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Analysis of lead and cadmium in cereal products and duplicate diets of a small group of selected Brisbane children for estimation of daily metal exposure

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ABSTRACT

Exposure to toxic metals such as lead (Pb) and cadmium (Cd) from foods is a concern for young children. The aims of the study were to analyse the levels of Pb and Cd in breakfast cereals, rice products and diets of selected children, and to estimate the daily intakes of Pb and Cd in these children. The samples (n = 82) of ready-to-eat breakfast cereals and rice products (n = 36) were collected and obtained from various markets in Brisbane, Australia. The samples for a duplicate diet study were collected for 3 consecutive days from normal healthy children (n = 15). The analysis was performed using ICP-MS after microwave digestion. The levels of Pb and Cd found in breakfast cereals and rice products ranged from < 0.01 to 0.25 mg/kg for Pb and < 0.01 to 0.11 mg/kg for Cd. The estimated daily intakes of Pb and Cd in children varied widely and ranged from 0.90 to 11.7 (5.6 ± 3.5 µg/day) for Pb and 0.98 to 9.5 (4.0 ± 2.2 µg/day) for Cd. The study shows low intakes of Pb and Cd in children.

1. Introduction

For the last few decades there has a rapid increase in industrial activities and this has led to significant increase of pollutants in the environment including heavy metal contaminants such as lead (Pb) and cadmium (Cd). Cd is widely used in manufacturing industry such electronics, pigments in plastics and Ni-Cd batteries; and Pb is used in soldering of various alloys, Pb batteries and colouring agents. These heavy metals will accumulate in the food chain and subsequently be taken up by humans through diet. The levels of Pb and Cd in food products could also increase as a result of vegetable and crop uptake of metals from soils [1,2].

A long term exposure to Pb and Cd is a major health concern particularly in young children for their growth and mental development. Cd is classified by the International Agency for Research on Cancer (IARC) as a human carcinogen (Group 1), and long-term exposure can cause increased cancer risk in lung, bladder and prostate [3,4]. High Cd exposure has been reported to cause itai-itai disease in Japan as a result of contamination of rice crop from the outflow of effluent of zinc mine containing Cd into rice field [5]. High Pb exposure can cause adverse health effect in humans and a long-term exposure in children can lead to low IQ, impaired neurobehavioral development and growth problems [6,7]. The IARC has classified Pb as potential carcinogenic substance for humans [8]. Since the elimination on the use of Pb petrol in cars and Pb paint in the 1970s, and regulation of drinking water standards around the world including Australia there has been a marked reduction of Pb exposure in the population [9].

The major route of exposure of Pb and Cd in humans includes diet, water and air dust. There have been many studies to assess the dietary intakes of Pb and Cd in a population including children. There are number of methods that are widely used for assessment of dietary intakes and these may include a total diet study, duplicate diet study, and diary record that may include food frequency questionnaire (FFQ). The selection of these methods will depend on the scale of the study, and in a wider population, a total diet study is used in many countries [10,11]. In this present study, a duplicate diet method was used as it has been shown to provide greater reflection on direct exposure of Cd and Pb from daily dietary intakes [6]. This was the first duplicate diet study conducted in Brisbane, Queensland for comparison with larger Australian total diet study. The levels of Pb and Cd in foods can vary significantly, and in this study the levels of Pb and Cd in selected core foods such as cereals were also investigated. The aims of the study were to analyse the levels of Pb and Cd in selected core food products such as breakfast cereals, rice products, and diets of selected children collected

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over three consecutive days for the assessment of the daily intakes of Pb and Cd in these children.

2. Materials and methods

2.1. Sample collection

The ready-to-eat breakfast cereals of various types (n = 82) and rice samples (n = 36) were obtained from various market outlets in Brisbane and Gold Coast in Queensland, Australia. These rice samples included Australian rice products and the imported rice from various countries. A duplicate dietary survey of food intakes by 15 normal healthy children with an average age of 7 years (4–12 y, 30% boys and 70% girls approximately) was conducted, in Brisbane, Australia. These children have not been exposed to Cd and Pb contamination from the environment. Informed consent was obtained from the parents and the ethical approval was obtained from the University of Queensland (Ref. No. 2006000409). The dietary food intakes and drinks were collected over 3 consecutive days for each subject for the study. Details of duplicate dietary study and food collection have been described elsewhere [12].

2.2. Sample preparation and analysis

The food samples were macerated and homogenized using a commercial food processor and a sub-sample was accurately weighed (0.25–0.5 g of dried sample and 2.0 g of liquid sample) into microwave digestion vessels. An aliquot of 4 mL of high-purity nitric acid (69% w/w) was added to each vessel and the digestion was heated using microwave digestion system (CEM MarsXpress, Mathews, NC). The digested solutions were diluted with ultra-pure water (Milli-Q Element System, Millipore, MA, USA), and analysis was performed by ICP-MS (7700 Agilent Technologies, Tokyo, Japan). Details of procedures for microwave digestion and ICP-MS analysis have been described elsewhere [13,14]. The certified reference materials (CRM) of rice flour (IRMM – 804), oyster tissue (NIST, SRM 1566b) and mussel tissue (NIST, SRM 2976) were used for quality control and assurance. These reference materials were treated similarly as the actual samples throughout the analysis.

