Food Standards Agency contract C101045:

Levels of arsenic in rice – literature review

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REPORT CONTENTS

REFERENCES 60

FIGURES

consumption of 1 L of water per day

TABLES

GLOSSARY

1. INTRODUCTION

A number of studies indicate, that in countries not suffering from high levels of arsenic in drinking water, that rice is a major contributor to inorganic arsenic in human diets¹⁻⁶. Although seafood is known to be high in total arsenic, the inorganic component is small^{3,7}. Rice on the other hand has a high proportion of inorganic arsenic^{3,8-12}, and rice is particularly susceptible to assimilating arsenic into its grain¹³.

This report sets out to assess the importance of arsenic in rice, particularly its inorganic component, to dietary arsenic intakes in a UK context. This involved considering the inorganic and total arsenic levels in UK available rice, the quantities of rice consumed by UK subpopulations (particularly those with high rice consumption rates), and placing this data into the context of the health risks posed by chronic inorganic arsenic exposure from rice.

Having considered the available information, the focus of future studies into arsenic in rice in a UK context is suggested.

2. ARSENIC IN THE PADDY FIELD ECOSYSTEM

2.1. Introduction

Rice is the most important grain crop worldwide, being the staple for around 50% of the world's population. It is grown widely in SE Asia, and with more discrete regional distribution in Southern Europe, Southern USA, South America and Africa. All soils, including rice paddies, naturally contain the element 14 . Whether the baseline levels of arsenic vary in soils between rice grow regions, potentially resulting in rice grain with different arsenic burdens, has yet to be ascertained.

2.2. Diffuse arsenic pollution

The natural arsenic soil burden can be added to by anthropogenic processes through both diffuse and point source contamination. Many paddy rice cultivation regions are situated on deltas and floodplains where diffuse pollution from feeding catchments are deposited in the sediments that accrete to form paddy soils. Thus, arsenic released into the upstream environment from industrial activities, sewage treatment works, pesticide application and fertilizer use may result in elevated levels in paddy soils. For example, floodplains in the South Central USA rice growing region are contaminated from diffuse pollution resulting from arsenical pesticide application⁵. Diffuse arsenic pollution of paddy environs, as yet, has not been adequately characterised.

2.3. Point source arsenic pollution

Point source pollution of paddies with arsenic has occurred in both USA and SE Asia. These point sources can be considered in four categories: pesticide, base and precious metal mining and processing, ground water contamination and municipal solid wastes.

2.3.1. Pesticide use

In the South Central US cotton belt, focussed on the Mississippi delta/floodplain and Texas, it has long been the practise to grow rice on soils previously used for cotton production where cotton was treated against boll weevil infection with arsenical pesticides, both inorganic (historically) and organic (still licensed), and as a desiccant to remove leaves before boll harvest⁵. It is suspected that this past arsenical pesticide/desiccant usage is the reason why South Central US rice has an average arsenic content almost double that of Californian rice, where the bulk of Californian rice is from northern California which does not have a history of cotton production⁵.

Although it remains to be ascertained in the literature, that there is little or no use of arsenical pesticides in paddy regions of developing countries. The reason for this is availability, cost and transport infrastructure.

Arsenical pesticides, besides their use in cotton production, were widely used in viniculture and orchards etc., and historic use may have contaminated southern European deltas and floodplains, though evidence for this is lacking.

2.3.2. Base and precious metal mining and processing

There are extensive regions of base and precious metal mining in SE Asia that coexist with subsistence rice farming¹⁶⁻¹⁷. Paddies can become contaminated from use of irrigation water impacted from mine runoff or overspill from mine tailing damns and from mine tailing damn collapse. Sediment redistribution from mine tailings also occurs due high rainfall events. Smelting of ores leads to atmospheric deposition and

subsequent mine soil pollution. Arsenic minerals are often associated with gold, silver, copper, zinc, lead and tin ores. Paddy soil contamination from mining related activity in China, with specific examples where high levels of arsenic in rice grain have resulted have been reported in the literature¹⁶⁻¹⁷.

2.3.3. Groundwater irrigation

Groundwaters of many of the major deltas and floodplains of SE Asia have naturally elevated levels of arsenic, including the Ganges, Brahmaputra, Mekong, Red, Pearl, and Irrawaddy River systems¹⁴. It has become the practise, particularly in Bangladesh and West Bengal, to irrigate paddy fields during the dry season with groundwater to provide two rice crops for year. Approximately half of Bangladesh is served with arsenic contaminated groundwater for irrigation purposes and this has led to extensive soil pollution with arsenic which has led to elevated arsenic in rice grain $12,18$.

2.3.4. Fertilizer application

Though not extensively investigated, fertilizer application may be a source of arsenic to paddies. A specific example of this is where municipal solid waste was applied to paddies in West Bengal, India resulting in elevated arsenic in paddy soil and, as a consequence, in rice grain¹⁹. It is not known how widespread this practise is.

2.4. Soil to grain transfer of arsenic

Lowland rice cultivation, that is paddy rice cultivation, is atypical of virtually all other major crops in that it is grown anaerobically (in wetland rice soils, flooding a field cuts off the oxygen supply from the atmosphere to the soil, which results in an anaerobic environment). The speciation of arsenic in the soil environment is dynamic where it can be biotically and abiotically interconverted between the dominant solution phase inorganic species of arsenate (As^v) and arsenite (As^{III}) , the oxidized and reduced form respectively²⁰. Inorganic arsenic can also be methylated through microbial action to give monomethylarsonic acid (MMA) and dimethyarsinic acid (DMA). All four species are present in the solution phase of paddy soils²⁰. These four species can also be assimilated by rice roots^{21} . The dynamic between solution phase and solid phase speciation is also important. Under oxidized conditions, where arsenate dominates, iron, as Fe^{III} , forms insoluble oxyhydroxide (FeOOH) mineral phases that have high affinity for arsenate, leading to low solution phase $\frac{14}{10}$. Under highly reduced conditions arsenic, as the reduced species arsenite, is precipitated from solution in sulphur minerals, primarily as arsenopyrite. At intermediate redox conditions, such as those found in paddy soils which continuously fluctuate between aerobic and anaerobic conditions, arsenic is mobilised from both pyrites and oxyhydroxides as the relatively mobile arsenite¹⁴. Thus, for aerobically grown crops the relatively immobile arsenate is the dominant plant available form, but for anaerobically cultivated rice, the more mobile arsenite is the dominant plant available form. This has a dramatic consequence for plant assimilation of arsenic as illustrated by Figure 2.1 which shows arsenic soil-shoot-root relationships for rice, wheat and barley cultivated on various British, French, Spanish and North American field sites.

Figure 2.1. Arsenic levels in root, shoot and grain of rice, wheat and barley surveyed from UK, USA and EU field sites.

* **Submitted for publication in Williams et al.13. Rice is represented by black circles.**

From Figure 2.1. it is observed that rice shoots and grain take up a lot more arsenic than wheat and barley from soil, even though wheat and barley have been found growing on much more contaminated soils (those of the SW of England) compared to rice, with the highest soil sample for wheat/barley being 50 fold more contaminated than for rice. For rice, when soil arsenic rises above around 5 mg/kg, arsenic export to the shoot increases dramatically, suggesting very high bioavailability of this element. Figure 2.1. is the first data set to compare rice grain arsenic uptake with other crops and, therefore, the first to reveal why arsenic is problematic in rice with respect to grain arsenic levels.

3. VARIATION IN ARSENIC IN GRAIN FROM DIFFERENT RICE PRODUCING REGIONS

3.1. Regional comparisons for arsenic in rice

An ongoing extensive survey of arsenic levels in rice from different origins has been conducted at the University of Aberdeen, with part of this work already in $5,11-13$. To date, the data base contains 850 market rice samples, with 788 white rice samples, excluding the field survey results that the database also contains. All data was analysed in our laboratory using uniform procedures and quality control, using rice flour certified reference material NIST 1568a. Details of procedures and quality control can be found in the Williams et al. Papers.

Table 3.1. summarizes the entire market rice (white, brown and red) rice findings based on country of origin. One-way Analysis of Variance (ANOVA) showed that there are highly significant differences $(P \le 0.001)$ between the different geographical regions. Egyptian rice had the lowest mean grain arsenic content of 0.051 mg/kg, whilst the highest mean level was for French rice at 0.24, a five-fold range in levels. The 95th percentile for Egyptian rice at 0.08 mg/kg was below the 5th percentile for Japanese, US and French rice (Figure 3.1.). The 95th percentile for Spanish and US rice were above 0.4 mg/kg, while this figure for French rice was 0.55 mg/kg.

Table 3.1. Mean rice arsenic concentrations in rice (white, brown and red) from different countries of origin (mg/kg)

When just looking at white rice, which constitutes the bulk of the survey, the only real change is that French rice levels drops below USA levels, making USA white rice the most contaminated in the survey (Table 3.2., Figure 3.1.). The reason for this large change in French rice between the whole survey and the white rice samples is due to the red rice (Figure 3.1.). This red rice (unmilled) has almost double the levels of arsenic compared to brown and white rice from the same region (Figure 3.1.). Where there were enough brown rice samples to compare with white rice, namely France, Italy and USA, levels in brown rice had means similar to, or slightly higher than, white rice. Ren et al. 22 have shown that rice bran contains considerably more arsenic, up to 10-fold higher, than milled rice. The bran only constitutes a small portion of the

grain biomass, but still contributes significantly to whole grain arsenic levels, raising arsenic levels compared to polished rice.

Country of origin	Number of samples	Mean arsenic	Standard deviation
		concentration	
	(n)	(mg/kg)	(mg/kg)
Egypt	110	0.05	0.06
Nepal	12	0.06	0.03
India	68	0.08	0.04
Pakistan	16	0.09	0.1
Ghana	34	0.1	0.08
Bangladesh	100	0.13	0.06
Australia	11	0.14	0.1
Thailand	48	0.14	0.04
China	92	0.15	0.06
Italy	28	0.16	0.07
France	21	0.19	0.04
Japan	26	0.19	0.08
Spain	51	0.19	0.14
USA	174	0.25	0.09

Table 3.2 Mean rice arsenic concentration in white rice from different countries of origin (mg/kg)

The market basket survey data presented in Figure 3.1. and Tables 3.1.-3.2. takes no account of regional variations within each country, or indeed in seasonal variations in rice crop production. The Chinese samples came from a range of regions, including some impacted by mine waste water. For the USA there are large differences in rice grain levels from the two rice growing regions: California (average rice arsenic level of 0.17 mg/kg) and South Central region $(0.27 \text{ mg/kg}$ average grain arsenic)⁵. The South central region produces 80% of US rice. For Bangladesh, around half the rice cultivation area is served by irrigation tubewells with elevated arsenic, leading to about three-fold higher levels of arsenic in the grain compared to low arsenic groundwater regions $^{12, 18}$.

