Ecotoxicology and Environmental Safety 197 (2020) 110601

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

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

Do arsenic levels in rice pose a health risk to the UK population?

Manoj Menon[a,*], Binoy Sarkar[b,1], Joseph Hufton[a], Christian Reynolds[a,2], Saul Vazquez Reina[c], Scott Young[c]

[a] *Department of Geography, University of Sheffield, Sheffield, S102TN, United Kingdom*
[b] *Department of Animal and Plant Sciences, University of Sheffield, Sheffield, S102TN, United Kingdom*
[c] *School of Biosciences, University of Nottingham, Sutton Bonington Campus, Leicestershire, LE12 5RF, United Kingdom*

ARTICLE INFO

Keywords:
Total arsenic
Arsenic speciation
Lifetime cancer risk
Rice consumption
Target hazard quotient
Margin of exposure

ABSTRACT

Consumption of rice and rice products can be a significant exposure pathway to inorganic arsenic (iAs), which is a group 1 carcinogen to humans. The UK follows the current European Commission regulations so that iAs concentrations must be $<$ 0.20 mg kg^{-1} in white (polished) rice and $<$ 0.25 mg kg^{-1} in brown (unpolished) rice. However, iAs concentration in rice used for infant food production or direct consumption has been set at a maximum of 0.1 mg kg^{-1}. In this context, this study aimed to evaluate iAs concentrations in different types of rice sold in the UK and to quantify the health risks to the UK population. Here, we evaluated 55 different types of rice purchased from a range of retail outlets. First, we analysed all rice types for total As (tAs) concentration from which 42 rice samples with tAs $>$ 0.1 mg kg^{-1} were selected for As speciation using HPLC-ICP-MS. Based on the average concentration of iAs of our samples, we calculated values for the Lifetime Cancer Risk (LCR), Target Hazard Quotient (THQ) and Margin of Exposure (MoE). We found a statistically significant difference between organically and non-organically grown rice. We also found that brown rice contained a significantly higher concentration of iAs compared to white or wild rice. Notably, 28 rice samples exceeded the iAs maximum limit stipulated by the EU (0.1 mg kg^{-1}) with an average iAs concentration of 0.13 mg kg^{-1}; therefore consumption of these rice types could be riskier for infants than adults. Based on the MoE, it was found that infants up to 1 year must be restricted to a maximum of 20 g per day for the 28 rice types to avoid carcinogenic risks. We believe that consumers could be better informed whether the marketed product is fit for infants and young children, via appropriate product labelling containing information about iAs concentration.

1. Introduction

Geogenic arsenic poses one of the most significant public health challenges, affecting 140 million people across 70 countries in the world (WHO, 2018). In particular, inorganic arsenic (iAs) is a group 1 carcinogen as advised by the International Agency for Research on Cancer (IARC). Also, iAs is included in the list of top 10 chemicals, or group of chemicals, of significant public health concern by the World Health Organisation (WHO 2016). Arsenic exposure affects almost every organ in the human body and produces a range of health effects, including skin lesions, cancer, diabetes and lung diseases (NRC, 2014). Risk assessment, therefore, requires a comprehensive understanding of absolute intake of arsenic from multiple sources such as food, water, soil, dust and air (Carlin et al., 2016), depending on the region. In particular, rice, the staple food for more than half of the world's population, has been shown to accumulate iAs in more significant amounts than other cereals (Carey et al., 2019; Liao et al., 2018; Meharg et al., 2008; Nunes and Otero, 2017). In regions where arsenic exposure through drinking water is minimal, rice and other foods rich in iAs can contribute significantly to human arsenic intake (54–85%) as shown in a US-based study (Kurzius-Spencer et al., 2013). Similarly, in the UK, arsenic exposure through drinking water is not widely reported except in private water supplies in Cornwall (Middleton et al., 2016). However, in the UK, arsenic exposure through the consumption of rice and rice products can be significant. Up to 90% of households in the UK buy rice; consumption of rice has increased by 450% since the 1970s, probably due to the growing Asian ethnic population and food diversification (Schenker, 2012; Rice Association, n. d). The *per capita* rice consumption in the UK is about 5.6 kg y^{-1} (i.e., 0.015 kg d^{-1}) which is slightly higher than across the European Union (4.9 kg y^{-1}) (OECD,

* Corresponding author.
E-mail address: m.menon@sheffield.ac.uk (M. Menon).
[1] Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, United Kingdom
[2] Centre for Food Policy; Myddelton Street Building, City, University of London, Myddelton Street, EC1R 1UW, London, United Kingdom

https://doi.org/10.1016/j.ecoenv.2020.110601
Received 19 January 2020; Received in revised form 1 April 2020; Accepted 5 April 2020

2015; Schenker, 2012); however, it varies significantly across the UK population. For example, Asian ethnic groups constitute 7.5% of the total population in England and Wales, and according to National Diet and Nutrition Survey (NDNS Years 1–9, 2008/09–2016/17), 42–43% of the sampled UK population consumed rice over a period of four days. In contrast, it was 73–78% for sub-population of Asian or Asian British ethnicity over the same period. Across the sampled UK population who did consume rice, adults (16+ years of age) consumed 0.036 kg d^{-1}, while children and infants (0–15 years of age) consumed 0.021 kg d^{-1}. The adults of the sampled sub-population of Asian or Asian British ethnicity consumed 0.047 kg d^{-1}, while children and infants of Asian or Asian British ethnicity consumed 0.028 kg d^{-1} (NatCen Social Research, 2019).

Regardless of ethnicity, rice and rice-based products are widely used for weaning and as an infant food due to nutritional benefits and relatively low allergic potential (Signes-Pastor et al., 2016a). Rice is also a preferred gluten-free choice for the Celiac disease affected population (one in every 100 people) in the UK (Munera-Picazo et al., 2014; National Health Service UK, 2020). Also, according to European Food Safety Authority (EFSA, 2014), children are 2–3 times more susceptible to arsenic risks than adults due to higher food and fluids consumption rates relative to their body weight (Guillod-Magnin et al., 2018).

It is essential to reduce the risk of arsenic exposure to humans through rice consumption (Carlin et al., 2016; Islam et al., 2016). Total arsenic concentration (tAs) in food products includes comparatively highly toxic inorganic (iAs) forms (i.e., As^{III} and As^{V}) as well as less toxic organic (oAs) forms (e.g., dimethylarsenic acid (DMA) and traces of monomethylarsonic acid (MMA)); all these arsenic species are commonly found in rice (Islam et al., 2016; Meharg et al., 2008; Norton et al., 2013). Rice is mainly grown under flooded soil conditions that are conducive to the reduction of As^{V} to As^{III}. The resulting lower-valent species, arsenous acid ($H_3As^{III}O_3$; pKa 9.2), is soluble in flooded soil and readily bioavailable to rice for uptake in the plant parts including grains (Bakhat et al., 2017; Islam et al., 2016).

In 2011, the US-EPA estimated lifetime cancer risk (LCR) through iAs intake and recommended 1.5 mg kg^{-1} bw d^{-1} as the upper limit for iAs oral intake rate (US-EPA, 2011; Jallad, 2019). Furthermore, Joint Expert Committee on Food Additives (JECFA) with Food and Agricultural Organisation (FAO) provided a Benchmark Dose Lower Confidence Limit ($BMDL_{0.5}$) of iAs as 0.003 mg kg^{-1} bw d^{-1} (FAO, 2011) for various cancers and skin lesions, which replaced the previous Provisional Tolerable Weekly Intake (PTWI) of 0.015 mg kg^{-1} bw d^{-1}. The EFSA identified a range of $BMDL_{0.1}$ (i.e. dose needed for 0.1% increase of various cancers and skin lesions of iAs between 0.0003 and 0.008 mg kg^{-1} bw d^{-1} (EFSA, 2009, 2014; Guillod-Magnin et al., 2018; Jallad, 2019; Rintala et al., 2014). Subsequently, the European Commission (EC, 2015) has set a maximum permissible limit of iAs in rice, which is currently followed in the UK. Based on this, the limits for iAs are 0.20 mg kg^{-1} in white or polished rice, and 0.25 mg kg^{-1} in parboiled or husked rice. However, rice destined to produce food for infants and young children must be < 0.10 mg kg^{-1}. Similarly, the US Food and Drug Administration (US FDA, 2016) has limited the iAs concentration of 0.10 mg kg^{-1} in infant rice cereals.

