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Review article

Arsenic accumulation in rice: Consequences of rice genotypes and management practices to reduce human health risk

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A B S T R A C T

Rice is an essential staple food and feeds over half of the world's population. Consumption of rice has increased from limited intake in Western countries some 50 years ago to major dietary intake now. Rice consumption represents a major route for inorganic arsenic (As) exposure in many countries, especially for people with a large proportion of rice in their daily diet as much as 60%. Rice plants are more efficient in assimilating As into its grains than other cereal crops and the accumulation may also adversely affect the quality of rice and their nutrition. Rice is generally grown as a lowland crop in flooded soils under reducing conditions. Under these conditions the bioavailability of As is greatly enhanced leading to excessive As bioaccumulation compared to that under oxidizing upland conditions. Inorganic As species are carcinogenic to humans and even at low levels in the diet pose a considerable risk to humans. There is a substantial genetic variation among the rice genotypes in grain-As accumulation as well as speciation. Identifying the extent of genetic variation in grain-As concentration and speciation of As compounds are crucial to determining the rice varieties which accumulate low inorganic As. Varietal selection, irrigation water management, use of fertilizer and soil amendments, cooking practices etc. play a vital role in reducing As exposure from rice grains. In the meantime assessing the bioavailability of As from rice is crucial to understanding human health exposure and reducing the risk.

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1. Introduction

Arsenic (As) is one of the most challenging environmental problems in drinking water as well as food crops especially in rice which affects millions of people worldwide viz. in Bengal Delta, China, Taiwan, parts of South America and South East Asia (Naidu et al., 2006; Nordstrom, 2002; Smedley and Kinniburgh, 2002). A significant number of populations rely upon groundwater for its drinking water supply as well as irrigation water supply for growing agricultural crops especially paddy rice and consequently exhibit extreme health effects of chronic As exposure (Bhattacharyya et al., 2003b). Arsenic in rice is a serious concern for >3 billion people across the world who consume rice as a staple food and millions of people may be at risk of developing As-related health problems (Meharg and Zhao, 2012). Fig. 1 shows the global rice production and per capita consumption in selected countries. Arsenic contents in rice varies widely, with most reported concentrations found in the range 0.02 to 0.90 mg kg^{-1} (Meharg and Zhao, 2012). Recent studies indicated that rice genotypes have wide variations in total grain-As concentrations and As speciation around the world (Al Rmalli et al., 2005; Huq et al., 2006; Meharg et al., 2009; Rahman et al., 2011; Rahman et al., 2009; Torres-Escribano et al., 2008; Williams et al., 2007b; Zavala and Duxbury, 2008; Zhu et al., 2008). Total As content in rice grains from Bangladesh varies from between 0.058 and 1.835 mg kg^{-1} (Meharg and Rahman, 2003). While this initial study shows that the As in rice grains can be as high as 1.835 mg kg^{-1}, recent studies show that As in rice grains was usually below 1.0 mg kg^{-1} (Rahman et al., 2009). Moreover, rice has been identified as an important source of inorganic As which may vary from 10% to 90% of total As (Williams et al., 2005; Williams et al., 2007c). Rice contains about ten times more As than other crops, and rice produced in regions such as Bangladesh, India, China and the US, often contains even higher levels of inorganic As (Meharg et al., 2008; Williams et al., 2005, 2007a).

Bioaccumulation of As by rice plants have been linked to a number of soil and environmental factors together with the nature of rice species. A number of field studies have shown substantial genetic variation among the rice genotype in grain-As concentration as well as As speciation. In rice, there are three interacting loci on chromosome 6 and chromosome10 which maintain the genetic variation to regulate As tolerance (Dasgupta et al., 2004). Other factors include soil-As loading, water management practices, nutrient management in soils etc. Accumulation of As from paddy soils and irrigation water poses a potential health risk to humans especially inorganic As species. They are classified as human carcinogens (IARC, 2004b) and several epidemiological studies confirmed the relationship between As exposure via drinking water and various health effects including skin and kidney disease, heart disease, diabetes mellitus, neurological, respiratory complications and gall bladder and lung cancers (Chen et al., 2015; Hopenhayn-Rich et al., 1996; Islam et al., 2015b; Sommella et al., 2013). Recent studies show that phosphate fertilizer is a major source of As in areas affected with chronic kidney disease in Sri Lanka (Jayasumana et al., 2014, 2015).

The association between a low level of As exposure and human health effects still needs to be confirmed. Given the many case examples of adverse impacts of As, there is a need to extend the existing studies that focus on an assessment of the presence of As in rice grains to the bioavailability of As from rice with a view to understanding their exposure and risk. A number of review articles were published on different aspects of As and rice (challenges of As in soil plant systems, geographical variation on As accumulation, management practices etc.). For this review we indexed articles in the Web of Science between 1956 and 2016 related to the keywords, As and rice, As and their phytotoxicity in rice, As and rice genotype, As management in paddy fields, water management and rice grain As, fertilizer management grain As in rice, As and human health risk from rice, bioavailability of As from rice, and cancer risk from ingestion of rice and retrieved them to obtain the maximum number of relevant articles (Fig. 2). There was no language restriction and we manually reviewed the references from the original research articles. We aimed to identify all related studies in our topics. The exclusion criteria we followed were: publications containing no original research (reviews, editorials, proceedings, non-research letters etc.), research related to performance of instruments, research related to As chemistry other than rice, research lacks of data on As exposure from rice and any duplications of the searched research articles in different categories. The main focus of this manuscript is that it deals with As and rice genotypes, growth and yield vs As contamination in irrigation water and soils, and how rice genotypes control As uptake and their speciation. It also discusses the major management practices that minimize As uptake by rice plants. Additionally, we also discuss about rice consumption and risk of As exposure to human health in the context of bioavailability and incremental lifetime cancer risk.

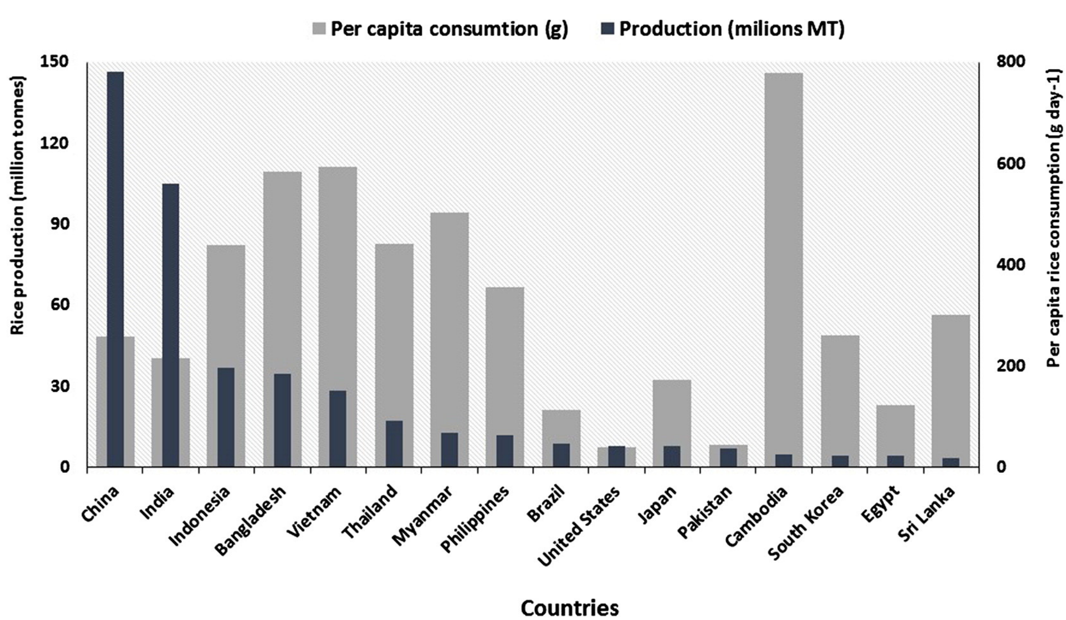

Fig. 1. The top rice producing and consuming countries of the world.
(data from USDA world rice production and consumption: https://www.worldriceproduction.com)

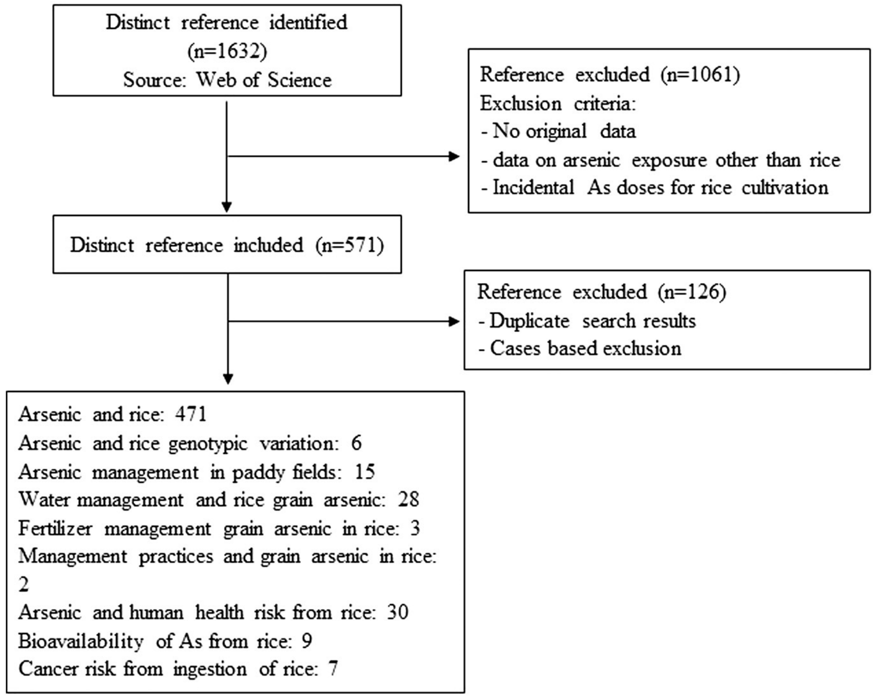

Fig. 2. Selection process used for reviewing As accumulation in rice: consequences of rice genotypes and management practices to reduce human health risk, 1965–2016.

