Cadmium and arsenic concentrations in Sri Lankan rice and their potential health risks


Highlights

- 186 newly improved and traditional rice samples were collected from the North-Central province in Sri Lanka.
- All rice grains, Cd concentrations were less than the JECFA standard values.
- HQ and CR values indicated that the Sri Lankan population, especially children, are at risk of potential non-carcinogenic and carcinogenic health effects.
- Comprehensive risk assessment is required based on the bioavailability/bioaccessibility and speciation exposure.
Cadmium and arsenic concentrations in Sri Lankan rice and their potential health risks

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Received: 09/08/2019; Accepted: 15/07/2020

Abstract: Arsenic (As) and cadmium (Cd) concentrations have been analysed in locally grown rice grains from North Central Province, Sri Lanka. Totally, 186 samples including newly improved and traditional varieties were collected during September-December 2016, and As and Cd concentrations were determined by electrothermal atomic absorption spectroscopy. The potential health risks were estimated by three indices namely Estimated Daily Intake, Target Hazard Quotient, and Cancer Risk. The values of As and Cd varied from 18.17-575.94 and 6.00-261.786 µg/kg, w/w respectively. The non-carcinogen risk, Target Hazard Quotient>1 in children was higher than the threshold value. The cancer risk of inorganic arsenic was also higher than the acceptable range for both children and adults, but children are more exposed to both elements than adults.

Keywords: ET-AAS; inorganic arsenic; cadmium; North-Central Province; rice cultivation.

INTRODUCTION

In Sri Lanka, rice is the most important staple food item, accounting for 39 % of total dietary energy supply. The consumption of contaminated food is a major pathway to exposure of contaminants like cadmium (Cd) and arsenic (As) for humans (Hensawang and Chanpiwat, 2017). According to data from the World Bank (WB) and the Food and Agriculture Organization (FAO), Sri Lanka is among the top 10 countries with the highest per capita rice consumption at 105 kg/year or about 300 g/day (Hu et al., 2016). Meharg et al., (2013) studied the variation of Cd in rice and human exposure in 12 countries and found that Sri Lankan populations were exposed to high weekly Cd intake from rice, which is close to or exceeds the Provisional Tolerable Weekly Intake (PTWI) value of 2.5 µg/kg published by the Joint Expert Committee for Food Additives (JECFA) of the FAO and World Health Organization (WHO) (EFSA, 2011). Though JECFA published the PTWI for As as 15 µg/kg, the European Food Safety Authority (EFSA) has stated that this value is no longer appropriate because JECFA had not reflected the presence of inorganic arsenic (iAs) which causes cancer, mainly in the lung and urinary bladder (EFSA, 2009). Although rice is rich in nutrients such as carbohydrate, vitamins, minerals etc, it is also a major exposure route of iAs. (Mondal et al., 2019). The studies showed elevated As levels in rice where the rice paddy fields are irrigated with contaminated water (Meharg et al., 2008). That means the chemical speciation of As is more important when explaining the health-related issues than total As level. Rice contains mainly iAs (As-III and As-V) and organic As (dimethyl arsinic acid and monomethyl arsenic acid) (Tenni et al., 2017). For monitoring purposes, European Union (EU/EC 2015) published the regulation for iAs as 0.20 mg/kg for non-parboiled milled rice (i.e. polished or white rice), 0.25 mg/kg for parboiled rice and husked rice, 0.30 mg/kg for rice waffles, rice wafers, rice crackers and rice cakes and 0.10 mg/kg for rice destined for the production of food for infants and young children (EU/EC, 2015).

The “Itai-Itai” disease in Japan during the 1950’s, resulting from the continued intake of Cd-contaminated rice, has provoked worldwide attention (Huang et al., 2009). Rice was seen to accumulate Cd and As more profoundly than other elements. The main reasons for this phenomenon were based on the genotype of rice, soil type, root system and silicon content (Song et al., 2015). There are many options and practices to manage the accumulation of As and Cd in rice, including soil amendments, as well as irrigation and fertilization management during growth (Chen et al., 2017). However, this task is difficult, because As and Cd concentrations correlate negatively. In Sri Lanka, during the last few decades, the soil has been contaminated by Cd and As due to the use of agrochemicals such as weedicides, fertilizers, etc. (Gunatilake et al., 2014; Bandara et al., 2010).

