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STUDIES ON SOME RESIDUES AND CONTAMINANTS AFFECTING THE SAFETY OF MILK AND SOME DAIRY PRODUCTS

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ABSTRACT

It is important that milk and milk products offered for sale do not contain residues at levels that exceed allowable maximum residue limits (MRLS), so this study aimed to determine the content of lead (Pb), cadmium (Cd), arsenic (As), copper (Cu) and zinc (Zn) in bulk tank milk, soft cheese, yoghurt, powdered ice cream and ice cream (30 samples each) collected from different small scale dairy plants at Kafr El-Sheikh governorate using atomic absorption spectrophotometer for analysis. The obtained results reveal that, the mean Pb levels in the examined bulk milk, soft cheese, yoghurt, powdered ice cream and ice cream samples were 5.5 ± 2.7, 0.2, 1.28 ± 0.55, 1.32 ± 0.6 and 0.0 ppm, respectively. These levels were higher in bulk milk, yoghurt and powdered ice cream than the permissible limits. The mean levels of Cd in the same examined samples were 5.40 ± 4.9, 0.1, 0.00, 0.35 ± 0.22 and 0.00 ppm, respectively. The obtained levels of Cd were higher in bulk milk, soft cheese and powdered ice cream than the permissible limits. For Cu, the mean levels were 5.39 ± 2.02, 2.58 ± 0.24, 1.28 ± 0.13, 2.34 ± 0.24 and 1.33 ± 0.43 ppm, respectively, while for Zn, they were 2.70 ± 0.19, 7.26 ± 0.67, 10.22 ± 0.44, 2.34 ± 0.24 and 2.50 ± 0.34 ppm respectively in the examined samples, respectively. Cu and Zn values were higher than the permissible limits. Arsenic could not be detected from any of the examined samples. Possible health risk of these metals was discussed.

Keywords: heavy metals, milk, dairy products

INTRODUCTION

Heavy metals represent one of the most important groups of pollutants. Toxic metals are present in the environment either naturally or as a consequence of industrial and/or agricultural activities. They find their way to milk through animal feed, water, air, during milk production, dairy processing or during packaging and storage (***Kahniki, 2007***).

Heavy metals such as Cd, Pb and As are not expected to have any direct contact with milk and milk products, except in accidental cases. Milk products contamination reflects the level found in fresh milk, taking into account concentration factors (***Hummelen, 2007***). Metals may be associated with particular milk fractions e.g. Pb and Cd bind strongly to casein. The use of specific milk fraction may thus concentration or remove metals (***Jansen, 1996***).

Excessive intake of heavy metals Pb to many cases of intoxication, ranged between the gastrointestinal disturbance to liver and kidney dysfunction and lung carcinoma (***Gracey and Collins, 1992***). For example, Pb causes encephalopathy in children usually followed by permanent CNS damage (***Carl, 1991***). While in adults avitaminosis, loss of weight and anaemia, liver dysfunction, kidney damage, GIT irritation tumor formation and cardiovascular problems (***Farag et al., 2009***). Chronic Pb poisoning is characterized by anaemia, muscular pains, changes in arterial elasticity resulting in hypertension, toxic effects on reproductive system and Pb nephropathy (***Goyer, 1993***).

Cadmium is an accumulative toxic metal, the main sites of deposition are liver and kidney (***Cussads, 1995***). Possible sources of environmental pollution are through galvanized pipes, pigments, rechargeable batteries, plastics, and effluents from electroplating works, however, food is the primary source of exposure (***WHO, 1995 and Dwivedi et al., 1997***). Anaemia is a common manifestation of chronic Cd toxicity due to its antagonism to Cu and iron (***IARC, 1994***).

Arsenic occurs naturally in the environment as an element of the earth's crust and combined with elements such as oxygen, chlorine and sulfur to form inorganic arsenic compounds. Chronic exposure to arsenic can Pb to dermatitis hyperkeratinization of exposed areas and lung cancer, while acute exposure can cause lung distress and death (***Osha, 2008***).

Copper is an essential element for all plants and animals. It is widely distributed and occurred in food in many chemical forms which affects its availability to the animals (***Watson, 1993***). Both deficiency and excess of Cu in the mammalian system results in untowards effects (***Hostynek et al., 1993***). High Cu levels Pb to various diseases include Mediterranean anaemia., cirrhosis, yellow atrophy of liver and Wilson's disease (hepatolenticular degeneration) (***Underwood, 1977 and Goyer, 1996***).

