High Percentage Inorganic Arsenic Content of Mining Impacted and Nonimpacted Chinese Rice

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Two approaches were undertaken to characterize the arsenic (As) content of Chinese rice. First, a national market basket survey ($n = 240$) was conducted in provincial capitals, sourcing grain from China's premier rice production areas. Second, to reflect rural diets, paddy rice ($n = 195$) directly from farmers fields were collected from three regions in Hunan, a key rice producing province located in southern China. Two of the sites were within mining and smeltery districts, and the third was devoid of large-scale metal processing industries. Arsenic levels were determined in all the samples while a subset ($n = 33$) were characterized for As species, using a new simple and rapid extraction method suitable for use with Hamilton PRP-X100 anion exchange columns and HPLC-ICP-MS. The vast majority (85%) of the market rice grains possessed total As levels $< 150 \text{ ng g}^{-1}$. The rice collected from mine-impacted regions, however, were found to be highly enriched in As, reaching concentrations of up to 624 ng g^{-1} . Inorganic As (As_i) was the predominant species detected in all of the speciated grain, with As_i levels in some samples exceeding 300 ng g^{-1} . The As_i concentration in polished and unpolished Chinese rice was successfully predicted from total As levels. The mean baseline concentrations for As_i in Chinese market rice based on this survey were estimated to be 96 ng g^{-1} while levels in mine-impacted areas were higher with ca. 50% of the rice in one region predicted to fail the national standard.

Introduction

Inorganic arsenic (As_i) is classified as a nonthreshold carcinogen with a linear dose–response for chronic low level exposure (1). Only relatively recently has rice, and rice-derived products, been recognized as a significant dietary source of As_i (2–15). In comparison to other cereal crops it exhibits a

particular susceptibility for As accumulation in the grain (10), levels which can be digested, absorbed, and subsequently metabolized *in vivo* (15–17). The problem is compounded by high consumption rates, typical in Asian, certain Latin American, and specific health related diets (9).

Farmers have been cultivating paddy rice (*Oryza sativa* L.) on China's east coast for approximately seven millennia (18). Rice is the predominant dietary staple in southeast Asia with China being the world's foremost producer, accounting for over one-third of global supply. However the bulk of the grain is used to meet domestic needs (~ 1.3 billion mouths), making China only the sixth largest exporter (19). Information on As_i concentrations in Chinese rice are sparse. Published studies exist regarding the As concentration of Chinese rice grain, mostly (20–23) focus on grains collected from land known to be As enriched, mainly resulting from mining or metal-processing industries requiring a posteriori knowledge of the sample area. Although of value, they do little to address the larger question concerning the safety of the average Chinese rice consumer.

Chinese standards for As_i in rice are probably the strictest in the world, which have been designed to protect a nation with high rice intakes (24). Maximum contaminant levels (MCL) are enforceable safety standards, derived from peer-reviewed data, they are not set specifically at levels known or anticipated to have no adverse health effects; rather they also account for technological feasibility and costs associated with compliance (1, 24). In China, MCLs for As_i in rice grain were originally set at 700 ng g^{-1} ; after a review of As_i exposure in 2005, this was reduced to 150 ng g^{-1} . At present, the proportion of Chinese rice that meets this requirement is undetermined.

In our study, the first of its kind, we screened market rice available in major cities for compliance with the MCL. As about half of the population in China still reside in rural settlements, we also sampled grain from local farmers in one of China's rice bowl regions (a prominent rice production zone), considering both mine-impacted and nonimpacted areas. New fast and convenient extraction and speciation methods were also developed and tested.

Material and Methods

Survey. Rice production in China is concentrated in southern and northeast regions (25). Polished (white) market rice was obtained from large cities in the southern provinces of Guangxi ($n = 39$), Guangdong ($n = 30$), Hong Kong ($n = 19$), Jiangxi ($n = 30$), Jiangsu-Shanghai ($n = 34$), Jiangsu ($n = 25$), and Guizhou ($n = 31$). Rice sourced from the northeast of China ($n = 32$) was obtained in Beijing markets and represented the provinces of Liaoning ($n = 5$), Heilongjiang ($n = 11$), Jilin ($n = 4$), and Hebei ($n = 2$) (Figure S3, Chinese map). In addition, three samples of US long grain rice were purchased in markets in Beijing as a comparison.

