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REVIEW PAPER

Arsenic Accumulation in Food Crops: A Potential Threat in Bengal Delta Plain

Sukalyan Chakraborty · Md. Osaid Alam · Tanushree Bhattacharya · Yogendra Nath Singh

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Abstract The Bengal Delta Plain formed by the sedimentation of three main rivers, Ganga, Brahmaputra and Meghna along with its several tributaries and distributaries have been found to contain significant amounts of arsenic in its groundwater and sediments due to hydrogeochemical, geological and biological factors which enhance the mobilisation of arsenic. India and Bangladesh have been severely affected by occurrences of vast number of arsenic toxicity- related diseases and deaths. The problem has gained a serious magnitude due to uptake, translocation and biomagnifications of arsenic in several food crops and vegetables. A number of associated factors including the type of plants affects the bioavailability, uptake and translocation of arsenic in crops, which has been discussed in the present communication. The present paper reviews the status of arsenic contamination, mobilisation and uptake in crops of India, Bangladesh specifically along with other arsenic- affected crops worldwide and related health impacts due to the consumption of arsenic laden water and food.

Keywords Arsenic · Bioavailability · Crops · Uptake · Health impacts

Introduction

In the year 1983, the first occurrence of arsenic-induced skin lesion was observed at School of Tropical Medicine in Calcutta and by 1987 several patients from West Bengal, India and Bangladesh were detected with arsenic-related diseases (Saha 1995). Since then cases of drinking water contaminated with arsenic cases have been reported from over seventy countries of the world with a potential risk to around 150 million people (Ravenscroft et al. 2009). The World Health Organization (WHO) has taken an initiative to set a drinking-water standard of 10 $\mu g\,L^{-1}$ (or 0.01 $mg\,L^{-1}$) in the last edition of WHO GDWQ (1993). About 20 major incidents of arsenic contamination in groundwater have been reported from all over the world including 10 countries from South and South-east Asia: India, Bangladesh, China, Nepal, Taiwan, Pakistan, Cambodia, Laos, Myanmar and Vietnam. These countries have suffered from arsenic calamity in the last few decades (Brammer 2008). Among all the reported cases, arsenic contamination of groundwater in the Bengal Delta affecting India and Bangladesh has been the most devastating one in history of human kind (Smith et al. 2000). In India, cases of arsenic toxicity due to drinking of arsenic-contaminated water were first reported from Chandigarh in early 1976 (Datta and Kaul 1976). In 1984, a large number of arsenic-induced skin lesion cases were reported from Kolkata, West Bengal. Since then many incidences of chronic arsenic toxicity have been reported in many States adjoining the upper, middle and lower Ganga and Brahmaputra plain. Arsenic contamination has been reported in the States of West Bengal, Bihar, Uttar Pradesh, Jharkhand, Chhattisgarh, Assam and Andhra Pradesh (Nickson et al. 2007). In the north eastern part of India, arsenic was detected in Assam, Arunachal Pradesh, Manipur, Nagaland and Tripura (Singh 2004). Fifteen districts in Bihar exhibited high arsenic contamination on either side of the banks of river Ganga (Kinniburgh and Smedley 2001). Arsenic contamination has been observed in eastern part of Ambagarh Chowki block of Rajnandgaon district in Chhattisgarh (Chaurasia et al. 2012). In Jharkhand, SOES has reported three blocks of Sahibganj, namely Rajmahal, Udhwa and Sahibganj to be arsenic

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S. Chakraborty (✉) · Md. O. Alam · T. Bhattacharya · Y. N. Singh
Environmental Science and Engineering Group, Birla Institute of Technology, Ranchi, Jharkhand 835215, India
e-mail: su_kalyanc@yahoo.co.uk

affected. 27 villages of Dhemaji and Karimganj district of Assam and areas of Thumbil and Imphal district of Manipur are also reported to be contaminated with arsenic. Occurence of large number of arsenic- induced skin lesions cases was reported from Ballia district of Uttar Pradesh during 2003. Significant amount of research conducted by SOES, Jadavpur University in recent years indicates that large stretches of the Gangetic plain in different states are arsenic affected, thousands are suffering from arsenic toxicity and a large number of people have been threatened by the severe health effects of arsenic toxicity (Chakraborti et al. 2004, 2013). UNICEF, in collaboration with the Public Health Engineering Department (PHED), Government of West Bengal, tested eight arsenic-affected districts of West Bengal (Nickson et al. 2007). Half of the districts of West Bengal (nine out of eighteen) has been found to be contaminated with arsenic (Samal 2005). Depending on the magnitude of arsenic concentration in the state of West Bengal, it has been divided into three categories namely severely affected, mildly affected and arsenic safe. The highest affected areas are in the districts of Nadia, North and South 24 Parganas, Murshidabad and Kolkata districts. A combined study was carried out by U.P. Jal Nigam and UNICEF in 249 blocks of 49 districts of Uttar Pradesh. In this study, it was discovered that the arsenic concentration was above permissible limit set as safe by Department of Drinking Water, GoI (0.05 mg L $^{-1}$) in 18 districts. 31 districts have arsenic in the range from 0.01 mg L $^{-1}$ (limit set by WHO) to 0.05 mg L $^{-1}$ (report of the Central Team on arsenic mitigation in rural drinking water sources in Ballia district, Uttar Pradesh State 14–17 September 2011, Ministry of Drinking Water and Sanitation Government of India, New Delhi).

