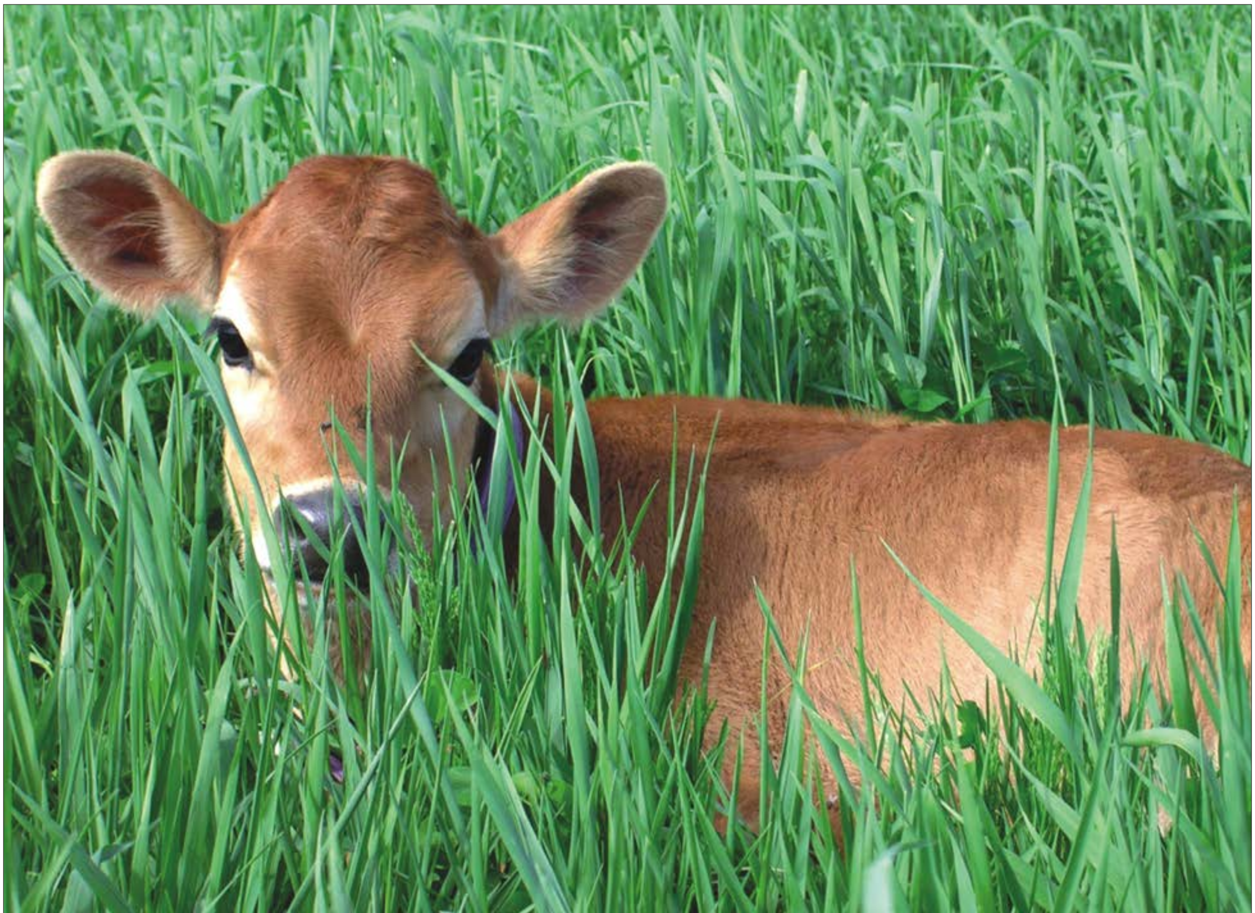

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Aluminium Content in Milk and Milk Products and its Leachability from Dairy Utensils

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ABSTRACT

Aluminium is non-essential element for humans and is considered to be a toxic metal. The present investigation was carried out to determine aluminium concentrations in milk and milk products, to estimate the intake of aluminium via consumption of milk and dairy products and to investigate the leachability of aluminium from utensils into milk products during processing and storage. A total of 85 milk and milk products samples were collected from farms, individual farmers and dairy shops in Beni-Suef governorate, Egypt. Mean aluminium concentrations in farm milk, market milk, kareish cheese, yoghurt and rice pudding were 19.93, 107.32, 52.36, 4.19 and 80.97 ppm, respectively. Aluminium intake through milk and milk products consumption was calculated to be 246.72 mg week^{-1} which corresponds to 205.5% of the PTWI. Processing and storage of milk in aluminium containers also raise aluminium content significantly. The results indicated the advantage of using stainless steel over aluminium utensils for processing and storage of milk products, especially those acidic in nature.