This basket survey has been broken down in Table 3.3. into country of purchase, specifically to highlight rice available in the UK. If anything, the arsenic levels in UK purchased rice where slightly lower than that purchased in the country of origin. From Table 3.3., the concentration of total arsenic in rice varies 5-fold dependent on country of origin, with USA being the highest (0.25 mg/kg) and India the lowest (0.05 mg/kg).

* **The central black line in the box is the median, while the red line is the mode.** The outer boundaries of the box are the 25th and 75th percentiles, the whiskers are the 10th and 90th percentiles and the dots the 5th and 95th percentiles.

Table 3.3. Descriptive statistics for rice purchased in the UK originating from different countries compared to rice purchased in the country of origin.

 $*$ reference: Rmalli et al.²³

To place this basket survey in context, other basket surveys of total arsenic content published in the literature have been tabulated in Table 3.4.. These comparisons, where the same countries are surveyed, are in agreement with the Aberdeen survey.

The $FSA²⁴$ commissioned a study into UK purchased weaning foods in which 6 pure rice flour samples where analysed for total arsenic with levels ranging from 0.150 to 0.276 mg/kg As, with a mean of 0.23 mg/kg. From Table 3.3. these concentrations suggest that this rice was sourced from US or European growing regions.

The mean total arsenic content in US rice range from 0.2 to 0.3 mg/kg, although the lower value was for cooked rice (Table 3.4.). However, not tabulated, is the US FDA arsenic total diet study published by Tao $\&$ Bolger²⁵ had arsenic in cooked rice ranging from 0.03 to 0.11 mg/kg for 18 samples, which is lower than all other studies, though they did find total arsenic levels ranging from 0.07 - 0.30 mg/kg (n=18) in crisped rice cereal. Arsenic levels in rice products will be considered in Section 3.4..

Also, in a Belgium based total dietary intake survey Robberecht et al.²⁶ found total arsenic levels in rice of up to 0.26 mg/kg (only ranges were given).

	Number of samples	Mean As (mg/kg)	Min. As (mg/kg)	Max. As (mg/kg)	Reference
USA	$\overline{4}$	0.30	0.22	0.46	3
USA	5	0.26	0.11	0.34	9
USA (Cooked)	6	0.21	0.10	0.30	8
Italy	8	0.17	0.08	0.27	27
Vietnam (white)	31	0.21	0.03	0.46	28
Vietnam (brown)	25	0.29	0.08	0.70	28
Australian (white)	11	0.26	0.12	0.78	28
Taiwan	407	0.08			29
Spain	$\overline{4}$	0.34	0.29	0.41	30

Table 3.4. Arsenic levels in rice from published surveys

Rice grain purchased for consumption in the market represents an integration, to an unknown degree, of rice supplies from an individual region, dependent on where mills are located, how the rice was shipped (in bulk or pre-packaged), the regional extent of farms supplying the mill, and the field or within region variation in grain arsenic levels. Field surveys of whole (brown) rice grains presented in Table 3.5., showing the extent of more localized variation of arsenic in grain in the samples regions.

Table 3.5. Descriptive statistics for whole (brown) rice surveyed at the field level by the University of Aberdeen, in press13 or unpublished.

3.2. Comparing rice with other grain crops

To determine how arsenic levels in rice grain compare to other grain crops available in the UK, a region with low soil arsenic (the south east of Scotland) and a region with high soil arsenic (Cornwall and south Devon) where surveyed for field sampled barley and wheat and compared with similar surveys of EU and US rice (Table 3.6.). The SW England soils with high arsenic where specifically targeted to examine the worst case scenario for barley and wheat in the UK, with soil levels, as related to shoot and grain concentrations, presented in Figure 2.1.. Average levels in Scottish wheat and barley where 0.03 and 0.04 mg/kg respectively, with these levels doubling in the arsenic affected regions of the SW England (Figure 3.6.). These levels compare to mean ranges in rice from field survey of 0.13 mg/kg in Californian rice to 0.32 mg/kg in French rice (Table 3.6.).

Other surveys confirm the ranges found for wheat and barley. A Netherlands survey that found a mean wheat grain level of 0.05 mg/kg d. wt. (assuming 15% water content) for 84 samples and a mean barley grain level for 0.08 mg/kg d. wt. (assuming 15% water content) for 45 samples³¹. Average US wheat grain levels are reported as 0.02 mg/kg d. wt.³². In UK field experimental plots of wheat grain grown under soil compaction and irrigation treatments, mean grain levels where ≤ 0.01 mg/kg for two successive harvests 33 .

Crop	Country	Region		min-max	mean	median	n
rice	France	Camargue	grain	$0.12 - 0.61$	0.32	0.34	22
			shoot	1.5-20.6	10.2	6.8	23
			soil	$5 - 10$	8	8	23
	Spain	Doñana	grain	$0.06 - 0.29$	0.16	0.15	25
			shoot	$0.8 - 9.8$	3.3	2.6	25
			soil	$4 - 11$	8	7	25
		Cadiz	grain	$0.07 - 0.21$	0.13	0.14	10
			shoot	$0.4 - 3.3$	1.4	1.2	10
			soil	$1 - 2$	$\mathfrak{2}$	$\overline{2}$	10
	USA	California	grain	$0.08 - 0.18$	0.13	0.11	9
			shoot	$0.4 - 1.3$	0.7	0.7	9
			soil	$2 - 4$	3	3	9
		Arkansas	grain	$0.08 - 0.43$	0.2	0.18	6
			shoot	$0.7 - 3.4$	1.5	1.3	6
			soil	$4 - 7$	6	6	6
wheat	UK	E Scotland	grain	$0.01 - 0.21$	0.03	0.02	29
			shoot	$0.0 - 0.21$	0.2	0.1	29
			soil	$3 - 18$	τ	6	29
		SW England	grain	$0.01 - 0.50$	0.07	0.04	37
			shoot	$0.1 - 1.6$	0.3	0.2	37
			soil	$6 - 201$	33	21	37
barley UK		E Scotland	grain	$0.03 - 0.05$	0.04	0.04	6
			shoot	$0.1 - 0.2$	0.1	0.1	6
			soil	$6 - 10$	τ	7	6
		SW England	grain	$0.01 - 0.54$	0.08	0.03	29
			shoot	$0.1 - 1.8$	0.4	0.2	29
			soil	6-546	57	25	29

Table 3.6. As distribution in rice, wheat and barley grain, shoot and soil by production region

3.3. Speciation of arsenic in rice grain

3.3.1. Arsenic species in the paddy field environment

The element arsenic has a dynamic speciation, with the inorganic forms arsenate and arsenite converted to organic forms by microbes, animals and plants. Figure 3.2. shows the common forms of arsenic found in biotic environments. Arsenobetaine is normally associated with marine environments, though it can be found in earthworms^{34} . Similarly, arsenosugars are marine associated, being found in high levels in seaweeds³⁵. The inorganic forms arsenate and arsenite are the dominant species in soil solution, the former predominant in aerobic soils and the later in

anaerobic soils 20 . Methylated species are also widely found in soils, particularly monomethyarsonic acid (MMA) and dimethylarsinic acid (DMA), which are found at high proportions in paddy soils²⁰. DMA was widely used as a pesticide, usually under its common name cacodylic acid, and is still licensed for use in the USA for lawn treatment and cotton production 5 .

Figure 3.2. Arsenic species in biotic environments

3.3.2. Concentrations of inorganic arsenic and DMA in rice grain

For rice grain, arsenic speciation is dominated by inorganic arsenic (arsenate and arsenite) and by $DMA¹¹$. It still is not known if the DMA is produced *in planta* or is assimilated from soil, though considering all evidence, especially the poor uptake of DMA by rice roots²¹, it seems that the former is more likely. It is likely that both arsenate and arsenite are present in grain, but as extraction and storage for analysis causes inter conversion of the two species²⁰, rice grain arsenate and arsenite are summed to give an inorganic arsenic concentration.

Summary statistics for all the market basket purchased rice that has been speciated at Aberdeen is presented in Table 3.7.. Part of this tabulation has been published¹², while the Chinese and Japanese survey is new. The Indian, US, EU and Italian rice were UK purchased. The distribution of inorganic arsenic in rice varied considerably with country of origin, and basically followed the total arsenic trend. For UK purchased rice, on average 50.1% of the total arsenic present in the grain was inorganic, but there was variation in this percentage value, ranging from 20-74% (n=28). This compares to the entire dataset where the average inorganic content was 53.9%, ranging from 20-89.5% (n=45).

Table 3.7. Arsenic speciation in rice determined by University of Aberdeen, denoted by country of origin and country of purchase.

The average total inorganic arsenic in UK purchased rice varied considerably from 0.03 mg/kg for Indian rice through to 0.09 mg/kg for US and 0.12 mg/kg for Italian (Table 3.7.). The highest average inorganic arsenic in rice value was found in Japanese purchased Japanese rice of 0.16 mg/kg.

There are only a limited number of datasets with which to compare the speciation outlined in Table 3.7.. Font et al.³⁶ surveyed 40 rice samples of Spanish origin purchased in Valencia. They found a mean inorganic content of 0.11 mg/kg and a range of 0.01-0.27 mg/kg. This survey did not report total arsenic content. Laparra et al. 30 also reported speciation in 4 Valencian samples where totals and speciation were reported. They found that inorganic arsenic varied from $0.10 - 0.20$ mg/kg for rice samples with total arsenic contents of $0.29 - 0.41$ mg/kg.

Figure 3.3. plots the Aberdeen UK inorganic arsenic survey with that for US purchased US rice published by Lamont et al.¹⁰ (n=39), Schoof et al.³ (n=4) and Heitkemper et al. 9 (n=5) From this combined survey the average inorganic arsenic content of US rice was 0.12 mg/kg with a range of 0.01-0.30 mg/kg for n=58.

Figure 3.4. reports a more detailed analysis of inorganic arsenic in rice, reporting its percentage contribution to total arsenic levels for white and brown rice. The market basket survey was supplemented by field survey and pot experiment samples, all conducted at Aberdeen. For both white and brown rice, total inorganic arsenic was negatively correlated with total arsenic, while the converse was true for DMA, being positively correlated with total arsenic. General Linear Modelling (GLM) ANOVA showed for the relationship between total arsenic and percentage inorganic arsenic that both rice type (brown or white) and total arsenic concentration (entered as a covariate) were significant with $p=0.03$ and $p<0.001$ respectively, with the interaction term being non-significant. For the corresponding test for percentage organic (DMA) arsenic, the interaction term was significant, showing that white and brown rice behaved differently. The extraction efficiency of TFA interaction term was also significant on GLM analysis, revealing further differences in arsenic behaviour, probably due to, as yet, further undefined differences in speciation.

