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35 Abstract

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36 Keywords separated by ' - '

Arsenic - Rice - Ecuador - Spain - Cooking - Arsenic species

37 Foot note information

Rice can reach high arsenic concentrations, and the cooking process can substantially reduce its concentration

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Electronic supplementary material

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RESEARCH ARTICLE

Effect of cooking on arsenic concentration in rice

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Abstract

This study assessed the effect of rinsing and boiling on total content of As (tAs) and of its inorganic and organic forms in different types of rice (polished and brown) from Spain and Ecuador. Rice was subjected to five different treatments. The results showed that the treatment consisting of three grain rinsing cycles followed by boiling in excess water showed a significant decrease in tAs content compared with raw rice. Regarding As species, it is worth noting that the different treatments significantly reduced the content of the most toxic forms of As. The estimated lifetime health risks indicate that pre-rinsing alone can reduce the risk by 50%, while combining it with discarding excess water can reduce the risk by 83%; therefore, the latter would be the preferable method.

Keywords Arsenic · Rice · Ecuador · Spain · Cooking · Arsenic species

Q4 Introduction

Q5 Arsenic (As) intake through the consumption of drinking water and rice (*Oryza sativa*) with high contents of this element constitutes a severe public health issue for almost half the world's population. This issue is particularly dramatic in South and Southeast Asian countries such as Bangladesh, where 35–77 million people are at risk of arsenic poisoning (Rahman et al., 2018).

Q3 **Hypothesis** Rice can reach high arsenic concentrations, and the cooking process can substantially reduce its concentration

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As is considered a class I carcinogen by the IARC (2012), and the main source of exposure is contaminated drinking water (Rahman et al., 2018, Davis et al., 2017, Rasheed et al., 2018). However, in countries where rice is a staple foodstuff (i.e., Southeast Asia, Latin America), rice is also a major source of As intake due to its capacity for As accumulation (Rahman et al., 2018, Williams et al., 2007a,b, Davis et al., 2017).

The predominant species of arsenic in rice grain are inorganic forms (AsIII, AsV) and dimethylarsinic acid (DMA) (Zhu et al., 2008, Signes-Pastor et al., 2016), with inorganic forms showing a much higher toxicity (Meharg and Zhao, 2012). Moreover, the toxicity of As has been observed to depend on other factors such as the amount of rice consumed (Mandal et al., 2019), type of rice (polished, brown, organic, etc.) (Segura et al., 2016, Yim et al., 2017, Meharg et al., 2008, Zhu et al., 2008), body weight (USEPA, 1989), and factors influencing the toxicity of the chemical, including genetic polymorphisms, life stage, gender, nutritional status, and concurrent exposure to other agents or environmental factors (NRC, 1999). More recently, the cooking method has also been observed to significantly influence As intake (Rahman et al., 2018).

Most studies on total arsenic (tAs) and its chemical forms have been performed mainly on raw rice grains (see e.g., Raber et al., 2012, Otero et al., 2016, Nunes & Otero, 2017, Chen et al., 2016, Dos Santos et al., 2017); however, its presence in cooked rice is worth considering, since this is the form in which it is consumed by the population (Jitaru et al., 2016).

It has been shown that the concentration of total arsenic content and of arsenic species may be altered during the preparation of food for human consumption. The number of studies on this regard is still limited and the observed uncertainty is high, as shown in Table S1 (Supplementary Material—main studies published over the last decades about the effect that cooking methods may have on arsenic content of rice). These studies considered the following sources of variability: (i) the arsenic content in the water used for cooking (no arsenic; low arsenic content; high arsenic content); (ii) the effect of rinsing the raw rice; and (iii) water-to-rice ratio (ranging between 1:2 and 1:10). Percentage of total arsenic remaining after cooking was the main indicator, with only a small fraction including speciation. Very large intra-study uncertainty is common, sometimes with percentages differing up to four times between the lowest and highest reported values (see, e.g., Althobiti et al. (2018)). Inter-study uncertainty is even higher, with difference in percentages of up to nine times for rice cooked under the same conditions.

Despite these uncertainties, some general conclusions are possible. Rinsing the rice before cooking can reduce total arsenic content between 78 and 97% of that of the untreated rice (Gray et al., 2015; Sharafi et al., 2019a), and between 75 and 91% for inorganic arsenic (Gray et al., 2015). When low arsenic water is used for cooking and the excess water is discarded (namely for water-to-rice ratios above 1:4), the cooked rice will have less total and inorganic arsenic than in the unprepared rice (Mandal et al., 2019; Sharafi et al., 2019b). The mean arsenic content of cooked rice decreases as the water-to-rice ratio increases, as long as excess water is discarded, indicating a clear dilution effect. Cooking with excess water is without doubt the best method to reduce exposure to arsenic. Ratios of 1:6 and above can reduce total and inorganic arsenic concentrations to about half of those in raw rice.

