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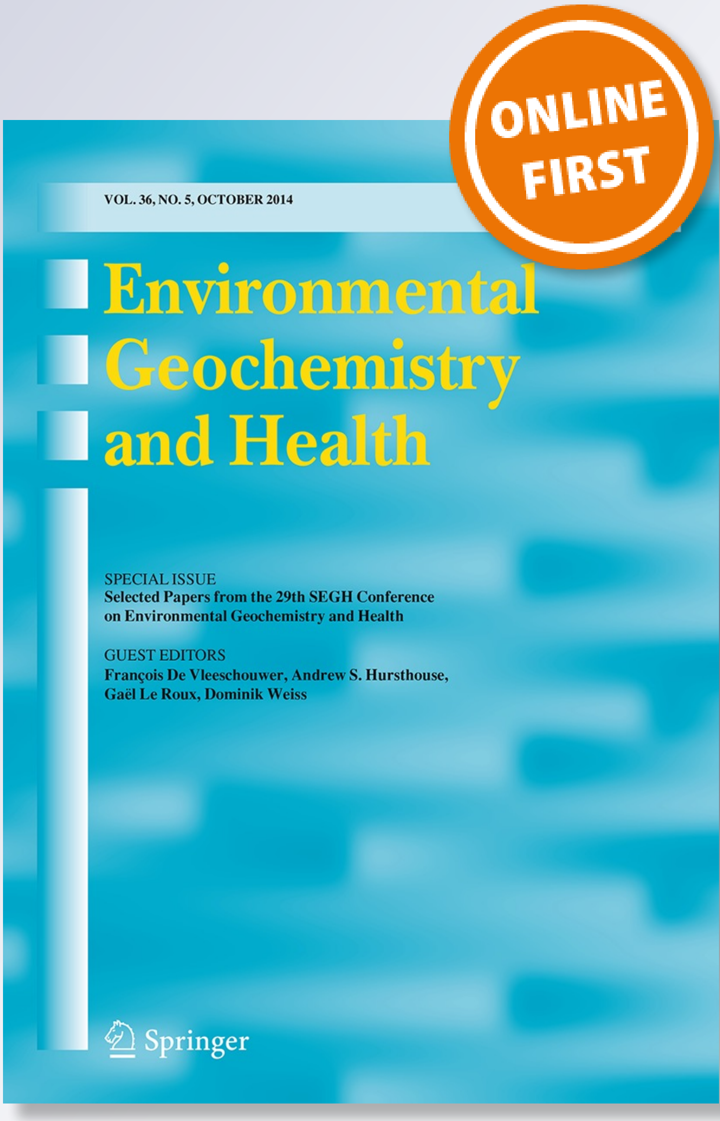

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ORIGINAL PAPER

The arsenic contamination of rice in Guangdong Province, the most economically dynamic provinces of China: arsenic speciation and its potential health risk

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Abstract Rice is a staple food in China, but it may contain toxic heavy metals. Hence, the concentrations of arsenic (As) species (As^{III}, As^{V}, MMA and DMA) were evaluated in 260 rice samples from 13 cities of Guangdong Province, the most economically dynamic provinces of China. The levels of sum concentrations of As species in rice samples varied from non-detect to 225.58 ng g^{-1}, with an average value of 57.27 ng g^{-1}. The mean concentrations of the major As species detected in rice samples were in the order As^{III} (34.77 ng g^{-1}) > As^{V} (9.34 ng g^{-1}) > DMA (8.33 ng g^{-1}) > MMA (4.82 ng g^{-1}). The rice samples of Guangdong Province were categorized as inorganic As type. Significant geographical variation of As speciation existed in rice samples of 13 cities of Guangdong Province by chi-square test ($p < 0.05$). The average human weekly intakes of inorganic As via rice consumption in Guangdong Province, southern China, were 1.91 μg kg^{-1} body weight. Hazard quotients of total As via rice consumption of adults in 13 cities ranged from 0.06 to 0.30, indicating the As contents in rice from Guangdong Province had no potential adverse impact on human health.

