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Arsenic biogeochemical cycling in paddy soil-rice system: Interaction with various factors, amendments and mineral nutrients

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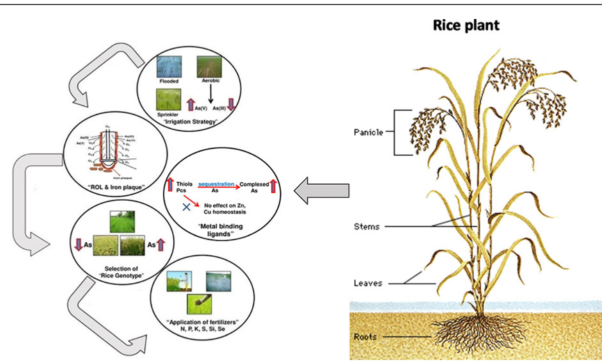
HIGHLIGHTS

- This review describes occurrence and geochemistry of arsenic (As) in paddy soil-rice system.
- Impact of biotic and abiotic factors on As (im)mobilization in paddy soil is elucidated.
- As interaction with organic/inorganic amendments, and mineral nutrients is discussed.
- Updated and key scientific information for limiting As uptake by rice is presented.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As) contamination is a well-recognized environmental and health issue, threatening over 200 million people worldwide with the prime cases in South and Southeast Asian and Latin American countries. Rice is mostly cultivated under flooded paddy soil conditions, where As speciation and accumulation by rice plants is controlled by various geo-environmental (biotic and abiotic) factors. In contrast to other food crops, As uptake in rice has been found to be substantially higher due to the prevalence of highly mobile and toxic As species, arsenite (As(III)), under paddy soil conditions. In this review, we discussed the biogeochemical cycling of As in paddy soil-rice system, described the influence of critical factors such as pH, iron oxides, organic matter, microbial species, and pathways affecting As transformation and accumulation by rice. Moreover, we elucidated As interaction with organic and inorganic amendments and mineral nutrients. The review also elaborates on As (im) mobilization processes and As uptake by rice under the influence of different mineral nutrients and amendments in paddy soil conditions, as well as their role in mitigating As transfer to rice grain. This review article provides

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critical information on As contamination in paddy soil-rice system, which is important to develop suitable strategies and mitigation programs for limiting As exposure via rice crop, and meet the UN's key Sustainable Development Goals (SDGs: 2 (zero hunger), 3 (good health and well-being), 12 (responsible consumption and production), and 13 (climate action)).

Contents

1. Introduction

Arsenic (As) contamination of terrestrial and aquatic systems represents a persistent global problem, especially in South and Southeast Asian and Latin American countries (Hussain et al., 2019b; Naidu et al., 2006; Thounaojam and Khan, 2020). Approximately 200 million people worldwide have been reported to be at risk of As poisoning, either directly from drinking As-contaminated groundwater or indirectly via ingestion of As-laced food crops, mainly rice (*Oryza sativa* L.) that is irrigated with As-contaminated groundwater (Bhattacharya and Bundschuh, 2015; Roy Chowdhury et al., 2020). Arsenic mobility and phytoavailability to rice increases under submerged paddy soil conditions due to transformation of arsenate (As(V)) to highly toxic arsenite (AsIII)) (Abedin et al., 2002a; Carrijo et al., 2019; Kumarathilaka et al., 2018; Meharg, 2004). In terrestrial ecosystems, both natural (weathering of rocks, hot springs) and anthropogenic (e.g., mining, smelting, coal burning, use of As-containing pesticides and herbicides in agriculture and irrigation with As-contaminated water) processes have contributed to As contamination of agricultural paddy soils (Ahmad et al., 2020b; Hussain et al., 2019b; Shahid et al., 2018; Shakoor et al., 2018; Upadhyay et al., 2019). The World Health Organization (WHO) has set a safe limit of As at 10 μg L^{-1} for drinking water and at 100 μg L^{-1} for irrigation water (WHO, 2011).

Rice is consumed as one of the major staple foods by around 3 billion people, mainly in Asian countries, to provide their energy (70%) and protein (50%) requirements. The Food and Agriculture Organization (FAO) has recommended a safe limit of 0.2 mg kg^{-1} dry weight (DW) in rice grain that was decreased from FAO's previous safe limit of 1.0 mg kg^{-1} DW (Codex Alimentarius Commission et al., 2016). Laborte et al. (2017) reported that about 115 countries in the world cultivate and produce rice, and around half of the regions in Asia have 2 to 3 rice crop planting seasons. In temperate climate, production of rice is limited to once cropping each year and the main rice growing season varies from country to country (Laborte et al., 2017). Rice plants are cultivated under flooded soil conditions in many countries, especially in Bangladesh, India, Pakistan and China, and this promotes the redox transformation of comparatively less toxic As(V) to more toxic, mobile and phytoavailable As(III) species (Abedin et al., 2002a; Nafees et al., 2011; Seddique et al., 2008; Suriyagoda et al., 2018).

The biogeochemical behavior of As in paddy soil-rice system makes it easily available for plant uptake and subsequent accumulation in rice grain (Halder et al., 2013; Halder et al., 2012; Herath et al., 2016; Hussain et al., 2019b; Shahid et al., 2018). Arsenic speciation and plant availability in the paddy soil environment is controlled by different factors such as redox potential (E_h), pH, organic matter content, phosphate (PO_4), sulfate (SO_4), iron (Fe) oxides, manganese (Mn), and microbial species (Abbas et al., 2018; Abedin et al., 2002b; Niazi et al., 2017; Upadhyay et al., 2019). Given the dynamic biogeochemistry of As in paddy soils, scientists have directed efforts to understand As uptake by rice plants in paddy soils (Bhattacharya et al., 2013). Under submerged soil conditions, microbial consumption of O_2 leads to the anoxic conditions which leads to As release by reductive dissolution of Fe (*oxy*)hydroxides (Herath et al., 2016). Overall, inorganic forms of As are taken up by rice root considerably in higher amount than that of organic As compounds (Bhowmick et al., 2018; Dawe et al., 2003; Islam et al., 2012; Islam et al., 2017). Total As content in uncontaminated soil usually ranges from 0.1 to 10 mg kg^{-1} (Zhao et al., 2010). The European Union (EU) recommends that the total As content of soils for agricultural purpose should be < 20 mg kg^{-1} (Kumarathilaka et al., 2020b). The presence of high As concentration in soil depends not only on soil As concentration, but also on various geo-environmental (biotic and abiotic) factors as mentioned above. The climatic and geomorphic features of a region such as precipitation, surface runoff, infiltration rate, and groundwater level; and their fluctuations also affect the migration and redistribution of As, although these factors are beyond the discussion of this review.

In recent years, there has been a great interest to examine the effect of different organic and inorganic amendments (e.g., biochar, compost, phosphate, farmyard manure) on As uptake by rice plants (Norton et al., 2017; Wan et al., 2019; Zia et al., 2017). Some recent research has also focused on As immobilization using different strategies (Table 1), such as biochar (Bakshi et al., 2018), water management practices, and foliar application of different elements (Norton et al., 2017; Wan et al., 2019; Zia et al., 2017). In an earlier review, Suriyagoda et al. (2018) discussed various physiological, biochemical and mechanistic aspects of As uptake and translocation in rice grains growing under paddy soil conditions. Recently, Kumarathilaka et al. (2020b) provided information on various agronomic/biological methods, water management practices and microbial inoculation to possibly decrease soil-plant transfer of As in rice crop. In another recent review, Kumarathilaka et al. (2018) elaborated, particularly As speciation dynamics, in paddy soil-rice environments, and Mitra et al. (2017) described mitigation approaches for reducing As threat in rice crop in comparison to other food crops. Previously, Bakhat et al. (2017) demonstrated the physiological and molecular aspects of As toxicity and accumulation in rice plants, and discussed some As mitigation strategies (e.g., water management, bioremediation) with a focus on the role of biochemical and molecular pathways.

