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Review Article

An Overview of Arsenic Dynamics in Lowland Rice Ecosystem

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ABSTRACT

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. Amongst the main contributing sources of As contamination of soil and water are geologic origin, pyritic mining, agriculture and coal burning. Soils contain both organic and inorganic arsenic species. Inorganic As species are more toxic to living organisms than organic forms. The majority of As in aerated soils exists as $H_2AsO_4^-$ *(acid soils) or* $HAsO_4^{2-}$ *(neutral and basic). However,* H_3AsO_3 *is the predominant species in anaerobic soils, where arsenic availability is higher and As (III) is more weakly retained in the soil matrix than As(V). The availability of As in soils is usually driven by multiple factors and processes such as the presence of Fe-oxides and/or phosphorus, (co)precipitation in salts, pH, organic matter, clay content, rainfall amount, etc. The available and most labile As fraction can potentially be taken up by plant roots, although the concentration of this fraction is usually low. The status of current scientific knowledge allows us to manage as contamination in the soil-plant system and to mitigate arsenic's effects. Hence it is imperative to understand the mechanisms of As uptake and translocation by rice and the present paper focuses on the journey of As from soil to human through the rice grains.*

Keywords: *Arsenic, Dynamics, Lowland, Management, Rice ecosystem*

INTRODUCTION

Arsenic (As) is a ubiquitous element that ranks 20th among the most abundant elements in the earth's crust. 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). 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 & Whitaker, 2002). Inorganic arsenic is present in soil, water, air, and food such that humans are constantly exposed to this contaminant (Mandal & Suzuki, 2002).

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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 and sometimes genotoxic effects (Smith et al., 1992; Martinez et al., 2011). On a global level, drinking contaminated water is the major source of exposure to this contaminant (Smedley & Kinninburgh, 2002). It is estimated that some 30 million people are exposed to waters contaminated with arsenic, a quarter of them showing symptoms associated with long-term exposure in at least five South Asian countries: Bangladesh, India, Nepal, Thailand, and Myanmar. According to the World Health Organization (WHO) guideline, the baseline limit of As concentration in safe drinking water is 10 microgram/litre (µg/L), which is similar to the European Union (EU), US Environmental Protection Agency (USEPA), Netherlands, and Germany limits of As in drinking water (Ahsan & Del Valls, 2011). However, in Bangladesh and many other developing countries, such as India, China, and Nepal, it is set to 50 µg/L (World Bank, 2005).

Arsenic poisoning via drinking of As contaminated groundwater is well documented (Smedley & 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. 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). Further, rice is one of the most efficient silica accumulators among all crop plants and As (III) too enters through silicic acid transporters in rice (Ma et al., 2008; Norton et al., 2010). However, accumulation of As in rice grains in quantities greater than recommended safe limits is influenced by various environmental, geochemical, and biological factors which controls As solubility, bioavailability and uptake in the soil–rhizosphere – plant system (Walter & Wenzel, 2002; Zhao et al. 2010).The situation is of grave concern considering the very high per capita rice consumption rate in As contaminated South East Asian countries ranging from 250 to 650 g of rice per day (Arslan et al., 2017). Hence it is imperative to understand the mechanisms of As uptake and translocation by rice (Li et al., 2011). The present paper focuses on the journey of As from soil to human through the rice grains.

GEOCHEMISTRY OF ARSENIC

Arsenic is a redox-sensitive element, which exists mainly in four oxidation states, + V (arsenate), + III (arsenite), 0 (arsenic), and – III (arsine) and occurs as cation as well as oxy-anion 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).