Keywords Arsenic speciation · Rice · Guangdong Province · Organic arsenic · Estimated weekly intake · Risk assessment

Kai Lin and Shaoyou Lu have contributed equally to this work.

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Introduction

Arsenic (As) is one of the most toxic metalloid metals that occurs in both inorganic and organic forms (Dopp et al. 2004; Hwang et al. 2010; Jomova et al. 2011), which has gained considerable attention globally in recent years due to its carcinogenic and other toxic properties (Gilbert-Diamond et al. 2011; Rahman et al. 2009). As contamination in subsoil and groundwater is a crucial problem in many countries, especially in Bangladesh, India and China (Rosen and Liu 2009). Unfortunately, contaminated groundwater irrigation was the primary reason which leads to increase of As in agronomic crops (Norton et al. 2013). Grain crops have been investigated in much detail, with respect to As accumulation (Gulz et al. 2005; Tuli et al. 2010; Zavala and Duxbury 2008). Rice is a very important agronomic crop, which provides food for around 50 % of the world's population (Meharg et al. 2009). Therefore, rice consumption is considered to be one of the main potential routes of dietary As exposure in

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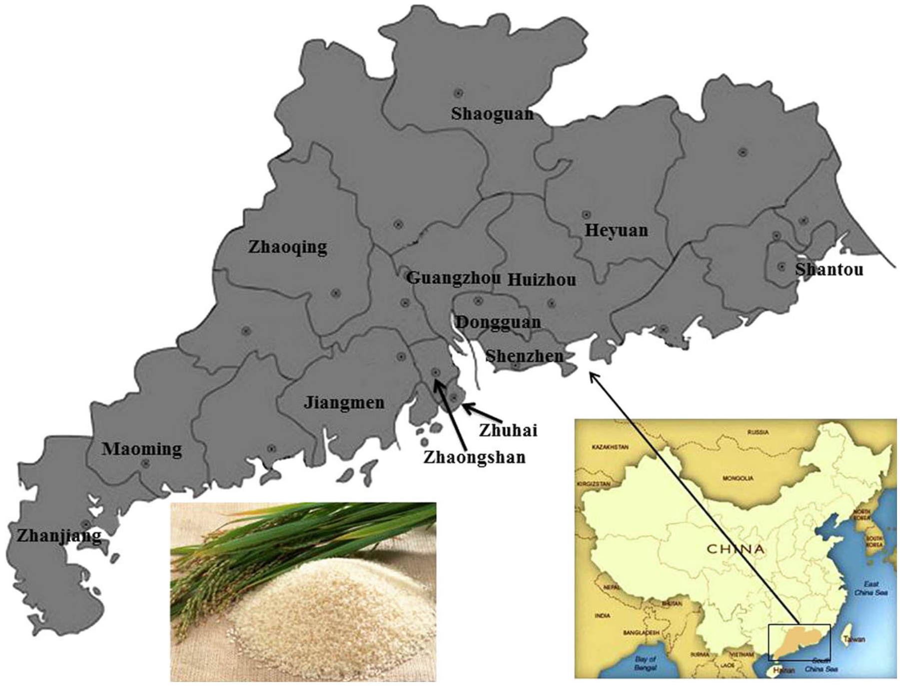

Fig. 1 Map showing the 13 cities of Guangdong Province, southern China, where the rice samples were collected

many parts of the world (Gilbert-Diamond et al. 2011; Williams et al. 2005).

As speciation accumulated in rice is important to evaluate the As levels and exposure risk assessment. Not only inorganic As species are found in rice, but organic forms of As are also detected in rice with variable proportions, such as monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) (Meharg et al. 2009). Zavala summarized the composition of As in the rice produced in different regions of the USA and found that the rice could be divided into two types: inorganic As type and DMA type (Zavala et al. 2008). Inorganic As content has been considered to be more toxic than MMA and DMA (Yamauchi 1984; Zavala et al. 2008), which is generally used to evaluate the health risk of As in rice.

