

# Assessment of dairy products consumed on the Arakmarket as determined by heavy metal residues

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## ABSTRACT

Dairy products are nutritious drink and can play a significant part in a healthy diet. The safety of dairy products decreases with increasing concentration of toxic compounds and environmental pollutants (especially heavy metals). In the present study, the contamination of Al, Sn, As, Cd, Hg and Pb in dairy products (pasteurized milk, yogurt, yogurt drink, cheese) consumed in Arak, Iran, 2013 was evaluated using inductively coupled plasma mass spectrometry (ICP-SFMS) method. To meet the aim, 60 samples of 5 different brands, consisting of pasteurized milk, yogurt, yogurt drink, cheese (n = 15) were selected and analyzed for heavy metals content. 100% of the samples were positive regarding the total average and range concentration of Al, Sn, As, Cd, Hg and Pb in dairy products was  $168.25 \pm 92.2$  (30.6 - 356.5),  $5.9 \pm 4$  (1.1 - 16),  $3.2 \pm 1.95$  (0.4 - 8.1),  $4.55 \pm 2.6$  (0.6 - 10.6),  $23.15 \pm 10.4$  (6.8 - 50.2) and  $15.4 \pm 8.53$  (3.1 - 40.2)  $\mu\text{g}/\text{kg}$ , respectively. 28.3% (17 of 60 samples) of dairy products samples had lead (Pb) greater than EU limit and national Iranian standard (20  $\mu\text{g}/\text{kg}$ ). Statistical analysis indicated except about As in pasteurized milk and cheese there was no significant difference between products in terms of heavy metals content.

## KEYWORDS

Heavy Metals; Milk; Cheese; Yogurt; Food

## Safety; ICP

## 1. INTRODUCTION

Milk and milk products are one of the main sources of minerals [1] and rich in calcium, phosphorus, vitamins and proteins [2], also are basic foods in the human diet. As an excretion of the mammary gland, milk can carry numerous xenobiotic substances (pesticides, antibiotics, drugs, heavy metals and various environmental contaminants), which is a risky factor for the health and safety of the consumer and new chemical medicines and antibiotics have negative side effects [3]. Heavy metals are essential for correct body metabolism however the ranges between their effective and toxic levels are small. Heavy metal contamination raises environmental concerns, such as entrance on the food chain, which can be potentially harmful to human's health. Trace elements are shown to have a multitude of toxic effects such as acute syndrome and neurotoxic effects [4,5]. Detection, determination and comparing of heavy metals in dairy products with standard to determine the safety of them have been a subject of numerous studies throughout last decades. Malhat *et al.* (2012) reported mean levels of 4.404 and 0.288  $\mu\text{g}/\text{g}$  for lead and cadmium, respectively, in cow milk collected from different sites in El-Qaliubiya governorate, Egypt [6]. Fatima *et al.* (2005) reported that the Pb was determined in two UHT samples with concentration well below the PTW limit defined by WHO [7]. Studies performed in the various foods in Arak city, showed that this area requires more attention to the foodstuff and regularly monitor pollutants in food [2,8,9]. The aim of

this study was to determine the contents of Al, Sn, As, Cd, Hg and Pb in pasteurized milk, yoghurt, yoghurt drink and cheese of five brands available in Iran in the region of Arak 2013.

## 2. MATERIALS & METHODS

### 2.1. Chemicals

All the reagents and chemicals Merck (Darmstadt, Germany) were used of analytical grade. De-ionized water was used throughout the work. Concentrated nitric acid (65%), were spectroscopic grades Merck (Darmstadt, Germany).

### 2.2. Apparatus

ICP-OES measurements were performed using an ICP spectrometer working in a simultaneous mode.

### 2.3. Sampling and Analysis

A total of 60 dairy product samples of 5 different brands included of pasteurized milk, yoghurt, yoghurt drink and cheese (n = 15) were randomly selected from local supermarket in Arak city, 2013. Samples were conditioned in sterile plastic container and kept at 4°C until analyses that were carried out in same day. Statistical analysis with SAS software (9.1) was carried out In order to determine significance difference between product's heavy metal content.

### 2.4. Sample Preparation

Six elements (Al, Sn, As, Cd, Hg and Pb) were determined in each dairy product samples. Prior to analysis samples must be decomposed with the appropriate methods procedure with digestcontaining reduced amounts of carbon residues. In order to minimize the effects of the organic matrix, closed vessel acid decomposition in microwave oven system was used. Moreover, it may provide faster, more efficient process and reduce or eliminate the risk of sample contamination and losses of analytes. A microwave assisted-acid digestion procedure was carried out, in order to achieve a shorter digestion time and using minimum amount of acid. Duplicate of 2.0 mL of each dairy product samples were taken into

microwave vessels but well shaken, opened and sampled by pouring directly from the original container into the microwavevessel. Added the samples to each vessels and 10 mL of a concentrated HNO<sub>3</sub>-H<sub>2</sub>O<sub>2</sub> (2:1, v/v) and kept samples for 10 min at room temperature till the samples were homogenized, and then placed the vessels in covered PTFE container. This was then heated following a one-stage digestion programmed at 80% of total power (900 W), for 3 - 5 min. After cooling, the resulting solutions were evaporated to semidried mass to remove an excess acid, and then diluted up to 50.0 mL in volumetric flasks and kept as a stock sample solution [10,11].