Hunan province is a major rice producer (26), with prolific ferrous and nonferrous metal industries that have been in operation, in some cases, for hundreds of years (27). Typical local daily rice consumption rates are among the highest in China at ca. 400 g/d wt per day (28). Two separate mining regions within Hunan and one in Daobanshan (Guangdong province, neighboring Hunan) were targeted for analysis (in total 157 samples), to determine background grain As concentrations. A region considered to be unaffected by prior mining activities was additionally sampled ($n = 38$).

Sample Preparation. Raw rice samples were washed with ultrapure water (18.2Ω), and then all grains (field and market)

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TABLE 1. Summary of Arsenic Species Characterisation and Extraction Recoveries of NIST Certified Reference Material (CRM) 1568a^a

	total As, ng g ⁻¹	As III, ng g ⁻¹	As V, ng g ⁻¹	DMA, ng g ⁻¹	MMA, ng g ⁻¹	species sum, ng g ⁻¹	recovery, %	n	refs
ultra pure water	290	49 ± 1	30 ± 2	143 ± 2	8 ± 0	230 ± 4	79 ± 1	3	
1% acetic acid	290	48 ± 7	34 ± 3	145 ± 11	9 ± 0	237 ± 22	82 ± 7	3	
1% nitric acid	290	67 ± 5	36 ± 1	162 ± 1	5 ± 1	271 ± 3	93 ± 1	3	
amylase and methanol:water	290	67 ± 4	39 ± 3	158 ± 5	13 ± 2	290 ± 2	96		*(29)
sonication probe	290	68 ± 3.7	21 ± 2	135 ± 4.1	8 ± 1	232	82 ± 4		*(30)

^a Recovery = (species sum/total As) × 100.

were oven-dried (70 °C) until a constant weight was reached. All husks were removed and the grains powdered.

Chemicals. The list of chemicals and their suppliers used in this study are detailed in the Supporting Information, page S3.

Total Digestion: Concentrated HNO₃. For concentrations of total As, oven-dried milled subsamples (0.1–0.2 g) were weighed into quartz glass digestion tubes, steeped in 2.5 mL of nitric acid, and allowed to stand overnight at room temperature. The samples were randomized prior to digestion and divided into batches consisting of ~40 samples. Heating was achieved using a hot block at 120 °C, until extracts were clear, and then made up to a volume of 10 mL with ultrapure water. GBW CRM 10010 Chinese rice flour was used to validate the analyses. Samples were again randomized prior to analysis.

Arsenic Speciation Extraction: 1% HNO₃. Three methods were evaluated, using NIST Certified Reference Material (CRM) 1568a US (Arkansas) long grain rice flour, for their suitability employed in conjunction with a microwave system, to extract As from rice grain while maintaining speciation integrity: 1% HNO₃, 1% acetic acid and ultra pure water (Table 1). On the basis of our findings, HNO₃ was selected as an extractant and subsequently tested on rice bran, husk, shoot, and root matrixes.

Milled subsamples (0.2000 g) were weighed into 50 mL polypropylene digest tubes and steeped in 10 mL of 1% nitric acid. The mixture was allowed to stand overnight. Samples were randomized and then heated in a microwave-accelerated reaction system (CEM Microwave Technology Ltd.). The temperature was gently raised, first to 55 and then to 75 °C, with holding times of 10 min. Finally the digest was taken up to 95 °C; this was maintained for 30 min before cooling. Upon reaching room temperature, samples were centrifuged at 3214g for 5 min. The supernatant was collected and passed through a 0.45 μm × 13 mm nylon filter (Membrana). To minimize any species transformation, samples were run within a few hours of filtration and kept in the dark and on ice.

NIST Certified Reference Material (CRM) 1568a US (Arkansas) long grain rice flour and GBW CRM 10010 Chinese rice flour were used to validate the analyses. Blank spikes (0.1 g of 1000 μg As g⁻¹) and sample spikes (0.200 g of US market rice, plus 0.1 g of 1000 μg As g⁻¹) for both As^{III} and As^V, and blanks were run with each extraction batch of ~35 samples. Samples were randomized prior to analysis. A subset of the speciated samples (~25%) were extracted and analyzed in duplicate to monitor extraction and analytical reproducibility.

Total Arsenic Detection for Rice Grain. Total As levels from the digest samples were derived using hydride generation atomic fluorescence spectrometry (HG-AFS) (model AF-610A, Beijing Ruili Analytical Instruments Co., Beijing, China). Samples were reduced with thiourea and hydrochloric and ascorbic acid solution before mixing with potassium borohydride to form arsenic hydrides. Arsenic peaks were integrated, using manufacturer issued software (AFS 610),

and their concentration was determined using a five-point calibration (0, 5, 10, 15, and 20 μg of As L⁻¹).

