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ORIGINAL ARTICLE

Rice is a potential dietary source of not only arsenic but also other toxic elements like lead and chromium

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Abstract Rice is a staple food and a good source of nutrition for half of the earth's population including Middle Eastern countries. However, rice may accumulate hazardous levels of toxic elements. In KSA, rice is imported from many countries; some of which suffer from arsenic contamination in their groundwater and soil. Despite the large daily consumption of rice in KSA, no investigations on the contamination of rice sold there are published so far. Additionally, reports on the contamination of rice with other toxic elements are rare in the literature. To investigate this issue, a total of 84 rice samples were collected from local markets in Almadinah Almunawarah, KSA ($n = 70$) and Brisbane, Australia ($n = 12$) and analyzed for arsenic and other elements by ICP-MS. The mean concentrations (mg kg^{-1}) for the KSA samples with concentrations $>$ LOQ were 0.136 for As (range 0.026–0.464, $n = 70$); Cd: 0.017 (0.003–0.046, $n = 64$); Pb: 0.029 (0.003–0.218, $n = 40$); Ni: 0.064 (0.042–0.086, $n = 5$); Mg: 157 (51.8–777, $n = 70$); Mn: 4.28 (0.960–10.9, $n = 70$); Fe: 7.07 (1.9–55.1, $n = 70$); Zn: 6.19 (1.15–13.5, $n = 70$); Cu 1.28 (0.508–2.41, $n = 70$); Se 0.202 (0.007–0.574, $n = 70$); Cr: 0.057 (0.010–0.184, $n = 19$); and Co: 0.012 (0.001–0.116, $n = 56$). Several samples were found to contain at least one element in excess of the Chinese MCL (0.2 mg $\text{kg}^{-1}$for Cd, Cr, Pb, and iAs each). A large variation in element concentration was observed for samples of different origins. In comparison, the American rice accumulated the highest arsenic concentration (mean 0.257 mg kg^{-1}) followed by the Thai rice (mean 0.200 mg kg^{-1}), the Pakistani rice (mean 0.147 mg kg^{-1}), the Indian rice (mean 0.103 mg kg^{-1}), and finally the Egyptian rice (mean 0.097 mg kg^{-1}). Additionally, 3 individual

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samples from Surinam, Australia, and France contained arsenic concentrations (mg kg^{-1}) of 0.290, 0.188, and 0.183. The findings of this investigation indicate that some of the rice varieties sold in KSA contain hazardous levels of arsenic and other toxic elements. For a better public health protection, concerned authorities are highly recommended to regularly monitor the concentrations of not only arsenic, but also other toxic elements (e.g. Cr, Cd, Pb) in rice grains.

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1. Introduction

Human beings are exposed to various types of chemical pollutants all the times. Metals and metalloids (e.g. lead, cadmium, mercury, chromium, and arsenic) are a special category of pollutants that can pose adverse health complications to living organisms (Nriagu, 1992; Stanley Manahan, 2000; Harrison, 2001; Järup, 2003). For instance, chronic exposure to lead may result in kidney damage as well as memory and metal disorders especially in children (Fewtrell et al., 2004; Lamb et al., 2008; Xie et al., 2013). Chromium on the other hand is a human carcinogen. Moreover, long-term exposure to this metal results in various health effects including oxidative stress and DNA damage (Sato et al., 2003; Nickens et al., 2010; Khan et al., 2012). Arsenic, the main focus of this work, is a metalloid that resulted in a major health concern at a global scale (Ng, Wang et al., 2003; Zhu et al., 2008). This metalloid is very mobile in the environment and occurs naturally in several organic and inorganic species that vary significantly in toxicity. Based on epidemiological human evidence, inorganic arsenicals (iAs) are classified as Group 1 human carcinogen (IARC, 1987; World Health Organization, 2001). Long term exposure to iAs, mainly in drinking water was shown to cause many illnesses including several types of cancers both in laboratory animals and humans (Ng et al., 1999; Karagas, 2001). On the other hand, many trace elements are considered as essential for the life of living organisms. However, the essentiality of such elements is valid only at certain levels of concentration, where they act as toxicants at higher levels or result in deficiency-related illnesses at lower concentrations. The essentiality vs. toxicity of several trace elements including Cr, Co, Cu, Fe, Mg, Mn, Se, and Zn has been thoroughly investigated by several scientists (Navarro-Alarcón and López-Martınez, 2000; Goldhaber, 2003; Fraga, 2005; Nielsen, 2007; Blust, 2011; Broadley et al., 2012). A summary of the essentially/toxicity of these elements is discussed in the following paragraphs with no further citation.

The essentiality and importance of Fe for the health of humans is undeniable. It is a very essential component of proteins that are involved in oxygen transport and very vital in regulating cell growth and their differentiation. Iron deficiency causes many health malfunctions such as anemia, fatigue, and decreased immunity. On the other hand exposure to excess concentrations of Fe is considered to be toxic.

Magnesium is one of several minerals that are needed in relatively large amounts by the human body mainly for healthy bones and teeth. It also plays important roles in the function of body enzymes and proteins. Deficiency in Mg may result in several health complications like muscle weakness, irregular heartbeat, high blood pressure, chronic alcoholism, and chronic diarrhea.

Zinc is an essential mineral and is largely involved in the body functions including the metabolism of the cell and is needed for the proper functioning of tens of body enzymes. It is also vital for the immune system, protein synthesis, DNA synthesis, and cell division.

Manganese is also an essential element with several benefits to the human body. It is important to the health of bones. It also acts as a co-enzyme assisting the metabolic activity in the body. Among the other benefits of Mn is its important role in the absorption of calcium and the proper functioning of the thyroid gland and sex hormones.

Selenium is a very important mineral and has many beneficial effects to the human body. This is mainly due to its antioxidant properties and its role in improving the immune system. High selenium blood levels have been liked with low rates of mortality from many types of cancer.

Copper is also one of the essential trace elements that are vital to the living organisms. It is vital to the proper functioning of body organs and its metabolic processes. Too little of this metal results in Cu deficiency with many related disorders including myelodysplastic syndrome, osteoporosis, cardiovascular disease, colon cancer, and many other chronic conditions. On the other hand exposure to excessive levels of Cu leads to health disorders such as stomach upset, nausea, diarrhea, headaches, and may cause liver and kidney damage.