MECHANISMS OF RELEASE OF ARSENIC INTO GROUND WATER

The primary sources of As are thought to be eroding coal seam and rocks containing sulfide minerals within the Himalayas whose weathering and transport leads to downstream deposition of As in Gangetic plains (Acharya et al., 1999). The minerals contained within these deposits are oxidized when exposed to the atmosphere, and much of their As content is transferred to secondary phases including iron (Fe) hydroxides, oxyhydroxides and oxides, collectively referred to as Fe oxides. As is released from Fe oxides into ground water through microbial processes (Fendorf et al., 2010). Hence As has been found to be distributed in wide spread areas of Gangetic plains spread across Uttar Pradesh, Bihar and West Bengal. Large areas of paddy soils are contaminated by As due to irrigation with As-tainted 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:

1. Mobilization of arsenic due to the oxidation of As-bearing pyrite minerals

Insoluble As-bearing minerals such as arsenopyrite (As-FeS) are rapidly oxidized when in contact with oxygen, releasing soluble As (III) and Fe (II) (Smedley & Kinniburgh, 2002) (Fig. 1).The oxidation of the As-bearing minerals is dependent on availability of oxygen. High oxygen in pyritiferrous system suffers by the excess pumping and water-table drawdown. 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.

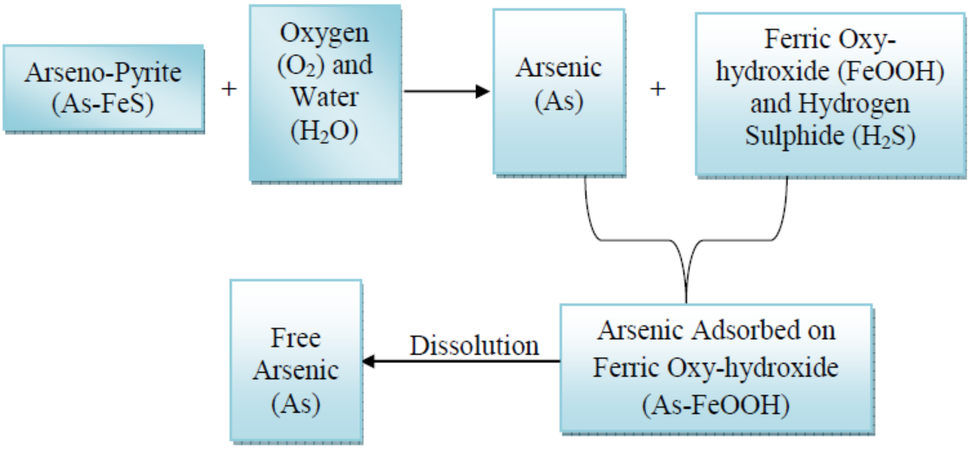

Fig. 1: Mobility of As due to oxidation of As-bearing pyrite minerals

2. Dissolution of As-rich iron oxyhydroxides (FeOOH) due to onset of reducing conditions in the subsurface:

During reductive dissolution mechanism, As gets adsorbed onto Fe (III)-oxyhydroxides and is released under reduction condition (Islam et al., 2004; Nickson et al., 1998) (Fig. 2). Moreover, dissolved oxygen in groundwater is consumed during microbial oxidation, inducing an increase in HCO_3 concentration (Liaoa et al., 2011; Kar et al., 2010). This microbial activity causes a reducing, alkaline environment where mobility and thereby the concentration of As increases.

Arsenic in Fe-oxyhydroxides is mobilized when the environment becomes anaerobic (Smedley & Kinniburgh, 2002). Bhattacharya et al. (1997) first proposed the reductive dissolution of Fe oxyhydroxides process for arsenic release. 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. 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 (hydro) oxides.

