

Arsenic Fate and Transport in the Groundwater-Soil-Plant System: An Understanding of Suitable Rice Paddy Cultivation in Arsenic Enriched Areas

P. K. Sahoo and A. Mukherjee

Abstract Irrigation with arsenic (As)-rich groundwater in agricultural soil is one of the major causes for As contamination in rice. This can be observed in Bangladesh where the highest levels of As were reported in rice grains. However, As risk assessment in rice based on the As contaminated groundwater and soil can be misleading, since As accumulation in plant is controlled by a number of factors in the soil–rhizosphere system. In this chapter, we have discussed the impact of As-rich groundwater on rice cultivation, and the major factors which control the fate of As in the soil–rhizosphere–plant system. Soil parameters such as pH, redox potential, and concentrations of phosphorous, sulfur, silica, and iron affect the availability of As in the soil–rhizosphere zone, thus, should be considered in the assessment of As toxicity for rice. Concentration of silica and phosphate compete with arsenite and arsenate respectively during uptake by plants. Environmental factor such as soil flooding condition is also one of the potential factors influencing arsenic accumulation in rice by increasing As mobility in the soil–rhizosphere. In biological factors, rice genotype, which controls the root aeration, is significant in affecting accumulation of As. Therefore, selection of suitable rice genotype besides growing rice in aerobic conditions would be important to minimize As accumulation in rice. The translocation of As from roots to above-ground parts is dependent on As speciation. Although, organic As is more readily translocated, its uptake is much lower compared to inorganic As; thus, inorganic As predominates in grains.

Keywords Arsenic · Rice · Bioavailability · Soil–Rhizosphere system · As uptake mechanism

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1 Introduction

Arsenic (As) is a ubiquitous element that ranks 20th among the most abundant elements in the earth's crust (Cullen and Reimer 1989). Its atomic number is 33, and the atomic weight is 74.92. Arsenic is a redox-sensitive element, which exists mainly in four oxidation states, +V (arsenate), +III (arsenite), 0 (arsenic), and -III (arsine). Unlike other toxic elements (Pb, Cu, and Co), As occurs as cation (positively charged ion) and oxyanion (negatively charged ions with oxygen) species. It can occur in the environment both in organic and inorganic forms. Inorganic forms usually associate with many minerals and other elements, especially oxygen, sulfur, and chlorine, while organic forms associate with carbon and hydrogen. The most important inorganic species are arsenate As(V) and arsenite As(III), while the most important organic species are monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA). The toxicity of As is mainly dependent on the forms of arsenic. Inorganic As species are more toxic to living organisms than organic forms (Meharg and Whitaker 2002). Arsenic toxicity in humans can cause severe health problems ranging from skin lesions to cancers of the brain, liver, kidney, and stomach, and many other cardiovascular and neurological diseases (Smith et al. 1992; Ng et al. 2003; Martinez et al. 2011).

Arsenic naturally occurs in many environmental media, such as rocks, soil, sediments, and surface/groundwater, and it can further be released into the aquatic and terrestrial ecosystem via natural and anthropogenic activities (Lombi et al. 2000). According to the World Health Organization (WHO) guideline, the baseline limit of As concentration in safe drinking water is 10 $\mu\text{g/L}$, which is similar to the European Union (EU), US Environmental Protection Agency (USEPA), Netherlands, and Germany limits of As in drinking water (Ahsan and DeIvalls 2011). However, in Bangladesh and many other developing countries, such as India, China, and Nepal, it is set to 50 $\mu\text{g/L}$ (World Bank 2005). Currently, Australia has set a strict national standard of 7 $\mu\text{g/L}$ for acceptable As concentration in drinking water (World Bank 2005). Arsenic poisoning via drinking of As contaminated groundwater is well documented (Smedley and Kinniburgh 2002); however, recent investigation revealed that rice, which is the staple food for nearly half of the world's population, is another potential As exposure pathway to human health (William et al. 2005; Zavala et al. 2008). It can add significant amounts of dietary intake of As than most crops because rice plants have the special ability to soak up As from soil compared to other cereals (Heikens 2006), and use of arsenic-contaminated groundwater for irrigation of rice cultivation (Heikens et al. 2007). However, accumulation of As in rice is influenced by various environmental, geochemical, and biological factors which controls As solubility, bioavailability, and uptake in the soil-rhizosphere-plant system (Walter and Wenzel 2002; Zhao et al. 2010).

