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Heavy metals residues and trace elements in milk powder marketed in Dakahlia Governorate

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Abstract

Fifty random milk powder samples were collected from different outlets in Dakahlia Governorate, Egypt and analyzed by Atomic Absorption Spectrophotometer to determine heavy metals residues and trace elements [lead (Pb), Cadmium (Cd), aluminium (Al), iron (Fe), selenium (Se) and manganese (Mn)]. The average concentration of Pb, Cd, Al, Fe, Mn and Se in examined milk powder samples were 0.791; 0.322; 1.57; 20.41; 0.497 and 0.014 ppm, respectively. All examined samples of milk powder had Pb, Cd, Fe, and Mn residues over the permissible limit. At the meanwhile, 96 and 58% of examined samples had Al and Se levels above the permissible limit, respectively. The calculated daily intake of Pb, Cd, Al, Fe, Mn and Se from consumption of 200 ml reconstituted milk powder per day were 158.5; 64.4; 313.4; 4082; 99.4 and 2.8 µg, respectively, which contributed about 31.64; 92.0; 26.12; 85.04; 1.99 and 3.5% from the Acceptable Daily intake of these elements, respectively. Public health significance and hazardous of these metals were discussed.

Introduction

Wide varieties and range of concentrations of metals are distributed everywhere in nature. While in human body, most of metals have come through food. However, not all consumed metals are retained, some pass straightly through and lost in feces and some are firstly absorbed and then lost in sweat, urine, bile excretion and in discarded hair and skin.

Heavy metals are persistent as contaminants in the environment and come to the fore front of dangerous substances causing health hazards in human. Pb, Cd and Al are among the most important of these elements. Industrial and agricultural processes have resulted in an increased concentration of heavy metals in air, water, soil and subsequently, these metals are taken by plants or animals and find their ways into food chain (Ahmad, 2002). The presence of heavy metals in dairy products may be attributed to contamination of the original cow's milk, which may be due to exposure of lactating cow to environmental pollution or consumption of feeding stuffs and water (Carl, 1991; Okada *et al*., 1997). Moreover, raw milk may be exposed to contamination during its manufacture (Ukhun *et al*., 1990; El-Batanouni and Abo El-Ata, 1996).

Among the metals found in the human body, only a small number are believed to be essential as Se, Fe and Mn (Lopez-Garcia *et al.*, 2009). Deficiency of any one of essential metal nutrients will result in specific biochemical lesions within cells of the body and subsequently development of certain characteristic clinical symptoms which will normally alleviate when the deficiency is corrected by supplying an adequate amount of the missing element. Although these metals are essential for health, excessive exposure may be hazardous (Protasowicki, 1992; Akhter *et al*., 2004). This is complicated even further if there is a very narrow range between the concentration at which metal is considered essential and considered toxic (Higharm and Tomkins, 1993).

Milk powder is 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. Usually, essential elements are normally added to milk powder during manufacture, in order to meet nutritional requirements (Oskarsson *et al*., 1995). Hence, it is necessary to control the level of the added elements since their excess may play a role as a potential source of exposure.

While the main quality parameters for milk powder are the microbiological and sensory characteristics, recent investigations have been recommended that the determination of heavy metals and trace elements in milk powder should be included to these parameters. (Karadjova *et al*., 2000; Martino *et al*., 2000; Orak *et al*., 2005). Since, as far to our knowledge, reports regarding heavy and trace metals determination in milk powder in Egypt is scarce,

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this work was undertaken to determine their levels in milk powder samples collected from Dakahlia Governorate, Egypt.

Materials and Methods

Collection of samples

50 random milk powder samples, locally packaged in Egypt, were collected from different outlets in Dakahlia Governorate, Egypt in their original package. Collected samples were taken to the laboratory without delay. Each sample was labeled to identify the source, site and date of sampling.

Preparation of samples

0.3 gm of each sample was transferred in clean and acid washed screw capped digestion tubes. Two tubes were prepared from each sample and all digestion tubes were identified for examination.

