Food Control 26 (2012) 6–14

Contents lists available at SciVerse ScienceDirect

Food Control

journal homepage: www.elsevier.com/locate/foodcont

Estimation of dietary intake and content of lead and cadmium in infant cereals marketed in Spain

Raquel Hernández-Martínez, Iñigo Navarro-Blasco*

University of Navarra, School of Sciences, Department of Chemistry and Soil Science, C/Irunlarrea 1, 31.080 Pamplona, Navarra, Spain

ARTICLE INFO

Article history:
Received 14 September 2011
Received in revised form
20 December 2011
Accepted 21 December 2011

Keywords:
Lead
Cadmium
Infant cereals
Dietary intake
Food analysis
Electrothermal atomic absorption spectrometry

ABSTRACT

Lead and cadmium have become highly toxic metallic elements. There is an obvious toxicological impact of these elements on infants since their intestinal absorption is significantly higher than in adults, thus it is desirable to quantify lead and cadmium levels in commonly consumed infant foods. Zeeman background correction, transversely-heated graphite furnace atomic absorption spectrometry, was used to determine both the lead and cadmium content of 91 Spanish infant cereals. Cereals were assessed in terms of different types, cereal predominant in formulation and whether it was obtained organically or conventionally. Preliminary analysis revealed a noticeably higher content of lead and cadmium (median, Q1–Q3) in organic cereals ($n = 17$, Pb: 26.07; 21.36–51.63; Cd: 18.52; 16.56–28.50 μg kg^{-1}) in relation to conventional ones ($n = 74$, Pb: 10.78; 6.43–19.33; Cd: 7.12; 4.40–11.77 μg kg^{-1}). Three formulations exceeded European lead maximum levels. Added ingredients (milk, cocoa, fruit and honey) to the cereal base provide lead enrichment. For cadmium, this pattern was observed by cereal based on cocoa, but also the raw materials contributed with a dilution phenomenon, decreasing the final cadmium concentration in infant cereal. Apart from several organically produced cereals, lead content showed a narrow variation, where gluten-free cereals provide lower cadmium content than formulations containing gluten. Dietary intakes of both elements were assessed in comparison with the reference intake values proposed by the EFSA Panel on Contaminants in the Food Chain. Organic infant cereals based on honey and cocoa supplied the highest risk intakes of lead and cadmium, respectively. In accordance with the actual state of knowledge on lead and cadmium toxicity and attending to the upper limits calculated from risk intake values set by EFSA, it seems prudent to call for a revision of both heavy metals content regulated by EC to set a maximum guideline values for infant cereal at 55 and 45 μg kg^{-1}, respectively.

1. Introduction

Residues of lead and cadmium are widely present in the environment as a result of anthropogenic processes. Lead is a metal largely used in industry, mainly in the manufacturing of pigments, coatings, containers, ointments, electric batteries and even liquor. Agricultural activities such as the excessive use of fertilisers and pesticides, irrigation with waste water, industrial applications (nickel–cadmium batteries, plastic stabilisers, dyes and pigments, coating of steel, various alloys) and urban life increase the content of cadmium in soils and waters. Therefore, clearly contributing to the contamination of raw materials used in the feeding industry (ATSDR, 2008; D'Mello, 2003; Reilly, 2002).

The evidence for the necessity of lead in animal and human nutrition is extremely limited (Anke, 1991; WHO/FAO/IAEA, 1996). However, the higher lead intestinal absorption (50%) of children in relation to adults (10%), and the retention rates of this potentially toxic element, 30% compared to 5%, is well-documented (González-Muñoz, Peña, & Meseguer, 2008; Reilly, 2002). Similarly, cadmium absorption and retention is also favoured in younger kids (Dabeka & Mckenzie, 1988).