When water rich in arsenic is used for cooking, the final product will be enriched in the substance, as almost all the arsenic present in contaminated cooking water may be retained during boiling of rice (FAO/WHO, 2011). In this case, the different studies show high variability, being strongly affected by the studied concentrations in cooking water.

If the rice is cooked until all water evaporates at boiling temperature, no relevant alteration in arsenic concentrations should be observed, apart from a small conversion of species (Gray et al., 2015; Raab et al., 2009). Conversion to other arsenicals during food preparation has been observed and may be significant after cooking at temperatures above 150 °C (Van Elteren and Šlejkovec, 1997), which may occur in some cooking treatments in which the food surface is in direct contact with the source of heat (grilling, frying, or baking) (Devesa et al., 2008).

The above observations agree with those of Bundschuh et al. (2012) and Cubadda et al. (2017) who identify the cooking method as maybe the most important process affecting both total arsenic concentrations and arsenic speciation. More research in this area is still necessary, with specific focus on iAs and on ready-to-eat food, since preparation and cooking can significantly affect bioaccessibility (Cubadda et al., 2017).

As mentioned, recent studies have mainly analyzed the presence of total As (tAs) and of its inorganic forms (iAs) after different rice-rinsing and rice-cooking processes (Mandal et al., 2019), but few studies have considered organic species of As (oAs), which are considered less toxic than iAs (Rasheed et al., 2018).

Taking into account that As toxicity depends essentially on the concentration of its chemical forms, the main aim of this study was to determine the effect of grain rinsing and boiling on the concentration of tAs and of its inorganic (iAs) and organic (oAs) forms in market basket samples of rice from Ecuador and Spain. For this purpose, five treatments were applied to brown and polished rice samples, combining rinsing in different volumes of water and different cooking methods (boiling with or without excess water). After each treatment, As was extracted and contents of tAs and of its inorganic (AsIII, AsV) and organic (dimethylarsinic acid, DMA; monomethylarsonic acid, MMA; and arsenobetaine, AsB) forms were determined.

Materials and methods

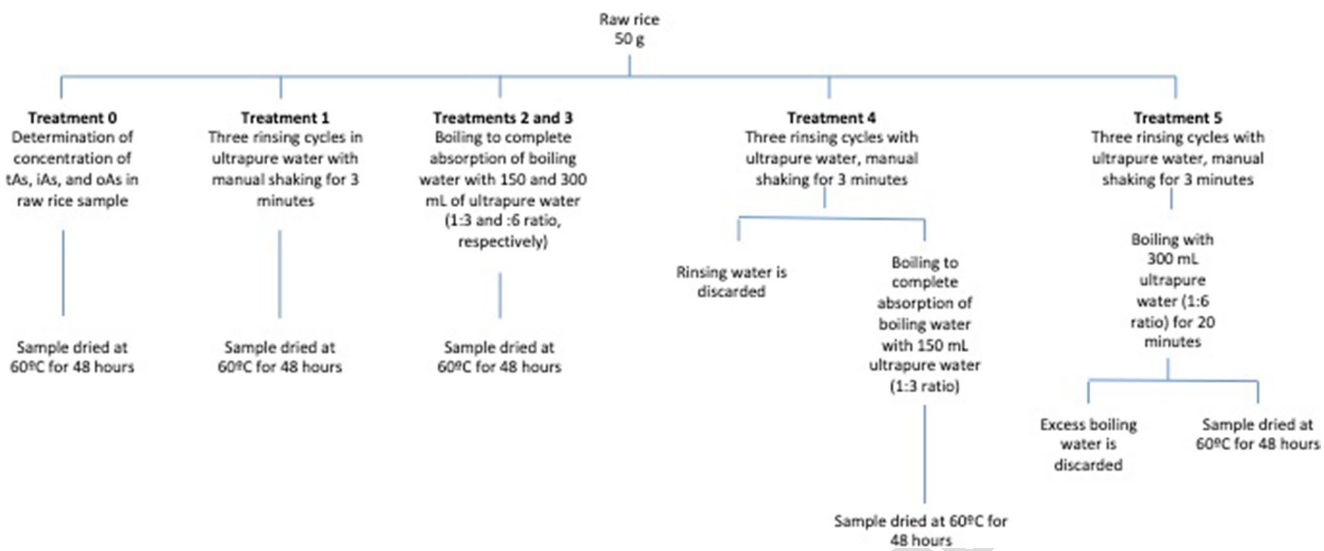
Treatment of rice grains

In practical studies, the statistics of the data are not known a priori, so the sample size is chosen parsimoniously to accommodate the available resources while complying with objectives of the experiment. Small sample sizes can increase the likelihood of a type II error skewing the results, which decreases the power of the study; but as the sample size grows above a certain size, the power of the test also increases, identifying small, impractical effects. As so, the option here was to start with a parsimonious sample size.

