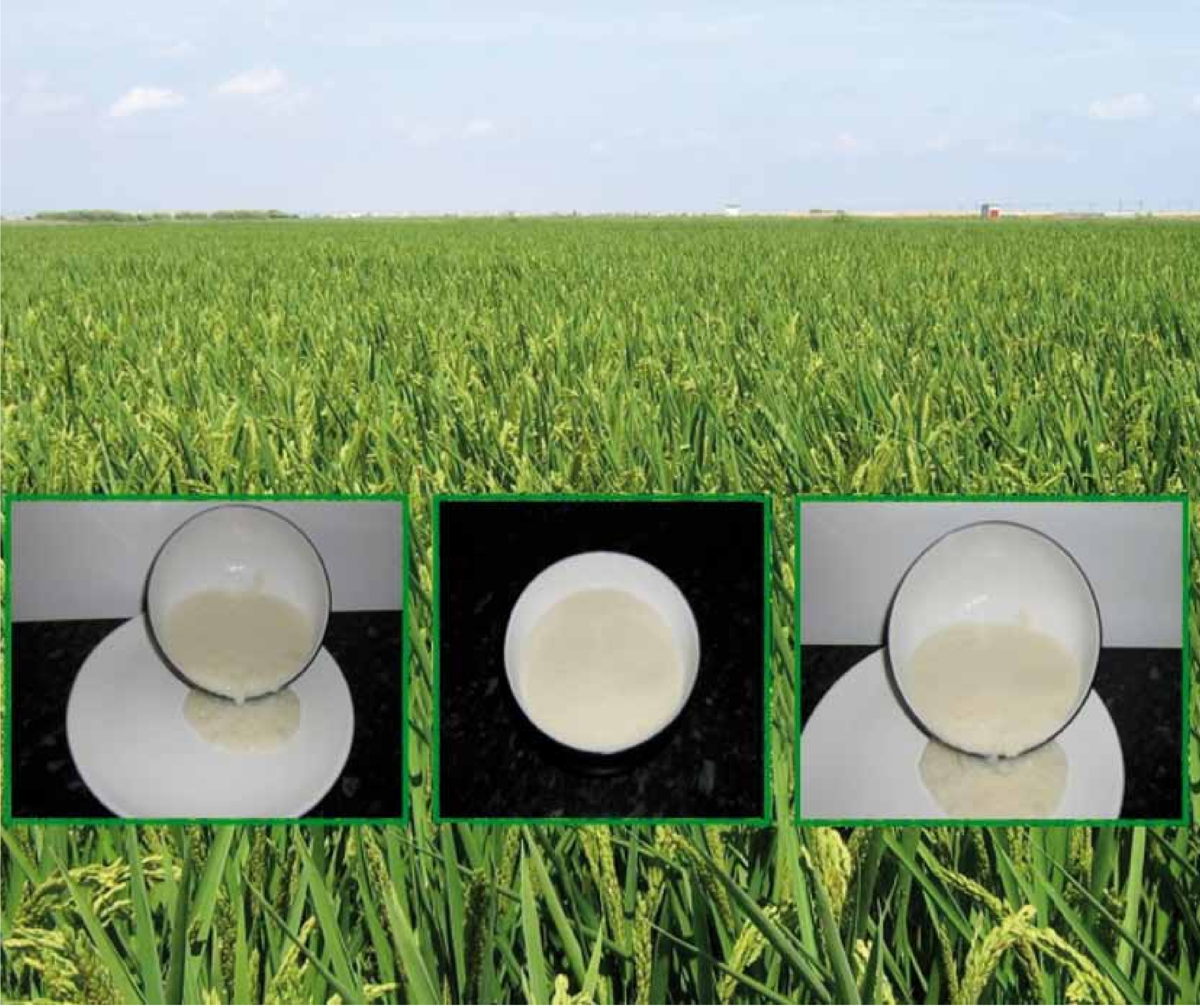
Journal of Environmental Monitoring

Cutting-Edge Research on Environmental Processes & Impacts

www.rsc.org/jem

Volume 11 | Number 11 | November 2009 | Pages 1889–2092



ISSN 1464-0325

RSC Publishing

1464-0325(2009)11:11;1-9

Arsenic speciation in Japanese rice drinks and condiments

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Received 16th June 2009, Accepted 7th September 2009
First published as an Advance Article on the web 9th October 2009
DOI: 10.1039/b911615j