Depending on its oxidation state, chromium can be either toxic or a source of minerals that is needed by the human body in trace amounts. Cr(VI) is toxic whereas Cr(III) is not. The latter is usually present in food.

Cobalt can also be beneficial for humans as it makes a major part in vitamin B12, but it can also be toxic as too high concentrations of this metal may result in negative implications to the human health.

The general population is exposed to arsenic and other toxic elements via ingestion of contaminated food or drink or through inhalation of polluted air (Shraim et al., 2003; Shrestha et al., 2003). Arsenic-contaminated water has affected the lives of hundreds of millions of people worldwide (Guo et al., 1997, 2001; Cantor et al., 2000; Kanel et al., 2005; Nickson et al., 2005; Ahamed et al., 2006; Shiber, 2007; Shinkai et al., 2007; Celik et al., 2008; Rahman et al., 2009). On the other hand, rice grains grown in arsenic-polluted soil or irrigated with arsenic-contaminated water have been recently reported to accumulate elevated levels of arsenic and can therefore contribute significantly to dietary arsenic intake (Mondal and Polya, 2008; Zavala et al., 2008; Zhu et al., 2008; Khan et al., 2009; Taskeen et al., 2009, Bhattacharya et al., 2010a,b, Rahman et al., 2010). For example, Mondal and Polya reported that the contribution of rice to the total arsenic intake in some parts of India is as high as that of arsenic-contaminated drinking water, indicating that As-tainted rice can be a significant source of arsenic (Mondal and Polya, 2008).

Noticeably, literature on the contamination of rice with toxic elements other than arsenic is very rare and interest in rice as a potential source of exposure to arsenic is very recent. Rice is a staple food for more than half the world's population around the globe as it is a good source of carbohydrates, thiamin, vitamin B6, and some essential elements like magnesium, zinc and copper. The estimated share of total calories obtained from rice is 20.5% globally, 29.2% in low-income countries and 31.6% in Asia (Wailes, 2005). The world's total production of rice in 2009 reached 682 million metric tons, 80% of which came from the following countries: PR of China (29%), India (20%), Indonesia (9%), Bangladesh (7%), Vietnam (6%), Myanmar (5%), and Thailand (5%). The largest exporter of rice in 2009 was Thailand (29% of the world's rice export) followed by Vietnam (20%), USA and Pakistan (10% each), India (8%), and the rest of countries (23%) (FAO, 2010). Interestingly, many of the rice-producing/exporting countries suffer from arsenic contamination in their groundwater or soil (Bhattacharya et al., 2010a,b; Khan et al., 2010; Liang et al., 2010; Singh et al., 2010; Fu et al., 2011; Kuramata et al., 2011; Rahman and Hasegawa 2011 Review).

Rice is grown mainly under flooded conditions, a practice that can lead to accumulation of arsenic and possibly other toxic elements in rice grains and other parts of the plant when the soil or irrigating water is polluted(van Geen et al., 2006; Khan et al., 2010). Such type of accumulation is attributed mainly to anerobic conditions in soil, which result in higher arsenic mobilization rates and increase in its bioavailability to rice (Takahashi et al., 2004; Neumann et al., 2011).

Once absorbed, the health risks associated with exposure to arsenic from rice are expected to be similar to that of arsenic that comes from contaminated water (Juhasz et al., 2006; Booth, 2009). Although both adults and children are exposed to arsenic via rice, children and babies in particular are more vulnerable to such exposure due to differences in body mass. In addition to the arsenic burden from consuming rice grains in main meals, children are exposed to arsenic through consumption of rice-based products such as rice-milk and breakfast cereals as reported by Meharg et al. (2008), where elevated levels of iAs have been reported in many baby rice-products sold in the UK.

In the Kingdom of Saudi Arabia (KSA) and most other Middle Eastern countries rice is consumed on daily bases. Moreover, rice is not grown locally but the needs of the consumers are met via import, mainly from some Asian countries where high levels of arsenic are reported in groundwater resources and soil. To date, there are no reports in the literature about the arsenic contamination in rice grains sold in KSA. Additionally, reports on the contamination of rice with toxic elements, other than arsenic, are very scarce in the literature. The aim of this work was to report on the concentrations of arsenic and other toxic elements in rice and rice-products sold in the local markets of Almadinah Almunawarah. Some rice samples were also collected from local markets in Brisbane (Australia) and others were obtained from arsenic-endemic areas in Bangladesh.

2. Experimental

2.1. Chemicals and reagents

Ultrapure water (18.2 M cm, Milli-Q, Gradient with Elix10, Millipore, USA) was used in all preparations and dilutions; nitric acid (90% fuming) was obtained from Fisher Scientific (New Jersey, USA); hydrogen peroxide (30%) was purchased from Panreac Quimica (Barcelona, Spain); ICP-MS multi-element calibration standards from Agilent Technologies (10 μg mL^{-1} each, USA); water analytical reference material (TM-25.3) from National Water Research Institute, Environment Canada; and rice certified reference material (IRMM-804) was obtained from the Institute for Reference Materials and Measurements (European Commission, Geel, Belgium).

2.2. Sample collection, digestion, and analysis

A total of 84 rice grain samples were analyzed: 70 samples were collected in Feb 2012 from local markets in Almadinah Almunawarah, KSA; 12 samples were collected in July 2010 from Brisbane, Australia; and 2 rice samples were obtained in person from arsenic-endemic areas in Bangladesh.

A representative portion (~50 g) from each sample with no prior treatment was powdered using a hand analytical mill (IKA A11 basic, Germany). About 1.0000 g of the powder was accurately weighed, placed in a Teflon vessel and digested using a microwave oven (Ethos 1 Advanced Microwave Digestion System from Milestone, Italy) as follows: 5.00 mL of conc. nitric acid was added followed by 5.00 mL of water and 1.00 mL of hydrogen peroxide (30%). The steps of the digestion program were: 1st step was run for 3 min at 80 °C, 2nd step: 9 min at 145 °C, 3rd step: 4 min at 180 °C, and last step: 15 min at 180 °C. After cooling, the clear solutions were diluted to 50.00 mL with water. With every batch of digested samples, a certified rice reference material and a blank were used and treated in the same way as the samples. About 10 mL of each digested solution was filtered through a 0.2 μm membrane filter just prior to ICP-MS analysis.