This review provides a brief overview of the biogeochemical cycling of As in paddy soils and information advancing knowledge on the importance of As interaction with various biotic and abiotic components in paddy soil. In addition, we discuss the use of different organic and inorganic amendments and mineral nutrients that could be used to reduce As uptake by rice. Key factors, including microbial processes, and mechanisms related to As speciation and volatilization in paddy soil-rice environments are also discussed to give a better understanding on As mitigation options in rice.

2. Sources, geochemistry and distribution of arsenic – A brief overview

Arsenic is a toxic metalloid that is widely present in the Earth's crust. Important minerals of As in the environment are arsenical pyrite (FeAsS), realgar (AsS) and orpiment (As_2S_3) (Khosravi et al., 2019; Natasha et al., 2021; Smedley and Kinniburgh, 2001). In addition to natural sources, anthropogenic activities that include agricultural use of As-based pesticides and herbicides, wood preservatives, mining and smelting, and coal combustion processes (Khosravi et al., 2019) release As into the environment. Geogenic As sources, particularly weathering of the parent material and alluvial sediments, have contributed in contamination of agricultural soils with As. For example, the weathering of As-containing minerals such as claudetite (As_2O_3) and bearsite (Be$_2$

Table 1
The interaction of different amendments on (im)mobilization of arsenic (As) in paddy soils.

Soil As contamination	Soil As (mg kg^{-1})	Soil amendment type	Arsenic removal efficiency (%)	References
Natural paddy soil	105	Titanium gypsum	38%	(Zhai et al., 2020a)
Natural paddy soil	47	Calcium-based magnetic biochaSr (Ca-MBC)	Increase As concentration by 40.6% (translocated from soil)	(Wu et al., 2020)
Natural paddy soil	33	Wet–dry cycle, Organic matter, gypsum, and hematite	Gypsum, and hematite reduced arsenic mobility in the soil by 8–60%, Organic matter increased arsenic mobility by 70–130% (translocated from soil)	(Yuan et al., 2020)
Natural paddy soil	138	Fe-Mn-Ce oxide–modified biochar composites (FMCBCs)	69.36% (roots) 51.42% (Stem) 47.27% (Leaves) 73.58% (grains)	(Lian et al., 2020)
Natural paddy soil	105	Combined amendment (CF) was prepared by mixing calcium sulfate ($CaSO_4 \cdot 2H_2O$) and ferric oxide (Fe_2O_3) at a ratio of 9:1. $CaSO_4$ and Fe_2O_3	55.5% and 69.3%	(Zhai et al., 2020b)
Natural paddy soil	1.76	Zinc oxide nanoparticles (ZnONPs) and zinc ions (Zn^{2+})	Reduced by 39.5% and − 83.3%	(Ma et al., 2020a)
Natural paddy soil	288	Goethite-modified biochar (GB)	32.2% - 46.6%	(Irshad et al., 2020)
Natural paddy soil	21	Fe-based biochar	18%	(Tang et al., 2020b)
Natural paddy soil	6.69	Hybrid rice varieties and conventional rice varieties	31% - 38%	(Cao et al., 2020)
Natural paddy soil	Not given	Bismuth-impregnated biochar and wheat straw biochar (WBC)	Reduced As mobility with Bismuth-impregnated biochar and wheat straw biochar results in the increase in the As mobility and bioavailability	(Zhu et al., 2019)
Natural paddy soil	132	woody peat and $Fe(NO_3)_3$	significantly decreased As and Cd in pore water and enhance the immobile phase of As and Cd	(Wang et al., 2019b)
Natural paddy soil	249	Biochar and zero-valent iron (ZVI)	47%	(Qiao et al., 2019)
Natural paddy soil	23	Soil microbial fuel cells (sMFC)	It enhances the As concentration	(Gustave et al., 2019)
Natural paddy soil	101	Alternate wetting and drying/Water management	84% and 81%	(Wang et al., 2019a)
Artificially spiked soil	10, 25, 50 and 100	Iron (Fe), phosphate (PO_4) and farmyard manure (FYM) and Rice genotypes	24% and 14%	(Irem et al., 2019)
Natural paddy soil	96	Organic fertilizer	Bioavailable As content increased by 3.2 times at −400 mV	(Shen et al., 2020)
Natural paddy soil	132	Poorly crystalline Fe oxides (PC-Fe), $FeCl_2$ + $NaNO_3$ and $FeCl_2$	54%, 52% and 46%	(Yu et al., 2017a)
Natural paddy soil	15 and 76.8	Biochar and oyster shell waste	62.3%	(Chen et al., 2018)
Natural paddy soil	15.7	Irrigation and rice genotype	17% to 35%	(Islam et al., 2017)
Natural paddy soil	1540	Gypsum and gypsum+lime	48–64%	(Kim et al., 2018)

$(AsO_4)(OH).4H_2O$) has been associated with the elevated As concentration in rice field (50 to 90 mg kg^{-1}) of Manipur, India (Chandrashekhar et al., 2016). The degradation of As minerals (e.g., FeAsS) and the associated secondary As-bearing Fe oxide minerals are the major causes of As contamination of rice in paddy soils globally, and particularly in South and Southeast Asia. Arsenic concentration in different soil sediments around the world with varying sampling depths is given in Table S1, Supplementary Information.

Arsenic is found in various natural reservoirs such as oceans, rocks, atmosphere, biota and soil, but > 99% of As is present in rocks and minerals (Francesconi and Kuehnelt, 2001). In sandy sediments, As concentration is usually found below 10 mg kg^{-1} and in peat and clay-rich sediments As levels may reach up to 100 mg kg^{-1} (Hussain et al., 2019b). For example, the source of As in India (West Bengal), China, Bangladesh, Taiwan, Vietnam, Pakistan and Nepal is attributed to the oxidation and reduction of As-rich Fe oxide minerals, deposited from Himalayas in the Bangal Delta, Ganges-Brahmaputra and Indus-Basin (Bhattacharya et al., 2007; Naidu and Bhattacharya, 2009; Naidu et al., 2006; Ormachea Muñoz et al., 2016). Under reduced conditions, alluvial sediments containing As are often associated with the migration of As into groundwater (von Brömssen et al., 2007), due to dissolution of minerals (Suriyagoda et al., 2018) (Fig. 1). Notably, these reactions control the presence of As in groundwater, which depends on various biotic and abiotic factors, including distribution of the peat deposits/dissolved organic carbon (DOC) (Shahid et al., 2017), groundwater movement, pH, bicarbonate, Fe/Mn oxides, as well as microbial species (Hossain et al., 2014; Sracek et al., 2004). Under anaerobic conditions like in paddy soils, As concentrations can increase and reach up to hundreds of micrograms per liter.

