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

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Abstract

Consumption of rice and rice products can be a significant exposure pathway to inorganic arsenic (iAs) which is a class 1 carcinogen to humans. The UK follows the current European Commission regulations so that iAs concentrations are <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 (carcinogenic and non-carcinogenic) 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; non-carcinogenic risk) 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 lowest threshold 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 maximum 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.

Capsule: Nearly half of the 55 rice samples marketed in the UK are unfit for infant food purposes, whereas iAs levels pose a minimal health risk to adults.

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

1.0 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 class 1 carcinogen as advised by the International Agency for Research on Cancer (IARC), and has been 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 compressive 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 per year (i.e., 0.015 kg d^{-1}) which is slightly higher than across the European Union (4.9 kg per year) (OECD, 2015; Schenker, 2012); however, it varies significantly across the 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 4 day period, while 73%-78% of the sampled sub-population of Asian or Asian British ethnicity consumed rice over a 4 day period. Across the sampled UK population who did consume rice, adults (16+ years of age) consumed 13.48 kg per year (0.036 kg d^{-1}), while children and infants (0-15 years of age) consumed 8.01 kg per year (0.021 kg d^{-1}). The adults of the sampled sub-population of Asian or Asian British ethnicity consumed 17.49 kg per year, (0.047 kg d^{-1}), while children and infants of Asian or Asian British ethnicity consumed 10.27 kg per year (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., 2016). 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, 2020). Also, according to European Food Safety Authority (EFSA, 2014), children are 2-3 times more susceptible to arsenic risks than adults due to greater food and fluid consumption rates relative to their body weights (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).

The iAs risk is linked to the daily intake of arsenic through and US-EPA (2011) recommendations for oral intake rate of 1.5 mg kg^{-1} bw d^{-1} as the upper limit for lifetime cancer risk (LCR) with an acceptable LCR range of 10^{-4} -10^{-6} (0.01-0.0001%), representing 1 in 10, 000 or 1,000,000 chance of getting cancer in human life time, respectively (Jallad, 2019). Furthermore, Joint Expert Committee on Food Additives (JECFA) with Food and Agricultural Organization (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, 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 & 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.

2.0 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^{o}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. Three sub-samples were drawn from each of the ground rice samples (three replications) and stored air-tight in Eppendorf tubes for further laboratory analysis.

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 (0.1 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 total arsenic 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 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 Risk calculations

The risk to humans from arsenic can be calculated using carcinogenic and non-carcinogenic risk parameters, both requiring estimated daily intake (EDI, mg kg^{-1} d^{-1}) which was calculated using Eq. 1 (Liao et al., 2018; Weber et al., 2019):

$$EDI = \frac{AC \times ADC}{bw} \quad \text{(Eq. 1)}$$

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} bw^{-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:

$$LCR = EDI \times SF \quad \text{(Eq. 2)}$$

The US EPA method for non-carcinogenic risk uses a target hazard quotient (THQ) calculated from EDI and a reference oral dose (RfD) (Eq. 3); a value of THQ less than one indicates no risk.

$$THQ = \frac{EDI}{RfD} \quad \text{(Eq. 3)}$$

The oral RfD for iAs set by the US EPA (0.0003 mg kg^{-1} d^{-1}) (US EPA, 1988) was used for assessing the non-cancerous risk, although the RfD value is still under evaluation.

The final assessment tool used in this study was the Margin of Exposure (MoE) (Guillod-Magnin et al., 2018; Jallad, 2011; Rintala et al., 2014) which was calculated as follows:

$$MoE = \frac{BMDL_{0.1}}{EDI} \qquad \text{(Eq. 4)}$$

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 carcinogenic and non-carcinogenic risks, respectively.

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 $P \leq 0.05$, "**" for $P \leq 0.01$, "***" for $P \leq 0.001$ and "****" for $P \leq 0.0001$.

3.0 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 (Figure 1). The high standard error for organic rice in Figure 1a was due to one wild rice sample included in this group. There was a significant difference observed in white rice (Figure 1b) and brown rice (Figure 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 Figure 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 Figure 1b; the concentration of tAs in brown rice was almost double that of wild or white rice.