An Inductive Coupled Plasma Mass Spectrometer (ICP-MS, 7500cx, Agilent Technologies, Japan) was used for the quantitative measurement of elements. The operational parameters applied are listed in Table 1.

The figures of merit of the analytical method were assessed using limits of quantitation (LOQ), calibration verification checks, accuracy, and precision.

Blank and multi-element calibration solutions of 1.00, 2.00, 2.50, 5.00, 10.0, 25.0, 50.0, and 100.0 μg L^{-1} were used to calibrate the instrument. A calibration verification check (CVC-50) using a 50.0 μgL^{-1} solution prepared from a stock solution different from that used for the preparation of the calibration solutions was utilized. A seven-sample blank replicate analysis was used to calculate the limit of quantitation (LOQ = standard deviation (SD) ×10). The precision of the method was assessed using replicate analysis of 3 different rice samples; whereas certified reference materials were used for evaluating the accuracy.

3. Results and discussion

3.1. Quality Control

The outcome of the quality control assessment is listed in Table 2. The LOQ range for the elements of interest is 0.002–0.117 μg L^{-1}. The R^2 values for the calibration curves are better than 0.999 for all elements except for Fe (0.9981). The mean values and standard deviations for the elements in

Table 1 ICP-MS operating parameters.

Sampler	Ni, standard
Skimmer	Ni, standard
Nebulizer	Micromist, standard
Plasma torch	Quartz, 2.5 mm, standard
Integration time (s × points)	
He mode	
Mg	0.05 × 3
Cu	0.50 × 3
Cr, Ni, As, Se	1.0 × 3
Mn, Fe, Co, Zn,	0.10 × 3
No gas mode	
Cd	1.0 × 3
Pb	0.10 × 3
Tune parameters	
RF power (W)	1550
Sample depth (mm)	8.4
Carrier gas (L min^{-1})	0.95
Makeup gas (L min^{-1})	0.2
Extract 1 (V)	1.0 (2.8 for He gas reaction mode)
Extract 2 (V)	−139.5
Discriminator (mV)	8.0
Reaction gas (He, mL min^{-1})	4.0
CeO/Ce (156/140,%oxide ratio)	1.05
Ce^{++}/Ce (70/140,%doubly charged ratio)	2.12
%RSD for m/z: 7, 59, 89, 205	< 2
Spray chamber temperature (°C)	2.0
Nebulizer pump (rps)	0.1

the CVC-50 solution are 50.2–54.5 and 0.580–3.62 µgL^{-1}, respectively. The accuracy results for the two certified reference materials (rice and water) varied between 75% and 102%. On the other hand the precision values measured as %RSD for all replicate analyses (i.e. CRM's and 3 rice samples) were between 2.1% and 14.3%. Overall, the outcome of the quality control tests is good ensuring high quality results.

3.2. Concentrations of arsenic and other toxic elements in rice samples

The concentrations of elements along with a statistical summary in rice samples are listed in Table 3.

The mean arsenic concentration in the KSA 70 rice samples was 0.136 mg kg^{-1} with a standard deviation (stdev) of 0.086 mg kg^{-1}, a median of 0.107 mg kg^{-1}, and a range of 0.026–0.464 mg kg^{-1}. The arsenic concentrations were found to vary widely with the country of origin as shown in Table 4. In comparison, the American rice contained the highest arsenic concentration mean of 0.257 mg kg^{-1} followed by the Thailand rice (mean 0.200 mg kg^{-1}), then the Pakistani rice (mean 0.147 mg kg^{-1}), then the Indian rice (mean 0.103 mg kg^{-1}), and finally the Egyptian rice (mean 0.097 mg kg^{-1}). Additionally, 3 individual rice grain samples from Surinam, Australia, and France contained arsenic concentrations of 0.290, 0.188, and 0.183 mg kg^{-1}.

On the other hand, the mean arsenic concentration in the rice samples collected from Brisbane markets (refer to Table 3) is 0.092 mg kg^{-1} (n = 12, SD 0.021 mg kg^{-1}, median 0.100 mg kg^{-1}, and range 0.054–0.116 mg kg^{-1}).

To assess the suitability of food items for human consumption, their levels of elements have to be checked against relevant established maximum contaminant levels (MCLs). Interestingly, the only available MCLs for toxic elements in rice grains are those established by the PR of China (Ministry of Health, 2012). Out of the elements analyzed in this study, MCL values were reported only for cadmium, chromium, lead, and inorganic arsenic (iAs), with a value of 0.2 mg kg^{-1} each.

Rice has been shown to accumulate various forms of arsenic like arsenite, arsenate, methylarsonic acid and dimethylarsinic acid that differ in toxicity to living organisms. The first two species (iAs) are more toxic than the other two species (Hughes, 2002; Hirano et al., 2004). Although only total arsenic concentrations were determined in this investigation, special attention has be given to rice samples with arsenic concentrations > 0.200 mg kg^{-1} (i.e. 4 samples from USA with As concentrations of 0.293, 0.363, 0.463, and 0.464 mg kg^{-1}; 4

Table 2 Figures of merit of the analytical method (nc: no certified value provided).

	^{24}Mg	^{52}Cr	^{55}Mn	^{56}Fe	^{59}Co	^{60}Ni	^{63}Cu	^{66}Zn	^{75}As	^{78}Se	^{111}Cd	^{208}Pb
LOQ, µg L^{-1}, n 7	0.117	0.005	0.025	0.093	0.002	0.030	0.019	0.077	0.003	0.011	0.003	0.011
Water CRM, µg L^{-1}												
Obtained mean	2712	20.7	23.8	26.9	25.9	14.58	25.2	42.6	29.1	29.4	28.2	26.91
(SD), n 4–5	135.3	0.92	1.02	2.39	1.06	0.41	1.25	0.46	0.55	0.36	0.42	0.54
Certified mean	–	24.5	25.4	29.5	28.0	15.5	27.6	41.9	27.6	27.9	24.0	27.8
Rice CRM, mg kg^{-1}												
Obtained mean	862.3	0.106	29.73	9.87	0.013	1.75	2.50	16.63	0.046	0.029	1.20	0.427
(SD), n 4–5	(10.1)	(0.012)	(1.03)	(0.55)	(0.001)	(0.07)	(0.40)	(0.52)	(0.008)	(0.001)	(0.02)	(0.026)
Certified mean	nc	nc	34.2	nc	nc	nc	2.72	23.1	0.049	0.038)	1.61	0.42
(SD)	–	–	(2.3)	–	–	–	(0.24)	(1.9)	(0.004)	(0.004)	(0.07)	(0.07)
Precision, n 6, mg kg^{-1}												
Sample no. R72	106.9	0.048	6.48	2.37	0.009	< LOQ	0.954	3.47	0.188	0.061	< LOQ	< LOQ
Mean (SD)	(6.61)	(0.032)	(0.41)	(0.81)	(0.002)	–	(0.11)	(0.23)	(0.016)	(0.006)	–	–
Sample no. R73	130.9	< LOQ	4.03	2.38	0.008	< LOQ	0.856	4.15	0.079	0.104	0.011	< LOQ
Mean (SD)	(2.88)	–	(0.13)	(0.14)	(0.001)	–	(0.06)	(0.24)	(0.006)	(0.008)	(0.001)	–
Sample no. R74	144.2	< LOQ	3.79	2.23	0.006	< LOQ	0.987	4.06	0.074	0.131	0.016	< LOQ
Mean (SD)	(15.4)	–	(0.31)	(0.29)	(0.0004)	–	(0.07)	(0.28)	(0.005)	(0.009)	(0.001)	–