Poor infrastructure and the absence of accessibility compels majority of the farmers in Bangladesh, Pakistan, and India to depend on existing groundwater resources for irrigation and rice crop production that might be contaminated with As (Brammer, 2009; Islam et al., 2012; Ravenscroft et al., 2009). As a result, soil receiving As-contaminated irrigation water from pumps also becomes contaminated with As over time. For example, As concentration of paddy soils in the range of 0.68–72 mg kg^{-1} have been reported in Bangladesh, Pakistan, and China (Table S1, Supplementary Information). Recharge of aquifers under the unconsolidated sediments by rainwater or flood water triggers reduction of Fe(*oxy*)hydroxides releasing As into groundwater (Shakoor et al., 2018). Consequently, the combination of natural enrichment and human disruption of groundwater resources leads to inorganic As release (Hu et al., 2013; Kumarathilaka et al., 2018; Nakaya et al., 2018). Arsenic through irrigation water is accumulated mainly in the upper soil layer (0–20 cm, i.e., rice plants rhizoplane) (Suriyagoda et al., 2018). Paddy soils are submerged for a longer period of time during the growth cycle of rice plants (3–4 months), and therefore, could undergo several physical and redox mediated chemical reactions as illustrated in Fig. 3 (Upadhyay et al., 2019; Yu et al., 2017a).

3. Paddy soils arsenic contamination and impact of various factors

3.1. Arsenic transfer from irrigation water in paddy soil-rice system

The accumulation of As in rice shoot and grain varies with irrigation water type. Groundwater, which is a major source of irrigation water, normally enters the rice fields through water channels. High As concentration is usually found in the irrigation water near entrance of water channel, and gradually decreases as a function of distance from the entrance (Neumann et al., 2014). At low flow rates, long residence time results in the aggregation of As bound to clay and Fe(*oxy*)hydroxides. Therefore, slow flow rates decrease As concentration along irrigation channel gradually (Dittmar et al., 2007). Conversely, a fast flow rate through the distribution channels leads to a short residence time, limiting the accumulation of As-containing colloids and subsequent settling (Fig. 3) (Neumann et al., 2010; Neumann et al., 2014).

Chemical changes can reduce total As concentration along the flushing channel. The basic principle is oxidation of As(III) to As(V) (Kabir et al., 2016) attained by diffusion of atmospheric oxygen in water used for irrigation. Iron(III) oxides can also help in oxidation of As(III) to As (V) (Roberts et al., 2004). Bioavailability of As can be increased by indigenous microbes present in the rhizosphere through siderophore production by solubilizing Fe(*oxy*)hydroxide that can bind As up to 160 mg kg^{-1} at the root-plaque interface (Kraemer, 2004). The formation of root-plaque can affect mobility and bioavailability of As in paddy soil-rice system, whereby both Fe and Mn co-exist at the root surface in root-plaques of rice plants. Iron concentrations in paddy soils are thought to play a major role in driving root plaque formation and concentration on rice roots. Therefore, rice genotypes grown on high Fe content paddy soil may have higher Fe plaques than the same rice genotypes grown on low Fe content paddy soil, causing As immobilization (Kraemer, 2004).

3.2. Effect of pH and redox potential (E_h)

Rice grain As concentration and soil pH showed a positive correlation with As in soil solution (Bhattacharya et al., 2002; Maity et al., 2017; Sandhi et al., 2017; von Brömssen et al., 2007). Changes in E_h and pH drive redox transformation and bioavailability of As in terrestrial

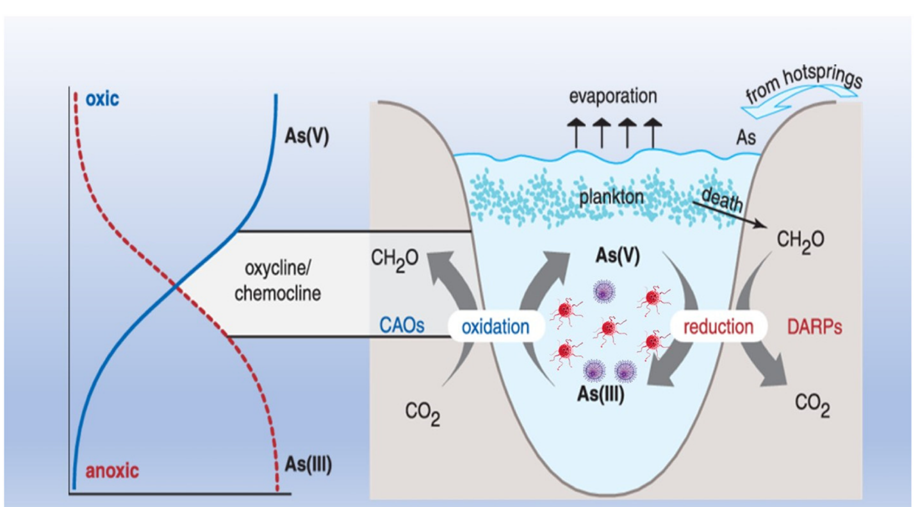

Fig. 1. Release mechanisms of arsenic (As) under different redox conditions (Modified from (Oremland and Stolz, 2003).

ecosystems, including the paddy soil-rice system (Fig. 1). Microbe-mediated redox transformation and dissolution of Fe(*oxy*)hydroxides are key factors for As speciation in paddy soil environment (Punshon et al., 2018; Punshon et al., 2017; Smedley and Kinniburgh, 2002). Under paddy soil conditions, E_h decreases (up to −200 mV) because oxygen as the electron acceptor is depleted and the development of anoxic conditions leads to dissolution of Fe(*oxy*)hydroxides (Meharg and Rahman, 2003), promoting release of sorbed As into soil solution. In conventional rice management, rice fields are subjected to alternate flooded and non-flooded conditions in the early stages or in final stages of rice growth (Kumarathilaka et al., 2018). In flooded soil conditions, oxidation and reduction reactions occur continuously resulting in fluctuating redox potential (E_h: +700 to −300 mV; between high and low E_h), thus triggering reduction of Fe(III), Mn(IV), SO_4, and methane generation (Frohne et al., 2011). If E_h becomes negative, as under paddy soils, the ratio of As(III)/As(V) in soil solution becomes high under flooded conditions (Nath et al., 2014).

3.3. Organic matter-arsenic interactions

Organic matter plays a significant role in determining As speciation in paddy fields (Rahaman et al., 2011). In rice cultivation, organic matter in the form of biofertilizer or rice stubble is added to soil and its roots are reintegrated into rice field after the crop harvest (Williams et al., 2011). Arsenic mobility is controlled by soil organic matter (SOM) content along with the chemical characteristics (Radloff et al., 2017).

Researchers have demonstrated that combined use of mountain thyme and poultry manure in rice fields contaminated with As reduced the total As content in rice tissue (Ajiboye et al., 2004; Batista et al., 2016; Bauer and Blodau, 2006; Dawe et al., 2003; Eiche et al., 2017). For instance, Fu et al. (2011) found a negative correlation between total As contents in rice tissue and SOM, with As concentration ranging from 20 to 1160 μg kg^{-1} DW (Tables S1 and S2, Supplementary Information). However, negatively charged functional groups in DOC can compete with As on Fe(III) oxides surface for available binding sites and increase mobility and uptake of As by rice plants. The concentration of DOC can also enhance activity of Fe(III) reducing bacteria that promotes the reduction of Fe(*oxy*)hydroxides, and causes As release into soil pore water (Bhattacharya et al., 2007; Bundschuh and Maity, 2015; Chen et al., 2014b; Chen et al., 2016).

