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Arsenic and cadmium concentrations in legumes and cereals grown in the North Central Province, Sri Lanka and assessment of their health risk

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Abstract

Background: A total of 181 samples of cereals and legumes, including mustard, maize, finger millet, sesame, cowpea/black-eyed pea, Urad dal /split bean, foxtail millet, long bean and green gram from the North Central Province of Sri Lanka were analysed for arsenic (As) and cadmium (Cd) levels using atomic absorption spectrophotometry (AAS).

Findings: The As and Cd level in each sample was not significantly different. The mean level was found to be 48.90 ± 34.63 μg/kg for As and 19.39 ± 9.508 μg/kg for Cd on weight basis.

Conclusion: None of the foodstuffs studied contained As and Cd in levels exceeding the maximum permissible levels currently in force. Human health risk assessment of As and Cd in the foodstuffs was conducted and computed values indicated that there is no health risk due to consumption of the varieties studied. Nevertheless, the values, especially Total Hazard Quotient (THQ) emphasize the requirement of a comprehensive total diet study.

Keywords: Cereals, Legumes, Cadmium, Arsenic, Non-carcinogenic health risk, Carcinogenic effects, Dietary exposure, THQ

Introduction

The Agency for Toxic Substances and Disease Registry (ATSDR), 2017 ranked arsenic (As) and cadmium (Cd) at number one and seven, respectively, in the hazardous substances list (ATSDR, 2017). Most As and Cd exposure of humans happens directly or indirectly by ingestion - through drinking water or contaminated food. As and Cd contaminated plant crops are mostly those that grow in contaminated soil (Zhao et al., 2010). After accumulation of these non-essential trace elements (NETEs) into the crops, transfer occurs via the food chain and finally reaches the apex consumer in the food chain such as humans. In the mid 1950s in Japan, the “Itai-Itai disease” was recorded after consumption of Cd contaminated soybean and rice (Huang et al., 2009). Consumption of Cd resulted in stomach irritation, vomiting and diarrhea, while long-term exposure caused kidney disease, cancer and fragility at birth. Arsenic exposure may cause skin lesions, neuropathy, gastrointestinal diseases, cardiovascular diseases, cancer, and other ailments (Corguinha et al., 2015).

Air, water, and soil contamination are the sources of NETEs in agricultural areas. Agricultural land inputs such as fertilizers and pesticides are responsible for high NETE contamination of soil (Gunatilake et al., 2014). The NETEs in soil are transferred through the agricultural crops as they are deposited in the edible parts. The transfer rate depends on many different factors such as climatic, plant species, and genotypes (Corguinha et al., 2015).

North Central Province is the largest province in Sri Lanka, and the majority of the people (65%) belong to the agricultural sector. Rice, the main crop of this area is cultivated by using the highly sophisticated ancient irrigation system from the Mahaweli River (Gunatilake et al., 2014).

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Apart from rice, other cereals, fruits, and vegetables are grown using seasonal tank water, rainwater and deep well water. The region is also associated with livestock farming.

Chronic renal failure (CRF) or chronic kidney disease (CKDu) of unknown etiology is the biggest problem among the farmers in North Central Province (Bandara et al., 2010). Scientific research has found a number of causes for CKDu, of which one was As and Cd contamination (Bandarage, 2013). Jayasumana et al. (2015) reported considerable amount (20.6–540.4 μg/Kg) of arsenic in rice, which is depend on the fertilizers and pesticides used during the cultivation.

Exposure of humans to contaminated food is a general health concern in Sri Lanka. In view of this, the study assessed the As and Cd levels in the edible part of nine foodstuffs including legumes and cereals grown in the North Central Province of Sri Lanka. To the best of our knowledge, this is the first paper dealing with NETEs in cereals in this area, which is relevant not only to food security but also to commercial and economic aspects.

Methods

Sample collection

A total of 181 samples of nine crop species were randomly collected from farmers from the North Central Province of Sri Lanka during two growing cycles of the year 2016. The basic information such as sampling location, common name, variety etc. was collected at same time. The samples were prepared as per general procedures followed by consumers. The inedible parts were first removed, the edible portion was then washed thrice in distilled water, dried at room temperature, packed in polythene bags, sealed and transported to the laboratory for further analysis. While people in the area use groundwater or tap water to prepare the cereals in the kitchen, in the study we used distilled water to avoid any contamination from water. The type and number of samples were as follow; mustard (10), maize (45), finger millet (50), sesame (26), cowpea/black-eyed pea (17), Urad dal/ split bean (11), foxtail millet/ Thanahal (7), long bean/Mea (5) and green gram (10).