2. Arsenic and rice plants

2.1. Sources and translocation of As in rice plants

Arsenic contamination occurs naturally in alluvial and deltaic sediments, as well as volcanic rocks and thermal springs and the weathering of such deposits can lead to mobilization of As (Nordstrom, 2002; Smedley and Kinniburgh, 2002; Welch et al., 2000). There are some anthropogenic sources of As that are released in the environment through mining, pesticide application, wood preservation, combustion of coal etc. Elevated concentrations of As are often derived from natural sources. Bangladesh is an example of As occurring naturally in alluvial sediments that is being mobilized into the groundwater. The natural sources of As in paddy fields are biogeochemical processes. The anthropogenic sources are from the use of As-contaminated irrigation water, mining activity, use of arsenical pesticides and fertilizers (Bhattacharyya et al., 2003a; Smedley and Kinniburgh, 2002). In pore water the concentration of As and their speciation are governed by soil redox potential, soil pH, soil organic matter status, nutrient concentration especially silicon (Si) and phosphorus (P), iron oxide and clay mineral contents (Bogdan and Schenk, 2009; Dixit and Hering, 2003; Smedley and Kinniburgh, 2002; Williams et al., 2011).

Rice is grown as a lowland crop in flooded paddy soils under reducing conditions where As availability is higher than under oxidizing conditions. In the flooded soil condition the mobility of arsenite [As(III)] is higher than arsenate [As(V)] due to the reductive dissolution of iron oxide or hydroxides (Takahashi et al., 2004). This reductive mobilization of As under the anaerobic conditions greatly enhances the bioavailability of As to rice, leading to excessive As bioaccumulation in rice grains and other plant parts (root and straw). Also widespread use of As-contaminated groundwater for the irrigation of paddy rice causes an additional health hazard to people who rely on this staple (Meharg et al., 2009; Mondal and Polya, 2008; Rahman et al., 2009; Williams et al., 2006). Paddy fields which are irrigated with As-contaminated groundwater act as net sinks of As from groundwater and a small amount returns to or replenishes the aquifer (Neumann et al., 2011). These paddy soils and As-contaminated irrigation water are both linked to elevated concentrations of As in the rice grains (Heikens et al., 2007; Panaullah et al., 2009; Xie and Huang, 1998). Furthermore, the deposition of As in irrigated soils poses a serious threat to sustainable agriculture in impacted areas (Heikens, 2006). However, grain-As concentrations are impacted by the combined influences of soil characteristics, environmental conditions, and crop management (Cheng et al., 2006; Khan et al., 2009; Liu et al., 2006; Panaullah et al., 2009; Xie and Huang, 1998; Xu et al., 2008) as well as rice varieties (Ma et al., 2008; Norton et al., 2009b; Zhang et al., 2008).

There is limited information on the uptake and translocation of As in rice plants (Fig. 3). The uptake and translocation of As in rice plants is greatly influenced by As species present in the rhizosphere (Arao et al., 2011). The presence of Si and P in soil as well as in pore water also impacted the As uptake by rice plants (Bogdan and Schenk, 2008). Two main pathways identified to uptake As in rice plants include phosphate transport pathways since As(V) is an analogue of phosphate and new evidence reveals that As(III) (silicic acid analogue) and undissociated methylated As species dimethylarsinic acid (DMA) and monomethylarsonic acid (MMA) enter into the root by aquaporin channels which are used as silicate transport (Li et al., 2009a; Ma et al., 2008; Wu et al., 2011b; Zhao et al., 2009). There is a significant variation in the uptake efficiency of different As species. The uptake efficiency of methylated As species is comparatively lower than inorganic As species but the translocation efficiency is higher for methylated As species (Raab et al., 2007). Research shows that transfer of As in rice grains is greater than other cereals (Williams et al., 2007c). This is due to the high soil/shoot ratio and this difference in the transfer ratio is probably due to differences in As speciation and dynamics in aerobic and anaerobic soils (Williams et al., 2007c). In rice, the export of As from the shoot to the grain appears to be under tight physiological control as the grain/shoot ratio decreases by more than an order of magnitude (from 0.3 to 0.003 mg kg^{-1}) and as As levels in the shoots increase from 1 to 20 mg kg^{-1}. Norton et al. (2010b) examined the relationships between P and As, and Si and As in a wide range of cultivars grown in As contaminated field trials in Bangladesh and China. They observed no correlation between shoot and grain speciation, with the inorganic form comprising 93 to 97% of As in the shoot and 63 to 84% in the grains (Norton et al., 2010b).

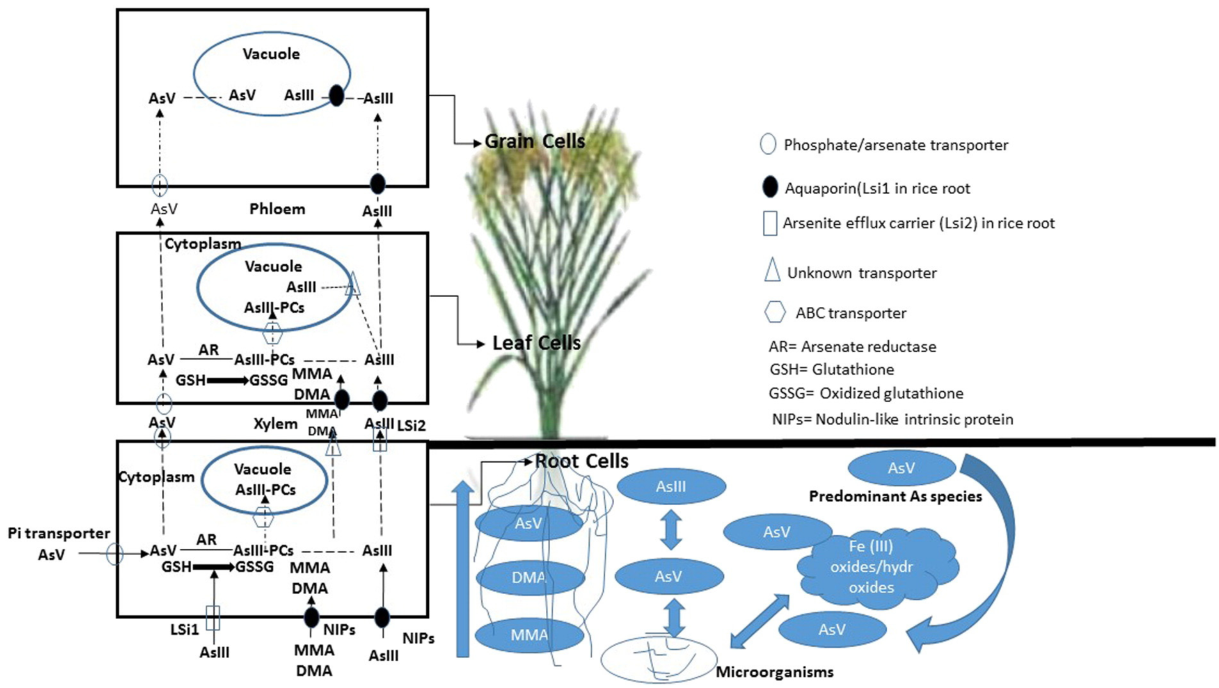

Fig. 3. Possible mechanisms of As uptake in rice plants.
(adopted and modified from Ali et al., 2009)

2.2. Arsenic phytotoxicity to rice plants

Arsenic is highly phytotoxic with the rice plants showing intolerance to elevated levels of soil-As resulting in a decrease in plant growth and crop yield (Barrachina et al., 1995; Carbonell-Barrachina et al., 1998). Various reports investigated the effect of As on growth of rice and most of them indicated that As has a significant negative effect on rice growth and yield. There are reports of rice grown either in soil or in solution culture with As or use of As-contaminated irrigation water which significantly reduces plant height, effective tiller number, shoot biomass, grain and straw yield reduction (Abedin et al., 2002a, 2002b; Frans et al., 1988; Hossain et al., 2007; Islam et al., 2004a; Khan et al., 2006; Liu, 1987; Marin et al., 1992; Milam et al., 1988; Tang and Miller, 1991; Tsutsumi, 1980; Wang and Forsyth, 2006). Rice yield was reduced by 10% at 12.9 mg As kg^{-1} soil application, and 50% at 52 mg As kg^{-1} application; no yield leading to death of plants occurred at 109–157 mg As kg^{-1} soil application (Yan-Chu, 1994) and 66% yield loss when mean soil solution As concentration was raised to 1.5 mg L^{-1} (Onken and Hossner, 1995). These findings demonstrate the critical role that soil solution As can play on rice plant growth and hence yield. Thus soil management strategies that change moisture levels such as via flooding or irrigation can impact rice growth. This is further evident from the studies conducted by Milam et al. (1988) who found a significant yield difference when rice grown was under field conditions followed by two irrigation options viz. continuous flooded and drained during mid-season. For instance, application of As @ 0, 2, 4 or 6 lb acre^{-1} results in an average grain yield of 1.32 ton acre^{-1} with continuous flooding and 1.78 ton acre^{-1} with mid-season drainage (Milam et al., 1988). However, As has also been found to enhance plant growth at concentrations below the toxicity threshold value. For example, application of As with the irrigation water up to 0.25 mg L^{-1} enhanced the plant height, panicle length, filled grains panicle^{-1}, thousand grain weight and finally the grain yield of Boro rice but further doses decreased the plant growth, yield and yield components (Islam et al., 2004a). Above 60% and 40% grain yield reduction for rice varieties Iratom 24 and BRRI dhan28, respectively, were found with 20 mg L^{-1} of As compared to control and the reduction in straw yield was also significantly higher for both rice varieties (Hossain, 2005). Maturity of rice grains was greatly delayed in the high As (57 mg kg^{-1}) plots with about 40–50% remaining green at the milk stage; as a result grain yield was very poor, only 2.0–2.5 t ha^{-1} (rough rice), which was almost half the standard yield, which is about 4.0–5.0 t ha^{-1} (Panaullah et al., 2006). A field study in China, observed that grain-As concentrations up to 0.72 mg kg^{-1} when grown in soils containing 68 mg As kg^{-1} (Xie and Huang, 1998). The concentrations of As in stems plus leaves were more closely related to soil total and available As than those of roots or grain (Kang et al., 1996). Another study from China indicated that As concentration in rice increased significantly with the addition of As in soil under field conditions with an isolation chamber (Wang et al., 2006).