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 (Figure 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 Figure 1) due to insufficient number of replicates. Both wild and brown rice types contained similar concentrations of iAs, which were different from the white rice (Figure 3a). An opposite trend was found for the concentration of oAs, where the white rice contained the highest concentration of oAs (Figure 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 (Figure 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 (Figure 4), and between rice types (Figure 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 (Figure 4a). However, the concentrations of As^{V} and DMA were similar under both cultivation methods (Figure 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 (Figure 5 a). Rice types also significantly influenced As^{v} concentrations ($P<0.0001$) and, as shown in Figure 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 (Figure 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 Figure 2 a-e). However, the R^2 value for organically grown rice (0.92; 6a) was higher than for non-organically grown rice (R^2 =0.68; 6b). 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 Carcinogenic and non-carcinogenic 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 $3.5x10^{-5}$ (i.e., 3.5 individuals per 100,000 of male population), $4.17x10^{-5}$ (4.17 per 100,000 of female population) and 3.25 $x10^{-4}$ (3.25 per 10,000 of infant population), respectively. The corresponding non-carcinogenic 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 < $1x10^{-4}$) 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. This also produced a desirable THQ (0.22) and the MoE values ~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 used in this scenario for adults (Table 1) are well above the UK average rice consumption rate of 0.036 kg d^{-1} for >16 years old, established by the NDNS (see the introduction), and it is very close to the consumption rate of 0.021 kg d^{-1} for <16 year old population. This is also true for Asian population (consumption rate is 0.047 kg d^{-1} for >16 years old. However, the rice consumption rate of <16 years old children from Asian communities is 0.028 kg d^{-1}, which will produce a MoE value of 0.74, increasing the risk of arsenic exposure.

4.0 Discussion

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. Hence, we did not compare the regional influence on arsenic and its species. However, the origin could be an important factor, as demonstrated in a recent 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 (Suppl. Table 1) available on the packaging.

There are some recent studies that looked at rice and rice products, especially rice-based baby food products. For instance, Rintala et al (2014) investigated iAs in eight brands of long grain rice and 10 brands of baby food products in Finland, and found that range of iAs concentrations 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., 2016) 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 0.06-0.14 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} (Supplemental Fig. 2a) 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 (Figure 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 >0.1mg 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. Figure 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 are unaffordable.

This study found that the arsenic health risk posed by rice consumption in the UK and EU populations is very low compared to risks faced in countries such as Bangladesh: 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) 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.

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. 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) used the 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^{-1} 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, their consumption rate 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 calculated the MoE through iAs and DMA concentrations, 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}, suggestign 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 with our findings for the 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^{-1} 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., 2018). 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.

5.0 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 risk analysis showed that the risks due to rice arsenic consumption is confined mainly to infants in the UK. We recommend that cosumers could be informed whether rice and rice products are suitable for infants and young children up to 5 years in the product description labels.

Acknowledgements

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Manuscript figure captions

Figure 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.

Figure 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 in a and b, respectively. The error bars indicate standard error of means (SEM).

Figure 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).

Figure 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 in a, b, and c respectively. The error bars indicate standard error of means (SEM).

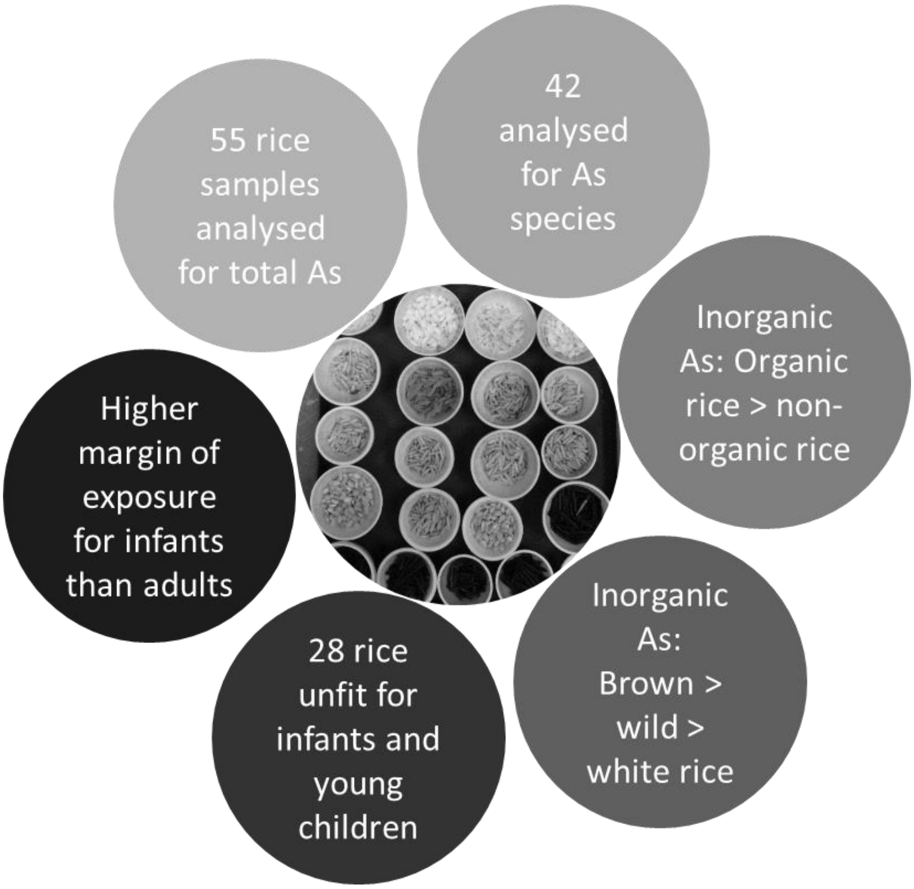
Figure 5 (a-c). Comparison of As^{III}, As^{V} and DMA concentrations in wild (n=4), white (n=26) and brown (n=13) rice as shown in in a, b, and c respectively. The error bars indicate standard error of means (SEM).

