur, Dhaka	Liquid
Dairy farm 8	F_8L	Shipon, Barishal	Liquid
Dairy Farm 9	F_9L	Alauddin, Barishal	Liquid

the calibration curves for quantitative elemental analysis in the respective samples and afterwards the generated X-ray spectra of the materials were stored into the computer.

2.3.2. Construction of calibration curve for validation of the method

EDXRF technique was used for the measurement of elemental concentration (Islam and Jolly, 2007) that is a direct comparison method in which standards are set to construct the calibration curves. In this method, both the standard and samples have to be of similar matrix, to produce identical sensitivity and thus matrix effects are nullified. Three lab-synthesized cellulose-based multi element standards (Cellu-1, Cellu-2, Cellu-3), were used to construct the calibration curves (Ali et al., 1985) on the basis of K X-ray and L X-ray line sensitivity as a function of its atomic number for carrying out elemental analysis in milk samples. Accuracy of the curves were justified by analyzing a groundwater sample under the constructed calibration curve and comparing the result by analyzing the same ground water sample with another method, TXRF (Total reflection X-ray Fluorescence) respectively. The results obtained for elements of interest in both the methods are shown in Table 2. All the results were found within the acceptable limit.

2.4. Data analysis

2.4.1. Calculation of Metal Pollution Index (MPI)

Metal pollution index (MPI) index was calculated following Ureso et al. (1997), which is the geometrical mean of concentrations of all the metals in the

Table 2. Elemental Concentrations of a Groundwater Sample measured by two different methods.

Elements	Elemental Concentrations in Groundwater (μg/ml)		
	EDXRF values	TXRF Values	Error (%)
K	6.24	5.64	9.62
Ca	82.36	86.87	−5.48
Ti	0.25	0.22	12.0
Mn	0.78	0.79	−1.28
Fe	0.96	1.06	−10.42
Ni	0.023	0.021	8.69
Zn	0.015	0.013	13.33
As	0.019	0.02	−5.26
Sr	0.358	0.322	10.06

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corresponding milk samples and represented by the following the equation (Eq. (1)) given below.

$$MPI\ (\mu g\ g^{-1}) = (Cf_1 \times Cf_2 \times \ldots \ldots \ldots \ldots \times Cf_n)^{1/n} \quad (1)$$

Cf_n represents the concentration of metal n in the measured sample.

2.4.2. Calculation of Daily Intake of Metals (DIM)

Daily intake of metal (DIM) has been calculated following Cui et al. (2004) (Eq. (2)).

$$DIM = \frac{(C_{metal} \times D_{food\ \ intake})}{B_{average\ \ weight}} \quad (2)$$

In this equation, C_{metal}, $D_{foodintake}$, and $B_{averageweight}$ stands for the heavy metal concentrations in milk (μg g $-^{1}$), daily intake of milk and average body weight respectively.

The required amount of milk for both children and adult is 300–800 ml/day based on Ca requirement in human body (ICUSD, 2015). In this study the maximum means 800 ml/day intake of liquid milk for both children and adult are considered. Again for powder milk 25 g is equivalent to 200 ml liquid milk, which was leveled in all the samples packets of powder milk. In this sense the required amount of powder milk for both children and adult is 37–100 g/day. So, 800 ml daily intake of liquid milk and 100 g of powder milk are considered for calculation of daily intake of heavy metals through milk consumption. WHO suggested the average body weight for adult is 70 kg and in this study the average body weight of children is taken 30 kg (1/3 of adult weight) (WHO, 1993).

2.4.3. Calculation of Health Risk Index (HRI)

Health Risk Index (HRI) is calculated on the basis of daily intake of metal (DIM) through food (milk) consumed and oral reference dose (Rf_D) suggested (Cui et al., 2004). Rf_D is estimated as per day exposure of metal to human body that has no hazardous effect during life time (US-EPA IRIS, 2006). Oral reference dose for Cr, Ni, Cu, Pb, Cd, Mn and Zn were 1.5, 0.02, 0.04, 0.004, 0.001, 0.033 and 0.30 (mg/kg bw/day) respectively (US-EPA IRIS, 2006); 10–60 (mg/kg bw/day) for Fe (US-EPA IRIS, 1998); 0.002 (mg/kg bw/day) for Hg (Friberg et al., 1994). The Reference Dose (Rf_D) for inorganic arsenic is 0.0003 (mg/kg bw/day) based on hyper pigmentation, keratosis and possible vascular complications in human (USEPA, 2002). Health Risk Index (HRI) for Cr, Ni, As, Cd, Hg, Pb, Mn, Cu, Zn and Fe by consumption of milk was calculated by following Cui et al. (2004) which is expressed by the Eq. (3).

$$HRI = DIM/Rf_D \quad (3)$$

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According to Food and Nutritional Board (2004), an index more than 1 is considered unsafe for human health.