Key words: Milk, aluminium, leaching, daily intake

INTRODUCTION

Aluminium is the third most abundant element in the environment, comprising 8.13% of the earth's crust. It is present naturally in soil, minerals and rocks and even in water and food. It does not appear to serve any essential biological function in the human body (Soni *et al*., 2001). The health safety of aluminium intake has long been a subject of controversial debate. For many years, it was not considered harmful to human health because of its relatively low bioavailability and its excretion in the urine is very efficient (Stahl *et al*., 2011). Recently, it has been associated with anaemia, osteomalacia and a neurological syndrome, commonly known as dialysis encephalopathy particularly in patients with chronic renal failure. In addition, there is evidence that aluminium is a cofactor in the development of serious brain disorders like Alzheimer's disease (Karbouj *et al*., 2009).

Humans are frequently exposed to aluminium, primarily from foods, water, airborne dust and pharmaceuticals (Semwal *et al*., 2006). Data from several countries summarized that adults are exposed to between 2 and 160 mg aluminium per day from various sources (Soni *et al*., 2001). Food is unquestionably the main source of aluminium intake. Aluminium enters the food chain through a number of natural and anthropogenic sources. The main sources of aluminium in the diet are cereal, desserts, beverages and milk products (Arruda *et al*., 1994).

Milk and milk products provide good quality nutrients necessary for a strong body and mind. However, the presence of toxic elements such as aluminium in milk and milk products may create health problems, especially for infants, children and old people who consume large quantity of those products (Gonzalez-Weller *et al*., 2010). Aluminium enters the milk and milk products from a variety of sources. Milk gets contaminated before milking, from the feed and fodder fed to the dairy cows. Additionally aluminium can be introduced into the milk and milk products during the production process or by contamination from the metal processing equipment (Soni *et al*., 2001; Deeb and Gomaa, 2011). The use of aluminium utensils for processing and storage of milk may increase substantially the level of this metal in milk and milk products (Semwal *et al*., 2006) and leaching of this metal from utensils is influenced by the quality of the containers, pH level, preparation conditions and the presence of complexing agents (Al Juhaiman, 2010).

The increasing demand of environmental and food safety has stimulated research regarding the risk associated with environmental contamination and consumption of foods contaminated with aluminium. As information on the levels of aluminium in milk and milk products in Egypt and on leaching of aluminium from processing utensils is scanty. Moreover, an additional insight into metal uptake and assessment of human risks associated with the consumption of milk and milk products are still needed. Therefore, this study was conducted to quantify the levels of aluminium in milk and milk products, to estimate the daily intake of this element through consumption of milk and milk products and to investigate the leachability of aluminium from utensils into milk products during processing and storage.

MATERIALS AND METHODS

Collection and preparation of samples: A total of 85 milk and milk products samples (15 farm milk, 15 market milk, 20 kareish cheese, 20 yoghurt and 15 rice pudding) were collected from local farms, individual farmers and dairy shops in Beni-Suef governorate, Egypt. All samples were collected in nitric acid-washed polyethylene containers. The samples were immediately transported to the laboratory in a cooler with ice packs and were stored at -20°C until analysis.

Milk and milk product samples (2 mL or g) were digested with nitric and perchloric acid mixture (HNO_3: $HCLO_3$ = 4:1 v/v) until a transparent solution was obtained (Patra *et al*., 2008). After digestion, samples were filtered and diluted to a suitable concentration. Three blank samples, where biosample was substituted by de-ionized triple distilled water, were run simultaneously with each batch of the digestion. Working standard solutions were made up by dilution of certified standard solutions to the desired concentration. All reagents used were of analytical reagent grade. Ultra high purity water was used for all dilutions. All glass and plastic wares were washed and kept overnight in 10% (v/v) nitric acid solution. Afterwards, it was rinsed thoroughly with ultra-pure water and dried.

