

Inorganic arsenic contamination of rice from Chinese major rice-producing areas and exposure assessment in Chinese population

Xiaowei Li¹, Ke Xie^{1,2}, Bing Yue¹, Yunyun Gong^{1,3}, Yi Shao¹,
Xiaohong Shang¹ & Yongning Wu^{1,2*}

¹Key Laboratory of Food Safety Risk Assessment, Ministry of Health; China National Center for Food Safety Risk Assessment, Beijing 100022, China

²Food Science and Engineering Department, Wuhan Polytechnic University, Wuhan 430023, China

³Institute for Global Food Security, School of Biological Sciences, Queen's University of Belfast, Belfast, UK

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This study examined the total arsenic (As_t) and inorganic arsenic (As_i) content in the main rice growing area of China. The results were compared with other countries and then used for dietary exposure assessment. A total of 446 rice samples from 15 main rice-growing provinces and autonomous regions of China were collected and then divided into unpolished and polished rice. Total arsenic and arsenic species were analyzed in a total of 892 subsamples using inductively coupled plasma-mass spectrometry (ICP-MS) and high performance liquid chromatography (HPLC) coupled ICP-MS, respectively. National As_t means were 255 μg/kg of unpolished rice and 143 μg/kg of polished rice. As_i was found to be the predominant species and mean levels were 209 μg/kg of unpolished rice and 108 μg/kg of polished rice, respectively. Exposure assessment to As_i in polished rice has been calculated for the margin of exposure (MOE), which highlights the fact that As_i levels in the Chinese rice should arouse public health concern.

arsenic speciation, exposure assessment, inorganic arsenic, rice

1 Introduction

There has been a growing concern over the incidence of human exposure to arsenic (As) world wide during the past four decades [1]. Traditionally, total As concentration was considered in assessing the exposure to As. Dietary exposure assessment to inorganic arsenic (As_i) has become a serious issue recently. Today, advances in chromatographic techniques coupled to inductively coupled plasma-mass spectrometry (ICP-MS) allow arsenic speciation analyses which lead to more accurate assessment methods of different forms of arsenic. The As species commonly found in

foods include As_i and organic As. As_i, primarily arsenite [As(III)] and arsenate [As(V)], are acutely toxic and carcinogenic [2–4]. Organic As, such as dimethylarsenic acid (DMA) and methylarsonic acid (MMA), are less toxic, but have also been considered as cancer promoters [2–5]. Other species of arsenic found in food, such as arsenobetaine, arsenicholine, and arsenosugars, are considered to be non-toxic [6].

Because of its high susceptibility to arsenic contamination [7] and as one of the most consumed cereals in the world, rice is an appropriate target food for arsenic speciation analysis [8,9]. The proportion of As_i in rice compared with total As appears to vary among countries and regions [10–12]. The concentration of As in rice was influenced by

*Corresponding author (email: wuyongning@cfsa.net.cn)

the soil and water of the growing region, agricultural practices, the variety of rice (e.g. *indica* and *japonica*), and even As-containing herbicides and pesticides use [12–18].

China is the largest rice-producing country in the world and rice is the most popular food in China. The history of rice planting in China goes back to the seventh millennia [19]. The rice yield in 2011 was more than 200 million tons, and the planting area was more than 29 million hectares [20,21]. Such a high yield and broad acreage led to the growing interest in frequent monitoring of both As contamination in rice [22], in shellfish [20] and the related agricultural practices [23,24], although much effort has been made to guarantee the safety of consumers.

As_i contamination in drinking water is a significant and internationally recognized public health concern. However, high consumption of rice with high As_i levels also contributes significantly to As_i intake. A recent study in China showed that the weekly As_i intake is about 5 µg/kg (bw), 60% of which is from rice [25]. Long-term ingestion of As_i has been associated with development of severe adverse health risks including cancer, skin lesions, developmental effects, cardiovascular disease, neurotoxicity and diabetes [26]. For frequent rice consumers in Asia, average daily consumption of rice can range from 200–900 g/d [27]. For Chinese consumers, polished rice consumption per capita (reference weight 63 kg) was estimated at 238 g/d, according to the National Nutrition and Health Survey of China in 2002 [28]. At this consumption level, As_i concentration in rice as low as 42 µg/kg may equal the As_i exposure level from drinking water of 10 µg/L As_i contamination.