This chapter presents the As levels in rice from different parts of the world, assesses the impact of irrigation with As rich-groundwater on soil and paddy rice, and discusses the major factors controlling As behavior in the soil-rhizosphere-plant systems, along with As toxicity to plants. Understanding of these aspects may be helpful to reduce As accumulation in rice grains.

2 Arsenic Concentration in Rice

Several surveys of As concentration in rice grains have been carried out in various countries (Table 1). It was found that As concentration in rice varied widely. The highest level of As, up to 1.85 mg/kg (0.05–1.85 mg/kg), was found in Bangladesh (Meharg and Rahman 2003). Other Asian countries also reported high levels (ranges) of arsenic in rice: China, 0.46–1.18 mg/kg (Sun et al. 2008); Taiwan, 0.1–0.63 mg/kg (Lin et al. 2004); Thailand, 0.06–0.5 mg/kg (Adamako et al. 2011); Japan, 0.07–0.42 mg/kg (Meharg et al. 2009); Korea, 0.24–0.72 mg/kg (Lee et al. 2008); and India, particularly from West Bengal, 0.085–0.66 mg/kg (Roychowdhury et al. 2002). Besides Asian rice, significant arsenic levels were reported from Venezuela (0.19–0.46 mg/kg) by Schoof et al. (1998), France (0.12–0.61 mg/kg) by Williams et al. (2007a), and USA (0.16–0.71 mg/kg) by Zavala et al. (2008).

Rice As values (ranges) from Europe, 0.13–20 mg/kg (Williams et al. 2005); Spain, 0.07–0.21 mg/kg (Williams et al. 2007a); Italy, 0.19–0.22 mg/kg (Williams et al. 2005); and Ghana <0.01–0.15 mg/kg (Adomako et al. 2011) fall into intermediate category. Some parts of the Asian rice also fall into this category, as follows: Philippines, 0–0.25 mg/kg (Williams et al. 2006); Vietnam, 0.09–0.17 mg/kg (Adomako et al. 2011); and Pakistan, 0.01–0.2 mg/kg (Adamako et al. 2011). On contrary, rice from Australia, 0.02–0.04 mg/kg (Williams et al. 2006); Canada, 0.02 mg/kg (Williams et al. 2005); and Egypt (0.02–0.08 mg/kg) (Meharg et al. 2007) is found to contain the lowest levels of As contamination (Table 1). The overall range found among all samples, the lowest grain As (0.01 mg/kg) is nearly two order magnitude difference with that of the highest concentration (1.835 mg/kg). According to the information available to us, in terms of As contamination levels in rice, there is no limit set by EU and US (Francesconi 2007). However, China has regulated the maximum contamination level (MCL) of As in rice as 0.15 mg/kg (Zhu et al. 2008). On comparison with Table 1, it was noticed that the mean values of most of the Asian, USA, and France rice were exceeded the MCL. However, higher enrichment in Asian rice, particularly in Bangladesh, which can be the significant source of dietary As for population of this area and population from other countries import rice from this region.

3 Arsenic in Groundwater

Arsenic contamination of groundwater has been identified in many countries, including Argentina, Bangladesh, China, Cambodia, India, Japan, Mongolia, Mexico, Nepal, Taiwan, Thailand, Vietnam, and USA (Mandal and Suzuki 2002; Smedley and Kinniburgh 2002). There are a number of people are at risk of chronic arsenic poisoning from drinking As-contaminated water in several parts of the world, however highest number of people at risk from Ganges Delta (Bangladesh and West Bengal, India) (Fig. 1) (Smedley and Kinniburgh 2005; Garelick and Jones 2008).