Table 3 Concentrations of elements (mg kg^{-1}) in all rice samples.

Sample no.	Grain type/color	Origin	^{24}Mg	^{52}Cr	^{55}Mn	^{56}Fe	^{59}Co	^{60}Ni	^{63}Cu	^{66}Zn	^{75}As	^{78}Se	^{111}Cd	^{208}Pb
Concentrations in rice samples collected from Almadinah Almunawarah markets, KSA														
R1	Long/White	USA	106	0.026	3.90	12.4	0.007	< LOQ	0.929	2.46	0.164	0.119	0.006	0.014
R2	Long/White	USA	98.1	< LOQ	0.964	1.95	0.007	< LOQ	0.572	1.56	0.155	0.022	< LOQ	< LOQ
R3	Medium/White	USA	208	< LOQ	8.47	2.77	< LOQ	< LOQ	1.38	6.07	0.139	0.043	0.008	< LOQ
R4	Long/White	USA	121	0.060	4.54	4.10	0.008	< LOQ	0.755	2.16	0.127	0.176	0.008	0.048
R5	Long/Yellow	USA	198	< LOQ	7.43	7.82	0.013	< LOQ	1.30	5.49	0.463	0.402	0.019	0.036
R6	Long/Yellow	USA	191	< LOQ	6.28	13.6	0.010	< LOQ	1.35	5.15	0.464	0.338	0.013	0.098
R7	Long/Yellow	USA	139	0.144	4.00	4.84	0.010	0.044	0.893	3.20	0.145	0.127	0.005	0.021
R8	Long/Yellow	USA	169	< LOQ	4.76	3.83	0.001	< LOQ	0.879	5.66	0.363	0.101	0.009	< LOQ
R9	Long/Yellow	USA	217	< LOQ	6.66	47.0	0.004	< LOQ	1.55	5.12	0.293	0.260	0.020	0.032
R11	Medium/White	Egypt	173	< LOQ	7.30	5.35	0.003	< LOQ	1.31	5.81	0.123	0.032	0.004	< LOQ
R12	Medium/White	Egypt	86.5	< LOQ	5.62	4.33	< LOQ	< LOQ	1.25	5.81	0.094	0.021	0.003	0.015
R13	Round/White	Egypt	118	< LOQ	6.21	4.75	0.007	< LOQ	1.43	6.72	0.154	0.044	0.005	0.031
R14	Round/White	Egypt	51.8	0.044	2.40	2.17	0.007	< LOQ	0.911	2.39	0.039	0.007	< LOQ	< LOQ
R15	Round/White	Egypt	123	0.010	3.32	2.01	0.008	< LOQ	1.35	3.47	0.074	0.017	< LOQ	0.017
R17	Long/Brown	India	777	0.056	10.4	11.8	0.010	0.085	1.77	7.12	0.060	0.202	0.018	0.025
R18	Long/Brown	India	732	< LOQ	10.9	11.7	0.007	< LOQ	1.66	11.0	0.089	0.205	0.032	0.015
R19	Medium/Red	India	205	< LOQ	3.07	10.3	0.020	< LOQ	0.811	2.72	0.088	0.084	0.016	0.067
R20	Long/White	India	108	0.089	1.07	4.56	0.008	0.063	1.20	1.78	0.082	0.116	0.006	0.022
R21	Long/White	India	124	0.026	1.44	2.58	0.005	< LOQ	1.17	2.00	0.085	0.258	0.005	0.031
R22	Long/White	India	168	0.079	4.35	4.46	0.006	< LOQ	1.44	4.46	0.104	0.150	0.007	0.033
R23	Long/White	India	124	0.017	3.82	2.63	0.014	< LOQ	1.15	4.97	0.026	0.167	0.011	0.014
R24	Long/White	India	156	0.022	3.01	2.99	0.004	< LOQ	1.16	4.04	0.079	0.169	0.003	0.017
R25	Long/White	India	132	0.015	4.48	2.45	0.005	< LOQ	1.40	5.49	0.061	0.253	0.014	< LOQ
R26	Long/White	India	143	0.053	1.58	3.44	0.006	0.042	1.09	1.82	0.068	0.154	0.008	0.015
R27	Long/White	India	172	< LOQ	4.80	5.82	0.019	< LOQ	1.55	8.21	0.099	0.239	0.018	< LOQ
R28	Long/White	India	159	< LOQ	5.18	6.83	< LOQ	< LOQ	1.55	9.89	0.046	0.488	0.026	< LOQ
R29	Long/White	India	121	< LOQ	3.76	2.54	< LOQ	< LOQ	1.01	5.89	0.068	0.286	0.023	< LOQ
R30	Long/White	India	155	< LOQ	4.13	2.90	< LOQ	< LOQ	1.11	7.07	0.065	0.360	0.020	< LOQ
R31	Long/White	India	95.5	0.184	1.96	55.1	0.111	0.086	1.35	1.43	0.071	0.053	0.046	0.218
R32	Long/White	India	103	0.038	1.17	3.49	0.006	< LOQ	1.08	1.85	0.104	0.223	< LOQ	0.020