Rice showed a physiological disorder called the straighthead disease, showing delayed heading date, shortened plant height and dramatically reduced grain yield which has been reported to be associated with As (Gilmour and Wells, 1980). The severity of straighthead increased significantly with the increase of soil As concentration. Straighthead caused approximately 17–100% sterile spikelet formation and about 16–100% reduction of grain yield (Rahman et al., 2008a). Literature suggests that As affects the photosynthetic pigments, chlorophyll-*a* and chlorophyll-*b* which is directly associated with rice yield and growth. Both chlorophyll-*a* and chlorophyll-*b* contents in rice leaf decreased significantly with the increase in soil As concentration of up to 30 mg kg^{-1}. No rice plant survived up to maturity stage in soil treated with 60 and 90 mg As kg^{-1} (Rahman et al., 2007a). The varieties Cocodrie, Mars, Kaybonnet, and Bengal were highly susceptible to straight head with ratings from 7.2 to 8.0 and grain yield reductions from 80 to 96%. Wells, Lagrue, Drew, Cypress, and Japan 92.09.31 were susceptible with ratings from 5.9 to 6.7 and yield reductions from 49 to 73% (Yan et al., 2005). Arsenic spiked soils having a concentration of 50 mg As kg^{-1} showed straighthead symptoms of rice causing 17–100% sterile spikelet's in BRRI dhan29 (Rahman et al., 2008a). Higher As also causes grain sterility in rice as reported in a field and glasshouse study in Bangladesh (Islam et al., 2004a). Grain yield and the occurrence of straighthead disease were cultivar-dependent and influenced by soil As level and water management practices (Hua et al., 2013).

Straighthead resistant cultivars yielded more and had lower grain-As than the susceptible ones. Elevated soil As with continuous flood management significantly reduced the grain yield of susceptible cultivars by > 89% due to substantially increased straighthead, which was induced by increased As content in grains. This study demonstrates that the selection of less As-susceptible cultivars and intermittent flood water practice could be an effective means to lower the As accumulation in grains and minimize the occurrence of the As-induced straighthead symptom and yield reduction.

2.3. Bio-accumulation of As in rice plants

The uptake or bioavailability and phytotoxicity of As is dependent on a number of factors including the source and the concentration of the element (NAS, 1977), nature of As species (Carbonell-Barrachina et al., 1998; Marin et al., 1992), soil properties such as clay content, pH and redox equilibrium conditions (Johnson and Hiltbold, 1969; Marin et al., 1993; Naidu et al., 2006) and type and amount of organic matter present (Mitchell and Barre, 1995). Adsorption–desorption and microbial processes are also predominant factors controlling the bioavailability of As (Naidu et al., 2006). The As content of rice plant parts generally follow the pattern: root > straw > husk > grain (Lei et al., 2012; Rahman et al., 2007b; Smith et al., 2008; Wang et al., 2006; Xie and Huang, 1998). Moreover, the concentration of As in all plant parts increased with an increase in soil As (Marin et al., 1992; Odanaka et al., 1987).

From a solution culture experiment it was observed that most of the As(V), As(III) and MMA accumulated in the root while DMA was readily translocated to the shoot (Marin et al., 1992; Odanaka et al., 1987). This was further demonstrated by Marin et al. (1993) who found rapid translocation of DMA up the plant with higher concentrations of As in rice shoots than in roots when rice plants were grown in DMA solution at 0–1.6 mg As L^{-1}. Irrespective of As chemical forms, root As concentration was 10.5 mg kg^{-1} in the 0.05 mg As L^{-1} treatment, which increased to 212.7 mg kg^{-1} in the 0.8 mg As L^{-1} treatment (Marin et al., 1992). Increasing concentration of As in irrigation water significantly increased As concentrations in root, straw, and rice husk (Abedin et al., 2002a; Carbonell-Barrachina et al., 1998; Hossain et al., 2007). A similar relationship between soil solution As and plant response was found in rice (Onken and Hossner, 1995). Application of As-contaminated irrigation water to the 1st crop Boro rice (dry season rice) had significant residual effects on the 2nd rice crop T. Aman (rain fed rice) and also increased the levels of As accumulation (Islam et al., 2004a). Arsenic present in irrigation water was found to enhance the bioaccumulation of As in rice plants (Bhattacharya et al., 2010) and higher accumulation of As was noticed in the root as compared to the straw, husk, and grain. The As concentration of rice produced in As-contaminated areas is 2 to 3 times higher than that produced in non-contaminated districts of Bangladesh (Hironaka and Ahmad, 2003) with total As concentrations ranging from 0.11 to 0.34 mg kg^{-1} in contaminated soils.

The range in concentrations of inorganic As in all unpolished rice was from 0.26 to 0.52 mg kg^{-1} dry weight. In the case of aromatic rice, it had a low level of As compared to other types of rice from different regions of Bangladesh (Ahmed et al., 2011). Rice samples from the districts of Pabna, Chapai Nawabganj, Rajbari, Faridpur and Gopalganj of the Gangetic floodplains of Bangladesh showed that 16% of the grain samples had no detectable As while on the other hand 14% grains had As level > 1 mg kg^{-1} (Islam et al., 2004b). Comparing varietal effects, the grain-As concentrations in IR 8 and BRRI dhan29 rice were higher in comparison with BRRI dhan28 and Parija and grain-As concentrations were always lower than in straw As (Islam et al., 2004b). Arsenic accumulation by rice plants growing with either As(III) or As(V) increased with increasing As treatment, irrespective of water regimes (Huq et al., 2006). However, the accumulation was greater in the As(III) treated soil than that in the As(V)-treated one.

In a study from West Bengal (India), high levels of As in the groundwater led to bioaccumulation of high levels of As in rice < 0.04–0.605 mg kg^{-1} (Roychowdhury et al., 2003). Another study has shown that the concentration of total As in cooked rice ranged from 0.33 mg kg^{-1} in the Jalangi block, Murshidabad to 0.38 mg kg^{-1} in the Domkal block, Murshidabad district west Bengal India (Roychowdhury et al., 2002). Paddy rice from North 24-Parganas India was detected at 0.50 mg kg^{-1} As (Signes-Pastor et al., 2009). Aman rice (wet season rice) in Bangladesh has been studied previously by different groups (Duxbury et al., 2003; Williams et al., 2006). The mean concentration of As in Aman rice was 0.125 mg kg^{-1} with a range of 0.072–0.170 mg kg^{-1} and in Boro rice (Irrigated dry season rice) it was 0.183 mg kg^{-1} (Duxbury et al., 2003), and the range of As levels in another study was 0.180–0.310 mg kg^{-1} (Williams et al., 2006). Arsenic levels in the rice grains of Bangladesh were typical of other regions of the world when rice was grown in an area where ground water and paddy soils contained low levels of As (Meharg and Rahman, 2003). In general, lower As content was found for Aman rice compared to Boro rice (Duxbury et al., 2003). However, it has also been reported that Aman rice from As affected areas have higher As content than Aman and Boro rice from low As affected areas (Roberts et al., 2011; Williams et al., 2006). The statistics of rice total As contents produced in different countries is presented in Table 1.

Table 1
Total As concentration (mg kg^{-1}) in rice produced in different countries of the world. [modified from Meharg et al. (2009) and Rahman et al. (2009)]

Countries	Total As			References
	No. of sample	Range (mg kg^{-1})	Mean (mg kg^{-1})	
Bangladesh	214	0.002–0.557	0.143	Rahman et al. (2009)
	144	0.020–0.330	0.130	Meharg et al. (2009)
	4	<0.005–0.020	0.011	Al Rmalli et al. (2005)
	78 (Boro)	0.108–0.331	0.183	Duxbury et al. (2003)
	72 (Aman)	0.072–0.170	0.117	
	10	0.040–0.270	0.136	Das et al. (2004)
	15	0.030–0.300	0.130	Williams et al. (2005)
	13	0.058–1.835	0.496	Meharg and Rahman (2003)
	133 (Boro)	0.040–0.910	–	Williams et al. (2006)
	189 (Aman)	<0.040–0.920	–	
	6 (Aromatic)	0.038–0.073	0.056	Rahman et al. (2014)
	35	0.050–2.050	−0.080	Islam et al. (2004b)
India	133	0.180–0.310	0.070	Meharg et al. (2009)
	11	0.041–0.605	0.232	Roychowdhury et al. (2003)
	23	0.079–0.546	0.033	
	8	0.120–0.663	0.358	Chowdhury et al. (2001)
China	11	0.283–0.725	0.501	Xie and Huang (1998)
	124	0.020–0.460	0.140	Meharg et al. (2009)
	2	0.460–1.180	0.820	Sun et al. (2008)
	43	0.052–0.253	0.129	Ma et al. (2016)
	165	0–0.665	0.116	Huang et al. (2015)
	446	0.033–0.437	0.143	Li et al. (2015)
	21	0.065–0.274	0.114	Liang et al. (2010)
Taiwan	5	0.010–0.630	0.100	Lin et al. (2004)
	–	0.010–0.140	0.050	
	2	0.190–0.210	0.200	Schoof et al. (1998)
	5	0.063–0.170	0.130	
	280	0.100–0.630	–	Lin et al. (2004)
Thailand	53	0.060–0.500	0.150	Adomako et al. (2011)
	54	0.010–0.390	0.140	Meharg et al. (2009)
Vietnam	31	0.032–0.465	0.208	Phuong et al. (1999)
Turkey	50	–	0.202	Sofuoglu et al. (2014)
Japan	26	0.070–0.420	0.190	Meharg et al. (2009)
France	33	0.090–0.560	0.280	
Italy	38	0.070–0.330	0.150	
Spain	76	0.050–0.820	0.200	
Egypt	110	0.010–0.580	0.050	
USA	163	0.030–0.660	0.250	
	134	0.100–0.660	–	Williams et al. (2007b)
	33	0.026–1.000	0.210	Heitkemper et al. (2009)
Australia	21	0.188–0.438	0.270	Rahman et al. (2014)
Pakistan	9 (Basmati)	0.073–0.088	0.082	

The mean and median grain-As levels for the South Central US were 0.30 mg kg^{-1} and 0.27 mg kg^{-1}, respectively (Williams et al., 2007b). In that study, 22 rice grain samples were analyzed from Camargue (France) reporting that the mean and median grain-As levels were 0.32 and 0.34 mg kg^{-1}, respectively. The total As was measured in 901 polished rice samples originated from 10 different countries (Meharg et al., 2009). The data showed that the lowest As content was in Egyptian rice (0.04 mg kg^{-1}) and the highest As was in US rice (0.25 mg kg^{-1}). Although, Bangladeshi rice contained 0.13 mg kg^{-1} of total As, 61% of this was present as inorganic As (Meharg et al., 2009). Rice grown in pots with high-As soil produced on average 17-fold higher grain-As than the same cultivars grown in a paddy field with low-As soil (Kuramata et al., 2011). Another result showed that when the soil and rice cultivar were the same, pot-grown rice grains contained larger percentages of DMA than field grown rice (Khan et al., 2010). This differences may be due to the redox potential differences between pot and field soil.