In China, As concentrations in 2.2 % rice samples were above maximum allowable concentration (Fang et al. 2014). As contents in rice of southern China around an abandoned tungsten mine and industrial districts were 0.15–1.09 and 0.5–7.5 mg kg^{-1}, respectively, which posed a high health risk to human health (Liao et al. 2005; Liu et al. 2010). Guangdong Province is one of the most economically dynamic provinces in China. However, it is also one of the most heavy metal-polluted areas due to its rapid economic growth and industrial development. The geometric mean concentration of As in soil of Guangdong Province even reached 10.4 mg kg^{-1} (Zhang et al. 2006), which is higher than the mean values of 9.6 mg kg^{-1} in China (Wei 1990). In this study, 260 rice samples were collected from thirteen cities of Guangdong Province. The purpose of this study was to assess As speciation in rice from the Guangdong Province in order to improve understanding of the health risk posed by As in rice in economically developed region, southern China.

Method and materials

Sample collection and preparation

Two hundred and sixty rice samples were collected from a range of geographic market basket and field surveys throughout 13 cities of Guangdong Province, southern China (Fig. 1). Twenty samples were collected from each city. Rice samples were rinsed with deionized water to remove dust and then dried by air flow at room temperature.

Arsenic speciation extraction

Five grams of rice samples was grinded for the preparation of As speciation extraction. One gram of rice samples was transferred into 10 mL Teflon tubes and then added 10 mL HNO_3 (0.15 M) to the vessel. The sealed vessels were placed in automatic digestion instrument to decompose for 2.5 h at 90 °C. After digestion process finished, the vessels were cooled to the room temperature and centrifuged for 15 min at 8,000 rpm. The supernatants were filtered by 0.45 μm organic membrane (polyvinylidene fluoride, Millipore, USA) and further purified by C_{18} column for analysis.

$$\mathrm{EWI} = \frac{\text{Rice Consumption (g day}^{-1}) \times \text{Concentrations of As Species } (\mu\text{g g}^{-1}) \times 7 \text{ days}}{\text{Average body weight (kg)}}$$

High-performance liquid chromatography-atomic fluorescence spectrometry (HPLC-AFS) analysis

The chromatographic separation was performed at controlled room temperature (20–22 °C) connected with PRPX-100 anion-exchange column (250 mm × 4.6 mm, 30 cm, Hamilton, Switzerland). Buffer solution with mixture of 1.79 ng L^{-1} Na_2HPO_4 and 6.05 ng L^{-1} NaH_2PO_4 was used as mobile phase at a flow rate of 1.2 mL min^{-1}. Mixture of 20 ng L^{-1} KBH_4 and 5 ng L^{-1} NaOH was used as hydride reducing agent and loaded by 5 % HCl. The main and auxiliary currents of lamp of atomic fluorescence spectrometer were the 60 mA and 30 mA, respectively. Carrier gas flow rate, pump speed and negative high voltage were set up 300 mL min^{-1}, 60 rpm min^{-1} and of 320 V, respectively.

Speciation quality control

Calibrations of As speciation standards (As^{III}, As^{V}, MMA and DMA) were performed daily spanning the entire concentration range of interest. Every extracts batch included preparation blanks, laboratory control samples, matrix duplicates and matrix spikes at two levels or in duplicate. The recovery ranges of As^{III}, As^{V}, MMA and DMA were 97–111, 86–120, 79–109 and 89–105 %, respectively.

Exposure risk assessment via rice consumption

Exposure risk assessment by estimated weekly intake (EWI) of As species from rice consumption was calculated by the following equation:

The average daily consumption of rice by people and average body weights of an adult in Guangdong Province were 372 g dry weight per day and 60 kg, respectively (Ma et al. 2005; Zhuang et al. 2009). Hazard quotient (HQ) was further applied to evaluate the potential human health risk, which was the ratio of EWI and the provisional tolerable weekly intake (PTWI) of total As [15 μg kg^{-1} body weight (BW)] (JECFA 2010).