2.4.4. Calculation of Hazard Index (HI)

The hazard index (HI) has been developed (US-EPA IRIS, 1989) to calculate the potential risk to human health through more than one heavy metal and estimated by the summation of the hazard quotients/health risk index (HRI) as described in the following equation (Eq. (4)).

$$HI = \Sigma HQ = HQ_{Cr} + HQ_{Mn} + HQ_{Ni} + HQ_{Cu} + HQ_{Zn} + HQ_{As} + HQ_{Cd} + HQ_{Hg} + HQ_{Pb} + HQ_{Fe} \quad (4)$$

It is considered that, sum of the multiple of metals exposure is proportional to the magnitude of the adverse effect. In this study, the hazard index for the toxic element Cr, Ni, As, Cd, Hg and Pb has been calculated to measure the toxicity of the milk items.

3. Result and discussion

3.1. Level of heavy metals in packaged liquid market milk samples

The mean, maximum, minimum level of the studied heavy metals in packed liquid milk samples are given in Table 3. The concentration of Cr in milk samples indicated that there was no statistically significant differences ($F_{cal.} = 0.333 < F_{crit} = 3.682$; $\alpha = 0.05$; $p = 0.772$) in different brands of packaged liquid market milk samples. This study also revealed that the concentration of Cr in MPL sample was too low to be detected by the systems (BDL 0.22 µg/ml). On the other hand FUL contains highest Cr concentration. It might be happened due to the reasons that the ultrahigh temperature treated milk samples contain more Cr than pasteurized milk samples. Zodape et al. (2012) reported that the concentration of Cr in cow's milk collected from Mumbai City, India was 0.175 µg/ml, which agreed well with the average Cr concentration found in the present study. In another study, Islam et al. (2015) reported to find 1.6 µg/ml of Cr in milk sample of Bangladesh around the vicinity of industries, which were much higher than the present values. The average concentration of Ni in different brands of milk samples was found to be 0.081 µg/ml ranging from 0.061 to 0.104 µg/ml. However, the *F* test revealed that the value of $F = 0.353$ found for this experiment was much larger than $F_{2,18,0.05} = 3.554$, which indicated that there was no statistically significant difference for Ni concentration in the different brands of packaged liquid market milk samples. On the other hand, commercial packed milk contains 0.053 µg/ml of Ni in Poland (Dobrzañskiet al., 2005), which was in line with the results found in this study. Reversely, *F*-test revealed that the concentration of As in the different brands of packaged liquid

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Table 3. Concentration of heavy metals in different types of milk samples.

Sample ID	Elemental concentration (µg/ml) in different types of milk samples									
	Cr	Mn	Ni	Cu	Zn	As	Cd	Hg	Pb	Fe
packed liquid milk (µg/ml) sample										
MPL	ND	0.032	0.061	0.065	0.754	0.034	0.010	ND	0.035	0.602
		0.029–0.035	0.058–0.064	0.063–0.067	0.752–0.755	0.031–0.035	ND-0.021		0.023–0.045	0.612–0.592
APL	0.101	0.044	0.071	0.064	0.672	0.034	0.016	0.036	0.046	0.968
	0.095–0.110	0.039–0.046	0.070–0.073	0.062–0.065	0.665–0.683	0.033–0.035	0.014–0.017	0.028–0.044	0.043–0.047	0.911–1.066
PPL	0.119	0.042	0.104	0.060	0.470	0.035	0.003	0.107	0.047	0.685
	0.106–0.129	0.025–0.059	0.098–0.107	0.052–0.067	0.067–0.493	0.034–0.036	ND-0.008	0.098–0.120	0.042–0.050	0.452–1.136
FPL	0.103	0.044	0.071	0.063	0.671	0.034	0.007	0.034	0.044	0.602
	0.095–0.110	0.038–0.046	0.070–0.073	0.060–0.065	0.667–0.683	0.033–0.035	0.002–0.012	0.028–0.044	0.043–0.045	0.612–0.592
AUL	0.170	0.075	0.095	0.062	0.504	0.035	0.006	0.018	0.050	1.491
	0.164–0.176	0.069–0.081	0.092–0.097	0.059–0.065	0.501–0.507	0.033–0.038	0.002–0.010	0.016–0.019	0.043–0.056	1.348–1.633
PUL	0.145	0.070	0.080	0.064	0.504	0.034	0.006	0.028	0.047	1.198
	0.133–0.170	0.068–0.075	0.078–0.095	0.062–0.065	0.486–0.522	0.032–0.035	ND-0.012	0.018–0.030	0.046–0.050	1.190–1.205
FUL	0.195	0.044	0.081	0.063	0.537	0.034	0.013	0.035	0.044	1.162
	0.192–0.199	0.038–0.051	0.079–0.082	0.060–0.065	0.512–0.562	0.033–0.035	0.011–0.015	0.027–0.043	0.043–0.045	1.143–1.181
Farmhouse raw liquid milk (µg/ml) sample										
F_1L	0.120	0.036	0.077	0.055	0.619	0.035	0.015	0.006	0.043	1.078
	0.178–0.201	0.026–0.046	0.073–0.080	0.054–0.055	0.602–0.635	0.034–0.035	0.013–0.016	ND-0.012	0.042–0.044	0.969–1.186
F_2L	0.202	0.043	0.079	0.063	0.708	0.038	0.004	ND	0.040	1.237
	0.192–0.211	0.035–0.050	0.078–0.080	0.060–0.065	0.687–0.735	0.036–0.039	0.003–0.005		0.039–0.042	1.235–1.238
F_3L	0.177	0.079	0.075	0.051	0.995	0.034	0.013	0.006	0.048	0.921
	0.162–0.191	0.081–0.093	0.074–0.075	0.049–0.052	0.989–1.001	0.033–0.034	0.005–0.020	ND-0.012	0.046–0.049	0.895–0.946

