Food Research 6 (1) : 233 - 237 (February 2022)
Journal homepage: https://www.myfoodresearch.com

FOOD RESEARCH

FULL PAPER

Determination of heavy metals in raw and pasteurized liquid milk of Bangladesh to assess the potential health risks

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Article history:
Received: 20 March 2021
Received in revised form: 24 April 2021
Accepted: 28 June 2021
Available Online: 13 February 2022

Keywords:
Heavy metals,
Milk,
Bangladesh,
Health risk,
Carcinogenic and non-carcinogenic risk

DOI:
https://doi.org/10.26656/fr.2017.6(1).191

Abstract

Milk is a balanced diet and the consumption of milk is increasing day by day in Bangladesh. For public health concerns, it becomes essential to detect the amount of heavy metals in milk and their effects on human health. For this purpose, the potential health risks of adults and children due to consumption of Cow raw milk and liquid Pasteurized milk were accessed. In this research, the concentrations of eight heavy metals namely Iron (Fe), Copper (Cu), Manganese (Mn), Zinc (Zn), Lead (Pb), Cadmium (Cd), Chromium (Cr) and Arsenic (As) in 64 samples of raw and liquid Pasteurized cow's milk were accessed. These samples are collected from 64 different administrative areas of Bangladesh. From this study, it was found that Cu showed the highest concentration and daily intake rate; while Pb showed the lowest concentration and daily intake among these metals. Except for Cr, the value of estimated daily intake (EDI) of the heavy metals was found to be lower than the permissible value. From the results of EDI, it can be concluded that the minor potential health risks may occur due to the consumption of raw and liquid pasteurized milk samples. From Target Hazard Quotient (THQ) values, it can be said that there is no chance of non-carcinogenic health risks. From the Target Carcinogenic Risk (TCR) values of As and Pb, it could be suggested that child milk consumers are sensitive to be exposed to carcinogenic health risk.

1. Introduction

Milk is a vital component of human nutrition and its consumption has risen in recent years (Khan *et al.,* 2014). The vital nutrients of milk including vitamins and minerals, proteins, lactose and essential fatty acids make it considered a complete food (Licata *et al.,* 2004). Though milk and milk products contain heavy metals in very small amounts, they become toxic during manufacturing and packaging due to alteration by different factors (Khan *et al.,* 2014). There are potential bio-accumulative risk factors in milk including aluminium, lead, cadmium, copper, chromium, zinc, and arsenic (Li *et al.,* 2005). Therefore, the risk to human health posed by exposure to food-based heavy metals varies from other types of pollutants in terms of long-term exposure, the inadequacy of decomposition, non-degradability and high accumulation levels along the food chain (Maas *et al.,* 2011). Raw milk contains 38 micro and trace elements which have been reported from different regions around the world (Nwankwoala *et al.,* 2002; Dobrzański *et al.,* 2005). Most of these elements have an essential role in human health. They act as enzymatic co-factors that can play vital roles in the human body's different physiological functions, and the lack of these elements in the whole physiological system can lead to problems (Schuhmacher *et al.,* 1991). Also, the deficiencies of these elements contribute substantially to the global burden of disease. When their amounts are present at higher levels, they may expose a detrimental effect on human health (Meshref *et al.,* 2014). On the other hand, toxic elements also expose negative effects at low concentrations while they are ingested over a long time (Celik and Oehlenschlager, 2007; Uluozlu *et al.,* 2009).

In our current study, Iron (Fe), Copper (Cu), Manganese (Mn), Zinc (Zn), Cadmium (Cd), Lead (Pb), Chromium (Cr) and Arsenic (As) are the selected heavy metals in milk samples. As babies and children are the main consumers of milk and milk products, the main concern of heavy metals is lead and cadmium because

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eISSN: 2550-2166
Published by Rynnye Lyan Resources

lead and cadmium are considered as potential carcinogens (Zhuang *et al.,* 2009).

In addition, due to the toxic level of these metals, babies and children are particularly at high risk for neurotoxic and developmental disorders. Arsenic exposure can also cause various acute problems in human health. Heavy metal emission is increasing day by day for industrialization, urbanization and agricultural mechanization, which has a negative effect on the livestock system and the quality of milk. Therefore, our study aimed to estimate the amount of heavy metals in raw and liquid Pasteurized milk samples and to evaluate their carcinogenic and non-carcinogenic health effects due to consumption of this milk. This analysis was performed using various standard methods and the findings were compared to find out their relationship.