3.4. Clay mineralogy

Low E_h under paddy soil conditions may lead to increased flocculation and dispersion of clay particles facilitating migration of clay to bottom of the plow zone and forming a hard clay pan (Kögel-Knabner et al., 2010). The presence of clay minerals results in fine structured soil with a relatively large surface area. Iron (oxy)hydroxides are mainly co-precipitated on the surface of clay particles that enhances the retention of As in paddy soil and decreases uptake by rice plants (Mohapatra et al., 2007). Surface runoff caused by heavy rains and the subsequent destruction of soil structure by tillage are the key causes of loss of the clay minerals during plantation of rice fields negatively affecting As immobilization (Kögel-Knabner et al., 2010; Regmi et al., 2013).

Various factors can affect sorption and redox forms of As in paddy soils, including the crystal structure and type of clay minerals. Secondary mineral deposits on surface of the clay minerals can change physico-chemical characteristics of mineral surfaces and impair the ability of clay mineral to adsorb As (Uddin, 2017). A long-term process (several months) can reduce extractability of As retained by the mineral and is considered to be important when restoring As-contaminated soils. Aging can be another vital factor that can impact the extractability and oxidation/reduction of As associated with clay minerals (Bhattacharya et al., 2002; Garnier et al., 2010; Regmi et al., 2013). Unfortunately, the long-term interaction between As and clay surfaces is not fully understood and needs further investigation of As speciation and micro-scale distribution in rhizosphere of rice plants by employing advanced X-ray fluorescence (XRF) and X-ray absorption spectroscopy (XAS) techniques.

3.5. Biotransformation pathways of arsenic: Microbial interactions

Microorganisms influence the forms of As in rhizosphere of rice plants directly and indirectly, and play a vital role in changing As biogeochemistry under flooded conditions (Gadd, 2010; Upadhyay et al., 2018). Under nutrient-restricted conditions, microorganisms can actively dissolve As-rich minerals to obtain nutrients for cell growth, thereby releasing As into pore water (Fig. 2) (Herath et al., 2020; Mailloux et al., 2009). Additionally, some favorable abiotic conditions could be generated through production of organic acids, polysaccharides and ligands that causes changes in As speciation and mobility in paddy soils for rice plant uptake (Cui et al., 2018; Guo et al., 2014; Upadhyay et al., 2018). Various bacterial species such as *Desulfovibrio strain* Ben-RB (Alam and McPhedran, 2019; Drewniak and Sklodowska, 2013), *Shewanella oneidensis* (Turpeinen et al., 2002), *Desulfosporosinus auripigmentum* (Cui et al., 2018), and *Streptococcus disintegrate* CN32 (Tsai et al., 2009) may promote formation of mineral-like As phases using As as a terminal electron acceptor (Wang et al., 2016). Jia et al. (2013) reported that the microbial-mediated redox processes could convert As from inorganic forms to organic species, e.g., MMA and DMA. The role of microbes in enhancing As bioavailability and their ability to carry out thiolation processes needs further exploration. Although As thiolation has been investigated in surface/sub-surface, sulfide-rich sediments and aqueous systems, it remains poorly understood under paddy soil conditions (Awasthi et al., 2018; Herath et al., 2018; Hussain et al., 2020a; Hussain et al., 2019b).

Volatile As species are developed due to the methylation of As or in the form of arsine (AsH_3) gas under paddy soil conditions where microorganisms play a vital role (Table 2). Although different metastable, intermediate volatile As forms are produced (i.e., monomethylarsine ($MeAsH_2$), dimethylarsine (Me_2AsH_2) and trimethylarsine (TMA)), the final volatile As form obtained is gaseous AsH_3 (Hu et al., 2020; Huang, 2014). It can be trapped by microbial/algal biofilms under reducing conditions, and as a result can be oxidized to inorganic As, thus completing As cycle in paddy soil-rice environments, either by precipitating or depositing produced inorganic As back into the soil (Cullen, 2014; Herath et al., 2020; Hussain et al., 2020b). With a decrease in E_h and addition of organic material, biomethylation of As in soil increases with microbial aid (Fig. 2).

For example, after adding straw and animal manure, an increase in As volatilization in paddy soil was observed to decrease by 2-fold. Likewise, fungal inoculation enhanced As remediation up to 7-fold in highly As-contaminated paddy soils, using *Penicillium* and *Ulocladium* spp. (Grob et al., 2018; Upadhyay et al., 2018). Similarly, biomethylation can effectively form volatile As compounds (e.g., alkyl-arsines), thus increasing As losses into the atmosphere. Various As compounds are produced after biomethylation, including mono-methyl, di-methyl, tri-methyl, As oxides. Volatile As species are more toxic and bioaccessible than the inorganic As forms; however, these are easily oxidized and demethylated in air. Hence, biological volatilization may provide an effective technique for remediation of As-contaminated soil (Table 2).

Arsenic volatilization mediated by microorganisms is inefficient hindering practical trials to apply it as a strategy for soil remediation. Gaseous As-hydrides evaporate from As-contaminated soil into the atmosphere slowly. Studies have shown that 0.5–70 μg As kg^{-1} soil was volatilized over a year (Turpeinen et al., 1999; Wu et al., 2002; Yang et al., 2018). Genetically modified bacteria have been applied as a way to increase As methylation and volatilization in soils. The arsM (S-adenosyl methyltransferase) was isolated from *R. palustris* and encoded in *S. desiccabilis* and *B. idriensis* in aqueous media resulting in 9-fold higher As losses (volatilization) compared to wild strains (Grob et al., 2018; Mestrot et al., 2016; Mestrot et al., 2013).

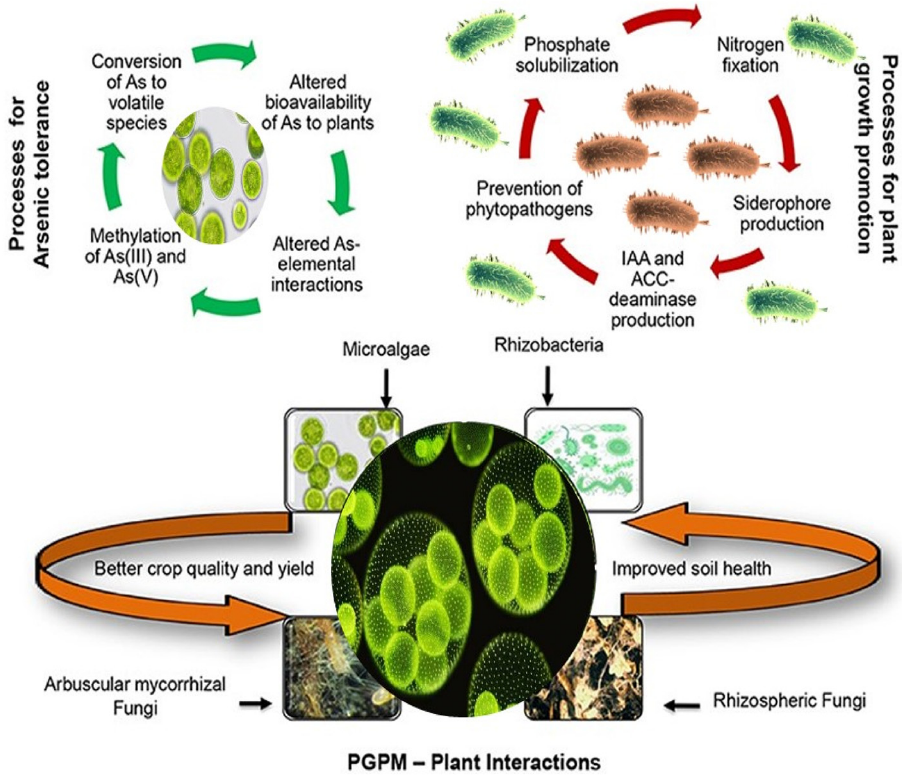

Fig. 2. Interaction between different microbial process and its efficiency in arsenic (As) stress tolerance and growth promotion of plants (Reproduced with permission from the publisher, Hussain et al. (2019b)).