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Table 3. (*Continued*)

Sample ID	Elemental concentration (μg/ml) in different types of milk samples									
	Cr	**Mn**	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Hg**	**Pb**	**Fe**
F_4L	0.208	0.025	0.084	0.057	0.992	0.033	0.001	ND	0.048	0.761
	0.190–0.223	0.024–0.025	0.082–0.085	0.056–0.058	0.982–1.002	0.033–0.034	ND-0.002		0.047–0.049	0.741–0.781
F_5L	0.199	0.090	0.081	0.056	0.802	0.034	0.005	ND	0.046	0.829
	0.196–0.203	0.081–0.099	0.079–0.082	0.053–0.059	0.801–0.803	0.033–0.035	0.004–0.006		0.044–0.048	0.805–0.853
F_6L	0.221	0.027	0.080	0.051	0.594	0.033	0.019	0.030	0.043	1.101
	0.216–0.225	0.027–0.028	0.078–0.082	0.048–0.054	0.591–0.596	0.032–0.034	0.018–0.020	0.031–0.028	0.042–0.044	1.094–1.108
F_7L	0.212	0.041	0.079	0.055	0.640	0.034	0.016	0.026	0.044	0.944
	0.205–0.218	0.033–0.049	0.073–0.085	0.052–0.027	0.637–0.643	0.035–0.036	0.012–0.020	0.018–0.034	0.043–0.044	0.923–0.965
F_8L	0.105	0.035	0.072	0.055	0.869	0.033	0.003	0.002	0.046	0.827
	0.089–0.120	0.035–0.035	0.071–0.073	0.052–0.057	0.857–0.884	0.032–0.035	0.001–0.005	ND–0.004	0.043–0.049	0.766–0.883
F_9L	0.158	0.051	0.078	0.059	1.059	0.038	0.013	0.008	0.037	1.138
	0.154–0.161	0.043–0.059	0.076–0.079	0.058–0.059	0.864–1.253	0.037–0.039	0.001–0.025	ND–0.016	0.033–0.040	1.127–1.149
Powder milk (μg/gm) sample										
Dnp	ND	0.921	2.519	2.526	29.516	1.701	ND	2.498	2.257	111.010
		ND-1.711	2.488–2.566	2.285–2.706	24.356–39.767	1.672–1.743		1.416–3.580	1.979–2.408	110.152112.047
FP	ND	1.737	2.421	2.511	27.976	1.766	0.022	4.407	2.265	16.996
		0.232–2.381	2.336–2.554	2.333–2.667	24.168–35.064	1.642–1.969	ND-0.065	3.504–5.31	1.905–2.266	16.742–17.250
DsP	ND	0.653	2.498	2.646	22.474	1.905	1.131	4.314	1.901	46.892
		0.391–0.893	2.476–2.540	2.569–2.747	14.389–26.013	1.822–2.039	ND-1.182	3.326–5.302	1.757–2.117	43.435–48.104
NP	ND	1.109	2.599	2.923	54.772	1.947	0.552	5.201	1.902	100.260
		0.804–1.373	2.511–2.632	2.905–2.951	54.085–55.197	1.858–2.046	0.184–1.131	5.054–5.528	1.773–2.138	98.586–103.408
DiP	5.780	1.207	3.587	3.213	25.917	1.926	0.928	5.012	2.774	29.676
	5.1946.450	1.035–1.409	3.551–3.613	3.052–3.359	25.058–26.716	1.645–2.071	ND-1.164	4.746–5.278	1.827–1.929	28.652–30.866

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Table 3. (*Continued*)

Sample ID	Elemental concentration (µg/ml) in different types of milk samples									
	Cr	**Mn**	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Hg**	**Pb**	**Fe**
PP	ND	2.215	2.823	3.277	25.473	1.763	0.326	1.015	2.147	32.022
		2.086–2.370	2.777–2.872	3.111–3.447	25.304–25.686	1.697–1.822	ND-0.564	1.001–1.030	2.092–2.196	30.822–34.281
MP	ND	0.598	2.926	3.222	22.770	1.828	0.421	3.117	2.768	26.371
		0.215–1.172	2.792–3.001	3.203–3.260	22.204–23.104	1.753–1.895	ND-0.884	2.555–3.679	1.866–2.188	25.815–27.033