Seven market basket samples of rice from Ecuador (2 samples) and Spain (5 samples), of which 4 samples corresponded to polished rice and 3 to brown rice, were subjected to six different treatments to determine the effect of rinsing and cooking methods on concentration of tAs and its species.

Treatments were designed considering previous studies (Jitaru et al., 2016, Mihucz et al., 2007, Naito et al., 2015, Raab et al., 2009; Fig. 1). For this, 50 g of rice was subjected to the following treatments:

- Treatment T0: Concentrations of tAs and its inorganic (iAs) and organic forms (oAs) in raw rice samples were determined.



Q6 **Fig. 1** Rinsing and cooking treatments applied to rice samples

- Treatment T1: Samples were subjected to three rinsing cycles with 300 mL Milli-Q water with manual shaking for 3 min.
- Treatment T2: Samples were directly boiled in 150 mL Milli-Q water (1:3 ratio) to complete absorption of the boiling water, with no prior rinsing.
- Treatment T3: Similar to treatment 2, but using 300 mL Milli-Q water (1:6 ratio).
- Treatment T4: Samples were subjected to three rinsing cycles with 300 mL (1:6) Milli-Q water with manual shaking for 3 min; rinsing water was then discarded, and rice was subjected to boiling to complete absorption in 150 mL (1:3) Milli-Q water.
- Treatment T5: Samples were subjected to a rinsing process similar to the one described in the previous item (treatment T4), and the rice was subsequently boiled in excess Milli-Q water (300 mL) for 20 min at 150 °C, discarding excess boiling water.

After each treatment, samples were dried at 65 °C to constant weight (~ 48 h), ground in an agate mortar, and stored in polyethylene bags at room temperature until analysis.

Determination of As concentration in grain rice

The content of tAs was determined in 0.5–1.0 g of previously ground samples. Samples were digested in a mixture of HNO_3 and H_2O_2 (Suprapur) (Meharg and Rahman, 2003): 5 mL HNO_3 (65%), 1 mL H_2O_2 (33%), and 5 mL Milli-Q water (*w/v*), and were left to rest overnight. Tubes were subsequently placed in a sample preparation block (Perkin Elmer SPB 48–50) at 95 °C for 3 h. The extract was filtered by 0.20 µm. The total As content was determined by an ICP-MS system (Agilent Technologies, Palo Alto, CA, USA).

Partitioning of As was carried out on 0.50 g of sample (dry weight), and it was extracted with 15 mL of 0.28 M HNO_3 (65%, Merck) by heating the samples at 95 °C for 90 min. The samples were then centrifuged at 10,000 rpm at 4 °C for 20 min. The supernatant was then filtered through a 0.45-µm filter and conserved at − 20 °C until analysis. Inorganic As (iAs: ΣAsIII, AsV) and organic As (oAs: Σ (DMA, MMA, AsB)) were analyzed by HPLC (Varian Prostar, Spectralab Scientific, Toronto, Canada) coupled to an ICP-MS system (Varian 820-MS).

Concurrently, the certified reference material (CRM) 1568b, rice flour, by NIST (USA), was analyzed. Mean values obtained for the different arsenic species were iAs: 0.109 ± 0.038 mg/kg; DMA: 0.218 ± 0.093 mg/kg; and MMA: 0.018 ± 0.001 mg/kg, corresponding to a 107%, 113%, and 118% percentage of recovery, respectively. The detection limit (DL) was 3.75 µg/kg for inorganic forms and 1.35–4.35 µg/kg for organic forms. The mean content and percentage of recovery of tAs in the CRM ($n = 4$) was 0.349 ± 0.102 mg/kg and 123%, respectively.

Methodology for ELTR estimation

Estimated daily intakes (EDI) for lifetime exposure were calculated by assessing the amount of iAs to which an individual is exposed per day and per kilogram of body weight (Eq. 1). Q7

$$EDI = \frac{iAs \cdot IR}{BW} \quad (1)$$

where iAs is the concentration of inorganic arsenic in rice (µg/kg), IR is the ingestion rate (kg/day), and BW is the body Q8 weight (kg). Ingestion rates for adults were obtained from the Food and Agriculture Organization "Food balance sheets (FAO, 2018). Anthropometric data were obtained from National Institute of Statistics in Ecuador (INEC, 2014) and from the recent nutrition study for the Spanish population (Bartrina and Rodrigo, 2018).