Rice imported and marketed in the UK include wild, white and brown rice, which can be organically or non-organically produced. Rice labels often contain additional information about the grain size classification (short, medium and long) set up by the UK government (HM Revenue and Customs, 2015) mainly for import and export purposes. The main aims of this research were to evaluate arsenic concentrations in various types of rice and to determine the arsenic exposure risk to the UK population from this source as there have been no previous studies that compared different rice types available in the UK retail outlets. The specific objectives of this investigation are listed below.

1. To assess and compare arsenic (total and its different species) concentrations in rice marketed in the UK, based on rice cultivation methods (organic or non-organic) as well as rice types (wild, white or brown).
2. To determine the risk to the UK population (adult males and females, and infants), based on reported consumption rates.

2. Methods

2.1. Collection and processing of rice samples

Fifty-five different rice types were purchased (0.5–1 kg packets) from various retailers such as major supermarket chains and online suppliers in the UK (the suppliers have been anonymised) during August–September 2018. Our sampling strategy was to obtain as many representative samples as possible from wild (n = 6), white (n = 36) and brown or unpolished (n = 13) rice under organic (n = 16) and non-organic (n = 39) categories (Supplemental Table 1). Though technically not a member of the rice family, wild rice (*Zizania* sp.) was included in this study due to its increasing presence in the UK retail stores. Note that we did not include 'ready to eat' rice brands or wild-white rice mixtures. Out of the 55 rice samples, 20 did not contain any specific information on their country of origin (Supplemental Table 1).

The moisture content of rice samples was determined using a gravimetric method (65 °C; up to 48 h); this was used to produce dry-weight based arsenic concentrations. For chemical analysis, approximately 150–200 g of rice was sampled and finely ground using a ball mill grinder (Retsch MM 200 Model Mixer Mill). Three sub-samples (~1–2 g) were taken for total arsenic analysis and arsenic speciation. To avoid cross-contamination, the grinding jars were cleaned thoroughly using acetone and ultrapure water (18.2 MΩ cm) and then left to dry before reuse.

2.2. Chemical analysis

2.2.1. Total arsenic (tAs) concentration

Samples (0.2 g dry weight) of rice powder were microwave-digested in 6 mL HNO_3 (Primar Plus grade, Fisher Scientific, U.K.) in perfluoroalkoxy (PFA) vessels (Multiwave; Anton Paar GmbH, St. Albans, U.K.). The digested samples were diluted to 20 mL, and then 1-in-10 with ultrapure water (18.2 MΩ cm), immediately before elemental analysis by inductively coupled plasma mass spectrometry (ICP-MS). Each digestion batch included operational blanks and certified reference material (NIST 1568b, rice flour) for quality assurance (QA) purposes. The average percentage recovery of tAs (0.285 mg kg^{-1}) was 104%. Multi-element analysis of diluted aliquots was undertaken by ICP-MS (Thermo-Fisher Scientific iCAP-Q; Thermo Fisher Scientific, Bremen, Germany).

2.2.2. Arsenic speciation

Based on tAs concentrations in 55 rice samples, 42 samples with tAs > 0.10 mg kg^{-1} were selected for further arsenic speciation analysis. On average, ~70% of the tAs in rice consists of the toxic iAs, and it rarely exceeds 85% mark (Islam et al., 2016). Thus, the benchmark of 0.10 mg kg^{-1} tAs would be well within the current lowest regulatory limit for infants (0.10 mg kg^{-1} iAs) in Europe. In other words, tAs < 0.10 mg kg^{-1} can be considered safe for the consumption for all age groups, including infants. The selected rice types in the speciation analysis included four wild, 13 brown and 25 white rice samples composed of both organically (n = 9) and non-organically (n = 33) grown categories.

Based on the above criteria, the arsenic speciation was carried out using a separate extraction and analysis from the tAs assay. Extraction of arsenic species from rice flour was undertaken using a method similar to that described by Huang et al. (2010). Approximately 1.5 g each of the 42 selected rice samples was suspended in 15 mL 2% nitric acid (Primar Plus grade, Fisher Scientific, U.K.) in polypropylene 'DigiTubes' (SCP Science, Quebec, Canada), and heated at 95 °C for 1.5 h

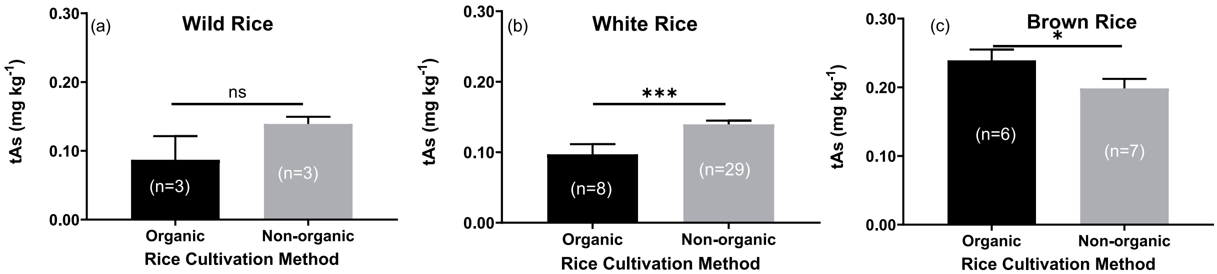

Fig. 1. (a–c). Comparison of total As (tAs) in organically and non-organically grown wild (a), white (b) and brown rice (c). The error bars indicate standard error of means (SEM); n is the number of samples used in the analysis indicated on each bar.

on a Teflon-coated graphite block digester (Model A3, Analysco Ltd, U.K.). Cooled suspensions were made up to 50 mL with ultrapure water (18.2 MΩ cm), and an aliquot (c. 6 mL) was syringe-filtered to < 5 μm for the speciation analysis. Arsenic speciation was undertaken using a coupled LC-ICP-MS (HPLC 5000 series, Thermo Scientific) with a PRP-X100 anion exchange column (PS-DVB/Trimethyl ammonium exchanger; 5 μm particle size; 4.6 mm ID; 250 mm length); the eluent was 20 mM $NH_4H_2PO_4$ and $(NH_4)_2HPO_4$ (analytical grade) at pH = 5.6, pumped at 1.5 mL min^{-1} in isocratic mode. Standards included 5.0 μg L^{-1} arsenite (As^{III}) and arsenate (As^{V}) (Spex Certiprep, Stanmore, U.K.), and 5.0 μg L^{-1} dimethylarsinic acid (DMA) and monomethylarsonic acid (MMA) (purity > 98%; Sigma/Merck, Darmstadt, Germany). Chromatography runtime was c. 13 min per sample. Based on the data obtained, we used concentrations of individual species to obtain the sum of inorganic (As^{III} and As^{V}) and organic (DMA and MMA) species for the statistical analysis and presentation of data.