Zinc is essential for biological function as protein synthesis and carbohydrate metabolism. A number of diseases and dermatological conditions are attributed to Zn deficiency, such as alopecia, acrodermatitis, and enteropthica (***Miyata et al., 1986***). On the contrary high levels of dietary Zn may interfere with the hepatic Cu storage and

may compete with calcium for intestinal absorption. The antagonistic effects of Zn against iron and Cu can result in suppression of hematopoiesis (***Osweiler, 1996***). Therefore, this study was conducted to throw light on the level of some heavy metals (Pb, Cd, As, Cu and Zn) in milk and some dairy products (soft cheese, yoghurt, powdered ice cream and ice cream) and compare the level of these metallic elements in the examined samples with MPL.

MATERIALS AND METHODS

Collection of samples:

One hundred and fifty random samples of bulk milk and some dairy products including soft-cheese, yoghurt, powdered ice cream and ice cream (30 samples of each) were collected from small scale dairy plants in Kafr El-Sheikh governorate during 2010 and 2011. Samples were transferred to the laboratory in ice-pack containers where kept frozen until analysis.

Preparation:

Each sample was prepared according to ***APHA (1992***), then was digested by wet way according to ***Tsoumbaris and Papadopulou (1994)*** using 5 grams from each sample.

Analytical procedures:

All samples were analyzed using air-acetylene unit in atomic absorption spectrophotometer GBC (Avanta) at Central Laboratory of Kafrelsheikh University for determination of Pb, Cd, As, Cu and Zn. The accuracy of the method was confirmed by analysis of standard reference

material calibration against standard solutions (Mono-element CPA chem.., Bulgaria) for all elements. Calibration graphs were linear in the ranges of 5-150 ppm for As, 1-5 ppm for Pb and Cu and 0.5-1.5 ppm for Zn and Cd. Blank absorbance values were monitored throughout the experiment and were subtracted from the readings before the results were calculated.

Statistical analysis:

Data groups for each element were statistically analyzed according to ***Petrie and Watson (1999).***

RESULTS

Table (1): Lead contents in the examined milk and dairy products samples and percent above maximum permissible limits (M.P.L.).

Types of samples	No. of examined samples	Lead levels (ppm)					M.P.L. (ppm)	No. & % of samples exceed M.P.L. (ppm)	
		Positive samples		Min.	Max.	Mean ± SEM			
		No.	%					No.	%*
Bulk milk	30	6	20.0	0.2	15.6	5.5±2.70	0.02 [a]	6	20
Soft cheese	30	1	3.33	0.2			0.3[a]	0	0
Yoghurt	30	5	16.66	0.3	2.9	1.28±0.55	0.2[b]	5	16.66
Powdered ice cream	30	13	43.33	0.1	6.4	1.32±0.6	0.3[c]	5	16.66
Ice cream	30	ND	ND	-	-	-	0.05[d]	0	0

a = Permissible limits (mg/kg) according to E.O.S.Q. (1993)

b = Cited after Carl (1991)

c = Egyptian standards No. 2360 (1993) and No. 1648 milk powdered (2001).

d = FAO/WHO (1993).

P.L. Permissible limits

M.P.L. Maximum permissible limites

ppm = mg/kg

ND Not detected

* % calculated to No. of examined samples

Table (2): Cadmium and arsenic contents in the examined milk and dairy products samples and percent above maximum permissible limits (M.P.L.).

Types of samples	No. of examined samples	Cadmium levels (ppm)					M.P.L. (ppm)	No. & % of samples exceed P.L. (ppm)		Arsenic level (ppm
		Positive samples		Min.	Max.	Mean ± SEM				
		No.	%					No.	%*	
Bulk milk	30	2	6.66	0.5	10.3	5.4± 4.9	0.05[a]	2	6.66	ND
Soft cheese	30	1	3.33	0.1	0.1	0.1	0.05[a]	1	3.33	ND
Yoghurt	30	ND	ND	-	-	-	0.03[e]	ND	ND	ND
Powdered ice cream	30	4	13.33	0.1	1	0.35± 0.22	0.05[f]	4	13.33	ND
Ice cream	30	ND	ND	-	-	-	0.05[d]	ND	ND	ND

a = Permissible limits (mg/kg) according to E.O.S.Q. (1993)
d = FAO/WHO (1993).
e = European maximum level
f = Citek *et al.* (1996)
P.L. Permissible limits
M.P.L. Maximum permissible limits
ppm = mg/kg
ND Not detected
* % calculated to No. of examined samples

Table (3): Copper contents in the examined milk and dairy products samples and percent above maximum permissible limits (M.P.L.).