2.4. Arsenic bio-accumulation and rice genotypes

There is a considerably wide variation among the rice genotypes to As sensitivity. Three interacting loci on chromosomes 6 (2 loci) and 10 (1 locus) have been identified to regulate As tolerance in rice (Dasgupta et al., 2004; Zhang et al., 2008). A number of field studies have shown substantial genetic variation in grain-As concentration as well as As speciation. Germination test and seedling growth by exposing eight Bangladeshi rice varieties to As(III) and As(V) showed that germination was slightly inhibited at 0.5 and 1 mg L^{-1}. At 2 mg L^{-1}, inhibition was >10% (Heikens, 2006). Root growth was inhibited by ~20% at 0.5 mg L^{-1} and As(V) was more toxic than As(III) (Meharg and Rahman, 2003). Another investigation showed the significant genotypic difference in response to As(V) toxicity in rice on root elongation, As(V) uptake kinetics, physiological and biochemical response and As speciation (Geng et al., 2005). Uptake kinetics data showed that P-deprived genotype 94D-54 had a little higher As uptake than P-deprived 94D-64, but the difference was not large enough to cause acute toxicity in P-deprived 94D-54. A significant genotypic variation was detected in the As concentrations in rice grains. There are also significant genotype-environment (location) interactions of the concentrations of As in grains, suggesting the importance of cultivar choice in producing rice with low As in grains for a given location (Cheng et al., 2006). Lei et al. (2012) evaluated the effect of As-contaminated soil on uptake and distribution of As in 34 genotypes of rice (including unpolished rice, husk, shoot, and root). The mean As concentrations in rice plant tissues were different among the 34 rice genotypes. The highest As concentrations were accumulated in rice root (196.27–385.98 mg kg^{-1} dry weight), while the lowest was in unpolished rice (0.31–0.52 mg kg^{-1} dry weight). The statistics of rice varietal differences on total As contents is presented in Table 2.

The rice varieties did not show significant differences in As accumulation in straw, husk, brown and polish grain when the concentrations of As in soil was low. However, at higher concentrations, different rice varieties showed significant differences. Arsenic translocation from root to shoot (straw) and husk was higher in hybrid varieties compared to those of non-hybrid varieties (Rahman et al., 2007c). The uptake and translocation of As from roots to grains involves a number of steps and checkpoints with these giving rise to potential differences among genotypes. Rice genotypes also show significant differences in iron plaque formation of a reddish brown coating on the root surface (Dwivedi et al., 2010; Geng et al., 2005; Liu et al., 2011b) as a result rhizosphere characteristics which may also play an important role. Significant negative correlations have been showed between As concentrations in straw or grain with root porosity or radial oxygen loss (ROL) from the roots among 20–25 rice cultivars (Mei et al., 2009; Wu et al., 2011a). The cultivars able to release more oxygen may maintain the rhizosphere at a higher redox potential and form more iron plaque on the root surface, which may reduce As uptake (Wu et al., 2011a). Mei et al. (2012) recently focused on genotypes with higher ROL which have a strong ability to reduce As accumulation in shoots and increase As tolerance by reducing As mobilization in the rhizosphere and by limiting As translocation.

There are significant genotypic differences in As concentrations of all organs, and polished grains are significantly affected by genotype and soil type (Ye et al., 2012). This result indicated that As concentration in grain was partially governed by As uptake and the transfer from root to grain. Some genotypes, such as *Japonica* rice had consistently low grain-As concentrations. An experiment with 216 rice genotypes showed that the averages of As contents for *Indica* rice were higher than those of *Japonica* rice. The ranges of As contents in *Indica* rice and *Japonica* rice were 0.021–0.296 mg kg^{-1} and 0.005–0.274 mg kg^{-1}, respectively (Jiang et al., 2012). When rice is grown in low As affected areas the results indicated that Boro (0.072 mg kg^{-1}) and Aman (0.086 mg kg^{-1}) rice contained 2 to 4-fold lower levels of As compared to high As affected areas of Bangladesh but inorganic As content (70%) was the same as the highly affected areas (Al-Rmalli et al., 2012). This suggests that irrespective of total As content it is the bioaccumulation

Table 2
Summarizing the varietal differences on grain-As accumulation.

Study type	Rice genotypes	Total grain As (mg kg^{-1})	References
Glasshouse study	BRRI dhan28	0.23 ± 0.05	Rahman et al. (2007a)
	BRRI dhan29	0.16 ± 0.08	
	BRRI dhan35	0.20 ± 0.03	
	BRRI dhan36	0.18 ± 0.05	
	BRRI hybrid dhan1	0.14 ± 0.04	
	BRRI dhan29	0.21–0.86	Khan et al. (2009)
	BRRI dhan33	0.23–0.75	
	BRRI hybrid dhan1	0.70 ± 0.01	Rahman et al. (2007b)
	BRRI dhan28	0.60 ± 0.01	
	BRRI dhan29	0.86 ± 0.03	Talukder et al. (2012)
	BRRI dhan32	0.37 ± 0.01	
	YY-1	0.66 ± 0.03	Liu et al. (2006)
	94D-64	0.32 ± 0.02	
	KY1360	0.32 ± 0.00	
	Gui630	0.43 ± 0.10	
	94D-54	0.41 ± 0.04	
	94D-22	0.36 ± 0.01	
	Hybrid	0.89–1.03	Ye et al. (2012)
	Japonica	0.24–0.63	
	Indica	0.43–0.66	
Field study	Local landraces with red bran	0.57 ± 0.08	Norton et al. (2009b)
	Local landraces with brown bran	0.42 ± 0.08	
	Red minikit	0.17–0.57	Bhattacharya et al. (2010)
	Megi	0.11–0.43	
	Aman:BR23, BRRI dhan33	0.10–0.22	Ahmed et al. (2011)
	Boro: BR3, BRRI dhan35	0.22–0.34	
	IR-68144-127	1.68 ± 0.38	Dwivedi et al. (2010)
	IR-68144-120	0.97 ± 0.14	
	CN1643-3	0.68 ± 0.02	
	CN1646-2	0.78 ± 0.02	
	IR-36	0.41 ± 0.19	
	IR-64	0.41 ± 0.02	
	Gotrabhog (IET-19226)	0.52 ± 0.06	
	Zhe 733	0.45 ± 0.07	Hua et al. (2011)
	Rondo	1.25 ± 0.12	
	Cocodrie	1.45 ± 0.98	
	9 non-glutinous	0.10–0.66	Kuramata et al. (2011)
	1 glutinous	0.14	
	Hybrid middle rice	0.31–0.47	Lei et al. (2012)
	Hybrid late rice	0.35–0.51	
	Normal late rice	0.39–0.50	
	Indica rice	0.02–0.30	Jiang et al. (2012)
	Japonica rice	0.01–0.27	

potential of plants that control grain As content. These investigators also reported that the As level in aromatic rice (0.049 mg kg^{-1}) from the Sylhet region was over 40% lower than that of non-aromatic rice (0.081 mg kg^{-1}); aromatic rice also contained higher levels of essential elements and consumption of aromatic rice may increase Se and Zn intake by 46% and 23% respectively (Al-Rmalli et al., 2012).

Arsenic concentration varied between rice subpopulations and ranges between 3 and 34 fold (Norton et al., 2012) and inorganic As correlated strongly with total As among a subset of 40 cultivars harvested in Bangladesh and China. Genetic variations have shown a large determining factor for grain-As concentration. The rice genotype Bala is tolerant to As contamination while the genotypes Cocodrie, Mars, Kaybonnet, Bengal, and Azucena are very much sensitive to As contamination (Yan et al., 2005). Other field trials at Faridpur and Sonargaon, Bangladesh, showed 4–4.6 fold variations in total grain-As among 76 cultivars including local landraces, locally improved cultivars and parents of permanent mapping populations (Norton et al., 2009b).

Environmental factors made the largest contribution to the variation in grain-As (61%), followed by genotype (6%) and genotype × environment interaction (19%) (Norton et al., 2009a). These results indicate a genotype × environment interaction across diverse environments, which is not surprising considering that As bioavailability in soil is greatly influenced by soil properties, the source of As contamination, water management and other environmental factors. Results showed that there was a significant genotype effect on the percentages of inorganic As and DMA in grain, but the influence by the environmental factor was greater (Norton et al., 2009a, 2009b). Another report showed that variety and environmental interaction had significant effects on grain-As concentrations in both wet and dry seasons and the relative variability due to the environment was greater than that due to varietal differences in both seasons (Ahmed et al., 2007). Environment and genotype × environment interaction accounted for 70–80% and 10–21% of the total variation, respectively while the variety explained only 9% of the total variation in As. A large environmental effect has been reported from a trial of 38 Bangladeshi cultivars grown at ten experimental sites across different agro-ecological zones of Bangladesh (Ahmed et al., 2011). Environment factors accounted for 69–80% of the variation in grain-As concentration, whereas genotype and genotype × environment interactions accounted for only 9–10% and 10–21% of the variability. Another report showed that total grain-As and As species [As(III) and DMA] varied widely among 25 diverse rice cultivars. It indicates that As concentration and speciation are mostly dependent on genotype, which accounted for about 70% of the variation in total grain-As (Pillai et al., 2010). All the studies identified a number of rice cultivars that accumulated relatively low levels of As across sites and it may be concluded that genetic stability is greater across seasons than across diverse sites (Ahmed et al., 2011; Norton et al., 2009a, 2009b; Pillai et al., 2010).