Arsenic Speciation and Total Arsenic Detection in Speciation Extracts. Arsenic speciation was assayed simultaneously by high performance liquid chromatography-inductively coupled plasma-mass spectrometry (HPLC-ICP-MS). Setup and procedures were adapted from Williams et al. (7). ICP-MS was used to determine the As content in a subset of speciation extracts. Further details are provided in the Supporting Information, pages S3–S5.

Analytical Quality Control Data. The recovery of As from Chinese rice flour CRM from total digestions were between 90–101%. The speciation recoveries ((species sum/total As) × 100) were as follows:

(i) *Extraction Trial.* NIST CRM 1568a using 1% HNO₃ 93 ± 1% (n = 3) (Table 1).

(ii) *Rice Survey CRM Recovery.* NIST CRM 1568a 96 ± 2% (n = 3), GBW CRM 10010 119 ± 5% (n = 3).

(iii) *Rice Survey Spike Recover.* (all n = 3): As^{III} blank spike 78 ± 2%, As^V blank spike 89 ± 3%, As^{III} sample spike = 82 ± 3%, As^V sample spike = 81 ± 2%.

The average relative difference, plus/minus one standard deviation ((replicate 1 – replicate 2)/(mean of replicate 1 and replicate 2) × 100) of the speciated samples extracted and analyzed in duplicate was 7 ± 6%. Further quality control data are shown in the Supporting Information (Tables S1 and S2.). All data is reported on a dry weight basis unless stated otherwise.

Results

Recoveries from As Speciation Extractions. The extraction solution strongly influenced speciation recoveries ((species sum/total As) × 100), which varied from 79% in water to 93% in 1% HNO₃. Little difference was observed between replicates (Table 1). Poor recoveries for 1% acetic acid and water could be attributed to the low levels of As^{III}, based on levels reported for NIST CRM 1568a, by Kohlmeyer et al. (29) and Sanz et al. (30) (Table 1), suggesting that these methods fail to extract all the As_i content. A solution of 1% HNO₃ proved successful in extracting As from rice shoot, bran, husk, and root, when exactly the same procedure used for the grain was employed on these plant sections, yielding recoveries of 94 ± 11 (n = 9), 91 ± 13 (n = 3), 81 ± 13 (n = 3), and 106 ± 4% (n = 6), respectively. Foster et al. (31) have successfully used 2% HNO₃ to extract As from marine and terrestrial plant matrixes, showing that extraction efficiencies were high and As^{III} and As^V species are maintained.

Market Rice Survey. Dietary Exposure to As in Chinese Cities. Differences in As levels between market rice from northern and southern China (Table 2) were not found to be significant (p = 0.077 Kruskal–Wallis). Mean concentrations between all the regions sampled were comparable, with averages only differing by a maximum of 31 ng g⁻¹ (Table 2). The lowest provincial average grain As levels were found in Hong Kong, Jiangsu, and Guizhou while Guangxi and Jiangxi were the highest. Cheng et al. (32) surveyed grain from 35 rice paddies near Shanghai, polishing the rice prior to analysis.

TABLE 2. Market Basket Survey of Arsenic Concentrations in Rice

region	province/city	total arsenic concentration of rice grain (ng g ⁻¹)				percentage of samples predicted to exceed 150 (ng As _i g ⁻¹) ^a
		mean	median	min-max	n	
far southeastern China		121	98	15-586	88	6
	Guangxi	155	127	41-586	39	10
	Guangzhou	102	97	59-245	30	3
	Hong Kong	80	83	15-138	19	0
upper southeastern China		114	114	29-250	89	1
	Jiangxi	145	145	68-250	30	3
	Shanghai	105	107	51-163	34	0
	Jiangsu	88	85	29-137	25	0
southwestern China		90	92	19-162	31	0
	Guizhou	90	92	19-162	31	0
northeast China		120	115	67-188	32	0
	Liaoning	108	89	68-162	5	0
	Heilongjiang	124	109	73-187	11	0
	Jilin	129	134	91-158	4	0
	Hebei	128	128	98-158	2	0
	northeast unknown province	118	115	67-188	10	0

^a Estimated As_i concentration derived from a linear regression of total arsenic against As_i, calculated from polished rice grain.

