# Contamination of raw fresh milk, market pasteurized milk and powdered milk by toxic heavy metals in Bangladesh

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***Abstract*- Milk is called the balanced nutrient food in the human diet. Micronutrients and macronutrients were found abundantly in milk, but contamination of heavy metals can occur in milk and milk products. The present investigation was carried out to determine concentrations of lead, chromium and nickel in raw fresh milk, market pasteurized milk and powdered milk of different brands. A total of 80 raw milk samples of cows from the household lactating cows in Chittagong, Narayanganj and Dhaka, 36 powdered milk samples and 36 market milk samples from Chittagong were taken for analysis. Lead (Pb), Chromium (Cr) and Nickel (Ni) concentration in raw fresh milk was found 0.17 mg/L, 0.04 mg/L and 0.15mg/L respectively. Where in brand market pasteurized milk the concentrations of Lead (Pb), Chromium (Cr) and Nickel (Ni) were 0.02 mg/L, 0.11 mg/L and 0.17 mg/L respectively. In powder milk of different brand samples the concentrations of Lead (Pb), Chromium (Cr) and Nickel (Ni) were 0.18 mg/L, 0.11 mg/L and 0.17 mg/L respectively. A comparison of level of those three heavy metals was done with different locations of Bangladesh and among different brands. The levels of those three heavy metals within most of the samples were found within permitted range of different approved authorities. But in some samples those limit exceeded the limits which can cause health risk to humans. The way of their entry to the human food chain is through the contaminated milk and powder milk due to the presence of industries and highway roads near the dairy farms, animal feeds contaminated with heavy metals and processing steps.**

***Index Terms*— Heavy Metals, Milk, Contamination, AAS.**

## I. INTRODUCTION

### **1.1** General

Milk and milk products are major constituents of the daily diet in Bangladesh as well as whole world. Those are used as food for supplement, food for treatment for vulnerable groups such as infants, school age children and old age people through nutritional program. Varieties of milk products are being prepared from milk in dairy enterprises and households everyday throughout the world including Bangladesh. Milk is the abundant sources of calcium, vitamin D, riboflavin, and phosphorus. Protein, potassium, vitamin A, vitamin B-12 and niacin are also found in good amounts. Heavy metal contamination in milk and other edible tissues is a matter of great concern for food safety and human health. Milk and milk products contaminated with heavy metals such as zinc, lead, cadmium, selenium, sulphur, iodine and possibly even more dangerous arsenic and cyanide (Belete, 2014). These metals are toxic in nature and even at relatively low concentrations can cause adverse effects for human health (Mahaffey, 1977). Heavy metal contamination in milk has been reported from different region in various studies (Gregorio and Siracusano, 1976). However, results vary according to the geographic distribution and difference in sampling methods and analytical techniques. Several international studies have also reported high levels of cadmium lead and zinc residues in milk (Alais, 2000). Many researchers have reported the instances of contamination of heavy metals in milk and milk products during processing. While the feeding of cattle on the contaminated feed and rearing of livestock in proximity to polluted surroundings were found to be responsible for heavy metal pollution in milk and milk products. The preceding lines reflect the sensitivity of threat being posed to general public health. This problem needs an immediate attention of health regulatory authorities and the researchers as well. There is an urgent need of local database or risk assessment studies in local food animals to assess the potential risk to humans from heavy metal residues because the Bangladesh have varying topographical and environmental conditions under which a considerable number of livestock are growing. Information is very limited on the concentration of Lead (Pb), Chromium (Cr) and Nickel (Ni) in farm produces non brand milk as well as different brand milk and powder milk of Bangladesh. However, very limited numbers of research works have been carried out in Bangladesh regarding heavy metals contamination in milk and powder milk. In this context present study was conducted to determine the status of heavy metals (Pb, Ni and Cr) in raw milk, market milk and powdered milk and to compare the concentrations of heavy metals (Pb, Ni and Cr) in raw milk, market milk and powdered milk. This study will help to determine the potential risks from the toxic effects of heavy metals and to make recommendations for future implementations by the local health regulatory authorities.

