



Assessment of Heavy Metals Residues in Milk Powder and Infant Milk Formula Sold in Mansoura City, Egypt

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Key words

ABSTRACT:

Heavy Metals, Residues, Milk Powder,

A total of 75 samples including, 25 samples each of full cream milk powder, skimmed milk powder and infant milk formula collected from Mansoura city, Egypt at the period between April and September 2014 were analyzed for measuring Lead, Chromium and Cadmium levels. Flame Emission Atomic Absorption Spectrophotometer could detect Lead, Chromium and Cadmium by means of 0.05 ± 0.01 , 2.70 ± 0.34 and 0.09 ± 0.02 mg/Kg in examined full cream milk powder, 0.03 ± 0.01 , 1.98 ± 0.24 and 0.03 ± 0.01 mg/Kg in examined skimmed milk powder and 0.03 ± 0.01 , 3.88 ± 0.34 and 0.04 ± 0.003 mg/Kg in examined infant milk formula, respectively. By comparing the concentration of heavy metals present in analyzed samples to their permissible limits, it was noted that most of tested samples contained heavy metals by levels exceeding maximum permissible limits proposed for them, hence milk powder and infant milk formula may carry a potential health hazards for humans which needs a rigorous monitoring program to prevent food contamination by these toxic heavy metals and to ensure that their levels did not exceed the legal limits for human consumption.

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1. INTRODUCTION

Milk is one of the most important food for human nutrition, as it contains all the macronutrients namely protein, carbohydrates, fat, sugars, vitamins (A, D and B groups) and trace elements particularly calcium, phosphate, magnesium, zinc and selenium (Buldini et al., 2002). Therefore, milk and dairy products are important components of human diets that are widely consumed by human children and adults especially elderly people around the World (Qin et al., 2009).

Powdered milks are one of the most essential dairy products needed by growing children. It contains both the basic and additional requirement needed by children especially during their developmental years. It is also one of the most popular dairy products due to long shelf life and its employment in the manufacture of many dairy products such as ice cream, cheese, evaporated milk, condensed milk and infant milk formula and also as an ingredient in many bakery products, processed meats and soups.

Heavy metals are persistent contaminants in the environment that can cause serious environmental and health hazards. They are released into the environment from natural as well as man-made activities. Some heavy metals (like copper and iron) are essential to nutritive maintain proper metabolic activity in living organisms; (lead and

cadmium) are non-essential and have no biological role also, chromium considered as non-nutritive non-toxic (Ayar et al., 2009). However, at high concentrations, they can cause toxicity to living organisms (Li et al, 2005).

Milk and dairy products become contaminated with heavy metals either through contamination of the original cow's milk, which may be due to exposure of lactating cow to environmental pollution or consumption of contaminated feed stuffs and water (Carl, 1991 and Okada et al., 1997). Also, trace metals may enter our foods from other sources as: (i) water used in food processing or cooking, (ii) equipment, containers and utensils used for food processing and (iii) packaging, storage and cooking. Intestinal absorption is dependent on the metal considered and is carried out by passive diffusion or active transport (USEPA, 2003). Moreover, raw milk may be exposed to contamination during its manufacture and packaging processes (Ukhun et al., 1990).

Milk powder may be contaminated by heavy metals at certain levels that may cause toxicity to consumers. This toxicity is attributed to accumulation of heavy metals in the body which are not metabolized to extractable products. Contamination of milk powder with heavy metals may cause a serious risk for human health because of the consumption of even small amount of metals can lead to considerable concentrations in human

body and exert their toxic effect by combination with one or more reactive groups essential for normal physiological function and cellular disturbances (Friberg and Elinder, 1988).

Heavy metals are toxic because they may have cumulative deleterious effects that can cause chronic degenerative changes (Ibrahim et al., 2006), especially to the nervous system, liver, and kidneys, and, in some cases, they also have teratogenic and carcinogenic effects (IARC, 1987). The mechanism of toxicity of some heavy metals still remains unknown, although enzymatic inhibition, impaired antioxidants metabolism, and oxidative stress may play a role. Heavy metals generate many of their adverse health effects through the formation of free radicals, resulting in DNA damage, lipid peroxidation, and depletion of protein sulfhydryls (e.g., glutathione) (Valko et al., 2005).

The presence of heavy metals as lead, cadmium and chromium even in low concentrations leads to metabolic disorders with extremely serious consequences and causing serious problems as it causes many health problems such as weakness, heart failure, cancer and also affects the kidneys (McCally, 2002 and Licata et al., 2004).

Since, as far to our knowledge, reports regarding heavy metals determination in milk powder in Egypt is scarce and due to the serious problems associated with ingestion of these metals in food, this study was planned to throw the light on determination of heavy metals in milk powder samples collected from Mansoura city, Dakahlia Governorate, Egypt.