Table 1 Arsenic concentration (in mg/kg) in rice grains from different countries

Country	Maximum	Minimum	Average	No. of samples	Study (Author/year)
Bangladesh	1.835	0.058	0.49	13	Meharg and Rehman (2003)
China	1.18	0.46	0.82	2	Sun et al. (2008)
India	0.66	0.085	0.3	10	Roychowdhury et al. (2002)
Taiwan	0.63	0.1	0.1	5	Lin et al. (2004)
Thailand	0.5	0.06	0.15	53	Adomako et al. (2011)
Korea	0.72	0.24	0.41	–	Lee et al. (2008)
Japan	0.42	0.07	0.19	26	Meharg et al. (2009)
Vietnam	0.17	0.09	0.12	18	Adomako et al. (2011)
Philippines	0.25	0	0.07	–	Williams et al. (2006)
Pakistan	0.2	0.01	0.09	5	Adomako et al. (2011)
Venezuela	0.46	0.19	0.3	4	Schoof et al. (1998)
Egypt	0.08	0.02	0.05	108	Meharg et al. (2007)
Spain	0.21	0.07	0.13	10	Williams et al. (2007a)
Europe	0.2	0.13	0.15		Williams et al. (2005)
France	0.61	0.12	0.32	22	Williams et al. (2007a)
USA	0.71	0.16	0.29	24	Zavela et al. (2008)
Italy	0.22	0.19	0.14	–	Williams et al. (2005)
Ghana	0.15	<0.01	–	7	Adomako et al. (2011)
Australia	0.04	0.02	0.03	5	Williams et al. (2006)
Canada	–	–	0.02	–	Williams et al. (2005)
Lebanon	0.07	0.01	0.04	11	Adomako et al. (2011)

“–”: not available

Scores of people from China, Vietnam, Taiwan, Chile, Argentina, and Mexico are likely at risk as well (Fig. 1).

3.1 Arsenic Mobility in Groundwater

Based on the As geochemistry, there are three major mechanisms controlling As mobility in the groundwater, which have been reported by various workers. These are:

Oxidation of Pyrite Mobilization of As due to oxidation of arsenic-rich pyrite minerals associated with sediment and rock materials have been identified by many workers (Williams et al. 1996; Kim et al. 2012) (Table 2). Insoluble As-bearing minerals such as arsenopyrite (FeAsS) are rapidly oxidized when in contact with oxygen, releasing soluble As(III) and Fe(II) (Smedley and Kinniburgh 2002). The oxidation of the As-bearing minerals is dependent on availability of oxygen. High oxygen in pyritiferous system suffers by the excess pumping and water-table draw-down. Chakraborti et al. (2001) and Sikdar et al. (2001) observed that water table declined in the Bengal basin because of the agricultural pumping that began in



Fig. 1 Worldwide distribution of arsenic contaminated regions, illustrating numbers of people at risk of chronic As exposure. (Adapted from Garelick and Jones 2008)

Table 2 As concentration in groundwater ($\mu\text{g/L}$) relations with major release mechanism reported from different countries

Country	As in groundwater	Major release mechanism	References
USA	Up to 12,000	Oxidation of As-rich sulfides	Schreiber et al. (2000)
Thailand	1.25–5114	Oxidation of As-rich sulfides	Williams et al. (1996)
Korea	23–178	Oxidation of As-rich sulfides	Kim et al. (2012)
Bangladesh	Up to 640	Reductive dissolution of Fe or Mn (hydro)oxides	Harvey et al. (2002)
Bangladesh	6–934	Reductive dissolution of Fe or Mn (hydro)oxides	Bibi et al. (2008)
India	0.77–1059	Reductive dissolution of Fe or Mn (hydro)oxides	Bhattacharyya et al. (2008)
Nepal	< 10–740	Reductive dissolution of Fe or Mn (hydro)oxides	Gurung et al. (2005)
China	0.6–572	Reductive dissolution of Fe or Mn (hydro)oxides	Guo et al. (2008)
Taiwan	Up to 12,000	Reductive dissolution of Fe or Mn (hydro)oxides	Chen and Liu (2007)
Vietnam	1–741	Reductive dissolution of Fe or Mn (hydro)oxides, oxidation of As-rich sulfides ^a	Naguyen and Itoi (2009)
USA	11–2620	Reductive dissolution of Fe or Mn (hydro)oxides, Desorption from Fe (hydro)oxides	Welch and Lico (1998)
India	0.82–538	Reductive dissolution of Fe or Mn (hydro)oxides, Desorption from Fe (hydro)oxides ^a	Kar et al. (2010)
Argentina	7–14,969	Desorption from Fe (hydro)oxides	Bhattacharya et al. (2006)

^a Minor mechanism

the 1970s. Subsequently, in 1978, first As contamination in the Bengal Basin was detected. The rate of oxidation of pyrite is dependent on the redox potential (Eh) and pH. The release of Fe from pyrite oxidation can form Fe oxides that can immobilize As.