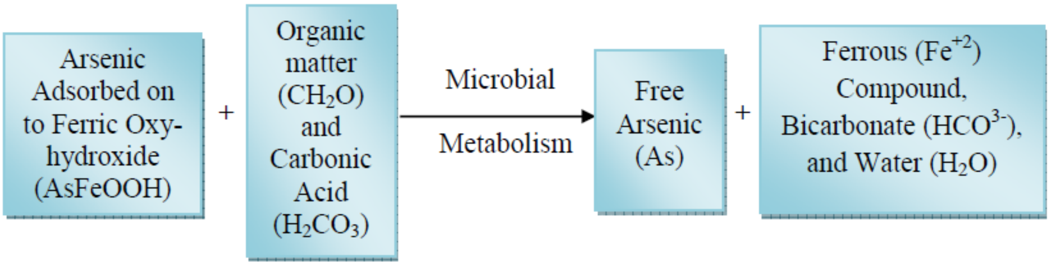

Fig. 2: Mobility of As due to dissolution of As-rich iron oxyhydroxides

3. Desorption of Fe-Oxydroxides

Besides anaerobic conditions for releasing As from Fe oxyhydroxides, there is also effect of pH, particularly at 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). The As concentration in groundwater (µg/L) in relation with major release mechanism reported from different countries are presented in Table 1.

Table 1: As concentration in groundwater (µg/L) in relation with major release mechanism reported from different countries

Country	As in ground water (µg/L)	Major release mechanism	Reference
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)
India	0.77 – 1059	Reductive dissolution of Fe or Mn (hydro)oxides	Bhattacharyya et al., (2009)
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	Naguyen and Itoi (2009)

FACTORS RESPONSIBLE FOR AVAILABILITY OF ARSENIC IN SOIL AND PLANT

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. The solubility and bioavailability of As in soil is influenced by various factors 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. As(III) is more toxic than As(V), and also much more toxic than MMA or DMA. 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. It was reported that the As availability to rice plants followed the order As(III) > MMA > As(V) > 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 controls arsenic mobility and speciation (Fitz & 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. 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. This is due to the reductive dissolution of Fe-oxyhydroxides, 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. 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).

pH

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 ($pH < 3$), arsenic mobility can increase due to the dissolution of arsenic binding species, such as Fe and Al oxycompounds. 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, and increases labile As in the rhizosphere, leading to more As accumulation in plants (Fitz & Wenzel, 2002).

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 (Mukhopadhyay & Sanyal, 2004). 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, the negative correlation between As concentration in rice grain and soil OM was found by Fu et al. (2011).

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. 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. Furthermore, dissolved organic matter can promote As release by changing the redox chemistry of site surfaces and As species. Selim Reza et al. (2010) reported that As is released by the reductive dissolution of Fe-Ox hydroxides linked to organic matter.

Soil Texture

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, and soils with clayey texture are supposed to be less toxic of arsenic to plants compared to sandy soils (Heikens et al., 2007).

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. However, their sorption property is strongly dependent on the environmental conditions (Mukherjee et al., 2009). Under flooding conditions, oxyhydroxides release As from soil by the reductive dissolution of Fe oxydroxides, 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-amorous fraction and rice. This indicates that As content in amorous/poorly crystalline Fe-oxides in soil can be an important factor for rice As.

Phosphate

Phosphate (PO_4) also controls As phytoavailability in agricultural fields. Generally, phosphate is a chemical analogue of As(V) (Fitz & Wenzel, 2002); thus, it competes with As(V) to occupy the same sorption sites in soils, leading to increase As solubility. Many studies reported that addition of PO_4 increases As mobility in soil, which can subsequently increase As uptake by plants. Contrary to this, PO_4 also competes with As(V) at the same transport during uptake, and can reduce arsenate accumulation in plants (Fitz & Wenzel, 2002).

ARSENIC IN SOIL-WATER-PLANT CONTINUUM

Arsenic dynamics under flooded rice culture is quite complex. A number of chemical and physical processes impact overall As dynamics and net As balance in flooded rice culture, e.g., irrigation protocols, adsorption/desorption, dissolution/precipitation, redox processes, As volatilization, vertical flow, lateral redistribution of soil and As resulting from surface flooding, repartitioning into surface water, and bypass flow as impacted by wetting/drying cycles.