Results and discussion

The sum concentrations of As species in rice

Table 1 shows the sum concentrations of As species from different cities of Guangdong Province. The levels of sum concentrations in rice samples varied from non-detect to 225.58 ng g^{-1} with an average value of 57.27 ng g^{-1}. From the frequency histogram (Fig. 2), the sum concentrations of As species between 40 and 50 ng g^{-1} were the most frequently detected scope with nearly 40 rice samples, followed by the ranges 0–10 and 50–60 ng g^{-1}. It was also obvious that the detect frequency became significantly less as the sum concentrations of As species increased from 100 to 230 ng g^{-1}.

To understand the pollution status of As species in rice of Guangdong Province, southern China, the sum concentrations ranges of As species were compared with similar results reported in the world. As shown by Table 2, the sum concentrations of As species in

Table 1 The average values and ranges of As species determination in white rice from different cities of Guangdong Province ($N = 20$ for each city, ng g^{-1})

City	Species sum		As^{III}		As^{V}		MMA		DMA	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Zhongshan	66.50	42.06–153.23	58.05	42.06–94.59	3.59	0–43.11	3.00	0–36.47	1.87	0–41.03
Huizhou	81.51	48.74–157.05	59.04	38.24–76.44	3.26	0–35.89	4.42	0–38.22	14.79	0–97.86
Shantou	105.24	39.75–210.14	71.41	39.75–114.64	8.49	0–43.11	7.03	0–50.45	18.31	0–73.50
Jiangmen	19.20	0–73.31	13.33	0–40.59	1.54	0–33.96	2.33	0–31.71	2.00	0–31.13
Zhanjiang	36.42	0.5–82.71	13.37	0.5-46.01	9.26	0–33.96	7.19	0–33.23	6.59	0–27.79
Dongguan	26.22	7.73–70.58	15.81	0–37.73	4.64	0–42.92	2.88	0–34.64	2.90	0–54.09
Maoming	51.02	4.74–121.08	40.86	4.74–71.78	4.77	0–55.42	1.79	0–39.40	3.61	0–79.38
Guangzhou	91.43	0–255.18	32.39	0–46.89	28.75	0–82.53	15.24	0–47.09	15.04	0–62.19
Shaoguan	74.65	0–225.58	38.08	0–57.26	14.51	0–79.52	9.35	0–62.32	12.71	0–62.19
Zhaoqing	60.51	0–132.11	38.41	0–72.45	13.42	0–91.41	1.40	0–30.84	7.27	0–55.71
Zhuhai	68.76	0–138.78	38.81	0–64.51	15.02	0–95.31	6.18	0–42.52	8.75	0–62.86
Shenzhen	37.95	0–166.13	20.33	0–59.98	6.68	0–75.00	0.76	0–16.82	10.18	0–68.21
Heyuan	25.09	0–106.13	12.11	0–46.20	7.53	0–86.72	1.12	0–24.56	4.33	0–63.67
Average	57.27	0–225.58	34.77	0–114.64	9.34	0–95.31	4.82	0–62.32	8.33	0–97.86

