Food Control 82 (2017) 196–202

Contents lists available at ScienceDirect

Food Control

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

Inorganic arsenic in rice and rice-based diets: Health risk assessment

Shofiqul Islam [a, b, c], Mohammad Mahmudur Rahman [a, b], M. Azizur Rahman [b, d], Ravi Naidu [a, b, *]

[a] Global Centre for Environmental Remediation (GCER), The University of Newcastle, University Drive, Callaghan, NSW, 2308, Australia
[b] Cooperative Research Centre for Contamination Assessment and Remediation of the Environment (CRC CARE), The University of Newcastle, University Drive, Callaghan, NSW, 2308, Australia
[c] Department of Soil Science, Bangladesh Agricultural University, Mymensingh, 2202, Bangladesh
[d] Centre for Environmental Sustainability, School of the Environment, University of Technology Sydney, P.O. Box 123, Broadway, NSW, 2007, Australia

A R T I C L E I N F O

Article history:
Received 7 April 2017
Received in revised form
15 June 2017
Accepted 17 June 2017
Available online 17 June 2017

Keywords:
Rice-based products
Young children
Inorganic As
Health risk
BMDL

A B S T R A C T

Total and inorganic arsenic (As) content in rice and rice-based diets (n = 59) obtained from supermarkets in South Australia were studied to investigate the contamination levels and whether consumption of these products pose potential health risks to young children and adults. Results show that of the 59 rice-based products, 31 (53%) exceeded the EU recommended value (100 μg/kg) of As for young children and 13 (22%) samples had higher than maximum level of 200 μg/kg recommended for adults. Arsenic content varies as rice crackers > baby rice > rice cakes > puffed rice > other rice-based snacks > ready-to-eat rice. Of the 6 categories of rice-based products, except ready-to-eat rice, all others exceeded the EU recommended value for young children. Even manufacture recommended servings deliver significant amounts (0.56–6.87 μg) of inorganic As. These amounts are within the range of $BMDL_{01}$ values indicated by the European Food Safety Authority (EFSA), which means the risk cannot be avoided for young children and adults considering the levels of total and inorganic As in rice-based products.

1. Introduction

Arsenic is a naturally occurring toxic element, which is ubiquitous in the environment and has been classified as a human carcinogen (group I), which with sustained exposure causes both carcinogenic and non-carcinogenic problems including nervous system disorder, developmental and reproductive issues, diabetes, heart disease and many more (IARC, 2004). Inorganic As [As(III) and As(V)] is more toxic compared to organic As [dimethylarsinic acid (DMA) and monomethylarsonic acid (MMA)]. Both As(III) and As(V) are potentially harmful to humans and animals, although As(III) is more toxic (Hindmarsh, McCurdy, & Savory, 1986; Vega et al., 2001). Humans' exposure to As mainly occurs through: firstly, drinking As-contaminated groundwater and consuming food crops grown in agricultural lands contaminated with As; and secondly, where contaminated irrigation water is used for cropping. Whenever, rice is the staple diet, As exposure from rice can be considerable and intake of inorganic As via this route is a significant risk factor, especially for populations who depend substantively on a rice diet (Mondal & Polya, 2008). Elevated concentrations of As have been detected in rice in several South-East Asian countries, which poses an additional risk to consumers (Correll, Huq, Smith, Owens, & Naidu, 2006; Mondal et al., 2010; Pal et al., 2009; Rahman, Ng, & Naidu, 2009; Williams et al., 2005). In rice grown in the Bengal Delta, the predominant As species present are usually inorganic and a small portion of organic As also exists (Williams et al., 2005). It was reported from West Bengal, India, that cooked rice with elevated As (>200 μg/kg) caused genotoxicity and it was found that As from cooked rice alone was responsible for the observed genetic effects in the study population (Banerjee et al., 2013). Due to the global food trade, millions of consumers are likely to be potentially exposed to As from the imported foods, especially rice and rice-based food products (Rahman, Rahman, Reichman, Lim, & Naidu, 2014).

In most cases, whole grain and polished rice, and bran are used to produce a variety of rice-based foods. These rice-based products are very popular worldwide especially for young children who are assumed to be the most vulnerable consumer group to As toxicity; they have a higher As body burden than adults (Rahman et al.,

* Corresponding author. Global Centre for Environmental Remediation (GCER), The University of Newcastle, University Drive, Callaghan, NSW, 2308, Australia.
E-mail addresses: ravi.naidu@crccare.com, ravi.naidu@newcastle.edu.au (R. Naidu).

http://dx.doi.org/10.1016/j.foodcont.2017.06.030
0956-7135/

2001). A study in the US suggests that rice is a potential source of As exposure for children (Davis et al., 2012). The impact of a mother's As consumption on fetal gene expression is evidenced by transcript levels in newborn cord blood (Fry et al., 2007). Studies have also provided evidence that children are more susceptible to the toxic effects of inorganic As (Vahter, 2008, 2009).

Several studies have reported that rice and rice-based products used for infants contained high levels of both total and inorganic As (Ljung, Palm, Grandér, & Vahter, 2011; Signes-Pastor, Carey, & Meharg, 2016; Sun et al., 2009). Research has shown that at least 50% of rice-based food products in the UK have As levels which exceeded those being proposed by the European Union (EU); 179 different products were tested and 53% of these products had As higher than the maximum level of 100 μg/kg recommended for young children (Signes-Pastor et al., 2016). The Commission Regulation (EU) 2015/1006 of 25 June 2015 has issued a limit value of 100 μg/kg for rice destined for the production of foods for infants and young children and 200 μg/kg for adults. This regulation has been in force since January 1, 2016 (EFSA, 2014). In 2014, the European Food Safety Authority (EFSA) investigated the dietary exposure of people in 21 European countries to inorganic As, and the data show that grain-based processed products are the main contributors of inorganic As for all age groups except infants and toddlers (EFSA 2014). The mean dietary exposure to inorganic As among these groups ranged between 0.20 and 1.37 μg/kg bw per day (EFSA 2014). The mean dietary exposure to inorganic As according to surveys in the adult population (including adults, elderly and very elderly) ranged from 0.09 to 0.38 μg/kg bw per day (EFSA 2014).