Rice has been demonstrated to be one of the major contributors to inorganic arsenic (i-As) intake in humans. However, little is known about rice products as additional source of i-As exposure. In this study, misos, syrups and amazake (a fermented sweet rice drink) produced from rice, barley and millet were analysed for total arsenic (t-As) and a subset of samples were also analyzed for As speciation. Rice based products displayed a higher i-As content than those derived from barley and millet. Most of the t-As in the rice products studied was inorganic (63–83%), the remainder being dimethylarsinic acid. Those who regularly consume rice drinks and condiments, such as the Japanese population and those who follow health conscious diets based on the Japanese cuisine, could reach up to 23% of the World Health Organization's Provisional Tolerable Daily Intake of i-As, by only consuming these kinds of products. This study provides a wide appreciation of how i-As derived from rice based products enters the human diet and how this may be of concern to populations who are already exposed to high levels of i-As through consumption of foods such as rice and seaweed.

Introduction

A number of surveys have reported the levels of total arsenic (t-As) and inorganic As (i-As) in rice.[1–4] Recent studies have also shown that As concentration may change due to rice processing operations, such as dehusking and cooking.[5–7] In addition, i-As has been reported in rice products such as baby rice, rice milk, rice crackers, rice wines, mirins and vinegars, puffed rice cereals and cereal bars,[8–10] which suggest that all products made with rice may pose an additional source of exposure to i-As. Consumption of rice has been associated with increased urinary excretion of As.[11] It has also been reported that the urine of Japanese populations, not occupationally exposed to As, contains a mean value of 173 μg t-As/l.[12] This value is quite high when compared with 8.0 μg t-As/l[13] and 65.4 μg t-As/l[14] in urine from European and US populations, respectively. These values are for people who are not occupationally exposed to arsenic. There is no doubt that the higher consumption of rice, rice-based products, seaweed could be a reasonable explanation for the higher urinary As levels in Japanese populations.[12] Although, some work has been reported on i-As content in rice and seaweed samples,[1–4,15] yet no studies have characterised rice condiments and drinks, such as amazake, miso or syrups, which are an important part of the Japanese diet. These products are also at the centre of Western health conscious diets based on the Japanese cuisine, such as vegan, macrobiotic and other whole food regimens.[16]

Misos can be made using rice, barley, wheat grain mixed with soya, and fermented using fungal cultures of *Aspergillus oryzae*.[17] Amazake, a rice drink, is also prepared from the same fungus. The aim of the present study was to examine the t-As and i-As contents of a range of rice condiments and amazake and estimate whether they contribute significantly to the daily intake of i-As.

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Experimental

Analytical grade nitric acid (HNO_3) (70%) was obtained for Fisher Scientific (UK). Monosodium arsenate (Na_2HAsO_4) and sodium arsenite ($NaAsO_2$) of reagent grade were purchased from Merck (UK). Methylarsonic acid (MA) was purchased from Chem Sevice MC (UK) and dimethylarsinic acid (DMA) was

Environmental impact

Arsenic concentration in rice can be very high compared to other cereals due to the nature of the plant and its growth conditions as well as due to environmental pollution such as the use of arsenic containing pesticides and contaminated irrigation water. This study provides information about total arsenic and arsenic species in rice-based products such as miso, syrup and amazake to better understand their health impact. Consuming such products can provide up to 23% of the Provisional Tolerable Daily Intake of arsenic. This is of concern to certain population who already consume arsenic rich products (rice and seaweed). Similar products from barley or millet, which contain lower levels of inorganic arsenic, can be alternatives until low arsenic containing rice is available.

purchased from Sigma Chemicals (UK). As internal standard, Indium with ammonium hydrophosphate ($NH_4H_2PO_4$) and ammonium dihydrogen orthophosphate (NH_4NO_3) from BHD (UK) was prepared.