Sample preparation and analysis

Samples were ground using a domestic food blender and sieved. Then 0.5 g of each sample was mineralized in Teflon digestion vessels in a closed microwave accelerated system (CEM, MARS 6, USA) using nitric acid (Sigma, USA) as a reagent. Analysis of As and Cd was carried out by atomic absorption spectrophotometry, AAS (Varian 240 FS, Victoria, Australia) assisted by graphite tube atomizer (GTA 120). The calibration curve range (μg/L), wavelength (nm), slit width (nm) and lamp current (mA) were as follow; for Cd, 0.5–3.0, 228.8, 0.5, 4 and for As, 5.0–50.0, 193.7, 0.5, 10 respectively.

In order to check the purity of chemicals and glassware, blank samples were run with each batch. Analytical quality control was achieved by using the Certified Reference Materials (CRM) IRMM 804 (rice flour) from the Institute for Reference Materials and Measurement (IRMM), Belgium. All reagent blanks, samples, and CRM were run in duplicate. Validation of the analytical method was performed using limit of detection, LOD (3 x standard deviation, SD) and limit of quantification, LOQ (10 SD), correlation coefficient of calibration curve (0.999 or better), relative standard deviation, RSD between duplicates (10% or below) and recovery of CRM (80–120%).

The data were analysed using Microsoft Excel (MS office, 2010) and SPSS (Version 17) software. The mean, median, minimum-maximum value and standard deviation were calculated for As and Cd in each cereal/ legume type. The mean comparison ANOVA test was applied to find the significant difference between species and metals (p-value < 0.05). All samples were considered for the statistical analysis and when the sample reading was below the LOQ values, half of LOD values were considered as a sample reading.

Health risk assessment

The health risk assessment based on the legumes and cereals were conducted according to the Integrated Risk Information System (IRIS) of the US Environmental Protection Agency (US EPA) guidance (IRIS, 2018).

Provisional Tolerable Weekly Intake (PTWI) of As and Cd through consumption of cereals and legumes was calculated using the following equation (Popovic et al., 2018).

$$\mathrm{PTWI} = \frac{C \times WC}{BW} \tag{1}$$

C = concentration of metals in legumes and cereals (mg/kg).

WC = average per capita cereal and legume consumption per week (kg/week).

BW = average body weight (kg).

The exposure rate of Cd and As through the legumes and cereals was calculated using the following equation (Hensawang and Chanpiwat, 2017)

$$\text{Exposure rate (ER)} = \frac{C \times IR \times ED}{BW \times AT} \tag{2}$$

C = concentration of metals in legumes and cereals (mg/kg).

IR = amount of legumes and cereal consumed (kg/year).

ED = period of time in contact with the metal (year).

BW = average body weight (kg).

AT = average time or period of exposure (day).

Non-carcinogenic health risk was calculated based on the Hazard Quotient (HQ) value using the equation is below (Hensawang and Chanpiwat, 2017).

$$\mathrm{HQ} = \frac{ER}{RfD} \qquad (3)$$

RfD = reference dose for Cd and As.

The carcinogenic risk was also calculated based on the following equation (Hensawang and Chanpiwat, 2017).

$$\mathrm{AELCR} = \frac{Exposure\ rate \times SF}{DL \times 365} \qquad (4)$$

AELCR = annual excess lifetime cancer risk.

SF = cancer slope factor (mg/kg, day).

DL = average human longevity.

Due to non-availability of the per capita consumption data, we assume that our population consume World Health Organization (WHO) recommended amounts for pulses, nuts, and seeds groups, viz., 30 g/day at least as a part of the 400 g of fruit and vegetables (Nishida et al., 2004). The mean body weight of Sri Lankan male is 61.4 kg and the female is 54.6 kg (MHNIM, 2015). The average Sri Lankan life expectancy is 75 years (CBSL, 2017). The RfD values for As (inorganic) and Cd are 3×10^{-4} and 1×10^{-3} mg/kg.day respectively and SF for As is 1.5 mg/kg.day (Hensawang and Chanpiwat, 2017).

Results and discussion

Method performance

The microwave assisted digestion, atomic absorption spectroscopy method used in this study shows the higher accuracy of different types of legume and cereal matrices. The mean As and Cd concentration of the standard reference material ($n = 10$) was for Cd: 1.875 mg/kg (recovery 116%) and for As: 0.056 mg/kg (recovery 114%). The LOQ value for Cd and As were 0.006 mg/kg and 18.17 μg/kg respectively.