2.5. Arsenic speciation and rice genotypes

Similar to its adverse impact on human health that varies with nature of speciation, toxicity of As to rice depends to a large extent on its speciations with significant difference between organic and inorganic As species. Therefore, the speciation and localization of As species and their distribution in rice grains are key factors controlling the bioavailability of As. As discussed above, the distribution of As varies between the various parts of the grain (husk, bran and endosperm) and are characterized by element specific distribution patterns. The major As species in roots, stems and leaves are As(III) and As(V), while As(III) and DMA (comprising 85 to 94% of the total As) are the major As species identified in the grain (Smith et al., 2008). Speciation of As in husk, bran and the endosperm shows As(III)-thiol complexes as the predominant form (Lombi et al., 2009). Depending on nature of As speciation, rice can be classified into two groups — those rich in inorganic As-type and those that are rich in DMA-type, depending on the speciation of As in the grain. The dominant form of As speciation in rice grains appears to depend on the total rice As content. Zavala et al. (2008) found that when rice contains low levels of As, the dominant species is As(III) but when rice contains high levels of As, the dominant form is DMA (Zavala et al., 2008). In the same report researchers also indicate much difference in the speciation of As in rice from the US and Europe and Asia. These researchers methylated As (a less toxic form) in the rice from the US whereas rice grown in Europe and Asia contains the predominant form which is toxic inorganic As. Inorganic As has been detected in European, Bangladeshi, and Indian rice at 64 ± 1% ($n = 7$), 80 ± 3% ($n = 11$), and 81 ± 4% ($n = 15$), respectively. A study from Australia showed that Asian rice contained relatively lower total As than Australian and Italian rice on sale in Australia. In Asian rice inorganic As is predominant (86–99%), whereas in Australian rice the average inorganic As and DMA in Australian-grown rice were found to be about 58–63% and 18–26% of the total As, respectively (Rahman et al., 2014). In another study an Australian rice variety Quest represents DMA that was 85–94% of the total As (Smith et al., 2008). The statistics of As species concentration in rice produced in different countries is presented in Table 3.

The proportion of organic and inorganic As in rice is also cultivar dependent (Williams et al., 2005). A study reported the higher amount of inorganic As in Boro rice (mean 82%) than in Aman rice (mean 66%) in Bangladesh (Williams et al., 2006). Brown rice has a higher proportion of inorganic As than white rice (Meharg et al., 2008) and in white rice As was generally dispersed throughout the grain, while in brown rice As was found to be preferentially localized at the surface. Another study showed that inorganic As is the major species in all rice cultivars (Kuramata et al., 2011) with few genotypic differences in the levels of total As and inorganic As in the grain. In soils with high As levels, the total grain-As increased, markedly with increased levels of DMA. Speciation of As in rice is also affected by root aeration and variation of genotypes (Wu et al., 2011a).

There is a significant genotype effect in the percentage of inorganic As and percentage of cacodylic acid respectively, in rice grains. An analysis of polished rice from various production regions of China showed that the inorganic As species was predominant, accounting for approximately 72% of the total As in rice, with a mean concentration of 82.0 ng g^{-1} (Liang et al., 2010). The concentrations of total grain-As and As species [As(III) and DMA] have been shown to vary widely among 25 diverse rice cultivars (Pillai et al., 2010). Arsenic concentration and speciation are mostly dependent on genotype, which accounts for about 70% of the variation in total grain-As. However, the proportion of organic species (DMA and MMA) in rice grains may vary depending on the source of grain and method of extraction. Nowadays not only inorganic As but also organic forms should receive more attention as the toxicity of the trivalent As metabolites MMA and DMA were found to be highly toxic to human cell lines (Petrick et al., 2000; Styblo et al., 2000).

3. Management practices to control As bio-accumulation

3.1. Fertilizers and other soil amendments

For successful crop production and reducing As toxicity, fertilizer amendments play an important role. Rice contributes >80% of dietary energy and is a source of many minerals, including selenium (Se), manganese, iron and zinc (Zn). There are many reports on the effect of silicon (Si) and Se on As accumulation. Silicon was reported to play a significant role in reducing As toxicity, because the two elements act as metabolic antipodes. A recent study suggests that a high level of As in soils decreases Se and Zn levels in rice (Norton et al., 2010a). Many forms of Si fertilizers are available, some of which are industrial by-products, such as basic slag from the steel manufacture processes (Takahashi, 2002). Silicon fertilizers have been widely used in different countries in the world especially in Japan to increase rice yield. Silicon fertilization can effectively decrease As accumulation in rice in the areas affected by As under field conditions. Addition of 20 g kg^{-1} of silica gel (SiO_2) to soil decreased As concentrations in straw and grain by

Table 3
Arsenic species concentration (mg kg^{-1}) in rice produced in different countries.

Countries	Types of rice	As species				Total As	References
		As(III)	As(V)	DMA	MMA		
Bangladesh	White	0.083 (0.010–0.210)	–	0.019 (0–0.050)	–	0.131 (0.030–0.300)	Williams et al. (2005)
	Brown	0.280	–	0.170	0.010	0.610	Meharg et al. (2008)
India	White	0.027 (0.020–0.040)	–	0.066	0.0007	0.046 (0.030–0.050)	Williams et al. (2005)
	Brown	0.040	–	<LOD	<LOD	0.070	
	Red	0.050	–	0.010	<LOD	0.080	
China	White	0.114 (0.051–0.302)	0.040	0.040 (0.009–0.147)	0.013 (0.007–0.013)	0.230 (0.019–0.586)	Zhu et al. (2008)
	Brown	0.210	–	0.090	0.010	0.360	Meharg et al. (2008)
Taiwan	White	0.247 (0.110–0.510)	–	0.037 (0.030–0.050)	0.032 (0.015–0.060)	0.383 (0.190–0.760)	Williams et al. (2005)
Thailand	Light yellow	0.080	–	0.060	<LOD	0.110	Williams et al. (2005)
Japan	White	0.071	0.013	0.011	0	0.095	Narukawa et al. (2008)
Spain	–	0.080	–	0.050	<LOD	0.170	Williams et al. (2005)
USA	White	0.076 (0.020–0.100)	0.042 (0.032–0.051)	0.077 (0.050–0.260)	<LOD	0.277 (0.170–0.400)	Williams et al. (2005)
	White	0.092 (0.079–0.101)	–	0.0137 (0.136–0.141)	–	0.329 (0.308–0.350)	Zhu et al. (2008)
	White	0.110	–	0.155 (0.040–0.302)	<LOD	0.280	Meharg et al. (2008)
	Brown	0.105 (0.060–0.140)	–	0.090 (0.010–0.150)	<LOD	0.225 (0.110–0.340)	Williams et al. (2005)
	Brown	0.170	–	–	0.010	0.440	Meharg et al. (2008)
Brazil	White	0.078 (0.040–0.156)	0.034 (0.016–0.062)	0.093 (0.039–0.258)	0.008 (0–0.029)	0.223 (0.109–0.376)	Batista et al. (2011)
	Boiling white	0.087 (0.045–0.127)	0.043 (0.024–0.060)	0.065 (0.017–0.139)	0.010 (0–0.051)	0.215 (0.108–0.367)	
	Brown	0.146 (0.139–0.151)	0.042 (0.037–0.051)	0.127 (0.070–0.206)	0.011 (0–0.018)	0.348 (0.271–0.428)	
Canada	Wild	0.045 (0.010–0.080)	–	0.010	<LOD	0.065 (0.020–0.110)	Williams et al. (2005)

78% and 16%, respectively (Li et al., 2009b). In the same experiment, Si addition decreased the concentration of inorganic As in grain by about 60% but increased the DMA concentration by 33% (Li et al., 2009b). This study also showed significant differences between two rice genotypes in grain-As speciation. A recent study investigating the effects of Si fertilization on As uptake and speciation in rice plants with different radial oxygen loss (ROL) showed that Si addition significantly reduced shoot and root total As and also decreased the inorganic As in shoots and had the tendency to increase DMA concentrations (Wu et al., 2015). Another investigation demonstrated the potential effects of Si fertilizers in reducing As in rice grains and found that the application of high NP + S-KSi 9000 (9000 kg ha^{-1}) significantly reduced the As concentration in rice grains by up to 20%, compared with the control (Wang et al., 2015a). Silica nutrition acts as a central player that restricts photosynthetic impairment in As-treated plants and in addition to limiting As uptake via modulation of the expression of genes with prime importance in As uptake and translocation (Sanglard et al., 2016).

Nitrate fertilization also has a positive role in reducing As uptake. A pot microcosm experiment showed that nitrate addition reduced As uptake by the rice plant. Nitrate may inhibit Fe(III) reduction and/or stimulate nitrate-dependent Fe(II) oxidation, leading to As co-precipitation with, or adsorption to, Fe(III) minerals in the soil (Chen et al., 2008). Considerable controversy exists in the literature regarding the effect of P on the availability and uptake of As by rice. For instance, P has been shown to increase, decrease or have no effect on the uptake of As. A number of pot and field experiments have shown that additions of phosphate fertilizer to soil decreased As uptake by rice (Abedin et al., 2002a; Hossain et al., 2009; Talukder et al., 2011; Wu et al., 2011b). However, in some cases phosphate was found to be effective in exchanging adsorbed As(V) or As(III) from the soil solid phase and from the iron plaque on the root surface, thus increasing the As availability to rice plants. Another report indicates that P influences As mobility and bioavailability which depends on the charge components of the soil (Bolan et al., 2013). Results showed that the addition of P resulted in an increase in As desorption, and the effect was more pronounced in the case of allophanic soil. In the case of both As-spiked soils and field contaminated sheep-dip soil, application of P increased the desorption of As, thereby increasing its bioavailability. Phosphorus concentration may promote As translocation from roots to shoots. High P concentrations decreased the percentages of As distribution in iron plaque from around 70% to 10%, while it increased the percentages of As in roots and shoots (Geng et al., 2005). Phosphate fertilization or silicon amendment of paddy soils could therefore be possibilities for reducing the uptake of As(V) and As(III) in the rice plant, respectively (Hu et al., 2005; Li et al., 2009b). However, a higher level of P application than plant requirements is not appropriate due to environmental concerns (Lee et al., 2015).