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market milk samples ($F_{cal.}$=17.706 < F_{crit} = 3.554; α = 0.05; $p < 0.0001$) as the $F_{cal.}$ = 17.706 value was much higher than F_{crit} = 3.554 and probability was very low ($p < 0.0001$). It might be happened due to the reasons that many places of Bangladesh are affected by arsenic. However, if cows are brought from those As affected areas then milk will be contaminated and if cows are brought from the arsenic affected areas then As concentration in milk samples should be lower. Therefore, a significant difference for As concentration in different milk samples was observed. Nevertheless, the average As concentration (0.034 μg/ml) for this study was found to be consistent with the reported result (0.029 μg/ml) in the literature (Dobrzañski et al., 2005). This study also revealed that Cd concentrations in milk samples were varied from 0.007 - 0.016 μg/ml with an average value of 0.009 μg/ml. Our findings are much lower than the reported values found in commercial packed milk samples in Palestine (Abdulkhaliq et al., 2012). The *F*-test shows that *F* ratio (3.14) for Cd concentration is close to F_{crit} = 3.554 at a level of α = 0.05, which indicting that Cd concentration in different milk samples are close to each other. This study also revealed that the concentration of Hg in MPL was below the detection limit (0.001 μg/ml), and PPL contained the higher Hg concentration (0.018–0.107 μg/ml) compared to other milk samples. Zodape et al. (2012) found Hg concentration in packed milk was 0.023 μg/ml, which were collected from Mumbai City, India. The sequence of Pb concentration in different brands of milk samples was found to be AUL > PPL = PUL > APL > FUL = FPL > MPL. However, *F* test revealed that Pb concentration in different brands of liquid packed milk were very much similar ($F_{cal.}$ = 3.257 < F_{crit} = 3.554; α = 0.05), and the average Pb concentration was found to be 0.045 μg/ml, that was also similar with the finding in literature for milk samples of Romania (Semaghiul et al., 2008). The concentration of Mn in different brand samples varies from 0.032–0.075 μg/ml, but statistically no significantly difference was observed ($F_{cal.}$ = 0.961 < F_{crit} = 3.6554; α = 0.05; $p = 0.959$). However, the ultra-high temperature treated milk samples contain more Mn than pasteurized milk samples. Mn concentration of liquid packed milk sample was 0.07 μg/ml in Romania (Semaghiul et al., 2008), which was similar with our findings. The concentration of Cu in different market liquid milk brand samples ranged from 0.060 to 0.065 μg/ml with an average value of 0.056 μg/ml, which was much lower with the reported results (0.46 μg/ml) of Peshawar, Pakistan (Ghosia et al., 2014), but very similar with the reported results (0.060–0.064 μg/ml) in literature by Tripathi et al. (1999). However, statistical analysis revealed that the concentration of Cu in different brand of liquid packed milk samples was significantly different ($F_{cal.}$ = 10.529 > F_{crit} = 3.655; $p < 0.0001$) at 99% confidence level as the F_{cal} value is much higher that F_{crit} value. Subsequently, Zn concentration in different market liquid milk brand samples were found to be 0.504 to 0.754 μg/ml, which was consistent with the finding (0.685 μg/ml) of Zodape et al. (2012) for the milk samples in India. The

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concentration of Fe in different brands of packaged liquid market milk samples were found be 0.602–1.492 μg/ml with an average value of 0.958 μg/ml, which was consistent with the reported results by Qin et al. (2009) for the Fe concentration (1.51 μg/ml) in commercial liquid milk samples in Japan.