It is extremely important to clarify the sample collection information when performing accurate exposure assessments because the origin of rice also determines the As level. Samples collected from a market may not originate locally. In order to ensure the authenticity of the sample location, 446 paddy rice samples from major rice-producing provinces and autonomous regions were collected in the local central grain storage granaries for this study. To avoid duplication, rice samples of the same province and autonomous region were collected from different granaries. Therefore, the 446 total samples collected from granaries in 15 provinces were good representative of the real situation of rice As contamination.

2 Materials and methods

2.1 Sample sourcing and preparation

Samples were collected in the major grain-producing provinces and autonomous regions. The number of samples from each province was based on its rice production level according to figures from the National Bureau of Statistics (2011). All samples were obtained from Fujian (*n*=14), Chongqing (*n*=14), Guangdong (*n*=28), Henan (*n*=12),

Zhejiang (*n*=15), Liaoning (*n*=13), Jiangxi (*n*=51), Jiangsu (*n*=48), Sichuan (*n*=40), Hunan (*n*=69), Hubei (*n*=43), Guangxi (*n*=30), Yunnan (*n*=17), Anhui (*n*=37) and Jilin (*n*=15). The sampling locations are presented in Figure 1. Paddy rice samples were randomly coded before being processed, i.e. by drying, shelling by hulling separator (Tianjin, China), polishing with a polishing machine (Tianjin, China) in the case of polished rice, and finally powdering using a micro-plant shredder (Tianjin, China). Chinese consumers often consume unpolished rice, so we compared As concentration in polished and unpolished rice. The reliability of the processing methods was evaluated using certified reference material.

2.2 Reagents and standards

Water used throughout the experiments was ultrapure deionized water (DIW) obtained from a milli-Q system (Millipore, USA). Nitric acid was purchased from Beijing institute of Chemical Reagent (BICR, China), BV-III grade. Ammonium dihydrogen phosphate (NH₄H₂PO₄) was of analytical-reagent grade (Beijing Chemical Works, China).

Total As standard solution was a 10 mg/L multi-element

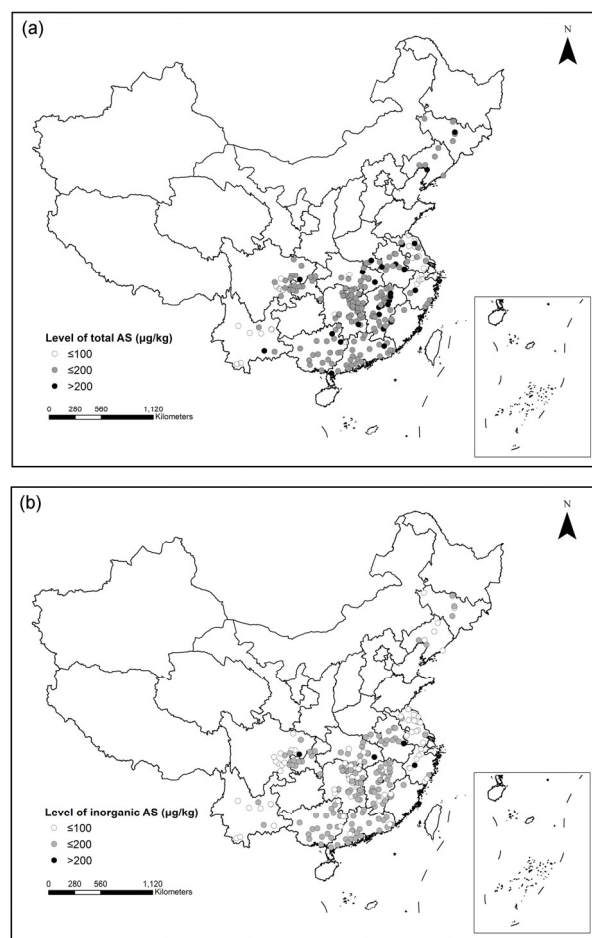


Figure 1 Geographic distribution map of arsenic levels in polished rice. (a) Total arsenic; (b) inorganic arsenic.

calibration standard 2A (Agilent technology, USA). Four different As species standard solutions were obtained from China Institute of Metrology (Beijing, China): As³⁺ (Arsenious Acid Solution GBW08666), As⁵⁺ (Arsenic Acid Solution GBW08667), MMA (Monomethylarsonic Acid Solution GBW08668), DMA (Dimethylarsinic Acid Solution GBW08669). The NIST 1568a rice flour (NIST, Gaithersburg, USA) was chosen as the Standard Reference Material (SRM).