## 3. RESULTS AND DISCUSSION

Levels of heavy metal in dairy products are summarized in **Table 1**. The results from samples showed that the pasteurized milk contained (mean ± S.D) of Al, Sn, As, Cd, Hg and Pb, 135 ± 85.7, 4.8 ± 3.6, 2.3 ± 1.6, 3.95 ± 2.3, 21.17 ± 9.6 and 12.5 ± 7.6 µg/l, respectively. Yoghurt drink contained of 157.9 ± 89, 5 ± 3.2, 2.9 ± 1.7, 4 ± 2.6, 20.3 ± 8.8 and 14.3 ± 7.6 µg/l, respectively, yoghurt contained of 186 ± 79.4, 6 ± 4, 3.3 ± 2, 4.8 ± 2.6, 23.8 ± 10.9 and 16.6 ± 8.9 µg/l, respectively and cheese contained of 194.2 ± 109.2, 7.5 ± 4.7, 4.3 ± 2.1, 5.4 ± 2.7, 27.2 ± 11.6 and 18.2 ± 9.6 µg/kg (**Table 1**). Accordingly, the highest and least amount of Pb samples was found in cheese, yoghurt, yoghurt drink and pasteurized milk samples, respectively. Mean and range concentration of Pb in samples was 18.2 ± 9.6 (3.8 - 40.2), 16.6 ± 8.9 (3.6 - 35), 14.3 ± 7.6 (3.1 - 29) and 12.5 ± 7.6 (3.2 - 27.5) µg/l or kg, respectively (**Table 1**). 28.3% (17 of 60 samples) of dairy product samples had lead (Pb) greater than EU limit and national Iranian standard (20 µg/kg). The amounts of lead in the samples, pasteurized milk are the best and cheeses have the highest levels of contamination. In the each product manufacturing process, biochemical content and their concentration can influence the metal concentrations. It is well known lead has a high affinity to caseins. In this study cheeses has the highest content of lead that this can be due to the high concentration of casein in it [12]. Except for asin pasteurized milk and cheese there is no significant difference (p < 0.05) there was no significant difference between products in terms of heavy metals content (**Tables 1** and **2**). The results

**Table 1.** The Comparisons between means of heavy metal content between various dairy products (µg/l).

	As	sn	Al	pb	Hg	Cd
Pasteurized Milk	2.28 <sup>a</sup>	4.76 <sup>a</sup>	134.9 <sup>a</sup>	12.5 <sup>a</sup>	21.16 <sup>a</sup>	3.94 <sup>a</sup>
Cheese	4.28 <sup>b</sup>	7.5 <sup>a</sup>	194.18 <sup>a</sup>	18.21 <sup>a</sup>	27.27 <sup>a</sup>	5.4 <sup>a</sup>
Yogurt	3.34 <sup>ab</sup>	6.07 <sup>a</sup>	186.02 <sup>a</sup>	16.56 <sup>a</sup>	23.81 <sup>a</sup>	4.79 <sup>a</sup>
Yogurt drink	2.95 <sup>ab</sup>	5.09 <sup>a</sup>	157.88 <sup>a</sup>	14.34 <sup>a</sup>	20.35 <sup>a</sup>	4.06 <sup>a</sup>
SD	0.48	1.01	23.61	2.18	2.65	0.66

**Table 2.** Characteristic of the statistical analysis method for dairy product samples (µg/l).

Sample	M <sup>1</sup>	Y <sup>2</sup>	C <sup>3</sup>	Yd <sup>4</sup>	M	Y	C	Yd	M	Y	C	Yd	M	Y	C	Yd	M	Y	C	Yd				
Mean	3.9	4.8	5.4	4.1	21.2	23.8	27.3	20.4	12.5	16.6	18.2	14.3	134.9	194.2	186.0	157.9	4.8	6.1	7.5	5.1	2.3	3.3	4.3	3.0
S.E	0.6	0.7	0.7	0.7	2.5	2.8	3.0	2.3	2.0	2.3	2.5	2.0	22.1	28.2	20.5	23.0	0.9	1.1	1.2	0.8	0.4	0.5	0.5	0.4
S.D	2.3	2.6	2.8	2.6	9.6	10.9	11.6	8.8	7.6	8.9	9.6	7.6	85.7	109.2	79.4	88.9	3.6	4.1	4.7	3.2	1.6	2.0	2.1	1.7
SV	5.3	6.9	7.6	6.7	92.0	119.1	134.4	77.3	58.2	79.0	91.7	57.6	7340.4	11918.5	6305.7	7905.3	12.7	16.6	22.0	10.6	2.5	3.9	4.5	2.9
Range	7.2	8.5	9.5	8.3	32.5	37.0	38.7	32.7	24.3	31.4	36.4	25.9	281.4	315.9	246.5	279.1	9.9	13.0	12.9	9.9	5.7	6.7	6.9	6.4
Min	0.8	0.7	1.1	0.6	8.5	9.0	11.5	6.8	3.2	3.6	3.8	3.1	30.6	40.6	93.5	32.5	1.1	2.0	3.1	1.6	0.4	0.9	1.2	0.7
Max	8.0	9.2	10.6	8.9	41.0	46.0	50.2	39.5	27.5	35.0	40.2	29.0	312.0	356.5	340.0	311.6	11.0	15.0	16.0	11.5	6.1	7.6	8.1	7.1
CI (95%)	1.3	1.5	1.5	1.4	5.3	6.0	6.4	4.9	4.2	4.9	5.3	4.2	47.4	60.5	44.0	49.2	2.0	2.3	2.6	1.8	0.9	1.1	1.2	0.9