R33	Long/White	India	127	< LOQ	1.44	8.09	< LOQ	< LOQ	1.05	3.85	0.220	0.415	0.006	< LOQ
R34	Long/White	India	156	< LOQ	1.52	9.75	0.004	< LOQ	1.40	3.84	0.114	0.514	0.014	0.024
R35	Long/White	India	162	< LOQ	2.26	7.60	0.002	< LOQ	1.52	5.26	0.120	0.298	0.019	0.039
R36	Long/White	India	106	< LOQ	4.54	6.18	0.002	< LOQ	1.38	8.01	0.120	0.300	0.031	< LOQ
R37	Long/White	India	168	< LOQ	4.01	7.34	0.010	< LOQ	1.51	8.34	0.100	0.413	0.023	0.014
R38	Long/White	India	133	< LOQ	4.72	5.93	0.004	< LOQ	1.62	9.30	0.145	0.285	0.025	< LOQ
R39	Long/White	India	185	< LOQ	1.77	6.23	0.003	< LOQ	1.47	4.41	0.226	0.389	0.013	0.016
R40	Long/White	India	138	< LOQ	5.17	6.53	0.005	< LOQ	1.70	9.14	0.134	0.204	0.019	< LOQ
R41	Long/White	India	163	< LOQ	1.73	8.64	0.003	< LOQ	1.33	3.75	0.212	0.273	0.014	0.019
R42	Long/White	India	222	< LOQ	5.35	5.84	< LOQ	< LOQ	1.71	9.89	0.103	0.262	0.027	< LOQ
R43	Long/White	India	218	0.081	5.62	8.57	0.027	< LOQ	1.43	8.92	0.108	0.212	0.025	0.015
R44	Long/White	India	167	< LOQ	4.89	5.54	< LOQ	< LOQ	1.54	10.5	0.097	0.343	0.030	< LOQ
R45	Long/White	India	162	< LOQ	2.23	8.62	0.003	< LOQ	1.45	4.03	0.134	0.269	0.016	0.042
R46	Long/White	India	144	< LOQ	3.79	2.23	0.006	< LOQ	0.987	4.06	0.074	0.131	0.016	< LOQ
R47	Long/White	India	184	< LOQ	4.66	5.84	< LOQ	< LOQ	1.32	9.78	0.084	0.574	0.023	0.014
R48	Long/White	India	150	< LOQ	4.57	5.02	0.004	< LOQ	1.21	8.74	0.060	0.429	0.020	< LOQ
R49	Long/White	India	139	< LOQ	5.08	7.49	0.012	< LOQ	1.49	8.47	0.066	0.171	0.022	0.003
R50	Long/Yellow	India	201	< LOQ	2.40	8.34	0.009	< LOQ	1.61	3.87	0.077	0.352	0.012	< LOQ
R51	Long/Yellow	India	177	< LOQ	1.84	7.78	0.116	< LOQ	1.19	3.83	0.105	0.306	0.034	0.015
R52	Long/Yellow	India	164	< LOQ	1.93	6.28	< LOQ	< LOQ	1.66	4.14	0.115	0.417	0.017	< LOQ
R53	Long/Yellow	India	131	< LOQ	4.03	2.38	0.008	< LOQ	0.856	4.15	0.079	0.104	0.011	< LOQ
R54	Long/Yellow	India	148	< LOQ	1.40	4.18	0.003	< LOQ	1.11	2.84	0.228	0.200	0.005	0.011
R55	Long/White	Pakistan	67.1	< LOQ	4.72	2.91	< LOQ	< LOQ	1.56	8.72	0.071	0.109	0.032	< LOQ
R56	Long/White	Pakistan	72.7	< LOQ	5.72	3.97	< LOQ	< LOQ	2.08	11.4	0.086	0.162	0.036	< LOQ
R57	Long/White	Pakistan	126	0.058	0.983	4.39	0.011	< LOQ	0.917	1.15	0.061	0.052	< LOQ	0.022
R58	Long/White	Pakistan	117	0.026	6.73	11.3	0.003	< LOQ	1.57	10.9	0.079	0.113	0.042	< LOQ
R59	Long/White	Pakistan	106	< LOQ	4.91	3.59	0.005	< LOQ	1.23	8.24	0.116	0.155	0.011	0.018
R60	Long/White	Pakistan	123	< LOQ	5.87	6.08	0.008	< LOQ	1.60	11.2	0.091	0.147	0.039	< LOQ
R61	Long/White	Pakistan	108	< LOQ	4.60	8.60	0.005	< LOQ	1.34	9.85	0.187	0.151	0.007	0.019
R62	Long/White	Pakistan	127	< LOQ	1.86	9.97	0.006	< LOQ	2.41	4.08	0.106	0.188	0.024	0.022
R63	Long/Yellow	Pakistan	167	< LOQ	2.57	5.52	< LOQ	< LOQ	1.51	2.77	0.141	0.250	0.010	< LOQ
R65	Long/White	Thailand	77.3	< LOQ	4.67	3.15	0.012	< LOQ	0.993	8.79	0.185	0.050	0.020	0.028

(*Continued*)

Table 3 (*continued*)