All commercial products analysed were imported from Japan and were purchased, in triplicate for each type, from health shops in the UK. There were 8 samples of commercial rice and barley misos, 8 samples of commercial barley and rice syrups, one sample of commercial *Aspergillus oryzae* inoculum (koji rice), two samples of commercial amazake and one sample of homemade amazake.

The homemade amazake was prepared in triplicate batches, following the instructions provided: 50 g of Italian rice was boiled with 300 ml of deionised water; then, 100 g of koji rice was added and mixed with the cooked rice. The mixture was incubated overnight at 30 °C under anaerobic conditions. The Italian rice used in this study was analyzed previously and showed a concentration of 0.15 mg/kg and 0.11 mg/kg for t- and i-As, respectively.[4]

For t-As measurements, 0.5 g of fresh samples were digested in 2 ml of nitric acid using a published methodology.[18,19] NIST SRM 1568a rice flour was used as certified reference material (CRM). Quality controls of CRM and blanks were run with each digest set.

Foster *et al.* (2007)[20] showed that As species in marine extracts were stable after microwave-assisted extraction using 2% nitric acid. A similar method, using 1% nitric acid was utilised in rice with good recoveries and minimal inter-conversion of As species.[21] Nitric acid (1%) was also used for extraction of As species as described below. Around 0.5 g of fresh sample was accurately weighed out into a 50 ml polypropylene digest tube and 10 ml of 1% nitric acid was added and left overnight. Samples were extracted in a microwave oven (CEM Mars 5, CEM Corp, Matthews, NC, USA). The temperature was raised first to 55 °C and held for 10 min; then raised to 75 °C and held for 10 min; finally the digestion was taken up to 95 °C and maintained for 30 min. Samples were cooled to room temperature and centrifuged at 3500 rpm for 1 h. The supernatant was filtered through 0.45 μm filter (Millipore) and kept at 4 °C until analysis. Quality controls of CRM and blanks were run with each of the extracts.

An ICP-MS spectrometer Agilent 7500 quadrapole (Agilent Technologies, UK) was used to determine the t-As concentration as described elsewhere[21,22] and As speciation was quantified by HPLC-ICP-MS. For As speciation, the chromatographic separation system consisted of a pre-column (11.2 mm, 12–20 mm) (Hamilton) and PRP-X100 10-mm anion exchange column (150 × 4.1 mm) (Hamilton). The mobile phase consisted of 6.66 mM ammonium hydrophosphate ($NH_4H_2PO_4$) and 6.66 mM ammonium nitrate (NH_4NO_3), adjusted to pH 6.2 using ammonia. Retention times for the As species were determined using a species mix comprising standard of 0.050 mg/l arsenite, DMA, MA and arsenate and they were 1.67, 2.42, 3.00 and 6.84 min, respectively.

Results and discussion

The recovery of t-As from NIST rice flour CRM was 0.27 ± 0.006 mg/kg (Table 1), 93% recovery of its certified value of 0.29 mg/kg. No CRM with certified speciation was available, but the NIST rice flour CRM speciation has been used in a number of studies, summarized by Williams *et al.* (2005),[23] and the characterisation of this CRM for this study (Table 1 and Fig. 1) was in agreement with these previous investigations.[23,24] The LOD for the As species were 0.006, 0.006 and 0.009 mg/kg dry weight for As (III), DMA and As (V), respectively.