2. Materials and methods

2.1 Sample collection and preparation

In this study, a total of 64 samples of cow's raw milk and 64 samples of liquid Pasteurized milk were collected from 64 administrative areas of Bangladesh. The 64 raw milk and liquid Pasteurized milk samples were collected from the local markets of 64 administrative areas in Bangladesh. By using the standard method, sterile glass bottles were used to collect samples. Samples were collected in sterile glass bottles, and instantly the glass bottles were kept in an icebox to maintain the temperature below 4°C. The samples were carried to the laboratory as soon as possible and stored at 4°C until analysis. The analysis process was started within 24 hours. The samples were maintained in glass bottles to avoid contact with any metal sources. In this research, the amount of selected heavy metals such as Copper (Cu), Cadmium (Cd), Iron (Fe), Manganese (Mn), Zinc (Zn), Chromium (Cr), Lead (Pb) and Arsenic (As) were estimated in milk samples. The research work was conducted at the Milk, Dairy and Fermented Food Products Research Section of the Institute of Food Science and Technology of Bangladesh Council of Scientific and Industrial Research, Bangladesh. This study was conducted from December 2018 to October 2019.

2.2 Sample digestion

The elements were extracted from samples by using a microwave digestion system (Berghof, Germany) with 5 mL of HNO3 acid (65% purity Merck, Germany) and 2 mL H_2O_2 (30% purity, Merck, Germany) as digestion reagents.

2.3 Analysis of sample

Atomic Absorption Spectrophotometer (iCE -3000 series, Thermo-Scientific, USA) was used for the analysis of heavy metal. The standard solution of analytical grade was purchased from Sigma-Aldrich, USA. The spectral lines were 228.67, 357.65, 324.57, 279.43, 217.35, 213.9, 248.30 and 193.7 nm for Cd, Cr, Cu, Mn, Pb, Fe, Zn and As respectively. Four standard solutions of each metal at 0.01, 0.1, 1.0, 5.0 parts per million (ppm) were prepared. The minimum detection limit for Cr, Zn was 0.005 mg/kg, Mn and Pb was 0.002 mg/kg and for Cd, Fe and Cu was 0.001 mg/kg. Certified reference materials (CRMs) of this method were purchased from Sigma Aldrich, USA and they are used to check the quality of the samples.

2.4 Health risks assessment for Bangladeshi peoples

2.4.1 Estimated daily intake (EDI)

The equation was used to calculate the EDI:

$$EDI = \frac{DFc \times Mc}{BW} \quad (1)$$

Here, DFc = Daily Food Consumption Rate ($gday^{-1}$); For Bangladeshi people, Adults = 27.31 $gday^{-1}$ and children = 25.53 $gday^{-1}$ (BBS, 2011); MC means the metal concentration in milk samples and BW represents the bodyweight of the population; 60kg for adult and 27 kg for children (Shaheen *et al.,* 2016).

2.4.2 Target hazard quotient (THQ)

The target hazard quotient (THQ) is calculated to estimate the potential risk of these heavy metals (USEPA, 2000). The equation is:

$$THQ = \frac{EDI}{RfD} \times 10^{-3} \quad (2)$$

Here, EDI represents the estimated daily intake of metals (mg/day), RfDs indicates reference dose; the reference doses for Fe, Cu, Mn, Zn, Pb, Cd, Cr and As are 0.7, 0.04, 0.05, 0.3, 0.004,0.001, 1.5 and 0.0003 mg/kg BW^{-1} day^{-1} 9 (USEPA , 2010). If the value of THQs is equal to or higher than 1, indicates the potential health risk for this heavy metal (Islam *et al.,* 2011). When consumers are exposed to two or more pollutants, they may suffer from adverse or interactive effects (Hallenbeck, 1993). The total THQ (TTHQ) of these heavy metals is the mathematical sum of the THQ values of each individual metal. TTHQ values can be estimated by the following equation:

$$TTHQ = THQ_{\text{toxic element 1}} + THQ_{\text{toxic element 2}} + \ldots + THQ_{\text{toxic element n}} \quad (3)$$