### **1.2** Heavy Metals

Heavy Metals are those elements which have density more than 5g/cm$^3$, atomic weight 63.546 to 200.590 (Kennish, 1992) and a specific gravity greater than 4.0 (Connell and Miller, 1984). Although heavy metals remain in ground water and soil they tend to accumulate and are very toxic at certain levels. Living organisms normally require some of these heavy metals up to certain limits and in case excess accumulation occurs it will lead to severe detrimental effects (Kennish, 1992) in animal and human including mental confusion, memory loss, fatigue, immune dysfunction etc. Historically the accumulation of heavy metals is not a new experience. Heavy metals enter the milieu through air emanation from coal burning plants, smelters and other industrial amenities (Fernandes et al., 2000; Beavington et al., 2004; Khillare et al., 2004; Al-Masri et al., 2006). Other than civil, natural processes also play an important role in decaying heavy metals in the ground water e.g. naturally occurring geological deposits of arsenic in ground water (Sanyal and Nasar, 2002; Ghosh et al., 2004). Once heavy metals are on the rampage to the environment, they remain for years to increase the chances of revelation to humans and livestock. Recent studies have shown that the modern products like cosmetics (Aslam et al., 1979; Hardy et al., 1998), mercury amalgam dental filling (Ellender et al., 1978; Chin et al., 2008), paints (Bastarche, 2003) and ground water residues (Sanyal and Nasar, 2002; Ghosh et al., 2004) of certain chemicals lead to chronic exposure to these heavy metals. Foodstuffs grown on contaminated soil or irrigated with impure water accumulate metal contents and are a big source of heavy metals exposure to the animals and humans (Ward and Savage, 1994).

### **1.3 Raw Fresh Milk**

Raw fresh milk is unpasteurized milk collected within 0-2 hours after milking that means no heat treatment applied to the milk.

### **1.4 Market Pasteurized Milk**

Pasteurized Milk is the milk that goes under pasteurization. Pasteurization is a process that kills harmful bacteria by heating milk to a specific temperature for a set period of time. In Bangladesh, HTST pasteurization is used for milk to inhibit microorganism, inactivate self-decomposition and increase self-life of milk.

### 1.5 Milk Powder

Powdered milk or dried milk is a manufactured dairy product made by evaporating milk to dryness. Different mechanical dryer are used for this purposes that preserve milk for longer.

## II. MATERIALS AND METHODS

### 2.1 General

The research work was carried out to determine contamination of raw fresh milk, market pasteurized milk and powdered milk by toxic heavy metals in Bangladesh. A cross-sectional study was conducted to determine the prevalence of heavy metals in raw milk from Dhaka, Narayanganj and Chittagong, powdered milk and market milk from Chittagong, Bangladesh. Dhaka, Narayanganj and Chittagong district were purposively selected for collecting milk samples. The raw milk samples were collected from Dhaka, Narayanganj and Chittagong district. The market milk and powdered milk samples were collected from available grocery shops and sweetmeat shops (brands showroom) of Bangladesh. The study was conducted during the period of July 2015 to December 2015.

### 2.2 Sample selection

A total of 80 raw milk samples of cows from the household lactating cows in Chittagong, Narayanganj and Dhaka, 36 powdered milk samples and 36 market milk samples from Chittagong were the target population. Raw milk, Market milk (A, B, C, D, E, and F) and Brand powder milk samples (X, Y and Z) were collected from different market areas of Bangladesh. Raw milk samples were collected and stored in sterile vials (10 ml). Samples were immediately transferred to laboratory, Chittagong Veterinary and Animal Sciences University through ice box. Samples were stored temporarily in refrigerator before laboratory evaluation. However, samples without transport media were stored at 0-4$^0$C for later laboratory evaluation.

### 2.3 The Method of Processing of milk samples

For the digestion of samples, approximately 10 mL of raw milk or 10gm of powder milk were digested with10ml of $HNO_3$ (65%) and 2ml of $H_2O_2$ (30%) in acid pre-washed Teflon vessels. After standing overnight, samples were digested using Microwave Lab Stations with 250W, 1min; 0W, 1min; 250W, 6min; 400W, 5min; 600W, 5min. After sufficient cooling, samples were moved to Teflon vessels and diluted to 35ml with deionized water. Milk powder GBW 10017 was used as reference material Analytical blanks were prepared with each batch of digestion set. All samples were prepared in triplicate run. In brief, 10ml of 6.0 mole/L HCl was added to the digested samples. They were slowly heated until color fading. Finally, all samples were diluted to 35ml with deionized water (Li-Qiang et al. 2009).

### 2.4 Detection and Estimation

The amount of (Pb, Ni and Cr) heavy metal was measured by Atomic Absorption Spectrophotometer (AAS) Model: ZEE nit 700P, Germany (Shahriar et al., 2014). All laboratory works were performed at Chittagong Veterinary and Animal Sciences University, Bangladesh. The obtained data were stored in Microsoft Excel 2007 and then exported into STATATM 11.0 (Stata Corporation, College Station, TX, USA) for statistical analysis. Descriptive analysis was performed by using percentages, mean and standard deviation for different variables. Finally one -way ANOVA was used to compare the level of heavy metal residues in milk of different regions of Bangladesh. The level of significance was set $\leq 0.05$.