A total of 60 samples (20 products × 3 replicates) were analyzed. They were classified within the following four categories: miso, syrup, amazake and fungal inoculum. Table 2 shows the average of three repetitions of As content (dry weight) in each samples belonging to the categories studied with their standard deviation and the percentages of their main ingredients. Miso products showed a range of t-As from 0.06 to 0.14 mg/kg and syrup products from 0.03 to 0.33 mg/kg. The highest t-As concentration (0.33 mg/kg) was found in rice sysrup, which also had the largest percentage of rice content (90%). Both rice miso and rice syrups showed higher levels of t-As than similar products prepared from barley. Furthermore, millet amazake had lower i-As compared to rice amazake. The lower i-As contents in cereals other than rice was not surprising because transfer of As from soil to grain has been reported to be an order of magnitude greater in rice than in wheat and barley.[18] Samples of rice miso (0.09–0.14 mg t-As/kg) and of rice syrups (0.10–0.33 mg t-As/kg) were within the ranges previously discussed. Rice used to prepare the samples was from Japan as indicated on food labels; Japanese rice has been reported to have an average of 0.19 mg t-As/kg.[4] Samples that do not indicate the origin of the rice are likely to be made with Japanese rice as well, as most of them belong to the same commercial brand. In addition, the t-As concentration in samples analysed agree with the total arsenic reported in a recent study[24] on Certified Reference Material "rice flour-unpolished" (No. 10a, 10b, 10c) derived from rice grown in different regions of Japan (0.17, 0.11, 0.15 mg t-As/kg). Furthermore, the percentages of i-As and DMA found in the samples analyzed in our study are similar to white rice flour from Japan.[24] Overall,

Table 1 Concentrations (mg/kg dry weight) of As species in rice fermented products

Product	Total As (mg/kg d wt)	Sum of speciation (mg/kg d wt)	Extr. Effic. (%)	DMA[a] (mg/kg d wt)	i-As (mg/kg d wt)
Brown rice miso	0.14 ± 0.006[b]	0.154 ± 0.008	107	0.044 ± 0.001	0.110 ± 0.009
Brown rice syrup	0.33 ± 0.006	0.281 ± 0.007	85	0.047 ± 0.001	0.234 ± 0.006
Amazake brown rice	0.12 ± 0.006	0.136 ± 0.007	113	0.033 ± 0.004	0.103 ± 0.014
Homemade amazake	0.13 ± 0.011	0.135 ± 0.011	103	0.031 ± 0.004	0.104 ± 0.008
Koji	0.12 ± 0.011	0.111 ± 0.007	92	0.041 ± 0.004	0.070 ± 0.002
NIST CRM 1568a	0.27 ± 0.006	0.262 ± 0.005	93	0.175 ± 0.005	0.087 ± 0.007

[a] No MA was found. [b] The standard errors are from triplicate analysis.

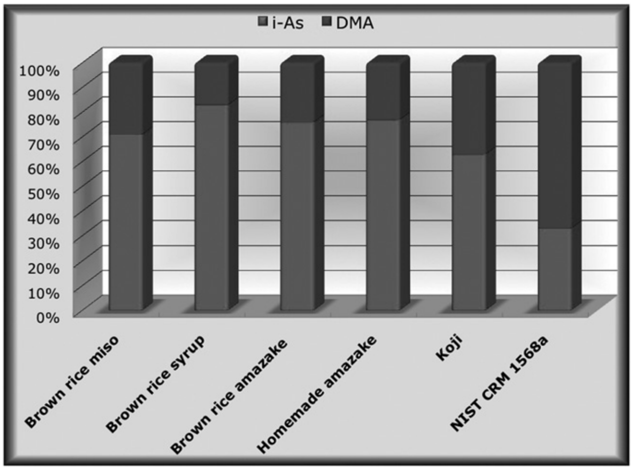

Fig. 1 Percentages of As species (i-As and DMA) in rice fermented products.