2.3. Health risk calculations

The risk to humans from arsenic is based on estimated daily intake (EDI, mg kg^{-1} d^{-1}) which is calculated as follows (Liao et al., 2018; Weber et al., 2019):

$$EDI = \frac{AC \times ADC}{bw} \tag{1}$$

where AC is the average concentration of iAs in rice (mg kg^{-1}), ADC is the average daily consumption rate of rice (kg d^{-1}), and bw represents the average body weight of the local population (kg). For the UK, bw values for adult males, adult females and infants (1-year-old) were taken as 83.6, 70.2 and 9 kg, respectively (Office of National Statistics, 2018).

The lifetime cancer risk (LCR) was calculated using EDI, and a slope factor (SF = 1.5 mg kg^{-1} d^{-1}) established by the United States Environmental Protection Agency (US EPA, 2011), which assumes daily exposure over an entire lifetime. The acceptable upper limit for LCR, set by the US EPA, is 1.0×10^{-4}. The LCR is given by Eq. (2):

$$LCR = EDI \times SF \tag{2}$$

The US EPA method for target hazard quotient (THQ) calculated from EDI and a reference oral dose (RfD) (Eq. (3)); The oral RfD for iAs set by the US EPA (0.0003 mg kg^{-1} d^{-1}) (US EPA, 1988) was used for calculating THQ.

THQ < 1 indicates no risk.

$$THQ = \frac{EDI}{RfD} \tag{3}$$

Finally, the Margin of Exposure (MoE) (Guillod-Magnin et al., 2018; Jallad, 2019; Rintala et al., 2014) was also calculated as follows:

$$MoE = \frac{BMDL_{0.1}}{EDI} \tag{4}$$

where $BMDL_{0.1}$ is Benchmark Dose Lower Confidence Limit and EDI is Estimated Daily Intake as per Eq. (1). The $BMDL_{0.1}$ is set at 0.0003 mg kg^{-1} bw d^{-1} for 0.1% increased incidence of various cancers as per EFSA, which is the same as RfD set by US EPA for THQ. In summary, the THQ is the inverse of MoE if $BMDL_{0.1}$ is set at 0.0003 mg kg^{-1} bw d^{-1}; hence the THQ values ideally be < 1 whereas the MoE > 1 to avoid iAs health risks.

Three different scenarios were tested to assess the risks to the UK population. The first scenario was based on the *per capita* consumption rate of rice in the UK (i.e., 0.015 kg d^{-1}) (Schenker, 2012) and the average iAs of 42 rice samples examined (0.13 mg kg^{-1}). In the second and third scenarios, we calculated the maximum permissible *per capita* consumption rates of rice for the above-mentioned age groups to avoid health risks.

2.4. Statistical analyses

GraphPad Prism (v 8) software was used to perform the statistical analysis and prepare the figures. Non-parametric tests, including Mann-Whitney test and Kurkal-Wallis Analysis of Variance (ANOVA), were used in combination with Dunn's multiple comparison test to compare different groups. In our presented graphs, statistical significance is presented as "ns" P > 0.05 (not significant), "*" for P ≤ 0.05, "**" for P ≤ 0.01, "***" for P ≤ 0.001 and "****" for P ≤ 0.0001.

3. Results

3.1. Total arsenic concentration in rice

Total arsenic (tAs) in the 55 rice samples (Supplemental Table 1; rice selected for speciation are indicated using*) analysed ranged from 0.01 to 0.37 mg kg^{-1} with an average of 0.15 (± 0.07) mg kg^{-1}. When we compared organic and non-organic rice cultivations for tAs in wild, brown and white rice types, the results showed no effect of rice cultivation method on tAs concentrations in wild rice (Fig. 1). The high standard error for organic rice in Fig. 1a was due to one wild rice sample included in this group. There was a significant difference observed in white rice (Fig. 1b) and brown rice (Fig. 1c) due to a change in the rice cultivation systems. In the case of white rice, non-organically grown rice contained a significantly higher concentration of tAs compared to organically grown white rice (P = 0.0004), and organically grown brown rice contained significantly more tAs compared to non-organic ones (P = 0.0189).

When data from all rice types were pooled together (i.e., wild, white and brown), there was no statistically significant difference between organically and non-organically grown rice categories (Supplemental Fig. 1a). Similarly, we statistically analysed the data using a non-parametric Kruskal- Wallis ANOVA test to compare wild, white and brown rice types irrespective of their cultivation methods. This analysis showed that rice type significantly influenced tAs levels (P < 0.0001), as shown in Supplemental Fig. 1b; the concentration of tAs in brown rice was almost double that of wild or white rice.

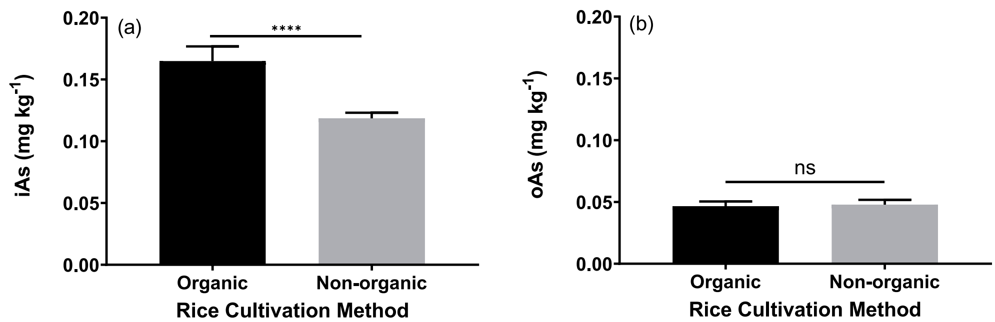

Fig. 2. (a & b) Comparison of inorganic (iAs) and organic As (oAs) concentrations in organically (n = 9) and non-organically (n = 33) grown rice as shown in a and b, respectively. The error bars indicate standard error of means (SEM).

3.2. Total inorganic and organic arsenic concentrations in rice

The average concentrations of iAs and oAs in the 42 rice types analysed were 0.129 ± 0.048 (range: 0.065–0.286) and 0.047 ± 0.034 (range: 0.009–0.203) mg kg^{-1}, respectively. On average, the iAs concentration in the tested varieties was 73% (± 1.2% SD) of tAs. Out of the 42 samples, 14 samples were below the infant maximum limit for iAs (0.1 mg kg^{-1}) with an average iAs concentration of 0.082 (± 0.012) whereas the average iAs concentration of the remaining 28 samples was 0.152 (± 0.041) mg kg^{-1}.

We present iAs (sum of As^{III} and As^{V}) and oAs (sum of DMA and MMA) concentrations when grown under two rice cultivation methods (Fig. 2 a & b); results showed a statistically significant ($P < 0.0001$) difference between the cultivation methods in the concentration of iAs but not oAs ($P = 0.355$). We were unable to compare iAs in wild, brown and white types of rice under organic and non-organic types (i.e. similar to Fig. 1) due to the insufficient number of replicates. Both wild and brown rice types contained similar concentrations of iAs, which were different from the white rice (Fig. 3a). An opposite trend was found for the concentration of oAs, where the white rice contained the highest concentration of oAs (Fig. 3b). Overall non-parametric ANOVA showed that rice type significantly influenced both iAs ($P < 0.0001$) and oAs concentrations ($P < 0.0048$). Comparison of these rice types showed that a significant difference was found between wild and white, and between white and brown rice for both iAs and oAs (Fig. 3 a & b).

3.3. Comparison of arsenic species (As^{III}, As^{V} and DMA) in rice

We compared concentrations of arsenic species (As^{III}, As^{V} and DMA) under different rice cultivation methods (Fig. 4), and between rice types (Fig. 5). MMA was present in traces or not detected in most of the samples, and hence was not included in this comparison. The As^{III} concentration of organically grown rice was significantly higher ($P < 0.0001$) than that of non-organically grown rice (Fig. 4a). However, the concentrations of As^{V} and DMA were similar under both cultivation methods (Fig. 4 b-c), and the differences were not statistically significant.