3.2. Level of heavy metals in liquid farmhouse raw milk sample

Raw cow's milk samples contain low amount of heavy metals. These metals can be contaminated through food staffs and during maintenance of milk in farmhouse. However, mean, maximum, and minimum concentration of different heavy metals in farmhouse raw liquid milk samples is presented in Table 3. This study revealed that the average Cr concentration in milk samples was found to be 0.178 μg/ml ranging from 0.105 to 0.212 μg/ml. In this study, concentration of Cr was found to be lower in the farm belongs to Barishal District and the highest concentration was found in the farmhouse of Mirpur area of Dhaka District. Concentration of Cr in raw cow's milk of China was found to be 0.17 μg/ml (Qin et al., 2009), which was in line with our findings. However, statistically no significant difference ($F_{cal.}$ = 0.667 < F_{crit} = 3.403; α = 0.05; p = 0.522) was found in different brand of liquid farmhouse raw milk samples. Statistically no significant difference ($F_{cal.}$ = 2.792 < F_{crit} = 3.403; α = 0.05; p = 0.081) was found for Ni concentration in the milk samples collected from the different farmhouses. Another study (Semaghiul et al., 2008) showed that raw cow's milk in Romania contains 0.04 μg/ml of Ni, that was lower than the average value (0.078 μg/ml) found in this study. The concentration of As (0.033–0.038 μg/ml)was almost similar in most of the liquid farmhouse raw milk samples. This study also revealed that the average As concentration (0.035 μg/ml) in the present study was lower than the reported As concentration (0.08 μg/ml) in cow's milk of Poland (Krystyna et al., 2011). The concentration of Cd indicated noteworthy differences between different liquid farmhouse raw milk samples (0.001–0.019 μg/ml). Concentration of Cd in raw cow's milk was 0.086 μg/ml in Egypt (Enb et al., 2009), which was higher than the present study. Pilarczyk et al. (2013) reported to find 0.004 μg/ml of Cd in milk sample collected from organic farm. This study revealed that the concentration of Hg in F_2L, F_4L and F_9L samples were below than the detection limit. On the other hand, the samples of F_6L and F_7L contain significantly higher concentration. Sample F_6L and F_7L, both were collected form Mirpur, Dhaka City. Hg concentration of raw cow's milk was too low in China and Japan (Qin et al., 2009), which was consistent with our findings (0.002 - 0.030 μg/ml). On the other hand, the concentration of Pb was almost similar (0.037–0.048 μg/ml) in most of the liquid farmhouse raw milk samples as well as there was no significant difference was found ($F_{cal.}$ = 1.788 < F_{crit} = 3.403; α = 0.05; p = 0.188). Our results were very close to the reported results for Pb concentration (0.05 μg/ml) in raw cow's milk samples in Poland (Krystyna et al., 2011). The concentration of Mn in different

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milk samples were found to be varied from 0.025–0.090 μg/ml with an average value of 0.047 μg/ml, that was consistent with the reported results (0.056 μg/ml) in the literature for Egyptian raw cow's milk (Enb et al., 2009). The concentration of Cu in different liquid farmhouse raw milk samples were found to be 0.051–0.063 μg/ml with an average value of 0.056 μg/ml, which was lower than the reported value for Cu (0.15 μg/ml) in raw cow's milk of Pakistan (Ghosia et al., 2014). However, this study revealed that Cu concentrations in different milk samples were statistically same ($F_{cal.} = 0.256 < F_{crit} = 3.403$; $p = 0.776$). The concentration of Zn ranges from 0.596–1.059 μg/ml, whereas raw cow's milk in Pakistan contained 1.93 ppm of Zn (Ghosia et al., 2014) that was higher than that of our findings. The concentration of Fe in powder milk samples were ranged from 0.761–1.237 μg/ml, which was in line with the finding (0.72 μg/ml) in powder milk samples in Romania (Semaghiul et al., 2008).

3.3. Level of heavy metals in powder milk sample

Powder milk was found to contain highest metal concentrations compared to liquid milk samples. Again metals like Zn, Cu, Mn and Fe are fortified in powder milk labeled in milk powder packet. As maximum powder milk available in local market was imported from different countries and the environment of those countries can be a reason of contamination. However, mean, maximum, minimum concentration of different heavy metals in powder milk samples are given at Table 3.The concentration of Cr in powder milk samples were too low to detect by the systems with an exception of Dip (5.780 μg/gm).Ni concentrations were almost similar in most of the powder milk samples (2.421–2.926 μg/gm)with an exception of Dip (3.587 μg/gm).Subsequently, statistically no significant difference was observed ($F_{cal.} = 0.159 < F_{crit} = 3.554$; $\alpha = 0.05$; $p = 0.853$). However one study conducted in powder milk of Romania showed 0.18 μg/gm of Ni concentration, which was lower than the value found in this study. The concentration of As indicated that there was a significant differences ($F_{cal.} = 7.486 > F_{crit} = 3.554$; $\alpha = 0.05$; $p = 0.004$) between powder milk brand samples ranging from 1.701–1.947 μg/gm respectively with an average value of 1.834 μg/gm, that is much higher than the reported value (0.009 μg/gm) in Ghana (Doreen, 2014). Reversely, no significant differences ($F_{cal.} = 0.862 < F_{crit} = 3.68$; $\alpha = 0.05$; $p = 0.442$) for Cd concentration in different brands of powder milk samples was observed. The average Cd concentration in this study was found to be 0.519 μg/gm, which was two times higher than that of Cd concentration (0.211 μg/gm) in powder milk of Pakistan (Ghosia et al., 2014). Again Hg concentration also indicated that there was no significant differences ($F_{cal.} = 0.956 < F_{crit} = 3.554$; $\alpha = 0.05$; $p = 0.403$) between powder milk brand samples. Study (Doreen, 2014) showed that the concentration of Hg in Ghana was 0.024 μg/gm, which was much lower than the present study. The concentration of Pb indicated significant differences