they are also similar to results for the brown rice with the exception that a very small percentage of MA (2–3%) was detected in their study[24] but not in ours. The reason for the absence of MA in our samples is not clear but the levels are so low that it may remain undetected or it could be due the use of a different variety of rice. However, what is clear from both studies is that Japanese rice and Japanese rice products contain a very high percentage of i-As. The fact that 56–74% of i-As in Japanese rice[24] is the more toxic As(III) is of particular concern for populations who consume these products along with other i-As rich foods. Although we are highly confident that Japanese rice has been used in the production of the foods analysed in our study, the possibility that Japanese rice grown in the USA or other parts of the world have been used as ingredients cannot be ruled out. Irrespective of the origin of the rice, the products contain high levels of inorganic arsenic which is the focus of this study. The highest levels of i-As in this study were found in the brown rice syrup (Table 1). This was expected because it has been previously shown that the level of i-As in brown rice can be much higher compared to white rice.[10] Brown rice, and wholegrain products in general, is very popular amongst the health conscious consumers in Western countries. Grain syrups, particularly rice, are used as an alternative to glucose and sucrose as sweeteners, particularly in products for the health conscious market[25] and for the Japanese population.

Both commercial rice amazake and homemade amazake showed similar t-As concentrations, 0.12 mg/kg and 0.13 mg/kg, respectively (Table 1). These concentrations may suggest that rice used for the commercial amazake had similar t-As concentration as the Italian rice (0.15 mg/kg) used to make the homemade version of the drink. As pointed out earlier, Japanese rice on average contains 0.19 mg t-As/kg, which is similar to Italian rice. Furthermore, the range of arsenic concentration in Italian and Japanese rice are similar; (0.07 to 0.33 mg t-As/kg for Italian rice and from 0.07 to 0.42 mg t-As/kg for Japanese rice).[4] This may offer a plausible explanation for the similarity in the arsenic content of the commercial and homemade (Italian) amazake.

Inorganic As (arsenate and arsenite) has been classified by the International Agency for Research of Cancer (IARC) as a human carcinogen.[26] Because of this, determination of As speciation in food is very important to realistically assess the risk

Table 2 Summary of t-As (mg/kg dry weight) in fermented products and the percentage of their moisture and main ingredients

				Main ingredients			
PRODUCTS	Average (mg/kg)	Standard Deviation	Moisture (%)	Rice (%)	Barley (%)	Soya (%)	Millet (%)
MISO							
[A] Barley miso	0.07[a]	0.02	36	0	48	32	0
[B] Brown rice miso	0.11	0.01	39	33	0	46	0
[E] Organic brown rice miso	0.14	0.01	47	40	0	n.a	0
[F] Organic barley miso	0.07	0.03	47	0	40	n.a	0
[K] Brown Rice miso	0.11	0.01	43	33	0	46	0
[K] Barley miso	0.07	0.02	43	0	38	35	0
[M] Barley miso	0.06	0.01	42	0	n.a	n.a	0
[N] Brown rice miso	0.09	0.01	42	n.a[b]	0	n.a	0
SYRUP							
[G] Organic rice syrup	0.10	0.01	15	72	n.a	0	0
[H] Brown rice syrup	0.33	0.01	16	90	n.a		0
[I] Organic brown rice syrup	0.08	0.01	15	—[c]	—	—	—
[J] Organic rice syrup	0.12	0.01	15	—	—	—	—
[O] Organic barley syrup	0.04	0.01	17	0	n.a	0	0
[P] Organic corn and barley syrup	0.03	0.01	15	0	18	0	0
[Q] Organic barley syrup	0.06	0.02	17	—	—	—	—
[R] Barely syrup	0.13	0.01	17	0	100	0	0
AMAZAKE							
[C] Brown rice amazake	0.12	0.01	39	28	0	0	
[D] Millet amazake	0.05	0.01	53	n.a	0	0	25
[T] Homemade amazake	0.13	0.02	57	21	0	0	0
KOJI							
[S] Koji	0.12	0.02	3	n.a	0	0	0

[a] Average of three repetitions. [b] n.a = ingredient used but percentage not available on the label. [c] "–" = ingredient not supplied.

posed by As in the diet.[27] A subset of samples, representing products with the highest t-As levels, was selected for As speciation; results are presented in Table 1. For all of the rice products selected for speciation, the predominant As species was inorganic (63–83%), with the remaining As being DMA (Fig.1). These percentages agree with the following relative amount; 88% i-As and 12% of DMA, reported for white rice flour from the northeast of Japan.[24] As was pointed out earlier, our results are also in overall agreement with the latter study for Japanese brown rice. It had been previously reported that i-As and DMA are the main As species in both white and brown rice[28,29] and it is considered that rice is the most important source of these As species in the human diet.[11]