2.3 Sample pretreatment

2.3.1 Total As

Rice samples (0.800 g each) were accurately weighed using an analytical balance and mixed with 5 mL of concentrated nitric acid for a minimum of 2 h pre-digestion. The mix was then subjected to digestion using a CEM MARS Xpress 5 microwave system (CEM, USA). During the microwave digestion, the temperature in the sample vessels was ramped to 120 °C over 5 min, held for 5 min, then the temperature was ramped to 160 °C over 5 min, then held for 5 min, at last to 190 °C over 5 min, held for 20 min, allowed to cool, then vented. Digested samples were then completely transferred into 50 mL Falcon tubes and diluted to 25 mL with ultrapure deionized water prior to the detection.

Agilent 7700x ICP-MS (Agilent technology, USA) with a pure He octopole reaction system (ORS) was used in total As analysis. No polyatomic interference as argon chloride interference was observed with this system.

2.3.2 Speciation extract and analysis

In this study we first examined and compared three widely-used extraction methods with 1% nitric acid and 3% trifluoroacetic acid condition, respectively: i.e., ultrasonic extraction at 75 °C for 2 h, microwave assisted extraction at 90 °C for 1 h, and heat-assisted ultrasound extraction at 90 °C for 2 h then ultrasonic extraction for 30 min. The extract efficiency of As_i was taken as the main criteria of method selection because As_i is the most harmful As species in rice. The results of As_i extraction (Table 1) showed no significant difference among these three methods. Heat-assisted ultrasonic extraction with 1% nitric acid was subsequently chosen as the extraction method in this study.

Table 1 Comparison of inorganic arsenic detection in NIST 1568a rice flour from different extraction methods

SRM1568a ^{a)}	Ultrasonication (75 °C, 2 h)	Microwave-assisted (90 °C, 1 h)	Heat-assisted (90 °C, 2 h)
1% (w/w) HNO ₃			
m_{As_i} (μg/kg)±SD <i>n</i> =7	112±1	114±3	115±2
3% (w/w) TFA			
m_{As_i} (μg/kg)±SD <i>n</i> =7	113±2	114±3	115±3

a) The reference amount of inorganic arsenic: 80–120 μg/kg.

Rice sample (1.000 g) was accurately weighed and steeped in 10 mL of 1% (w/w) nitric acid overnight. The resulting mixture was heated in an oven at 90 °C for 2 h, followed by ultrasonic extraction for 30 min. Then the extract was centrifuged at 8000 r/min for 5 min at 4 °C. The supernatant was filtered through a 0.45 μm nylon syringe with a polypropylene pre-filter before sampling.

The As speciation was analyzed using an Agilent 1260 high performance liquid chromatography coupled with Agilent 7700x ICP-MS. The As peaks were integrated with Mass hunter (Agilent technology, USA). The Chromatographic condition for As speciation was based on Heitkemper *et al.* [10] with minor modification. A mobile phase of 15 mmol/L NH₄H₂PO₄, pH 6.0 (adjusted with phosphate acid) at a flow rate of 1.0 mL/min was used. The column was a Hamilton PRP-X100 (Hamilton, USA) with an injection volume of 100 μL.

2.4 Quality control

2.4.1 Total As analysis

The As standards ranging from 0 to 100 ng/mL were prepared in 3% HNO₃ (v/v) in ultrapure water and used to generate calibration curves. External calibration was employed with a seven-point calibration curve to quantify total Arsenic. A 50 ng/mL Ge solution was chosen as an internal standard for Arsenic speciation determination. One QC standard was reanalyzed after every 15 samples to ensure accuracy. Two blanks and two SRM NIST 1568a rice samples were analyzed along with each batch of sample (*n*=36). The analyzed value of total As in SRM NIST 1568a was 0.30±0.01 mg/kg (*n*=20), which compares closely to the certified value of 0.29±0.03 mg/kg.

2.4.2 As speciation analysis

Quantification was achieved using external calibration, including five to seven mixed standards ranging in concentration from 0 to 50 ng/mL of each As(III), DMA, MMA, and As(V) prepared in DIW. The solution was prepared immediately before use. Calibration curves were generated by manually integrating individual peaks using manufacturer issued software (Mass hunter, Agilent technology, USA). Two blanks and two SRM 1568a rice samples were analyzed along each batch of sample to confirm extraction efficiency and monitor interspecies conversion. Recovery of As species in SRM NIST 1568a rice samples was compared with the results of other studies in the literature that analyzed the same SRM (Table 2) [22,29–31].