The relationship between percentage speciation and total grain arsenic does show scatter, with inorganic arsenic component varying from 4-94% for brown rice and 20- 86 for white, with the corresponding means (\pm S.E.) being 54 \pm 3 and 52 \pm 2.4% respectively. The linear regression between total arsenic and percentage inorganic arsenic can be used to further refine this prediction.

For white rice this relationship is:

percentage inorganic arsenic = $60.4 - 57.9$ * total arsenic (mg/kg), $r^2 = 0.123$

Where r is the residual from the mean.

and for brown:

percentage inorganic arsenic = $76.7 - 64.6$ * total arsenic (mg/kg), $r^2 = 0.267$

*** All the UK data is from Williams et al.11. Dark green symbols are for UK purchased Indian, dark brown for UK purchased US rice, light brown for UK purchased Italian rice, the dark pink symbol for UK purchased Thai rice, the dark blue symbol for UK purchase Spanish rice and light grey symbols for European rice not denoted by country. For US purchased rice, light green** symbols are from Lamont¹⁰, blue symbols from Heitkemper et al.⁹ and pink **symbols from Schoof et al.3 .**

Figure 3.4. Speciation in Aberdeen surveyed brown (filled symbols) and white (open symbols) rice by HPLC-ICP-ms.

* **The shapes and colours of the symbols code for the following: Bangladesh (triangles), China (stars), Europe (diamonds), India (squares), USA (hexagons). All white rice (n=40) was purchased from markets for direct human consumption. For the brown rice (n=45), the market rice has a blue outline to the symbol, field collected rice has a green edge and pot grown rice a red edge. Solid line is for brown rice regression, dashed line for white rice regression. Graph A is for percentage inorganic arsenic, B for percentage organic (DMA) arsenic, and C for percentage extraction efficiency (sum of HPLC measured species/total arsenic).**

With respect to the threat that arsenic in rice poses to the human diet, assuming that inorganic arsenic is more problematic than organic $(DMA)^5$, then polishing rice reduces the total arsenic burden of the grain and its inorganic content. Brown rice is unpopular in staple/subsistence rice diets, and tends to be more popular in developed countries as a wholegrain alternative to white rice. While this raises concerns about brown rice *per se*, it is clear that products made from rice bran and germ, such as rice milk and direct consumption of bran and germ as dietary fibre supplements, are of particular concern and need further attention. Our preliminary analysis of the major

UK available brand of rice bran, used similarly to other cereal brans as a fibre fortifier, found total arsenic levels of 0.55 mg/kg. Speciation has yet to be determined.

3.4. Arsenic in rice products besides grain

With the exception of weaning and infant products which have recently been reviewed by the FSA^{24} , rice products have received little attention. Rice flour is used in many processed food products, where it normally will be diluted.

Products which are primarily rice based may have similar arsenic contents to rice grain. These include:

- Puffed rice breakfast cereals
- Puffed rice cakes
- Rice wine
- Rice vinegar
- Mirin fermented rice liquid
- Rice miso fermented rice paste
- Amazake (amassake) a milk substitute created from fermenting sweet rice
- Rice syrup
- Rice malt
- Rice bran
- Rice bran oil
- Rice bran solubles
- Rice bran pickles
- Rice milk a cow and soya milk substitute

The University of Aberdeen has started a survey of these products and data is being collected at present. Preliminary results for total arsenic levels in liquid products are presented in Table 3.8. show that many contain total arsenic levels above 0.01 mg/L. In particular all rice milks, which are used as a dairy milk substitute, had levels above 0.01 mg/L, with some above 0.03 mg/L. As this product is drunk like milk or used on breakfast cereals or in cooking as a milk substitute, unlike rice vinegars and mirin which are used in small quantities as condiments, levels may give rise to high intakes.

Rice milk has become popular in the health food market as cow milk replacement as it is low in hormones and has no lactose. It has a more bland taste than soya milk, and hence its increasing popularity. Soya allergies also cause a dependence on rice milk. The rice milk samples presented in Table 3.8. still have to be speciated, but as rice contains over 50% inorganic arsenic on average (Section 3.3.2.), the inorganic arsenic content could be high.

Table 3.8. Total arsenic levels in liquid rice products

One published study shows that arsenic levels in bran are high, ranging from 0.55 to 1.05 mg/kg in Chinese bran, produced from brown rice with arsenic levels ranging from 0.08 -0.39 mg/kg²². Initial unpublished data from Aberdeen shows that UK purchased bran products are high in arsenic with mean levels of 0.55 mg/kg total arsenic. Although speciation is still to be conducted, bran is enriched in arsenite (Section 3.3.2.). Figure 3.4. shows that brown rice has a higher inorganic arsenic content than white rice.

Reports of arsenic levels in other rice products are limited. Crisped rice cereals are widely consumed. Tao and Bolger²⁵ found total arsenic levels of 0.07-0.34 mg/kg in crisped rice cereal. Robberecht et al. 2002 found up to 0.58 mg/kg in rice biscuits from a Belgium food basket survey. Rice puddings may also have similar arsenic levels to rice grain. Williams et al.¹¹ found that EU produced pudding, flaked and ground rice contained 0.12, 0.14 and 0.20 mg/kg total arsenic respectively, with 66,

44 and 51% of this total being as inorganic arsenic, respectively. Pre-prepared rice based puddings, such as tinned rice pudding, will reflect these arsenic levels and speciation.

Many of the fermented rice products are consumed in traditional Japanese diets, which have been modified for macrobiotic diets³⁸. In macrobiotic diets the ferment products are used as condiments, milks and sweets; rice milk is used as a cow milk substitute for cereals and puddings; rice syrups and malts used as sweeteners; and boiled/steamed brown rice is the staple.

3.5. Infant foods and weaning products

Table 3.9. reports the University of Aberdeen unpublished data on arsenic levels in baby rice. Four different makes of rice were examined from three different manufacturers, with arsenic levels in rice varying from 0.128 through to 0.494 mg/kg. Specific brands, had higher arsenic levels than others. These levels are similar to those reported in baby rice from the $FSA²⁴$ survey, where levels in pure baby rice varied from $0.069 - 0.276$ mg/kg. The FSA²⁴ survey also looked at other baby food items containing rice and found that these had similar levels to pure baby rice, with the highest arsenic levels found in organic rice cakes at 0.404 mg/kg (Table 3.10.). The $FSA²⁴$ looked at over 200 separate baby food items. All items formulated with rice were above the $80th$ percentile with respect to total arsenic levels, while all pure baby rice products were above the $90th$ percentile, with this percentile approximately being at 0.100 mg/kg arsenic (Figure 3.5.). The median for the entire survey was only 0.006 mg/kg, showing how high in arsenic rice and rice products are with respect to other baby foods.

This elevation in total arsenic levels in baby rice products from the $FSA²⁴$ survey is further demonstrated by the descriptive statistics presented in Table 3.11.. All products containing rice are elevated, with the only comparable total arsenic levels being for fish products.

The FSA has commissioned a study specifically looking at inorganic arsenic speciation in weaning foods and formulae for infants³⁹, and this data is tabulated in Table 3.12. It confirms the high levels of total arsenic in baby foods containing rice observed in tables 3.9-11..

When products contained rice, the percentage of this total arsenic present as inorganic was considerable, reaching up to 94%, higher than any values measured in market purchased rice observed to date (Table 3.7.). The average inorganic content reported in baby rice containing foods of 0.135 mg/kg (Table 3.12.) was higher than that observed for pure rice grain (Table 3.7.), and the percentage inorganic content was higher than for any source of origin of rice grain (Table 3.7.).

Table 3.9. Total arsenic levels in baby rice; Meharg, unpublished data.

Product	Sample number	Total arsenic	Standard
		concentration	deviation
	(n)	(mg/kg)	(mg/kg)
Rice	7	0.200	0.041
$Rice + milk$	1	0.069	0.000
Biscuits	8	0.015	0.014
Breakfast cereal	26	0.030	0.032
Rice porridge	1	0.217	0.000
Cereal bar	5	0.018	0.012
Rice bars	4	0.250	0.123
Desserts	11	0.015	0.021
Rice desserts	1	0.056	0.000
Fish	7	0.145	0.083
Follow on milk	13	0.005	0.006
Fruit puree	7	0.011	0.016
Growing milk	4	0.002	0.002
Infant formulae 1	11	0.003	0.002
Infant formulae 2	13	0.006	0.007
Meat	45	0.020	0.031
Other milk	6	0.006	0.001
Pasta	15	0.018	0.032
Rice pasta	1	0.064	0.000
Rusks	7	0.005	0.003
Vegetables	6	0.005	0.005
$Vegetables + rice$	1	0.0063	0.000

Table 3.11. Descriptive statistics for total arsenic levels in baby foods from FSA24 survey

Figure 3.5. Arsenic levels reported in FSA²⁴ "Survey of metals in weaning foods **and formulae for infants".**

* **Symbols coloured green denote those without rice in the "products as described column" except for fish products which are denoted in blue, red symbols are for pure rice products and orange symbols for mixed rice products.**

Table 3.12. Total and inorganic arsenic determined in weaning foods and formulae for infants as reported in FSA FD 06/12³⁹

3.6. Arsenic in rice grain imported into the UK

Table 3.13. lists the tonnage and percentage of rice imported from each country that has over 1% in 2005 of all UK imports. The first thing to note is that there is a large change between 2005 and 2006 with respect to where rice is imported from. Over the two years import from India was the largest, accounting for 21-22%, with the USA being second largest in 2005 at 17.9%, but falling to 12.6% in 2006. This is directly related to the exclusion of US rice since September 2006 due to the presence of GMcontamination and has "caused enormous disruption"⁴⁰. The USA has been the most important source of long grain rice to the UK, and its lack of availability has tended to increase the price of other sources^{40}. This disruption, i.e. internal redistribution of supplies within Europe, may also explain the large drop in Italian imports into the UK from 17.5% to 12.7%. There was a large increase in Spanish rice imported into the UK in 2006, and a smaller increase in rice from the Netherlands, Pakistan and Thailand in that year. Overall, the total tonnage of rice imported into the UK fell from 2005 to 2006 by 534,453 to 485,448 tonnes.