Rice is known to be a “natural arsenic accumulator”. Rice plants take up large amounts of silicon from the soil to strengthen the stems and husks, which protect the plant against pests. Arsenic and silicon are chemically similar in soil conditions found in flooded rice paddies. Thus, arsenic is literally integrated into a rice plant as it grows. Arsenic is concentrated in the outer layers, or husks, of the grain. So, higher levels of arsenic are found in brown rice as compared white rice. Inorganic arsenic species are retained in soils by adsorption on mineral oxide surfaces, with Fe-oxides generally considered to be the major sink for As in paddy rice soils when they are oxidized. Under the reducing conditions of the paddy, Fe-oxides dissolve and inorganic As is released into the soil-water matrix from which it can be assimilated by the growing rice plant. Uptake of As by rice is complicated by various chemical and physiological processes that occur in the rice paddy, namely

- a change in oxidation state of As from arsenate (As-V) to arsenite (As-III) as reduction in the paddy intensifies
- the formation of oxidized Fe plaque on rice root surfaces that readsorbs As, but in competition with phosphate
- the possible formation of insoluble As sulfur species
- competitive uptake of phosphate and arsenate through the same ion channel
- competitive uptake of arsenite and silicate through a general aquaporin channel and
- microbial methylation of inorganic As to mono- and dimethyl-As species that have different rates of uptake than those of inorganic As species.

With the exception of hyper accumulators such as certain ferns, the translocation of inorganic As from the roots to the above ground parts is limited. Organic As is more readily translocated but the uptake is much lower compared to inorganic As. In pot experiments with rice plants exposed to As added via As-V in irrigation water, plant parts were ranked according to the As concentrations as follows: root > straw > husk > grain. Concentrations in all plant parts increased with the exposure concentration (Abedin et al., 2002).

MITIGATION OPTIONS ON ARSENIC ENTRY IN FOOD CHAIN

The following practices have shown great potential in mitigating the arsenic contamination hazards.

1) Using surface water sources
2) Exploring and harnessing alternative arsenic free aquifer
3) Removal of arsenic from ground water using arsenic treatment plants/filters
4) Rain water harvesting
5) Agronomic management like use of balance fertilizer, irrigation management, use of organic manure.

CONCLUSION

Arsenic is a natural trace element found in the environment. In some cases and places, human activities have increased the soil concentration of As to levels that exceed hazard thresholds. Amongst the main contributing sources of As contamination of soil and water are geologic origin, pyritic mining, agriculture, and coal burning. Soils contain both organic and inorganic arsenic species. Inorganic As species include arsenite and arsenate, which are the most abundant forms found in the environment. The majority of As in aerated soils exists as $H_2AsO_4^-$ (acid soils) or $HAsO_4^{2-}$ (neutral and basic). However, H_3AsO_3 is the predominant species in anaerobic soils, where arsenic availability is higher and As(III) is more weakly retained in the soil matrix than As(V). The availability of As in soils is usually driven by multiple factors. Among these factors is the presence of Fe-oxides and/or phosphorus, (co)precipitation in salts, pH, organic matter, clay content, rainfall amount, etc. The available and most labile As fraction can potentially be taken up by plant roots, although the concentration of this fraction is usually low.

FUTURE LINE OF WORKS

The status of current scientific knowledge allows us to manage As contamination in the soil-plant system and to mitigate arsenic's effects. Phytoremediation is an emerging technology suitable for reclaiming As-contaminated soils and waters. Phytoextraction has been used to clean As-contaminated soils, although its applicability has not yet reached maturity. Phytostabilization has been employed to reduce environmental risk by confining As as an inert form in soils and has shown success in both laboratory experiments and in field trials. Phytofiltration has been used to treat As-enriched waters. Such treatment removes As when it is accumulated in plants grown in or on water. In agricultural food production, appropriate soil management and plant variety/species selection can minimize As-associated human diseases and the transfer of As within the food chain. Selecting suitable plants for use on As-contaminated soils may also enhance alternative land use, such as for energy or raw material production.

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