Geological and geochemical investigations have unearthed the fact that the release of arsenic in different spheres of the earth emanates from both anthropogenic activities and natural rock or mineral weathering processes (Bundschuh et al. 2011). The problem gets more aggravated because arsenic once present in groundwater and soil can be bioaccumulated by the terrestrial and aquatic biota which are either consumed by humans or incorporated into fodder for livestock (Meharg and Jardine 2003; Huq and Alam 2005). Thus, arsenic once present in the environment gradually reaches the upper level consumers and gets biomagnified in each trophic level (Ahsan and Del Valls 2011). Simultaneously, it is also a fact that since 30–40 percent net cultivable land is under irrigation, and more than 60 % of this irrigation is catered by groundwater (Huq and Alam 2005), the risk of arsenic-contaminated water being used is high. Places where there is a high concentration of arsenic ($\geq$0.01 mg L $^{-1}$), the transfer of arsenic from soil to water and plant is a serious problem (Rahman et al. 2011a, b). It is a major threat to the sustainable agriculture practices and food security. Especially the accumulation of arsenic in rice is viewed as an emerging disaster for South-East Asia, where rice is a staple food (Meharg 2004). Studies have reported the transfer of arsenic into the edible parts of rice and wheat and some vegetables and fruits when irrigated with arsenic-contaminated groundwater (Meharg 2004; Samal 2005; Bhattacharya et al. 2012; Kar et al. 2013). Though it is now a well-known fact that the use of groundwater for irrigation leads to bioaccumulation and metabolism of arsenic in the agricultural crops, there is very less information about the release of arsenic from sediment to groundwater and the degree of bioaccumulation of arsenic in crops due to irrigation by arsenic-contaminated groundwater. In this paper, we have tried to collect information on the main factors responsible for release of arsenic from sediment to groundwater, their bioavailability and transfer and extent of bioaccumulation in different crops grown in the Bengal Delta plain.

Geological Setting of Ganga-Brahmaputra-Meghna Plain

Since geological condition of an area is mainly responsible for natural release of arsenic, study of the geology of BDP, which is our focus area, is very important. The BDP or the world's largest Ganga-Brahmaputra-Meghna (GBM) plain is a fluvio-deltaic basin been formed by the sedimentation (channel–interchannel and overbank deposits: paleochannels, ox-bow lakes and inland lakes) of the rivers Ganges, Brahmaputra and Meghna with their several tributaries and distributaries (Jha and Bairagya 2011; Uddin and Huda 2011) (Fig. 1). This is a marginal structural foreland basin that developed by the subduction of the Indian plate beneath the Eurasian and Burmese plates (Kuehl et al. 2005). GBM is surrounded by the Precambrian Peninsular shield as well as the mid-Mesozoic Rajmahal basaltic hills in the west (India), Precambrian Shillong Plateau in India (north) and the Mio-Pliocene Tripura and Chittagong Hills in Bangladesh (east and south-east) (Kuehl et al. 2005; Jha and Bairagya 2011). The Bengal Basin is said to be divided into two major parts namely the Pleistocene uplands and Holocene lowlands, where the Pleistocene uplands stand for the Paleo-GBM plains and the Holocene lowlands consist of alluvial fans, flood and delta plain (Uddin and Lundberg 2004; Mukherjee and Fryar 2008). The transportation and deposition of sediment have occured as fine-grained sediments. The sedimentation has been followed by Holocene sea-level change from the Pleistocene Ice age which ultimately formed the marshes and wetlands along with the end mangrove vegetation and peat swamps in the southern part of the plain (Ravenscroft 2003; Kuehl et al. 2005). The depth of the water tables in aquifers of the GBM is generally noticed <15 m below ground level (Ravenscroft et al. 2005). Previous studies have reported that the shallow groundwater of

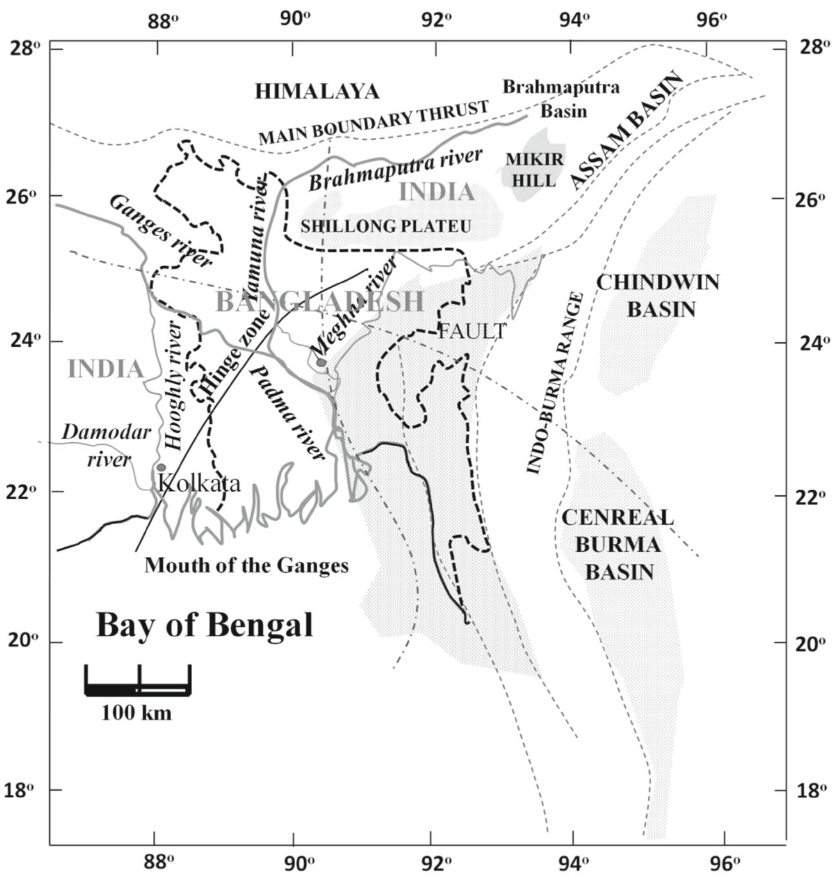

Fig. 1 Geological setting of Ganga-Brahmaputra-Meghna plain: Adjoining areas in the eastern part of the Indian subcontinent, the major tectonic elements surrounding the GBM plain (after Johnson and Alam 1991; Alam et al. 2003)

the aquifer has grey sediments that originated from Holocene age, whereas deeper sediment was yellowish or brownish with less contamination (Ravenscroft et al. 2005; Uddin and Huda 2011).