TABLE 3. Field Survey of Arsenic Concentrations In Rice

province/city	area ^a	mine impacted	total arsenic concentration of rice grain (ng g ⁻¹)				Percentage of samples predicted to exceed 150 (ng As _i g ⁻¹) ^b
			median	mean	min - max	n	
Hunan	a	-	159	63	9-367	38	5
	b	+	184	215	74-484	40	55
	c	+	255	303	157-624	22	28
Daobanshan(Guangdong)		+	186	190	74-447	95	23

^a Hunan areas b + and c + refer to Hunan mining regions I and II, respectively, while a - denotes the control.

^b Estimated As_i concentration derived from a linear regression of total arsenic against As_i, calculated from unpolished rice grain.

The average grain concentrations were 120 ng g⁻¹ and ranged from 60-190 ng g⁻¹. This is similar to the levels we report for this area in this study (Table 2).

The vast majority (85%) of the rice grains possessed As levels < 150 ng g⁻¹ and therefore are compliant, irrespective of As speciation, with the Chinese MCL for As_i of 150 ng g⁻¹. Only 2% exceeded 200 ng As g⁻¹ while ~50% were ≤ 100 ng As g⁻¹. The mean content of As was 114 ng g⁻¹ (Table 2), although this is lower than averages obtained from other market basket surveys by Meharg et al. (12) conducted for (non-As contaminated) Bangladesh (120 ng g⁻¹, n = 83), Thailand (140 ng g⁻¹, n = 50), Italy (160 ng g⁻¹, n = 28), Spain (180 ng g⁻¹, n = 50), Japan (190 ng g⁻¹, n = 26), USA (270 ng g⁻¹, n = 198), and France (280 ng g⁻¹, n = 33); it is still over twice that reported for Egypt (50 ng g⁻¹, n = 100), Pakistan (60 ng g⁻¹, n = 13), and India (50 ng g⁻¹, n = 100).

A study from Taiwan (33) currently provides the most information on the As content of Chinese rice; however, the survey was conducted for a specific subpopulation and therefore not necessarily applicable to other regions of China. Nearly 400 samples of Taiwanese origin were analyzed. Market (packaged) and polished rice collected from storage barns averaged 100 ng g⁻¹ fresh weight (f wt) (n = 266) and 50 ng g⁻¹ f wt (n = 137), respectively.

Field Rice Survey. Dietary Exposure to As in Hunan Province. Much of the field-collected unpolished rice was found to be considerably As enriched. The highest level was 624 ng As g⁻¹ (Table 3.). There were differences between the sample areas, with both Hunan mining regions showing larger mean As concentrations than Daobanshan and the control

zone, respectively. Hunan mining region II exhibited mean grain levels (303 ng As g⁻¹) ca. twice that of the local background (163 ng As g⁻¹) (Table 3) despite the baseline levels being higher than reported for other provinces (34). Findings from comparable surveys reveal similar trends. Arable soils close to the Dongjeong mine (South Korea) were found to be elevated in As, resulting in average grain As levels of 200 ng As g⁻¹ (n = 3) (35) while the local background was ~90 ng As g⁻¹ (36). Rice grown in the vicinity of a thallium-mercury-arsenic mine in Guizhou showed a 2- to 3-fold increase in grain As compared to a control site (37).

Speciated As Concentrations in Chinese Rice. As^{III}, As^V, DMA (dimethylarsinic acid), and MMA (monomethylarsonic acid) were the only species detected in the rice extracts. As^{III} was predominant in Chinese rice while in the US grown rice DMA prevailed (Table 4). The data was not adjusted for recoveries. In each sample measured, DMA was detected, and in three grains MMA was additionally found. NIST CRM 1568a contained MMA, but there was none in the US market rice. The most elevated samples speciated (three samples from Guangxi province) were calculated to possess As_i concentrations of 380, 329, and 353 ng As g⁻¹, being twice that of the MCL.