2. MATERIALS AND METHODS

2.1. Collection of samples:

A total of 75 samples from 3 different brands of full cream milk powder, skimmed milk powder and infant-based formula were randomly purchased from groceries, supermarkets and pharmacies distributed in Mansoura city, Dakahlia, Egypt at the period between April and September 2014. All samples were still valid for consumption. The samples were kept in their original packages and were individually packed into a clean polyethylene bag. Each sample was labeled to identify the source, site and date of sampling, then transferred to the laboratory of Food Hygiene and Control Department, Faculty of Veterinary Medicine, Mansoura University, wherein the preparation of the samples were done to be ready for analysis of lead, Chromium and cadmium.

2.2. Preparation of samples:

2 ml of 30 % HNO₃ was added to 100 ml of deionized water to prepare dilution water. All glassware was initially washed with detergent and water, then washed by diluted water and rinsed several times with deionized water, then allowed to be dried.

1g of the sample was added into 100 ml beaker, then 5 ml of HNO₃ (16 N) and 5 ml of H₂O₂ (30% V/V) were added to the sample. The mixture was left to allow the reaction to proceed, then placed on a hot plate and heated to 60°C for 30 minutes. The beaker was removed and allowed to cool for 5 minutes and then 10 ml of HNO₃ and 5 ml of H₂O₂ were added. The beaker was placed on a hot plate and the temperature was increased gradually. The addition of HNO₃-H₂O₂ was continued until samples became clear. The digested samples were filtered through Whatman filter paper No. 42. The clear filtrate of each sample was transferred into 100 ml capacity volumetric flask (Agemian et al., 1980).

For detection of lead, cadmium and chromium, the diluted standard solution for each metal in strength of 0.005, 0.01 and 0.1 ppm were prepared from 1000 ppm stock spectrosol solution. All standards were made in 1% HNO₃.

2.3. Measurement of heavy metals:

All filtered samples were analyzed for their heavy metals content according to methods of (Agemian et al. 1980 and Perkin-Elmer, 1980) by using "contraAA® 700 High-Resolution Continuum Source atomic absorption spectrometer for flame, hydride, graphite tube and HydrEA technique Atomic Absorption Spectrophotometer" at the central laboratory, Faculty of Agriculture Sba-Basha, Alexandria University, Egypt. The apparatus has digital absorbance and concentration readout capable of operating at wavelengths of 217, 228.8 and 357.8 nm for Lead, Cadmium and Chromium, respectively.

2.4. Statistical analysis:

Data was collected, arranged, summarized and then analyzed using the computer program SPSS/PC+(2001).

3. RESULTS AND DISCUSSION

Table (1): Heavy metals concentration in examined full cream milk powder samples (mg/Kg).

Heavy metals	N	Minimum	Maximum	Mean±SE
Lead	N=25	0.002	0.15	0.05±0.01
Chromium	N=25	N.D	6.95	2.70±0.34
Cadmium	N=25	0.02	0.32	0.09±0.02

Table (2): Heavy metal concentration in examined skimmed milk powder samples (mg/Kg).

Heavy metals	N	Minimum	Maximum	Mean±SE
Lead	N=25	N.D	0.12	0.03±0.01
Chromium	N=25	N.D	4.68	1.98±0.24
Cadmium	N=25	N.D	0.18	0.03±0.01

Table (3): Heavy metal concentration in examined infant milk formula samples (mg/Kg).

Metal	N	Minimum	Maximum	Mean±SE
Lead	N=25	N.D	0.12	0.03±0.01
Chromium	N=25	1.06	8.54	3.88±0.34
Cadmium	N=25	0.02	0.09	0.04±0.003

Table (4): Frequency distribution of heavy metals in total of 25 full cream milk powder samples.

Metal	Permissible limit mg/kg	Within Permissible limit		Over Permissible limit	
		No of samples	%	No of samples	%
Lead	0.02 ^b	12	48	13	52
Chromium	2.0 ^a	9	36	16	64
Cadmium	0.05 ^c	8	32	17	68

a = National Standards of People's Republic of China, GB 2762-2012

b = Codex Standard 108-1981

c = Egyptian standard No. 1648 milk powder (2000).

Table (5): Frequency distribution of heavy metals in total of 25 skimmed milk powder samples.

Metal	Permissible limit mg/kg	Within Permissible limit		Over Permissible limit	
		No of samples	%	No. of samples	%
Lead	0.02 ^b	13	52	12	48
Chromium	2.0 ^a	15	60	10	40
Cadmium	0.05 ^c	4	16	21	84

a = National Standards of People's Republic of China, GB 2762-2012

b = Codex Standard 108-1981

c = Egyptian standard No. 1648 milk powder (2000).

Table (6): Frequency distribution of heavy metals in total of 25 infant formula samples.