No provisional tolerable daily intake is currently accepted for inorganic arsenic: the World Health Organization concluded that the former value of 2.1 μg/kg day was no longer considered health-protective (FAO/WHO, 2011). Estimated lifetime health risks (ELTR) were calculated; these are proportional to EDI, and the proportionality coefficient is known as the cancer slope factor (CSF), equal to 1.5×10^{-3} $(\mu g/kg\ day)^{-1}$ (USEPA, 1995):

$$ELTR = EDI \times CSF \tag{2}$$

Statistical analysis

One- or two-way analyses of variance with Fisher's HSD post hoc test were performed to test differences between means, with a significance level of $\alpha = 0.10$. This significance level was deliberately chosen, instead of the more traditional 0.05, as the authors were willing to accept a 10% chance of incorrectly finding that an innocuous and very cheap treatment is beneficial for human health when it is not. From a statistical point of view, $\alpha = 0.05$ or 0.1 is equally valid (Koch and Link, 1971; Gibbons and Coleman, 2001).

The Doornik-Hansen test was used to test normality. Concentration values were log-transformed before statistical testing.

Results

Effect of rinsing and/or boiling on total arsenic content

Significant differences in tAs concentrations were found between treatments ($F(5,36) = 2.4$, $p < 0.10$) for polished rice. Cooking with no previous rinsing and without removing excess water (treatments T2 and T3) did not significantly reduce the content of tAs in cooked rice compared with raw rice (Fisher's HSD, $n = 36$, $p > 0.10$). Contrarily, rinsing before cooking can efficiently reduce tAs concentrations in cooked rice (Fisher's HSD, $n = 36$, $p < 0.10$). The different treatments ordered by increasing efficiency would be T2 > T4 > T5 (Fig. 2).

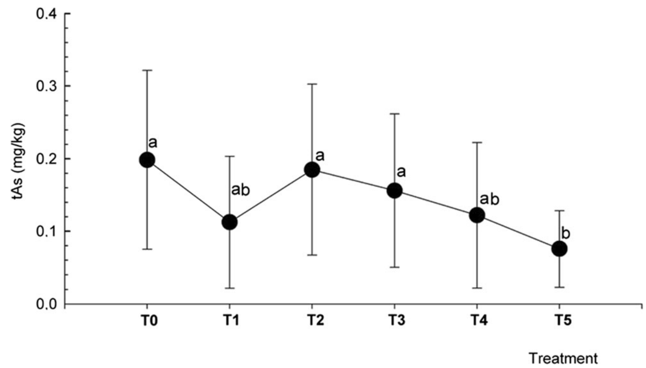


Fig. 2 Decreases in total arsenic concentration by type of treatment for the whole set of samples. Different letters indicate significant differences at the $p < 0.10$ level

Treatment T0 The concentrations of tAs in rice from Spain (polished rice 0.163–0.234 mg/kg, $n = 4$; brown rice 0.231–0.438 mg/kg, $n = 2$) were higher than for those from Ecuador (polished rice 0.090 mg/kg, integral rice 0.165 mg/kg, $n = 1$). These results are consistent with those obtained in previous studies suggesting that rice from Spain usually shows high tAs contents (see, e.g., Meharg and Zhao, 2012, Signes-Pastor et al., 2016, Torres-Escribano, et al., 2008). Nevertheless, it is worth noting the low concentrations of tAs obtained for Spanish organic rice (0.067 mg/kg) (Table 1). By type of rice, the concentration of tAs in raw polished rice (T0) (0.138 ± 0.076 mg/kg^{-1}) was lower than in brown (0.278 ± 0.142 mg/kg) (Table 1).

Treatment T1 (rinsing only) Treatment T1, along with treatment T5, was the process that removed the greatest amount of tAs. The concentration of tAs in polished rice after rinsing (T1) ranged between 0.024 and 0.097 mg/kg, while for T5, it ranged between 0.02 and 0.075 mg/kg. These values correspond to a 39% and 59% reduction, respectively (Table 1). The concentration of tAs ranged between 0.025 and 0.097 mg/kg for polished rice and between 0.095 and 0.310 mg/kg for brown rice.

The treatment consisting in boiling with 150 mL to complete absorption with no previous rinsing (T2) showed an 8.4% reduction in tAs (range 4–18%). The highest percentage of reduction (17%) was found for one polished rice sample from Spain. The concentration of tAs after treatment T2 ranged between 0.081 and 0.193 mg/kg for polished rice and between 0.151 and 0.423 mg/kg for brown rice.