Occurrence and Distribution of Arsenic in BDP

Arsenic contamination in the Bengal basin is massive in its areal extent and the number of people affected who have been affected by it. Fifty out of sixty-four districts in Bangladesh and nine out of eighteen districts in West Bengal including around 3,500 villages have been detected with arsenic concentration above 0.05 mg L^{-1} (Chakraborti et al. 2002; Rahman et al. 2005). According to Ravenscroft et al. (2005), groundwater in a large area of southern, central and eastern Bangladesh is contaminated with arsenic. He further stated that the arsenic content in Holocene alluvium of rivers Ganga, Brahmaputra and Meghna exceeds 200 times the WHO permissible value for drinking water of 0.01mg L^{-1} of arsenic and roughly 25 % of wells in Bangladesh exceed the national standard (0.05 mg L^{-1}). Department of Public Health Engineering, Government of Bangladesh (DPHE) 1999 has summarised the degree of arsenic contamination in the entire country of Bangladesh and has concluded that 1 % in Barisal, 56 % in Chittagong, 22 % in Dhaka, 31 % in Khulna, 10 % in Rajsahi, 24 % in Sylhet and 25 % in overall Bangladesh had arsenic contamination in the wells greater than 0.05 mg L^{-1}. In West Bengal (India), Murshidabad and Nadia are two out of the nine arsenic-affected districts, where arsenic concentration in groundwater exceeds 0.05 mg L^{-1} (Chowdhury et al. 2000; Chakraborti et al. 2002). 26 percent wells having more than 0.05 and 58 percent wells having more than 0.01 mg L^{-1} of arsenic have been reported by Nickson et al. (2007). It was reported by Mandal et al. (1996) that a single Rural Water Supply Scheme (RWSS) in Maldah district (another arsenic-affected district in West Bengal) was alone capable of withdrawing 147 kg of arsenic from groundwater in a year and about 6.4 tons of arsenic in a year from the shallow large-diameter tubewells in use for agricultural irrigation in Deganga block, North 24-Parganas district (Chakraborti et al. 2002). Arsenic concentrations as high as 1.9 mg L^{-1} have been reported in Bihar, India, (Ghosh et al. 2007) and 2.63 mg L^{-1} in Nepal (Shrestha et al. 2003). Contamination of groundwater above WHO permissible limit (0.01 mg L^{-1}) has been detected in 49 districts of Uttar Pradesh (Report of the Central Team on arsenic mitigation in rural drinking water sources in Ballia district, Uttar Pradesh State 14–17 September 2011, Ministry of Drinking Water and Sanitation Government of India New Delhi).

Arsenic in Water-Soil-Plant System

Arsenic-contaminated irrigation water and fertilizers are the main source of arsenic in agricultural soil (Yuossef and Meharg 2011). The study of As uptake in plants is significant in two ways: On the one hand, it indicates the fraction in the soil which is readily available for the plant species, and in turn provides the information from a toxicological point of view and on the other it also indicates the potential of a certain plant species for phytoremediation and cleanup of arsenic from a contaminated site. Recently, it has become apparent that arsenic-contaminated irrigation water has added significant amount of arsenic in the topsoil and in crop plants, which pose serious threat to sustainable agricultural practices in the Bengal Delta Plain (BDP) (Dittmar et al. 2010; Khan et al. 2010). Long-term use of As-contaminated irrigation water generally results in arsenic accumulation in the soil, which in turn if absorbed by the crops, may enter the food chain and biomagnified in each trophic level, thus posing additional human health risks. In West Bengal most of the crop fields are contaminated with arsenic through irrigated water (Christopher and Haque 2012). The bioavailable fraction of arsenic in the soil only gets transferred to crops. Under oxic soil conditions (Eh > 200 mV; pH 5–8), As is commonly present in the +5 oxidation state. However, As^{II} is the predominant form under reducing conditions (Marin et al. 1993). Inorganic As forms are more toxic than organic, and arsenite (As^{II}) is more toxic than arsenate (As^{V}) (World Health Organization, 1981).Total metal concentration alone is insufficient to assess its environmental impact in contaminated soils (Sadiq 1986). There are some factors that determine the bioavailability, uptake and toxicity of arsenic in plants indicated in the next section.

Factors Regulating the Bioavailability, Uptake and Toxicity of Arsenic in Plants

Soil Moisture and Iron

Soil moisture plays a major role in determining the chemical form and species of arsenic that will be present in soil. Deuel and Swoboda (1972) found that there was an increase of As^{II} in soil solution over time under flooded soil conditions, which they attributed to the release of As during dissolution of iron oxyhydroxide minerals that have a strong affinity for As^{V} under aerobic conditions. In reducing environment interchange of the trivalent and the pentavalent form is also possible, and in such conditions arsenic is mainly taken up by plant roots as arsenite which is more toxic than arsenate. Flooding of paddy soils leads to mobilisation of arsenite into the soil solution and enhanced As bioavailability to rice plants (Xu et al. 2008). Fe plays a pivotal role in the availability of arsenic in the form of Fe oxyhydroxides on soil particulate surfaces or root surface of plants which have high affinity for arsenic (Zhao et al. 2010). Reductive dissolution of Fe oxyhydroxides under reducing condition releases the adsorbed arsenic which leads to enhanced arsenic availability to plants (Geen et al. 2004). As^{V} and As^{II} get adsorbed mainly to iron (hydr)oxides (FeOOH) present in the soil and As^{V} is bound strongest. FeOOH is mainly present in the clay size soil fraction ($<2\,\mu m$) and clayey soils, therefore, generally have a higher arsenic content compared to sandy soils (Mahimairaja et al. 2005).

As Speciation in the Soil

As exists in the environment in various organic and inorganic forms (species). The most important inorganic species are arsenate (As^{V}) and arsenite (As^{II}). Monomethylarsonic acid (MMA) and dimethylarsonic acid (DMA) are the most common organic species in the soil, though present in low quantities compared to inorganic As (Abedin et al. 2002). Depending on the pH and the presence of special substances like Fe oxyhydroxides as mentioned earlier, the interchange in valence states is possible in aqueous solutions. The arsenite concentration in flooding paddy soils for instance is generally higher than those of arsenate found in uncontaminated aerobic soils. Organic As is more readily translocated but the uptake is much lower compared to inorganic As (Carbonell-Barrachina et al. 1998).