2.4.3 Target carcinogenic risk (TCR)

When the carcinogens are to be exposed over a lifetime, the probability of cancer of an individual can be developed (USEPA, 1989). The acceptable risk level

eISSN: 2550-2166

ranges from 10^{-4} to 10^{-6} for carcinogens. The equation is:

$$TCR = \frac{EFr \times ED \times EDI \times CSF^{\circ}}{BW \times AT} \times 10^{-3} \quad (4)$$

Where, EFr means the frequency of exposure (365 days/year), ED express duration of exposure (for adults 70 years and for children 14 years) which is the average human lifetime. BW is the average body weight (where adults 60 kg; children 27 kg), AT is the average time for carcinogens, for adults = (365 days/year × 70 years) and for children= (365 days/year × 14 years) and CSF° means the oral carcinogenic slope factor (USEPA, 2010) which is 1.5 mg/kg/day and 0.0085 mg/kg/day for Arsenic and lead respectively. TCR for As and Pb was calculated because they are carcinogenic.

3. Results and discussion

In this analysis, the concentrations of Copper (Cu), Iron (Fe), Manganese (Mn), Zinc (Zn), Lead (Pb), Cadmium (Cd), Chromium (Cr) and Arsenic (As) were calculated in 64 samples of cow's raw (non-brand) milk and 64 samples of cow's liquid brand milk specifically for adults and children. The concentrations of metals are presented in Tables 1 and 2 respectively. The mean value of micro-minerals and toxic metals had shown in the descending order of Cu>Fe>Zn>Mn and Cr>As>Cd>Pb respectively for both raw milk and liquid Pasteurized milk samples. Among raw milk (non-brand) and liquid Pasteurized milk samples, Cu had shown the highest concentration, including 0.742±0.377 for raw milk samples; whereas 0.807±0.522 for liquid Pasteurized milk samples. On the other hand, Pb had shown the lowest concentration like 0.013±0.004 for both raw milk and liquid Pasteurized milk samples. The daily intake of selected eight heavy metals from milk consumption was calculated for the purposes of health risk assessment. The EDIs for adults and children through the consumption of raw and liquid Pasteurized milk is shown in Tables 3 and 4. EDI was calculated for two population groups, specifically adults and children through two types of milk consumption, namely raw milk (non-Pasteurized) and liquid Pasteurized milk. The daily intake of Cu was shown to have the highest value (mg/day) including 0.3377 mg/day for adults and 0.7016 mg/day for children, through raw milk consumption. Through the consumption of Pasteurized milk, the daily intake of Cu was also shown to have the highest value (mg/day) like 0.3673 mg/day for adults and 0.7630 mg/day for children. On the other hand, the daily intake of Pb was shown to have the lowest value (mg/day) like 0.0059 mg/day for adults and 0.0123 mg/day for children for both raw and pasteurized milk samples. The permissible value of these selected heavy metals is presented by Malhat *et al.* (2012). From the findings, it can be observed that only the daily intake of Cr of both adults and children exceeds their permissible limit from the consumption of both raw milk and liquid Pasteurized milk. On the other hand, the value of the daily intake of remaining heavy metals is below their permissible limit. On the basis of these results, it can be concluded that Cr is the component that makes a minor contribution to the potential health risk through the intake of raw and liquid brand milk samples. The value of the daily intake of these heavy metals through the consumption of raw and Pasteurized milk samples may be stated below that the value found in the article of (Muhib *et al.* 2016).

3.2 Non-carcinogenic health risk

The target hazard quotient (THQ) was estimated to know the non-carcinogenic health risks. In this study, the estimated THQs of children and adults from raw milk consumption are presented in Table 5 and the estimated THQs of children and adults from liquid Pasteurized milk consumption are presented in Table 6. From the results, the estimated THQ value of all the metals is below the threshold value of 1through the consumption of both raw and liquid Pasteurized milk for both adults and children. It can be said that there are no possible non-carcinogenic health risks associated with these metals. The findings are agreed with Islam *et al.* (2015) and Muhib *et al.* (2016). TTHQ was accessed in order to know the total potential risk of non-carcinogenic effects caused by more than one factor. In this study, the TTHQs value for adults and children from raw milk consumption are 0.03431 and 0.22157 respectively; also for adults and children from brand milk consumption are 0.08992 and

Table 1. The concentration of heavy metals in raw milk (64 samples) results in ppm (mg/kg)