Iron-oxide minerals have a significant impact on As solubility, retention, release and As dynamics in flooded rice culture (Loeppert et al., 2005; Takahashi et al., 2004). Iron plaque diminishes the inhibition effect of phosphate on paddy rice's As(V) uptake (Chen et al., 2005). Several recent studies have shown that the formation of iron plaque on the rice root surface leads to the sequestration of As and reduction in As transfer to shoot and grain (Chen et al., 2005; Liu et al., 2004, 2005; Voegelin et al., 2007). The accumulation of As on plaque in close association with the root might increase the potential bioavailability of As (Voegelin et al., 2007). Iron oxides or hydroxides are an important component in the soil–paddy rice system with a strong influence on As biogeochemistry. Hossain et al. (2009)) found that when As was added to the soil, an addition of ferrous sulphate alleviated As toxicity in rice plants, possibly by enhancing the formation of iron plaque on the root surface (Hu et al., 2007). Sometimes sulphur itself plays an important role in As detoxification and mobility from root to shoot. The nutritional status of sulphur can influence As detoxification and mobility within a plant due to the role of thio-rich compounds (e.g. phytochelatins) in complexing and sequestering As(III) (Zhao et al., 2010). Plants deficient in sulphur were found to have a greater root-to-shoot translocation of As (Liu et al., 2010; Zhang et al., 2011). Another result shows that rice plants treated with sulphur fertilizers show an apparent increase of iron plaque formation and small decreases in the concentrations of As in rice tissues (Hu et al., 2007). Thus the proper supply of S nutrition may be helpful in the prevention of As accumulation in the aerial parts of a plant as well as As induced toxicity (Dixit et al., 2016).

Organic matter has many important roles in crop production. However, there are some controversies regarding the effect of organic matter on As uptake and mobilization. Mobilization of As from paddy fields is governed by organic matter (Williams et al., 2011). Generally microbes utilizing the organic matter consume oxygen that leads to a decrease in redox potential, which in turn leads to As dissolution from FeOOH (Rowland et al., 2009; Smedley and Kinniburgh, 2002). It has another two roles in As availability in soils, desorbing As species from soil surface exchange sites (Weng et al., 2009), and dissolved organic matter complexing As species (Liu et al., 2011a; Williams et al., 2011). Recently, it was reported that total grain-As is higher when rice was grown in soil with high As contamination and high organic matter, with the increase of organic As species. The results indicate that the application of organic matter in paddy soils with elevated soil As may lead to an increase in the accumulation of As within rice grains (Norton et al., 2013). Biochar is a pyrogenic carbon material produced by the combustion of biomass under oxygen limited conditions. It has an important role in soil remediation for example by reducing As mobility and bioavailability in soils because of its unique properties (high surface area and cation exchange capacity) (Ahmad et al., 2014; Zhou et al., 2013). It also improves soil fertility and helps carbon sequestration (Mohan et al., 2014; Wang et al., 2015b). There are several methods developed to modify the biochar to increase its sorption capacity to As. However, the methods used are relatively complex and costly. Thus, additional investigations are needed to develop simple and cost-effective methods to modify biochar.

3.2. Water management

Water management techniques may also prove to be highly effective in combating the problem of excessive accumulation of As in rice grains. Besides saving water it also means less input of As into the paddy field from irrigation of As-contaminated groundwater. Selecting appropriate water management practices and rice cultivars according to the level of As contamination in soils will benefit food security as well as high yields. In the dry season, irrigation is required for rice crops. In As-affected areas like Bangladesh As-contaminated groundwater is widely used to irrigate rice crops (Saha and Ali, 2007), which has resulted in elevated As concentrations in soils and rice grains (Hossain et al., 2008; Islam et al., 2007; Meharg and Rahman, 2003), and significant yield losses due to As phytotoxicity (Khan et al., 2009; Panaullah et al., 2009; Tripathi et al., 2013). A study was conducted on the impact of As contaminated irrigation water on soil-As content and rice productivity over two dry seasons in the area of the Faridpur district, Bangladesh with soil-As levels ranging from about 10 to 70 mg kg^{-1}. A simple mass balance calculation using the current water As level of 0.13 mg As L^{-1} suggested that 96% of the added As was retained in the soil and yield declined progressively from 7–9 to 2–3 t ha^{-1} (Panaullah et al., 2009).

It has been estimated that 900 to 1360 tons of As per year is brought onto the arable land of Bangladesh due to irrigation with As-contaminated groundwater (Ali, 2003). In paddy soils, the reductive mobilization of As under the anaerobic conditions greatly enhances the bioavailability of As to rice that leads to excessive accumulation of As in rice grains and straw. In comparison, soils maintained at aerobic conditions generally have very low levels of As in the soil solution. Under aerobic conditions the least As accumulation was found in rice straw and grain (Li et al., 2009b) but anaerobic conditions enhanced As uptake in rice plants (Talukder et al., 2012). Research showed that aerobic soil conditions resulted in a 10-fold lower amount of As uptake among a set of several hundred global cultivars as compared to a flooded field (Norton et al., 2012). Similar results from a greenhouse experiment, maintaining soil under aerobic conditions decreased As concentration in rice grains and straw by 10–20 fold, and 7–63 fold, respectively, compared with continuously flooded rice (Li et al., 2009b; Xu et al., 2008). Further investigation of the effect of imposing a period of aerobic conditions during either the vegetative or reproductive growth showed that it decreased grain-As by 80% and 50%, respectively (Li et al., 2009b). The pot study also showed that an aerobic treatment 3 weeks before and after heading was effective in reducing grain-As concentration (Arao et al., 2009). It was also observed that intermittent flooding reduced As uptake (23.33% in root, 13.84% in shoot and 19.84% in leaf) at panicle initiation stage, instead of continuous flooding (Rahaman et al., 2011).

Under aerobic soil conditions, As(V) is the dominant species, whereas under submerged soil conditions the predominant species is As(III). A comparative study using flooded versus intermittent flooding by Hua et al. (2011) showed that the combination of water management practices that allow periodic aeration of the soil and use of cultivars that are low accumulators of As can reduce As in the grain. A recent field study showed that in flooded treatment, As(III) in the pore water was the predominant As species, accounting for 87.3–93.6% of the total As, whereas in the non-flooded and Alternate Wetting and Drying (AWD) treatments, As(V) was the dominant species, accounting for 90–96% and 73–83%, respectively (Das et al., 2016). In another field study in Bangladesh, the site employing intermittent irrigation showed lower grain-As content than the site under continuously flooded conditions (Stroud et al., 2011). A field study at Stuttgart, Arkansas, US showed that grain contained 41% lower As in an intermittently flooded paddy than in a continuously flooded paddy (Somenahally et al., 2011). There is a 13% increase in grain yield over the conventional cultivation method and, importantly, As concentrations in grain and straw are decreased by 62% and 86%, respectively (Talukder et al., 2011). A glasshouse study in Bangladesh indicates that the use of AWD techniques increases the grain yield at around 40% in As contaminated soils over water management options (Islam et al., 2015a). These literatures provide proof of the concept that water management can be a highly effective tool in controlling As bioavailability in paddy soils and the subsequent accumulation in rice grains. Arsenic concentrations in rice grains are also cultivar dependent and influenced by water management (Hua et al., 2011). Therefore, the selection of less As-responsive rice cultivars and the use of saturated water management in paddy fields could be an effective means of minimizing As accumulation in rice grains, thus reducing health risks of As exposure.

4. Human health exposure to As through rice

4.1. Rice consumption and exposure to As

Drinking water was considered the major exposure route of As but a number of studies have established that rice is also a significant exposure pathway of daily As intake (Halder et al., 2012; Jackson et al., 2012; Mondal and Polya, 2008; Rahman et al., 2011, 2009; Williams et al., 2006). Rice can be a major food source in the daily diet of the Asian population and can reach 67% of the daily Asian diet (Adomako et al., 2011; Brandon et al., 2014; EFSA, 2009; Li et al., 2011). Fig. 4 illustrated the process of As contamination in rice and the human health risk. Recently, dietary exposure studies have been reported in many countries. The estimated daily intake varies significantly from country to country. In Asian countries, the consumption of rice is very high at around 588 g day^{-1} (Correll et al., 2006; FAOSTAT, 2007) and it contributes a relatively high amount of As compared to other foods (Schoof et al., 1999). Depending on the age group, according to EFSA, rice accounted for 5% to 15% of the dietary intake of inorganic As (EFSA, 2014). According to Mondal et al. (2010) rice is the major route of inorganic As exposure in some blocks of West Bengal India and rice itself

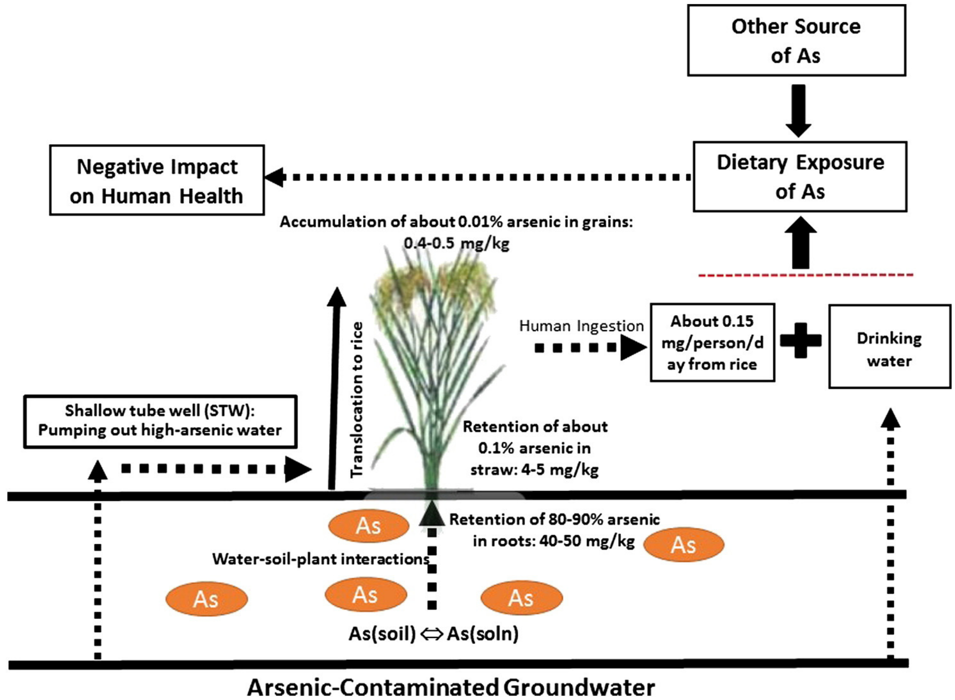

Fig. 4. Schematic illustration of As contamination in rice and the human health risk.

may contribute 12 to 34% of the total As. The regulation sets out limits for the adult population at 0.20 mg kg^{-1} for white rice, and 0.25 mg kg^{-1} for brown rice. Based on EFSA (2014) concentration data 0.089 mg kg^{-1} contribute 0.465 μg kg^{-1} BW day^{-1} As in high rice consumers (300 g raw rice).