Analysis of the prepared samples

For determination of Pb, Cd, Fe and Se, the first tube of each prepared sample was digested according to Tsoumbaris and Papadop (1994). While, for determination Al and Mn, the procedure was carried out on the second tube according to Dabeka and Mckenzie (1992). All filtered samples were analyzed for their metal contents according to methods of Medina *et al*. (1986) by using "perkia-Elmer Atomic Absorption Spectrophotometer model d 2380, USA, 1998" at the micro analytical laboratory, Department of Chemistry, Faculty of Science, Mansoura University, Egypt. Instrumental analysis of Pb, Cd, Fe and Se and were conducted by air lacetylene Flame Atomic Absorption Spectrophotometer (FAAS). While for determination of Al and Mn, Graphite Furnace Atomic Absorption Spectrophotometer (GFAAS) was used. The analytical detection limits of Pb, Cd, Fe, Se, Al and Mn for the used instrumentation were 0.02 ppm, 0.0006 ppm, 0.05 mg/kg, 0.002 ppm, 0.02 ppm and 0.01 ppm, respectively.

Calculation of the daily metal intake

The daily metal intake was estimated from the consumption of milk powder and the mean data of the present study were combined with the consumption data obtained from Nutrition Institute, Cairo, A.R.E. (1996). Comparison of the calculated daily metal intake from milk powder with the Acceptable Daily Intake (ADI) values recommended by FAO\WHO (1972, 1974c, 1982, 1987, and 1989), FAO\WHO (1999), Food and Nutrition Board (1980), Dreosti (1986) and Pennington (1987).

Table 1. Statistical analytical results of heavy metals and trace elements residues (ppm) in examined milk powder samples (N = 50)

Metals	No. of examined samples	% of positive samples	Minimum	Maximum	Mean	Mean ± S.E
Lead	50	100.0	0.45	1.85	0.791	0.791±0.057
Cadmium	50	100.0	0.10	1.45	0.322	0.322±0.039
Aluminium	50	100.0	0.33	2.83	1.57	1.57±0.094
Iron	50	100.0	15.35	29.65	20.41	20.41±0.621
Manganese	50	100.0	0.35	1.05	0.497	0.497±0.030
Selenium	50	100.0	0.005	0.025	0.014	0.014±0.001

Table 2. Previous related results as compared to results of this study

Similar values were reported by	Higher values were reported by	Lower values were reported by
Morrison (1988); Finoli and Rondinini (1989); Cabrera *et al.* (1995) and Jorhem and Engman (2000)	Madeha *et al.* (1994) and Hamouda (2002)	Ukhun *et al.* (1990); Metrovic *et al.* (1992); Fathi *et al.* (1995); Elena *et al.* (1999); Abdou and Eman (2001) and Abdulkhaliq (2012).
Morrison (1988) and Abdou and Eman (2001)	--	Fathi *et al.* (1995); Elena *et al.* (1999); El-Malt (2001) and Abdulkhaliq (2012)
Woollard *et al.* (1990) and Bloodworth *et al.* (1991)	Garcia *et al.* (1999).	Onianwa *et al.* (1996); Elena *et al.* (1999) and Amal (2000)
Jorhem (2000) and El-Prince and Abd El-Mohsen (2001)	Jorhem and Engman (2000).	Abdulkhaliq (2012); Park (2000); Sharkawy and Manal (2002); Ukhun *et al.* (1990).
Murthy and Rhea (1971) and Garcia *et al.* (1999).	Abdou and Eman (2001) and El-Prince and Abd El-Mohsen (2001)	Ukhun *et al.* (1990); Cabrera *et al.* (1995) and Elena *et al.* (1999)
Golubkina (1996)	Cabrera *et al.* (1996).	Elena *et al.* (1999) and Garcia *et al.* (1999)

Statistical analysis

All the data analyzed using SPSS/PCT (Foster, 2001).