Sample no.	Grain type/color	Origin	^{24}Mg	^{52}Cr	^{55}Mn	^{56}Fe	^{59}Co	^{60}Ni	^{63}Cu	^{66}Zn	^{75}As	^{78}Se	^{111}Cd	^{208}Pb
R66	Long/White	Thailand	78.7	< LOQ	7.52	5.17	0.011	< LOQ	0.960	13.5	0.200	0.093	0.015	0.015
R67	Long/White	Thailand	65.6	< LOQ	4.43	3.75	0.014	< LOQ	0.902	9.35	0.181	0.063	0.007	< LOQ
R68	Long/White	Thailand	86.9	< LOQ	6.16	6.65	0.018	< LOQ	0.931	10.7	0.195	0.075	0.007	0.012
R69	Long/White	Thailand	101	< LOQ	8.09	4.57	0.022	< LOQ	0.626	11.5	0.250	0.100	0.008	< LOQ
R70	Long/White	Thailand	83.1	< LOQ	4.13	7.79	0.023	< LOQ	0.508	7.05	0.190	0.055	0.013	0.022
R10	Medium/White	Australia	107	0.048	6.48	2.37	0.009	< LOQ	0.954	3.47	0.188	0.061	< LOQ	< LOQ
R64	Long/White	Surinam	156	< LOQ	3.67	5.65	0.010	< LOQ	1.21	12.4	0.290	0.096	0.008	0.018
R16	Long/White	France	182	< LOQ	4.65	4.95	< LOQ	< LOQ	1.56	8.61	0.183	0.247	0.013	< LOQ
Statistical summary for rice samples collected from Almadinah Almunawarah markets, KSA[a]														
		n	70	19	70	70	56	5	70	70	70	70	64	40
		mean	157	0.057	4.28	7.07	0.012	0.064	1.28	6.19	0.136	0.202	0.017	0.029
		SD	111	0.045	2.17	8.11	0.020	0.021	0.339	3.21	0.086	0.133	0.010	0.035
		median	139	0.048	4.45	5.53	0.007	0.063	1.32	5.57	0.107	0.174	0.015	0.020
		min	51.8	0.010	0.96	1.95	0.001	0.042	0.508	1.15	0.026	0.007	0.003	0.003
		max	777	0.184	10.9	55.1	0.116	0.086	2.41	13.5	0.464	0.574	0.046	0.218
Concentration in rice samples collected from Brisbane, Australia														
R71	Medium/White	Australia	162	0.063	11.9	3.68	0.017	1.64	2.55	10.2	0.106	0.066	0.034	0.055
R72	Medium/White	China	156	0.075	8.36	4.27	0.018	1.64	2.30	10.9	0.093	0.032	0.002	0.296
R73	Medium/White	China	95.3	0.390	11.8	3.57	0.013	1.66	1.73	8.65	0.104	0.028	0.013	0.016
R74	Medium/White	China	133	0.094	7.85	4.45	0.015	1.76	1.63	10.5	0.064	0.028	0.028	0.354
R75	Long/White	Pakistan	121	0.065	7.04	3.52	0.017	1.77	3.94	10.8	0.065	0.079	0.043	0.025
R76	Long/White	Thailand	72.7	0.055	6.13	1.75	0.017	1.64	1.54	10.8	0.101	0.035	0.019	0.025
R77	Long/White	Thailand	89.0	0.130	7.34	3.98	0.026	1.63	1.61	10.9	0.112	0.056	0.150	0.300
R78	Long/White	Thailand	63.7	0.095	6.15	2.59	0.017	1.54	1.93	10.6	0.099	0.032	0.011	0.236
R79	Long/White	Thailand	109	0.047	10.6	6.47	0.019	1.76	1.27	13.0	0.080	0.031	0.046	0.054
R80	Long/White	Thailand	96.6	0.033	8.77	2.70	0.022	3.01	5.32	18.3	0.116	0.040	0.015	0.019
R81	Medium/White	USA	155	0.041	13.1	3.67	0.017	2.13	6.23	13.5	0.106	0.030	0.008	0.041
R82	Medium/White	USA	216	0.075	14.8	2.99	0.017	1.72	2.53	8.44	0.054	0.040	0.095	0.019
Statistical summary for rice grain samples collected from Brisbane, Australia[a]														
		n	12	12	12	12	12	12	12	12	12	12	12	12
		mean	123	0.097	9.49	3.64	0.018	1.83	2.72	11.4	0.092	0.041	0.039	0.120
		SD	44	0.096	2.88	1.17	0.003	0.402	1.61	2.60	0.021	0.017	0.043	0.133
		median	115	0.070	8.57	3.62	0.017	1.69	2.12	10.8	0.100	0.033	0.024	0.047
		min	63.7	0.033	6.13	1.75	0.013	1.54	1.27	8.44	0.054	0.028	0.002	0.016
		max	216	0.390	14.8	6.47	0.026	3.01	6.23	18.3	0.116	0.079	0.150	0.354
R83	Medium/Brown	Bangladesh	315	0.909	9.64	24.7	0.029	2.03	4.91	5.44	0.143	0.062	0.004	0.089
R84	Medium/white	Bangladesh	128	0.491	3.64	14.3	0.020	1.50	2.42	5.71	0.304	0.028	0.015	0.020

n gives the number of samples with concentrations ⩾LOQ (limit of quantitation).

[a] Only the results that are ⩾LOQ were included in the statistics.

samples from India: range 0.212–0.228 mg kg^{-1}; 2 from Thailand: 0.200 and 0.250 mg kg^{-1}; and 1 from Surinam: 0.290 mg kg^{-1}). This type of variation in As concentration in rice grains was also observed by many investigators. For instance, a 7-fold difference in As concentration was reported for 901 white rice samples collected from 10 countries in 4 continents. The median arsenic concentration for the Egyptian and Indian rice was the lowest with values of 0.04 and 0.07 mg kg^{-1}, respectively, while the highest content was reported for the USA and France rice samples with median values of 0.25 and 0.28 mg kg^{-1}, respectively (Meharg et al., 2009). High levels of As have also been reported for rice grains grown in some parts of the USA (e.g. reported means for Texas and Arkansas rice were 0.258 and 0.190 mg kg^{-1}) (Zavala and Duxbury, 2008). Some Chinese rice has also been shown to accumulate high levels of As, where a mean arsenic concentration of 0.092 mg kg^{-1} (range 0.005–0.309 mg kg^{-1}) was reported for 282 brown rice grains collected from Hainan Island, China (Fu et al., 2011). Similarly, a high As mean concentration of 0.223 mg kg^{-1} (range 0.109–0.376 mg kg^{-1}) was reported for 44 different rice samples from Brazil (Batista et al., 2011). Relatively high As concentrations have also been reported for 95 rice samples collected from Southwestern Bangladesh with a mean As concentration of 0.23 mg kg^{-1} and a coefficient of variation of 53 (Rahman et al., 2010). However, higher levels of arsenic have been reported for rice samples collected from Bangladeshi arsenic-endemic areas (mean As concentration was 0.358 mg kg^{-1}, range 0.046–1.110 mg kg^{-1}, $n = 46$) (Smith et al., 2006). This has also been supported by the results of the 2 rice samples collected from As-endemic areas in Bangladesh as shown at the end of Table 3 (As concentrations of 0.143 and 0.304 mg kg^{-1}). Rice grain samples from arsenic-endemic areas in West Bengal, India were also reported to contain high concentrations of As with a mean value of 0.45 mg kg^{-1} ($n = 21$, range 0.19–0.78 mg kg^{-1}) for Boro rice and a mean concentration of 0.33 mg kg^{-1} ($n = 8$, range 0.06–0.60 mg kg^{-1} for Aman rice (Bhattacharya et al., 2010a,b).