The treatment consisting in boiling with 300 mL to complete absorption with no previous rinsing (T3) led to a 30% reduction in the concentration of tAs (range 7–49%). The concentration of tAs ranged between 0.084–0.122 mg/kg for polished rice and between 0.113–0.362 mg/kg for brown rice.

Rinsing and boiling to complete absorption (T4) decreased tAs content by 53% in polished rice, with the highest percentage of reduction (74%) found in rice from Spain. The concentration of tAs ranged between 0.060 and 0.074 mg/kg for polished rice and between 0.128 and 0.0338 mg/kg for brown rice.

Table 1 Total As concentration (mg/kg) by treatment

Site	Type of rice	Treatment					
		T0—raw rice	T1—only washing	T2—cooked to dryness (1:3)	T3—cooked to dryness (1:6)	T4—washing and cooked to dryness (1:3)	T5—washing and cooked with excess water (1:6)
Ecuador	Brown	0.165	0.101	0.151	0.113	0.128	0.081
	Polished	0.090	0.097	0.810	0.084	0.065	0.075
Spain	Polished	0.233	0.080	0.192	0.122	0.060	0.049
	Brown	0.231	0.095	0.219	0.171	0.128	0.076
	Brown	0.437	0.310	0.422	0.362	0.338	0.184
	Polished	0.163	0.080	0.153	0.084	0.074	0.042
	Polished	0.067	0.025	0.073	0.072	0.061	0.020

Rinsing and boiling in excess water (T5) was the treatment with the greatest reduction in tAs content, with a mean value of 63% and significantly lower tAs content compared with raw grain for this set of samples (Fig. 2). The greatest percentage of reduction was found in polished rice from Spain (79%). The concentration of tAs decreased to values of 0.020–0.075 mg/kg for polished rice and 0.076–0.184 mg/kg for brown rice.

Effect of rinsing and/or cooking on the content of forms of arsenic

The statistically significant differences found among arsenic concentrations in rice were due to changes in contents of both iAs and oAs between raw rice (T0) and rice subjected to treatment T5 ($F(5,247) = 3.1$, $p < 0.01$; Fisher's HSD, $n = 247$, $p < 0.10$) (Fig. 3).

Treatment T0 The content of iAs forms varied substantially both between countries and between types of rice (Table 2 and Table S2). The concentration of iAs was higher in Ecuadorian than in Spanish rice, while the concentration of oAs was higher in brown rice from Ecuador (0.166 mg/kg, $n = 1$), followed by polished rice from Ecuador (0.135 mg/kg, $n = 1$), brown rice from Spain (median value 0.118 mg/kg, $n = 2$), and polished rice from Spain (median value 0.059 mg/kg, $n = 4$).

The predominant iAs form was As(III), with values ranging from 0.027 to 0.131 mg/kg, except in brown rice from Ecuador, where As(V) was slightly higher. The concentration of As(V) ranged between 0.023 and 0.088 mg/kg for the whole set of samples. The highest oAs concentration was found in brown rice from Spain (0.127 mg/kg), whereas the lowest was found in polished rice from Ecuador (0.024 mg/kg); meanwhile, the concentration of oAs in polished rice from Spain showed similar values to those found in brown rice from Ecuador (0.062 and 0.068 mg/kg, respectively). The predominant oAs form was DMA, whose concentrations represented 85–100% of oAs. MMA and AsB showed very low concentrations, which were below the detection limit in most cases (Table 2, Table S1, and Fig. 3).