	Fe	Cu	Mn	Zn	Pb	Cd	Cr	As
Max.	0.785	2.268	0.298	0.204	0.02	0.045	0.942	0.091
Min.	0.346	0.128	0.033	0.114	0.005	0.021	0.116	0.005
Mean	0.520	0.742	0.112	0.151	0.013	0.032	0.548	0.053
SD	0.115	0.377	0.064	0.021	0.004	0.005	0.229	0.029

Table 2. The concentration of heavy metals in pasteurized milk (64 samples) results in ppm (mg/kg)

	Fe	Cu	Mn	Zn	Pb	Cd	Cr	As
Max.	0.711	2.781	0.235	0.201	0.02	0.041	0.834	0.089
Min.	0.315	0.132	0.031	0.105	0.007	0.018	0.108	0.005
Mean	0.512	0.807	0.102	0.148	0.013	0.027	0.457	0.043
SD	0.109	0.522	0.050	0.024	0.004	0.006	0.225	0.026

eISSN: 2550-2166

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Table 3. Estimated daily intake of heavy metals (mg/day) from raw milk (non-pasteurized) consumption by the population of Bangladesh

Individuals	Fe	Cu	Mn	Zn	Pb	Cd	Cr	As
Adult	0.2367	0.3377	0.0509	0.0687	0.0059	0.0146	0.2494	0.0241
Child	0.4917	0.7016	0.1059	0.1428	0.0123	0.0303	0.5182	0.0501

Table 4. Estimated daily intake of heavy metals (mg/day) from pasteurized milk consumption by the population of Bangladesh

Individuals	Fe	Cu	Mn	Zn	Pb	Cd	Cr	As
Adult	0.2330	0.3673	0.0464	0.0674	0.0059	0.0123	0.2080	0.0196
Child	0.4841	0.7630	0.0965	0.1399	0.0123	0.0256	0.4321	0.0407

Table 5. Target hazard quotient (THQ) value of raw milk samples for non-carcinogenic health risks

Individuals	Fe	Cu	Mn	Zn	Pb	Cd	Cr	As
Adult	0.00034	0.00844	0.00102	0.00023	0.00148	0.0146	0.00017	0.0080
Child	0.00070	0.01754	0.00212	0.00048	0.00308	0.0303	0.00035	0.1670

Table 6. Target hazard quotient (THQ) value of Pasteurized milk samples for non-carcinogenic health risks

Individuals	Fe	Cu	Mn	Zn	Pb	Cd	Cr	As
Adult	0.00033	0.00918	0.00093	0.00023	0.00148	0.0123	0.00014	0.06533
Child	0.00069	0.00918	0.00193	0.00047	0.00308	0.0256	0.00029	0.13567

0.17691 respectively. In fact, a TTHQ < 1 does not indicate acute health effects of consumers (Islam, Ahmed and Al-Mamun, 2015). The TTHQ values of both raw and pasteurized milk samples were enough less than 1. Therefore, there is no risk of adverse effects.

3.3 Carcinogenic health risk

The TCR values for As exposure was 6.025E−07 and 0.7833E−06 for adults and children respectively, from raw milk consumption. And the values of As for liquid brand milk were 4.9E−07 and 0.2611E−06 for adults and children, respectively; while for both raw and liquid brand milk samples, the TCR values of Pb was 8.3583E−10 for adults and 0.8722E−09 for children. The risk of cancer above 10^{-4} is considered undesirable, the risk of cancer below 10^{-6} is generally considered minimal and the risk of cancer between 10^{-4} to 10^{-6} is generally considered an acceptable range (USEPA, 2000, 2010). It was found that the TCR value of Pb was within the acceptable range to the negligible range. While the TCR value of As was within the acceptable range to the unacceptable range. This study indicates that children are more vulnerable than adults in Bangladesh to exposure to harmful or non-essential elements by daily food intake.

4. Conclusion

Milk is an important food for people of all ages because of its high nutritional value. The emission of heavy metals is increasing day by day because of continuous industrialization. As the consumption of milk is increasing day by day, it is necessary to detect the amount of heavy metals in milk and their effects on human health. This study will help the consumers to assess their potential health risks through the consumption of milk.

Conflict of interest

The authors declare no conflict of interest.

Acknowledgement

The study was part of the R&D work of "Milk, Dairy and Fermented Product Research Section" of the Institute of Food Science and Technology (IFST), BCSIR. The instrument used in this study was supported by the ADP project allocated by the Ministry of Science of Technology, Bangladesh.

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