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($F_{cal.} = 5.44 > F_{crit} = 3.554$; $\alpha = 0.05$; $p = 0.0.014$) between powder milk brand samples. However, the reported Pb concentrations in powder milk samples in Palestine was were 0.93 μg/gm (Abdulkhaliq et al., 2012), which was higher than the present study. This study also revealed that Mn concentration was almost similar ($F_{cal.} = 2.87 > F_{crit} = 3.554$; $\alpha = 0.05$; $p = 0.0.082$) in different brands of milk powder. The average Mnconc was found to be 1.206 μg/gm, which was much higher than that of Mn conc. (0.29 μg/gm) in powder milk sample of Romania (Semaghiul et al., 2008). On the other hand, the average Cu concentration in different brands of the powder milk samples was varied from 2.511–2.215 μg/gm with an average value of 2.903 μg/gm, 5 times higher than that of the reported result (Abdulkhaliq et al., 2012). Bosnak and Pruszkowski (2006) reported to contain 45.7 μg/gm of Zn in USA powder milk, which agrees with the present study. The concentration of Fe ranges from 16.996–111.010 μg/gm, indicated no significant differences ($F_{cal.} = 0.0087 > F_{crit} = 3.554$; $\alpha = 0.05$; $p = 0.991$) between different powder milk brand samples. Abdulkhaliq et al. (2012) found that in Palestine powder milk contained 12.91 μg/gm of Fe concentration.

3.4. Correlation matrix analysis

The Pearson's correlation matrices among the studied metals (Cr, Mn, Ni. Cu, Zn, As, Cd, Hg, Pb and Fe) in the different types of milk (packaged liquid, farm liquid and powder milk) samples were performed using SPSS 22 (IBM Corp., USA) software. This study revealed that Fe had significantly ($\alpha = 0.05$) positive correlation with most of the elements, i.e., Mn ($r = 0.574$, $p < 0.01$), Ni ($r = 0.723$, $p < 0.01$), Cu ($r = 0.733$, $p < 0.01$), Zn ($r = 0.869$, $p < 0.01$), As ($r = 0.768$, $p < 0.01$), Cd ($r = 0.575$, $p < 0.01$), Hg ($r = 0.709$, $p < 0.01$) and Pb ($r = 0.707$, $p < 0.01$) but no significant correlation with Cr was observed at a 95% confidence level (Table 4). Subsequently this study revealed that there was no significant correlation between Cr and Mn, between Cr and Cu, between Cr and Zn, between Cr and As, between Cr and Hg, between Cr and Pb (Table 4). Therefore, it has been suggested that Cr was not correlated with other metals. However, rest of the metals has significantly positive correlation between each other in different type of milk samples analyzed in this study (Table 4), suggesting a common source of these metals.

3.5. Metal pollution index

Metal Pollution Index (MPI) has been calculated using Eq. (1) for the toxic element Cr, Ni, As, Cd, Hg, Pb in different milk samples, which is shown in Fig. 2. Among different milk samples Metal Pollution Index (MPI) as shown in Fig. 2 followed a decreasing sequence of Dip (2.698) > NP(1.739) > DsP(1.706) > DnP (1.700) > MP(1.562) > PP(1.334) > FP(0.99) > MPL(0.095) > F_3L(0.071) > F_2L

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Table 4. The Person's correlation matrices among among the studied metals (Cr, Mn, Ni. Cu, Zn, As, Cd, Hg, Pb and Fe) in the different types of milk (packaged liquid, farm liquid and powder milk) samples.

	Cr	Mn	Ni	Cu	Zn	As	Cd	Hg	Pb	Fe
Cr	1									
Mn	.241	1								
Ni	.413*	.860**	1							
Cu	.324	.876**	.994**	1						
Zn	.194	.803**	.892**	.906**	1					
As	.299	.860**	.988**	.992**	.926**	1				
Cd	.498*	.510*	.795**	.773**	.674**	.790**	1			
Hg	.408	.708**	.903**	.886**	.902**	.923**	.807**	1		
Pb	.372	.845**	.992**	.987**	.872**	.982**	.744**	.891**	1	
Fe	.053	.574**	.723**	.733**	.869**	.768**	.575**	.709**	.707**	1

* Correlation is significant at the 0.05 level (2-tailed).
** Correlation is significant at the 0.01 level (2-tailed).

(0.068) > PUL(0.056) > F_4L(0.055) > F_6L(0.049) > F_7L(0.047) = FUL(0.047) > APL(0.043) = PPL(0.043) > AUL(0.038) > FPL(0.037) > F_1L(0.035) = F_3L (0.035) = F_9L(0.035) > F_8L(0.020).