Types of samples	No. of examined samples	Copper levels (ppm)					M.P.L. (ppm)	No. & % of samples exceed P.L. (ppm)	
		Positive samples		Min.	Max.	Mean ± SEM			
		No.	%					No.	%*
Bulk milk	30	30	100	0.8	52.8	5.39±2.02	0.10[h]	30	100
Soft cheese	30	30	100	1.1	7.5	2.58±0.24	0.3[a]	30	100
Yoghurt	30	29	96.66	0.1	2.4	1.28±0.13	0.4[f]	24	80
Powdered ice cream	30	30	100	1.4	6.6	2.34±0.24	3.0[a]	5	16.66
Ice cream	30	30	100	0.2	13.5	1.33±0.43	0.4[f]	27	90.00

h = IDF 1979. International Dairy Federation. Bulletin, Chemical residues in milk and milk products.
a = Permissible limits (mg/kg) according to E.O.S.Q. (1993)
f = Citek *et al.* (1996)
P.L. Permissible limits
M.P.L. Maximum permissible limits
ppm = mg/kg
ND Not detected
* % calculated to No. of examined samples

Table (4): Zinc contents in the examined milk and dairy products samples and percent above maximum permissible limits (M.P.L.).

Types of samples	No. of examined samples	Zinc levels (ppm)					M.P.L. (ppm)	No. & % of samples exceed P.L. (ppm)	
		Positive samples		Min.	Max.	Mean ± SEM			
		No.	%					No.	%*
Bulk milk	30	30	100	1	4.9	2.7±0.19	3.28[h]	9	30
Soft cheese	30	30	100	0.8	14.1	7.26±0.67	12.0[g]	2	6.66
Yoghurt	30	30	100	4.4	15.1	10.22±0.44	5.0[a]	28	93.33
Powdered ice cream	30	30	100	0.2	26.4	7.48±1.11	15.0[a]	2	6.66
Ice cream	30	30	100	0.8	7.7	2.5±0.34	5.0[a]	3	10

a = Permissible limits (mg/kg) according to E.O.S.Q. (1993)

g = Csapo (2002)

h = IDF 1979. International Dairy Federation. Bulletin, Chemical residues in milk and milk products.

P.L. Permissible limits

M.P.L. Maximum permissible limits

ppm = mg/kg

ND Not detected

* % calculated to No. of examined samples

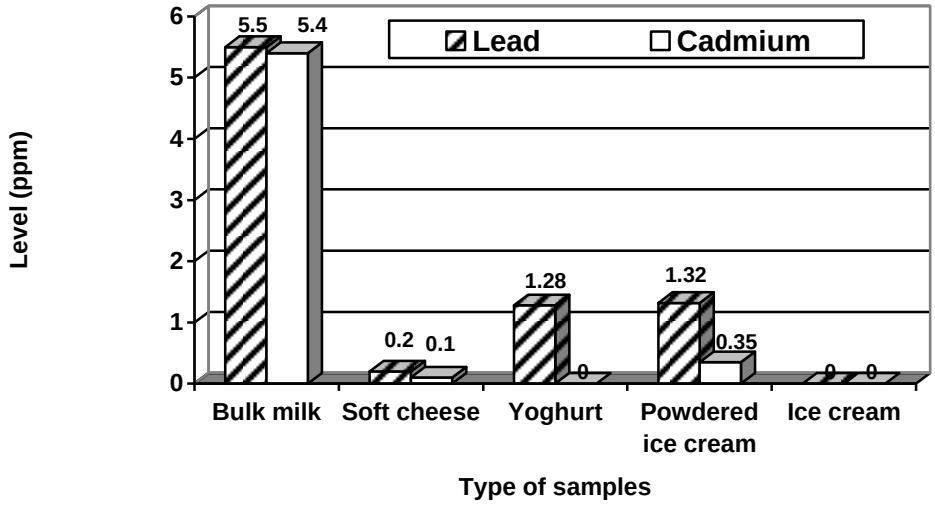

Fig. (1): Mean values of lead and cadmium (ppm) in the milk and dairy products samples.

