Title: 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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Short Title: Lead and cadmium in cereals and intakes in children

Highlights:

- The levels of lead and cadmium varied widely in ready-to-eat breakfast cereals
- Low levels of lead and cadmium were found in rice products
- Daily intakes of lead and cadmium were relatively low in selected children

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.

Keywords: Cadmium intake, lead intake, children, diets, cereal products, rice

Introduction

For the last few decades there has been 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 as 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 of 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 over three consecutive days for the assessment of the daily intakes of Pb and Cd in these children.

Materials and Methods

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].

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.

Statistical Analysis

The statistical analysis was performed 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.

Results

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.

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).

Cd and Pb intakes in children

A wide variation of Cd intakes (Fig. 1, A) 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. 1, B). 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).

Discussion

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 [16,17,18]. 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 suggested that Pb could come from other major ingredients and additives added to cereals [17].

Rice is considered a staple food for mostly Asian countries and in Australia rice is commonly used as additional cereal ingredients for food products such as rice-based breakfast cereals. Rice is also grown in some parts of Australia and its consumption has increased over the years. The levels of Cd and Pb in Australian rice found in this study were relative low, and most were below the LOQ level (0.01 mg/kg) (Table 2). The levels of Cd in rice in this study are comparable to levels reported in other studies [25]. High levels of Cd in rice have been reported for Bangladesh (1.13 mg/kg) and India (0.80 mg/kg), and because of higher rice consumption in these countries this would contribute considerably to Cd exposure [26].

It has been reported that levels of Pb in Australia rice can vary widely ranging from 0.016 to 1.25 mg/kg with the mean of 0.375 mg/kg [27]. The present study has also shown that low levels of Cd and Pb were found in imported rice products from other countries (Table 2). Others have reported that Pb levels in rice from India can vary widely and range from 0.002 to 0.637 mg/kg (mean, 0.267 mg/kg), and from Thailand it can range from 0.096 to 0.921 mg/kg (mean, 0.419 mg/kg) [27]. The high level of Pb in rice could be the result of

adventitious contamination of metals including Pb during processing and milling of rice. The high level of Pb in rice could be attributable to uptake of heavy metals by rice crop grown near the industrial area. In some parts of China, rice crops grown in near heavy industrial areas have been shown to cause a significant increase and wide variations of Pb levels in rice and can be as high as 2.75 mg/kg [25,28,29]. The elevated levels of Cd and Pb in rice products are a cause of concern in many countries, particularly in children because of their greater consumption of food in relation to their body size and rapid development. As a staple food, rice can contribute significantly to the intakes of heavy metals [30].

Cd and Pb intakes in children

There has been a concern that a consumption of foods such as cereal and rice products as staple foods can contribute significantly to the intakes of Cd and Pb if these foods are contaminated with these elements [30,31]. In Australia there have been little studies on the levels of Cd and Pb in cereal products and their contribution to Cd and Pb daily intakes in young children. In this study, the estimated daily intakes of Cd and Pb in children were conducted using a duplicate diet method and collection of food consumed over the 3 consecutive days. The mean daily intakes for Cd (4.0 µg/day) and Pb (5.6 µg/day) found in this study in children were generally low, and the daily intakes were varied widely ranging from 0.98 to 9.5 µg/day for Cd and 0.90 to 11.7 µg/day for Pb (Table 3). No significant differences were found for Cd intakes ($p=0.31$) and Pb intakes ($p=0.159$) between boys and girls when the non-parametric statistics of Mann-Whitney test was used. The values of median Cd daily intake and the 25% and 75% percentiles for children were calculated at 4.14 mg/kg, 2.41 mg/kg and 4.70 mg/kg, respectively. For Pb, the median intake and the 25% and 75% percentiles were calculated at 4.59 mg/kg, 2.72 mg/kg and 9.46 mg/kg, respectively.

The distribution of Cd and Pb daily intakes with the mean and median values for boys and girls is shown in Fig. 1.