Different rice types significantly ($P < 0.0001$) influenced As^{III} concentrations. Both wild and white rice types did not show any significant difference, but they were significantly lower in As^{III} concentration than the brown rice (Fig. 5 a). Rice types also significantly influenced As^{V} concentrations ($P < 0.0001$) and, as shown in Fig. 5b, wild rice showed the greatest concentration of As^{V}, followed by brown and white rice. The differences between these rice types were statistically significant. The concentration of DMA was also influenced by rice type ($P = 0.0019$), and average DMA concentrations followed the order white > brown > wild rice with a significant difference between wild and white, as well as between white and brown rice (Fig. 5c). The difference in DMA between wild and brown rice was not statistically significant.

3.4. Relationship between total, inorganic and organic arsenic in rice

On average, iAs constituted 73% of the total sum of all species (iAs + oAs), but the range was 36–95% in the rice samples examined. The relationship between iAs and the total of all species (iAs + oAs) was linear and statistically significant ($P < 0.0001$) in all cases for different types of rice (Supplemental Fig. 2 a-e). However, the R^2 value for organically grown rice (0.92) was higher than for non-organically grown rice ($R^2 = 0.68$). Similarly, R^2 values for different rice types were also different (0.97 for brown, 0.88 for wild and 0.66 for white rice).

3.5. Health risks

We considered three scenarios for the human health risk assessment of rice arsenic, as described in Table 1. The first scenario was based on the reported *per capita* consumption rate of rice in the UK (i.e., 0.015 kg d^{-1}) (Schenker, 2012) and the mean iAs concentration (0.13 mg kg^{-1}) of the 42 rice samples examined. Accordingly, the lifetime cancer risks (LCR) for UK adult males, adult females and infants were 3.5×10^{-5} (i.e., 3.5 individuals per 100,000 of male population),

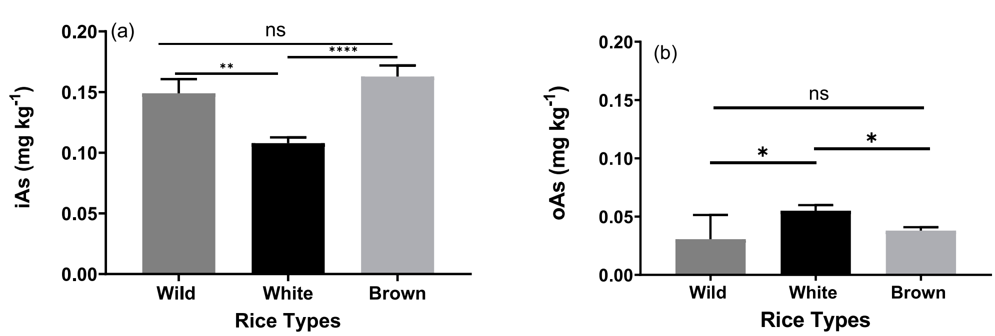

Fig. 3. (a & b) Comparison of wild (n = 4), white (n = 25) and brown (n = 13) rice in their inorganic (iAs) and organic As (oAs) concentrations as shown in a and b, respectively. The error bars indicate standard error of means (SEM).

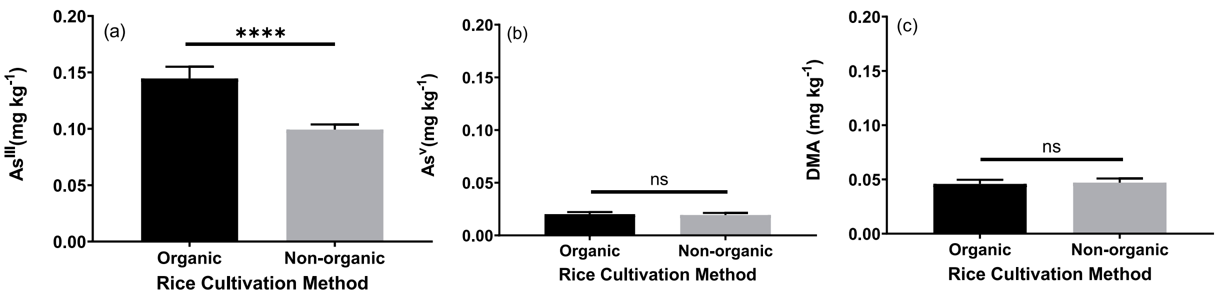

Fig. 4. (a–c). Comparison of As^{III}, As^{V} and DMA concentrations in organically (n = 9) and non-organically (n = 33) grown rice as shown in a, b, and c respectively. The error bars indicate standard error of means (SEM).

4.17×10^{-5} (4.17 per 100,000 of female population) and 3.25×10^{-4} (3.25 per 10,000 of infant population), respectively. The corresponding target hazard quotients (THQs) were 0.08, 0.09 and 0.72, respectively. The MoE values were also > 1 in all groups. The risk nearly doubled when we considered the maximum iAs concentration (0.29 mg kg^{-1} of a brown short-grained organic rice) found in the present study.

However, to avoid carcinogenic risks (i.e., LCR $< 1 \times 10^{-4}$) for men, women and infants, the consumption rates must not exceed 0.043, 0.036 and 0.0046 kg d^{-1}, respectively, as shown in the second scenario. These values correspond to a weekly maximum consumption rate of 0.301, 0.252 and 0.0322 kg for men, women and infants, respectively. In this scenario, THQ and MoE were 0.22 and 4.5 for all groups.

If we consider THQ or MoE, rice consumption rate must be < 0.19, 0.16 and 0.02 kg d^{-1} for men, women and infants, respectively, to avoid any health risks (Scenario 3). However, at this rate of consumption, the LCR would increase by a factor of four for all groups. Note that ADCs derived in this scenario for adult male and female (Table 1) were well above the average rice consumption rates for > 16-year-old population (the UK average = 0.036 kg d^{-1}; Asian or Asian British ethnic communities = 0.047 kg d^{-1}) as shown by the NDNS survey (see the introduction). However, ADC derived for infants was very close to the current average consumption rate of 0.021 kg d^{-1} for < 16 years old. However, if we use the rice consumption rate of < 16 years old children from Asian communities (i.e. 0.028 kg d^{-1}), the MoE will be 0.74, increasing the risk of arsenic exposure.

4. Discussion

4.1. Arsenic concentrations in rice

This is the first study, which has quantified differences in human health risks from iAs using a substantial number of rice samples marketed in the UK. Even though our overall strategy was to obtain as many samples as we could, we were not able to obtain an equal number of samples from all rice types. This was because most supermarket chains and online retailers have similar product ranges mostly dominated by white and non-organic rice types in comparison to the others. To increase the sample size from organic types, we bought additional samples from a few organic health food online suppliers. Wild rice (pure without mixing with white rice) was only available through online retailers as they were not available in any major supermarket chains. Thus, our sample numbers also reflected the availability or popularity of various rice in the UK. The study could not successfully relate the risk to the origin of rice samples because 20 out of the 55 samples analysed did not contain this information on their packaging labels. However, the origin could be an important factor, as demonstrated in a recent compressive study (Carey et al., 2019) where the authors reported that lowest iAs concentrations were found in rice sourced from East Africa and the Southern Indonesian islands. However, rice sourced from South American rice types were universally high in iAs. However, none of our samples originated from the above regions as per the information available (Suppl. Table 1) on the packaging. A study from Italy (Sommella et al., 2013) which examined 101 rice types (mostly different varieties of short-grain *japonica*) and iAs in their samples ranged from 0.08 to 0.11 mg kg^{-1} by variety. Though not shown in this paper, 15 of our samples were short-grain rice, and the average iAs was 0.125 ± 0.065 (range: 0.060–0.336) mg kg^{-1}; however, our samples are from a diverse range of suppliers and origins are not known for some samples. In another comprehensive study (Signes-Pastor et al., 2016b) from the Iberian Peninsula (Portugal and Spain) in which samples were collected from field-grown rice (20 samples) as well as the market (144 samples composed for white, brown and parboiled). The market-derived samples showed higher iAs in brown rice (0.053–0.247 mg kg^{-1}) in comparison to white rice (0.027–0.175 mg kg^{-1}), and parboiled rice iAs range was very similar to the white rice. The sample means for white and brown rice were 0.071 and 0.157 mg kg-1, respectively, in comparison to 0.108 and 0.163 mg kg-1 in this study.