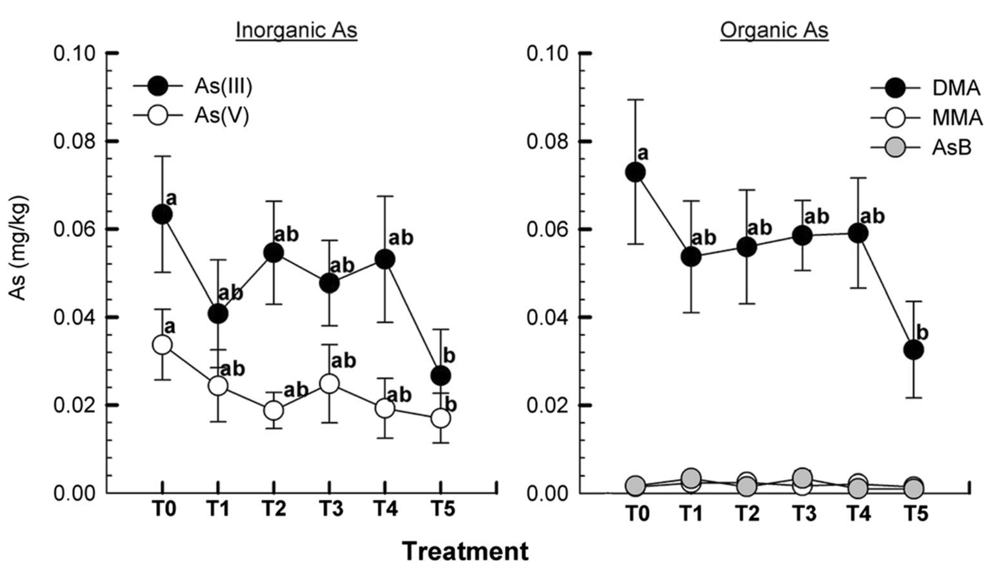


Fig. 3 Decrease in the concentration of arsenic forms by type of treatment for the whole set of samples. Different letters indicate significant differences at the $p < 0.01$ level

Table 2 Concentration of As species by rice milling step (brown and polished) and by treatment

		As(III)		As(V)		DMA		MMA		AsB	
		Brown mg/kg	Polished	Brown	Polished	Brown	Polished	Brown	Polished	Brown	Polished
T0	Mean ± SD	0.09 ± 0.04	0.05 ± 0.04	0.05 ± 0.04	0.026 ± 0.004	0.10 ± 0.07	0.05 ± 0.03	0.003 ± 0.002	< LOD	0.005 ± 0.004	< LOD
	Median	0.078	0.040	0.038	0.025	0.065	0.051	0.003		0.005	
T1	Mean ± SD	0.11 ± 0.09	0.02 ± 0.01	0.04 ± 0.03	0.014 ± 0.007	0.08 ± 0.05	0.04 ± 0.01	0.003 ± 0.002	0.002 ± 0.002	0.006 ± 0.005	0.003 ± 0.002
	Median	0.070	0.020	0.047	0.013	0.052	0.038	0.002	0.002	0.006	0.002
T2	Mean ± SD	0.08 ± 0.03	0.05 ± 0.03	0.03 ± 0.02	0.014 ± 0.004	0.08 ± 0.04	0.04 ± 0.02	0.003 ± 0.002	0.001 ± 0.001	0.001 ± 0.001	0.003 ± 0.001
	Median	0.075	0.034	0.030	0.013	0.060	0.042	0.003	0.001	0.002	0.002
T3	Mean ± SD	0.07 ± 0.04	0.04 ± 0.03	0.06 ± 0.04	0.013 ± 0.006	0.07 ± 0.04	0.05 ± 0.02	0.007 ± 0.007	0.001 ± 0.001	0.002 ± 0.001	0.002 ± 0.001
	Median	0.067	0.033	0.057	0.011	0.074	0.046	0.007	0.001	0.002	0.002
T4	Mean ± SD	0.09 ± 0.04	0.05 ± 0.04	0.03 ± 0.03	0.015 ± 0.008	0.08 ± 0.04	0.03 ± 0.02	0.005 ± 0.004	< LOD	< LOD	0.001 ± 0.001
	Median	0.067	0.054	0.024	0.015	0.062	0.021	0.005		n.a	0.002
T5	Mean ± SD	0.04 ± 0.02	0.02 ± 0.04	0.03 ± 0.02	0.009 ± 0.005	0.07 ± 0.05	0.02 ± 0.03	0.003 ± 0.002	< LOD	0.001 ± 0.001	< LOD
	Median	0.039	0.006	0.037	0.008	0.043	0.017	0.004		0.002	

LOD limit of detection, *SD* standard deviation, *n.a.* not analyzed

Treatment T1 The mean concentration of iAs was 0.031 mg/kg ($n = 4$) for polished rice from Spain and 0.159 mg/kg ($n = 2$) for brown rice, corresponding to a mean percentage of reduction of 50% and 8%, respectively (Supplementary Table 1, Fig. 3). The concentration of iAs in samples from Ecuador was 0.040 mg/kg ($n = 1$) in polished rice and 0.142 mg/kg ($n = 1$) in brown rice, corresponding to a 70% and 14.5% reduction, respectively. Mean reduction in iAs content with treatment T1 was 40% both for AsIII and for AsV. The concentration of oAs (DMA) was reduced by 15–37%. Content of oAs was higher in rice from Spain (polished 0.045 mg/kg; brown 0.104 mg/kg) than in rice from Ecuador (polished 0.028 mg/kg; brown 0.058 mg/kg) (Table 2).