Arsenic in sludge (bio-solids) can be converted by microorganisms through multiple processes like redox reactions and biomethylation (Ojedokun and Bello, 2016). Soil microorganisms obtain their energy from soil organic carbon pool (Bhowmick et al., 2018; Kim et al., 2018) that is involved in reduction of As. Organic As compounds can also be present in the environment, but their concentrations are low. The retention of As(III) by inorganic soil components is not as strong as that of As(V), hence microbial-mediated reduction ultimately leads to highly mobile and bioavailable As species (Park et al., 2011).

3.5.1. Arsenic methylation pathways in rhizosphere and microbial cells

Microorganisms including bacteria, fungi, eukaryotic algae, and Archaea are responsible for conversion of As(III) to methylated As species. Microbial genes that code for As(III) methylation (arsM) for As treatment lead to formation of mono-, di-, tri- and tetra-methyl As species in paddy soil environments (Wang et al., 2014b). In reduced conditions, several factors may control As methylation processes. Firstly, diversity and activity of various anaerobic microbial species may enhance with As concentration; secondly, As(III) transformation increases under flooded conditions, and as such it is utilized as substrate and transform to methylated As form; thirdly, reductive dissolution of Fe/Mn oxides lead to desorption of methylated As (i.e., MMA(V) and DMA(V)) into soil solution (Zhao et al., 2013). It has been reported that rice plants do not have the ability to methylate As species in plant tissues. Therefore, the methylated As compounds present in rice are produced by microbial-mediated methylation of As in plant rhizosphere (Zhao et al., 2013).

In addition, various cellular mechanisms may occur in microorganisms that help them in methylation-demethylation cycling of As in paddy soils. For example, S-adenosylmethionine (SAM) and methylcobalamin have been reported in As methylation as both act as methyl donor for As methylation process under paddy soil conditions (Bentley and Chasteen, 2002). In microbial cells, As(V) could be reduced to As(III) by GSH followed by As(III) catalysis via methyltransferase enzyme that accepts methyl group from SAM to form MMAs(V) and subsequently MMAs(III) (Zakharyan and Aposhian, 1999). Methylcobalamin can convert As(III) directly into MMAs(V) and

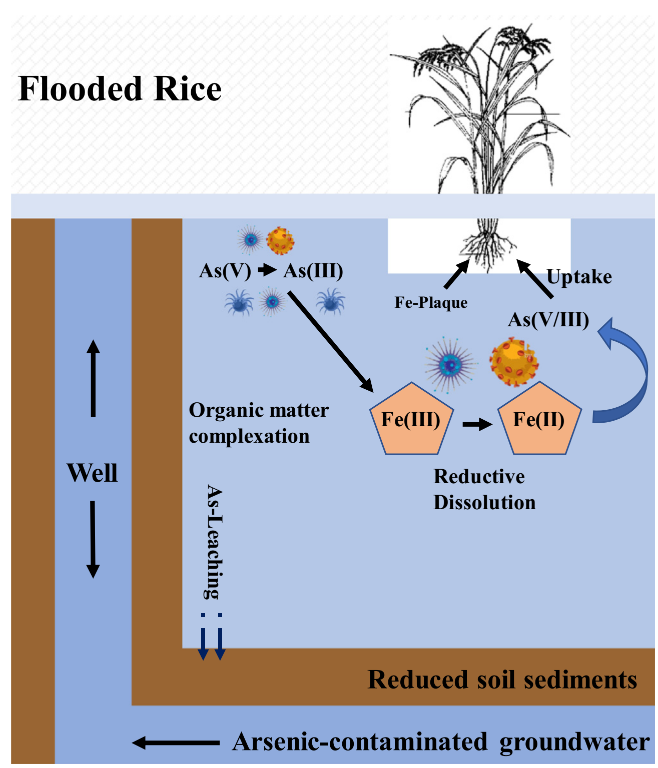

Fig. 3. Impacts of arsenic (As)-contaminated irrigation water on As uptake by rice in paddy soils.

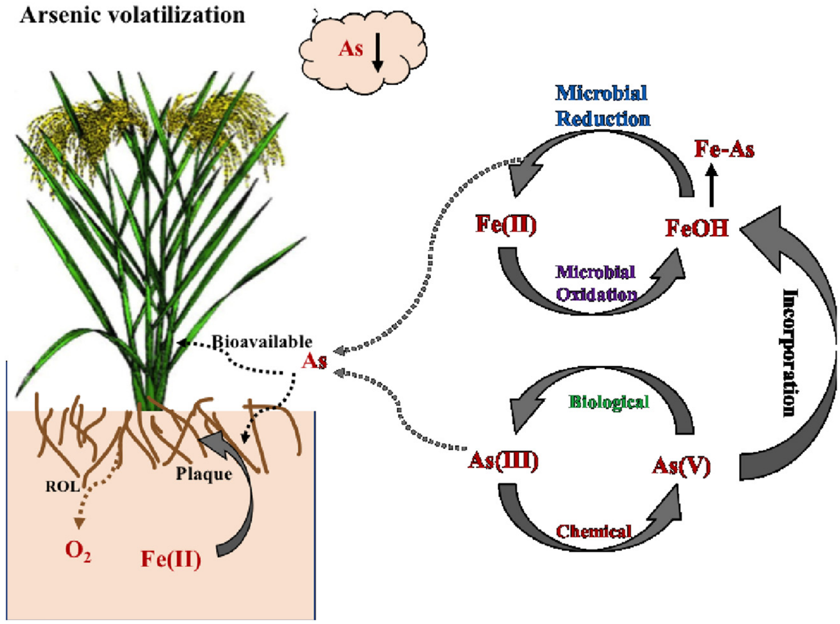

Fig. 4. Arsenic (As) interaction with iron (Fe) amendments in paddy soil conditions and uptake by rice plants (Modified from Wang et al. (2018b)).

DMAs(V) under reducing soil conditions without involvement of methyltransferase enzyme, in the presence of GSH (Zakharyan and Aposhian, 1999).