2.3. Statistical analysis

The statistical analysis was preformed using the GraphPad Prism 5 (GraphPad Software, Inc., CA, USA) statistical package. The values for sample mean, median, 25% and 75% percentiles were calculated. For statistical analysis, the concentration values of less than the limit of detection at 0.01 mg/kg for Cd and Pb in these samples were assigned to zero values. The Mann-Whitney test of non-parametric statistics was used for analysis for significant difference of intakes for Cd and Pb between boys and girls.

3. Results

3.1. Quality control and assurance

The results of Cd and Pb found in this study from the standard reference materials were satisfactory and within the range of the reference values with the recoveries ranged from 88 to 100% (Table 1). This laboratory has, on regular basis, participated with the proficiency trial programs for quality control and assurance, organised by the Australian National Measurement Institute (NMI) for analysis of metals including Cd and Pb in food products. Our results for Cd and Pb were satisfactory and within the assigned consensus values at < 1.0 z-score [15]. The limit of quantification (LOQ) for Cd and Pb in this study was found to be at 0.01 mg/kg. The LOQ was estimated from ten times the standard deviation of the reagent blanks (n = 20) and by taking into account of typical sample weight and dilution.

Table 1
Recoveries (%) of cadmium and lead from standard reference materials (SRM).

	Concentration (mean ± s.d., mg/kg)	
Materials	Cadmium	Lead
Rice flour (CRM IRMM - 804)		
This study (n = 7)[a]	1.46 ± 0.023	0.37 ± 0.018
Refeference value	1.61 ± 0.07	0.42 ± 0.07
Recovery (%)[b]	*90.7*	*88.1*
SRM 2776 (NIST Mussel tissue)		
This study (n = 10)	0.83 ± 0.015	1.14 ± 0.049
Reference value	0.82 ± 0.16	1.19 ± 0.18
Recovery (%)	*101.2*	*95.8*
SRM 1566b (NIST Oyster tissue)		
This study (n = 10)	2.38 ± 0.043	0.284 ± 0.011
reference value	2.48 ± 0.08	0.308 ± 0.009
Recovery (%)	*96.0*	*92.2*

[a] N = number of replicates.
[b] Recovery is calculated as: (determined mean/reference mean) × 100.

3.2. Levels of Cd and Pb in cereal products

The levels of Cd in ready-to-eat breakfast cereals found in this study were generally low and varied widely. About 64% of all products had (n = 82) Cd below the LOQ level (0.01 mg/kg) (Table 2). With the exception of one sample (0.246 mg/kg Pb), low levels of Pb were found in these breakfast cereals. A wide variation of Pb levels was also found in cereals and about 29% of all products were below the LOQ level (0.01 mg/kg) (Table 2). Generally, low levels of Cd and Pb were also found in the Australian rice and imported rice from other countries sold in Australia (Table 2).

3.3. Cd and Pb intakes in children

A wide variation of Cd intakes (Fig. 1A) and one 5 year old girl had intake (9.5 μg/day) considerably higher than other children. A wide variation of Pb intakes was also found in these children, and 5 children had intakes considerably higher than the others (Fig. 1B). The mean daily intakes of Cd (4.0 μg/day) and Pb (5.6 μg/day) were generally low and comparable to other countries (Table 3).

4. Discussion

4.1. Levels of Cd and Pb in cereal products

The levels of Pb and Cd found in these breakfast cereals were generally comparable with reported values for cereal products [16–18]. Breakfast cereals are considered beneficial and about two-thirds are consumed by the Australian children [19,20]. It should be noted that some of the ready-to-eat breakfast cereals are not entirely made up of cereals but a mixture of plain or mixed flakes, puffed grains, processed grains, and fruit/flake mixtures with or without other ingredients. In this study, one sample of breakfast cereals was found to contain Pb at 0.25 mg/kg. This unexpected higher Pb level in this sample could be result of adventitious contamination during food processing prepared from various ingredients [21]. The Food Standards Australia New Zealand has set the maximum permitted levels of Cd in wheat and rice at 0.1 mg/kg, and Pb at 0.2 mg/kg for cereals, pulses and legumes [22]. High levels of Pb in wheat (0.22–0.24 mg/kg) and barley (0.57 mg/kg) have been reported in other countries [16,23]. Rice-based products have been reported to contain high Pb at 0.363 mg/kg, and this has caused concern in children and infants who would consume the products at greater amount [24]. Others have also reported that infant cereals with added honey can contain Pb as high as 1.027 mg/kg and