Phosphate

Arsenic and Phosphorus are both Group VA elements and thus have similar electron configurations and chemical properties. Both compete for the soil sorption sites and thus the solubility in groundwater gets affected (Smith et al. 2002). Plants find it difficult to distinguish between arsenate and phosphate. Thus, uptake of arsenate and phosphate by plants is very likely to be competitive. The effect of PO_4 additions to aerobic soils on the uptake of As depends on the balance between competition for sorption sites and competition for uptake.

Sulphate

Sulphur helps to detoxify arsenic through complexation of arsenite with thiol-rich peptides. This complexation also helps to restrict translocation of arsenic from roots to shoots. So, maintaining the sufficient S nutrition may be important in arsenic-contaminated environment. Wei et al. (2010) reported that though S had no effect on *Pteris vittata* biomass, it significantly increased its arsenic uptake. The explanation put forward by Marschner (1995) was that addition of S being an essential macronutrient, serves as a precursor for GSH formation in plants. The increased GSH pool apparently renders plants substantially more resistant to different stresses. This is perhaps the main factor that might help plant species to accumulate more arsenic from the soil.

pH

For a given plant species, pH is an important factor determining the arsenic uptake. It influences plant growth and As uptake (Marschner 1995). According to Bech et al. (1997), pH is a major factor in determining arsenic availability in arsenic-contaminated soil. Arsenate exists as an oxyanion at neutral pH and Arsenite is neutrally charged at pH 7.0. It leads to the formation of $Ca_3(AsO_4)_2$, $Mn_3(AsO_4)_2$, $AlAsO_4$ and $FeAsO_4$ (Sadiq et al. 1983). Thus, the solubility of As depends on its oxidation state and solution pH. At alkaline pH, iron hydroxide has greater affinity for the arsenate anion than manganese and, therefore, binds the available arsenic. However, above pH 8.5, as pH increases adsorption of As^{V} decreases, while the opposite occurs for As^{III}. But at pH around 4, the adsorption for As^{V} on FeOOH is maximum, whereas for As^{II} the optimum pH is 7–8.5 (Mahimairaja et al. 2005). In reducing environment, iron or manganese are rendered more soluble and thus lead to release of arsenic ions adsorbed to them. In addition, as arsenate is reduced to arsenite, it binds less strongly to the hydroxide solids, which in turn increases the availability of arsenic for plants (Marquez et al. 2011).

Volatilization

As may be lost from the soil via the formation of volatile arsenic components by soil microbes under anaerobic conditions (Mestrot et al. 2009; Mahimairaja et al. 2005). WHO (2001) estimated this removal to be in the order of 12–35 % per year. The extent to which this process is relevant to flooded paddy fields with their distinct soil conditions is, however, still unknown.

Distance from the Source of Irrigation

Lawgali and Meharg (2011) have found that distance from the source of irrigation also affects the metal concentration in soil and crops, (grain/soil arsenic ratios). Crops irrigated near the source were found to contain higher arsenic than those growing far from the source (Hossain et al. 2008). Even transfer of soil arsenic to rice grain was found to be higher in gold mining regions compared to non-goldmining regions in a study in Southern Ghana (Adomako et al. 2010).

Source of Arsenic in the Bengal Delta Plain

Two principal hypotheses are generally proposed by scientists explaining about the origin of arsenic in groundwater of Bengal Delta Plain (Bagla and Kaiser 1996): According to the first hypothesis, the arsenic is derived from the oxidation of As-rich pyrite in the shallow aquifer as a result of lowering of water table due to overabstraction of ground water for irrigation (Mallick and Rajgopal 1996). The other hypothesis suggests that the arsenic is released by desorption from ferric hydroxide minerals in the aquifer sediments under reducing conditions (Bhattacharya et al. 1997). The sources of arsenic are believed to originate mainly from the parent rocks materials like arsenopyrite or arsenic-rich sea water protruding across large tracts of the Bengal delta which has been entrapped in the sediments by adsorption. Acharya (1999) stated that during the Pliestocene and Holocene time large volume of sediments was laid down in the GBM delta complex by the large rivers that built up the area and aquifer systems. The sediments input and its distribution were largely influenced by tectonic activities of Himalayas and climatic changes in the regions (Acharya 1999). During the quaternary period, formation of peat, peaty clay and carbonate mud within the quaternary sediments is mainly thought to be due to prevalence of back swamp, lagoon and lake in the Bengal delta. Islam and Tooley (1999) have postulated the origin of sedimentary pyrite with peat formation in a lagoon and swampy environments which occured during the putrefaction stage of fallen vegetation. Hydrogen sulphides (H_2S) were released in the decay process enhanced by sulphate reducing bacteria, which attacked the protein part of the organic matter and soluble sulphates thereby releasing arsenic in each instant. Arsenic contamination in groundwater of flood plains and deltaic plains of Holocene sediments of Bangladesh is generally thought to be due to the rapid oxidation, increased solubility and transportation in runoff of arsenic sulphides owing to meteoric precipitation in moist climates (Koyama 1975). The second hypothesis explains the ‘reduction hypothesis’, in which the reduction of As-rich Fe-oxides results from increased oxygen demand possibly related to human disturbances, or buried peat (McArthur et al. 2001). Here, the occurrence of arsenic on extensive FeOOH and lesser MnOOH coatings both in the grains of finer and coarser facie reveals the source of arsenic other than parent minerals. Tanvir et al. (2012) in his study of hydrogeochemistry and arsenic contamination of groundwater of Haor basin, Bangladesh concluded that As is released to groundwater through dissolution of FeOOH and MnOOH under reducing environment, facilitated by microbial degradation of organic matter. Apart from these two hypothesis, Ravenscroft et al. (2001) have reported that the competitive exchange of phosphate from fertilizer over the past fifteen years in

Bangladesh is also one of the reasons of arsenic desorption in groundwater.