The accelerated Mahaweli development program and irrigation supplied the water for most of the paddy rice cultivation in the North Central Province (NCP), where the highest chronic kidney disease was reported (Weeraratne and Wimalawansa, 2015). Although some studies of As and Cd in Sri Lankan rice have been published (Meharg et al., 2013; Kariyawasam et al., 2016; Herath et al., 2014), they are often limited in the geographic locations. The objective of the present study was to examine the As and Cd concentrations in improved (IRV) and traditional (TV) rice varieties, available in different locations in NCP, Sri Lanka, and to estimate the dietary intakes and carry out a health risk assessment of Cd and As exposure through the consumption of rice.

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MATERIALS AND METHODS

Sample collection and metal analysis

Rice samples (n=186), including 44 traditional samples, were collected from the paddy fields in 13 different locations of Anuradhapura and Polonnaruwa districts of NCP, Sri Lanka during the year 2016 (Figure 1). Sampling location, rice variety, irrigation water resource, and the farming system (organic or not) were recorded. Raw paddy was dehulled and ground using a domestic blender and processed for analysis. All glassware used was cleaned and dried properly. All the standards and reagents were prepared using ultrapure water (Barnsted, Easy pure LF system, Dubuque, USA). All the chemicals used were analytical reagent grade or better (Sigma Aldrich, USA). The working stock solutions to calibrate for Cd (3 µg/L) and As (50 µg/L) were prepared by appropriate dilution of stock standard solution of Cd and As (1,000 mg/L from Fluka, Switzerland) and the calibration curve plotted the automatic dilution of working stock solution within the range of 0-3 µg/L for Cd and 0-50 µg/L for As.

A portion of 0.5-1.0 g of rice flour was taken in a Teflon tube and 10 mL nitric acid was added. After 15 mins of free digestion, the tubes were placed in a microwave accelerated system (CEM, Mars 6, Matthews, USA) and the digested samples were used to prepare 50.0 mL aqueous solutions. Simultaneously, Certified Reference Materials (CRM), from the Institute for Reference Materials and Measurements, Belgium (IRMM 804, rice flour) was also run in the same manner and all the experiments were carried out in duplicate.

An Atomic Absorption Spectrophotometer (AAS) (Varian 240 FS, Varian Inc., Mulgrave, Victoria, Australia), supported with a Graphite Tube Atomizer (Varian, GTA-120) and PSD-120 autosampler was used to determine the Cd and As levels (As analysis with nickel (Ni); as a modifier). Cd and As were determined at 228.8 nm and 193.7 nm, with 4 mA and 10 mA of lamp current and 0.5 mm slit width respectively. The statistical analysis was performed using SPSS-20 software, including mean comparison and one-way ANOVA. All samples were considered for statistical analysis. When the sample reading was below Limit of Quantification (LOQ), half of the LOQ value was used for calculation.

Method performance

The accuracy of Cd and As analyses was verified using CRM. The certified values of Cd and As in CRM were 1.61 mg/kg and 0.049 mg/kg respectively. Comparing the certified value, the recovery was 114 % for As and 116 % Cd, which fitted within the 95 % confidence interval. The Limit of Detection (LOD) was established through the 3ϭ criterion (ϭ is the standard deviation of 11 measurements of blank), while the LOQ was established through the 10 ϖ criterion. The LOQ of As and Cd were 18.17 and 6.00 µg/kg, w/w respectively. Further, the uncertainty (k=2) of the As and Cd were ±11.0 % and ±8.8 % respectively and Relative Standard Deviation (RSD) was maintained below 15 %. The analytical performance was acceptable for the measurement of heavy metals in the rice grains.

Health risk assessment

The health risk assessment in this study was based on the Integrated Risk Information System (IRIS) of the United States Environmental Protection Agency (USEPA). Arsenic speciation was not performed in this study. However, from the toxicological point of view iAs is important and the
amount of iAs was around 80-90% of the tAs (Al-Saleh and Abduljabbar, 2017). For the health risk assessment, we assumed that 80% of the tAs in our samples were iAs.

The Estimated Daily Intake (EDI) of tAs, iAs, and Cd from rice was calculated according to the following equation (Ma et al., 2017),

$$\text{EDI} = \frac{C \times DI}{BW}$$  \hspace{1cm} (1)

Where, EDI is Estimated Daily Intake of metals (µg/day, kg BW), C is metal concentration of the rice (µg/kg), DI is Daily Intake of rice (0.3 kg/day), and BW is average Body Weight of Sri Lankan adults (60 kg) and school-age children (35 kg).