<sup>1</sup>Pasteurized Milk; <sup>2</sup>Yogurt; <sup>3</sup>Cheese; <sup>4</sup>Yogurt drink.

were nearly agreement with [13], that reported Pb and Al contamination in milk samples 41.8 - 58.7 and 1460 - 1985  $\mu\text{g}\cdot\text{L}^{-1}$ . [14] also pointed out rate of contamination in raw milk samples of 15 different area in Iran, about 90% of the samples were less than the newly established Codex standard (20 ng/ml). Iwegbue et al. (2008) reported that all analyzed pasteurized bovine milk samples were above the one permitted by the current Brazilian legislation. In similar following studies in Romania in different country was reported average concentration of Pb varied from 2.73  $\mu\text{g}/\text{kg}$  in pasteurized milk to 4.05  $\mu\text{g}/\text{kg}$  in hard cheese; that Pb were found at concentration levels less than WHO tolerated values (25  $\mu\text{g}/\text{kg}/\text{week}$ ) [15]. In dairy products from sheep milk collected in Two Regions of Southern Italy, chromium was the highest levels in milk and lead was the highest in fresh, mature cheese and in ricotta. The compounds detected at the lowest concentrations were cadmium in milk, fresh and mature cheese (0.05  $\mu\text{g}/\text{g}$ , 0.05  $\mu\text{g}/\text{g}$  and 0.06  $\mu\text{g}/\text{g}$ ) and chromium in ricotta (0.03  $\mu\text{g}/\text{g}$ ) [12]. in milk of cattle and goats for Pakistan, concentration of lead and cadmium ranged from  $42.687 \pm 0.051$  and  $0.084 \pm 0.003$  mg/L, respectively and showed that the residual levels of Cd and Pb have been found higher in goat milk [16]. in Egypt, was reported that lead and cadmium contents of all the cow milk samples were 4.404 and 0.288  $\mu\text{g}/\text{g}$ , respectively [6]. In Pakistan, milk heavy metals (Cadmium, Chromium, Lead and Nickel) concentrations were found above those normally associated with suitability for human consumption [17]. Tona et al. (2013) in Nigeria, showed that all the milk and milk products samples analyzed contained residues of Pb and Cd heavy metals. heavy metal contamination in raw milk samples exceeded EU limit with percentage of 100% for lead in Lubuskie Province, Poland [18]. Also, mean levels for cadmium, lead, and mercury in the breast milk of Saudi women were 1.732  $\mu\text{g}/\text{L}$ , 31.671  $\mu\text{g}/\text{L}$ , and 3.1  $\mu\text{g}/\text{L}$ , respectively [19]. Licata et al. (2004) Reported that the concentrations of Pb in all milk samples in Calabria, Italy were lower than EU limit (0.10 to 9.92  $\text{Ag}/\text{kg}$ ). There are some difference between our study and others. As a reason Several factors such as exhaustion gases, industry wastes and waste waters polluted to the plants affected on heavy metal level of plant and other edible parts of feed animals and then, heavy metal is secreted to milk and subsequently entered individuals through consumption of milk and other dairy products. Also there is no information about feedstuff given to cows in sampling regions, but depending on the Planting area for agricultural products (i.e. air pollution, soil, water, etc.), dairy Production Equipment, Storage and transport containers, are important supplies of heavy metal contamination. It seems that the use of stainless steel containers, appropriate packaging and agricultural products grown in soils

with low heavy metal pollution could play major role in reduction of heavy metal content in dairy product.

#### 4. CONCLUSION

As regards the fact that the amount of heavy metals in most of dairy products examined was above EU limit, it is highly recommended that controlling measures should be taken promptly to reduce contamination such as implementing a food control systems (i.e. GAP and HACCP), educating dairy farmers and manufacturers. Additionally, it is suggested that subsequent studies should be conducted on heavy metals contamination in different stages of milk preparing and which factors could be involved in heavy metals contamination. The presence of heavy metals emphasizes the need for regular monitoring and a more stringent food safety management system (FSMS) in order to control the heavy metals at the lowest possible levels.

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