The Cd intakes were lower than the reported values (mean range: 10.3-13.3 $\mu\text{g/day}$) by Callan and others [32] for children in Western Australia, but comparable with estimated values from the Australian total diet study (TDS) [33]. The estimated intakes from the Australian TDS were calculated and converted to daily intakes ($\mu\text{g/day}$) from the $\mu\text{g/kg bw/day}$, based on average body weight (bw) of 52 kg of 12 years old girls and 49 kg for 12 years old boys (Table 3). The Cd intakes in this study were also comparable to intakes from Germany but generally lower than other countries, and considerably lower than the reported high intakes for children in Thailand and Poland (Table 3). The EFSA Panel on Contaminants in the Food Chain (CONTAM) has maintained the tolerable weekly intake (TWI) of Cd at 2.5 $\mu\text{g/kg bw/week}$ (equivalent to 0.36 $\mu\text{g/kg bw/day}$) and Pb at 0.5 $\mu\text{g/kg bw/day}$ for young children which is a benchmark dose level (BMDL_{01}) with 1% extra risk [34, 35]. For comparison with the EFSA values the theoretical estimation of TWI for this study, calculated on the basis of mean intakes of Cd at 4 $\mu\text{g/day}$ (28 $\mu\text{g/week}$) and Pb at 5.6 $\mu\text{g/day}$, and using average body weight of 23 kg for average age of 7 years old, it can be estimated that TWI value for Cd would be at 1.2 $\mu\text{g/kg bw/week}$ (0.17 $\mu\text{g/kg bw/day}$) and Pb at 0.24 $\mu\text{g/kg bw/day}$. These theoretical estimated values are considerably lower than the EFSA values for Cd and Pb. It should be emphasised that the estimated daily intake values in this study are only as guideline for comparison with EFSA values for Cd and Pb daily exposure.

The Pb intakes in this study were comparable to intakes reported in other studies for Australia and for Germany and Japan (Table 3). High Pb intakes (41.0 $\mu\text{g/day}$) have been reported in Chinese children, and the major contributors of Pb come from rice and wheat flour [6,36].

Vegetables and crops including rice grown near the industrial areas have been reported to cause increase in Pb levels which can significantly contribute to Pb daily intakes [2,37]. Relatively high Pb intakes were reported for children in Poland (25.9 $\mu\text{g/day}$) and Spain (23.0 $\mu\text{g/day}$) (Table 3). The major contributors to Pb intakes for Polish and Spanish children come from cereals, vegetables and meat [31,38]. Because of ever increasing food variety available to infants and young children there is a concern that other food items such as candies have been reported to contain high Pb levels that contributed significantly to Pb intakes and subsequently caused elevated levels of blood Pb in children [39,40].

Generally, the low levels of exposure from Cd and Pb in diets of these children in comparison to other countries are possibly due to the food products produced in Australia are grown in regional rural areas, away from any industrial activities and emissions [41]. However, vegetables grown in the vicinity of Australian metal smelter contaminated sites have been shown to accumulate heavy metals including Cd and Pb [42].

Conclusions

Wide variations of Cd and Pb levels in cereal products were found in this study, and the levels were generally low, particularly for breakfast cereals. However, because breakfast cereals are consumed more by young children there is still a concern that major food components in breakfast cereals such as cereal types, food additives and ingredients could have a potential source of metal contaminants including Cd and Pb. Because of changing dietary patterns and increasing food varieties available for infants and young children, there is a need to regularly monitor the levels of Cd and Pb in these food products. The study also found the daily intakes of Cd and Pb in children were generally low and comparable to other Australian studies. Even though the intakes of Cd and Pb were relatively low in children in

this study, given their accumulative nature, it is important that the long-term exposure of these elements be kept to minimal. The known deleterious effects of these metals on bioavailability of essential dietary minerals need also be considered because of young children's unique sensitivity and vulnerability during growth and development.

Acknowledgement

The authors would like to thank H. Yates, R. Sumner and X. Yang for their assistance with sample preparation and analysis. This research did not receive any specific grant from funding agencies in the public, commercial, or not-to-profit sectors.

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(A)

(B)

Fig. 1. The distribution of intakes with mean and median values ($\mu\text{g/day}$) for Cd (A) and Pb (B) of 15 children at different age and gender estimated from duplicate diet study.