Table 3.13. Tonnage and percentage contribution on a country basis of major sources of rice imported into the UK in 2005-2006.

Source: DEFRA

It is unclear from these import data, particularly for European importers, where the rice has originated. The Netherlands is not a rice producing country yet was the $4th$ largest importer in 2005 and the $2nd$ largest in 2006. It cannot be certain, therefore, that all Spanish, French and Italian imports all originate from these countries although, unlike The Netherlands, they are rice producers. For intercontinental importers such as USA, India, Pakistan, Thailand and Egypt, it is likely that the rice imported from these countries is produced by these countries, given transport costs etc.

With respect to arsenic levels in imported grain, all European and USA sources must be considered elevated compared to African and SE Asian importing countries (Figure 3.1. and Table 3.2.). The African and SE Asian market accounted for 34.3% of imports in 2005, rising to 39.0% in 2006. The mean levels of arsenic in Egyptian

uncooked white rice was 0.05 mg/kg, 5 fold lower than USA uncooked white rice at 0.25 mg/kg (Table 3.2.), a 5-fold difference.

4. The effects of cooking rice on its arsenic content

4.1. Rational for conducting rice cooking studies in the context of arsenic contamination.

There has been interest in how cooking of rice affects arsenic content and speciation, with a primary focus on regions of the globe that suffer elevated arsenic levels in water used for cooking such as West Bengal/Bangladesh^{30, 41-44} and USA⁸. These invariably find that when rice is cooked in waters containing inorganic arsenic that the rice is very effective at binding the arsenic in the cooking water, leading to an increased arsenic burden in the ingested rice. This has little relevance for the UK context where public water supplies are low in arsenic, but it does suggest that rice is effective at retaining arsenic present in the grain.

4.2 Rice cooking studies

As there are only a limited number of studies on the effects of rice cooking on arsenic, each study will be considered individually below.

Bae et al.⁴¹ were the first to broach this subject. Their study was based in Bangladesh using traditional cooking techniques. Market rice was boiled in an aluminium pot. The rice (500g) had around 2.5 L of water added, with the rice absorbing around 1.2 L of this water. The water had 0.23-0.37 mg/L inorganic arsenic, causing the arsenic content of the uncooked rice to rise from 0.17 to 0.21-0.31 mg/kg As on cooking. They found that the arsenic content of the rice increased by 10-35% more than predicted simply by water absorption, and concluded that arsenic concentrated in the rice during the cooking process, probably due to loss of solubilised material (such as starch), effectively reducing the dry mater yield. No speciation was reported in this study.

In an Aberdeen University unpublished study total arsenic levels in an uncooked American long grained rice sample increased from 0.232 ± 0.004 mg/kg d. wt. to 0.274 ± 0.009 mg/kg d. wt. (n=4) on cooking where 530 ml of water were used to cook 300 g of uncooked rice. The mineral water contained 0.003 mg/L total arsenic, contributing up to 0.0053 mg/kg to this arsenic increase in cooked rice, the rest is due to grain mass loss on cooking. Speciation on these samples will be conducted.

For the Ackerman et al.⁸ study a water to rice ratio of 1:1 to 4:1 was used, as per cooking instructions on the rice packet. The water was distilled and deionised, and they used either arsenic free water or water with 0.022 mg/L added. After cooking rice samples were oven dried. Arsenic was speciated by HPLC-ICP-MS. Unfortunately, this study did not report arsenic levels or speciation in the uncooked rice, therefore, the effects of cooking on total arsenic levels and speciation could not be ascertained. They found that 89-105% of arsenic added in the cooking water was absorbed to the rice. This was higher than for Bae et al.⁴¹ Bangladesh study, but the Bangladesh arsenic level in the cooking water was an order of magnitude higher.

A more revealing study with respect to the affect of cooking on arsenic levels in rice was conducted by Rahman et al.⁴². They looked at four rice samples, two with relatively low arsenic (0.21 and 0.24 mg/kg total arsenic) and two high arsenic rice samples (0.57 and 0.69 mg/kg total arsenic). For low arsenic rice low arsenic water was used in parboiling and cooking at a concentration of 0.04 mg/kg, while for the

high arsenic rice high arsenic water was used at 0.13 mg/L. Both these water sample would be considered elevated in a UK context. Also, note that the water used in cooking and parboiling was the water used for paddy field irrigation, and thus explains why the "low" arsenic rice is relatively elevated compared to other regions of Bangladesh^{12}. Parboiling did not change the total arsenic content of the rice, regardless of water source used. This may be because the rice still has its hull when parboiled, and the hull may have a high affinity for arsenic. When cooked with excess water (250 ml of water to 50 g rice), arsenic content of the low arsenic rice tended to stay unchanged for both the parboiled and non parboiled, but decreased for both parboiled and non-parboiled high arsenic rice (by 15-35%). When cooked with limited water (100 ml water to 50g rice) however, arsenic content increased in both the high and low arsenic rice. Interpreting this data in a UK context is not possible because the water used in cooking was arsenic contaminated.

A further Bangladeshi study on cooked rice, which included speciation, was conducted by Smith et al.⁴⁴. This study only reported levels in cooked rice collected from households from regions with very high arsenic in the drinking and irrigation water (0.2-0.5 mg/L) and cannot be interpreted beyond the context of estimating dietary intakes of arsenic from rice in Bangladesh.

Perhaps the most relevant of the Bangladesh/West Bengal studies in a UK context is that of Sengupta et al.⁴³ who cooked a range of rice in water containing 0.003 mg/L using traditional high water volume cooking technique and a low volume cooking technique. In the traditionally method rice was first washed until the rinsing water was clear, this reduced the arsenic content of the rice by 28%. Then rice was cooked in a 1:6 rice to water volume with excess water discarded. This removed, including that lost in the rinse step, 57% of the arsenic. Using no rinse and a 1:1.5-2 L rice-water volume, the arsenic content of the rice stayed unchanged.

A European study was conducted using rice purchased in Valencia, Spain, where the arsenic was measured in raw and cooked rice, where inorganic speciation was determined and where deionised (i.e. arsenic free) water was used³⁰. For cooking, 131 g of rice was boiled to dryness with 500 ml of water. Cooked rice samples varied little in terms of total (79-107%) and inorganic (70-130%) arsenic contents.

Finally, a limited study with high grain arsenic, produced by growing rice under glasshouse conditions with elevated soil arsenic, was cooked in arsenic free water at a rice to water ratio of $1:2.5⁴⁵$. The study found that total arsenic levels fell in the rice from 1.25 to 0.48 mg/kg, but the DMA (at 86%) and inorganic arsenic (at 14%) content stayed constant.

In summing up these cooking experiments, it appears where rice is cooked in a small volume of low arsenic water, the arsenic content and speciation of rice differs little between raw and cooked rice. Where large volumes of water are used in cooking, with discarding of the cooking water, rice levels decrease by around about 50%. However, none of these studies are complete in terms of knowledge required for UK consumer conditions where rice steaming is also popular. They do not compare white and brown rice, all do not fully account for speciation (if at all), and those that do consider speciation are too limited to make generic conclusions from. Where levels of arsenic

in cooking water are low, cooking water contributes relatively little to the cooked rice arsenic burden.

5. Human exposure to arsenic in rice

5.1 Human bioavailability of arsenic in rice

Assessing the bioavailability of arsenic from rice is crucial to understanding human exposure. Only one study has addressed this problem on an animal model⁴⁵ while others have used cultured cells as model systems³⁰, and enzymatic extraction⁸⁻³⁰. Direct measurement of human exposure from rice has been assed by urine sampling experiments conducted at the University of Aberdeen.

The Juhasz et al.⁴⁵ used female large white swine $(20-25 \text{ kg})$ from which routine blood samples were taken. The pigs were fed low arsenic swine pellets (500 g/d) during the experiment. Pure arsenic salt solutions (arsenate, arsenite, MMA and DMA) where then orally gavaged or delivered intravenously (IV), while rice (170- 270g) was ingested as feed, with blood samples taken up to 26 hours after dosing. The experiments were conducted on triplicate animals. Pharmokinetic modelling of IV administered arsenic compared to orally dosed arsenic was then performed. The orally gavaged pure salts differed considerably in their bioavailability, with arsenite having 104%, arsenate 94%, DMA 33% and MMA 17% bioavailability. Two types of rice were used in the study, one grown in the glasshouse with high soil arsenic, having a total arsenic content in the cooked rice fed to the pigs of 0.48 mg/kg, 86% of this being DMA and 14% inorganic. The total (i.e. organic and inorganic summed as speciation was not determined on the bloods) was 33%. The second rice experiment consisted of low arsenic basmati that was boiled in 1 mg/L arsenate to give 1 mg/kg rice all speciated as inorganic arsenic. Here the bioavailability to the swine was 89%. The difference in bioavailability may reflect both differences in speciation (as orally gavaged pure DMA is only 33% transferred to the blood stream) and to the arsenic being complexed in a different manner in the rice whose arsenic was solely biologically incorporated (i.e. no additional arsenic in cooking water).

If arsenic speciation in the bloods had been conducted for these experiments, perhaps the relative bioavailabilities of inorganic versus DMA could have been established, though this would depend on *in vivo* methylation which could have confused this assessment.

Laparra et al.30 developed an *in vitro* assay, as compared to the pig *in vivo* study of Juhasz et al.45. The *in vitro* study consisted of lyophilising cooked rice and subjecting it to simulated gastric conditions where the rice is incubated with pepsin and then an intestinal step where it was incubated with pancreatin and bile extract. Then the soluble extract was removed and total arsenic determined. This arsenic extract was also exposed to a monolayer of immobilized Caco-2-cells, cultured human colon cells, seeded onto a polycarbonate surface. The experiments were conducted on rice that had been dosed with inorganic arsenic in the cooking water. However, the bioaccessible fraction, the fraction mobilized by enzymatic digestion, varied from 83% to 120%, showing that both the bioincorporated and the additional dosed arsenic where highly bioaccessible, indeed of the 8 samples tested only 3 had less than 100% recovery. However, only 4-18% of this bioaccessible arsenic, which was primarily inorganic due to the manner in which it was dosed, was assimilated by the Caco-2 cells. This contrasts strongly with the *in vivo* bioavailability of arsenic to swine⁴⁵, where inorganic arsenic dosed into rice via cooking was 89% mobilised into the

bloodstream. This suggests that the Caco-2-cell model is a poor predictor of bioavailability. Only inorganic arsenic and total arsenic levels were assessed.