Cadmium level in legumes and cereals

The concentration (μg/kg, wet weight basis) of Cd in the foods studied are presented in Table 1 as mean, median and range. There was no significant difference in Cd levels between legumes and cereals ($p < 0.05$). The highest mean (37.52 μg/kg) and median (27.68 μg/kg) Cd level was in sesame (cereal) while the highest range was observed in maize (cereal), <LOQ-122.2 μg/kg. Apart from cowpea/black-eyed pea, all the samples had Cd levels of more than 10 μg/kg. Cd levels have been reported in maize at < 0.01 mg/kg (dry weight) in Nigeria (Akinyele and Shokunbi, 2015), 0.07 mg/kg (dry weight) also in also Nigeria (Onianwa et al., 2000) and 0.007 mg/kg (dry weight) in Canada (Dabeka and McKenzie, 1995). Cadmium concentration in the sample of Spanish legume (lentils, bean, chickpea, and green peas) was not detected 0.018 mg/kg (Cabrera et al., 2003). This shows that the Cd concentrations obtained in this study are within the range reported in the literature. Commercially used phosphate fertilizers have a high levels of Cd and are extensively used in rice cultivation and are possible to add into the irrigation system (Diyabalanage et al., 2016), and this may be the reason for the record values of Cd in legumes and cereals from the North Central Province.

Arsenic level in legumes and cereals

Generally As is detected in all foods. The level is higher in fish, shellfish, and seaweed than cereals and vegetables (Hernández and Navarro, 2013). The total As (tAs) in legumes and cereals detected in this study are presented in Table 1. The recorded mean tAs level in all varieties was below 0.1 mg/kg, except in the case of sesame. Corguinha et al. (2015), reported 19, 47 and 65 μg/kg (dry weight) of tAs in wheat, corn, and soybean respectively. Total arsenic exposures in the United States in between 2006 and 2008 were estimated for 16 population subgroups and ranged from 0.14 to 0.45 μg/kg/day (Jara and Winter, 2014). Liu et al. (2009), found a mean tAs of 0.11 ± 0.02, 0.14 ± 0.14 and 0.16 ± 0.046 mg/kg

Table 1 Concentrations (μg/kg, wet weight) of As and Cd in legumes and cereals

Species	Cd (μg/kg)			As (μg/kg)		
	Mean	Median	Range	Mean	Median	Range
Mustard	13.67	7.220	<LOQ-42.52	<LOQ	<LOQ	<LOQ-61.84
Maize	10.37	4.882	<LOQ-122.2	90.94	<LOQ	<LOQ-475.7
Finger millet	22.38	11.11	<LOQ-120.5	52.67	<LOQ	<LOQ-602.5
Sesame	37.52	27.68	<LOQ-121.5	105.2	<LOQ	<LOQ-519.7
Cowpea	7.930	3.036	<LOQ-34.61	60.25	<LOQ	<LOQ-502.5
Urad dal	21.21	14.16	<LOQ-39.34	47.95	<LOQ	<LOQ-198.5
Foxtail millet	22.34	22.57	<LOQ-49.99	29.40	<LOQ	<LOQ-127.1
Long bean	11.53	3.316	<LOQ-44.17	<LOQ	<LOQ	<LOQ
Green gram	27.53	16.85	<LOQ-117.4	46.69	<LOQ	<LOQ-348.0

Table 2 PTWI of As and Cd considering the recommended legumes and cereal per capita consumption (30 g/day)

Metal	PTWI (expressed as % of PTWI or TWI)		Established PTWI	Established TWI
	Male	Female		
As	1.1	1.3	15	–
Cd (PTWI)	0.9	1.1	7	–
Cd (TWI)	2.7	3.0	–	2.5

(dry weight) in wheat grown in three areas in China. Škrbić and Onjia (2007), evaluated the content of different microelements in wheat grain cultivated in 14 regions of Serbia and found a range from < 50–162 (mean 82) μg/kg (dry weight). These results were comparable to the present results in cereal and legume species grown in Sri Lanka.