Rintala et al. (2014) investigated iAs in both long grain rice (and rice-based baby food products) in Finland and found that the range of iAs concentrations in rice samples was 0.09–0.28 mg kg^{-1}. Although not shown in this paper, we analysed the data based on the grain length (23 long; 4 medium and 15 short grains samples) and iAs range in long-grain rice was 0.045–0.213 mg kg^{-1}, fitting well with the findings by Rintala et al. (2014). However, this study did not include baby food products; such studies have been conducted earlier (Signes-Pastor et al.,

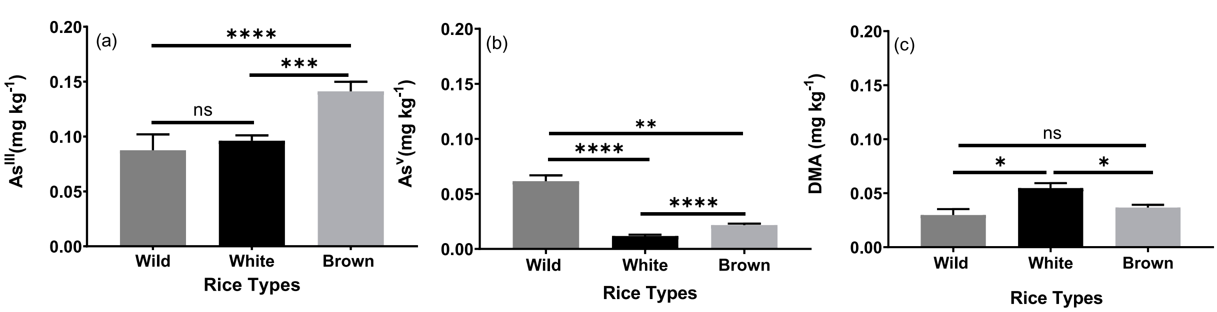

Fig. 5. (a–c). Comparison of As^{III}, As^{V} and DMA concentrations in wild (n = 4), white (n = 26) and brown (n = 13) rice as shown in a, b, and c respectively. The error bars indicate standard error of means (SEM).

Table 1
Lifetime Cancer Risk (LCR), Target Hazard Quotient (THQ) and Margin of Exposure (MoE) under different scenarios. Key: AC = Average concentration of As_{io} in rice (mg kg^{-1}); ADC = Average daily consumption rate of rice (kg); BW = Average body weight of the local population; and EDI = Estimated daily intake. Scenario 1 is based on current per capital consumption rates of 0.015 kg day^{-1} in the UK. Scenario 2 is maximum ADC to avoid LCR. Scenario 3 is ADC based on THQ and MoE.

Target Population	AC (As_{io}) (mg kg^{-1})	ADC (kg)	BW (kg)	EDI (mg kg^{-1} day^{-1})	LCR	THQ	MoE
Scenario 1							
Adult Male	0.13	0.015	83.6	2.3×10^{-5}	3.50×10^{-5}	0.08	12.86
Adult Female	0.13	0.015	70.2	2.8×10^{-5}	4.17×10^{-5}	0.09	10.80
1 year old infant	0.13	0.015	9	2.2×10^{-4}	3.25×10^{-4}	0.72	1.38
Scenario 2							
Adult Male	0.13	0.043	83.6	6.6×10^{-5}	1.0×10^{-4}	0.22	4.5
Adult Female	0.13	0.036	70.2	6.6×10^{-5}	1.0×10^{-4}	0.22	4.5
1 year old infant	0.13	0.0046	9	6.6×10^{-5}	1.0×10^{-4}	0.22	4.5
Scenario 3							
Adult Male	0.13	0.192	83.6	3.1×10^{-4}	4.47×10^{-4}	1.0	1.00
Adult Female	0.13	0.162	70.2	3.1×10^{-4}	4.50×10^{-4}	1.0	1.00
1 year old infant	0.13	0.0208	9	3.0×10^{-4}	4.50×10^{-4}	1.0	1.00

2016a, 2016b) in the UK. Investigations that compared organically and non-organically grown rice types for arsenic health risk assessment are rare. Our findings are similar to a market-based study conducted in Brazil by Segura et al. (2016) which showed no difference between tAs for organic or non-organically (i.e., conventionally) grown rice; however, they found that iAs was 41–45% higher in organically produced husked or polished rice than the corresponding samples from conventionally produced rice. In contrast, a study conducted by Rahman et al. (2014) in Australia, found significantly higher tAs and iAs in organic brown rice compared to non-organic brown rice, similar to our findings. Although we do not have details of the source or amount of organic matter (OM) added during cultivation of the rice samples analysed, the addition of OM in lowland rice may play a significant role in increasing arsenic mobility and plant uptake. Addition of OM can reduce the redox potential of rice soils, which can trigger arsenic dissolution as arsenite (As^{III}) from adsorbed arsenate (As^{V}) forms in the soil (Islam et al., 2016; Rowland et al., 2009; Smedley and Kinniburgh, 2002). Based on this, we can expect to have more tAs and iAs when rice is grown organically. However, previous experimental data have suggested the opposite conclusion (Ma et al., 2014; Norton et al., 2013) and indicated an increase in oAs, which suggested that organically grown rice could be a healthier option for human consumption. Here we show that iAs increased significantly in organically grown rice, more specifically As^{III}, which supports the recognised mechanisms of arsenic reduction, desorption and increased availability of iAs (As^{III} and As^{V}) compared to the methylated forms (DMA and MMA) (Raab et al., 2007).

Arsenic data on wild rice are sparse in the literature. The first study on wild rice examined 26 rice types from Michigan state in the US (Nriagu and Lin, 1995) for arsenic (tAs) and other trace elements, and found that tAs ranged from 0.06 to 0.14 mg kg^{-1} with an average of 0.066 mg kg^{-1}. In our study, the tAs range was found to be 0.01–0.22 mg kg^{-1} with an average of 0.11 (± 0.078, n = 18) mg kg^{-1}. A study from Wisconsin, USA, reported a similar average tAs concentration in seeds of wild rice (Bennett et al., 2000). Two further studies investigated arsenic species in wild rice and reported concentrations of 0.08 mg kg^{-1} (Heitkemper et al., 2001) and 0.01 mg kg^{-1} (Williams et al., 2005) of iAs compared to our average value of 0.15 mg kg^{-1} iAs, which was significantly higher than white rice. More recently, a study from Valencia, Spain, did not detect any iAs in the wild rice examined (Torres-Escribano et al., 2008).