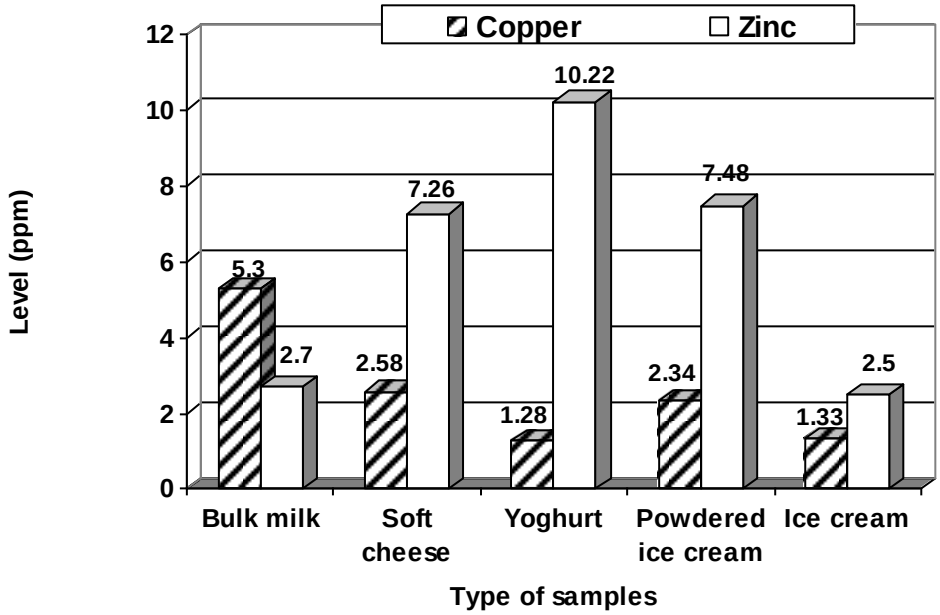

Fig. (2): Mean values of copper and zinc (ppm) in the milk and dairy products samples.

DISCUSSION

Contamination of food and milk by heavy metals is one of the major problems affecting human health. The main source of these heavy metals is the overuse of fertilizers and increase industrial activity. Industrial and agricultural use of heavy metals and their compounds resulted in environmental pollution and presence of metal residues in food chain (***Tork, 1996***).

Results recorded in (Table 1 and Fig. 1) reveal that Pb was detected in the examined samples of bulk milk (20%), soft cheese (3.33%), yoghurt, (16.66%) and powdered ice cream (43.33%) with mean values $\pm$ SEM of 5.5 $\pm$ 2.7, 0.2, 1.28 $\pm$ 0.55 and 1.32 $\pm$ 0.6, respectively, but not detected in ice cream samples.

In comparison with the permissible limits of Pb in bulk milk (0.02 ppm) and powered ice cream (0.3 ppm) according to Egyptian standard 6 (20%) and 5 (16.66%) of the examined samples exceed the MRL, while the only one positive sample of cheese is lower than the MPL (0.3 ppm). Regarding yoghurt, the five positive (16.66%) samples exceed the MRL of Pb (0.2 ppm) as reported by (Carl, 1991) (Table 1). Consequently, bulk milk, yoghurt and powered ice cream represent high risk of consumer.

The given results in (Table 2 and Fig. 2) reveal that Cd was detected in 2 (6.66%), 1 (3.33%) and 4 (13.33%) out of the examined bulk milk, soft cheese and powdered ice cream samples with mean values $\pm$ SEM of 5.4 $\pm$ 4.9, 0.1 and 0.35 $\pm$ 0.22 respectively, but not detected in yoghurt and ice cream samples.

The obtained results were higher than those recorded by ***El-Prince and Sharkawy (1999), Pavlovic et al. (2004) and Maas et al. (2011).*** According to Egyptian standard, the positive samples of bulk milk (2) and soft cheese (one exceeded the MPL of Cd (0.05 ppm), also the four positive samples of powdered ice cream exceeded the same limit that mentioned by ***Citek et al. (1995)***. Presence of Cd in bulk milk may be attributed to grazing of dairy animals near high ways or animal held in industrial areas, as Cd widely used in industry and has the ability to spread through environment (***Venalainen et al., 1996 and Daganoc, 1999***). Also, Cd contaminates the food chain via the use of the sewage sludge and the phosphate compounds as land's fertilizers that results in increased Cd level in animal's feed and milk (***Robards and Worsfold, 1991 and Morcombe et al., 1994***).

Exposure to arsenic is mainly via food intake and drinking water. Long term exposure to As is related to risk of various forms of cancer (mainly skin cancer) and numerous non-cancer diseases including skin lesions, diabetes, chronic cough and toxic effect on liver, kidney, cardiovascular system and peripheral and central nervous system (***Jarup, 2003, Vahter, 2007 and Sobeih and Hegazy, 2011***). Arsenic could not be detected from any of the examined bulk milk and dairy products samples (Table 2).