Sample analysis: The Al content of the digested samples was determined using flame atomic absorption spectrophotometer (Thermo Solaar M6 A.A. spectrometer, Thermo Electron). The instrumental parameters for Al were: Wave length 309.3 nm, bandpass 0.5 nm, lamp current 100% and nitrous oxide-acetylene flame.

Estimated Daily Intake (EDI): The daily intake of Al for an adult person (60 kg b.wt.) was calculated as follows:

$$\text{EDI} = \frac{C_{\text{metal}} \times W_{\text{food}}}{\text{Body weight (b.wt.)}} (\text{mg/kg b.wt./day})$$

where, C_{metal} (mg kg^{-1}, on fresh weight basis) is the concentration of Al in contaminated foods, W_{food} represents the daily average consumption of food and b.wt. represents the body weight. The average daily consumption per adult person were considered to be 200 mL, 22, 14 and 106 g of milk, kareish cheese, yoghurt and rice pudding, respectively (Saleh *et al*., 1998; FAO., 2009; Al-Ashmawy, 2011).

Experimental study: Milk, yoghurt and kareish cheese were processed individually in aluminium and stainless steel utensils to determine the leaching of aluminium from dairy utensils. Raw milk (5 L) was divided into 5 equal parts. The first part was set as a control (raw milk, analyzed before processing). The second and third parts were individually heated to boiling temperature for 10 min in aluminium and stainless steel utensils while the fourth and fifth parts were separately stored at 4°C for 24 h in aluminium and stainless steel utensils.

Two batches of yoghurt were prepared (one batch in aluminium cookware and the other batch in stainless steel cookware). Raw cow's milk (1.5 L) was heated in a water bath at 90°C for 30 min, cooled to about 42°C and inoculated with 2% of yoghurt starter cultures (Yoflex, Chr. Hansen). The inoculated milk was incubated at 43°C until a firm coagulum was formed (5 h).

In the same manner, two batches of kareish cheese were prepared (one batch in aluminium cookware and the other batch in stainless steel cookware). It was prepared by incubation of raw milk (1.5 L) at 30°C for 2 h, followed by rennet addition and further incubation until coagulation. After that, the curd was salted (5 g/100 mL original milk) while ladling into cheesecloth's for overnight drainage of the whey.

Raw milk, boiled milk, cold stored milk, yoghurt and kareish cheese samples were collected in nitric acid-washed polyethylene containers. The samples were stored at -20°C until analysis. All analyses were performed in duplicate.

Data analysis: Concentrations were expressed as Mean±Standard Error (SE), minimum and maximum values. All calculations were performed with the SPSS pocket program for windows (version 16, 2007). One-way analysis of variance (ANOVA) and Duncan's multiple range tests were used to determine significant differences in the measured attributes at $p<0.05$.

RESULTS AND DISCUSSION

The aluminium contents of milk and milk products are given in Table 1. Significant levels of aluminium were detected in all the samples analyzed. The levels of aluminium in the farm milk samples were between 0.51 and 104.93 ppm, with a mean value of 19.93±7.2 ppm. Two thirds of these samples contained aluminium concentration less than 10 ppm. More than one quarter (26.7%) had aluminium concentrations between 10 and 50 ppm. Only 6.6% had greater than 50 ppm (Fig. 1). The market milk examined here revealed aluminium concentrations between 1.07 and

Table 1: Concentrations of aluminium (ppm) in milk and milk products samples

Products	No. of samples	Min.	Max.	Mean±SE
Farm milk	15	0.51	104.93	19.93±7.2
Market milk	15	1.07	270.63	107.32±26.9
Kareish cheese	20	1.40	251.38	52.36±1.70
Yoghurt	20	2.66	7.42	4.19±0.34
Rice pudding	15	1.70	361.36	80.97±2.60