Treatment T2 Reduction in iAs ranged between 14.5 and 49%, with higher iAs content in rice from Ecuador (polished 0.105 mg/kg; brown 0.097 mg/kg, $n = 1$, Supplementary Table 1) than in rice from Spain (polished 0.042 mg/kg, $n =$ 4; brown 0.109 mg/kg, $n = 2$). The mean percentage of reduction after this treatment was 21.5% for AsIII and 47% for AsV; therefore, the concentrations of AsIII reached higher values than AsV in all the studied types of rice. The content of oAs (mainly DMA) decreased by 24–38%, with median values of 0.052 mg/kg for polished rice from Spain, 0.106 mg/kg for brown rice from Spain, 0.052 mg/kg for brown rice from Ecuador, and 0.018 mg/kg for polished rice from Ecuador (Fig. 3, Table 2, Supplementary Table 1).

Treatment T3 The results for iAs concentration were similar to those found with treatment 2. The content of iAs decreased by 20–39%, while oAs decreased by 4–31%. The concentration of iAs was higher in rice from Ecuador (polished 0.108 mg/kg, brown no data, Supplementary Table 1) than in rice from Spain (median values: polished 0.038 mg/kg, $n = 4$; brown 0.124 mg/kg, $n = 2$). The percentage of reduction was 25% for AsIII and 52% for AsV. The concentration of oAs ranged between 0.058 mg/kg for polished rice from Spain and 0.082 mg/kg for brown rice from Spain. For polished rice from Ecuador, the concentration of oAs was 0.025 mg/kg.

Treatment 4 The reductions of iAs ranged between 8 and 65%. The median concentration of iAs was 0.029 mg/kg ($n = 4$) in polished rice from Spain and 0.117 mg/kg ($n = 2$) in brown rice from Spain, corresponding to a 55% and 8% decrease, respectively. For Ecuadorian rice, the percentages of reduction of iAs reached values around 20%, with concentrations of 0.131 mg/kg for brown rice and 0.103 mg/kg for polished rice. Reductions in AsIII reached mean percentages of 29%, while for AsV, it was 46%. The percentages of reduction for oAs were similar to those for iAs (3–48%), with mean concentrations of 0.059 mg/kg in polished rice from Spain, 0.102 mg/kg I in brown from Spain, 0.013 mg/kg in polished rice from Ecuador, and 0.056 mg/kg in brown rice from Ecuador.

Treatment 5 This treatment showed the highest percentages of reduction of As (iAs 29–90%, oAs 4–85%), with significantly lower concentrations of iAs forms and DMA compared with those found in raw grain rice (Fig. 3). The median concentration of iAs in rice from Spain was 0.006 mg/kg for polished rice and 0.064 mg/kg for brown rice, while in rice from Ecuador, it was 0.096 mg/kg for polished rice and 0.086 mg/kg for brown rice. This was the only treatment in which the percentage of reduction was higher for AsIII (62.5%) than for AsV (51%).

Mean oAs concentrations were 0.044 mg/kg in polished rice from Spain and 0.089 mg/kg in brown rice from Spain. For Ecuadorian rice, the obtained oAs concentrations were 0.006 mg/kg for polished rice and 0.034 mg/kg for brown rice.

Estimated daily intakes and estimated lifetime health risks

The low concentrations of iAs in Spanish rice and the relatively low importance of rice in the diet of the Spanish population (22.8 g raw rice/day; FAO, 2018) result in an estimated daily intake (EDI) of polished rice of 19.4 ng/kg day for raw rice (T0); 10.0 ng/kg day for T1; 9.6 ng/kg day for T2–T4; and 3.2 ng/kg day for T5. The corresponding ELTR values are in the same order as above: 2.9×10^{-5}, 1.5×10^{-5}, 1.4×10^{-5}, and 4.8×10^{-6}. Pre-rinsing the rice or cooking it using the traditional method in Spain (T2–T4) seems to be sufficient to reduce ELTR by 50% compared with the assessed concentration in raw rice. The T5 cooking method reduces ELTR by 83% compared with T0 and therefore constitutes the preferable method.

Discussion

Arsenic in cooked rice

The results of tAs concentration in rice from Ecuador and Spain are consistent with previously published data that suggest that iAs content in rice from Spain is usually high (Torres-Escribano et al., 2008, Meharg and Zhao,2012, Signes-Pastor et al., 2016); in fact, it was higher than in rice from Ecuador. However, the predominant species of As in Spanish rice is DMA, whose toxicity is lower than that of inorganic forms of As (Suriyagoda et al., 2018).

Content of tAs and iAs was also higher in brown than in polished rice, consistently with the fact that As is mainly concentrated in the outermost portion of the grain (pericarp and aleurone layer), which is removed in polished rice (Meharg et al., 2008, Zhu et al., 2008, Carey et al., 2010).