Predicting Grain Inorganic As Concentrations. When biplots of total As and As_i were produced (Figure S1) from our data for polished and unpolished rice, linear regressions were statistically significant and explained 94% and 72% of total variation, respectively. The resulting models were then validated by using total and As_i data from a Taiwanese survey by Schoof et al. (2) combined with Chinese rice samples

TABLE 4. Arsenic Speciation of Chinese Rice^a

origin ^b	As III, ng g ⁻¹	As V, ng g ⁻¹	DMA, ng g ⁻¹	MMA, ng g ⁻¹	species sum, ng g ⁻¹	total As, ^c ng g ⁻¹	recovery, ^d %
Guangxi (market)	302	77	102	13	495	586	84
	245	84	147	—	476	510	93
	291	63	93	8	454	550	83
	165	55	112	—	343	329	104
	86	37	31	—	153	154	100
Hong Kong (market)	84	42	38	—	164	138	119
	76	33	23	—	132	120	110
	63	34	21	—	118	106	112
Jiangxi (market)	87	38	34	7	167	164	102
	96	25	31	—	152	184	83
	87	34	28	—	150	190	79
	83	39	27	—	149	157	95
Shanghai (market)	131	42	38	—	226	187	121
	65	49	85	—	199	145	138
	51	28	31	—	109	129	85
	58	27	17	—	102	152	67
Guizhou (market)	89	52	25	—	166	150	111
	72	33	16	—	121	128	94
	47	33	9	—	88	19	471
Hunan (mine impacted)	214	43	60	—	318	484	66
	181	43	60	—	283	385	74
	171	62	11	—	244	321	76
	134	46	29	—	209	325	64
	102	30	27	—	159	201	79
	68	30	26	—	123	139	88
	76	24	10	—	111	196	56
	46	17	15	—	79	159	49
	174	51	11	—	237	243	98
Guandong (Mine Impacted)	129	37	47	—	213	249	86
	111	32	16	—	159	219	73
	83	26	47	—	156	251	62
	71	36	41	—	149	151	98
	80	35	27	—	141	131	107
	54	24	20	—	98	95	103
	63	40	18	—	121	102	119
Chinese CRM	101	51	136	—	289	350	82
US (market) bought in Beijing	95	32	141	—	269	329	82
	79	42	133	—	254	308	82

^a A subsample ($n = 10$) was extracted and analyzed in duplicate. The average relative difference, plus/minus one standard deviation, ((replicate 1 – replicate 2)/(mean of replicate 1 and replicate 2) $\times 100$) was $7 \pm 6\%$. ^b All market rice is polished while all mine-impacted rice is unpolished. ^c Total As = concd HNO₃ digestion. ^d Recovery = (species sum/total As) $\times 100$.

characterized for total and As_i (Figure S2). The linear regressions of predicted and actual As_i for polished and unpolished grain were significant ($p < 0.001$) with R^2 values of 85% and 99%, respectively, following 1:1 ratios, throughout the sample range (Figure S2).

The models were applied to the complete market basket and field surveys. Predicted means of As_i in market rice were 96 ng As_i g⁻¹ ($n = 240$) while median values were 90 ng As_i g⁻¹ with only 2% of the samples exceeding the Chinese MCL, meaning 98 were compliant. Only 2 samples collected from control sites in Hunan were predicted to exceed the Chinese MCL while in contrast 22% and 25% of the rice from Dabaoshan and Hunan mining region II, respectively, were above this level. Alarming ~50% of the rice from Hunan mining region I failed national standards based on our estimates (Table 3).

The average recoveries ((species sum/total As) $\times 100$) for polished rice were 99% with a standard error of 4. Recoveries for unpolished rice, however, were ~80%. Arsenic is concentrated in the bran layers of rice grain; therefore, unpolished can be considered to be more elevated in As than polished rice (38). On the basis of average brown to white As ratios calculated from Rahman (39), it can be estimated that bran layers increase overall grain As levels by ~20%, and that nearly all of the bran As is inorganic in nature (data unpublished).

The presented data could equate to the As_i content of the Hunan and Guandong rice that would be directly consumed, i.e. bran layer removed. When the predicted As_i levels in market rice and mine-impacted grain are compared, considerable disparity or inequality in potential dietary exposure to a recognized carcinogen is apparent (Tables 1 and 2, Figure S4).

Discussion

Arsenic species extraction from rice grain, bran, husk, shoot, and root matrixes can be achieved rapidly using 1% HNO₃ in combination with microwave heating. Our method is advantageous to other techniques such as methanol:water and TFA extraction, being more cost-effective, safer to handle, and allowing for easier disposal. Fast and effective characterization of rice As speciation is currently required, as many fundamental aspects of *in planta* As dynamics lay unresolved; for example, whether DMA and MMA grain content is controlled by cultivars, environment, or their interaction has yet to be ascertained. Figure S5. shows no obvious influence of total As on the proportion of organic As in Chinese rice, and regressions were nonsignificant. Two studies report Chinese rice as having a high proportion of total As in the organic form (8, 40). However, in light of this more extensive

survey, the proportion of As_i in the majority of Chinese rice is high (Table 4 and Figure S5) even when considerably arsenic elevated.