Reductive Dissolution of Fe Oxyhydroxides Arsenic in Fe-oxyhydroxides is mobilized when the environment becomes anaerobic (Smedley and Kinniburgh 2002). Bhattacharya et al. (1997) first proposed the reductive dissolution of Fe oxyhydroxides process for arsenic release. After that, many other studies have been reported this mechanism (Welch and Lico 1998; Harvey et al. 2002; Bibi et al. 2008; Gurung et al. 2005; Chen and Liu 2007; Bhattacharyya et al. 2008; Guo et al. 2008; Kar et al. 2010) (Table 2). McArthur et al. (2001) stated that the good correlation between As and HCO_3^- in groundwater is an indication of reduction. Other studies from Bengal basin reported that low Eh, O_2 , NO_3^- , and SO_4^{2-} , and high Fe and Mn in groundwater are the indication of reducing conditions (Bhattacharya et al. 1997; Dowling et al. 2002). Similarly, Kim et al. (2009) and Sahoo et al. (2013) stated that the higher arsenic concentration in groundwater associated with lower Eh, NO_3^- , and

SO_4^{2-} , and higher alkalinity, Fe, and PO_4 is the indication of reductive dissolution of Fe (hydr)oxides.

Desorption of Fe-Oxyhydroxides Besides anaerobic conditions for releasing As from Fe oxyhydroxides, there is also effect of pH, particularly at $\text{pH} > 8.5$, which can cause mobilization of As from Fe-oxides. This is identified by some workers (Bhattacharya et al. 2006; Kar et al. 2010) (Table 2).

4 Impact of As-Rich Groundwater on Paddy Cultivation

Arsenic concentration in paddy fields can occur naturally via weathering processes, and/or by anthropogenic activities such as mining (Zhu et al. 2008), pesticide use, fertilizer application (Williams et al. 2007b), and irrigation with As contaminated groundwater (Mehrag and Rahman 2003; Williams et al. 2006). However, irrigation with As contaminated groundwater plays a dominant role, especially in Asian countries where groundwater-irrigated rice cultivation has increased dramatically over the past few decades to maintain the country's food security. In the long term, use of this As-contaminated irrigation water can cause elevated As concentration in paddy soils and crops (Heikens et al. 2007; Hossain et al. 2008; Baig et al. 2011). Several field studies from Bangladesh and West Bengal showed that irrigation with As-rich groundwater increases the As levels in the top soil (Mehrag and Rahman 2003; Norra et al. 2005; Williams et al. 2006; Roychowdhury et al. 2002). Studies from Argentina, Chile, and Taiwan (Chen et al. 2002; Tu and Ma 2004) also reported that As contents in agricultural soils are significantly higher than the normal levels due to irrigation with As-rich groundwater.

There are a number of studies that investigated the impact of irrigation with As-rich groundwater on soil and its subsequent impact on rice grains (Mehrag and Rahman 2003; Williams et al. 2006; Dittmar et al. 2010). Mehrag and Rahman (2003) reported the positive relations among As in irrigation water, soil, and rice from Bangladesh. Williams et al. (2006) did an extensive sampling of rice throughout Bangladesh, and found that high As concentration in rice is associated with As contaminated irrigation water. In another study from Murshidabad district of Bangladesh, Roychowdhury et al. (2005) reported the positive relations among As concentrations in irrigated water, soil, and plants. Recently, Singh et al. (2010) observed the positive relations between irrigation water and rice. However, this relationship is conflicting in some cases. Norra et al. (2005) found no significant relationship between rice grains from West Bengal, India, where plants are irrigated with low and high concentrations of As containing water. Van Geen et al. (2006) reported that there was no evidence of a proportional transfer As from soil to rice grains, despite the soil irrigated with As rich-groundwater. Furthermore, Williams et al. (2007a) reported elevated As concentration in rice grains in background paddy soil. These observations indicate that As content in rice is not directly dependent on total concentration of As in soil and irrigation water, but may be with other factors in the soil-rhizosphere-plant system, which are relevant.