Supplementary Figure Captions

Figure 1 (a & b). Total As (tAs) contents based on rice culture method (a), and type of rice (b). The error bars indicate standard error of means (SEM) and n is the number of samples used in the analysis.

Figure 2 (a-e). Linear regression models established to predict iAs from tAs concentrations for various groups of rice. (a) all data combined; (b) organic rice; (c) non-organic rice; (d) brown rice; (e) white rice.

Graphical abstract



Highlights

• Total As was determined in 55 rice types and 42 for As species.

• Organic rice contained significantly more iAs compared to non-organic rice.

• The concentration of iAs rice types was brown > wild > white.

• 28 rice types were found to be unfit for infant food production or consumption.

Figures

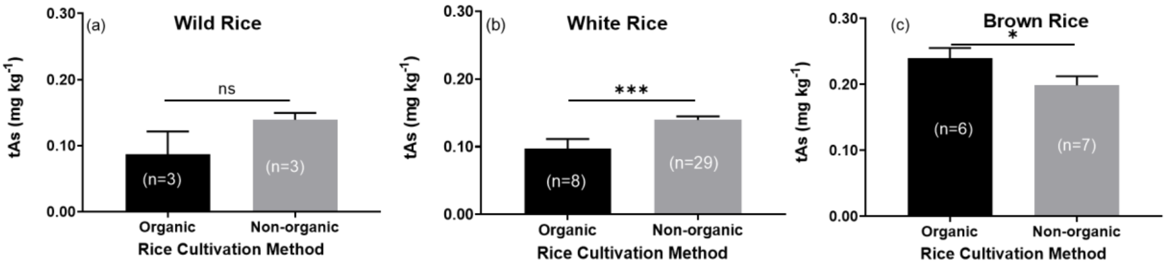


Fig. 1

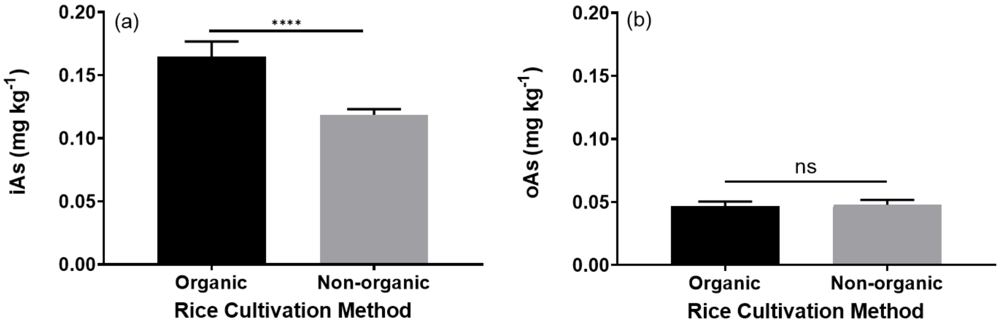


Fig. 2

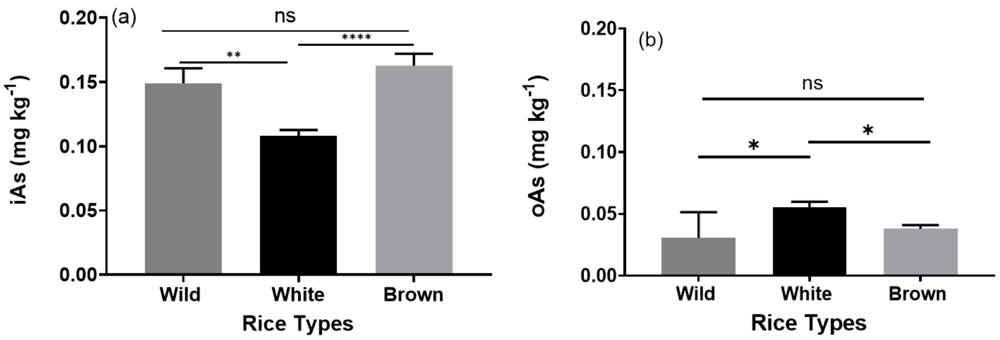
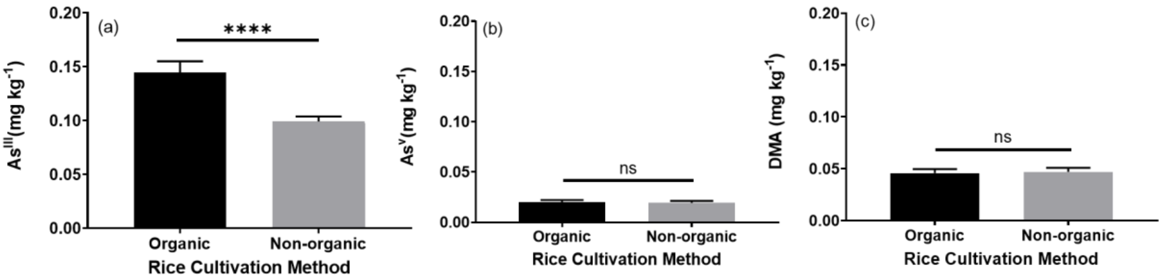


Fig. 3


(a)
As^{III}(mg kg^{-1})