Results and Discussion

Analysis of whole milk powder samples indicated their contamination by some heavy metals residues, exhibiting a wide array of hazardous impacts on human health. Beyond certain limits, all metals turned to be toxic to human body. This could be applied to essential minerals like Fe; Mn and Se, as well as non-essential metals and metalloids like Pb and Cd compounds. In relation to many previously reported surveys worldwide, Tables 1 and 2 summarized results obtained in this study and correlated our findings with results of these surveys.

The statistical analysis of obtained data (Table 3) revealed that all examined milk powder samples having Pb, Cd, Fe and Mn levels above the recommended permissible limit. The obtained results lied above the permissible limit recommended by Wenlok *et al.* (1979), Egyptian Standard (1993) and Egyptian standard (2001). While, 48 (96%) out of 50 examined samples having Al levels above the permissible limit recommended by Pennington (1987). However, 29 (58%) out of 50 examined samples having Se levels above the permissible limit recommended by Thorn *et al.* (1976).

Table 3. Frequency distribution of heavy metals and trace elements residues in examined milk powder samples (N = 50)

Metals	Permissible limit mg/kg (ppm)	Within permissible limit		Over permissible limit	
		No. of samples	%	No. of samples	%
Lead	0.3[a]	0	0.0	50	100.0
Cadmium	0.05[a]	0	0.0	50	100.0
Aluminium	0.5[b]	2	4.0	48	96.0
Iron	2.7[c]	0	0.0	50	100.0
Manganese	0.10[d]	0	0.0	50	100.0
Selenium	0.011[e]	21	42.0	29	58.0

a: Egyptian standard (2001)
b: Pennington (1987)
c: Egyptian standard (1993)
d: Wenlock *et al*. (1979)
e: Thorn *et al*. (1976)

Table 4. Comparison of Acceptable Daily Intake (ADI) value of heavy metals and trace elements with the calculated daily intake from milk powder

Metals	ADI µg/70 kg person	Mean concentration of metals (µg/L) in examined samples	Calculated daily intake of metals from consumption of 200 ml reconstituted milk powder per day [f]	
			µg/day/person	%
Lead	500[a]	791	158.5	31.64
Cadmium	70[a]	322	64.4	92.0
Aluminium	1200[b]	1567	313.4	26.12
Iron	4800[c]	20410	4082	85.04
Manganese	5000[d]	497	99.4	1.99
Selenium	80[e]	14	2.8	3.5

a: FAO/ WHO, Joint Expert Committee on Food Additives WHO Technical Report Series, 1972, 1974c, 1982, 1987 and 1989.
b: Pennington (1987).
c: FAO/ WHO, Joint Expert Committee on Food Additives, 1999.
d: Food and Nutrition Board (1980).
e: Dreosti (1986).
F: Daily consumption of milk for adult person according to Nutrition

From the above mentioned results, it is evident that examined milk powder samples were contaminated with variable amounts of heavy metals. Higher values in milk powder may have been arisen from contamination during handling, exposure 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).

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; Caggiano *et al*., 2005). Oxidation of containers and equipments were affected by some parameters such as pH, quality of raw materials of containers and equipments. Enhancement of oxidation will increase the metal contents of samples. Metal contents of original milk will subsequently affect the metal levels in milk powder.

Increased Pb 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 Pb from heavy traffic, consumption of contaminated feeding stuffs and water (Okada *et al.*, 1997). Moreover, raw milk may be contaminated from metallic Pb from Pb soldered cans (El-Batanoni and Abo El-Ata, 1996). Pb was mainly associated with caseins in cow's milk and freezing or heating did not cause significant changes in distribution of Pb in cow's milk and milk products (Mata *et al.*, 1996) but desiccation process produces significant increases in Pb concentration (Moreno-Rojas *et al*., 1994).

Milk and dairy products usually contain very low concentration of Cd except when diary animals consumed contaminated feeds and water (Cabrera *et al*., 1995; Okada *et al*., 1997). Moreover, contamination during storage, marketing and leaching from containers may be considered as a source of Cd in milk and other dairy products. The distribution change of Cd after heat treatment of milk due to the formation of complexes between the whey proteins and the metal or to the desegregation of the Cd bound to casein micelles.