5 Arsenic Behavior in Soil–Plant System

5.1 Soil

Arsenic is naturally present in soil. Its average concentration in non-contaminated soils is ~ 5 mg/kg; however, the concentration can increase up to 27,000 mg/kg or more than that in contaminated soil (McLaren et al. 1996). The solubility and bioavailability of As in soil is influenced by various factors (Fitz and Wenzel 2002; Zhao et al. 2009), which are discussed below.

As Speciation

Arsenic in soil exists in both inorganic and organic forms. The most common inorganic As species in soil are As(V) and As(III), while organic forms are MMA and DMA (Zhao et al. 2009). As(III) is more toxic than As(V), and also much more toxic than MMA or DMA (Zhao et al. 2010). Inorganic As species are dominating in paddy soils, whereas organic species are in low quantities (Fitz and Wenzel 2002). Inorganic species can be converted to organic form by methylation linked with microbial action in paddy soil (Takamatsu et al. 1982). Each species has different solubility and bioavailability. Marine et al. (1992) reported that the As availability to rice plants followed the order $\text{As(III)} > \text{MMA} > \text{As(V)} > \text{DMA}$. Meharg and Whitaker (2002) also observed that both As(III) and MMA are more available to rice plants. This indicates that speciation of As in soil environment is essential to assess As toxicity to plants.

Redox Potential

The balance of reducing and oxidizing condition (redox status) in the soil is important because it controls arsenic mobility and speciation (Fitz and Wenzel 2002). Under oxidizing conditions (aerobic), As(V) usually dominates, which has strong affinity for soil minerals such as Fe-oxhydroxides, leading to decrease As solubility and bioavailability to plants (Takahashi et al. 2004; Xu et al. 2008). However, under reducing conditions, such as soil flooding, As(III) is the most common inorganic species, and its mobility is sharply increased in the soil-rhizosphere (Takahashi et al. 2004). This is due to the reductive dissolution of Fe-oxhydroxides, and relatively high abundance of Fe reducing bacteria and algae in reduced soil, which help in As solubility via reduction of As(V) and methyl As species to more soluble As(III) species (Horneman et al. 2004; Mahimairaja et al. 2005). Therefore, in flooded soil, As is readily available for plant uptake. This may be the cause for higher As content in lowland crops usually growing in a reducing environment, such as paddy rice, than upland cereal crops (Williams et al. 2007a). However, changing irrigation practice can manage As uptake in plants (Das et al. 2008). Somennahally et al. (2011) con-

ducted an experiment by changing the irrigation process, and found that rice takes up less As in intermittently flooding than continuous flooding. Other studies also supported that irrigation practice can significantly control As accumulation in plants (Arao et al. 2009; Roberts et al. 2011; Rahaman et al. 2011; Talukdar et al. 2011).

pH

The sorption of As to metal-oxyhydroxides is generally affected by soil pH (Fitz and Wenzel 2002; Quazi et al. 2011), but there is not any universal agreement on this issue. Arsenic (AsV) tends to be sorbed by Fe–Al oxyhydroxides at near neutral to acidic pH (Mukherjee et al. 2009). However, at extremely acidic pH ($\text{pH} < 3$), arsenic mobility can increase due to the dissolution of arsenic binding species, such as Fe and Al oxycompounds (Signes-Pastor et al. 2007). Furthermore, the mobility of As in soil can increase at higher soil pH, above pH 8.5, because at high pH, mineral surfaces (mainly Fe-oxides) become progressively negatively charged that facilitate desorption of As from Fe-oxides (Marine et al. 1992; Streat et al. 2008), and increases labile As in the rhizosphere, leading to more As accumulation in plants (Fitz and Wenzel 2002). Campbell et al. (1985) observed that uptake of arsenic in plants increases at higher soil pH. In a recent work, Ahmed et al. (2011) also observed the positive relationship between grain As and soil pH.