The non-carcinogenic risk to the population with long term intake of rice was estimated as an index of Target Hazard Quotient (THQ), which was calculated from the following equation (Ma et al., 2017).

$$\text{THQ} = \frac{\text{EDI}}{RfD} \times 10^{-5}$$  \hspace{1cm} (2)

Where RfD (mg/BW kg.day) is an oral Reference Dose for the element, regulated by USEPA. It is $1 \times 10^{-3}$ for Cd and $1 \times 10^{-4}$ for iAs.

Of the two metals studied, USEPA did not quantitatively estimate the oral exposure carcinogenic slope factor for Cd. Hence, Cancer Risk (CR) associated with rice consumption was calculated only for As by using the following equation (Ma et al., 2017).

$$\text{CR} = \text{EDI} \times \text{SF}$$  \hspace{1cm} (3)

Where, SF (BW.kg.day/µg) is the cancer Slope Factor set by USEPA (for iAs 1.5 mg/kg.day).

RESULTS AND DISCUSSION

Geographical variation of Cd and As concentrations

The average Cd and As concentrations in rice obtained from NCP districts are presented in Table 1. The concentrations of Cd in the two districts and areas are not significantly different ($p<0.05$). The mean value of Cd is 38.192 µg/kg while the median value is 28.257 µg/kg. The range of Cd in rice is <LOQ-261.786 µg/kg (Table 1). Among the 186 rice samples from the NCP, no sample exceeded the JECFA maximum allowable limit of Cd (400 µg/kg) of fresh weight. The threshold Cd level proposed by the European Commission is 200 µg/kg of fresh weight (Corguinha et al., 2015) and only one sample exceeded this level. Meharg et al., (2013) evaluated the Cd content in rice from several countries and observed the mean Cd concentration of rice in Sri Lanka to be 81 µg/kg. However, in the literature, a high amount of Cd in rice: 231 µg/kg in China (Ma et al., 2017), 480 µg/kg in imported rice in Iran (Naseri et al., 2015) and 329 µg/kg in brown Jasmine rice in Thailand (Hensawang and Chanpiwat, 2017) have been reported. Earlier studies in China and Japan have shown that the genotypes have an effect on Cd concentration in rice (Arao and Ae, 2003; Cheng et al., 2006) and such differences may be related with morphological and physiological characteristics. This did not seem to be the case in our study, even though we analysed 34 traditional and new rice varieties.

The concentration of tAs in rice from NCP ranged from <LOQ-575.94 µg/kg with the mean of 38.26 µg/kg and median <LOQ (Table 1). ANOVA for the 8 samples in the Nochchiyagama area of Anuradhapura was significantly different ($p<0.05$) from the mean value of the area. However, the Nochchiyagama samples, had 121.56 and 104.06 µg/kg higher mean and median values of As, respectively. The

<table>
<thead>
<tr>
<th>District</th>
<th>Area</th>
<th>n</th>
<th>Avg. Cd concentrations, (µg/kg, w/w)</th>
<th>Avg. As concentrations, (µg/kg, w/w)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Galkulama-Thirappane</td>
<td>22</td>
<td>22.817±18.379</td>
<td>35.48±48.14</td>
</tr>
<tr>
<td></td>
<td>Mahakandarawa-Galenbindanuwewa</td>
<td>24</td>
<td>31.983±30.358</td>
<td>88.01±112.25</td>
</tr>
<tr>
<td></td>
<td>Kahatagasgiliya-Horowpathana</td>
<td>3</td>
<td>37.147±40.687</td>
<td>&lt;LOQ</td>
</tr>
<tr>
<td></td>
<td>Nachchaduwa</td>
<td>11</td>
<td>61.158±56.228</td>
<td>&lt;LOQ</td>
</tr>
<tr>
<td></td>
<td>Rajanganaya</td>
<td>21</td>
<td>22.103±23.595</td>
<td>46.94±54.04</td>
</tr>
<tr>
<td></td>
<td>Nochchiyagama</td>
<td>8</td>
<td>43.340±34.926</td>
<td>121.56±87.09</td>
</tr>
<tr>
<td></td>
<td>Polonnaruwa Town</td>
<td>7</td>
<td>71.908±42.692</td>
<td>52.56±56.25</td>
</tr>
<tr>
<td></td>
<td>Jayanthipura-Hingurakkgoda</td>
<td>20</td>
<td>28.475±60.450</td>
<td>75.96±130.72</td>
</tr>
<tr>
<td></td>
<td>Diyabeduma</td>
<td>20</td>
<td>38.749±27.641</td>
<td>12.32±14.51</td>
</tr>
<tr>
<td></td>
<td>Bakamuna</td>
<td>5</td>
<td>35.152±14.688</td>
<td>&lt;LOQ</td>
</tr>
<tr>
<td></td>
<td>Siripura</td>
<td>12</td>
<td>47.461±47.245</td>
<td>&lt;LOQ</td>
</tr>
<tr>
<td></td>
<td>Dimbulagala</td>
<td>3</td>
<td>39.410±27.488</td>
<td>&lt;LOQ</td>
</tr>
<tr>
<td></td>
<td>All NCP rice</td>
<td>186</td>
<td>38.192±36.659</td>
<td>38.26±74.61</td>
</tr>
</tbody>
</table>