Microorganisms can also demethylate As into inorganic arsenical compounds in paddy soil conditions. In a previous study, it was demonstrated that *Burkholderia* sp. MR1 can transform DMAs(V) to MMAs(V), but it was unable to further demethylate MMAs(V) to inorganic As species under paddy soil environments (Lehr et al., 2003). However, *Streptomyces* sp. MD1 was able to transform MMAs(V) to inorganic As(V) species. This indicates that demethylation of methylated

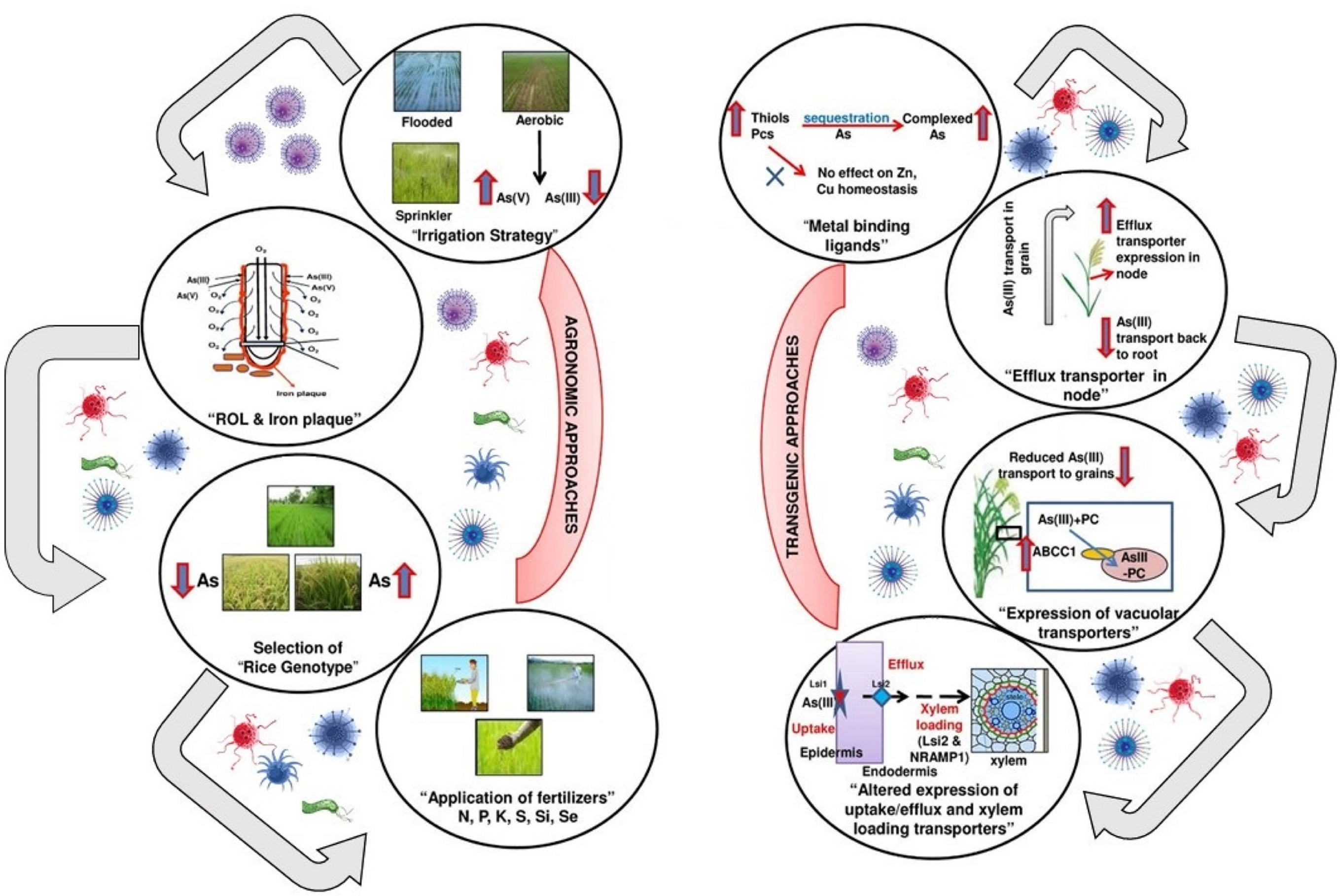

Fig. 5. Schematic diagram of multiple mechanisms carried out by the application of various organic and inorganic amendments, and the role of multiple transporters on arsenic (As) uptake and effluxes in rice plants (Modified from Awasthi et al. (2017b)).

Table 2
The role of microbes in biotransformation and transformation pathways of arsenic in paddy soils.

Microbial species	Source	Major As species	Reference
Pseudomonas putida (with *R. palustris arsM*)	Paddy soil	DMAs(V)	(Chen et al., 2014a)
Streptomyces sp.	Rice rhizosphere	MMAs(V), DMAs(V)	(Kuramata et al., 2015)
Proteobacteria, *Gemmatimonadales*, and *Firmicutes*	13 paddy soils collected across Southern China	*Arsenite* (*As*(*III*)) *oxidase genes* (*aioA*), *respiratory arsenate* (*As*(*V*)) *reductase genes* (*arrA*), *As*(*V*) *reductase genes* (*arsC*), and *As*(*III*) *S-adenosylmethionine methyltransferase genes* (*arsM*)	(Zhang et al., 2015b)
Arsenic methylation bacteria	5 paddy soils with low-As concentration	*Arsenate reduction genes* (*ars*) dominated in all soil samples, and significant correlation existed between the abundance of *arr* (*arsenate respiration*), *aio* (*arsenite oxidation*), and *arsM* (*arsenite methylation*) genes	(Xiao et al., 2016)
Sulfate-reducing bacteria	Mekong Delta paddy soil	DMAs(V)	(Reid et al., 2017)
Arsenic methylated-genes (microbes)	Paddy soils, Pakistan	*aioA* and *arsM with Tri-As*	(Hashmi et al., 2020)
Bradyrhizobium, Bryobacter, Candidatus Solibacter, Geobacter, Gemmatimonas, Halingium, and *Sphingomonas*	Mining site	–	(Li et al., 2020)
Geobacter and *Hydrogenophaga*	Paddy soil (Fe-plaque)	*arsC* (encoding As(V) reductase) and *arsB* genes (encoding As(III) efflux membrane protein)	(Hu et al., 2019)
Multiple bacterial strains	Paddy -rice system	*Geo, arsC* and *arsM*	(Xue et al., 2020)
Fe(III) reducing bacteria *Geobacteraceae* and sulfate reducing gene *dsrA*	Paddy soil	–	(Tang et al., 2020a)
Proteobacteria, *Bacteroidetes*, *Geobacter*, *Sphingomonas*, *Streptomyces*, and *Rhodopseudomonas*	Paddy soil	arsM	(Yan et al., 2020)
Geobacter and *Shewanella; Gammaproteobacteria*	Paddy soil	*arsM* gene	(Yang et al., 2020)

As species is coordinated by multiple microorganisms performing their specific functions during demethylation process (Table 2). However, recent research showed that some of microbial species (such as *Nostoc* sp.) can perform both these processes by converting dimethyl As to monomethyl As followed by transformation into inorganic As species (Yan et al., 2015).

The addition of methylated As (i.e., MMAs(V) and DMAs(V)) into paddy soil increases volatilization losses of As considerably since biotransformation of methylated As to volatile As forms occurs quickly (Mestrot et al., 2013). Arsenic is more volatile under flooded soil conditions than in the aerobic soils. This is partly because under flooded soil conditions the activity and abundance of As methylating microorganisms increases (Di et al., 2019).