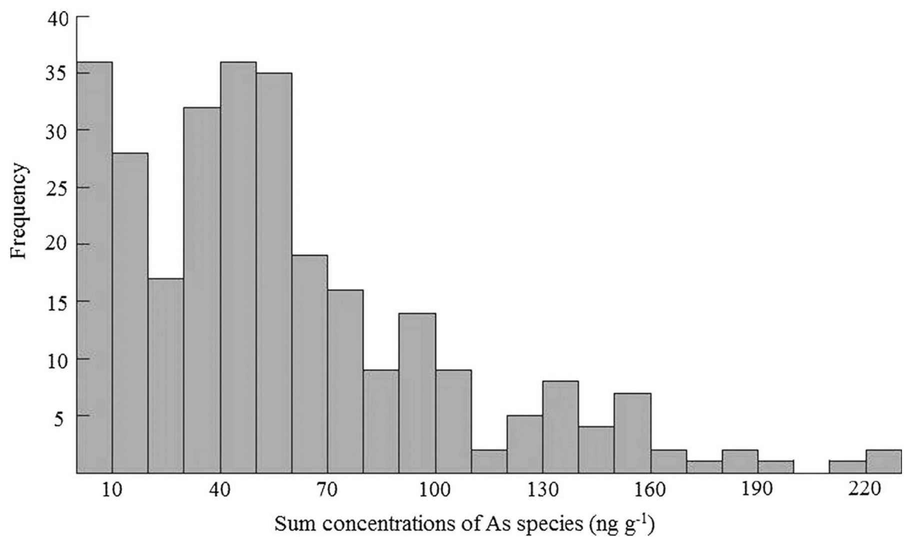

Fig. 2 Frequency histogram of the sum concentrations of As species in rice samples from Guangdong Province, southern China

Guangdong Province, southern China, in this study were higher than those in India (Williams et al. 2005), but lower than those of Brazil (Batista et al. 2011), USA (Zavala et al. 2008), Malaysia, Vietnam and Thailand (Nookabkaew et al. 2013). The As bioaccumulation in rice was affected by many factors. Pot experiments by Williams showed that the proportions of DMA^{V} in the rice are significantly dependent on rice cultivar ($p = 0.026$) and that plant nutrient status is affected by As exposure (Williams et al. 2005). In America, As pesticide application was most prevalent in the early- to mid-twentieth century. In a field trial under US growth conditions, rice sprayed with arsenical pesticides could accumulate As species up to 2,500 ng g^{-1} in the grain (Wauchope et al. 1982). In China, the amount As pesticide application was small, but they were forbidden to use in recent years. Environment pollution in mining area in China also

Table 2 Comparison of As species determination in white rice from different countries (ng g^{-1})

Samples	Species sum		As^{III}		As^{V}		MMA		DMA		References
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	
Brazil	212	98–357	78	40–156	34	16–62	8	0–29	93	39–258	Batista et al. (2011)
USA	257	169–382	86	49–122	17	3–95	1.4	0–13	155	40–302	Zavala et al. (2008)
Malaysia	97	86–116	64	59–68	7	<2–17	<2	<2–4	25	19–31	Nookabkaew et al. (2013)
Vietnam	109	80–164	86	44–130	5	<2–26	<2	<2–4	16	6–25	Nookabkaew et al. (2013)
Thailand	116	18–250	81	14–154	4	<2–8	<2	<2–6	29	2–86	Nookabkaew et al. (2013)
India	32	20–40	27	20–40	–[a]	–	66	–	0.7	–	Williams et al. (2005)
China	230	19–586	114	51–302	40	24–84	1	7–13	40	9–147	Zhu et al. (2008)
Guangdong, China	57	0–226	35	0–115	9	0–95	5	0–62	8	0–98	This study

[a] Means data were not available

played an important role in the As contamination in soil, which may be responsible for the As accumulation in rice. For example, the sum concentrations of As species of the mining impacted rice of Daobanshan, Guangdong Province, ranged from 98 to 237 ng g^{-1} with an average value of 164 ng g^{-1} (Zhu et al. 2008), which was higher than those in this study. The level of As contamination in rice samples was relatively low in this study compared with reported value in the world.

For the 13 cities where the rice samples were collected, the sum concentrations of As species were found in Shantou in the range of 39.75–210.14 ng g^{-1} with an average value of 105.24 ng g^{-1}, followed by Guangzhou and Huizhou. Average sum concentration of As species in Jiangmen (19.20 ng g^{-1}), Heyuan (25.09 ng g^{-1}) and Dongguan (26.22 ng g^{-1}) was <30 ng g^{-1}. These results indicated that significant geographical variation of As contamination existed in rice samples of Guangdong Province, southern China, by chi-square test ($p < 0.05$). This phenomenon may be related to the heavy metal pollution in these cities. For examples, Shantou is very famous for electronic waste dispose in China, and 35.6 % of soil in that area exceeded the threshold values (Yan et al. 2007). Heyuan is mainly with mountainous region and hills, and it is less contaminated by heavy metals.