LOQ: Limit of Quantification
Table 2: Cd and As concentrations in traditional and newly improved rice varieties.

<table>
<thead>
<tr>
<th>Rice type</th>
<th>n</th>
<th>Avg. Cd concentrations (µg/kg)</th>
<th>Avg. As concentrations (µg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>New</td>
<td>142</td>
<td>32.31±32.731</td>
<td>45.23±79.74</td>
</tr>
<tr>
<td>Traditional</td>
<td>44</td>
<td>33.757±52.524</td>
<td>37.26±36.38</td>
</tr>
</tbody>
</table>

Table 3: Estimated Daily Intake, Target Hazard Quotient and Cancer Risk of As and Cd through rice consumption by Sri Lankans.

<table>
<thead>
<tr>
<th>Metal</th>
<th>RI</th>
<th>Adults</th>
<th>Children</th>
<th>HQ</th>
<th>Adults</th>
<th>Children</th>
<th>CR</th>
<th>Adults</th>
<th>Children</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cd</td>
<td></td>
<td>0.19</td>
<td>0.33</td>
<td>0.19</td>
<td>0.33</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>tAs</td>
<td></td>
<td>0.19</td>
<td>0.33</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>iAs</td>
<td></td>
<td>0.15</td>
<td>0.26</td>
<td>0.51</td>
<td>0.87</td>
<td></td>
<td>0.00028</td>
<td>0.00049</td>
<td></td>
</tr>
</tbody>
</table>

The highest amount of tAs (575.94 µg/kg) was reported in the sample obtained from the Jayanthipura - Hingurakkoda (Polonnaruwa district). There is no threshold level for total As published by the Codex Alimentarius Commission or the European Union for rice. Ma et al., (2017) found an average of 107 µg/kg rice grown in Guangdong Province, China. Rahman et al., (2011) evaluated the total As level in rice grown in Bangladesh and found a range from 74-301 µg/kg on a dry weight basis, while Williams et al., (2006) detected As concentrations ranging from <40-920 µg/kg in rice from 25 districts of Bangladesh, which are much higher than our study.

**Traditional vs newly improved rice varieties**

In 1960, the International Rice Research Institute (IRRI) introduced new rice varieties while Sri Lankan rice research institutes, and consequently the Bathalagoda centre, also introduced a number of new varieties. These varieties have high yield potential and thus are more popular than traditional varieties (TV) (Jayasumana et al., 2015). In this study, we analysed 10 TVs (n=44), namely, Kuruluthada, Pachchaperumaal, Suvandel, Madathawalu, Kahawatun, Rathkanda-El, Kulu-heenati, Kaha-mala, Pokura sama, and Suda-heenati. There was no significant difference between TVs for Cd concentration, while it was significant for As concentration (Table 2). Jayasumana et al., (2015) reported a low amount of As in TVs (11.6-64.2 µg/kg) that were cultivated without using agrochemicals. Similar studies were done by Kariyawasam et al., (2016), who analysed 5 types of TVs for As and Cd, also cultivated under organic farming conditions. The results for both As and Cd of the above studies are below the LOQ (LOQ-As: 40 µg/kg and Cd: 10 µg/kg). However, it is not possible to compare the present results with the reported values as those samples were obtained from controlled cultivation.

**Health risk assessment**

Based on the average BW (adults and children) and rice consumption, the EDI of Cd, tAs and iAs through rice consumption are summarized in Table 3.