Ackerman et al. 8 used a very similar enzymatic approach as Laparra et al. 30 to look at the bioaccessible fraction, the difference being that the Ackerman study conducted detailed speciation for both inorganic and DMA fractions. They found that the enzymatic extraction liberated, on average, 94% of arsenic present in five different sample of cooked rice (boiled in arsenic free water). There was only limited change in percentage liberation of the individual arsenic species.

The *in vitro* studies have their obvious limitations^{8, 30} but are illustrative of the potential for arsenic in rice grain to be mobilized in the gut. The *in vivo* pig study was limited in that speciation was incomplete and in that very unusual rice samples were used. True bioavailability of arsenic could be conducted on humans in a totally nonintrusive manner by simply conducted a mass balance on food inputs and excretion outputs for a cohort eating high rice diets, such as a traditional in SE Asia. Such an approach was trialled at the University of Aberdeen and the initial unpublished data presented here. Detailed speciation on food intake and urine are still to be conducted.

The Aberdeen trial consisted of two groups of cohorts, both of which went on a fish/seafood, seaweed, chicken, mushrooms and rice exclusion diet for 7 days before trial commenced. No other limitations were placed on diet. After this week, one group continued with the exclusion diet, while another switched to a rice diet where 300 g packet weight American long grain rice at 0.232. mg/kg d.wt. was consumed per day, flavoured with soy sauce, and supplanted by apples and bananas, with one water source used for both drinking water and rice cooking water. The 300 g weight was chosen as this reflects traditional and UK high rice consumers rice intake (see Section 5.2.1.). Arsenic levels were measured in all food items consumed by the rice eating cohort, and they were all low in arsenic in comparison to the rice $(*LOD - 0.01*$ mg/kg d. wt.). For the rice eating cohort, weight of food consumed, volumes of water drunk and volume of urine excreted during the trial were recorded to enable mass balance calculations to be conducted. Figure 5.1. summarizes the findings and shows that the total arsenic in urine for those on the rice diet for the first urine pass of the day dramatically increased during the 5 days of the trial, with concentration of total arsenic increasing 7 fold and the absolute quantity of arsenic excreted increasing 5 fold. Urine is an excellent biomonitor of arsenic exposure $46,47$ and this study shows that arsenic exposure from a mixture of inorganic arsenic and DMA (see Section 3.3.2.), remembering that DMA was poorly absorbed in gavaged pigs Juhasz et al.⁴⁵, increased 5 fold (in absolute terms) during the study. Detailed recording of intra day variation in urine levels for each person was also recorded, but the data are not presented.

Figure 5.1. Excretion of total arsenic for cohorts eating 300 g of American long grain rice (total arsenic content of 0.232 mg/kg) per day (open symbols) and those on an exclusion (including rice) diet.

*** The figure reports the total arsenic levels in the first urine pass of the day, expressed as both arsenic concentration in urine and the absolute amount of urine passed. Error bars represent the standard error of the mean.**

5.2. Quantities of rice ingested in the UK

Rice consumption rates in the UK can be derived from DEFRA's Expenditure and Food Survey (EFS) (which reports household purchase of rice on a per capita basis) and the National Diet and Nutrition Survey (NDNS) (which records daily food consumption per person $)^{48-51}$.

Considering the EFS data first, when this data is considered by ethnic composition of the household, large differences in rice consumption occur. Table 5.1. shows the detailed ethnic breakdown of this survey, with data expressed on a daily purchase (assumed here to be also consumption) rate, show that there is considerable variation in rice consumption within these broad ethnic classes. For example, mixed race Caribbean's purchased $1/8th$ the quantity of dried rice compared to mixed race Africans, yet purchased over 2.5 fold more takeaway rice than they did dried rice, where mixed race African's purchased no takeaway rice.

The largest rice purchasing/consuming group was Asian-Bangladeshi who purchased total rice amounting to 251 g/d (all as dried rice) on a per capita basis, over 30 times more than the average White-British person (Table 5.1.). Considering dried rice alone Asian-Bangladeshi's purchased 60 times more of this commodity compared to White-British. The second largest consumption group, with under half the Bangladeshi rice consumption, is "other ethnic background" at 118 g/d. This "other" category will include people of Middle eastern origin, South Americans and Pacific Islanders etc, many of whom come from cultures where rice is the staple. All ethnic groups, excluding mixed, had rice consumption rates at least 3-fold higher than White-British.

The subdivision of rice consumption rates within the British-Asian community was confirmed by Wharton et al.⁵² who investigated dietary patterns in Moslem, Sikh and Hindu pregnant women in a maternity hospital, Birmingham, UK. They report the most commonly eaten food (10% or more) per meal. For Pakistani Moslems and for Sikhs rice was not above this 10% threshold for any meal. For Hindus, 25% had rice at the first main meal and 38% at the evening meal. For Bangladeshi Moslems, 100% of the cohort consumed rice at the first main meal and 69% at the evening meal.

Asian-Bangladeshi's make up 0.5% of the UK populace, while the "other" category make up 0.4% of the population (Table 5.3.). Therefore, these very high rice consuming groups make up 0.9% of the UK populace. The total non-white and mixed race percentage of the UK population in 6.7% of the UK population, and these can be considered as high rate rice consumers.

A traditional Bangladeshi diet has a rice consumption rate ranging from 400-650 g/d^{53} , where a traditional Chinese diet typically contains around 180-300 $g/d^{17,29}$. As can be seen from Table 5.1., these traditional rice consumption rates are reduced by these ethnic groups in the UK.

In a nutrient intake study for African-Caribbean populace of inner city Britain rice (rice and rice + peas) contributed to 9% of energy intake, whilst for White European and Pakistani origin, rice did not figure in the to 10 foods contributing to total energy intake⁵⁴, confirming the high rice intakes observed in the African-Caribbean community outlined in Table 5.1..

Table 5.1. Daily adult rice purchase (g/d) from the Expenditure & Food Survey database broken down into detailed ethnic grouping .

* **Data from DEFRA Expenditure & Food Survey. Averages for the three years ended 31st March 2005, Updated on: 25/05/2006. Supplied by the Food Statistics Branch - Tel 01904 455067 Email: familyfood@defra.gsi.gov.uk**

From the NDNS records rice consumption rates on a daily rate were extracted, broken down into adult, young person (4-18y) and toddler (1.5-4.5y) basis. It also enables consumption percentiles to be calculated. Table 5.3. reports the findings for the mean, 95th and 99th percentile. The 99th percentile rice consumption rate for adults at 134 g/d is lower, by 53%, than average Bangladeshi per capita rice purchase of 250.6 g/d (Table 5.1). Bangladeshis represent 0.5% of the population (Table 5.2.) and their rice consumption rates are higher than the $99th$ percentile for the UK populace, indicating that the NDNS and the EFS databases are consistent. Young people and toddlers consume more rice on a body weight basis than adults (Table 5.3.), double and 3-fold at the mean respectively, though this drops a bit at the 99th percentile. Body weights are directly based on the personal information given by respondents who reported any rice consumption that week and are not specific to the different population groups.

Ethnic rice consumption rates for children (infant, toddlers and young people) vary greatly from the norm, with mean Japanese child consumption rates being 4 times higher than the US mean, and above the US $90th$ percentile⁴. For the US, Hispanic children and babies (<1 year old) had higher rates of rice consumption than non-Hispanics, with this disparity altering as children get older⁵⁵. Thus, the high rates of toddler rice consumption at the 95 and 99th percentiles in Table 5.3. probably reflects ethic variation in the rice consumption of children.

The FSA outlines food size portions⁵⁶, expressed on a boiled weight basis (boiled:raw conversion used by the FSA is 3:1, Table 5.3.). A small boiled weight portion is 100g, medium 180 g, and large 290 g. A takeaway rice portion is 300 g.

Table 5.2. Ethnic composition of the UK in 2001. Data from National Statistics Online.

The UK data can be compared with data from the US Department of Agriculture Continuing Survey of Food Intake by Individuals (CSFII) database. Tsuji et al. 4 report graphically rice consumption rates for the US population based on the CSFII database. This data is expressed on a wet weight basis (i.e. boiled). They found that there was a very strong relationship with age for both the quantity of rice taken at any one sitting per user, and the quantity of rice ingested per capita at the 90th percentile, indicating that this 90th percentile is consuming one rice meal per day. At the $90th$ percentile the per capita and per user graph were almost identical up to the 40-49 age group where the per capita line fell below the per user line. Rice consumption peaked in 20-29 year olds with a per capita and per user consumption rate of over 150 g/d , falling to 100 or below at age 70. The mean intakes also showed this trend with the

per capita consumption being above 40 g/d for 20-29 year olds, falling to below 20 g/d for the 70+ age group.

* **Data is the sum of boiled rice and raw daily rice consumption converted into raw rice, using a 3:1 boiled:raw rice conversion factor. Body masses of 65 kg, adult; 33.8 kg, young person; 12.5 kg toddler; was used in the consumption on a body weight basis calculation. Body masses were directly recorded from respondents.**

Batres-Marquez and Jensen⁵⁷ reported rice consumption rates, on a dry weight basis, in the US based on the CSFII survey (Table 5.4.). White, non-Hispanic consumed the least amount of rice. The group "others" consisting of Asians, Pacific Islanders and Native Americans ate more than 115g d.wt. of rice per day compared to the average US intake (which included non-rice consumers over the 2 survey days) of 11.4 g/d. d.wt.. For rice consumers the average consumption rate was 61.2 g/d d.wt., over 5 times the average.

For low income population the overall average increases to 13.3g/d d.wt. while the average for rice consumers is 67.3 g/d d.wt.. However, for all groups except the "other", rice consumption falls as a percentage of these mean intakes, whilst the "other" category substantially increases. The average rice intake for the other category is 166.5 g/d d.wt.. This is lower than the 251 g/d consumed by UK Bangladeshi's (Table 5.2.).

Batres-Marquez and Jensen⁵⁷ found that being born outside the US, i.e. first generation immigrants, had higher rice consumption rates than average (Table 5.4.).

Rice consumption rates should also be considered, but to the author's knowledge are unavailable, for other target groups that have high rice intake rates. Specifically people with gluten intolerances, Celiac disease, tend to substitute wheat and other gluten containing substances with rice as rice is the most palatable alternative source of carbohydrate⁵⁸. Celiac disease is prevalent in Northern European, and affects 1 in 133 of the US population. There is a considerable market for rice products such as rice biscuits, rice wafers, rice pasta, crisped ice cereals etc. for this market (see Section 3.4.). Also, those on health conscious diets tend to eat more rice, particularly brown rice products. This is typified by macrobiotic diets, which are based on traditional Japanese diets but with an emphasis on whole grain, which have a high daily intake of rice (normally 2 meals a day), have a high reliance on fermented rice products and use rice milk as a substitute for animal milk 38 .