A recent study on Australian rice, notably brown rice, found As content substantially higher than imported rice on sale in Australia which poses potential serious health risk to Asian adult immigrants (Rahman et al., 2014). A few studies have been conducted on potential health risk of As to children from their rice-based food products in Europe and the US, but no information is available about the health risk of toxic inorganic As in products produced in Australia. This is especially so given that such products besides being consumed here are also exported to other countries, for example China and South Korea. For this reason, it is crucial to investigate the As levels in rice-based diets available in Australia and its associated health risk to both adults and young children. Considering that As is carcinogenic and highly toxic, reliable assessments of the health risks associated with the intake of As through rice-based products consumption are urgently needed, especially for children. In this study, we determine the levels of As (total and inorganic) in a variety of rice and rice-based products, and to assess inorganic As exposure and its associated health risks to both adults and young children.

2. Materials and methods

2.1. Selection of samples

Fifty-nine rice products in 6 categories commonly found in supermarkets were analyzed for total and speciated As. Samples of rice crackers [cracker is a baked food typically made from rice flour (n = 10)], rice cakes [rice cake is a kind of food item made from puffed rice that has been shaped, condensed, or otherwise combined into a single object (n = 11)], puffed rice [puffed rice is a type of puffed grain made from rice, commonly used in breakfast cereal or snack foods (n=6)], baby rice [ground rice cereals consumed by baby to young children (n = 4)], ready-to-eat rice [cooked rice ready to eat, only need 2–3 min heat using microwave oven (22)] and other rice-based snacks [rice biscuits, rice hoops etc.(6)] of 25 different popular commercial brands were purchased from supermarkets in Adelaide, South Australia during February 2015. The samples' expiry date (best before use) was between April 2015 and March 2016.

2.2. Sample preparations

With the exception of ready-to-eat rice, all rice-based products were oven dried (60 °C) and ground with mortar and pestle before microwave digestion for analysis. Between each samples, mortar and pestle were washed in tap water, deionised water and dried in a fan forced oven. For total As examination, the powdered samples were weighed accurately to 0.5 g into microwave digestion tubes to which 2 mL MQ water (ELGA Labpure), 3 mL trace analytical grade HNO_3 (70%), obtained from Fisher Chemicals and 2 mL H_2O_2 (30%, Ajax Finechem) were added and allowed to sit overnight (12 h). A microwave digestion system with 40 rotors (MARS 6, CEM) served for the digestion of the samples. Batches of 30 samples were prepared including a blank and a standard reference material (SRM 1568b rice flour) obtained from the National Institute of Standard and Technology (NIST), USA. Firstly, the temperature was raised to 55 °C, and held for 10 min, and then 75 °C, and again held for 10 min. Finally, temperature was lifted to 90 °C and held for 30 min. The ready-to-eat rice was directly weighed and followed the same digestion procedure as described above. The digests were diluted to 10 mL using 0.1% HNO_3 and then passed through a 0.45 μm syringe filter (MCE, Agilent Technologies). Finally, 10 mL aliquot of the digest from each tube was transferred to a plastic tube for instrumental analysis.

Arsenic speciation analysis (inorganic As, MMA and DMA) was carried out with the procedure utilized by Signes-Pastor et al. (2016). Briefly, the powdered samples were weighed accurately to 0.25 g into microwave digestion tubes to which 10 mL of 1% conc. HNO_3 (trace analysis grade) was added and allowed to sit overnight (12 h). Batches of 30 samples were prepared including a blank and a SRM (1568b rice flour), which has inorganic As [As(III) and As(V)], DMA and MMA concentrations certified. The samples were then digested in the microwave digestion system. The digests were diluted to 10 mL and filtered by a 0.45 μm syringe filter. Lastly, 1 mL aliquot of the digest from each tube was transferred to a 2 mL HPLC glass vial (amber) for chromatographic analysis. For both total As and its speciation we have run three replicates of each samples.

2.3. Instrumentation

An Agilent 7500ce (Agilent Technologies, Tokyo, Japan) inductively coupled plasma mass spectrometer (ICP-MS) coupled with auto-sampler (ASX-520, CETAC Technologies) and integrated samples introduction system (ISIS) was used to determine the amount of As in the digested samples. An Agilent 1100 liquid chromatography system (Agilent Technologies, Tokyo, Japan) equipped with a guard column and a Hamilton PRP-X100 separation column, coupled with ICP-MS served to determine As species. The detection limit (DL) of the total As was 0.01 μg/L. The detection limit for speciation in As standard solutions [As(III), As(V), DMA and MMA] by the IC-ICP-MS ranged between 0.1 and 0.3 μg/l (Chen, Akter, Rahman, & Naidu, 2008).

2.4. Quality control: analysis of standard reference materials (SRM)

We found the concentration of total As in rice flour was SRM 270 ± 13 (n = 6) μg/kg, which indicates 95% recovery of the certified value of 285 ± 14 μg/kg. Similar SRM was used to validate the results of As speciation in rice-based samples included in this study. Our results revealed that inorganic As recovery was around 98%. The certified values for inorganic As, MMA and DMA of SRM 1568b