The general population was divided into two groups, namely children (average weight 35 kg) and adults (60 kg). It was assumed that the rice consumption was the same for the two groups. Accordingly, the order of EDI for Cd, tAs, and iAs is higher in children than adults. When comparing the present results with the previous research on risk assessments of rice consumption in Asian countries, including China, Iran and Taiwan, it was found that As exposure in the adult Sri Lankan population is approximately two times lower than the EDI of the Chinese population, four times lower than the Iranian population, while it is two times higher than the Thai population. The As-EDI value of the Thai adult population, reported by Hensawang and Chanpiwat, (2017) was equal to 0.784 µg/day kg BW for brown jasmine rice. For the Chinese adult population, it was 0.44 µg/day kg BW for iAs (Chen et al., 2018; Naseri et al., 2015). In the case of Cd exposure, the EDI value in China is nine times (1.66 µg/BW.kg. day) higher than our study while Iranian and Thai exposures are four times higher (0.76 and 0.784 µg/day kg BW) than in the present study (Naseri et al., 2015; Hensawang and Chanpiwat, 2017; Ma et al., 2017). However, this comparison does not mean anything because the higher exposure rates, based on the human digestion system, are mainly dependent on three factors: the contaminant concentrations in rice, the ingestion rate of rice and the body mass of the individual (Meharg et al., 2013; Hensawang and Chanpiwat, 2017). Diyabalalage et al., (2016) studied EDI values for As in different zones from rice varieties and found that, based on Sri Lankan adults 60 kg body weight and 284 g daily rice consumption, derived values were similar to the current studies (0.20 µg/day kg BW), while being two times higher for Cd (0.40 µg/day kg BW).

The levels of non-carcinogenic health risk from the two metals studied are given in Table 3. Theoretically the THQ value was calculated according to equation (2) and compared with the threshold value of 1. Based on previous studies, if the THQ≥1, there is a potential health risk and hence it is necessary to take protective action (Gladyshchev et al., 2009). To estimate the health risk for more than one non-essential trace elements (NETEs) the Hazard Index (HI) is developed and is based on the cumulative value of each THQ. Though the THQ of As and Cd are below 1 for each...
metal and population group, there is a risk in the children’s group. The HI value for As and Cd for children is 1.20 (>1). Even in the adults’ group, the HI value is trending towards 1 (0.70) when considering only the two metals. Overall, it can be concluded that the Sri Lankan population is at risk from exposure through rice consumption.

Table 3 summarizes the cancer risk for iAs exposure through rice consumption. Even though the International Agency for Research on Cancer (IARC) has classified Cd as a known human carcinogen, the USEPA does not quantitatively calculate the oral slope factor for Cd (Al-Saleh and Abduljabbar, 2017). Hence, in this study, CR was calculated only for iAs. The CR-iAs values for adults and children are 2.87×10⁻⁴ and 4.92×10⁻⁴ respectively. The cancer risk is acceptable when the CR is in the range of 1×10⁻⁶ (1:1,000,000) chance of developing cancer in a human lifetime to 1×10⁻⁴ (1:10,000) chance of developing cancer in a human lifetime (Ma et al., 2016). They also highlighted that long-term exposure of iAs may cause skin, lung and bladder cancer. Our results indicate a significant CR to those consuming rice grown in NCP, as the main food.

It is important to emphasize that this value may be overestimated because we assumed that 100% of ingested As and Cd is bioavailable. It is necessary to carry out this kind of study for other nonessential metals, other population groups and based on total diet. The other limitation is that the metal concentrations were measured in untreated samples. Some studies have pointed out that the soaking and rinsing of rice with water help to minimize the metal concentration. These kinds of effects depend on the metal speciation and also vary with grain variety (Al-Saleh and Abduljabbar, 2017). However, in the worst-case scenario, this result could be appropriate.

CONCLUSION

Rice samples (n=186), including 44 traditional samples were collected from NCP, Sri Lanka and analysed for Cd and As contents. Considering all the varieties, As ranged between <LOQ-575.94 while Cd was <LOQ-261.786 µg/kg on a wet weight basis. The Cd concentrations in all rice samples were well below the JECFA level of 400 µg/kg, while there is no guidance for tAs. Considering HQ and CR values, there are indications that the Sri Lankan population, especially children, are at risk of potential non-carcinogenic health effects and carcinogenic health effects from iAs exposure. Some assumptions of this study may have overestimated the risk and hence it is suggested that studies about bio-accessible amounts and speciation analysis of As and Cd are necessary to obtain a more comprehensive idea regarding public health concerns of As and Cd due to rice consumption in Sri Lanka.

AVAILABILITY OF DATA AND MATERIALS

Please contact the corresponding author for data requests.

DECLARATION OF CONFLICT OF INTEREST

The authors declare that they have no competing interests

ACKNOWLEDGEMENT

Financial assistance of District Secretariat Office of Anuradhapura, Sri Lanka and analysis facilities provided by National Aquatic Resources Research and Development Agency (NARA), Sri Lanka are acknowledged.

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