Table 4 Mean, standard deviation (SD), median, and range concentrations (mg kg^{-1}) for some toxic elements in the rice samples sold in Almadinah Almunawarah, KSA arranged by country of origin.

Country of origin	*n* [a]	Mean ± SD	Median	Range
Arsenic				
USA	9/9	0.257 ± 0.142	0.164	0.127–0.464
Thailand	6/6	0.200 ± 0.025	0.192	0.181–0.250
Pakistan	9/9	0.147 ± 0.055	0.151	0.052–0.250
India	38/38	0.103 ± 0.048	0.093	0.026–0.228
Egypt	5/5	0.097 ± 0.044	0.094	0.039–0.154
Surinam	1/1	0.290		
Australia	1/1	0.188		
France	1/1	0.183		
Cadmium				
USA	8/9	0.011 ± 0.006	0.006	0.005–0.020
Thailand	6/6	0.012 ± 0.005	0.010	0.007–0.020
Pakistan	8/9	0.025 ± 0.014	0.028	0.007–0.042
India	37/38	0.018 ± 0.009	0.018	0.003–0.046
Egypt	3/5	0.004 ± 0.001	0.004	0.003–0.005
Surinam	1/1	0.008		
Australia	0/1	< LOQ		
France	1/1	0.013		
Lead				
USA	6/9	0.041 ± 0.030	0.034	0.014–0.098
Thailand	4/6	0.019 ± 0.007	0.019	0.012–0.028
Pakistan	4/9	0.020 ± 0.002	0.020	0.018–0.022
India	22/38	0.031 ± 0.044	0.018	0.003–0.218
Egypt	3/5	0.021 ± 0.008	0.017	0.015–0.031
Surinam	1/1	0.018		
Australia	0/1	< LOQ		
France	0/1	< LOQ		

[a] *n* is the number of samples with concentrations > LOQ out of the total number of samples from the specified country of origin.

Arsenic finds its way to rice grains from As-contaminated soil or As-laden irrigation water or both (Mondal and Polya, 2008; Zavala et al., 2008; Zhu et al., 2008; Khan et al., 2009; Taskeen et al., 2009; Bhattacharya et al., 2010a,b, Rahman et al., 2010). For instance, an investigation by Khan et al. reported an increased pattern of As concentrations in rice grains with an increase in As concentration in the irrigation water, soil, or both (Khan et al., 2009).

In addition to As, rice samples investigated in this work were found to contain variable levels of toxicologically relevant elements such as Ni, Cd, and Pb as shown in Tables 3 and 4. The source of these toxic metals can be anthropogenic, natural, or both (Qian et al., 2010; Lei et al., 2011).

Nickel is one of many metals that are known to cause cancer and many other health complications to humans (Denkhaus and Salnikow, 2002; Henderson et al., 2012). Luckily, almost all of the KSA samples (93%) contained no detectable Ni, while the rest of the samples ($n = 5$) contained low Ni concentrations with a mean of 0.064 mg kg^{-1}, SD 0.021 mg kg^{-1}, median 0.063 mg kg^{-1}, and range 0.042–0.086 mg kg^{-1}. In comparison, elevated levels of Ni were detected in the Brisbane samples ($n = 12$) with a mean of 1.83 mg kg^{-1}, SD 0.402 g kg^{-1}, median 1.69 mg kg^{-1}, and range 1.54–3.01 mg kg^{-1}.

Low levels of cadmium were also detected in all the rice samples but none has exceeded the Chinese MCL for Cd (i.e. 0.2 mg kg^{-1}). The mean Cd concentration in the KSA samples was 0.017 mg kg^{-1} ($n = 64$, SD 0.010 mg kg^{-1}, range 0.003–0.046 mg kg^{-1}), whereas the mean Cd level in the Brisbane samples was 0.039 mg kg^{-1} ($n = 12$, SD 0.043 mg kg^{-1}, range 0.002–0.150 mg kg^{-1}). The highest Cd concentration of 0.150 mg kg^{-1} was recorded for one of the Thailand samples. Similar to arsenic, cadmium has been classified by IARC as a human carcinogen and exposure to high levels of Cd severely irritates the digestive system, causes vomiting and diarrhea, and damages the lungs (Jarup et al., 1998; Nawrot et al., 2006; Åkesson, 2011; Lee et al., 2011).

More than half of the KSA samples contained detectable concentrations of lead (i.e. 40 out of 70 samples), but the concentrations are low in most samples. The mean Pb concentration in the samples was 0.029 mg kg^{-1} (SD 0.039 mg kg^{-1}, median 0.020 mg kg^{-1}, and range 0.003–0.218 mg kg^{-1}). Only one sample of Indian origin contained Pb concentration (0.218 mg kg^{-1}) in excess of Chinese MCL (i.e. 0.2 mg kg^{-1}). On the other hand, all the Brisbane samples contained detectable levels of Pb with a mean concentration of 0.120 mg kg^{-1} (SD 0.133 mg kg^{-1}, median 0.047 mg kg^{-1}, and range 0.016–0.354 mg kg^{-1}). Four of the samples contained elevated levels of Pb (0.236–0.354 mg kg^{-1}) in excess of the Chinese MCL.

Due to the widespread contamination of As in groundwater worldwide, most of the investigations that dealt with contamination of rice have concentrated on this metalloid with much less attention focused on other toxic elements that have the tendency to accumulate in rice grains. In one of these few articles that addressed this issue, a survey of the concentrations of some toxic elements such as Cd, Pb, Hg, and As in Chinese rice was carried out between 2005 and 2008, the mean concentration reported for Cd was 0.05 mg kg^{-1}; Pb 0.062 mg kg^{-1}; Hg 0.0058 mg kg^{-1}; and As 0.119 mg kg^{-1} (Qian et al., 2010). In another investigation, the reported levels of Cd, Pb, and As in different types of rice samples collected from Swedish retail market were 0.20 mg kg^{-1} for As, 0.024 mg kg^{-1} for Cd, and 0.004 mg kg^{-1}for Pb (Jorhem et al., 2008). Elevated levels of Co and Ni have been also reported for rice grains grown on treated soils in India (means for Co and Ni were 0.49, and 0.48 mg kg^{-1}, respectively) (Bhattacharyya et al., 2008). Rice-based baby food was also found to contain high levels of some toxic metals as shown by a Pakistani investigation, where reported concentration ranges (mg kg^{-1}) for Al, Cd, Ni, and Pb were 4.77–35.20, 0.0256–0.883, 0.124–0.332 and 0.053–0.091, respectively (Kazi et al., 2010).