As speciation

The present results of As speciation indicated that the major As species detected in all rice samples of Guangdong Province were in the order As^{III} (34.77 ng g^{-1}) > As^{V} (9.34 ng g^{-1}) > DMA (8.33 ng g^{-1}) > MMA (4.82 ng g^{-1}) (Table 1). Compared to As species concentrations in rice samples from other regions in the world, the concentrations of As^{III} and DMA in rice were at relative low level in this study (Table 2). But the concentrations of As^{V} and MMA were at relative high level. The inorganic As concentrations (As^{III} + As^{V}) of total 260 samples ranged from non-detect to 157.75 ng g^{-1} with an average value of 44.11 ng g^{-1}. Maximum level of inorganic As in polished rice (Rice without bran and aleurone layer) was proposed at 200 ng g^{-1} by the sixth session of the Codex Committee on Contaminants in Foods (CCCF 2012). Hence, inorganic As-polluted level in rice samples from Guangdong Province, southern China, was less than the suggested maximum level by the Codex Committee on Contaminants in Foods.

Table 1 shows that maximum average concentrations of As^{III} and DMA in rice samples from Shantou were 71.41 and 18.31 ng g^{-1}, respectively. The maximum average concentrations of As^{V} and MMA were detected in Guangzhou with 28.75 and 15.24 ng g^{-1}, respectively. Figure 3 shows the composition ratios of As^{III}, As^{V}, MMA and DMA, indicating that the concentrations of As^{III} in 11 cities were higher than 40 % except those of Zhanjiang and Guangzhou. Maximum proportion of As^{III} was 87.29 %, which was obtained at Zhongshan. The percentages of DMA in rice samples of 13 cities ranged from 2.80 to 26.82 %. These results further confirmed that geographical variation of As speciation existed in rice samples of Guangdong Province, southern China.

The rice could be divided into two types, depending on the form of As in the rice: inorganic As type and DMA type (Zavala et al. 2008). Hence, the correlation relationships between the As species and sum of As species for 260 rice samples are shown in Fig. 4. The rice samples of Guangdong Province were divided as

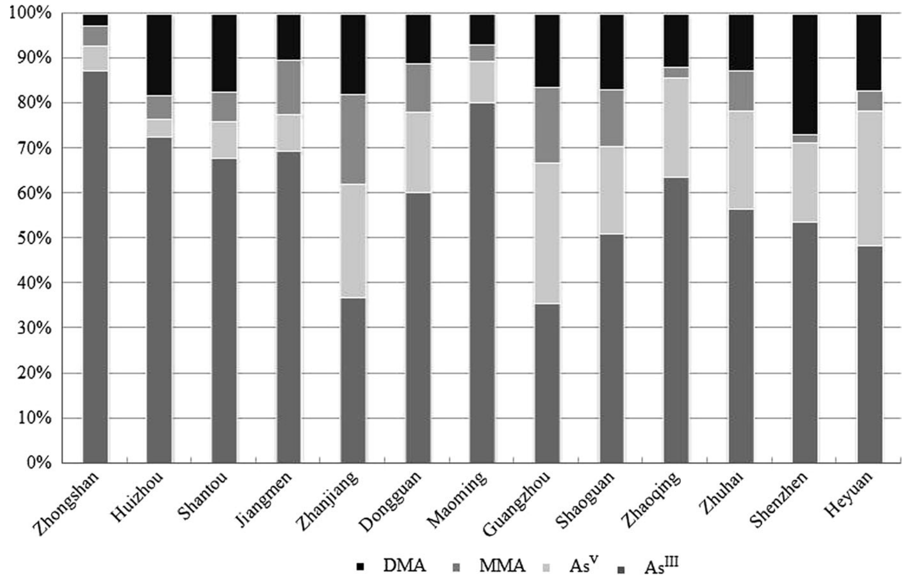

Fig. 3 Composition ratios of As^{III}, As^{V}, MMA and DMA in 260 rice samples of 13 cities from Guangdong Province, southern China