## III. RESULTS AND DISCUSSION

### 3.1. Concentration of Pb, Ni and Cr in raw milk

Among the three heavy metals Lead (Pb) was at higher concentration (0.17 mg/L) whereas Chromium (Cr)was at lower concentration (0.04 mg/L) and Nickel (Ni) was also determined as (0.15mg/L) in the raw milk (Table 3.1).

**Table 3.1: Concentration of Pb, Ni and Cr in raw milk**

| Heavy metals | N | Concentration(mg/L) Mean ± SD | Min – Max (mg/L) |
|---|---|---|---|
| Pb | 80 | 0.17±0.10 | 0 – 0.39 |
| Ni | 80 | 0.15±0.13 | 0 – 0.71 |
| Cr | 80 | 0.16±0.06 | 0 – 0.11 |

### 3.2. Concentration of Pb, Ni, and Cr in raw milk of different areas

The concentration of Pb in Dhaka district was at higher concentration (0.26 mg/L) in raw milk when it was lower concentration in raw milk of Chittagong (0.11 mg/L). There was significant (P<0.01) difference among Pb concentration in raw milk of three districts. (Table 3.2). In raw milk Ni concentration in Narayanganj was at higher (0.19 mg/L) level. However it was lower concentration in raw milk of Dhaka (0.12 mg/L). Difference among Ni concentration in raw milk of three districts was insignificant (P>0.05). (Table 3.2). Cr in Narayanganj was at higher concentration (0.05 mg/L) in raw milk and it was lower concentration in raw milk of Chittagong (0.02 mg/L). Chromium concentration in raw milk of three districts was significant (P<0.05). (Table 3.2)

**Table 3.2: Concentration of Pb, Ni and Cr in raw milk of different areas**

| Heavy metals | Areas | N | Concentration(mg/L) Mean ± SD | p-value |
|---|---|---|---|---|
| Pb | Dhaka | 30 | 0.26 ± 0.08 | 0.000 |
| | Chittagong | 24 | 0.11 ± 0.03 | |

| | | | | |
|---|---|---|---|---|
| | Narayanganj | 26 | 0.13 ± 0.09 | |
| | Dhaka | 30 | 0.12 ± 0.06 | |
| Ni | Chittagong | 24 | 0.13 ± 0.10 | 0.348 |
| | Narayanganj | 26 | 0.19 ± 0.02 | |
| | Dhaka | 30 | 0.14 ± 0.01 | |
| Cr | Chittagong | 24 | 0.12 ± 0.02 | 0.048 |
| | Narayanganj | 26 | 0.15 ± 0.04 | |

**3.3. Concentration of Pb, Ni and Cr in market milk**

Among the three heavy metals Ni was found at higher concentration (0.17 mg/L) as Pb was at lower concentration (0.02 mg/L) and Cr was also determined as (0.11 mg/L) in brand market milk. (Table 3.3)

**Table 3.3: Concentration of Pb, Ni and Cr in market milk**

| Heavy metals | N | Concentration(mg/L) Mean ± SD | Min – Max (mg/L) |
|---|---|---|---|
| Pb | 36 | 0.02 ± 0.01 | 0 – 0.10 |
| Ni | 36 | 0.17 ± 0.10 | 0 – 0.24 |
| Cr | 36 | 0.11 ± 0.03 | 0 – 0.44 |

**3.4. Concentration of Pb, Ni and Cr in different market milk**

The concentration of brand X market milk sample was (0.15 mg/L). Difference among Pb concentration in market milk sample was insignificant (P>0.05) ( Table4.4) .Ni in brand Z market milk sample was at higher concentration (0.83 mg/L) among market milk samples when it was lower concentration in brand X market milk sample (0.43 mg/L).The concentration of brand Y market milk sample was (0.65 mg/L).There was insignificant (P>0.05) difference among the Ni concentration in brand market milk samples. (Table 3.4)

**Table 3.4: Concentration of Pb, Ni and Cr in different market milk**

| Heavy metals | Brands | N | Concentration(mg/L) Mean ± SD | p-value |
|---|---|---|---|---|
| | X | 12 | 0.15 ± 0.11 | |
| Pb | Y | 12 | 0.38 ± 0.55 | 0.60 |
| | Z | 12 | 0.13 ± 0.04 | |
| | X | 12 | 0.43 ± 0.39 | |
| Ni | Y | 12 | 0.65 ± 0.52 | 0.86 |
| | Z | 12 | 0.83 ± 0.14 | |
| | X | 12 | 0.16 ± 0.24 | |
| Cr | Y | 12 | 0.12 ± 0.04 | 0.64 |
| | Z | 12 | 0.24 ±0.15 | |

In case of Cr, it was at higher concentration in brand X market milk sample (0.16 mg/L) but it was at lower concentration in brand Y market milk sample (0.12 mg/L). In brand Z market milk sample the concentration was (0.15 mg/L). Cr concentration in brand market milk samples was insignificant (P>0.05). (Table 3.4)