Rice grown in Bangladesh, the world's hot spot for As poisoning, contains about 80% inorganic As, and people there eat 450 g day^{-1} (Potera, 2007). But the average daily rice consumption for adult ranges from 400 to 650 g (Ahsan and Del Valls, 2011; Rahman et al., 2006). This is one of the highest per capita rice consumption figures in the world. Consumption of As-contaminated rice may contribute as much as 60% of the daily dietary As intake based on conservative As concentrations in rice of Bangladesh (Meharg, 2004). When rice ingestion is assumed at 0.432 kg day^{-1} for a 60 kg adult, assuming bioavailability of 90% when cooked (Juhasz et al., 2006), then a grain inorganic As level of 0.053 mg kg^{-1} and 0.267 mg kg^{-1} would equate to 16% and 81% of the MTDI, respectively. A survey data showed that the average rice consumption rates for adult males and females were 432 and 420 g day^{-1} (uncooked basis) and there is no significant difference between adult males and females and this consumption rate may contribute 0.049 and 0.047 mg inorganic As day^{-1} for adult males and females, respectively (Rahman et al., 2011). Ohno et al. (2007) reported that adult males and females consume 776 and 553 g of cooked rice daily, based on their study from Bangladesh and rice contributes 56% of the total As intake which is greater than the total As intake from drinking water (13%). Another study from Bangladesh showed that adults (male or female) consumed 495 g day^{-1} of rice and this rice and water consumption contribute 0.888 and 0.706 mg inorganic As day^{-1} for adult males and females, respectively (Rahman et al., 2009). Fig. 5 indicates the daily intake of As from food (rice and vegetables) and drinking water. According to Smith et al. (2006b) rice and drinking water contribute 1.186 mg As daily in the study of Bangladesh. The intake of As through rice as a food source was 0.20 to 0.35 mg day^{-1} per adult (Rahman et al., 2008b). In India and Bangladesh the consumption of 420 g rice combined with 2 L of drinking water containing 0.05 mg of As L^{-1}, would equate to a dietary exposure for the people of India and Bangladesh of 0.121 and 0.155 mg As day^{-1}, respectively (Williams et al., 2005). Roychowdhury et al. (2003) reported 0.693 and 0.560 mg As daily intake for adult males and females, respectively in the Murshidabad district in West Bengal India and the daily intake of total As from water and food was 4.5 times higher than the tolerable daily intake with food itself contributing 27%.

The mean daily intake of inorganic As in a Cambodian study was 0.00187 mg kg^{-1} BW and none of the individuals exceeded the $BMDL_{0.5}$ of 0.003 mg kg^{-1} BW from rice consumption alone (Gilbert et al., 2015). In the Chinese population, the daily intake of inorganic As was approximately 0.042 mg day^{-1}, with rice as the largest contributor of inorganic As, accounting for approximately 60% of the total As (Li et al., 2011). But in Taiwan the average intake value was 0.079 to 0.104 μg kg^{-1} BW day^{-1} (Chen et al., 2015). A study by Torres-Escribano et al. (2008) in Spain showed that the rice with the highest inorganic As (0.253 mg kg^{-1}) would contribute 0.148 mg inorganic As intake from rice which is 99.2% of the TDI value. In Finland, the estimated inorganic As intake from long-grain rice and rice-based baby foods were close to the lowest $BMDL_{0.1}$ value of 0.0003 mg kg^{-1} BW day^{-1} set by the European Food Safety Authority (EFSA) for every age group (Rintala et al., 2014). There are average rice consumption figures of 15 g day^{-1} in France, 24 g day^{-1} in the United States (US), and even 218 g day^{-1} in China by comparison (Meharg et al., 2009). In Brazil the inorganic As intake from rice consumption was estimated at 10% of the PTDI values with a daily consumption 88 g of rice (Batista et al., 2011).

4.2. Rice processing and their effect on As contents

Traditionally rice is cooked with a substantial amount of excess water. Arsenic content in cooked rice is the main determinant in case of actual dietary exposure. Many studies have reported a significant decrease of As in brown and polished rice after washing several times. Recently, a study by Naito et al. (2015) reported that washing three times may reduce 71–83% of the total and inorganic As in polished rice. Sengupta et al. (2006) reported that washing of long grain rice 5 to 6 times may remove 28% of total As whereas washing long grain white rice 3 times removed 8–17% of total As (Mihucz et al., 2007). It differs significantly from that of raw rice depending on the As content in cooking water and processing methods (Pal et al., 2009; Rahman et al.,

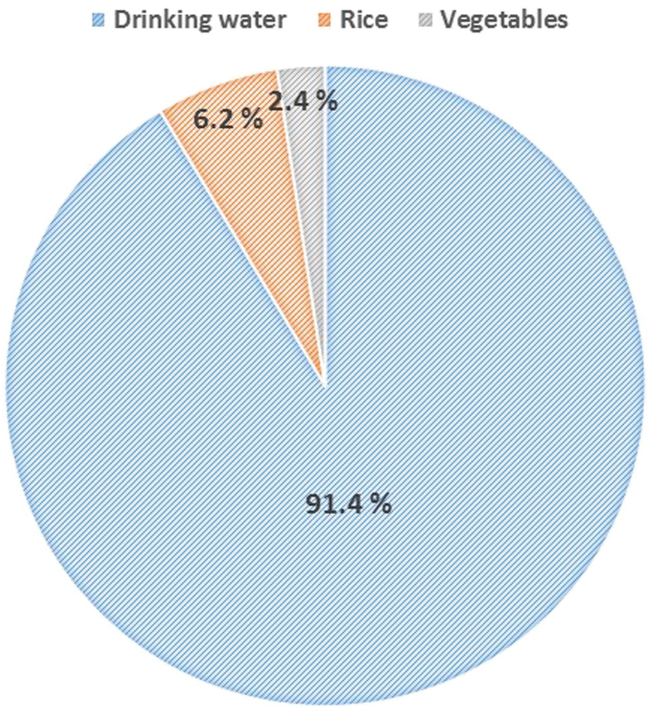

Fig. 5. Intake of As from food (rice and vegetables) and drinking water per day. (adopted from Rahman et al., 2013)

2006). The effect of cooking on speciation of As in grain lacks clarity given widely different results in the literature. Several reports indicate that the differences in As content in raw and cooked rice would be either rice varietal differences or concentration of As in cooking water (Huq and Naidu, 2003; Meharg and Rahman, 2003). The type of rice, percentage of water absorbed and the manner of preparation also affects the total As in cooked rice (Ackerman et al., 2005), however the duration of cooking also may affect the total As concentration in cooked rice (Rahman et al., 2006). In fractions of parboiled and non-parboiled rice grains the order of As concentrations was; rice husk > bran brown rice > raw rice > polish rice (Rahman et al., 2007c). From a household survey processing of rice (parboiling and milling) reduced As concentrations by an average of 19% (Duxbury et al., 2003).

Absorption of As-contaminated water during the rice cooking process may significantly increase the amount of As in cooked rice (Ackerman et al., 2005), which is often overlooked when calculating daily As intake values. Cooking rice in excess water efficiently reduces the amount of inorganic As in the cooked rice on an average by 40% from long grain polished, 60% from parboiled and 50% from brown rice (Gray et al., 2015). Another report from Rahman et al. (2006) indicates that when rice is cooked with excess water and the gruel was discarded, the concentration of As in cooked rice decreased. They attributed this to the release of water soluble As during cooking at high temperature and decantation of cooking water after cooking. This was further confirmed by cooking rice with limited water and the gruel (cooking water) was absorbed in rice. This led to a slight increase in As concentration in cooked rice (Rahman et al., 2006). The mean As concentration in 14 raw Aman rice samples was 0.153 mg kg^{-1} with a range of 0.074–0.302 mg kg^{-1} but after cooking the mean value is 0.139 mg kg^{-1} so there was no significant difference in the mean As concentrations between raw and cooked rice as As free pond water was used for cooking rice (Rahman et al., 2011).

Research shows that the cooking of rice with water does not make significant changes in total and inorganic As contents. On average 87% of inorganic As is present in cooked rice (Smith et al., 2006a) but Ohno et al. (2007) reported that it ranges from 97 to 102%. Pal et al. (2009) reported that cooking procedures did not change the nature of As in cooked rice. They showed that 95% of the recovered As present in cooked rice is inorganic when rice from contaminated areas and traditional cooking methods were used. On the other hand, when white rice was cooked without washing, the As levels in raw and cooked rice were almost the same and this indicate that total As and inorganic As in rice was decreased by washing only (Naito et al., 2015). Sengupta et al. (2006) showed that total As content in cooked rice depends strongly on the method of cooking when cooked with low As water. There was up to a 57% reduction of total As from rice by subsequent washing 5 to 6 times and high-volume (water: rice = 6:1) cooking followed by discarding excess water. Raab et al. (2009) reported that decanting of excess water after high-volume cooking effectively decreases total As and inorganic As in rice. In general the larger the water:rice cooking ratio, the greater the removal of more inorganic As. At a water-to-rice cooking ratio of 12:1, 57 ± 5% of inorganic As could be removed. Recently percolating technology proved highly effective in removing inorganic As from the cooking rice, with up to 85% removed from individual rice types (Carey et al., 2015).