Table 5.4. Dry weight rice consumption rates in the US from the US CSFII database considered by ethnic origin, from Batres-Marques and Jensen53

5.3. Total diet studies and arsenic intake from rice

A Belgium based total diet study found that second to fish, that rice was the main source of arsenic to the diet, although the data presentation is limited and no breakdown by arsenic speciation was given²⁶.

Williams et al.⁵ modelled exposure to the US populace from inorganic arsenic in rice based on detailed survey of US rice samples. The study considered average rice consumption by the whole population and Asians who consume on average in 25 and

115 g d. wt. of rice per day dry weight⁵⁷. At 0.1 mg/kg inorganic arsenic in rice, Asians were consuming more than 0.01 mg/d, greater than the maximum allowed under the drinking water standard of 0.01 mg/L per day based on ingestion of 1 L of water 55 .

Schoof et al.³ conducted a US market basket survey of inorganic arsenic which "confirmed that rice has higher inorganic arsenic concentrations than most other foods. Consequently, diets that rely heavily on rice may contain the most inorganic arsenic." In this study, the inorganic arsenic content of rice greatly exceed all other foods tested, the next lowest foods (flour [unspecified], watermelon and grape juice) were 7-fold or more lower in inorganic arsenic. Fish products such as tuna and shrimp that had very high total arsenic levels had inorganic arsenic levels at least 30-fold lower than rice.

Meacher et al.¹ used the USDA's CSFII to estimate that inorganic arsenic dietary intakes of arsenic in adults, which ranged from $1.8 - 11.4 \mu\text{g/d}$ for males and 1.3-9.4 μ g/d and females at the 10th and 90th percentile respectively. Mean intake was 6.3 ± 10.6 and $5.2 \pm 9.3 \mu$ g/d for males and females respectively. The 95th percentile for both men and women is above 15.9 and 13.2 µg/d for men and women respectively. In 3 out of the 4 US regions with low arsenic in drinking water, food accounted for over 50% of dietary inorganic arsenic intakes. Indeed, in the southern region food accounted for 79% of inorganic arsenic intake. This study did not specifically breakdown exposures from individual food types.

Arsenic intake from a US Total Diet Study based on food surveys between 1991-1996 was published by Tao and Bolger²⁵. However, the arsenic levels in rice they report, 0.03-0.11 mg/kg, are very low compared to other studies (see Section 3.3.2.). Also, when calculating inorganic content of arsenic in rice they assumed that 100% of the total arsenic content was inorganic, except for seafoods where 10% inorganic was assumed. Rice has approximately 50% inorganic content (see Section 3.3.2.), where the inorganic content of seafood is much lower than 10% ³. Given these inherent weaknesses, and the fact that this type of study has been superseded $1-4,6$, the results of this study will not be considered further.

Meliker et al.² conducted a detailed study on 440 adults from the state of Michigan, USA, recording what they ate and drunk, and then used literature estimates of the inorganic arsenic content of these foods and drink to estimate dietary exposure. However, the study was not structured with respect to demography as more than 80% of participants were over 60, 87% were male and over 90% were white, so the results must be interpreted with this in mind. In particular, only 2% of the study group was African America/Black while the population of this group in Michigan counties studied was 10%. Similarly the study group contained on 2% Asian/Asian American with the actual population being 6%. In low arsenic in drinking water regions of Michigan 57% of inorganic ingestion was from food "with almost all the intake from rice". Rice consumption rates found in this study must be born in mind given the fact that this study was for a mainly old, white and male subsection of the Michigan populace. The authors found 8.1, 32.40, 32.40 and 793.8 g/day rice consumption for the 10^{th} , 50^{th} , 90^{th} and maximum intake respectively. The percentiles for inorganic arsenic content of rice used in this modelling were 0.05, 0.11, 0.18 and 0.27 mg/kg respectively, with the data obtained from $Lamont¹⁰$.

Tsuji et al.⁴ modelled exposure to the US populace again using the CSFII data and based inorganic content of arsenic in foods on Yost et al.⁶, and considered the 40 food types that made up to 90% of US exposure according to Schoof et al.³. The Foods and Residue Evaluation (FARE) model was used in modelling. At the $95th$ percentile inorganic arsenic intake from rice was 6.1 µg/d for adults, while this figure was 3.3 µg/d for children of 1-6 years, by far the highest food source, and similar to water when exposure was truncated to below 0.01 mg/L for water. The mean values for children and adults from rice were 1 and 1.67 µg/d for children and adults respectively.

No such total diet intake survey including rice has been published for the UK. The FSA54 conducted a total dietary intake of arsenic study where inorganic and total arsenic exposure was modelled using the NDNS food intake data combined with analysis of 20 food groups for total and inorganic arsenic (Table 5.4.). None of these food groups contained rice. There is a "Miscellaneous cereals" group, where details of cereals analysed are not given, but it is assumed that none of these contained rice as the maximum total arsenic levels found where 0.026 mg/kg, where the lowest rice samples analysed, Egyptian rice, contained on average 0.05 mg/kg total arsenic (Table 3.1.). Some samples of rice were included in the cereals group, although in a smaller proportion to other cereals and hence not affecting the totals. Therefore, there will be some double counting when considering rice intakes.

Table 5.5. Consumer dietary exposures to total and inorganic arsenic estimated from the 1999 Total Diet Study from FSA report 51/0458 for food items excluding with rice data calculated from this study using NDNS rice consumption data presented in Figure 5.2. and inorganic arsenic concentrations in rice ranging from 0.03 mg/kg in Indian rice to 0.12 mg/kg for Italian rice purchased in the UK (Table 3.7.). NOTE: people at the 95th percentile for inorganic arsenic **intakes excluding rice are not necessarily the same cohort of rice intake at the 95th percentile.**

The ranges of inorganic arsenic intake from rice from Table 5.5. are comparable in quantity to total inorganic arsenic intakes from the sum of 20 food groups excluding rice, accounting for approximately 50% of total inorganic arsenic consumption. This is true across all consumer groups. Rice is the dominant source of inorganic arsenic intake into the UK diet, accounting for around 30-60% of total inorganic arsenic intake.

Tsuji et al.⁴ modelled exposure to the US populace at the $95th$ percentile inorganic arsenic intake from rice was 6.1 µg/d for adults, equivalent to 0.094 µg/kg/d, assuming a 65 kg adult (Table 5.2.). This compares to a range of 0.030 – 0.119 for UK adults (Table 5.5.) at the 95th percentile. This US figure was 3.3 μ g/d for children of 1-6 years, or 0.097 µg/kg/d assuming a body weight of 33.8 kg (Table 5.2.), compared to the estimated range for the UK populace of $0.065 - 0.260 \mu$ g/kg/d. The mean values for children and adults from rice were 0.03 and 0.026 µg/d for children and adults respectively for the US Tsuji et al.⁴ study, compared to ranges of $0.018 -$ 0.071 and $0.009 - 0.035 \mu$ g/kg/d for the UK populace for children and adults respectively. Thus, the US and UK modelling of inorganic arsenic exposure from rice are within similar ranges.

Dietary exposure to inorganic arsenic was modelled for US children aged 1-6 using the FARE model using food consumption rates from the USDA's Continuing Surveys of Food Intakes by Individuals (CSFII) for 39 food types shown to account for at least 90% of US dietary intake of inorganic arsenic⁶. Mean dietary intake were $3 \mu g/d$ inorganic arsenic with a range of 1.6 - 6.2 for the $10th$ and $95th$ percentiles respectively. This exercise showed that four food groups were the primary contributors of inorganic arsenic to diet, which was grain and grain products (excluding rice) at 27.5%, fruit and fruit juices at 20.9%, dairy products at 14% and rice and rice products accounting for 19.8% . However at the $95th$ percentile, rice and rice products accounted for 49.9% of inorganic arsenic intake, that is 3.1 µg/d.

Yost et al.⁶ state that "Where a given arsenic source results in exposures within the background dietary range, any mitigation measures should be considered carefully to determine whether mitigation would result in an actual reduction in total exposure to inorganic and be a public health benefit". Yost et al.⁶ used a figure of 0.074 mg/kg for the inorganic rice content of rice (based on 4 samples), with this figure being lower than that reported in other studies for US rice (see Section 3.3.2.).

Tsuji et al.⁴ modelled inorganic arsenic exposure in children $(1-6 \text{ years})$ using the CSFII database of food consumption and inorganic arsenic levels reported in Yost et al.⁶, which itself was derived from Schoof et al.³. For inorganic arsenic in rice a value of 0.079 mg/kg was used, below the value reported by other studies (see Section 3.3.2.). They estimated that mean child arsenic exposure for the 1-6 yr age group was 3.5-3.7 μ g/d and the 90th percentile of 5.9-6.2 μ g/d inorganic arsenic.

Tao and Bolger²⁵ estimated exposure of arsenic in children from rice ingestion, but as they considered 10% of seafood arsenic was inorganic and 100% of all other foods arsenic was inorganic, this study is not reliable.

Tao and Bolger²⁵ estimated exposure of arsenic in children from rice ingestion, but for reasons outlined above, this exercise is not reliable.

5.4 Food arsenic standards

5.4.1. Focus on the risk from inorganic arsenic

Arsenic is a chronic carcinogen (as well as an acute toxin at higher concentrations) and decades of exposure to elevated levels lead to a host of illnesses including bronchitis, hypertension, miscarriage, skin hypo and hyper pigmentation, skin, bladder and lung cancers^{46,47,60,61}. It is the inorganic forms of arsenic that are thought to be of concern with respect to these illnesses, but evidence suggests that organic forms should receive more attention^{62,63}, including $DMA^{61,65,66}$. This is because it is thought that in the human body that DMA (V) can be reduced to DMA (III) by compounds such as glutathione and enzymatically via arsenic reductases. Like inorganic arsenic, it is the reduced species, or at least the redox cycling between the reduced and oxidised state, that is though to give rise to carcinogenicity⁵⁹⁻⁶². However, as is standard in the literature, and because no chronic dose response curves have been established for DMA, only exposure to inorganic arsenic will be considered.