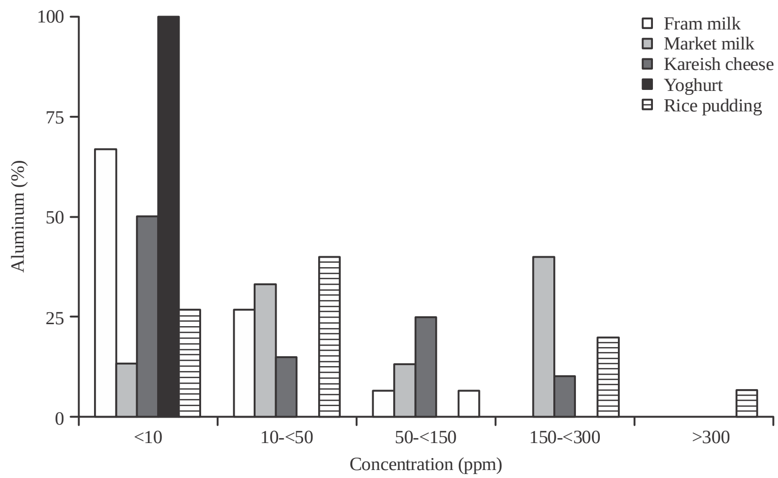

Fig. 1: Frequency distribution of aluminium levels in milk and milk products

270.63 ppm, with a mean value of 107.32±26.9 ppm. One third of these samples contained aluminium concentrations between 10 and <50 ppm. Forty percent had aluminium concentrations between 150 and <300 ppm (Fig. 1).

Many reports dealing with the aluminium contamination level in raw milk have been accumulated. In those studies, aluminium levels were reported as 3.76, 1.18, 7.427, 0.84, 0.37 and 0.501 ppm by Guler (2007), Birghila *et al*. (2008), Ayar *et al*. (2009), El-Mossalami and Noseir (2009), Gonzalez-Weller *et al*. (2010) and Abd-El Aal *et al*. (2012), respectively. Al-Ashmawy (2011) reported aluminium levels of bulk farm milk and market milk at 0.004 and 0.081 ppm, respectively. Aluminium concentrations measured in the market milk were significantly higher ($p \leq 0.05$) than those found in the farm milk (Table 1). A similar observation was reported by Al-Ashmawy (2011). In the present study, the aluminium levels obtained turned out to be higher than the reviewed studies. Large amounts of aluminium contamination can be resulted either from the abundant and traditional use of aluminium containers for production and storage of raw milk or by external environmental contamination during the phases of collection and transport of milk from the farms to the dairy shops.

Regarding kareish cheese, the aluminium contents were in the range of 1.4-251.38 ppm. Mean aluminium level was 52.36±1.7 ppm. Fifty percent of these samples contained aluminium concentration below 10 ppm. One quarter had aluminium concentrations between 50-<150 ppm (Fig. 1). The results obtained in the present study were higher than those obtained by Ayar *et al*. (2009) and Gonzalez-Weller *et al*. (2010) and lower than those reported by Deeb and Gomaa (2011) who determined a mean aluminium concentration of 57.58±3.44 mg kg^{-1} for kareish cheese.

Kareish cheese is one of the most popular types of soft cheese consumed in Egypt. The aluminium contents of kareish cheese were higher than those of the farm milk and lower than those of the market milk, but statistically not significant ($p \leq 0.05$) (Table 1). Aluminium contamination of kareish cheese can be resulted from the milk portion. This may be ascribed to the fact that the aluminium is preferentially bound to caseins and fat and consequently shift mostly to the curd. Also, a certain accumulation during the production process through aluminium utensils, salt and the environment can not, however, be excluded (Coni *et al*., 1996). Additionally, the acidity of cheese can enhance corrosion of aluminium in pots and increases the rate of contamination with aluminium (Elbarbary and Hamouda, 2013).

Table 2: Intake of aluminium via consumption of milk and milk products

Products	Food intake	Average daily Al intake (mg day^{-1})	Average weekly Al intake (mg $week^{-1}$)	PTWI (mg kg^{-1} b.wt.)	Contribution to PTWI (%)
Farm milk	0.2 L	3.99	27.902	0.470	23.25
Market milk	0.2 L	21.46	150.250	2.500	125.21
Kareish cheese	0.022 kg	1.15	8.060	0.130	6.72
Yoghurt	0.014 kg	0.06	0.410	0.007	0.35
Rice pudding	0.106 kg	8.60	60.100	1.001	50.10
Total		35.26	246.720	4.110	205.50

Table 3: Aluminium levels (ppm) in raw milk and milk products after processing in aluminium and stainless steel utensils

		Boiling			Kareish cheese			
Utensil	Raw milk	5 (min)	10 (min)	Yoghurt	Curd	Fresh	Whey	Storage at 4°C for 24 h
Aluminum	3.01	3.86	7.998	4.72	6.76	7.75	7.21	8.93
Stainless steel		2.98	3.380	3.15	3.51	4.97	6.31	3.61

In yoghurt, aluminium levels ranged from 2.66 to 7.42 ppm. All yoghurt samples had aluminium concentrations below 10 ppm (Fig. 1). These values were higher than those reported by Lopez *et al*. (2000) and Gonzalez-Weller *et al*. (2010) and lower than those reported by Ayar *et al*. (2009). Aluminium levels were significantly lower ($p \leq 0.05$) in yoghurt than in market milk and rice pudding (Table 1). Lower aluminium levels in yoghurt may be attributed to the fact that the yoghurt is fermented and stored in plastic containers.