**3.5. Concentration of Pb, Ni and Cr in powder milk**

Among the three heavy metals Pb was found at higher concentration (0.18 mg/L) and Cr was at lower concentration (0.11 mg/L) and Ni was also determined (0.17 mg/L) in brand powder milk samples. (Table 3.5)

**Table 3.5: Concentration of Pb, Ni and Cr in powder milk**

| TABLE I. Heavy metals | TABLE II. N | TABLE III. Concentration(mg/L) TABLE IV. Mean ± SD | TABLE V. Min – Max (mg/L) |
|---|---|---|---|
| TABLE VI. Pb | TABLE VII. 36 | TABLE VIII. 0.20 ± 0.18 | TABLE IX. 0 – 0.86 |
| TABLE X. Ni | TABLE XI. 36 | **TABLE XII.** 0.19± 0.17 | TABLE XIII. 0 – 0.62 |
| TABLE XIV. Cr | TABLE XV. 36 | TABLE XVI. 0.11 ± 0.10 | TABLE XVII. 0 – 0.34 |

**3.6. Concentration of Pb, Ni and Cr in different powder milk**

The concentration of brand powdered milk samples A, B, C and E was 0.17, 0.19, 0.22, 0.10 (mg/L) respectively. Pb concentration in brand powder milk sample was insignificant

(P>0.05) (Table 3.6). Ni in brand E powdered milk sample was higher concentration (0.78 mg/L) in powder milk samples since it was lower concentration in brand C powder milk samples (0.07 mg/L). The concentration of brand powdered milk samples A, B, D, and F was 0.23, 0.19, 0.30, and 0.14 (mg/L) respectively. There was insignificant (P>0.05) difference among Ni concentration in brand powder milk samples. (Table 3.6)

**Table 3.6: Concentration of Pb, Ni and Cr in different powder milk**

| Heavy metals | Brands | N | Concentration(mg/L) Mean ± SD | p-value |
|---|---|---|---|---|
| Pb | A | 6 | 0.17 ± 0.07 | 0.72 |
| | B | 6 | 0.19 ± 0.15 | |
| | C | 6 | 0.22 ± 0.14 | |
| | D | 6 | 0.32 ± 0.46 | |
| | E | 6 | 0.10 ± 0.15 | |
| | F | 6 | 0.06 ± 0.45 | |
| Ni | A | 6 | 0.23 ± 0.29 | 0.70 |
| | B | 6 | 0.19 ± 0.02 | |
| | C | 6 | 0.17 ± 0.07 | |
| | D | 6 | 0.43 ± 0.30 | |
| | E | 6 | 0.78 ± 0.13 | |
| | F | 6 | 0.14 ± 0.23 | |
| Cr | A | 6 | 0.17 ± 0.86 | 0.25 |
| | B | 6 | 0.13 ± 0.33 | |
| | C | 6 | 0.18 ± 0.99 | |
| | D | 6 | 0.02 ± 0.24 | |
| | E | 6 | 0.02 ± 0.30 | |
| | F | 6 | 0.12 ± 0.19 | |

The concentration of Cr in brand C powdered milk sample was at higher concentration (0.18 mg/L) among brand powder milk samples as it was lower concentration in brand D and E samples (0.02 mg/L). The concentration of Cr in brand powder milk samples A, B and F were 0.17, 0.13 and 0.12 (mg/L) respectively. There was insignificant (P>0.05) difference among Cr concentration in brand powder milk samples.

## IV. CONCLUSION

The present study includes the investigation of the levels of heavy metals in milk and powder milk from Bangladesh. The overall prevalence of heavy metals was 96% in milk samples. There was significant (P<0.01) difference among Pb and Cr concentration in raw milk samples. Heavy metals such as Cr, Pb, and Ni are a threat to public health. In the present study the concentration of lead(Pb) (0.17mg/L) in raw milk samples and nickel(Ni) in brand E powder milk (0.78 mg/L) was higher in Dhaka. The results also suggest that brand Y market milk (0.38mg/L) had higher concentration of lead among the brand market milk samples. The way of their entry to the human food chain is through the contaminated milk and powder milk. Further studies are necessary to determine the standard permissible limits for heavy metals in all types of consumable milk and dairy products. This necessitates that all the effort including awareness creation, effective surveillance, monitoring heavy metals in animal derived products is employed. In addition, the dairy scientists, veterinarians, farmers, might be beneficial from the present findings. Therefore, it can be concluded that the present results will contribute in awareness building regarding public health to a great extent. Finally, the present study will contribute in understanding the level of heavy metals in milk and powder milk.

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