Regardless of the place of origin of rice, with reasonably large sample size, we have demonstrated that brown or unpolished rice contained significantly higher concentrations of tAs and iAs compared to white rice. Our findings are in agreement with previous observations (Batista et al., 2011; Islam et al., 2016; Meharg et al., 2008; Rahman et al., 2014; Zhu et al., 2008). This is due to the presence of the bran in brown rice (Meharg et al., 2008), although a US market-based study, which compared polished and unpolished (brown) rice, found no statistical difference in tAs concentration (Williams et al., 2007). In terms of arsenic speciation, brown rice accumulated more As^{III} (Fig. 5a) compared to wild or white rice whereas As^{V} concentrations were significantly higher in wild rice compared to the others, which warrants further research on uptake mechanisms. In particular, concentrations of the less toxic DMA species were significantly lower in wild and brown rice, compared to white rice, suggesting that DMA accumulates more in the starchy interior part of the rice and less in the bran of brown or wild rice. Further studies on wild rice are required to understand the mechanisms behind the accumulation of higher concentrations of As^{v} in comparison to white and brown rice (Fig. 5b). The findings from this study should be taken into consideration when advocating the consumption of brown rice for increased dietary fibre, minerals and B-vitamins in the bran (Schenker, 2012).

In a recent review, Liao et al. (2018) demonstrated that only one-third (11 out of 30) of the reported studies on carcinogenic risk assessment of rice arsenic were based on measured concentrations of iAs. The rest of the studies estimated iAs based on either regression equations, or in most cases it was assumed that iAs was ~80% of tAs. Based on our data for 42 rice types, on average, iAs constituted 73.46% (± 11.91) of the sum of all species of arsenic. This could enable the saving of the substantial analytical costs involved in arsenic speciation, in a limited number of labs in the UK, by selecting rice types based on tAs > 0.1 mg kg^{-1} for speciation. In other words, rice types with tAs < 0.1 mg kg^{-1} cannot be regarded as unsafe for consumption, especially for infants, and we found only 13 such samples out of 55. The linear regression equations developed in this study (Suppl. Fig. 2 a-e) could be used to predict iAs based on tAs concentrations for various groups of rice in regions where arsenic speciation facilities are not available or unaffordable.

4.2. Health risks

According to the EFSA (EFSA, 2014), the mean dietary iAs exposure infants are limited by the lack of an adequate number of surveys; according to two dietary surveys (one of which had only 16 participants). Based on the available data, the mean dietary exposure to iAs for infants ranged from 0.0.00024 to 0.00043 mg kg^{-1} bw d^{-1} (min-max Lower Bound or LB). The 95th percentile dietary exposure based on single qualifying study ranged from 0.00054 to 0.00167 mg kg^{-1} bw

d^{-1} (Lower to Upper Bound or UB). Based on *per capita* rice consumption (0.015 kg with average iAs concentration 0.13 mg kg^{-1}), the EDI for infants was 0.000217 mg kg^{-1} bw d^{-1} (scenario 1) which is in agreement with these EFSA (2014) findings.

Based on 15 dietary surveys conducted in 14 countries, mean dietary exposure to iAs for adults (> 18 - < 65 years old) ranged from 0.00011 to 0.00017 mg kg^{-1} bw d^{-1} (min LB- max LB) and from 0.00024 to 0.00038 mg kg^{-1} bw d^{-1} (min UB- max UB). The 95th percentile dietary exposure estimates ranged from 0.00018 to 0.00032 mg kg^{-1} bw d^{-1} (min LB- max LB) and 0.00044–0. mg kg^{-1} bw d^{-1} (min UB-max UB). Based our scenario 1 used in this study, the UK adult (male and female) population exposure (EDI) is at least an order magnitude lower than above values, and more close to scenario 3.

We used three widely popular risk assessments (LCR, THQ and MoE), and using multiple assessments are often found to be useful in understanding the risks posed by iAs in different age groups. Based on LCR results obtained, risks posed by rice consumption in the UK is very low compared to risks faced in countries such as Bangladesh. For instance, the LCR is 50 in 10,000 in Bangladesh compared to 2 in 10,000 in the EU (Liao et al., 2018; Meharg et al., 2009; Nunes and Otero, 2017). While an average UK citizen consumes ~100 g (uncooked dry weight) rice a week, this could be as high as 850 g (uncooked) rice per week for South Asian people (Khokhar et al., 2013) aggravating their LCR by a factor of 4.

More recent papers used MoE (Guillod-Magnin et al., 2018; Rintala et al., 2014), whereas others used all three methods (e.g. Jallad, 2019). Rintala et al. (2014) a worst-case scenario for MoE using maximum iAs in long-grain rice (0.28 mg kg^{-1}) and baby products (0.21 mg kg^{-1}) and used the lowest $BMDL_{0.1}$ of 0.0003 mg kg^{-1} bw d^{-1}). They found MoE was ≤1 for adult men and women and for children who consumed different rice in different forms (porridge or non-porridge products). However, the consumption rate in the studied population was 4–5 times higher than the average *per capita* rice consumption in the UK, and we used an average iAs concentrations in rice as opposed to maximum concentrations found in our study.

Similarly, a recent comprehensive study based on rice and rice-based products (105 samples) from Switzerland (Guillod-Magnin et al., 2018) found that the concentrations of tAs and iAs were significantly higher in brown rice compared to white rice samples. They analysed the MoE through iAs and DMA and in several scenarios tested, iAs intake was found to be higher than EFSA's $BMDL_{0.1}$ lower limit of 0.0003 mg kg^{-1} bw d^{-1}, suggesting that health risk by iAs for certain toddlers through the consumption of rice and rice products could not be excluded. Their findings are in agreement from our first scenario where we found infants are likely at risk from iAs exposure compared to adult male or female groups. The MoE based on $BMDL_{0.1}$ 0.0003 mg kg^{-1} bw d^{-1} is the most conservative assessment although if we use the upper limit of 0.008 mg kg^{-1} bw d^{-1}, the MoE will increase dramatically; using this value, for example, in Scenario 1, MoE will rise to 342, 288 and 36 for UK adult male, female and infants, respectively.

We can conclude that out of 55 rice types studied, 28 exceeded the infant maximum limit for iAs stipulated by the European Commission, and are therefore unsuitable for the production of baby food products or direct feeding (Carey et al., 2019). Based on the MoE, we recommend the consumption of these 28 rice types may be restricted to ~20 g d^{-1} for infants in order to minimise the risks. Therefore, it is appropriate that manufacturers and suppliers inform consumers about iAs concentrations in marketed rice and rice products made for infants and young children up to 5 years old.

5. Conclusions

This study examined arsenic concentrations in 55 rice types marketed in the UK in which we compared cultivation methods (organic or non-organically grown) and various types of rice (wild, white/polished and brown/unpolished). The total arsenic (tAs) concentrations in organic white rice were significantly lower than non-organic types, whereas the opposite was true for brown rice. However, inorganic arsenic (iAs) concentration of organically grown rice was significantly higher than non-organically produced rice. The order of accumulation of iAs in different rice types was brown > wild > white. Out of 55 rice types studied, 28 exceeded infant iAs maximum limit stipulated by the European Commission as unsuitable for the production of baby food products or direct feeding. Our study showed that health risks due to rice arsenic consumption are confined mainly to infants in the UK. We recommend that consumers could be better informed whether rice and rice products are suitable for infants and young children up to 5 years in the product description labels.

Author contributions

MM designed the study including sample collection, moisture analysis, prepared graphs and statistical analyses including the risk calculations and wrote the manuscript. BS helped to design the study, rice collection as well as reviewed the manuscript. JH prepared all rice samples through ball milling and was responsible for sample storage and postage of samples, and reviewed the paper; CR contributed provided UK-wide rice consumption database, and reviewed the paper. SVR contributed to the analysis of total arsenic and its species in their labs at the University of Nottingham and SY provided overall supervision of the arsenic analyses, and reviewed the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The project was funded by the Science and Technology Research Council (STFC) Food Network (Grant No: ST/P003079/1), United Kingdom (https://www.stfcfoodnetwork.org/). We also would like to acknowledge Prof Ed Rhodes (The University of Sheffield) and Prof Andrew Meharg (Queen's University, Belfast) for their support and advice throughout the research.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecoenv.2020.110601.