Maximum allowable values for As have been established in drinking water under regulatory processes. US and EU water regulation and World Health Organization have a maximum limit of 0.01 mg/L of t-As in drinking water.[30] However, there is no EU, US or WHO standards for t-As or i-As in food.[31] The level of 1 mg/kg of t-As is often cited with respect to rice and that level was established by UK regulations in 1959,[32] 28 years before i-As was classified as carcinogenic to humans by IARC.[26] This peculiar situation clearly shows the lack of updated regulations regarding "safe" levels of i-As in food. On an equivalence bases, 100 g of 0.1 mg/kg i-As in food equates to 1 L of 0.01 mg/L water, remembering that the water limit was set in the EU based on the assumption that 1 L of water is consumed per day.[8–10] Although more research is needed to understand As species bioaccessibility from food, several studies have already reported that over 90% of the As from rice is bioavailable in mammals.[33,34]

The parameter most commonly used for evaluation of As risk assessment is the Provisional Tolerable Weekly Intake (PTWI) established by FAO/WHO: 15 μg i-As/week/kg body weight.[35] The PTWI value can be transformed into the Provisional Tolerable Daily Intake (PTDI) of i-As as follows: TDI = PTWI/7 days = 2.14 μg i-As/day/kg body weight. According to a market basket method carried out in different regions of Japan, the daily intake of the following food groups: sugar and confectionery, pulses and their products (*i.e.* soy products such as misos), and beverages were 33.4, 58.5, 562.2 g, respectively.[36] If it is considered that the "sugar and confectionery" products are all derived from rice syrup, the "pulses and their products" includes rice based miso, and at least half of the beverages is rice amazake, then these food products may increase the i-As intake by up to 0.009, 0.004 and 0.016 mg i-As/day, respectively. For someone with a body weight of 60 kg, who only consumes rice syrup, rice miso and amazake, the PTDI value will be 0.128 mg i-As/day. Thus 23% of the PTDI can be reached by just consuming these rice based products. Furthermore, this exceeds by three-fold the limit set in the EU based on the assumption that 1 L of water, with a maximum concentration of 0.01 mg t-As/L, is consumed per day. It is important to highlight that the Japanese diet already has a very high level of i-As intake directly from rice and seaweed[37,24] and the additional intake, from rice miso, rice syrup and amazake, may pose increased health risks for certain dietary groups. Indeed, the t- and i-As levels reported in urine samples from Japanese populations are much higher[12] than those reported for populations in Europe and North America.[13,14] The findings of this study also have implications for health conscious consumers in other countries who eat these products in addition to brown rice which can comprise between 50–60% of their main meals.[16]

Conclusion

This study has shown that rice based miso, syrups and amazake from Japan can contain significant concentrations of i-As, which can further increase the daily human exposure to the most toxic As species (arsenite and arsenate). The Japanese population already have much higher concentrations of total and i-As in their urine than populations in Europe and USA, perhaps related to their high consumption of rice and rice based products. This experimental situation is suggestive of a high dietary exposure to As, some of which is likely to come from the type of rice-based products analysed in the present study. The study has also shown that similar products derived from barley and millet display lower i-As contents. Thus, it may be advisable, for those already exposed to a high level of i-As, to eat miso, syrups and amazake that are prepared from barley, soya, millet or wheat, at least until low i-As containing rice is available.

Acknowledgements

Antonio José is an Early Stage Researcher of the AquaTRAIN Marie-Curie Network funded under the European Commission. Sixth Framework Programme (2002–2006) Marie Curie Actions, Human Resources & Mobility Activity Area, Research Training Networks. This paper reflects the views of the authors but not necessarily those of the European Community, which is not liable for any use that may be made of the information contained therein.

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