5.4.2. Arsenic risk assessments

5.4.2.1. US

The NRC⁵⁹ established that the inorganic arsenic dose response curve for lung and bladder cancers from drinking water were linear. The risk posed by the bioavailable inorganic arsenic in foods should be identical to that derived from drinking water, with all studies to date indicating that inorganic arsenic availability from rice is high (see Section 5.1.). Theoretical maximum-likelihood estimates of excess lifetime risk for inorganic arsenic, expressed as an incidence per 10,000 people were calculated based on consumption of 1 L of water per day, based on Taiwanese epidemiological studies. This dose response modelling was used as the scientific rational of reducing arsenic in US drinking water from the old standard of 0.05 mg/L to 0.01 mg/L. The NRC⁵⁵ dose response curves are illustrated graphically in Figure 5.2.

The current US EPA skin cancer slope is 1.5 per mg/kg/d excess lifetime risk for inorganic arsenic, but a slope of 3.67 per mg/kg/d has been used in recent US EPA $assessments⁴$. The slope for internal cancers is though to be higher⁴. These revised slopes, based for a 65 kg person, and the slopes for internal cancers determined by the NRC 55 presented together in Figure 5.3..

The EPA has an upper limit of acceptable risk for cancer of 1 in $10,000^4$. At the 95th percentile a 6.1 μ g/d inorganic arsenic ingestion rate for rice calculated by Tsuji et al.⁴ US population, at a slope of 1.5 per mg/kg/d, for a 65 kg person, equates to an excess skin cancer risk of 1.4 in 10,000. Using the slope of 3.67 per mg/kg/d, this equates to an excess cancer rate of 3.4 in 10,000, for a 65 kg person. The mean arsenic ingestion rate for rice from Tsuji et al.⁴ is 1.67 μ g/d, 27% of the 95th percentile, allowing cancer rates to be adjusted accordingly, i.e. at a slope of 3.67 per mg/kg/d the excess skin cancer rate from mean levels of inorganic arsenic ingestion from rice is 0.9 in 10,000. The situation is more problematic for children $(1-6)$ with a mean and $95th$ percentile inorganic arsenic intake from rice of 1 and 3.1 µg/d, assuming an average body mass of 15 kg^{25} .

Figure 5.2. NRC⁵⁹ theoretical maximum-likelihood estimates of excess lifetime **risk, expressed as an incidence per 10,000 people calculated based on consumption of 1 L of water per day.**

Infants (6-11 months) eat more rice on a per kg body weight basis, 10 g/d wet wt. on average⁴ and have a lower body weight, 7 kg on average²⁵.

If UK Bangladeshi adults are considered, the highest UK rice consuming group for a 65 kg person, consuming 0.25 kg of rice per day (Table 5.2.), assuming an inorganic content in rice of 0.1 mg/kg for US long grain rice (Section 2.3.2.), the excess cancer risk would be 5.8 and 14.1 per 10,000 based on a slope of 1.5 and 3.67 per mg/kg/d respectively. This calculation is only for the mean. Data for other percentiles is not available, but if the maximum consumption rate for a traditional Bangladeshi diet is considered of 0.65 kg/d⁵³, this rate increases to 15 and 35.9 per 10,000 at a slope of 1.5 and 3.67 per mg/kg/d respectively. It must also be remembered that surveys of US have found maximal levels above 0.3mg/kg inorganic arsenic (Figure 3.3.).

This UK Bangladeshi calculation is based on the assumption that they are consuming rice with inorganic levels around 0.1 mg/kg, typical of US and EU rice (Figure 3.3., Table 3.7.. A personal communication from Parvez Haris (Haris per. Com.), a UK expert on food consumption by UK ethnic community, particularly Bangladeshis²³, states that around 95% of UK Bangladeshis, and 99% of Indian/Bangladeshi restaurants, use American long grain rice. Information on arsenic levels and speciation has not been conducted at present.

Figure 5.3. Current EPA cancer slopes⁴ for a 65 kg person are plotted with **NRC59 theoretical maximum-likelihood estimates of excess lifetime risk, expressed as an incidence per 10,000 people calculated based on consumption of 1 L of water per day. The WHO cancer slope is from WHO67.**

For the other UK consumers, assuming consumption of US or EU rice with an inorganic arsenic content of 0.1 mg/kg (Section 3.3.2.), based on a 65 kg body mass, the relative skin cancer rate can be extrapolated from this UK Bangladeshi risk calculation. For example, for white ethnic origin, based on ingestion of 8 g of rice per day, the UK Bangladeshi risks are multiplied by 8g/250g, resulting in an excess cancer risk of 0.2 and 0.5 in 10,000 for a 1.5 and 3.67 per mg/kg/d slope.

5.4.2.2. World Health Organization

The WHO estimated the cancer risk from arsenic using a multistage model and found that the excess lifetime skin cancer risk of 10^{-5} was estimated to be 0.17 μ g/L inorganic arsenic in drinking water 67 . They estimated that a provisional drinking water guideline value of 0.01 mg/L has an excess cancer risk of 6 in 10^{-4} (60 in 10,000) presumably based on ingestion of 1 L of water per day $(1L * 10^{-5} * 10 \mu g/L / 0.17 \mu g/L)$

 $= 5.9 \times 10^{-4}$), i.e. consumption of 10 µg/d inorganic arsenic. This is higher than the US EPA slopes (see Figure 5.3.).

The average UK Bangladeshi is consuming 25 µg arsenic per day, assuming 0.25 kg rice consumption (Table 5.2.) and a rice inorganic arsenic content of 0.1 μ g/kg (Section 3.3.2.); and this estimated excess lifetime risk from WHO calculations is 150 in 10,000.

The WHO have not ratified their Provisional Maximum Tolerable Daily Intake (PMTDI) for inorganic arsenic of 0.002 mg/kg/d which was first established in 1983⁶⁷. This equates to a cancer rate of $30 - 73.4$ in 10,000 using the lower and upper US EPA cancer slopes of 1.5 and 3.67 per mg/kg/d.

This WHO PMTDI is consistent with the WHO water standard of 0.05 mg/L, based on roughly 3 L water consumption per day for agricultural workers in hot climates. For a 65 kg person drinking this quantity of water, inorganic ingestion is 2.3 µg/kg/d. This 0.05 mg/L water standard is 5-fold higher than the EU legal limit³⁷, and assumes 3L water consumption rather than 1L per day used in developed nations⁵⁹, making the WHO PMTDI 15-fold higher than EU or US water limits in terms of exposure. Also, in 1993, the WHO suggested a provisional guideline value of 0.01 mg/ L^{67} , which it has subsequently failed to ratify 46,47 .

The EPA cancer assessment data is more up to date than that used by Joint FAO/WHO Expert Committee on Food Additives (JECFA) and WHO, as it includes human epidemiology data for lung and bladder cancer (IARC has concluded that arsenic also causes lung and bladder cancers and the Committee on Toxicology (COT) have taken the view that the WHO PTWI for inorganic arsenic might not be sufficiently protective and, therefore, the **A**s **L**ow **A**s **R**easonably **P**racticable (ALARP) principle should be used. The EPA assessment is more precautionary but has not been agreed by UK experts.

5.4.2.3. EU arsenic standards

The EU standard for arsenic, speciation not specified, in drinking water is mandatory at 0.010 mg/ L^{37} . No food standard is available. Assuming 1L of water consumption per day, the maximum arsenic consumption per day in the EU from water should be below 0.01 mg/d. The average UK Bangladeshi already exceeds this through rice intake alone by 2.5 fold for inorganic arsenic and 5 fold for total arsenic.

5.4.2.4. UK standards for arsenic in food

Food total arsenic limits in the UK are set at 1 mg/kg, irrespective of arsenic speciation, a standard that was set in 1959^{68} . Based on 50% of arsenic in rice being inorganic (Section 3.3.2.), this would set the maximum level of inorganic arsenic permissible in rice to be around 0.5 mg/kg. Again using UK Bangladeshis, the most at risk subpopulation in Britain from arsenic in rice, at 0.25 kg/d rice consumption, this equates to an inorganic arsenic intake of $125 \mu g/d$, 5-fold higher than the actual consumption rate based on a rice inorganic arsenic content of 0.1 mg/kg. The excess cancer risk, for a 65 kg person from ingesting 125 µg/d inorganic arsenic is predicted at 73.8-184.6 in 10,000 based on US cancer rate slopes of 1.5 – 3.67 in 10,000 per mg/kg/d inorganic arsenic consumed. These slopes are conservative compared to the NRC^{59} internal and WHO⁶³ cancer slopes. If the maximal traditional Bangladeshi rice

consumption rate is used of 0.65 kg/d, for a 65 kg person, at 1 mg/kg total arsenic in diet equates to a inorganic cancer rate of 192 and 480 in 10,000 using the lower and higher US EPA cancer rate slopes⁴. This is a very conservative assessment for illustration purposes regarding the consequences of using this 1 mg/kg standard, though rice at 1.8 mg/kg has been recorded in Bangladesh¹⁸.

5.4.2.5. Chinese standards for inorganic arsenic in rice

Modern food arsenic standards have been set for countries in the Far East that have high rice and fish consumption rates. The Peoples Republic of China FAIRS standards for maximum levels of arsenic in foods are reported in Table 5.6. the level for inorganic arsenic in rice is set at 0.15 mg/kg⁶⁹. From the US rice survey of Williams et al.⁵ 20% of US rice would fail this standard.

Table 5.6. Standards for the maximum levels of arsenic in foods, Peoples Republic of China64.

5.5. UK specific risk assessment and exposure to inorganic arsenic from consuming rice

The sections above outline US cancer risk assessments (Section 5.4.2.1.), WHO arsenic ingestion standards (Section 5.4.2.2.) and EU drinking water standards (Section 5.4.2.3.); the risk posed by inorganic arsenic consumption via rice in the UK in the context of these risk assessments is summarized for rice purchase data (Table 5.7.) and UK rice consumption data (Table 5.8.). The US EPA has a target goal of risk

associated with background diet, water or soil exposure of 1 in $10,000^4$. This 1 in 10,000 threshold is just exceeded for Indian rice for only the highest rice consuming group, Bangladeshis, when a slope of 1.5 per mg/kg/d is used, increasing when the higher slope of 3.67 per mg/kg/d is used (Table 5.7.). When US or Italian rice was used, more ethnic groups exceed the 1 in 10,000 threshold, even at the lower slope, with Asian-Bangladeshi and Other ethnic background greatly exceeding it.

When considered in the context of the WHO PMTDI of $2 \mu g/kg/d$, maximal contribution, again for Bangladeshis, was only 23%, but as pointed out in Section 5.4.2.2. this PMTDI is around 15 fold higher than standards set for US and EU drinking waters. Assuming ingestion of 1L of 10µg/L drinking water from domestic sources per day, i.e. the maximum level allowed under EU law³⁷, that is 10 μ g/d, inorganic arsenic exposure from rice consumption exceeds this $10 \mu g/d$ by 2-3 fold for Bangladeshis ingesting either US or Italian rice, and falls below this threshold for Indian rice for this high exposure group.