4. Arsenic interaction with inorganic and organic amendments in flooded soil conditions

4.1. Inorganic amendments-arsenic interaction

4.1.1. Gypsum

A few studies have focused on the immobilization of As under paddy soil conditions, whereby gypsum was used in combination with other amendments (Zhang et al., 2015a; Qayyum et al., 2017; Zhao et al., 2018). Fernandez-Martinez et al. (2008) examined the effect of DOC coagulants as metal fixatives for plant growth. Gypsum as a DOC coagulant has been tested for the efficiency of simultaneously immobilizing Cd, Pb, and As in agricultural soils surrounding mining areas. The incorporation of gypsum at 3% (wt/wt) into soil resulted in better immobilization of As than that of Cd and Pb (Kim et al., 2018). Lime, compost, and a mixture containing lime and gypsum were used to evaluate uptake of these toxic elements by *A. gigas* and *A. macrocephala*. Arsenic uptake was lower compared to Pb and Cd under the impact of gypsum. Results indicated that gypsum immobilized As up to 76%, Cd by 54% and Pb 46% (Table 1) (Kim et al., 2018; Zhao et al., 2018).

4.1.2. Lignite

Lignite application in the remediation of As under paddy soil environments has been partially explored, but its application in immobilization of other potentially toxic elements has been investigated (Rehman, 2007; Rehman et al., 2017). Arsenic sorption potential of various geological materials such as lignite, bentonite, shale, and Fe-rich sand was evaluated in batch incubation experiments (Mar et al., 2013; Rehman, 2007) (Table 1). Results showed that, among these geological materials, lignite was the most promising material for As(V) immobilization followed by bentonite, shale, and Fe-rich sand. However, the amount of As(III) adsorbed by all these geological materials was found to be lower than that of As(V). Thus, it is important to take into account the As species present before applying inorganic amendments for remediation and restoration programs of As in paddy soil environments.

4.1.3. Iron-based modifiers

Among many inorganic modifiers that can help immobilize As in paddy soils, Fe-containing modifiers have very strong impact on the sorption properties of soil and that is why these are considered promising in regulating As mobilization under paddy fields (Irem et al., 2019; Matsumoto et al., 2016; Matsumoto et al., 2015; Zou et al., 2018). As soon as the density of surface sites is reduced sufficiently to question the occupancy of As sites, the reduction of Fe leads to desorption of As. However, for Fe(*oxy*)hydroxides such as ferrihydrite, hematite, and magnetite, whose redox chemistry can change under submerged paddy soil conditions, making As adsorption-desorption complicated. In addition, the regulated release of As(III) from ferrihydrite in pore water varies with reaction time. Prolonged Fe(III) reduction and dissolution processes can ultimately lead to release of As(III) into soil pore water in paddy soils. Essentially, three competitive processes occur during the reaction process that together determine dissolved concentration of As(III): (i) desorption due to chemical imbalance during pore water exchange (flow); (ii) Fe phase retention for transformation; and (iii) As release from the dissolved ferric phase (Table 1) (Burton et al., 2009; Fendorf et al., 2010; Johnston et al., 2015; Niazi and Burton, 2016). Iron minerals can also decrease As mobility (e.g., Fe sulfate, zero-valent Fe (Fe(0)), red mud, steel shot) minimizing As solubility and toxicity and limiting As uptake by rice (Fig. 4) (Nagar et al., 2013).

Use of Fe(*oxy*)hydroxides reduces solubility and uptake of As under paddy field conditions mainly due to adsorption and co-precipitation pathways. Notably, the effectiveness of Fe(*oxy*)hydroxides in reducing absorption of As by rice plants in paddy soils is attributed to the type, size, crystallinity of Fe (oxy)hydroxides, soil E_h, pH, (Fig. 4), as well as amount of the root exudates or inorganic and organic ligands (Burton et al., 2009; Johnston et al., 2015; Niazi and Burton, 2016).

Iron-based amendments have been used for sequestering As in paddy soils to reduce its accumulation by rice (Table S4, Supplementary Information) (Juang et al., 2020; Yu et al., 2017a; Zou et al., 2018). For example, Yu et al. (2017a) noted that the poorly crystalline Fe oxides (PC-Fe) were a more promising (54%) Fe amendment than that of $FeCl_2$-$NaNO_3$ (52%) and $FeCl_2$ (46%) in decreasing rice grain As content over control at the filling stage. Moreover, data from 2% Fe oxide application are very consistent. Farrow et al. (2015) applied (2%) Fe oxide to As-contaminated soil and found that Fe oxide based amendments significantly decreased As concentration in rice grain (67%, i.e., from 0.16 to 0.05 mg As kg^{-1} DW), suggesting that the Fe oxide (hematite, magnetite) amendment to be feasible for reducing grain As content. However, the authors indicated that the application of these Fe oxides (2% wt/wt) in their study was not economical for field application, because the As immobilization ability of magnetite and hematite mixture was lower compared to granular Fe(III)-hydroxide and some other forms of Fe oxide minerals (Farrow et al., 2015). Irem et al. (2019) reported that accumulation of As in rice grain followed a linear trend in BR-1 rice genotype, whereas a parabolic relationship was observed in Bas-385. Arsenic concentration in the rice grain was reduced in both genotypes that exhibited a positive response with Fe sulfate amendments.

4.1.4. Manganese (Mn)

Remediation mechanisms for As in paddy soils can generally be divided into immobilization and removal that can be achieved by the application of certain amendments. Mn-based amendments could be an effective and cost-effective approach to transform As(III) species into less toxic and mobile As(V) and immobilize As in paddy soil (Xie et al., 2020) (Table 1). Manganese oxide is considered as an important component in soil that can oxidize the As(III) form due to its high reactivity (Fig. S1, Supplementary Information), poor crystal structure, and large surface area (Carrijo et al., 2018; Carrijo et al., 2019; Parikh et al., 2010). Therefore, despite the lack of direct experimental evidence, the Mn-dependent pathway could possibly be the key transformation process of As(III) oxidation in rice root-plaques. Komárek et al. (2013) reported that amorphous Mn oxides decreased As bioavailability to rice plants grown in paddy soils contaminated with different As contamination origins. Maximum adsorption capacity of amorphous Mn for As(V) was found to be at pH 7–8. The adsorption capacity was reduced at low pH (especially at pH 4) due to dissolution of amorphous Mn oxides (Kögel-Knabner et al., 2010; Komárek et al., 2013). At pH ranging from 4 to 7, poorly crystalline Mn oxide demonstrated the greatest ability to transform As speciation in solution. In the low pH range, oxidation of As is associated with reductive dissolution of Mn oxides, and as such As(V) precipitates and becomes insoluble on surface of the Mn oxides and/or dissolved Mn_2O colloidal particles. Application of Mn oxides can decrease dissolved As concentration in soil solution and rice tissue, because of their ability to oxidize As(III) to As(V) in paddy soil facilitating As adsorption on the Mn oxides surface (Bhattacharya et al., 2002; Martínez-Villegas and Martínez, 2008). Amorphous Mn oxides are primarily As stabilizers in soils with circa neutral to alkaline pH and under acidic pH As mobilization could be increased in soil solution phase (Amirnia et al., 2019; Frohne et al., 2011; Suda and Makino, 2016; Xu et al., 2017).

4.2. Organic amendments-arsenic interaction

4.2.1. Biochar

Biochar can (im)mobilize As in soils and sediments, although its impact on biogeochemical cycling of As in As-contaminated rice fields is not well understood (Javed et al., 2018). Biochar is considered to be low-cost adsorbent, that is carbonaceous and stable, and attained by the pyrolysis (>250 °C) of a variety of biowastes under little or no oxygen conditions (Amen et al., 2020; Sohi et al., 2010). Biochar has been reported to be an effective material for improving soil properties such as nutrient retention, contaminant immobilization, and carbon storage (Mohan et al., 2014) (Table 1). However, the majority of published studies have examined the effect of biochar on mobilization processes of As in soil under limited settings and with static pH, E_h, and soil moisture situations. Whereas, in paddy settings changes in soil E_h, pH, and mineral chemistry are dynamic. A few studies have examined the ability of biochars and some other materials (e.g., clay, alumina, bio-charcoal, perlite, shell limestone, organic fertilizer) to immobilize As in floodplain soils (Mohan et al., 2014; Wen et al., 2020; Yang et al., 2020). In addition, biochars are negatively charged and As in soil environment are oxyanions limiting adsorption capacity of biochar (Kumarathilaka et al., 2020a; Zama et al., 2018).