Organic Matter

Organic matter (OM) can have a profound effect on As solubility in the soil since it tends to form insoluble and soluble complexes with As (Mukhaopadhyay and Sanyal 2004). Pikaray et al. (2005) reported that organic matter has a greater potential for As sorption due to formation of organo-As complex. Thus, high OM containing soil can reduce As availability to plants. Considering this effect, Rahaman et al. (2011) conducted an experiment, and observed that organic amendments significantly reduce arsenic content in rice grains. This can be supported by Fu et al. (2011) who found the negative correlation between As concentration in rice grain and soil OM.

On the contrary, organic matter can increase arsenic mobility in soil by forming water-soluble complexes with As, leading to inhibit As sorption on mineral surfaces (Bauer and Blodau 2006). Dissolved organic matter contains negative charge, which has high potential to compete with As for sorption sites in the soil, thus, it can increase As mobility (Lin et al. 2008). Furthermore, dissolved organic matter can promote As release by changing the redox chemistry of site surfaces and As species (Wang and Mulligan 2006). Selim Reza et al. (2010) reported that As is released by the reductive dissolution of Fe-Ox hydroxides linked to organic matter. Others studies also supported that As solubility in soil is positively related with organic matter (Hunag et al. 2006; Harvey et al. 2002; Rowland et al. 2006). This may be the cause for the positive relationship between rice and OM reported by Bhattacharya et al. (2010a), and elevated grain As in paddy soil even at background soil (Williams et al. 2011).

Soil Texture

Soil texture is another important factor that can influence arsenic mobility (Fitz and Wenzel 2002; Quazi et al. 2011). In general, clay or finer texture soils have much more surface area than coarse or sandy soils. In addition, Fe oxides are mainly present in the clay size soil fraction; therefore, clayey soils can have a higher As retention potential compared to sandy soils (Fitz and Wenzel 2002; Mahimairaja et al. 2005; Heikens et al. 2007), and soils with clayey texture are supposed to be less toxic of arsenic to plants compared to sandy soils (Heikens et al. 2007). Sheppard (1992) observed that phytotoxicity of As is five times more in sands and loams than clay soils. Similar studies have reported that plant uptake of As is high in loamy sand and low in silty clay loam (Woolson 1973; O'Neil 1995).

As Bound to Fe–Mn Oxides

Oxides of Fe and Mn are common in soils, and these phases are very efficient in sorbing As due to their high sorption capacity (Dudas 1987). However, their sorption property is strongly dependent on the environmental conditions (Mukherjee et al. 2009). Under aerobic conditions, the affinity of the oxyhydroxides is very high for As. However, under flooding conditions, oxyhydroxides release As from soil by the reductive dissolution of Fe oxyhydroxides, leading to more As available for plant uptake (Fitz and Wenzel 2002; Takahashi et al. 2004). Fu et al. (2011) found the positive relations between Fe–Mn bound As and grain As. Moreover, amorphous or poorly crystalline Fe oxides are much more readily dissolved than the crystalline Fe oxides under flooding conditions (Biswas et al. 2003). Ahmed et al. (2011) observed the significant positive relations between As in Fe-amorphous fraction and rice. This indicates that As content in amorphous/poorly crystalline Fe-oxides in soil can be an important factor for rice As.

Phosphate

Phosphate (PO_4) is an important parameter in agricultural field not only as an important nutrient, but also it controls As phytoavailability (Woolson et al. 1973; Peryea and Kammereck 1995; Bodgan and Schenk 2009). Generally, phosphate is a chemical analog of As(V) (Fitz and Wenzel 2002); thus, it competes with As(V) to occupy the same sorption sites in soils, leading to increase As solubility (Smith et al. 2002; Fitz and Wenzel 2002). Many studies reported that addition of PO_4 increases As mobility in soil (Peryea and Kammereck 1995; Smith et al. 2002; Campos 2002; Cao et al. 2003; Signes-Pastor et al. 2007), which can subsequently increase As uptake by plants (Bogdan and Schenk 2009; Islam and Jahiruddin 2010). However, it is conflicting at some point, since, PO_4 also competes with As(V) at the same transport during uptake, and can reduce arsenate accumulation in plants (Fitz and Wenzel 2002). This is supported by several works (Abedin et al. 2002a; Tu and Ma 2003; Lihong and Guilan 2009).



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