5.6. Minimizing inorganic arsenic exposure from rice

From the risk assessments conducted in Table 5.7. and 5.8 it is only high rice consumers that are at serious risk from ingesting arsenic from rice. High rice consumers most at risk are UK Bangladeshis (Table 5.7), followed by the "Others" ethnic category. When the NDNS data is modelled (Table 5.8) at the $95th$ percentile the risk posed is equivalent to the "Other Asian background" category in the EFS modelled data (Table 5.7), the 3rd highest exposure group from arsenic in rice.

It is also clear from the data presented in Table 5.8. that young people and toddlers are at higher risk, approximately 3-fold, than adults from cancer. There is no rice consumption data for infants for the UK, though from Tsuji et al.⁴, the per body weight consumption of infants is higher still than toddlers and young adults in the USA. At the $99th$ percentile, considering consumption of Italian rice, the highest inorganic arsenic content measured so far in UK purchased rice, a toddlers risk of cancer ranges from 8.6 to 21 per 10,000 based on the 1.5 and 3.67 per mg/kg/d slopes (Table 5.8)

Other high rice consuming groups not characterised in EFS and NDNS surveys, besides infants will also include people with gluten and dairy intolerances (see Chapters 3 and 5).

The simplest way to minimize inorganic exposure to these groups is to source their rice from low arsenic regions such as India, Pakistan, Thailand and Egypt (Table 3.7.). The reduction in cancer risks by consuming low inorganic arsenic rice, such as Indian, is illustrated in Tables 5.7 and 5.8. A 3- and 4-fold reduction in cancer risk rate is achievable by using Indian rice as compared to US or Italian rice respectively.

The use of the EFS household survey is justified when looking at different ethnic groups as these groups are not represented well in the NDNS survey of individuals. As the EFS data are at household level the individual is probably best estimated in the sub-populations where rice is a staple food (e.g. Asian) and less well estimated where it is more likely to be an occasional food and not necessarily consumed by the whole household (e.g. White-British).

*** Inorganic arsenic data are expressed as a percentage of WHO PMTDI of 2.14 µg/kg/d** inorganic arsenic⁶⁷ and EU water standard of 0.01 mg/L³⁷ assuming 1L drinking water **consumption per day (note that the EU standard is for total arsenic not just inorganic, thus the comparison presented here is conservative). Cancer rates are calculated from US EPA slopes of 1.5 per mg/g/d and 3.67 per mg/kg/d⁴ .**

Table 5.8. Inorganic arsenic cancer risk assessment performed using NDNS data presented in Table 5.3. using inorganic arsenic levels in US (0.09 mg/kg), Italian (0.12 mg/kg) and Indian (0.03 mg/kg) as presented in Table 3.7.

*** Inorganic arsenic data are expressed as a percentage of WHO PMTDI of 2 µg/kg/d inorganic arsenic67 and EU water standard of 0.01 mg/L37 assuming 1L drinking water consumption per day (note that the EU standard is for total arsenic not just inorganic, thus the comparison presented here is conservative). Cancer rates are calculated from US EPA slopes of 1.5 per mg/g/d and 3.67 per** $mg/kg/d^4$.

6. Suggestions for future studies

6.1. General conclusions

This review suggests areas that could be focussed on to obtain a more detailed assessment of the risks posed from inorganic arsenic ingestion from rice.

Rice is the highest source of inorganic arsenic to the UK diet. At the $50th$ and $95th$ percentiles rice accounts for around 30-60% of total dietary intake of inorganic arsenic, whilst the sum of 20 other food groups make up the rest of this contribution (Table 5.5).

Inorganic, and total, arsenic content of rice varies greatly dependent on the region of origin (Table 3.7.).

Toddlers and young people are also at a 3-fold greater cancer risk of arsenic exposure from rice compared to adults (Table 5.8.). Their rice consumption patterns are better quantified than infants.

Rice purchase data (Section 5.2.) indicates that the "average" adult populace is not at risk from arsenic consumption from rice. However, specific ethnic groups consume more rice than the UK average and their rice consumption patterns need to be defined more closely, such as rice consumption distribution curve, rice variety/country of origin preferences and rice cooking methodologies. The Bangladeshi and "Others" group according to DEFRA Expenditure and Food Survey classification have particularly high rice consumption rates (Table 5.1.) and thus higher predicted rates of arsenical disease (Section 5.6.).

The review also identified the health food market may be at risk from excessive arsenic exposure through rice consumption, particularly those adhering to macrobiotic, vegan or similar diets, or diets where rice is used as a substitute for other grains due to other grain intolerances (Section 3.4.). Not only is rice grain of concern, but the wide range of rice products that are available such as fermented rice products, milk, breakfast cereals, crackers and cereal bars. Diets that may be of concern are where rice milk is substituted for milk due to lactose intolerance or due to other dairy avoidance rationales. Rice milk arsenic levels exceed EU statutory water limits (Section 3.4).

Quantification and speciation of crisped rice products, particularly in breakfast cereals is not well defined (Section 3.4.), and may be an important source of arsenic to the diet and higher crisped rice consumption quartiles.

Surveys of arsenic levels in rice, particularly with respect to speciation, are more detailed for other countries than the UK, though a limited survey has been presented by Williams et al.¹¹. Thus a more comprehensive survey of arsenic speciation in UK rice would enable a more accurate picture of arsenic exposure from rice to be established. This survey could focus on what the general population is exposed to, and what specific high rice consuming groups (Bangladeshis, "Others", macrobiotic or other high rice health diets, children). Also, quantifying total levels and speciation in rice products, most notably crisped rice products and rice milk, will be important for certain classes of consumers.

The rice preparation habits of key rice consumers, including the "average" consumer needs to be ascertained, and then replicated under laboratory conditions to determine how this affects arsenic speciation and concentration.

Bioavailability is an issue and has yet to be resolved in detail (Section 5.1.).

6.2. Specific research suggestions

6.2.1. Rice consumption rates and geographical origin of rice consumed for subpopulations than ingest large quantities of rice

The largest gap in knowledge in assessing inorganic arsenic consumption rates from rice is not arsenic species levels in rice and rice products as these are either known or can be estimated, though some of the more unusual rice products need to be considered. The largest gap in knowledge is quantification of rice ingestion, along with the geographical origin of that rice, for high rice consuming groups. Average rates of rice consumption are available for Bangladeshis from DEFRAs Expenditure and Food Survey (Table 5.2.), but percentile data, or ideally the whole distribution curve needs to be extracted. Caution needs to be taken when looking at the percentile distribution as it is a purchase survey, and thus shoppers who buy in bulk at lower or higher than weekly shopping frequency will skew both he upper and lower percentiles. The average should be fine as irregularities in shopping frequency will be averaged out. There is also a problem with the detail of ethnic origin in the Expenditure and Food Survey database as the "Others" category, the second largest rice consuming group, is too broad and it is suspected that the range of rice consumption of the subpopulations that make up this group is considerable.

The key subpopulation is the Bangladeshis, making up 0.5% of the UK population while the "Others" group only comprises 0.4% (Table 5.3.). Thus a detailed rice consumption survey of Bangladeshis, conducted by questionnaire, would reveal actual ingestion rates on a per person basis (as opposed to rice purchased per capita averaged by household) enabling a consumption distribution curve to established, along with brand and type of rice consumed so that inorganic arsenic content of that rice can be properly quantified.

The health food/grain intolerance market consumers also needs to be quantified in terms of percent of the population, and the quantity, nature and origin of rice products consumed assessed – again by questionnaire.

For children, and infants (6-11 months) in particular, rice consumption habits also need to be assessed, including processed rice products such as crisped rice cereals. Crucial to this is assessing if there is ethnic variation in the amount of rice consumed by infants. This ethnic variation in rice consumption rates is also likely to extend to children.

Crisped cereal rice, and other rice products such as cereal bars, need to be assessed for the general consumer market, as this may be an important, and as yet unquantified, route of inorganic arsenic exposure to the genera; populace.

Recommendations:

a. Survey by questionnaire rice consuming habits of UK Bangladeshis

- **b.** Identify and survey key groups in the "Others" ethnicity category
- **c.** Survey by questionnaire rice consuming habits of UK health food/grain/dairy intolerance/avoidance market
- **d.** Survey by questionnaire rice consuming habits of UK children, with a focus on ethnic origin

This set of recommendations is a large task and maybe should form a more detailed survey. The Bangladeshi component is the most tractable and could be achieved at low cost and quickly through questionnaire.

6.2.2. Quantification of arsenic species in rice grain on the UK market

Having identified the subpopulations within the UK at risk from increased inorganic arsenic ingestion through rice consumption - Bangladeshis, "Others", health food/grain intolerance market consumers, children – speciation and quantification of arsenic should focus on grain and other rice products consumed by this market. This could be done by targeting Halal shops, Asian and Chinese supermarkets, Health food shops (including online shops), and baby, rice and health food sections of major supermarkets.

This constitutes a large body of work and for this reason could be restricted to look at the highest exposure group, namely Bangladeshis, obtaining bulk purchased rice for the Bangladeshi market from Halal and Asian supermarkets.

Recommendations:

- **a.** Survey arsenic species concentration in Bangladeshi purchased rice
- **b.** Survey arsenic species concentration in other high rice consuming groups rice grain and rice products

6.2.3. Arsenic species concentrations in rice of different origin

The proposed Bangladeshi/"other" high rice consuming groups survey could feed into a wider survey to look at the quantities of arsenic species in grains from major sources of rice imports into the UK market, as outlined in Table 3.13.. This could be done by surveying major UK supermarkets. This survey would need to consider rice type and brown and white rice, as colour has a considerable role in quantity and quality of arsenic species present in rice (Section 3.3.2.).

Recommendation:

• To survey arsenic species concentrations in rice from major UK importers

6.2.4. Alteration in concentration and speciation of arsenic during rice cooking

Volume of water used to cook rice and the arsenic content of that water can affect speciation and total arsenic content of the cooked rice (Section 4.2.). High and low volume cooking techniques, as discussed in Section 4.2. could be used to look at effects of cooking on brown and white rice. Differences in rice type (Basmati verses long grain for example) could also be explored in a factorial experiment:

 (cooking water volume) x (arsenic concentration in cooking water) x (rice colour) x (rice type)

Recommendation:

• To investigate how cooking alters rice arsenic concentration and speciation

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