The total As concentration was compared with the sum of concentrations of each As species to ensure extraction efficiency (Recovery). The limit of detection (LOD) for total As was about 0.02 μg/kg which was three times the average blank value (*n*=20). The limit of quantification (LOQ) for total As was 0.06 μg/kg, which was ten times the average blank value (*n*=20). Based on the same principle the

Table 2 Comparison of arsenic species detection in NIST 1568a rice flour from different studies

Species sum ($\mu\text{g}/\text{kg}$ mean \pm SD)	As ³⁺ ($\mu\text{g}/\text{kg}$ mean \pm SD)	DMA ($\mu\text{g}/\text{kg}$ mean \pm SD)	MMA ($\mu\text{g}/\text{kg}$ mean \pm SD)	As ⁵⁺ ($\mu\text{g}/\text{kg}$ mean \pm SD)	Recovery (%)	Reference
289 \pm 4.0	73 \pm 7.0	167 \pm 8.0	5 \pm 6.0	45 \pm 5.0	99.7 \pm 1.0	Present study
271 \pm 3.0	67 \pm 5.0	162 \pm 1.0	5 \pm 1.0	36 \pm 1.0	93 \pm 1.0	[22]
272.8 \pm 9.9	63.4 \pm 3.5	144 \pm 4.5	14.9 \pm 3.9	50.3 \pm 2.9	94.1	[29]
288.2	54.7 \pm 1.4	165 \pm 8.0	14.8 \pm 1.8	53.7 \pm 3.3	99.4	[30]
286.4 \pm 6.2	68.3 \pm 3.7	135.4 \pm 4.1	8.1 \pm 1.3	20.5 \pm 2.3	82.3 \pm 1.6	[31]

LOD for As_i was 2.40 $\mu\text{g}/\text{kg}$.

Researchers responsible for this experiment participated in the Food Analysis Performance Assessment Scheme [32]. All Z-scores achieved were lower than 0.5 for total As and As_i determination in polished rice.

2.5 Statistical analysis

The total As (As_t) and As speciation levels among different provinces and autonomous regions, different types of rice were compared using student *t*-test or ANOVA test or Kruskal-Wallis Test. SPSS 13.0 was used for statistical analysis. $p < 0.05$ was considered to be statistically significant.

3 Results

3.1 Total As results

The total As concentration in rice samples is as reported in Table 3. Total As content ranges from 83 to 739 $\mu\text{g}/\text{kg}$ in unpolished rice, and from 33 to 437 $\mu\text{g}/\text{kg}$ in polished rice. Because the mean concentrations of total As in both types of rice are comparable across all provinces and autonomous regions, national means were 255 $\mu\text{g}/\text{kg}$ for unpolished rice and 143 $\mu\text{g}/\text{kg}$ for polished rice. For unpolished rice, Fujian and Chongqing had the lowest average total As levels, whilst Jilin and Jianxi the highest average. For polished rice, Jiangsu and Yunan had the lowest average total As levels, whilst Zhejiang and Henan the highest average.

3.2 As speciation results

As_i and DMA levels of all samples were above the LOD. Only one sample had a MMA level above the LOD. A summary of the speciation results is shown in Table 4. As_i concentration of unpolished rice ranged from 71 to 567 $\mu\text{g}/\text{kg}$; of polished rice from 28 to 217 $\mu\text{g}/\text{kg}$. Mean concentrations of As_i in both types of rice were comparable across all the provinces and autonomous regions. The national means were calculated at 209 $\mu\text{g}/\text{kg}$ of unpolished rice and 108 $\mu\text{g}/\text{kg}$ of polished rice. DMA concentration ranged from below LOD to 156 $\mu\text{g}/\text{kg}$ in unpolished rice, and below LOD to 128 $\mu\text{g}/\text{kg}$ in polished rice. All but one of the polished rice samples (from Jiangxi province, at 17 $\mu\text{g}/\text{kg}$) had MMA levels below LOD. The mean extraction efficiencies

were 94% and 91% in unpolished and polished rice, respectively. For unpolished rice, Fujian and Chongqing had the lowest average As_i levels, whilst Jilin and Hunan the highest average. For polished rice, Jiangsu and Yunan had the lowest average As_i levels, whilst Jiangxi and Chongqing the highest average.