3.6. Health risk assessment

The health risk assessment associated with heavy metal (Cr, Ni,As, Cd, Hg, Pb, Mn, Cu, Zn, Fe) in different milk samples were calculated (Table 5). The result showed that the health risk index (HRI) for Cr, Ni, Mn, Cu, Zn and Fe in all types of powder milk samples for children are lower than 1 and the health risk index for

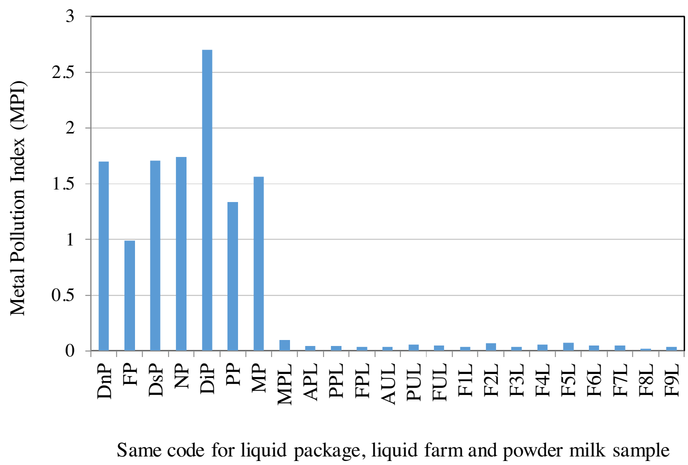

Fig. 2. Metal Pollution Index (MPL) in different types of milk samples.

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Table 5. Health Risk Assessment in milk sample.

Sample ID	Elements										
		Cr	Ni	As	Cd	Hg	Pb	Mn	Cu	Zn	Fe
Powder milk samples											
DnP	Adult	0.000	0.180	0.810	0.000	1.785	0.806	0.040	0.090	0.141	0.003
	Child	0.000	0.420	1.890	0.000	4.164	1.881	0.093	0.211	0.329	0.006
FP	Adult	0.000	0.173	0.841	0.031	3.148	0.809	0.045	0.090	0.133	0.000
	Child	0.000	0.404	1.962	0.073	7.345	1.888	0.105	0.209	0.311	0.001
DsP	Adult	0.000	0.179	0.907	0.901	3.082	0.679	0.028	0.095	0.107	0.001
	Child	0.000	0.416	2.117	2.103	7.190	1.584	0.066	0.221	0.250	0.003
NP	Adult	0.000	0.186	0.928	0.789	3.715	0.679	0.048	0.104	0.261	0.002
	Child	0.000	0.433	2.164	1.840	8.669	1.585	0.112	0.244	0.609	0.006
Dip	Adult	0.006	0.256	0.917	1.326	3.587	0.741	0.052	0.115	0.123	0.001
	Child	0.013	0.598	2.140	3.093	8.370	1.728	0.122	0.268	0.287	0.002
PP	Adult	0.000	0.202	0.840	0.466	0.725	0.767	0.096	0.117	0.121	0.001
	Child	0.000	0.471	1.959	1.087	1.692	1.789	0.224	0.273	0.283	0.002
MP	Adult	0.000	0.209	0.871	0.601	2.227	0.739	0.026	0.115	0.108	0.001
	Child	0.000	0.488	2.031	1.403	5.195	1.723	0.060	0.269	0.253	0.002
Liquid market milk samples											
MPL	Adult	0.000	0.035	0.130	0.114	0.000	0.263	0.011	0.019	0.029	0.000
	Child	0.000	0.092	0.340	0.300	0.000	0.263	0.029	0.488	0.754	0.000
APL	Adult	0.001	0.041	0.130	0.183	0.206	0.307	0.015	0.018	0.026	0.000
	Child	0.002	0.095	0.302	0.427	0.480	0.307	0.036	0.427	0.597	0.000
PPL	Adult	0.001	0.060	0.133	0.034	0.612	0.314	0.015	0.017	0.018	0.000
	Child	0.002	0.139	0.311	0.080	1.427	0.313	0.034	0.400	0.418	0.000
FPL	Adult	0.001	0.041	0.130	0.080	0.194	0.293	0.015	0.018	0.026	0.000
	Child	0.002	0.095	0.302	0.187	0.454	0.293	0.036	0.420	0.596	0.000
AUL	Adult	0.001	0.054	0.133	0.069	0.103	0.333	0.026	0.018	0.019	0.000
	Child	0.003	0.127	0.311	0.160	0.240	0.333	0.061	0.413	0.448	0.001
PUL	Adult	0.001	0.046	0.130	0.069	0.160	0.313	0.024	0.018	0.019	0.000
	Child	0.003	0.107	0.302	0.160	0.374	0.313	0.057	0.427	0.448	0.001
FUL	Adult	0.002	0.046	0.130	0.149	0.200	0.293	0.015	0.018	0.021	0.000
	Child	0.004	0.108	0.302	0.347	0.467	0.293	0.036	0.420	0.477	0.001
Liquid farm milk samples											
F1L	Adult	0.001	0.044	0.133	0.171	0.034	0.123	0.013	0.016	0.024	0.000
	Child	0.003	0.103	0.311	0.400	0.080	0.287	0.029	0.037	0.055	0.001
F2L	Adult	0.002	0.045	0.145	0.046	0.000	0.114	0.015	0.018	0.027	0.000
	Child	0.004	0.105	0.338	0.107	0.000	0.267	0.035	0.042	0.063	0.001