4.3. Bioavailability and bioaccessibility of As from rice grains

Knowledge of As bioavailability is important to minimize the uncertainty in the risk of As. Bioavailability is often used as the main indicator of the potential risk that chemicals pose to the environment and human health (Naidu et al., 2008). The objectives of all bioavailability studies are to obtain the best possible estimate of the amount of available As that poses a potential risk to human health. However, most of the methods suffer from draw-backs, e.g. if the systems are artificially constructed and may not always simulate actual human physiological conditions. In an attempt to overcome these limitations, human volunteers or experimental animals have been used (Caussy, 2003). There are a limited number of in vivo studies using animals but they are almost all soil related with incidental high levels of As used (Bradham et al., 2011; Brattin and Casteel, 2013; Denys et al., 2012; Laird et al., 2013). Arsenic is relatively rapidly excreted from the body primarily through the urine (Cascio et al., 2011) and most of the inorganic As, As(III) and As(V), are metabolized to DMA and MMA prior to excretion through the urine. Therefore, monitoring the urinary excretion of As is one of the easy ways to determine bioavailability. Direct measurement of human exposure from rice has been assessed by urine sampling. There is only one, limited rice eating trial where rice consumption was measured and urinary excretion monitored in humans (Meharg and Zhao, 2012).

Naidu and his co-researchers assessed As bioavailability in cooked rice (glasshouse grown rice and supermarket bought rice) using an in vivo swine model (Juhasz et al., 2006). Arsenic speciation shows that supermarket bought rice contains 100% inorganic As whereas 86% of the glasshouse grown rice contains dimethylarsinic acid (Juhasz et al., 2006). Only 33 ± 3% of As was bioavailable in greenhouse grown rice cooked in water contaminated with sodium As(V) as the low absolute bioavailability of dimethylarsinic acid whereas in supermarket bought rice, As bioavailability was high (89 ± 9%). The bioavailability using in vivo swine models showed that the bioavailability in rice is highly dependent on As speciation, which in turn can vary depending on rice cultivars, As in irrigation water and cooking water. These findings suggest that rice genotypes rich in DMA are likely to pose less risk from exposure to As and subsequent toxicity.

The bioaccessibility of As from cooked rice with an in vitro dynamic digestion process and parboiled rice, which is most widely consumed in South Asia, showed a higher percentage of As bioaccessibility (59% to 99%) than non-parboiled rice (36% to 69%) and most of the As bioaccessible in the cooked rice (80% to 99%) was easily released during the first 2 h of digestion (Signes-Pastor et al., 2012). The estimation of the As intake through cooked rice based on the As bioaccessibility highlights that a few grams of cooked rice (<25 g dry weight per day) cooked with highly As-contaminated water is equivalent to the amount of As from

2 L of water containing the maximum permissible limit (0.0010 mg As L^{-1}). With As(V) and DMA being the predominant species present in cooked rice, very similar trends in the bioaccessibility of these As species were observed as upon digestion of the pure As standards range between 85% and 90% at pH 1.8 (Alava et al., 2013). The use of an enzymatic approach indicates that on average 94% of As is liberated during enzymatic extraction (Ackerman et al., 2005). The mean As concentration is 0.275 ± 0.161 mg kg^{-1} ($n = 31$) and rice samples with relatively high total As (>0.20 mg kg^{-1}, $n = 18$) has shown bioaccessibility ranging from 53% to 102% (He et al., 2012). Laparra et al. (2005) evaluated the bioavailability of inorganic As in cooked rice to better understand the possible health risks derived from the consumption of rice. The contents of total As in raw rice (0.05–0.53 μg g^{-1}) increased considerably after cooking (0.88–4.21 μg g^{-1}), as a consequence of the presence of As(V) in the cooking water. The total As content of the bio-accessible fraction (1.06–3.93 μg g^{-1}) demonstrates the high bio-accessibility of As from cooked rice (>90%) (Laparra et al., 2005).

4.4. Associated health risk from the ingestion of rice

It is evident from existing literature that the presence of As in the environment has adverse consequences to human health. Arsenic is a chronic carcinogen and exposure to elevated levels causes to serious illness including different types of cancers (IARC, 2004a, 2004b). Inorganic As in drinking water has been studied and is linked to human carcinogenesis and exposure. Fairly constant levels of a mixture of As metabolites are generally excreted in urine, i.e. 10–30% inorganic As, 10–20% MMA, and 60–70% DMA (Vahter, 1999) and it is associated with various internal cancers viz. liver, bladder, kidney, and lungs as well as other health problems, including skin cancer and diabetes (Guo et al., 1997; IARC, 2004a). Inorganic As is metabolized by consecutive reduction and oxidative methylation in the liver and is largely excreted via urine (Yamauchi et al., 1989). This process is considered to be a detoxification mechanism because the major methylated metabolites MMA and DMA are easily excreted and are less acutely toxic than the inorganic species (Gebel, 2002; Thomas et al., 2001). In a clinical study of 18,000 persons in Bangladesh and 86,000 persons in West Bengal India from As-affected areas showed that of these, 3695 (20.6% including 6.11% children) in Bangladesh and 8500 (9.8% including 1.7% children) in West Bengal had arsenical dermatological features (Rahman et al., 2001). A large portion of the total population is highly vulnerable to various internal cancers. However, the exposure time to develop arsenicosis varies from case to case reflecting its dependence on As level in drinking water and food, nutritional status, genetic variant of the human being and other compounding factors (Anawar et al., 2002).

Arsenic contamination of rice grains and the low concentration of micronutrients in rice have been recognized as a major concern for human health (Lombi et al., 2009; Williams et al., 2009). Therefore, rice itself plays a potential exposure route to humans but there is very limited human data as evidence for this impact on humans directly from rice. Banerjee et al. (2013) reported that cooked rice with 0.2 mg kg^{-1} As was associated with genotoxic effects measured in micronuclei in urothelial cells. A study from India showed that the chronic daily intake (CDI) and health risk index (HRI) were >1 for rice indicating the potential health risk from the middle Ganga plains of India (Kumar et al., 2016). Several studies reported the incremental lifetime cancer risk from the consumption rice. Excess lifetime cancer risks for rice consumption by country is presented in Table 4. The US EPA has an upper limit of acceptable risk for cancer from any given source of 1 in 10,000 (Kavcar et al., 2009; Tsuji et al., 2007). This is a useful figure with which to consider cancer risks from inorganic As of rice. Tsuji et al. (2007) also calculated that at the 95% 6.1 μg day^{-1} inorganic As ingestion rate for rice, at a slope of 1.5 mg kg^{-1} BW day^{-1}, for a 65 kg person, this equates to an excess skin cancer risk of 1.4 in 10,000 and using the slope of 3.67 per mg kg^{-1} day^{-1}, this equates to an excess cancer rate of 3.4 in 10,000. The probabilistic risk assessment model calculated median increased lifetime cancer risk due to cooked rice intake was 7.62 per 10,000 and was attributable to eating rice and rice contributes about 44% to this median risk (Mondal and Polya, 2008). The risk levels of As from rice for the people of Bangladesh, China and India had median risks of 22, 15 and 7 in 10,000, respectively (Meharg et al., 2009). A study from rural Bangladesh showed that cooked rice is the second highest contributor towards the adverse health risk with an overall risk 7.59×10^{-4} (Khan et al., 2012). In the adult Chinese population, the incremental life-time risk of cancer from food intake was 106 per 100,000 (Li et al., 2011). Chen et al. (2015) reported that the incremental lifetime cancer risks for the Taiwan population were 7.9 and 10.4 per 100,000 for females and males, respectively. Meanwhile, the non-cancer health hazard index for the daily intake of rice showed that the toxic risk due to As was 7.8 times greater than the reference dosage (Lee et al., 2008). Thus, rice is a major potential source and exposure pathway of As in the As affected areas where people are exposed to very low levels of As via drinking water.

5. Concluding remarks

Naturally occurring As has been detected in many parts of the world, and populations of south-east Asian countries are adversely affected through drinking water and food. During the last 50 years there has been a major shift in the food dietary intake by people largely related to global migration that has seen many Asians relocate to Western countries — both as students and as professionals. This has resulted in a significant change in food dietary intake with rice now being a staple food for half of the world's populations. As a consequence, consumption of inorganic As through rice poses significant health risks including cancers to humans. Paddy rice accumulate As from the soil and irrigation water and the ingestion of this As-contaminated rice acts as an exposure route to humans in any stage of life. Arsenic also has a negative effect on the growth, yield and quality of rice. Recognition of the poisoning of thousands of people in Bangladesh, West Bengal, India and parts of China, has led to a substantial progress in the understanding of As bioactivity, their exposure pathways, mechanism of bioaccumulation,

Table 4
Excess lifetime cancer risks from rice consumption by country.

Country	Polished rice consumption (g day^{-1})	Median inorganic As content of rice (mg kg^{-1})	Country specific rice inorganic As intake (mg day^{-1})	Country specific rice excess cancer rate per 10,000	References
Bangladesh	445	0.081	0.036	22.10	Meharg et al. (2009)
China	218	0.109	0.024	15.20	Meharg et al. (2009)
India	192	0.059	0.011	6.90	Meharg et al. (2009)
	–	–	–	7.62	Mondal and Polya (2008)
Italy	17	0.071	0.001	0.70	Meharg et al. (2009)
Taiwan	–	–	–	1.04	Chen et al. (2015)
US	24	0.088	0.002	1.30	Meharg et al. (2009)
	–	–	0.006	1.4–3.4	Tsuji et al. (2007)

speciation pattern and their toxicity levels in soil-water-plant systems. Arsenic speciation draws major attention because toxicity totally depends on the speciation pattern and it varies from variety to variety and location to location. However, environmental variation is more prominent than genotypic variation. Potential strategies are required to minimize As exposure from water, soil and food. Different management practices such as varietal selection, fertilizer and nutrient management, irrigation water management and cooking practices showed significant variation on As bioaccumulation, speciation and their bioavailability. There is an urgent need to develop a mitigation strategy to minimize As accumulation in rice grains. So, it is crucial to reduce As transfer from soil to rice grains to reduce human exposure risk from rice. Because As intake from rice represents an important route of exposure, especially for people consuming a large amount of rice in their diet, mitigation measures to reduce As accumulation in rice are urgently needed through genetic variation or identification of rice varieties which accumulate low As and effective water management strategies. Current knowledge of As contamination including bioavailability and bio-accessibility should be used to minimize As exposure.

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