The mean levels of Cu in the examined bulk milk, soft cheese, yoghurt, powder ice cream and ice cream samples were detected as follows 5.39 $\pm$ 2.02, 2.58 $\pm$ 0.24, 1.28 $\pm$ 1.13, 2.34 $\pm$ 0.24 and 1.33$\pm$ 0.43 ppm, respectively. Higher results were reported by ***Deeb (2010) and Adetunji and Salawu (2008)*** while lower levels were reported by ***Citek et al. (1995) and Nasr et al. (2007)***. (Table 3 and Fig. 2)

Egyptian standard (E.O.S.Q., 1993) cited MPL for Cu as 0.3 ppm in soft cheese and 3.0 ppm in powdered ice cream. Accordingly, 30 (100%) and 5 (16.66%) of the examined samples exceed these limits (Table 3). Moreover, all bulk milk samples exceed the MPL of Cu 0.1 ppm cited by IDF (1979). Comparing our results with the PML (0.4 ppm) of Cu reported by ***Citek et al. (1995),*** 24 (80%) and 27 (90%) of yoghurt and ice cream samples exceeded this limit (Table 3).

Although Cu is an essential trace element (at low concentration), it is toxic at high level. Daily intake of an excessive dose of Cu may Pb to Wilson's disease which is manifested by destruction of nerve cells, liver cirrhosis, ascitis, oedema and hepatic failure (***Gossel and Bricker, 1990***). Moreover, high level of Cu in milk has an adverse effect on lactic acid bacteria and production of dairy products with bad quality (***Bottazzi et al., 2000***).

The mean levels of Zn in the examined bulk milk, soft cheese, yoghurt, powdered ice cream and ice cream samples were recorded as follows 2.70 ± 0.19, 7.26 ± 0.67, 10.22 ± 0.44, 7.48 ± 1.11 and 2.50 ± 0.34 ppm, respectively lower values were reported by ***Moreno Rojas et al. (2000), El-Prince and Abdel-Mohsen (2001) and Jeena Zagroska (2007).*** Nearly similar results were obtained by ***Ukic (1998);*** and higher results which exceeded the MRLs were recorded by ***Miedzobrodzka et al. (1995).*** **(Table 4 and Fig. 2)**

Egyptian Standard, (***E.O.S.Q.C., 1993***) cited MPL for Zn as 5.0 ppm in yoghurt, 15 ppm in powdered ice cream and 5.0 ppm in ice cream. Accordingly, 28 (93.33%), 2 (6.66%) and 3 (10%) of the examined samples exceed these limits (Table 4). While in bulk milk samples 9 (30%) exceed the MPL of Zn (3.28) cited by ***IDF (1979)***. Moreover, 2 (6.66%) soft cheese samples exceeded the MPL of Zn 12.0 ppm cited by (***Csapo and Csapone, 2002***) (Table 4).

High level of dietary Zn may interfere with the hepatic Cu storage and may compete with calcium for intestinal absorption. The antagonistic effects of Zn against iron and Cu can result in suppression of hematopoiesis (***Osweiler, 1996***)

In conclusion, high concentrations of Pb and Cd were detected in bulk milk and dairy products. It is necessary to warning about the hazardous effects of these toxic elements on human being. The consuming of such products considered as an additional source of exposure beside the direct sources of air, water and plants. Moreover, the effect of consumers may contribute through disturbing the levels of essential elements (Cu and Zn) in vital foods.

Therefore, the preventive measures for minimizing the pollution of milk and milk products with such metals are of significant concern and regular examination of milk and dairy products and their load for heavy metals should be evaluated according to the international guidelines.

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دراسات على بعض المتبقيات والملوثات المؤثرة على سلامة اللبن وبعض منتجاته

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يعتبر التلوث بالمعادن الثقيلة من أخطر أنواع التلوث لما له من أثر سلبي على الجهاز العصبي والدم وتأثيره على مستوى الذكاء وبخاصة الأطفال ولما لهذه المعادن من أثر تراكمي في الجسم أجريت هذه الدراسة لتقدير بقايا المعادن الثقيلة فى اللبن الخام وبعض منتجاته حيث تم تجميع 150 عينة (30 عينة من كل نوع) من اللبن الخام والجبن الطري والزبادي وبودرة الأيس كريم والأيس كريم من المصانع الصغيرة بمحافظة كفرالشيخ خلال عامى 2010 ، 2011. تم تحليل هذه العينات لتقدير مستوى كل من الرصاص والكادميوم والزرنيخ والنحاس والزنك باستخدام جهاز الامتصاص الذرى وأوضحت النتائج وجود ارتفاع نسبة الرصاص والكادميوم عن الحد المسموح به فى العينات وغياب الزرنيخ من جميع العينات ، أما نسبة النحاس والزنك فى العينات موضع الدراسة كانت أعلى نسبيا من المسموح به. وقد تم مناقشة النتائج وخطورة تواجد هذه المعادن على الصحة العامة للإنسان وكذلك الاحتياطات اللازمة لتجنب هذه المخاطر.