(*Continued*)

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Table 5. (*Continued*)

Sample ID	Elements										
		Cr	Ni	As	Cd	Hg	Pb	Mn	Cu	Zn	Fe
F3L	Adult	0.001	0.043	0.130	0.149	0.034	0.137	0.030	0.015	0.038	0.000
	Child	0.003	0.100	0.302	0.347	0.080	0.320	0.070	0.034	0.088	0.000
F4L	Adult	0.002	0.048	0.126	0.011	0.000	0.137	0.009	0.016	0.038	0.000
	Child	0.004	0.112	0.293	0.027	0.000	0.320	0.020	0.038	0.088	0.000
F5L	Adult	0.002	0.046	0.130	0.149	0.046	0.131	0.031	0.016	0.031	0.000
	Child	0.004	0.108	0.302	0.347	0.107	0.307	0.073	0.037	0.071	0.000
F6L	Adult	0.002	0.046	0.126	0.217	0.172	0.123	0.009	0.015	0.023	0.000
	Child	0.004	0.107	0.293	0.507	0.400	0.287	0.022	0.034	0.053	0.001
F7L	Adult	0.002	0.045	0.130	0.183	0.149	0.126	0.014	0.016	0.024	0.000
	Child	0.004	0.105	0.302	0.427	0.347	0.293	0.033	0.037	0.057	0.000
F8L	Adult	0.001	0.041	0.126	0.034	0.011	0.131	0.012	0.016	0.033	0.000
	Child	0.002	0.096	0.293	0.080	0.027	0.307	0.028	0.037	0.077	0.000
F9L	Adult	0.001	0.045	0.145	0.057	0.000	0.106	0.018	0.017	0.040	0.000
	Child	0.003	0.104	0.338	0.133	0.000	0.247	0.041	0.039	0.094	0.001

Cr, Ni, As, Pb, Mn, Cu, Zn and Fe for adult are lower than 1 indicating safe for the consumer. However As, Hg, Pb found to show HRI value greater than 1 for children for all types of samples. Again for Cd, all samples showed HRI value greater than 1except DnP and FP. In this regards, a suggestion can be given to intake 45 g of powder milk per day by the children and thus the HRI value will be reduced and will remain within acceptable range. In case of adult, HRI value for Hg was found greater than 1 for all samples and for Cd it is lower than 1 for all samples except DiP.

4. Conclusion

Important information of toxic elements (Cr, Ni, As, Cd, Hg, Pb, Mn, Cu, Zn, and Fe) in different types of cow's milk available in Bangladesh is computed in the present study. The elemental analysis showed a significant variation depending on the source and type of the milk samples. ANOVA (two ways) test revealed that the different heavy metal concentration in different types of milk samples were significantly different. This study also revealed that powder milk samples were found to contain a higher metal concentration than the commercially available liquid milk samples. More over the commercially available milk sample showed lower metal concentration than the liquid milk collected from different farm house, which might be attributed by the fact that in most cases there is a trend to mix water with the milk sample that may cause metal contamination. Among the farm

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milk samples, F_8L and F_9L collected from Barishal District showed lower value of toxic elements than the milk collected from the larger city Dhaka District, which may be due to the location of farm house from where the milk samples were collected. From nutritional point, the liquid milk collected from Barishal contains Mn, Cu, Zn and Fe in an amount sufficient to fulfill the requirement of human. Calculated metal pollution index (MPI) indicated that powder milk contained more concentration of the mentioned elements than the liquid milk collected from retail market and farm house. On the other hand, 'Diploma' contained the highest value among all the powder milk samples, which was really alarming. The result of calculated HRI for powder milk reveled that Cr, Ni, Mn, Cu, Zn and Fe have a value lower than 1 (both children and adult) but HRI value for the toxic element As, Cd, Pb and Hg are more than 1 for the case of children, and hence there is a possibility of health hazard. Therefore it may be suggested to consume less amount of powder milk. The HI (hazard index) value of the studied elements showed a higher value for children than the adult and hence the powder milk showed the highest value.

The overall study concluded that liquid milk is more suitable than commercially available powder milk and suggested to reduce its consumption especially for children. Moreover special attention should be given to heavy metals contamination as once they present in concentration greater than the acceptable daily intake, it may be difficult to reduce them to an acceptable level.

Declarations

Author contribution statement

Yeasmin Jolly: Conceived and designed the experiments, Wrote the paper.

Iftekhar Ahmad: Conceived and designed the experiments.

Shahriar Iqbal, M.S. Rahman, J. Kabir, S. Akter: Performed the experiments.

Competing interest statement

The authors declare no conflict of interest.

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