References

Bakhat, H.F., Zia, Z., Fahad, S., Abbas, S., Hammad, H.M., Shahzad, A.N., Abbas, F., Alharby, H., Shahid, M., 2017. Arsenic uptake, accumulation and toxicity in rice plants: possible remedies for its detoxification: a review. Environ. Sci. Pollut. Res. 24, 9142–9158. https://doi.org/10.1007/s11356-017-8462-2.

Batista, B.L., Souza, J.M.O., De Souza, S.S., Barbosa, F., 2011. Speciation of arsenic in rice and estimation of daily intake of different arsenic species by Brazilians through rice consumption. J. Hazard Mater. https://doi.org/10.1016/j.jhazmat.2011.04.087.

Bennett, J.P., Chiriboga, E., Coleman, J., Waller, D.M., 2000. Heavy metals in wild rice from northern. Wisconsin. Sci. Total Environ 246, 261–269. https://doi.org/10.1016/S0048-9697(99)00464-7.

Carey, M., Meharg, C., Williams, P., Marwa, E., Jiujin, X., Farias, J.G., De Silva, P.M.C.S., Signes-Pastor, A., Lu, Y., Nicoloso, F.T., Savage, L., Campbell, K., Elliott, C., Adomako, E., Green, A.J., Moreno-Jiménez, E., Carbonell-Barrachina, Á.A., Triwardhani, E.A., Pandiangan, F.I., Haris, P.I., Lawgali, Y.F., Sommella, A., Pigna, M., Brabet, C., Montet, D., Njira, K., Watts, M.J., Meharg, A.A., 2019. Global sourcing of low-inorganic arsenic rice grain. Expo. Heal 1–9. https://doi.org/10.1007/s12403-019-00330-y.

Carlin, D.J., Naujokas, M.F., Bradham, K.D., Cowden, J., Heacock, M., Henry, H.F., Lee, J.S., Thomas, D.J., Thompson, C., Tokar, E.J., Waalkes, M.P., Birnbaum, L.S., Suk, W.A., 2016. Arsenic and environmental health: state of the science and future research opportunities. Environ. Health Perspect. https://doi.org/10.1289/ehp.1510209.

European Commission, 2015. Commission Regulation (EU) 2015/1006 of 25 June 2015 amending Regulation (EC) No 1881/2006 as regards maximum levels of inorganic arsenic in foodstuffs (Text with EEA relevance) - publications Office of the EU. URL. https://publications.europa.eu/en/publication-detail/-/publication/4ea62ae9-1bc8-11e5-a342-01aa75ed71a1/language-en accessed 12.19.18.

European Food Safety Authority, 2009. Scientific opinion on arsenic in food. EFSA panel on the contaminants in the food chain. EFSA J 7 (10), 1351.

European Food Safety Authority, 2014. Dietary exposure to inor- ganic arsenic in the European population. EFSA 12 (3), 3597.

Food and Agriculture Organization of the United Nations (FAO), World Health Organization (WHO), 2011. Safety evaluation of certain contaminants in food. In: Seventy-second Meeting of the Joint FAO/WHO Expert Committee on Food Additives. WHO/FAO, Geneva Available from: http://apps.who.int/food-additives-contaminants-jecfadatabase/chemical.aspx?chemID=1863.

Guillod-Magnin, R., Brüschweiler, B.J., Aubert, R., Haldimann, M., 2018. Arsenic species in rice and rice-based products consumed by toddlers in Switzerland. https://doi.org/10.1080/19440049.2018.1440641.

Heitkemper, D.T., Vela, N.P., Stewart, K.R., Westphal, C.S., 2001. Determination of total and speciated arsenic in rice by ion chromatography and inductively coupled plasma mass spectrometry. J. Anal. At. Spectrom 16, 299–306. https://doi.org/10.1039/b007241i.

HM Revenue and Customs, 2015. Classifying rice for import and export - GOV.UK. URLhttps://www.gov.uk/guidance/classifying-rice accessed 12.19.18.

Huang, J.H., Ilgen, G., Fecher, P., 2010. Quantitative chemical extraction for arsenic speciation in rice grains. J. Anal. At. Spectrom. https://doi.org/10.1039/c002306j.

Islam, S., Rahman, M.M., Islam, M.R., Naidu, R., 2016. Arsenic accumulation in rice: consequences of rice genotypes and management practices to reduce human health risk. Environ. Int. 96, 139–155. https://doi.org/10.1016/J.ENVINT.2016.09.006.

Jallad, K.N., 2019. The hazards of a ubiquitary metalloid, arsenic, hiding in infant diets: detection,. Speciation, Exposure, and Risk Assessment. Biol. Trace Elem. Res. 190, 11–23. https://doi.org/10.1007/s12011-018-1510-z.

Khokhar, S., Ashkanani, F., Garduño-Diaz, S.D., Husain, W., 2013. Application of ethnic food composition data for understanding the diet and nutrition of South Asians in the UK. Food Chem. 140, 436–442. https://doi.org/10.1016/J.FOODCHEM.2012.10.034.

Kurzius-Spencer, M., O'rourke, M.K., Hsu, C.H., Hartz, V., Harris, R.B., Burgess, J.L., 2013. Measured versus modeled dietary arsenic and relation to urinary arsenic excretion and total exposure. J. Expo. Sci. Environ. Epidemiol. https://doi.org/10.1038/jes.2012.120.

Liao, N., Seto, E., Eskenazi, B., Wang, M., Li, Y., Hua, J., 2018. A comprehensive review of arsenic exposure and risk from rice and a risk assessment among a cohort of adolescents in Kunming, China. Int. J. Environ. Res. Publ. Health 15, 1–17. https://doi.org/10.3390/ijerph15102191.

Ma, R., Shen, J., Wu, J., Tang, Z., Shen, Q., Zhao, F.-J., 2014. Impact of agronomic practices on arsenic accumulation and speciation in rice grain. Environ. Pollut. 194, 217–223. https://doi.org/10.1016/J.ENVPOL.2014.08.004.

Meharg, A.A., Lombi, E., Williams, P.N., Scheckel, K.G., Feldmann, J., Raab, A., Zhu, Y., Islam, R., 2008. Speciation and localization of arsenic in white and brown rice grains RN - environ. Sci.Technol. 42, 1051–1057. Environ. Sci. Technol. 42, 1051–1057. https://doi.org/10.1021/es702212p.

Meharg, A.A., Williams, P.N., Adomako, E., Lawgali, Y.Y., Deacon, C., Villada, A., Cambell, R.C.J., Sun, G., Zhu, Y.-G., Feldmann, J., Raab, A., Zhao, F.-J., Islam, R., Hossain, S., Yanai, J., 2009. Geographical variation in total and inorganic arsenic content of polished (white) rice. Environ. Sci. Technol. 43, 1612–1617. https://doi.org/10.1021/es802612a.

Middleton, D.R.S., Watts, M.J., Hamilton, E.M., Ander, E.L., Close, R.M., Exley, K.S., Crabbe, H., Leonardi, G.S., Fletcher, T., Polya, D.A., 2016. Urinary arsenic profiles reveal exposures to inorganic arsenic from private drinking water supplies in Cornwall, UK OPEN. https://doi.org/10.1038/srep25656.

Munera-Picazo, S., Burló, F., Carbonell-Barrachina, Á.A., 2014. Arsenic speciation in rice-based food for adults with celiac disease. Food Addit. Contam. Part A Chem. Anal. Control. Expo. Risk Assess. https://doi.org/10.1080/19440049.2014.933491.