Uptake and Translocation of Arsenic in Plants

Arsenic normally has a low mobility with respect to translocation from roots to shoots (Zhao et al. 2009). It is generally believed that As concentrations decrease in the order of roots > stems and leaves > husks > grain in rice (Liu et al. 2006). The pattern of accumulation and translocation of As in plant tissue depends on plant species and habitat. Arsenic is primarily found in the roots rather than in the shoots of plants. For example, more than 15 times higher As concentration in *Spartina pectinata* (Rofkar and Dwyer 2011), and 28–75 times higher As concentration in rice (Rahman et al. 2007) were found in the roots than in the shoots. However, some species, such as radish, have a higher concentration of As in the shoots compared to the roots (Smith et al. 2008).

Arsenic after entering plants through its root system first gets seated in the apoplast and then into the plant cytosol. In case of arsenate, high-affinity phosphate transporters help in the translocation of arsenate into the cell cytoplasm (Meharg and Jardine 2003). Aquaglyceroporins also enable the plants to uptake organic As like MMA and DMA (Rahman et al. 2011a, b). According to Tripathi et al. (2007), first, arsenate from the soil is converted into arsenite inside the root by gluthatione and then complexation occurs between arsenite and phytochelatins. Differences in the As content in husk and bran of Chinese rice samples were investigated by Lombi et al. (2009). Zhao et al. (2009) proposed xylematic transport of As from root to shoot to be the cause for high concentration of As in the husk and through the phloem for accumulation in the grains. In *P. vittata* rapid translocation of arsenic from roots to fronds and arsenic reduction from arsenate (As^{V}) to arsenite (As^{II}) in the fronds are believed to be one of the mechanisms for arsenic detoxification (Zhao et al. 2003). Sequestration of arsenic into the vacuoles, which are away from sites of metabolism has been found to be another strategy of detoxification by certain plants (Wang et al. 2002).

Arsenic Uptake by Specific Crops

The first known arsenic hyper-accumulating plant, *Pteris vittata* L., also known as Chinese brake fern, was discovered by Komar et al. (1998) from an arsenic-contaminated site. Several plants have been identified to considerably accumulate arsenic like *P. Vittata*, *P. cretica*, *Pityrogramma calomelanos* (Francesconi et al. 2002), *Pteris longifolia* and *Pteris umbrosa* (Meharg, 2003; Zhao et al. 2002). In the category of vascular plants, the dicots appear to transfer more arsenic to their aerial parts than monocot plants (Otte 1991). Translocation of arsenic from roots to pods was found to be very little in tomato and bean plants, which preferred to concentrate arsenic more in their roots (Cobb et al. 2000). A similar pattern was observed in Tamarik (*Tamarix pamiflora*) and Eucalyptus (*Eucalyptus camaldulensis*), where the roots accumulated more arsenic compared to the shoot (Tossell et al. 2000). Among the major crops cultivated and consumed in Asia, considerable work has been done on rice. Rice (*Oryza Sativa*) plants have a special mechanism in their roots to exude phytosiderophores in the rhizosphere soil under iron-deficient condition to increase iron bioavailability and uptake (Ishimaru et al. 2006). This indirectly increases arsenic bioavailability and uptake. The rice cultivation conditions also favour arsenic uptake in rice plant (Xu et al. 2008). It was estimated to add 0.05 kg ha-1 arsenic in the requirement of 1,000 mm of irrigation water in paddy field (Meharg and Jardine 2003). Apart from these some studies have identified various vegetables to uptake arsenic from soil. Bhattacharya et al. (2012) worked with rice, wheat (*Triticum aestivum*) and vegetables in arsenic-affected area of Nadia district of West Bengal. He found that wheat plants accumulate comparatively less amount of arsenic (0.129 mg kg^{-1} dry weight) in comparison to rice and most of the vegetables. In wheat, the concentration of arsenic is low and more or less constant in different parts of wheat plant. Sadiq (1986) reported that the growth of maize is significantly correlated with the water-extractable As but not with the total arsenic in calcareous soils. Baig et al. (2010) reported that the translocation of total As in different parts of maize was found in the order of Root > Shoot > Grain. Furthermore, the trend of arsenic accumulation is found to be higher in leafy vegetables and lower in fruity vegetables (Farid et al. 2003). In most types of vegetables, arsenic is present entirely in inorganic form throughout the Bengal region (Halder et al. 2013).

Hyperaccumulation of Arsenic

There are many hyperaccumulators of arsenic reported so far. Many of these are ferns. Most of the hyperaccumulator ferns are of genus *Pteris*. There are also some grasses which act as hyperaccumulator. Apart from these arsenic uptake in selected vegetables, grains and pulses have been studied by various authors around the world. Table 1 shows a non-exhaustive list of hyperaccumulators of arsenic.

Arsenic Affecting Food Stuffs in the BDP (Bengal Delta Plain)

In rural Bengal, it was observed by Halder et al. (2012) that the total daily intake of inorganic As (TDI-iAs) exceeds the provisional tolerable daily intake (PTDI) value of

Table 1 Arsenic accumulation in different edible plant species (crops and vegetables)