Health risk assessment in consumption of legumes and cereal

There is no As threshold level proposed by the World Health Organization (WHO) Codex Alimentarius Commission or the European Union (EU) for cereal and legumes. In generally tAs is analysed for dietary estimation, although it is the inorganic As (iAs) that shows major toxicity signs (Hensawang and Chanpiwat, 2017). The EU regulation number 2015/1006 establishes that the iAs level for rice should not exceed 200 μg/kg (non-parboiled milled polished or white rice) and 250 μg/kg (parboiled rice and husked rice) (EU/EC, 2015). However, we were unable to perform As speciation analysis due to non-availability of the facilities and lack of funds.

The Joint Expert Committee on Food Additives (JECFA) proposed 400 μg/kg wet weight as a threshold level of Cd in rice while the European Food Safety Authority (EFSA) proposed 200 μg/kg for rice (Corguinha et al., 2015). Applying the Cd regulated limits for the legumes and cereals studied; no samples exceeded the JECFA level or even the EFSA level.

The study carried out a health risk assessment with the derived values by comparing with the available toxicological guidelines. JECFA established the PTWI values while EFSA established the TWI values. For As, the PTWI value is 15 μg/kg.bw. For Cd, the PTWI value is 7 μg/kg.bw while the TWI is 2.5 μg/kg.bw. Legumes and cereals do not significantly contribute to the PTWI or TWI value (Table 2). Legume and cereal consumption represent less than 1% of PTWI and less than 3% TWI for Cd while it is less than 1.3% of the PTWI for As.

The computed non-carcinogenic health risk of As and Cd indicates that there is a slight risk from eating legumes and cereals (Table 3). According to earlier studies, if the Total Hazard Quotient (THQ) ≥1, there is a potential health risk and it is necessary to intervene and take protective action (Gladyshev et al., 2009). The THQ calculated as a sum of HQ for each element was 0.6238 for males and 0.7015 for females. The dominating metal that could make the greater contribution to a higher THQ was As. The carcinogenic health risk from As in cereals and legumes is well below 1×10^{-6} (Table 3) which is considered as the lowest acceptable level of risk (Popovic et al., 2018).

It is important to emphasize that this value may be an erroneous estimation as it was calculated based on the WHO recommended per capita legume and cereal consumption. The real consumption value may be less than this. Also, we assumed that 100% of the ingested As and Cd were bioavailable and did not consider the potential reduction in amounts while preparing and cooking. However, in the worst-case scenario, this result could be appropriate and thus highlights the requirement of total diet study from the toxicological viewpoint.

Conclusion

This study examined the toxicological risk due to two non-essential trace elements (As and Cd) in nine different commonly consumed legumes and cereals in Sri Lanka. The results showed no significant difference in As and Cd levels among the samples studied. Arsenic was the most abundant non-essential trace element and it was the major contributor to non-carcinogenic health risk. The calculated PTWI, ER, HQ and AELCR values are below the published international regulations. However, it is necessary to note the contribution of THQ value through consumption of legumes and cereals. Some assumptions of this study may overestimate the risk and hence a comprehensive Total Diet Study (TDS) based on the toxicological endpoint should be carried out.

Table 3 ER, HQ and AELCR values in As and Cd through consumption of legumes and cereals

Metal	ER		HQ		AELCR	
	Males	Females	Males	Females	Males	Females
As	1.67×10^{-4}	1.88×10^{-4}	0.558	0.627	9.16×10^{-9}	1.03×10^{-8}
Cd	6.63×10^{-5}	7.46×10^{-5}	0.066	0.075	–	–

Abbreviations

AAS: Atomic Absorption Spectrophotometer; ACL: Analytical Chemistry Laboratory; AELCR: Annual excess lifetime cancer risk; As: Arsenic; ATSDR: Agency for Toxic Substances and Disease Registry; Cd: Cadmium; CKDu: Chronic kidney disease of unknown etiology; CRM: Certified Reference Materials; EFSA: European Food Safety Authority; GTA: Graphite tube atomizer; iAs: Inorganic; IRIS: Integrated Risk Information System; IRMM: Institute for Reference Materials and Measurement; JECFA: Joint Expert Committee on Food Additives; LOD: Limit of detection; LOQ: Limit of quantification; NARA: National Aquatic Resources Research and Development Agency; NETEs: Non-essential trace elements; PTWI: Provisional Tolerable Weekly Intake; tAs: Total Arsenic; TDS: Total Diet Study; THQ: Total Hazard Quotient; US EPA: US Environmental Protection Agency; WHO: World Health Organization.

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Availability of data and materials

Please contact corresponding author for data requests.

Authors' contributions

All authors contributed to the analysis, writing and improvement of the manuscript. All authors read and approved the final manuscript.

Competing interests

The authors declare that they have no competing interests.

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