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Organic
Non-organic
Rice Cultivation Method
(b)
As^{V}(mg kg^{-1})
ns
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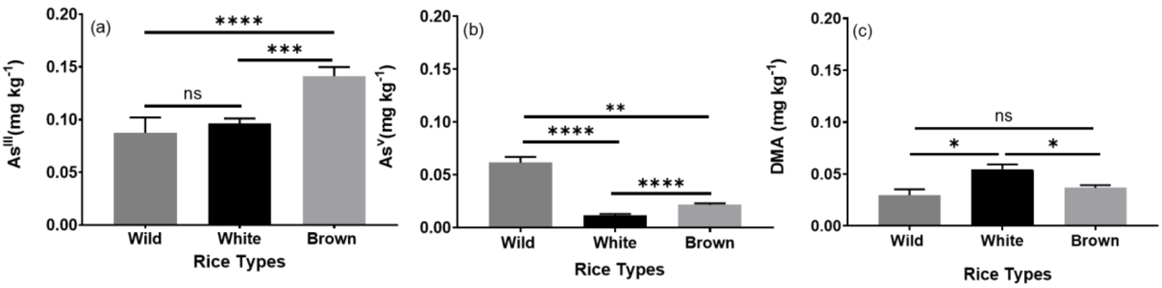
Non-organic
Rice Cultivation Method
(c)
DMA (mg kg^{-1})
ns
Organic
Non-organic
Rice Cultivation Method

Fig. 4


(a)
As^{III}(mg kg^{-1})
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Wild
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Brown
Rice Types
(b)
As^{V}(mg kg^{-1})
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Wild
White
Brown
Rice Types
(c)
DMA (mg kg^{-1})
ns
*
*
Wild
White
Brown
Rice Types

Fig. 5

Tables

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.

Scenario 1.

Target Population	AC (As_{io}) (mg kg^{-1})	ADC (kg)	BW (kg)	EDI (mg kg^{-1} day^{-1})	LCR	THQ	MoE
Adult Male	0.13	0.015	83.6	$2.3x10^{-5}$	$3.50x10^{-5}$	0.08	12.86
Adult Female	0.13	0.015	70.2	$2.8x10^{-5}$	$4.17x10^{-5}$	0.09	10.80
1 year old infant	0.13	0.015	9	$2.2x10^{-4}$	$3.25x10^{-4}$	0.72	1.38

Scenario 2.