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

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

^aN=number of replicates^bRecovery is calculated as: (determined mean/reference mean) x 100

Table 2 Cadmium and lead levels (mg/kg) in breakfast cereal products and rice

Breakfast cereal products	Cadmium		Lead	
	Concentration (range)	Median (25%, 75% percentile)	Concentration (range)	Median (25%, 75% percentile)
Corn-based cereal (n=15)	<0.01-0.029	<0.01 (<0.01, 0.021)	<0.01-0.17	0.031 (0.014, 0.065)
Mixed grain cereal (n=24)	<0.01-0.048	<0.01 (<0.01, 0.021)	<0.01-0.15	0.024 (<0.01, 0.069)
Rice-based cereal (n=9)	<0.01-0.021	<0.01 (<0.01, 0.01)	<0.01-0.095	<0.01 (<0.01, 0.058)
Wheat-based cereal (n=34)	<0.01-0.107	<0.01 (<0.01, 0.107)	<0.01-0.246	0.031 (0.014, 0.066)
Rice (Australian rice & other countries)				
Australia (n=10)	<0.01-0.015	<0.01 (<0.01, <0.01)	<0.01-0.024	<0.01 (<0.01, <0.01)
India (n=4)	0.012-0.050	0.022 (0.013, 0.045)	<0.01-0.014	0.012 (<0.01, 0.034)
Pakistan (n=5)	<0.01-0.044	0.017 (0.01, 0.036)	<0.01-0.025	<0.01 (<0.01, 0.018)
Thailand (n=8)	<0.01-0.018	<0.01 (<0.01, 0.014)	<0.01-0.021	0.014 (<0.01, 0.019)
USA (n=4)	<0.01	<0.01 (<0.01, <0.01)	<0.01-0.018	0.01 (<0.01, 0.016)

Table 3 The daily intakes of cadmium and lead in children by country

Country	Age group (years)	Intake (µg/day)		Reference
		Cadmium	Lead	
Australia				
(Brisbane)	4 - 12 (both sexes)	4.0 ± 2.2 (0.98-9.5)	5.6 ± 3.5 (0.90-11.7)	This study
Australia (NSW)	6 - 11 (both sexes)	-	6.44	[43]
Australia (WA)	8-13 (boys)	10.34-14.3 ^a	4.75-6.10 ^a	[32]
	8-13 (girls)	12.36-14.2 ^a	5.01-6.18 ^a	
Australia				
(national)	12 (boys)	5.4 -14.2 ^b	0.98-21.1 ^b	[33]
	12 (girls)	4.7-11.4 ^b	0.52-29 ^b	

Brazil	7 - 14 (both sexes)	0.9	0.6	[44]
Canada	1 - 4 (both sexes)	8	15	[45]
	5 - 11 (both sexes)	12	20	
China	1.9 - 7 (both sexes)	4.1 ± 8.9 ^c	41.0 ± 31.7 ^c	[6]
China	2 - 6 (both sexes)	9.8	-	[46]
France	3 - 14 (both sexes)	2.04	12.8	[47]
Germany	1.5 - 5.3 (both sexes)	6.3 ± 2.9 (1.6-14)	5.8 ± 5.1 (0.58-29)	[48]
India	6 - 10 (both sexes)	2.1 ± 2.29	16.5 ± 1.7	[49]
		13.2 ± 5.2 (4.19-		
Japan	3 - 6 (both sexes)	22.4)	5.34 (1.22-9.25)	[50]
Japan	3 - 6 (girls)	11.6	2.4	[51]
	3 -6 (boys)	12.0	2.1	

Korea	0 - 6 (both sexes)	9.0 ± 4.7 ^d	11.1 ± 3.3 ^d	[52]
Korea	4 - 10 (both sexes)	11.2	8.2	[53]
The Netherlands	4 - 7 (boys)	8 ± 3 ^e	11 ± 9 ^e	[54]
	4 - 7 (girls)	7 ± 3	12 ± 11	
	7 - 10 (boys)	10 ± 3	16 ± 12	
	7 - 10 (girls)	9 ± 3	14 ± 14	
Poland	7 - 13 (both sexes)	32.5 ^f	25.9	[38]
Spain	4 - 9 (both sexes)	13.17	23.04	[31]
	9 mon - 2 (both			
Thailand	sexes)	4.18-40.4	-	[55]

^aMean range values

^{b,c,d,f}Values were calculated and converted from µg/kg bw/day to µg/day by taking into account of average body weight (bw) of the age group

^eGeometric mean values