Plant species	Degree of accumulation (mg kg^{-1} dry weight)	Reference
Pteris Vittata (Fern)	27,000	Wang et al. (2002)
Agrostis capillaries (Common bent)	100	McCutcheon (2003)
Agrostis tenerrima (Agrostide fluette in French and Vallico in Spanish)	1,000	
Sarcosphaera coronaria (cup fungus)	100–7,000	Stijve et al. (1990); Borovička (2004)
Oryza sativa (Rice)	0.04–0.27	Das et al. (2004)
	0.30	Norra et al. (2005)
	0.16–0.58	Bhattacharya et al. (2009)
	0.334–0.451	Bhattacharya et al. (2012)
	0.362	Roychowdhury et al. (2002)
	0.70	Norra et al. (2005)
	0.872	Samal (2005)
	0.010–0.190	Bhattacharya et al. (2012)
	0.141	Wang et al. (2012)
	0.096	Halder et al. (2012)
Triticum aestivum (Wheat)	0.198	
	0.654	
	0.293	
	0.162	Alam et al. (2002)
Pisum sativum (Pea)	0.279	
Lens culinaris (Lentil)	0.257	
Brassica juncea (Mustard seed)	0.021	
Solanum tuberosum (Potato)	0.126	
Brassica oleracea Botrytis (Cauliflower)	0.344	
Allium cepa (Onion bulb)	0.003	
Solanum melongena (Brinjal)	0.085	
Spinacia oleracea (Spinach)	0.407	
Momordica charantia (Bitter gourd)	0.372	
Allium sativum (Garlic)	0.209	
Raphanus sativus (Radish)	0.258	
Curcuma longa (Turmeric)	0.301	
Capsicum frutescens (Green Chilli)	0.184	
Arum pictum (Arum)	0.091	
Amaranthus sp. (Amaranth)	0.084	
Brassica oleracea capitata (Cabbage)	0.012	
Carica papaya (Papaya)	0.0174	
Abelmoschus esculentus (Lady's finger)	0.015	
Cucurbita pepo (Pumpkin)	0.037	
Phaseolus vulgaris (Beans)	0.012	
Solanum lycopersicum (Tomato)	0.038	
Citrus limon (Lemon)	0.010	
Momordica charantia (Bitter gourd)	0.012	
Brassica oleracea capitata (Cabbage)	0.125	
Daucus carota (Carrot)	0.007	
Raphanus sativus (Chinese radish)	0.008	
Cucumis sativus (Cucumber)	0.017	
Solanum melongena (Egg plant)	0.008	
Vigna unguiculata (Long bean)	0.013	

Table 1 continued

Plant species	Degree of accumulation (mg kg^{-1} dry weight)	Reference
Ipomoea purpurea (Morning glory)	0.138	
Brassica juncea (Mustard green)	0.27	
Brassica rapa (Petsai)	0.14	
Luffa cylindrica (Sponge gourd)	0.22	
Benincasa hispida (Winter melon)	0.15	
Musa acuminate (Banana)	0.16	
Oryza sativa (Rice) fruit	0.30	
Amaranthus *sp.* (Amaranth) leaf	0.08	
Basella alba (Indian spinach)	0.08	
Lagenaria siceraria (Bottle gourd) leaf	0.05	
Moringa oleifera (Drumstick) leaf	0.14	
Colocasia esculenta (Taro) leaf	0.10	
Spinacia oleracea (Spinach)	0.10	
Vicia faba (Broad bean)	0.04	
Abelmoschus esculentus (Ladies finger)	0.05	
Solanum lycopersicum (Tomato)	0.07	
Amaranthus sp. (Amaranth)	0.08	
Lagenaria siceraria (Bottle gourd)	0.08	
Carica papaya (Green papaya)	0.10	
Musa balbisiana (Green Banana)	0.08	
Capsicum frutescens (Green Chilli)	0.10	
Brassica oleracea botrytis (Cauliflower)	0.20	
Benincasa hispida (White gourd)	0.15	
Solanum melongena (Brinjal)	0.27	
Lens culinaris (Lentil)	0.10	
Solanum tuberosum (Potato)	0.3	
Daucus carota (Carrot)	0.2	
Alocasia macrorrhizos (Giant taro)	0.3	
Arum pictum (Arum tuber)	0.2	
Amorphophallus paeoniifolius (Elephant foot)	0.19	
Raphanus sativus (Radish)	0.19	
Lagenaria siceraria (Bottle gourd) leaf	0.09	
Colocasia esculenta (Taro) leaf	0.5	
Lagenaria siceraria (Bottle gourd) leaf	0.09	
Colocasia esculenta (Taro) leaf	0.16	
Moringa oleifera (Drumstick) leaf	0.02	
Solanum melongena (Brinjal)	0.4	
Benincasa hispida (Ash gourd)	0.1	
Trichosanthes cucumerina (Snake gourd)	0.43	
Solanum tuberosum (Potato)	0.09	
Amaranthus sp. (Amaranth)	0.36	
Abelmoschus esculentus (Ladies finger)	0.42	
Carica papaya (Green papaya)		
Musa paradisiacal (Plantain)		
Colocasia esculenta (Taro) lati		
Colocasia esculenta (Eddoe)		
Amorphophallus paeoniifolius (Elephant foot)		
Typhonium trilobatum (Ghotkol)		

0.002 μg day $^{-1}$ kg $^{-1}$ Body weight of the participant, recommended by the WHO. Norra et al. (2005) conducted a study in the intensively cultivated agricultural area of BDP, West Bengal and found the concentration of As was 169–178 mg kg $^{-1}$ in the rice root which was more than 20 times higher than value of 7.7 mg kg $^{-1}$ measured at the uncontaminated reference site. In West Bengal, it was found that the arsenic accumulation in leafy vegetables is more than in the non-leafy and root vegetables (Halder et al. 2012). Similarly, high accumulation of arsenic in leafy vegetables was found in separate studies conducted by Williams et al. (2006) from different parts of Bangladesh and Roychowdhury et al. (2002) from arsenic-affected areas of Murshidabad, West Bengal. However, Roychowdhury et al. (2002) reported that the arsenic concentration in fleshy vegetable is low. He also found that the mean highest concentration of arsenic was in potato skin (292.62 μg g^{-1}) and leaf of vegetables (226.18 μg g^{-1}). The concentration of arsenic was observed as higher in cooked vegetables than in raw (Khan et al. 2010). Alam et al. (2003) conducted a study in heavily As-contaminated village in Samta of Bangladesh and found that the Bioconcentration factors (BCF) values were below 1 for Pb, Cd, Zn, Cu as well as As. In most of the cases, the BCF value decreased with increasing metal concentrations in soil. The calculated BCF values for arsenic in ladies finger (0.001), potato (0.006), ash gourd (0.006), brinjal (0.014), green papaya (0.030), wild aroid leaf (0.034) and snake gourd (0.038) were reported by Alam et al. (2003).