There was a significant correlation between As_t and As_i concentration in both unpolished (Spearman $r=0.937$, $p < 0.001$) and polished rice (Spearman $r=0.885$, $p < 0.001$) in this study.

4 Discussion

4.1 Comparison of As levels between unpolished and polished rice

Total As mean levels in all unpolished rice were higher than in the polished rice. However, the degree of total As reduction in polished rice vs. unpolished rice varies among provinces and autonomous regions from 3.2% to 64.2% ($p < 0.001$). The polishing process involves the removal of bran from rice. Generally speaking, total As in bran is higher than in other parts of rice, although great variation among provinces exist, which is primarily owing to the diversity of the rice growing environment. Soil, water, and environmental pollution by industrial and agricultural production vary among provinces, which led to variation in As content of rice in each province and autonomous regions. That is, the different concentrations of arsenic in rice are to some extent a reflection of the regional environmental pollution. The geographic distribution of total As and As_i levels in polished rice are presented in Figure 1.

The effect of the polishing process on As reduction also varied from sample to sample, another reason for the geographical variation in total As levels. Further investigations are needed to evaluate the influence of polishing on the change of the As concentration in rice.

Zhu *et al.* [22] analysed 262 polished rice samples collected from supermarkets of China, and reported the mean total of As was 114 $\mu\text{g}/\text{kg}$, and the lowest provincial average total As level was found in Jiangsu, highest in Guangxi. The mean concentration of As for polished rice obtained by Liang *et al.* [33] was 114.4 $\mu\text{g}/\text{kg}$, which was similar to Zhu *et al.* but lower than the national means reported in this study. The provincial ranks for their total As mean levels were similar between this study and the others. The total As levels did not increase significantly between 2008 to 2014.

Table 3 Comparison of total arsenic in rice samples with varieties from different provinces and autonomous regions of China

Provinces and autonomous regions	Rice species' varieties	Grain process	Total arsenic concentration of rice grain ($\mu\text{g}/\text{kg}$)			<i>n</i>
			Mean	Median	Min-max	
Fujian	<i>Indica</i>	unpolished rice	147	139	90–314	14
		polished rice	142	127	73–268	14
Guangdong	<i>Indica</i>	unpolished rice	202	184	125–329	28
		polished rice	161	148	84–225	28
Guangxi	<i>Indica</i>	unpolished rice	302	285	171–739	30
		polished rice	151	141	95–303	30
Yunnan	<i>Indica</i>	unpolished rice	200	184	107–477	17
		polished rice	85	73	47–206	17
Chongqing	<i>Indica</i>	unpolished rice	184	182	155–218	14
		polished rice	171	169	147–186	14
Sichuan	<i>Indica</i>	unpolished rice	218	223	83–320	40
		polished rice	103	104	35–209	40
Jiangsu	<i>Japonica</i>	unpolished rice	187	175	95–341	48
		polished rice	83	82	33–166	48
Zhejiang	<i>Indica</i>	unpolished rice	277	260	183–399	15
		polished rice	190	169	87–333	15
Jiangxi	<i>Indica</i>	unpolished rice	309	296	170–619	51
		polished rice	175	166	88–437	51
Henan	<i>Japonica</i>	unpolished rice	216	206	147–387	12
		polished rice	185	172	139–285	12
Hunan	<i>Indica</i>	unpolished rice	308	297	174–730	69
		polished rice	142	138	78–256	69
Hubei	<i>Indica</i>	unpolished rice	246	259	109–341	43
		polished rice	137	146	52–210	43
Anhui	<i>Indica</i>	unpolished rice	263	248	162–397	37
		polished rice	178	170	101–305	37
Liaoning	<i>Japonica</i>	unpolished rice	196	184	164–293	13
		polished rice	171	154	128–303	13
Jilin	<i>Japonica</i>	unpolished rice	426	384	272–612	15
		polished rice	152	143	106–237	15

4.2 The total arsenic and arsenic speciation in rice

As_t and As_i concentration were found to be correlated in both unpolished and polished rice in this study (Figure 1). Interestingly, the reduction of total As concentration during polishing process was primarily due to reduction of As_i . The percentage of As_i in unpolished rice was higher than in polished rice, as also reported by Meharg *et al.* [7]. In contrast, the percentage of DMA in polished rice was greater than in unpolished rice. Overall, the losses in arsenic species between unpolished rice and polished rice were greater for inorganic arsenic than for organic arsenic.