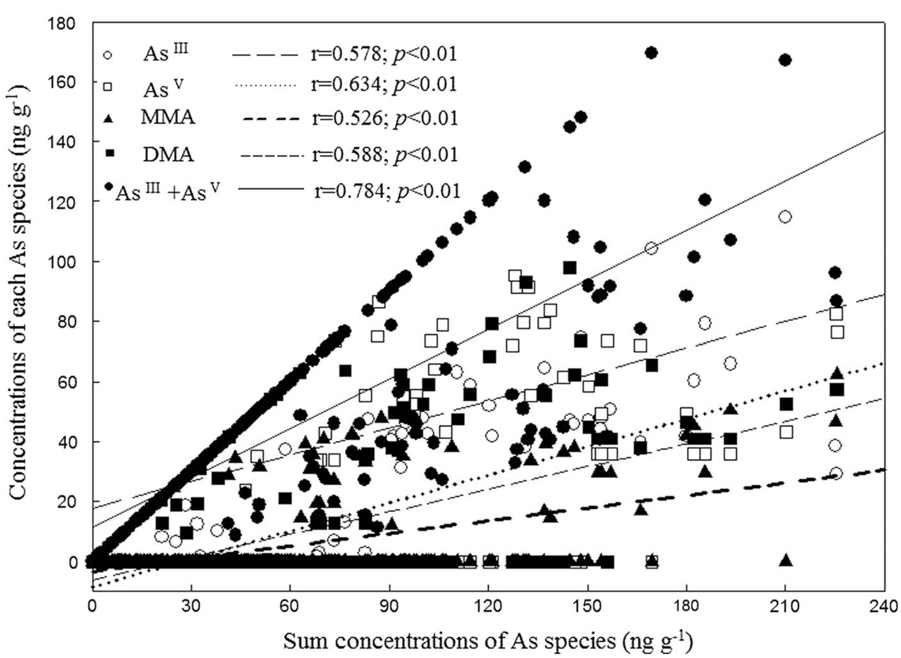

Fig. 4 Correlation between As species and sum of species in 260 rice samples

the inorganic As type since there was a high correlation between inorganic As (As^{III} and As^{V}) and the sum of As species. This agreed with the finding that most rice samples from Guangdong Province have a higher inorganic As concentration compared with DMA concentration in rice (Fig. 3). But Zavala et al. (2008) surveyed the content of As species in a single rice samples from China and categorized it to DMA-type rice. The opposite conclusions may due to the number and sites of rice samples from China.

Exposure risk assessment

Rice is the most important staple food in the China (Peterson et al. 1991). Hence, rice consumption is one of the most common exposure routes for daily intake

Table 3 Estimated weekly intake of As species and potential health risk via rice consumption

City	As^{III}		As^{V}		MMA		DMA		Species sum	Hazard quotient
	WI[a]	% of PTWI[b]	WI	% of PTWI	WI	% of PTWI	WI	% of PTWI	WI	
Zhongshan	2.52	16.80	0.16	1.04	0.13	0.87	0.08	0.54	2.89	0.19
Huizhou	2.56	17.08	0.14	0.94	0.19	1.28	0.64	4.28	3.54	0.24
Shantou	3.10	20.66	0.37	2.45	0.31	2.03	0.79	5.30	4.57	0.30
Jiangmen	0.58	3.86	0.07	0.45	0.10	0.67	0.09	0.58	0.83	0.06
Zhanjiang	0.58	3.86	0.40	2.68	0.31	2.08	0.29	1.91	1.58	0.11
Dongguan	0.69	4.57	0.20	1.34	0.12	0.83	0.13	0.84	1.14	0.08
Maoming	1.77	11.82	0.21	1.38	0.08	0.52	0.16	1.04	2.21	0.15
Guangzhou	1.41	9.37	1.25	8.32	0.66	4.41	0.65	4.35	3.97	0.26
Shaoguan	1.65	11.02	0.63	4.20	0.41	2.70	0.55	3.68	3.24	0.22
Zhaoqing	1.67	11.11	0.58	3.88	0.06	0.41	0.32	2.10	2.63	0.18
Zhuhai	1.68	11.23	0.65	4.35	0.27	1.79	0.38	2.53	2.98	0.20
Shenzhen	0.88	5.88	0.29	1.93	0.03	0.22	0.44	2.94	1.65	0.11
Heyuan	0.53	3.50	0.33	2.18	0.05	0.32	0.19	1.25	1.09	0.07
Guangdong Province	1.51	10.06	0.41	2.70	0.21	1.40	0.36	2.41	2.49	0.17