The characteristic health effects that result from ingestion of arsenic-contaminated groundwater or food are slowly manifested and the diagnosis is usually simple (Smith et al. 2000). Rahman (2002) reported skin lesions due to diffuse melanosis, hyperpigmentation and keratosis, to be commonly recognised health effects. In a recent study, Mitra and Bose (2002) found that 82 % of patients had moderate to severe skin lesions, out of which 72 % were young adults. It was also observed that men show more clinical presentation of skin lesions than women under equal exposure level (Mitra and Bose 2002). The ingestion of inorganic arsenic (As^{II} and As^{V}) may induce carcinogenesis and cardiovascular disorders in West Bengal, India (Guha Mazumder et al. 1998). Maity et al. (2012) found that the mean arsenic concentration in urine samples of exposed population was 366 μg L^{-1} in comparison with 172 μg L^{-1} in unexposed population of Moyna village and Ardebok villages of North 24 Parganas district, West Bengal. Some plants may accumulate high levels of arsenic even at soil concentrations near the background level of 5–40 mg As kg $^{-1}$ (Adriano 1986). For instance, (Bhumbla and Keefer 1994) found arsenic concentrations of 6–12 mg As kg^{-1} (fresh weight) in alfalfa and pasture grass grown on soil containing 25–50 mg As kg^{-1}. Tsutsumi (1983) found arsenic concentrations up to 149 mg kg^{-1} in rice straw at an arsenic concentration of 312 mg kg^{-1} in soil. Kar et al. (2013) calculated bioaccumulation factor (BAF) for a number of plants and found BAF value in order of mustard > amaranth > tomato > rice > spinach > maize > sweet potato > basella > peas > onion > egg plant> bean. He also reported that in case of rice, a strong correlation ($R^2 = 0.78$, $p < 0.01$) between the As content of rice roots and available soil As was observed. The average contribution to total arsenic intake from drinking water was 13 %, whereas from cooked rice it was 56 %, thus making it clear that rice contributed most to the daily arsenic intake (Ohno et al. 2007). Rice is a staple crop of Bengal Delta Plain and West Bengal is the leading rice producing state of India. So the high arsenic concentration may be recognised as a disaster for the region. Arsenic concentration was found to be up to 2.0 mg kg^{-1} by Meharg and Rahman (2003) and Islam et al. (2004), much higher than guidelines by WHO permissible limits (1 mg kg^{-1}) in rice. Islam et al. (2012) found that the Boro rice (winter rice) grains contain 1.7 times higher arsenic concentration in comparison to Aman rice (Monsoon rice) grain. His study also suggested that both Boro and Aman rice access the same arsenic soil pools in Bangladesh when grown in same field in different seasons. In another study by Rahman et al. (2011a, b) average As concentrations in uncooked and cooked rice were found to be 153 and 139 g kg^{-1} (dry weight), respectively. Roychowdhury et al. (2002) in his study of Murshidabad, West Bengal, India concluded that arsenic concentration in vegetables, cereals, herbs and spices ranged between <0.0004 and 0.693 mg kg $^{-1}$. Bhattacharya et al. (2012) in a study conducted in Murshidabad district of West Bengal found the accumulation of arsenic to follow the order potato > rice grain (Boro) > arum > amaranth > radish > rice grain (Aman) > lady's finger > cauliflower > brinjal > papaya > spinach > cabbage > mustard seed > pumpkin > onion bulb > pea > wheat > garlic > lentil > beans > green chilli > tomato > bitter gourd > lemon > turmeric. Samal et al. (2011) reported the daily arsenic intake from rice, vegetable and pulses in Nadia district of West Bengal as 126 μg day^{-1}, 26 μg day^{-1} and 1.75 μg day^{-1}, respectively. Santra et al. (2013) focussed on the daily total arsenic uptake in cooked food, with arsenic-contaminated water along with drinking water and its potential health risk. They have reported a total intake of daily 560 μg and 393 μg of arsenic for adult and children, respectively.

Arsenic Exposure and Health Impacts

The exposure of human beings to arsenic mainly results from drinking water and food. So it is essential to estimate the average consumption of water and food per day to get an idea about the cumulative exposure of arsenic per person per day. Hels et al. (2003) have reported the average food consumption of Falshatia (Manikganj) and Jorbaria (Mymensingh) of