3.3. Concentrations of essential elements in rice samples

Despite its ability to accumulate some toxic elements, rice is known to be a good source of some essential elements to the human body including Fe, Mg, Zn, Cu, Cr, Co, Mn, and Se. The essentiality vs. toxicity of such elements was discussed in the introduction section above.

The concentrations of Fe, Mg, Zn, Cu, Cr, Co, Mn, and Se plus other elements in the rice samples are listed in Table 3.

The mean Fe concentration in KSA samples was 7.07 mg kg^{-1} (n = 70, SD 8.11 mg kg^{-1}, median 5.53 mg kg^{-1}, and range 1.95–55.1 mg kg^{-1}). A lower mean Fe concentration was found for the Brisbane samples (mean 3.64 mg kg^{-1}, SD 1.17 mg kg^{-1}, range 1.75–6.47 mg kg^{-1}, n = 12). This difference may be due to variation in the source.

Magnesium concentrations are the highest among the rice samples followed by Fe, Zn, and Mn. All the samples contained Mg with a mean concentration of 157 mg kg^{-1} (SD 111 mg kg^{-1}, median 139 mg kg^{-1}, and range 51.8–777 mg kg^{-1}). Two of the Indian rice (brown) contained high Mg levels of 777 and 732 mg kg^{-1}. The mean Mg concentration in the Brisbane samples was 123 mg kg^{-1} (range 63.7–216 mg kg^{-1}).

Zinc concentrations in Brisbane samples (mean 11.4 mg kg^{-1}, SD 2.6 mg kg^{-1}, and range 8.44–18.3 mg kg^{-1}) were relatively higher that those found in KSA samples (mean 6.19 mg kg^{-1}, SD 3.21 mg kg^{-1}, and range 1.15–13.5 mg kg^{-1}). This variation is also a result of variation in the source.

The mean Mn concentration in the KSA samples was 4.28 mg kg^{-1} (n = 70, SD 2.17 mg kg^{-1}, range 0.96–10.9 mg kg^{-1}) whereas the mean Mn concentration for the Brisbane samples was 9.48 mg kg^{-1} (n = 12, SD 2.88 mg kg^{-1}, range 6.13–14.8 mg kg^{-1}).

The mean Se concentration in the KSA samples was 0.202 mg kg^{-1} (n = 70, SD 0.133 mg kg^{-1}, median 0.174 mg kg^{-1}, and range 0.007–0.574 mg kg^{-1}), whereas the mean Se concentration for the Brisbane samples was 0.041 mg kg^{-1} (n = 12, SD 0.017 mg kg^{-1}, range 0.028–0.079 mg kg^{-1}).

The mean Cu concentration in the KSA samples was 1.28 mg kg^{-1} (n = 70, SD 0.339 mg kg^{-1}, and range 0.508–2.41 mg kg^{-1}) whereas the mean concentration in the Brisbane samples was 2.72 mg kg^{-1} (n = 12, SD 1.61 mg kg^{-1}, and range 1.27–6.23 mg kg^{-1}).

Total Cr concentrations in most of the KSA rice samples (73%) were below the LOQ and the mean total Cr concentration in the rest of the samples (n = 19) was 0.057 mg kg^{-1} (SD 0.045 mg kg^{-1}, median 0.048 mg kg^{-1}, and range 0.010–0.184 mg kg^{-1}). Two of these samples; one of USA origin and the other of Indian origin contained relatively elevated levels of Cr (i.e. 0.144 and 0.184 mg kg^{-1}, respective) but lower than the Chinese MCL for Cr (i.e. 0.2 mg kg^{-1}). As for the samples that were collected from Brisbane markets (n = 12), all were found to contain Cr concentration >LOQ with a mean of 0.097 mg kg^{-1} (SD 0.096 mg kg^{-1}, median 0.070 mg kg^{-1}, and range 0.033–0.390 mg kg^{-1}). Elevated Cr levels in excess of the Chinese MCL for Cr were found in one of the Chinese samples (0.390 mg kg^{-1}).

Finally, Co was found in 80% of the KSA samples with a mean concentration of 0.012 mg kg^{-1}, SD 0.020 mg kg^{-1}, median 0.007 mg kg^{-1}, and range of 0.001–0.116 mg kg^{-1}. Similarly low Co concentrations were found in the Brisbane samples (Table 3).

Different rice grain cultivars have been shown to accumulate variable levels of nutritional elements as reported in the literature. For example the Fe and Zn contents of some Taiwanese rice cultivars ranged from 3.90 to 28.1 mg kg^{-1} for Fe and 26.2 to 29.0 mg kg^{-1} for Zn (Jeng et al., 2012). In another investigation, the mean concentrations of Mg, Mn, and Zn in Thai white jasmine rice were 40.5 ± 4.9, 7.3 ± 0.9, and 21.8 ± 4.7 mg kg^{-1}, respectively (Parengam et al., 2010). Concentrations of minerals similar to those reported in this investigation have been stated for 25 rice brands collected from Jamaican markets. The reported mean values (in mg kg$^{-1)}$ for both white and brown rice, respectively were 0.08 and 0.157 for Cr, 1.65 and 2.96 for Cu, 22.3 and 20.1 for Fe, 371 and 1205 for Mg, 10.5 and 26.5 for Mn, 0.108 and 0.131 for Se, and 15.6 and 20.2 for Zn (Antoine et al., 2012).

4. Conclusions

The results of this investigation indicate that some rice varieties sold in the local markets of Almadinah Almunawarah, KSA contain hazardous levels of one or more of the toxic element (e.g. arsenic, chromium, and lead). Additionally, variation in the country of origin of rice resulted in large variation in concentration of both the toxic and essential elements in rice grains. In order to reduce the exposure of the local populations to toxic elements through rice consumption, the concerned authorities are strongly recommended to periodically monitor the levels of toxicologically relevant elements (e.g. arsenic, cadmium, chromium, and lead) in imported rice.

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