Nat Cen Social Research, MRC Elsie Widdowson Laboratory, 2019. National Diet and Nutrition Survey Years 1-9, 2008/09-2016/17. [data Collection], fifteenth ed. https://doi.org/10.5255/UKDA-SN-6533-15. UK Data Service. SN: 6533.

National Health Service, UK, 2020. Coeliac disease - NHS [WWW document]. URLhttps://www.nhs.uk/conditions/coeliac-disease/ accessed 1.31.19.

Norton, G.J., Adomako, E.E., Deacon, C.M., Carey, A.-M., Price, A.H., Meharg, A.A., 2013. Effect of organic matter amendment, arsenic amendment and water management regime on rice grain arsenic species. Environ. Pollut. 177, 38–47. https://doi.org/10.1016/J.ENVPOL.2013.01.049.

NRC, 2014. Critical Aspects of EPA's IRIS Assessment of Inorganic Arsenic: Interim Report. National Academies Press, Washington DC.

Nriagu, J.O., Lin, T.-S., 1995. Trace metals in wild rice sold in the. United States. Sci. Total Environ 172, 223–228.

Nunes, L.M., Otero, X., 2017. Quantification of health risks in Ecuadorian population due to dietary ingestion of arsenic in rice. Environ. Sci. Pollut. Res. 24, 27457–27468. https://doi.org/10.1007/s11356-017-0265-y.

OECD, 2015. OECD iLibrary | Rice projections: consumption, per capita [WWW Document]. URL. https://www.oecd-ilibrary.org/agriculture-and-food/oecd-fao-agricultural-outlook-2015/rice-projections-consumption-per-capita_agr_outlook-2015-table125-en accessed 3.16.20.

Office for National Statistics, UK, 2018. The average briton. [WWW document]. URL. https://www.ons.gov.uk/aboutus/transparencyandgovernance/freedomofinformationfoi/theaveragebriton, Accessed date: 17 March 2020.

Raab, A., Williams, P.N., Meharg, A., Feldmann, J., 2007. Uptake and translocation of inorganic and methylated arsenic species by plants. Environ. Chem. 4 (197). https://doi.org/10.1071/EN06079.

Rahman, M.M.A.M., Rahman, M.M.A.M., Reichman, S.M., Lim, R.P., Naidu, R., 2014. Arsenic speciation in australian-grown and imported rice on sale in Australia: implications for human health risk. J. Agric. Food Chem. 62, 6016–6024. https://doi.org/10.1021/jf501077w.

Rintala, E.M., Ekholm, P., Koivisto, P., Peltonen, K., Venäläinen, E.R., 2014. The intake of inorganic arsenic from long grain rice and rice-based baby food in Finland - low safety margin warrants follow up. Food Chem. 150, 199–205. https://doi.org/10.1016/j.foodchem.2013.10.155.

Rice in the UK - Rice Association WWW Document, n.d. URL http://www.riceassociation.org.uk/content/1/3/rice-in-the-uk.html (accessed 2.8.19).

Rowland, H.A.L., Boothman, C., Pancost, R., Gault, A.G., Polya, D.A., Lloyd, J.R., 2009. The role of indigenous microorganisms in the biodegradation of naturally occurring petroleum, the reduction of iron, and the mobilization of arsenite from West Bengal aquifer sediments. J. Environ. Qual. 38 (1598). https://doi.org/10.2134/jeq2008.0223.

Schenker, S., 2012. An overview of the role of rice in the UK diet. Nutr. Bull. 37, 309–323. https://doi.org/10.1111/j.1467-3010.2012.02002.x.

Segura, F.R., de Oliveira Souza, J.M., De Paula, E.S., da Cunha Martins, A., Paulelli, A.C.C., Barbosa, F., Batista, B.L., 2016. Arsenic speciation in Brazilian rice grains organically and traditionally cultivated: is there any difference in arsenic content? Food Res. Int. 89, 169–176. https://doi.org/10.1016/j.foodres.2016.07.011.

Signes-Pastor, A.J., Carey, M., Meharg, A.A., 2016a. Inorganic arsenic in rice-based products for infants and young children. Food Chem. 191, 128–134. https://doi.org/10.1016/j.foodchem.2014.11.078.

Signes-Pastor, A.J., Carey, M., Carbonell-Barrachina, A.A., Moreno-Jiménez, E., Green, A.J., Meharg, A.A., 2016b. Geographical variation in inorganic arsenic in paddy field samples and commercial rice from the Iberian Peninsula. Food Chem. 202, 356–363. https://doi.org/10.1016/j.foodchem.2016.01.117.

Smedley, P., Kinniburgh, D., 2002. A review of the source, behaviour and distribution of arsenic in natural waters. Appl. Geochem. 17, 517–568. https://doi.org/10.1016/S0883-2927(02)00018-5.

Sommella, A., Deacon, C., Norton, G., Pigna, M., Violante, A., Meharg, A.A., 2013. Total arsenic, inorganic arsenic, and other elements concentrations in Italian rice grain varies with origin and type. Environ. Pollut. 181, 38–43. https://doi.org/10.1016/j.envpol.2013.05.045.

Torres-Escribano, S., Leal, M., Vélez, D., Montoro, R., 2008. Total and inorganic arsenic concentrations in rice sold in Spain, effect of cooking, and risk assessments. Environ. Sci. Technol. 42, 3867–3872. https://doi.org/10.1021/es071516m.

US EPA, ORD, I.R.I.S.D, 1988. Arsenic, inorganic CASRN 7440-38-2 | IRIS | US EPA, ORD.

United States Food and Drug Administration (US FDA), 2011. Age Dependent Adjustment Factor (ADAF) Application. US FDA, Washington D. C Available from: https://hero.epa.gov/hero/index. cfm/reference/details/reference_id/783747.

United States Food and Drug Administration (US FDA), 2016. Questions & Answers: Arsenic in Rice and Rice Products. US FDA, Washington D. C Available from: https://www.fda.gov/food/foodborneillnesscontaminants/metals/ucm319948.htm.

Weber, A.M., Mawodza, T., Sarkar, B., Menon, M., 2019. Assessment of potentially toxic trace element contamination in urban allotment soils and their uptake by onions: a preliminary case study from Sheffield, England. Ecotoxicol. Environ. Saf. 170, 156–165. https://doi.org/10.1016/J.ECOENV.2018.11.090.

WHO, 2018. ARSENIC PRIMER guidance on the investigation & mitigation of arsenic contamination. Avialble from https://www.unicef.org/wash/files/UNICEF_WHO_Arsenic_Primer.pdf .

WHO | Ten chemicals of major public health concern, 2016. https://www.who.int/ipcs/assessment/public_health/chemicals_phc/en/.

Williams, P.N., Price, A.H., Raab, A., Hossain, S.A., Feldmann, J., Meharg, A.A., 2005. Variation in arsenic speciation and concentration in paddy rice related to dietary exposure. Environ. Sci. Technol. 39, 5531–5540. https://doi.org/10.1021/es0502324.

Williams, P.N., Raab, A., Feldmann, J., Meharg, A.A., 2007. Market basket survey shows elevated levels of as in South Central U.S. processed rice compared to California: consequences for human dietary exposure. Environ. Sci. Technol. 41, 2178–2183. https://doi.org/10.1021/es061489k.

Zhu, Y.G., Sun, G.X., Lei, M., Teng, M., Liu, Y.X., Chen, N.C., Wang, L.H., Carey, A.M., Deacon, C., Raab, A., Meharg, A.A., Williams, P.N., 2008. High percentage inorganic arsenic content of mining impacted and nonimpacted Chinese rice. Environ. Sci. Technol. https://doi.org/10.1021/es8001103.