Lead and cadmium exposure during childhood cause anaemia, abdominal pain, neurological and adverse developmental effects, learning disabilities, kidney damage, hypertension, and changes in vitamin D metabolism. In addition, the International Agency for Research on Cancer (IARC) has classified lead and cadmium as category 2A and 1 carcinogens, respectively (IARC, 1993, 2006).

The consumption of lead and cadmium-contaminated foods is the main source of human exposure to these elements (Capar, Mindak, & Cheng, 2007; González-Muñoz et al., 2008; Wilhelm, Wittsiepe, Schrey, Budde, & Idel, 2002). In view of their toxic effects on infants, the monitoring of chemical safety through lead

Abbreviations: TWI, tolerable weekly intake; IARC, International Agency for Research on Cancer; Q1, first quartile; Q3, third quartile.

* Corresponding author. Tel.: +34 948 425600; fax: +34 948 425740.
E-mail address: inavarro@unav.es (I. Navarro-Blasco).

0956-7135/$ – see front matter © 2012 Elsevier Ltd. All rights reserved.
doi:10.1016/j.foodcont.2011.12.024

and cadmium analysis of the most widely used infant foods, especially infant cereals, is of paramount importance. With this purpose different analytical methods such as atomic spectrometry, electroanalytical techniques, mass spectrometry or nuclear activation analysis could be used (WHO/FAO/IAEA, 1996). But electrothermal atomic absorption spectroscopy has been selected because of its high sensitivity and low detection limit, established as one of the most common methods to determine cadmium and lead in food after dry or wet ashing of organic matter (Ajtony, Bencs, Haraszi, Szigeti, & Szoboszlai, 2007; Correia, Oliveira, & Oliveira, 2000; Nazari, 2008).

In this sense, lead and cadmium levels in food products are controlled by Regulation 1881/2006 of the 2006 European Union Council, which set maximum levels in cereals of 0.2 and 0.1 mg kg^{-1} of lead and cadmium, respectively (EC, 2006a). Cereal products are the largest contributors to dietary exposure of both lead and cadmium in children and infants. The Joint Expert Committee on Food Additives (FAO/WHO) initially set the provisional tolerable weekly intake (PTWI) of 25 and 7 μg kg^{-1} body weight $week^{-1}$ for lead and cadmium, respectively (WHO, 2000, 2006). Although, subsequently the expert committee (WHO, 2010) concluded that the lead PTWI could no longer be considered health protective and withdrew it; also they considered that a monthly guidance value for cadmium was more appropriate, establishing a tolerable monthly intake of 25 μg kg^{-1} body weight $month^{-1}$. In this regard, the EFSA Panel on Contaminants in the Food Chain (CONTAM) has identified developmental neurotoxicity as the main potential adverse effect of lead in young children at a benchmark dose level ($BMDL_{01}$) of 0.5 μg kg^{-1} body weight day^{-1}. Moreover, the EFSA Panel confirmed the tolerable weekly intake (TWI) of cadmium at 2.5 μg kg^{-1} body $weightweek^{-1}$. Therefore, lead and cadmium adverse effects are unlikely to occur in the global population but the possibility of a risk to some subgroups of the population, such as infants and children up to age seven, cannot be excluded (EFSA, 2011).

Thus, taking into account all the above-mentioned aspects and because of the scarce amount of studies reported in the literature, the main aims of the present work were to determine the lead and cadmium concentration in most infant cereals marketed in Spain. Furthermore, to evaluate the levels of these contaminants in terms of different types, cereal predominant, organic or conventional production method, and to assess the dietary intake of both potentially toxic elements in comparison with reference values and tolerable weekly intake respectively, as proposed by the CONTAM Panel (EFSA, 2011).

2. Materials and methods

2.1. Instrumentation

Milestone Ethos Plus microwave with segmented rotor of high pressure (HPR-1000/10S) and temperature sensor (Milestone, Sorisole, Italy) was used for digestion of the infant cereal samples.