Also, the mean levels of Al were higher in examined samples and this may be attributed to migration of this element from packaging materials into the milk products (Poonam *et al*., 1997). The high levels of Fe in milk powder may be attributed to its fortification with Fe. The Se content of milk depends largely on animals feed and its geographic distribution (Klaassen *et al*., 1986).

The results recorded in Table (4) indicated that the average concentration of Pb, Cd and Al were 791, 322 and 1567 µg/L, respectively. These concentrations gave a daily intake of about 158.5, 64.4 and 313.4 µg/person, respectively and these quantities representing 31.64, 92.0 and 26.12% of the acceptable daily intake recommended by Pennington (1987) and FAO/WHO (1987). At the same time, the average concentration of Fe, Mn and Se were 20410, 497 and 14 µg/L, respectively. These concentrations supplied a daily intake about 4082, 99.4 and 2.8 µg/person, respectively and these quantities representing 85.04, 1.99 and 3.5 of acceptable daily intake recommended by Food and Nutrition Board (1980), Dreosti (1986) and FAO/WHO (1999). From aforementioned results, it could be concluded that the daily intake of Pb, Cd, Al and Fe were relatively high, but for Mn and Se were relatively low.

Regarding the public health hazards of the detected metals, Pb is a potent neurotoxin for which no safety threshold has yet been found (US Environmental Protection Agency, 2003). *In-vitro* exposure to Pb and during infancy irreversibly affects development of the nervous system, causing reduced learning disabilities. Cd has estrogenic properties and causes an increased incidence of cancer in mice (Johnson *et al*., 2003). Chronic exposure to Cd and Pb is associated with kidney damage in adults (Navas-Acien *et al.*, 2009). Infants, particularly those born prematurely, have reduced renal function and their developing kidneys are more susceptible to damage caused by excessive Cd and Pb in their diet. Al is a potent neurotoxin; and long-term feeding of Al-containing foods to infants caused impaired mental development at 18 months (Committee on Nutrition, 1996; Klein *et al.*, 2004). Also, excessive Al in the blood is selectively incorporated into the bones of infants, resulting in a weakened bone structure (Bernardo *et al.*, 2010). Al can only be removed from blood via the kidneys, and severe renal disorders can result in accumulation of Al in the blood, a situation potentially exacerbated by the action of higher levels of Cd and Pb on the kidneys. Thus, the monitoring of these elements is a high priority, internationally (Ikem *et al.*, 2002; Navarro-Blasco and Alvarez-Galindo, 2003; Ursinyova and Masanova, 2005; US FDA, 2008; Wojciechowska-Mazurek *et al*., 2008; Winiarska-Mieczan, 2009; Al Khalifa and Ahmad, 2010; Burrell and Exley, 2010).

In this sense, the determination of trace elements (Fe, Mn and Se) in food is of great importance since the deficiency or excess of metals could promote several clinical disorders resulting public health problems, as respiratory system cancer, skin disorder, anemia, depression of growth, impaired reproductive performance, heart failure and gastrointestinal disturbances, fatigue, decreased immunity and even death (Fraga, 2005; Yebra *et al*., 2008; Tokalioglu and Gurbuz, 2010).

Conclusion

Investigations revealed that extremely toxic heavy metals (Pb; Cd and Al) were detected above the toxicity levels. It is seemly necessary to warn about the hazardous effects of these toxic elements on both child and adult. The consumption of such milk powder is considered as additional source of exposure beside the direct sources of air, water and plants. Therefore, a strict regular monitoring of heavy metals contamination of milk and milk products is recommended in order to (i) establish the true contribution of milk and milk products to the dietary intake of heavy metals, (ii) recognize their possible health hazardous effects, their bioaccumulation during chronic exposure and their mobilization and secretion in milk.

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