[a] *WI* weekly intake of As species (μg kg^{-1} body weight)

[b] *PTWI* provisional tolerable weekly intake (PTWI) of total As [15 μg kg^{-1} body weight (BW)] (JECFA 2010)

of heavy metals (Fu et al. 2013). The maximum level of inorganic As in rice permitted by national standard of China was 150 ng g^{-1} (GB2715-2005, China). Two rice samples of the total 260 samples having the level of inorganic As exceeded national standard of China (150 ng g^{-1}), which were both found in rice samples of Shantou. Table 3 shows the human EWI of each As species via rice consumption in 13 cities from Guangdong Province, southern China. The human weekly intakes of inorganic As ($As^{III} + As^{V}$) via rice consumption in Jiangmen (0.65 μg kg^{-1} BW) were lower than those in other regions of Guangdong Province, southern China. The highest human weekly intakes of inorganic As ($As^{III} + As^{V}$) via rice consumption were found in Shantou with a value of 3.47 μg kg^{-1} BW. The average human weekly intakes of inorganic As and organic As (MMA and DMA) via rice consumption in Guangdong Province were 1.91 and 0.57 μg kg^{-1} BW, which were about 12.76 and 3.81 % of the PTWI, respectively. The average weekly intakes of inorganic As ($As^{III} + As^{V}$) in this study were found to be lower than those by Thai (2.62 μg kg^{-1} BW; 17.45 % of the PTWI) (Nookabkaew et al. 2013), Vietnamese (7.40 μg kg^{-1} BW; 49.26 % of the PTWI) (Agusa et al. 2009) and Indian (3.71–11.13 μg kg^{-1} BW; 24.73–75.33 % of the PTWI) (Halder et al. 2012). The HQ values of total As via rice consumption of adults in 13 cities based on the mean and maximum values of total As in rice were in the range 0.06–0.30 (mean value 0.17) and 0.20–0.65 (mean value 0.41), respectively. HQ > 1 indicates potential adverse health effects (Leung et al. 2008). Therefore, the As contents in rice of Guangdong Province had no potential adverse impact on human health.

Conclusions

Guangdong Province is one of the most 14 polluted areas of heavy metals in China. Hence, in this study, the concentrations of As species in 260 rice samples from Guangdong Province, southern China, were analyzed using HPLC-AFS. The inorganic As concentrations ($As^{III} + As^{V}$) of total 260 samples ranged from non-detect to 157.75 ng g^{-1} with an average value of 44.11 ng g^{-1}. The rice samples of Guangdong Province were categorized as inorganic As type. The average human weekly intakes of inorganic As and organic As (MMA and DMA) of Guangdong Province from rice were 1.91 and 0.57 μg kg^{-1} BW, which were about 12.76 and 3.81 % of the PTWI, respectively. Among the rice samples from 13 cities, the As concentrations in rice from Shantou were highest, which may be due to the pollution from electronic waste dissembling industry. Fortunately, the local government has realized the problem and taken measures to soil remediation and reduce the pollution of heavy metals. The heavy metal in grains still needs to study in future to evaluate the risk from As exposure in rice over time.

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