The Chinese government has set up the maximum level (ML) for As_i contaminant in rice as 0.2 mg/kg [34]. All the provincial mean levels of As_i in polished rice were lower than this ML. However, many provincial levels of As_i in unpolished rice samples were higher than the ML. Because unpolished rice is considered to be more nutrient-rich than polished rice [35], it is more favoured and more frequently recommended by international and national food safety authorities. Thus, it is evident that the Chinese consumers may

be exposed to As_i levels above the ML. Further research is urgently required to understand the negative impacts of As exposure from unpolished rice on human health.

4.3 Inorganic arsenic concentration among different countries and regions

Williams *et al.* [12] reported As_i concentration in rice from several countries and regions: Canada (65 $\mu\text{g}/\text{kg}$, 20–110 $\mu\text{g}/\text{kg}$), India (46 $\mu\text{g}/\text{kg}$, 30–50 $\mu\text{g}/\text{kg}$), Taiwan of China (383 $\mu\text{g}/\text{kg}$, 190–760 $\mu\text{g}/\text{kg}$), and Bangladesh (131 $\mu\text{g}/\text{kg}$, 30–300 $\mu\text{g}/\text{kg}$). Additionally, Meharg *et al.* [7] reported As_i concentration in polished rice from Thailand (140 $\mu\text{g}/\text{kg}$, $n=50$), Italy (160 $\mu\text{g}/\text{kg}$, $n=28$), Spain (180 $\mu\text{g}/\text{kg}$, $n=50$), and France (280 $\mu\text{g}/\text{kg}$, $n=33$). Mean level of As_i in polished rice of China in this study is lower than France, Spain and Italy, but higher than those of Thailand, Bangladesh, Canada and India. It appears that the As_i concentration of rice in this study was lower than those of Taiwan of China, Italy, Spain and France, similar to Bangladesh and Thailand, but higher than Canada and India. This implied that As_i concen-

tration may be related to soil background and agricultural processes, which are similar around southeast Asia. However this comparison is not conclusive. The mean As_i concentrations of polished rice measured in this study were compared with other countries according to information from FAO/WHO Food Standard Programme, i.e., Codex Committee on Contaminants in Food (Table 5) [36–38]. Data of this study is lower than those of EU and Japan but higher than those of Australia and USA, possibly due to the fact that DMA is the dominant type in the USA rice whilst As_i is the dominant type in the rice of other countries.

4.4 Exposure assessment of As_i in rice

FAO/WHO Joint Expert Committee on Food Additives (JECFA) has withdrawn the previous provisional tolerance weekly intake (PTWI) for As_i with 15 µg/(week kg (bw)) as reference health standard on 2010, because the PTWI (equivalent to 2.1 µg/(d kg (bw))) is in the region of the benchmark dose lower confidence limit for 0.5% response, i.e., BMDL_{0.5} from lung cancer epidemiological study (3 µg/(d kg (bw)) with range of 2–7 µg/(d kg (bw))). Therefore, it is not a suitable approach to use the PTWI as a reference

Table 4 Comparison of arsenic species in rice samples from different provinces and autonomous regions of China

Provinces and autonomous regions	Grain fraction	DMA (µg/kg)	MMA (µg/kg)	Arsenite+arsenate (µg/kg)	Species sum (µg/kg)	Total digest As (µg/kg)	Organic As (%)	Inorganic As (%)	Extraction efficiency (%)
Fujian	unpolished rice	16	–	120	136	147	11	82	93
	polished rice	13	–	108	122	142	9	76	85
Guangdong	unpolished rice	19	–	169	188	202	9	84	93
	polished rice	16	–	131	147	161	10	81	91
Guangxi	unpolished rice	28	–	260	289	302	9	86	96
	polished rice	26	–	118	144	151	17	78	95
Yunnan	unpolished rice	20	–	175	195	200	10	88	98
	polished rice	15	–	65	81	85	18	76	95
Chongqing	unpolished rice	26	–	133	158	184	14	72	86
	polished rice	22	–	131	155	171	13	77	91
Sichuan	unpolished rice	22	–	183	206	218	10	84	94
	polished rice	15	–	82	97	103	15	80	94
Jiangsu	unpolished rice	23	–	155	178	187	12	83	95
	polished rice	19	–	66	85	90	21	73	94
Zhejiang	unpolished rice	59	–	195	255	277	21	70	92
	polished rice	32	–	120	153	190	17	63	81
Jiangxi	unpolished rice	42	–	247	290	309	14	80	94
	polished rice	24	–	135	160	175	14	77	91
Henan	unpolished rice	51	–	153	204	216	24	71	94
	polished rice	23	–	121	145	185	12	65	78
Hunan	unpolished rice	23	–	265	288	308	7	86	94
	polished rice	17	–	107	124	142	12	75	87
Hubei	unpolished rice	32	–	203	235	246	13	83	96
	polished rice	25	–	106	131	137	18	77	96
Anhui	unpolished rice	30	–	225	255	263	11	86	97
	polished rice	26	–	140	166	178	14	78	93
Liaoning	unpolished rice	30	–	154	187	199	15	77	94
	polished rice	17	–	109	128	173	10	63	74
Jilin	unpolished rice	50	–	288	377	426	12	68	88
	polished rice	28	–	115	143	152	18	75	94