Rice pudding is a product made of whole milk, rice, sugar and starch. The aluminium content of rice pudding ranged from 1.7 to 361.36 ppm, the mean value was 80.97±2.6 ppm. More than one quarter of these samples contained aluminium concentrations below 10 ppm. Forty percent of the samples had aluminium concentrations between 10 and <50 ppm. One sample had an aluminium concentration of 361.36 ppm (Fig. 1). In general, the aluminium contents of rice pudding were higher than those of farm milk, kareish cheese and yoghurt. Considering the aluminium levels found in milk, the determined aluminium values in rice pudding can be attributed to the milk portion. Contamination caused by rice, sugar, starch and processing equipment can not, however, be excluded.

Since aluminium is ubiquitous in the environment and is used in a variety of products and processes, daily exposure of the population to aluminium is inevitable (Soni *et al*., 2001). Recently, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a Provisional Tolerable Weekly Intake (PTWI) of 2 mg kg^{-1} b.wt. (FAO/WHO., 2011) which means that for an average 60 kg person, a daily intake of 17.14 mg of aluminium is tolerable. In the present study, the aluminium daily intakes from farm milk, market milk, kareish cheese, yoghurt and rice pudding were 3.99, 21.46, 1.15, 0.06 and 8.6 mg day^{-1}, respectively. These values are equivalent to 23.25, 125.21, 6.72, 0.35 and 50.1% of the PTWI (Table 2). The biggest contribution to the intake of aluminium came from market milk and rice pudding, whereas, yoghurt contributed the least. The estimated total intake of aluminium via consumption of milk and milk products was calculated to be 246.72 mg $week^{-1}$ which corresponds to 205.5% of the PTWI (Table 2). The increased aluminium consumption above the recommended PTWI may become a threat to humanity.

Humans are frequently exposed to aluminium. One of the potential sources of additional dietary aluminium is the aluminium utensils (Semwal *et al*., 2006). The concentrations of aluminium in milk and milk products after processing in aluminium and stainless steel utensils are presented in Table 3. Aluminium level in milk boiled for 10 min in aluminium cookware was approximately twice higher than that of the raw milk, whereas, leaching of aluminium during boiling in stainless steel cookware was found to be negligible. Regarding yoghurt production, aluminium level in yoghurt manufactured in aluminium cookware was 1.5 fold higher than that manufactured in stainless steel cookware. Aluminium level in the kareish cheese processed in aluminium cookware was twice higher than that processed in stainless steel cookware. It was observed that the nature of the food has a great effect on the extent of aluminium leaching from the containers. High acidity products such as cheese and yoghurt resulted in greater leaching of aluminium into the food from the utensils. Similar observation has also been reported by Semwal *et al*. (2006). It was also observed that curdling gives rise to increased concentrations of aluminium in the curd and end product compared to raw milk. Similar observation has also been reported by Coni *et al*. (1996). In contrary, Elbarbary and Hamouda (2013) reported that the aluminium level decreases from 0.561 mg kg^{-1} in raw milk to 0.401 mg kg^{-1} in fresh cheese. It was also observed that the duration of heat treatment has a great effect on the aluminium leaching from the utensils. Furthermore, the results obtained suggest that leaching of aluminium was higher from aluminium than stainless steel utensils, indicating the advantage of using stainless steel utensils for heating, processing and storage of milk and milk products, especially those acidic in nature such as yoghurt and cheese.

CONCLUSION

The results clearly indicated that milk and milk products in Beni-Suef governorate are significantly contaminated with aluminium which may carry high health hazards to the consumers. Market milk, rice pudding and farm milk contribute greatly to the total weekly intake of aluminium than yoghurt and kareish cheese. With respect to the leaching of aluminium, the results indicated that processing and storage of milk in aluminium utensils raise aluminium content, so the use of such utensil is not recommended for processing and storage of milk products, especially those acidic in nature such as yoghurt and cheese.

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