Bangladesh to be 450 g rice/day, 200 g vegetables/day, 60 g roots/tubers/day, 40 g fish/day and 30g meat/day. In West Bengal similarly, Roychowdhury et al. (2002) reported the daily As intake of 180 μg day^{-1} for adults 97 μg day^{-1} for children (10 years old) through foodstuff and 400 and 200 μg day^{-1} via drinking water. Watanabe et al. (2004) reported the daily intake of As by adults to be approximately 600 μg day^{-1}. Some of the effects of arsenic poisoning on human health reported are melanosis, keratosis, hyperkeratosis, oedema, gangrene and skin cancer (Maity et al. 2012). Melanosis and keratosis are the most common forms among the affected people (Maity et al. 2012). Chronic exposure to inorganic arsenic affects several disorders upon different biological systems like on gastrointestinal system, respiratory system, neurological system, cardiovascular system, reproductive system, hematopoietic system, hormonal system, renal system, etc. which ultimately leads to cancer (Maharjan et al. 2005). Even symptoms like nausea, anorexia, diarrhoea and abdominal pain in cases of chronic arsenic toxicity have been reported (Aleksunes and Manautou 2007). Significant peripheral vascular disorders have also been reported due to arsenic exposure (Srivastava et al. 2009). Peripheral neuritis was found in 74(47.4 %) out of 156 patients due to drinking of arsenic- contaminated water (0.05–14.2 mg L^{-1}) (Guha Mazumder et al. 1998). Haematological abnormalities and anaemic conditions have been reported in acute and chronic arsenic poisoning (Guha Mazumder 2008). About one third of the biological samples (urine, hair, nail, skin-scales) (n = 27,000), when analysed contained arsenic above its normal level value and about 9.8 % of the population (n = 86,000) have been identified as designated patients with clinical disorders (Roychowdhury et al. 2002). Estimates indicated around one lakh cases of skin lesions caused by arsenic in Bangladesh (Smith et al. 2000). Significant correlation was found between total arsenic intake through food and drinking water and the concentration of arsenic in hair (Islam et al. 2012). Long-term exposure to arsenic can cause serious health problems including internal cancers of the skin, lungs, bladder and kidney, which can pose a threat to life. These cancers can occur without skin lesions. Most of the deaths caused by arsenic are expected to be from lung cancer. In a study conducted by Mitra and Bose (2002), he reported that 80 % of the studied population had hyperkeratosis with or without nodular skin lesions. He also examined the Biopsy specimens from One hundred and twenty three (82 %) patients and found that they had moderate or severe skin lesions. Sites of appearance of skin lesions, in order of frequency, were trunk, including chest (38 %), hands only (18 %), both hands and feet (15 %), feet only (13 %) and chest only (11 %). Common symptoms were weakness, chronic cough, chest pain, abdominal pain, joint pain, itching, loss of appetite, insomnia, shortness of breath and frequent urination with burning. In another study conducted on 240 married women residing in four As-affected villages in North 24 Parganas district of West Bengal showed occurrences of stillbirth and miscarriage in pregnant women were noticed (Sen and Chaudhuri 2008).

Chemical and Microbial Methods for Arsenic Removal from Water and Soil

The methods to treat arsenic mainly rely on few chemical reactions. Atmospheric oxidation or chemical oxidation that alters the chemical form followed by precipitation is one of the known methods of treating arsenic contamination in water (Bissen and Frimmel 2003; Zaw and Emett 2002; Dutta et al. 2005). Many treatment technologies depend on oxidation as a first step of treatment. Chemicals used for the process include mainly hypochlorite, permanganate gaseous chlorine and ozone. After this step arsenic can effectively be removed by adsorption and co-precipitation. Coagulation is generally regarded a good option for arsenic removal (Wickramasinghe et al. 2004; Hering et al. 1997). The commonly used coagulants are alum or ferric chloride (Hering et al. 1996) and ferric sulphate. This step is simple in operation and effective over a wider range of pH and the chemicals are widely available. Since in many areas of BDP, arsenic is often found with high levels of Fe and Mn, removal of these elements by flocculation, sedimentation and filtration can significantly remove arsenic. Adsorption and ion exchange are yet another method of arsenic removal (Yadava et al. 1988; Yan et al. 2000). The commonly used media are activated alumina, activated carbon, iron and manganese-coated sand, activated carbon, kaolinite clay and hydrated ferric oxide. Ion exchange, another form of adsorption is also a potential technology for arsenic removal. It involves the reversible displacement of an ion adsorbed onto a solid surface by a dissolved ion. Various anion exchange resins (sulphate-selective and nitrate-selective resins) capable of removing arsenic (arsenate) concentration below 1 mg L^{-1} are commercially available. Several researchers have found that copper-doped materials have strong, specific affinity for arsenate (e.g. Rajakovic and Mitrovicm 1992; Ramana and Sengupta 1992). Apart from this mesoporous silica sorbent which makes use of Cu(II)-based functional groups has been suggested by Fryxell (1999) which has a higher ion exchange capacity (75 mg As g^{-1}) than conventional resins. Other than this, Membrane filtration, including, reverse osmosis electrodialysis and nanofiltration have also been proved to be capable of removing arsenic (Velixarov et al. 2004; Wickramasinghe et al. 2004).

Microbial treatment of arsenic-contaminated water and soil has also gained importance in the last few decades. Microbial processes mediate the cycling of metals in the environment by redox reactions, differing solubility using different complexation reactions, changing pH and adsorption or uptake of a substance from the environment (Smith et al. 1994). Oxidation of arsenite to arsenate by *Bacillus arsenoxydans, Pseudomonas*, *Xanthomonas* and *Achromobacter* has been identified by researchers (Hambsch et al. 1995; Mokashi and Paknikar 2002; Battaglia-Brunet et al. 2002) as a detoxification mechanism. Biomethylation of arsenic is another strategy by microbes which help in arsenic detoxification. Biomethylation of arsenic can form volatile methylated compounds like mono-, di- and trimethyl arsine, respectively, and nonvolatile methylated compounds, e.g. methyl arsonate and dimethyl arsinate (Bentley and Chasteen 2002). *Candida humiculus* methylates the widely known wood preservative, chromated copper arsenate (CCA) to trimethyl arsine (Cullen et al. 1984). Several other bacterial strains are also reported by researchers which help in oxidising or detoxifying arsenic from water or soil (Gihring and Banfield 2001; Maity et al. 2011). Several studies are continuing on arsenic removal and detoxification by microbes.

Conclusion

Bengal Delta Plain (BDP) supporting a huge population of India and Bangladesh and dependent on agriculture for their food as well as livelihood has been severely affected by arsenic contamination in its groundwater and sediment. The sustainable development of agriculture and food security has been seriously jeopardised in several areas of these two countries due to arsenic contamination due to several natural and anthropogenic phenomenons. Bioavailable arsenic has reached significant levels due to several factors related to soil and plant species. Uptake and translocation of arsenic in the edible crops and vegetables have been noticed to be markedly different. Huge number of arsenic-related diseases including cancer, teratogenic effects and deaths has been reported from both the countries in the last few decades. Situations arising from arsenic toxicity via food chain are expected to worsen further if proper corrective actions are not taken immediately. Integrated knowledge and management about arsenic levels in irrigation water, soil and crops are required to identify the behaviour of arsenic in soil, its uptake and accumulation in agro ecosystems. This information would help in the evaluation of the current situation and would serve as a baseline for upcoming days.

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