Preliminary washing (treatment 1) removed 39–59% of the total arsenic, 40% of the inorganic arsenic, and between 15 and 37% of organic forms. These values are about 10% above the values reported by other authors (see Table S1). Raab et al. (2009) investigated total arsenic and inorganic arsenic in different rice types (basmati, long-grain, polished (white), and wholegrain (brown)) after being cooked in non-contaminated water. The effects of rinse washing, low water volume (rice-to-water ratio of 1:2.5), and high water volume (rice-to-water ratio of 1:6) cooking were investigated. Rinse washing was effective at removing about 10% of the total and inorganic arsenic from basmati rice, but was less effective for other rice types. Sengupta et al. (2006) tested the three major rice-cooking procedures in practice globally, using low arsenic water (tAs < 0.003 mg/L). Preliminary washing removed 28% of the rice arsenic. The results were not influenced by water source (tube well, dug well, pond, or rain), cooking vessel (aluminum, steel, glass, or earthenware), or the absolute weight of rice or volume of water. Naito et al. (2015) studied the traditional Japanese rice cooking method by cooking washed rice until dry (rice-to-water ratio of 1:1.4). Again, rinse washing was effective at removing 16–24% of tAs and 12–29% of iAs.

The most commonly used rice-cooking method in Spain and in Ecuador is using a volume of water that will result in all the water being absorbed or evaporated (Torres-Escribano et al., 2008). Rice cooked by boiling to complete absorption (treatments 2 and 3) constituted the least effective treatment to remove As from rice (Fig. 3), which is also consistent with results by other authors (Sengupta et al., 2006, Torres-Escribano et al., 2008, Raab et al., 2009; Ackerman et al., 2005) (see Table S1 for a more exhaustive list). Contrarily, this cooking method may even result in an additional increase in As content with respect to raw rice if the boiling water has an abnormally high content, as occurs in many South Asian countries (e.g., Bangladesh and India) (Meharg and Zhao, 2012; Mandal et al., 2019).

Significant decreases in tAs and iAs content in rice grain were only obtained when rice was rinsed and cooked in excess water (1:6 ratio; treatment 5). The mean percentage of total arsenic removed for the whole set of samples (62%) is in agreement with results obtained by previous studies under similar rinsing and cooking conditions: 57% (Sengupta et al., 2006), 54% (Mihucz et al., 2007), and 65% removal (Raab et al., 2009). Nevertheless, it is also worth highlighting that simply rinsing rice grains before cooking leads to a substantial removal of tAs, particularly of iAs, going from a ratio of $iAs/tAs_{raw\text{-}rice} = 0.49$ to $iAs/tAs_{T1} = 0.44$ for the whole set of samples. This is mainly due to the fact that iAs is accumulated in the outermost portion of the grain, while DMA is found in the inner endosperm (Carey et al., 2010). Raab et al. (2009) also found that rinsing and cooking with excess water specifically reduces iAs but has no effect on DMA.

Risk assessment: estimated excess lifetime risk

Previous studies have estimated daily intake (EDI) for the Ecuadorian population as a whole, which is almost twice that of Europe but from one-half to one-third that of Brazil, Bangladesh, and India. Estimated excess lifetime risk (ELTR) for adults was 3.0×10^{-4}, while for infants, it varied between 10×10^{-4} in rural areas and 20×10^{-4} in urban areas (Nunes & Otero, 2017). Nevertheless, these estimations were based on iAs content in raw grain. However, considering the percentage of iAs that is lost with each treatment, EDI and ELTR decreased substantially when calculated for cooked rice. Thus, simply rinsing rice grains before cooking reduced ELTR by 50%, while rinsing and cooking in excess water led to an 83% decrease. This scenario is more realistic and less dramatic than calculations based on As contents in raw grain.

Conclusions

Rinsing and boiling rice in excess water and simply rinsing rice grains with As-free water are two efficient methods to significantly reduce As intake in the population. According to our results, the rinsing of rice before cooking can reduce the content of total and of inorganic arsenic by a substantial amount (up to 40–59% of total arsenic and 40% of inorganic arsenic). When rinsing and cooking in excess water are used, the reductions are even more pronounced, of up to 62% for total and inorganic arsenic. This observation can have significant impacts on risk estimates as exposure to the hazard is reduced by the same amount.

In summary, rinsing rice grains before cooking can reduced health risk by 50%, while rinsing and cooking in excess water can promote a reduction of 83% in the risk. This scenario is more realistic and less dramatic than calculations based on As contents in raw grain.

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