Metal	Permissible limit mg/kg	Within Permissible limit		Over Permissible limit	
		No of samples	%	No. of samples	%
Lead	0.02 ^b	12	48	13	52
Chromium	2.0 ^a	3	12	22	88
Cadmium	0.05 ^c	18	72	7	28

a = National Standards of People's Republic of China, GB 2762-2012.

b = Codex Standard 108-1981

c = Egyptian standard No. 1648 milk powder (2000).

3.1. Full cream milk powder:

The results of atomic absorption spectrometry revealed that the levels of heavy metals in examined samples of full cream milk powder ranged from 0.002-0.15 mg/Kg with a mean of 0.05 ± 0.01 mg/Kg for Lead, 0.00-6.95 mg/Kg with a mean of 2.70 ± 0.34 mg/Kg for Chromium and 0.02-0.32 mg/Kg with a mean of 0.09 ± 0.02 mg/Kg for Cadmium (Table 1).

The mean level of Lead in our study is lower than that stated by (Mohamed, 2005) who could detect lead in examined milk powder samples by a mean level of 0.76 ± 0.07 mg/Kg and by (Salah, 2013) who found that the average concentration of Lead in examined milk powder samples collected from different outlets in Dakahlia Governorate, Egypt was 0.79 mg/Kg. Also, higher lead levels were conducted by (Abd- El Aal, 2012) who found that the mean values of Lead in milk powder samples distributed in Zagazig city, Egypt during three periods of storage were 0.48, 0.57 and 0.80 mg/Kg, respectively. Meanwhile, lower Lead value at 0.034 mg/kg was estimated by (Al-Zahrani, 2012). Very low level of Chromium (0.02 mg/Kg) was detected by (Farid et al., 2004) in cow's milk powder in Saudi Arabia. Another low chromium values at 0.18 mg/Kg and 0.28 mg/Kg were estimated in milk powder by (Birghila et al., 2008) in Romania and by (Qin et al., 2009) in China, respectively.

Higher values of Cadmium at 0.33 ± 0.06 mg/Kg were estimated by (Mohamed, 2005) in examined milk powder samples and by (Salah, 2013) who found that the average concentration of Cadmium in examined milk powder samples collected from different outlets in Dakahlia Governorate, Egypt was 0.32 mg/Kg. Also, higher cadmium levels were estimated by (Abd- El Aal, 2012) who found that the mean values of Cadmium in milk powder samples distributed in Zagazig city, Egypt during three periods of storage were 0.225, 0.330 and 0.345 mg/Kg, respectively.

The heavy metal contents varies widely due to many factors such as differences between species, characteristics of the manufacturing practices and possible contamination coming from the equipments during the process (Yuzbasi et al., 2003 and Caggiano et al., 2005). Higher values of heavy metals in milk powder may have been arisen from contamination during handling and processing. The processing steps mainly involve boiling and frying in steel or Aluminium-ware from which such

contamination may have results (Onianwa et al., 1996). Increased Lead concentration in milk powder may be attributed to contamination of original cow's milk used for manufacture of these products (Nasef, 2002). The contamination of original cow's milk may be due to excessive exposure of lactating cows to environmental Lead from heavy traffic, consumption of contaminated water and feed stuffs (Okada et al., 1997). Moreover, raw milk may be contaminated from metallic Lead from Lead soldered cans (El-Batanoni and Abo El-Ata, 1996).

By comparison the concentration of heavy metals present in analyzed full cream milk powder to the permissible limits (mg/kg) of each tested metals, we found that 48% (12/25), 36% (9/25) and 32% (8/25) of tested samples were within the permissible limits proposed for Lead, Chromium and Cadmium, respectively, meanwhile, 52% (13/25), 64% (16/25) and 68% (17/25) of samples were above the permissible limits for Lead, Chromium and Cadmium, consecutively (Table 4).

Not all samples in our study contained Lead and Cadmium above the permissible limits which were in contrast to that reported by (Abdallah, 2005) in Damietta city/Egypt and (Amer et al., 2006) in Sharkia Governorate, Egypt, who found that all of examined samples of whole milk powder contained lead and cadmium by levels above the recommended permissible limits. Also, (Amer and Aiad, 2012) revealed that obtained Lead and Cadmium levels in all examined samples of milk powder collected from Alexandria city, Egypt were complied with Egyptian Standards (1993). Meanwhile, (Salah, 2013) found that all examined samples of milk powder collected from different outlets in Dakahlia Governorate, Egypt had Lead and Cadmium residues over the permissible limit.

3.2. Skimmed milk powder:

It is noticed from Table (2) that examined samples of skimmed milk powder contained heavy metals by levels ranged from 0.00-0.12 mg/Kg with a mean of 0.03 ± 0.01 mg/Kg for Lead, 0.00-4.68 mg/Kg with a mean of 1.98 ± 0.24 mg/Kg for Chromium and 0.00-0.18 with a mean of 0.03 ± 0.01 mg/Kg for Cadmium.