Table 5 Comparison of rice As_i content (µg/kg) in different countries^{a)}

	EU	Japan	China	Australia	USA
Year	2004, 2006, 2007, 2008	2003, 2004, 2005	2011	1998	1980, 1981, 2001, 2002
<i>n</i>	142	600	446	46	60
Maximum	860	370	217	150	157
Minimum	0	40	28	15	25
Average	118.1	154.8	108.2	80	91.2
Median	104.5	150	107.8	75	95
95th	203.8	250	166	150	142.3

a) Data from CCCF, 2011, 2012.

health standard. Margin of exposure ratio (MOE) is a more suitable approach for carcinogens such as arsenic. Toxicological risk evaluations on dietary As_i exposure have been carried out by both the European Food Safety Authority (EFSA) [39] with proposed BMDL_{0.1} (from 0.3 to 8 µg/(d kg (bw))) and the JECFA (WHO) [40] with proposed BMDL_{0.5} (3 µg/(d kg (bw))), respectively. Hence, the MOE strategy was used to assess the dietary exposure of As_i in polished rice from JECFA (WHO) [40] to establish the maximum limit of As_i in polished rice by the United Nations Food and Agriculture Organization and the World Health Organization (Codex Committee on Contaminants in Food) [36–38].

The MOE value of As_i in rice was calculated based on the exposure from the estimated daily intake of As_i in rice divided by the BMDL_{0.5} suggested by the JECFA in 2010. Estimated daily intake of As_i was calculated on the basis of the level of As_i in polished and unpolished rice, respectively. Assuming that the daily polished rice consumption per capita (reference body weight 63 kg) is 238.3 g [28] for the Chinese adults, the estimated MOE value in polished rice for Chinese people ranges from 28.3 down to 3.7. The average MOE and median MOE of polished rice was 7.3 and 7.4, respectively, while the 95th percentile MOE value for polished rice was 4.8. The MOE value in unpolished rice for Chinese people ranges from 11.2 to 1.4. The average MOE and median MOE of unpolished rice was 3.8 and 3.9, respectively, while the 95th percentile MOE value for polished rice was 2.4. All the MOE values obtained in this study were under 100. It is therefore reasonable to consider that the risk of adverse effects by As_i exposure of Chinese rice is relatively high, and reduction measures should be taken. The Chinese government has established the ML for As_i in rice is 0.2 mg/kg [34]. The Codex Alimentarius Commission (CAC) also agreed with its Codex Committee on Contaminants in Food [36–38] to establish the international food standard on the ML for As_i in rice, and the work group is led by China as chaired by authors. The ML for As_i in polished rice with 0.2 mg/kg has been passed at 5/8 steps in CCCF for the CAC adoption [38].

5 Conclusions

In conclusion, the national mean levels for total As were 143 µg/kg of polished rice and 255 µg/kg of unpolished rice, respectively. As_i was the predominant species in Chinese rice, and the national mean levels were 108 µg/kg of polished rice, and 209 µg/kg of unpolished rice, respectively. The As contamination of Chinese polished rice from this study was mostly within the safety limit. However the contamination of As, and particularly the high proportion of toxic As_i in unpolished rice is a major food safety concern. These data highlight the need for research to evaluate the negative impact on human health of consumption of unpolished rice in China. Furthermore, appropriate programs

should be established for a long-term effective monitoring and reduction of As_i in Chinese rice.

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