The analytical determination of lead and cadmium was carried out using an atomic absorption spectrophotometer Perkin Elmer A Analyst 800 (Norwalk, CT, USA) equipped with a transversely-heated graphite furnace atomiser, Zeeman background correction and an AS-800 autosampler. Single lead and cadmium hollow cathode lamps providing resonance lines of 283.3 and 228.8, operated at 10 and 4 mA, respectively with a slit width set at 0.7 nm. All measurements were carried out by integrated absorbance peak area using pyrolytically coated graphite tubes with end caps supplied by Perkin Elmer. The graphite furnace temperature programs are summarised in Table 1. Argon was used as the inert gas, the flow rate being 250 mL min^{-1} during all stages except atomisation, when the flow was stopped.

Table 1
Graphite furnace programmes for the determination of lead and cadmium in infant cereal samples.

Stage	Temperature (°C)		Ramp (s)		Hold time (s)		Gas flow rate (mL min^{-1})	
	Pb	Cd	Pb	Cd	Pb	Cd	Pb	Cd
1. Drying 1	110	130	10	15	30	45	250	250
2. Drying 2	130	–	20	–	30	–	250	–
3. Calcination	950	400	10	10	20	20	250	250
4. Atomisation	1350	1600	0	0	5	5	0	0
5. Cleaning	2400	2450	1	1	5	3	250	250
6. Cooling	20	–	–	–	–	–	250	250

2.2. Reagents and solutions

The highest purity reagents available were used throughout. Ultrapure deionised water type Milli-Q with a specific resistivity of 18 MΩ cm was used for the preparation of the standard and sample solutions. Concentrated 65% nitric acid (analytical grade, Merck, Barcelona, Spain) was additionally purified by sub-boiling distillation in a quartz sub-boiling still (Hans Kürner, Rossenheim, Germany). All materials were nitric acid-washed (10%, v/v) for at least 3 days and later thoroughly rinsed with ultrapure water before use.

The lead and cadmium stock standard solution (1000 mg L^{-1}) were both from Merck. The working standard solutions were prepared in sub-boiled nitric acid to match the acid concentration of digestion solutions by serial dilutions from the standard stock solution.

A mixed solution of magnesium nitrate–ammonium dihydrogenphosphate (0.6 g $Mg(NO_3)_2 \cdot 6H_2O$ suprapur, Merck and 1 g $NH_4H_2PO_4$ suprapur, Merck diluted in 100 ml ultrapure water) was used as matrix chemical modifier. 10 μL of the mixed modifier solution was injected prior to each determination.

The Certified Reference Materials NCS ZC 73008 Rice and NCS ZC 73009 Wheat (China National Analysis Center for Iron and Steel, Beijing, China) was used for validation of the methodology.

2.3. Infant cereal samples

Ninety-one samples of different infant cereals marketed in Spain from eight distinct conventional (Hero baby, Milupa, Nestlé, Nutriben, Nutricia, Ordesa, Puleva and Sandoz-Sanutri) and two organic (Biocrecimiento and El Granero Integral) manufacturers were analysed. The sampling was carried out from 2009 to 2010. During that time, these formulations were the most popularly consumed by neonates. Infant cereal samples were provided for free by the household or purchased in pharmacies and specialised organic feeding shops from Pamplona (Navarra, Spain).

The infant cereals studied were either organic ($n = 17$) or conventional ($n = 74$) cereals. Moreover, different types of infant cereals were classified according to the varieties used throughout the different stages of growth in a progressively diversified diet for infants aged from four months. These included: gluten-free based infant cereals ($n = 23$, a specialised formula which is normally based on rice and corn and specially designed for infants from 4 to 6 months), infant cereals with fruits ($n = 5$, a formula composed with- or without-gluten based cereals and dehydrated fruits added, designed for infants from 4 to 6 months of age), infant cereals with milk ($n = 15$, a formula containing follow-up infant formula which constitutes the principal liquid source of nourishment for infants from six months of age), infant multicereals (a product formulated to