Very low level of Lead was detected by (Abdulkhaliq et al., 2012) who found that the mean concentrations of Lead was 0.002 mg/kg in examined Skimmed milk powder in Ramallah City, Palestine.

Our mean values of Cadmium was higher than that reported by (Abdulkhaliq et al., 2012) who found that the mean concentrations of Cadmium was 0.00001 mg/kg in examined Skimmed milk powder from Ramallah City, Palestine and by (O'Keeffe et al., 2001) who couldn't determine cadmium in any samples of skimmed milk powder.

It is obvious from Table (5) that heavy metals levels in 52% (13/25), 60% (15/25) and 16% (4/25) of examined skimmed milk powder were within the permissible limits proposed for Lead, Chromium and Cadmium, respectively, meanwhile, 48% (12/25), 40% (10/25) and 84% (21/25) of samples have heavy metals concentration above the permissible limits stated for Lead, Chromium and Cadmium, respectively.

3.3 Infant milk formula:

It is indicated from our study that tested samples of infant milk formula were contaminated by heavy metals with concentrations ranged from 0.00-0.12 mg/Kg with a mean value of 0.03±0.01 mg/Kg for Lead, 1.06-8.54 mg/Kg with a mean level of 3.88±0.34 mg/Kg for Chromium and from 0.02-0.09 mg/Kg with a mean concentration of 0.04±0.003 mg/Kg for Cadmium (Table 3).

Our results of lead is nearly similar to that reported by (Moreno-Rojas et al., 2002) who found that the mean concentrations of lead were 0.026±0.008 mg/Kg of examined Spanish infant formulas. Lower level of Lead was found in the study achieved in Philippines by (Cruz et al., 2009) in which, all tested samples of infant formulas for infants aged from 6 to 12 months were negative for Lead. Meanwhile, higher lead levels at 0.38±0.22 mg/kg and 0.109 mg/kg were estimated in Iran by (Behrooz et al., 2009), in Brazil by (De Castro et al., 2010), respectively and in Zagazig city, Egypt by (Abd- El Aal, 2012) who found that the mean values of Lead in infant formula samples during three periods of storage were 0.410, 0.561 and 0.815 mg/kg, respectively. The finding of Chromium was in contrast to the study conducted by (Bamidele et al., 2007) in which, Chromium was not detected in any tested sample. Also, very low value of Chromium (0.001 mg/Kg) was estimated by (Solidum et al., 2012) in powdered children's milk in Philippines.

A higher Cadmium level of 0.359±0.215 mg/kg in examined infant formula was calculated in Iran by (Behrooz et al., 2009) and by (Abd- El Aal, 2012) who found that the mean values of Cadmium in infant milk formula samples distributed in Zagazig city, Egypt during three periods of storage were 0.210, 0.280 and 0.285 ppm, respectively. But,

lower Cadmium level at 0.033 mg/kg was detected by (De Castro et al., 2010) in analyzed infant formulas from Brasilia, Brazil.

Our finding indicated that the obtained levels of Lead, Chromium and Cadmium were within the permissible limits in 48% (12/25), 12% (3/25) and 72% (18/25) of examined infant milk formula samples, successively and were above the permissible limits in 52% (13/25), 88% (22/25) and 28% (7/25) of the tested samples, respectively (Table 6).

By comparing our obtained levels of Lead and Cadmium to the permissible limits we found that not all samples were within the permissible which was different than that reported by (Amer and Aiad, 2012) who revealed that the obtained Lead and Cadmium levels in all examined samples of infant formula collected from Alexandria city, Egypt were complied with Egyptian Standards (1993). Also, our findings was not similar to that reported by (Solidum et al., 2012) who found that the Cadmium and Chromium contents in all examined samples of children's milk powder are within the safe limits recommended by the World Health Organization.

There are several sources of contamination of infant formula with heavy metals; among these sources drinking water used to mix infant powdered formula which may add Cadmium and Lead significantly to the concentrations in the ready-made products (Ljung et al., 2011). From the previously mentioned results, it is evident that the metal contents of dairy products are variable because of the possible contamination from the equipment during processing, packaging, and storage. So, it is necessary to control the manufacturing process at each step, in order to determine the source and levels of contamination and to ensure the desired product quality (Ayar et al., 2009).

Conclusion:

The current study concluded that milk powder and infant milk formula distributed in Mansoura city may carry a potential health hazards for humans due to the majority of these examined samples contained Lead, Chromium and Cadmium by levels exceeded the maximum permissible limits proposed for them. Therefore, a rigorous monitoring program should be followed for estimation of these heavy metals in food to ensure that their levels did not exceed the legal limits for human consumption.

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