Based on speciation and concentration results, mean and median baseline levels of As_i in Chinese rice were determined to be 96 and 90 ng As_i g⁻¹ or at ~2/3 of the Chinese MCL. US rice bought in markets in Beijing, despite exhibiting high proportions of organic As, were found to have As_i levels averaging 132 ± 17 ng As_i g⁻¹ (not adjusted for recovery), which is marginally higher than the Chinese rice. Other work found mean levels in US rice to vary from 74 ng As_i g⁻¹ f wt to 112 ng As_i g⁻¹ f wt (3, 7, 41); however, a cautionary approach is required when using generalized average arsenic levels for US rice, as there is considerable nationwide variation in grain arsenic concentrations (9).

The reduction of US drinking water MCLs from 50 µg L⁻¹ to the WHO endorsed level of 10 µg L⁻¹ followed the largest and most comprehensive review of chronic As exposure and associated cancer risks to date (1, 42). Cancer risk models used in the assessment were specifically adjusted to consider As_i from food (1); therefore, the drinking water standards afford no direct protection against other dietary sources of As_i. Once As_i has been absorbed through intestinal membranes and enters the circulatory system, its source is immaterial. This thereby enables the US drinking water MCL for As, with its associated excess cancer risk, to be used as an appropriate measure of other dietary As_i exposures, if those sources are readily bioavailable.

US cancer risks associated with exposure to As_i in drinking water were initially modeled with a US water consumption rate of 2 L per day (42, 43); however, it was subsequently adjusted after review because it did not address differences in US water consumption patterns. National dietary surveys (CSFII) were used in simulations of entire lifetime exposure that encompassed population consumption variability, in addition to differences in gender and age; based on this, the average daily intake was determined to be 1 L (1). Therefore the daily lifetime exposure for the average US citizen from drinking water at the statutory safety limit would be 10 µg. Exposure to As_i from the daily consumption of 400 g/d wt (average rice consumption in rural Hunan) (28) of rice or rice product with a concentration of 150 ng As_i g⁻¹ could result in a dietary intake from rice alone of 60 µg, or 6 times the exposure for US citizens from drinking water at their MCL. So although China is the first and only country to presently endorse a maximum limit in grain of 150 ng As_i g⁻¹, this should not be considered an act of unnecessarily restrictive legislating. When placed in context, this standard is comparable to the now largely obsolete drinking water MCLs of 50 µg L⁻¹ once adopted by many countries, including the US, EU member states, and China.

In accordance with the US EPA who set maximum contaminant level goals (MCLG) of zero for As_i in drinking water (1), we propose the setting of an MCLG for As in rice, especially the rice intended for populations with high rice consumptions. Although complete removal of As_i from rice is unfeasible, a 5-fold reduction in the background As_i concentration of Chinese rice would result in grain levels comparable to those found in other cereal crops (10, 44). Exposure to As_i from the daily consumption of 400 g of rice or rice product with a concentration of 20 ng g⁻¹ could result in a dietary intake from rice alone of 8 µg (lower than average predicted As_i dietary exposures for the US population estimated at 10 µg per day (1)).

The risk posed to subpopulations who are consuming large amounts of rice with elevated As_i contents needs to be further addressed. In areas either naturally or anthropogenically enriched in As, exposure is already likely to be high. Wang et al. (45) found elevated levels of As in the hair and blood of a Chinese community that resided close to a smelter

district. This concurs with other studies in the southwest of England, an area renowned for metal mining (46, 47). In a recent study by Lee et al. (11) they concluded that rice, grown on contaminated soils, was the primary source of As exposure for local villagers that lived in the vicinity of an abandoned gold and silver mine (Myungbong, South Korea).

In conclusion, the vast majority of polished Chinese market rice falls well within nationally set safety standards for grain As_i, exhibiting average levels lower than a number of other major rice producing countries. However, in contrast to this, localized contamination (especially in mining regions) of rice is at present a poorly characterized exposure pathway for As_i.

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Supporting Information Available

Chemical lists, analytical methodologies, sampling maps, grain As models, summaries of speciation data, and quality control and method development data. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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