Target Population	AC (As_{io}) (mg kg^{-1})	ADC (kg)	BW (kg)	EDI (mg kg^{-1} day^{-1})	LCR	THQ	MoE
Adult Male	0.13	0.043	83.6	$6.6x10^{-5}$	$1.0x10^{-4}$	0.22	4.5
Adult Female	0.13	0.036	70.2	$6.6x10^{-5}$	$1.0x10^{-4}$	0.22	4.5
1 year old infant	0.13	0.0046	9	$6.6x10^{-5}$	$1.0x10^{-4}$	0.22	4.5

Scenario 3.

Target Population	AC (As_{io}) (mg kg^{-1})	ADC (kg)	BW (kg)	EDI (mg kg^{-1} day^{-1})	LCR	THQ	MoE
Adult Male	0.13	0.192	83.6	$3.1x10^{-4}$	$4.47x10^{-4}$	1.0	1.00
Adult Female	0.13	0.162	70.2	$3.1x10^{-4}$	$4.50x10^{-4}$	1.0	1.00
1 year old infant	0.13	0.0208	9	$3.0x10^{-4}$	$4.50x10^{-4}$	1.0	1.00

Supplementary figures

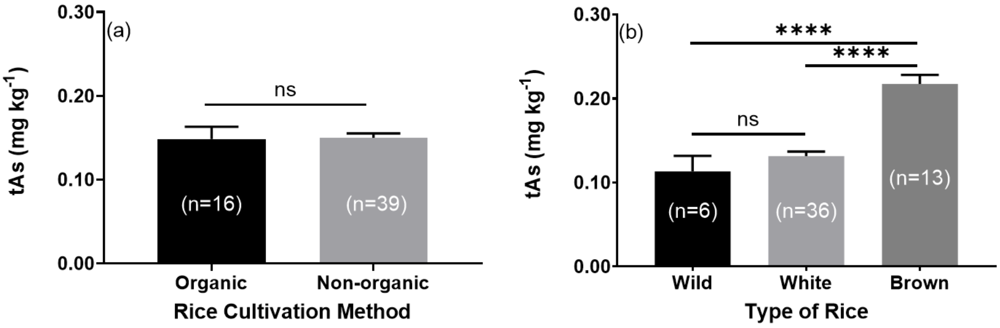

Fig. S1

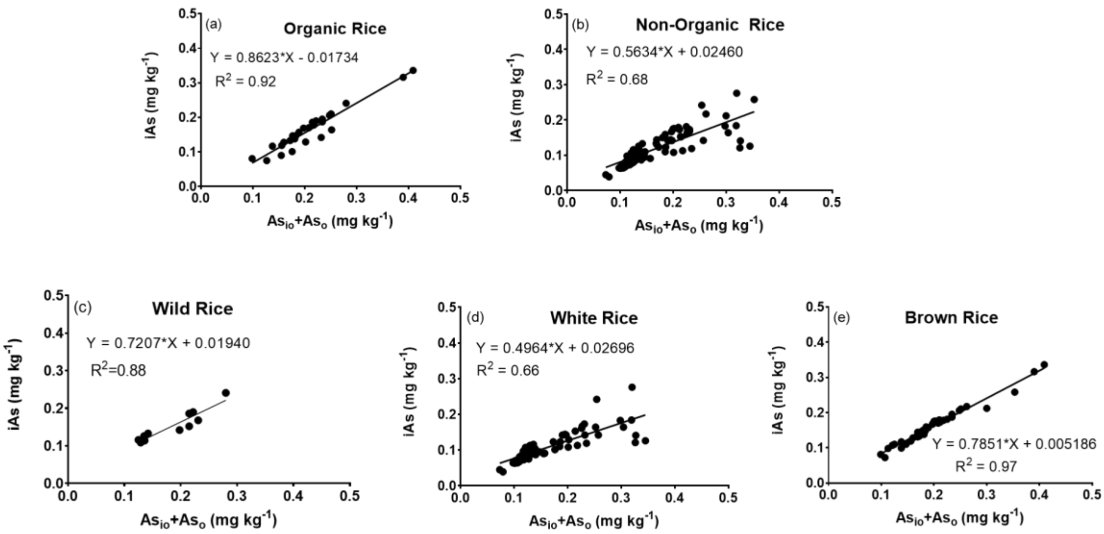

Fig. S2