Given these specific challenges, when using biochar to immobilize soil As, impregnating biochar with Mn oxide to fix As in soil may be a better option since Mn oxides can limit the flow of As through anion exchange and convert As(III) into As(V) that is stable and less toxic (Yu et al., 2017b). Most of the studies showed that biochar can adsorb various organic and inorganic contaminants in soil and water, the kinetics of which mainly depends on specific properties of biomass used and pyrolysis temperature for biochar production (Niazi et al., 2017; Savage et al., 2018). Qiao et al. (2018) evaluated the influence of various modified biochars (i.e., $FeSO_4$, hydroxysulfate (biochar-FeOS), $FeCl_3$ (biochar-$FeCl_3$), zero-valence Fe (biochar-ZVFe)) on As immobilization in soil by determining $NaHCO_3$-extractable As pool and soil pH (Table 1). Mixing of soil with biochar-ZVFe, biochar-FeOS, and biochar-$FeCl_3$ significantly decreased the $NaHCO_3$-extractable As pool in soil (13–30%, 10.9–28.3% and 17.9–35.1%, respectively), while there was no effect on soil pH. Treatment with biochar-FeOS, biochar-$FeCl_3$, and biochar-ZVFe reduced concentration of non-specifically adsorbed and specifically adsorbed As phases in soil and increased As content in Fe/Al poorly crystalline pools (Qiao et al., 2018; Wen et al., 2020). Under reduced conditions, microbial-mediated reduction of Fe(III) can lead to release of adsorbed As(V) into soil solution (Qiao et al., 2018). Future research is warranted to prepare functionalized biochars that will allow for effective immobilization for specific toxic elements, like As anions, in soil under paddy soil environments.

4.2.2. Other organic amendments

Agricultural biowaste materials, especially those containing cellulose have a great metal and metalloid sorption capacity (Mehmood et al., 2021). The basic components of biomass of agricultural wastes include hemicellulose, lignin, lipids, proteins, monosaccharides, hydrocarbons, and starch, which contain various functional groups. These functional groups contribute to binding of toxic elements, like As (Hossain et al., 2020; Nawab et al., 2018). A series of experiments were carried out using farmyard manure (FYM) to improve paddy soil for 10, 25 or 50 days of treatment. The 0.5 M $NaHCO_3$ was used to extract available As from soil (Arao et al., 2009). Data showed that the $NaHCO_3$-extractable As pool increased after 25 days and it decreased after 50 days of incubation. Also, it was observed that the soils amended with high amounts of FYM had relatively higher bioavailable As, as well as more As concentration in rice compared to the soils receiving less amount of FYM. Methylated As species in rice were observed to increase in soils with the high FYM application. Recently, it has been suggested that methylated As forms can be linked to the soil under microbial action and may not be produced in plants (Hossain et al., 2020;

Wasserman et al., 2018). Hence, the increase in proportion of methylated organic As forms in cereals like rice under flooded situations can be attributed to the complex As biogeochemical processes prevailing in paddy soils (Ma et al., 2016).

Cow dung based manure can also stimulate activity of indigenous microorganisms which have a strong influence on As fate and transform As into soluble and volatile species through different pathways under submerged soil conditions (Huang et al., 2012). Cow dung derived FYM is considered to be eco-friendly and inexpensive soil amendment, and a source of nutrition (e.g., nitrogen, phosphorus, sulfur). It can attract bacteria, fungi, protozoa, worms, nematodes and arthropods, thus improving the natural As biogeochemical process (Banik et al., 2006) (Fig. 5) (further detailed discussion on As (im)mobilization processes by amendments is given in Text S1, Supplementary Information).

Microorganisms are the main cause of biochemical degradation process during vermicomposting while earthworms can enhance the activity and population of microorganisms (Kaur et al., 2010). Rahaman et al. (2011) demonstrated that the concentration of As in rice shoot decreased significantly after application of poultry manure, lathyrus, and vermicompost together into the As-contaminated paddy soil. Arsenic content varied between 0.70 and 1.67 mg kg^{-1} DW in rice grain (Table S3, Supplementary Information) that was above the Food and Agriculture Organization/World Health Organization safe limit of As in rice grain (0.2 mg kg^{-1} DW).

4.2.3. Advantages and disadvantages of the organic and inorganic amendments

Although Fe-based materials have ability to immobilize As in paddy soil conditions, the high synthesis cost, less eco-friendly nature of dissolved Fe at high concentration, widely unavailable and un-stability in reduced soil conditions with possible release of adsorbed As may be possible limitations of their field scale application (Mulligan et al., 2001). In addition, application of biochar in remediating As-contaminated soils has been an emerging research topic over the past decade (Qiao et al., 2018; Yu et al., 2017b). Various organic amendments including biochar, cow dung, biogas slurry and farmyard manure have great potential to sequester As under varying environments as discussed in above section (Irem et al., 2019). Cow dung and farm yard manure are widely available, low-cost materials and may be applied to immobile As in paddy soil to reduce As accumulation, as well as supplement the nutrients for rice plants (Hussain et al., 2019a). However, their As remediation potential and economic feasibility have not been examined yet in paddy soil-rice system. Based on a rough estimate, organic amendments may be available at US $2–30 ton^{-1} (Ojedokun and Bello, 2016). Also, lignite could have potential to immobilize paddy soil As, although it is not well understood (Mohan and Chander, 2006). Lignite is freely available and its estimated cost is US $25 metric ton^{-1} which is lower than the Fe-based amendments (US $25–$300 ton^{-1} material treated) (Table S4, Supplementary Information) (Mohan and Chander, 2006).

Notably, organic amendments could be more advantageous than Fe-based amendments since these: do not produce contaminated sludge residues, supply micro- and macro-nutrients for crop growth, improve soil physical, biological and chemical health, environmentally-friendly in nature, cheap and abundantly available.

5. Effect of mineral nutrition on arsenic availability and uptake by rice

Some strategies have been developed to reduce the translocation and uptake of As by rice plants under paddy soil conditions. Most of these strategies rely on changing the physical and chemical properties of the soil using various mineral elements. Similarly, the use of N, P, Se, Si, and other elements are reported to affect the accumulation and immobilization of As. This section discusses the effects of various nutrient-based modifiers to minimize the accumulation of As in rice plants (Fig. 5).

5.1. Phosphorus (P)

The dynamics of As in rice rhizosphere is controlled by state of plant phosphate (PO_4) concentration that regulates the formation of Fe plaque and absorption of As(V) via PO_4 transporters (Fig. 5). Previous studies have shown a competitive inhibitory effect between phosphate and As(V) in soil or inside the plants (Ahmad et al., 2020a; Amna et al., 2020). Due to similarity in chemical properties, Fe plaques may adsorb oxyanionic As species via the ligand exchange mechanism due to competition with phosphate, although the effect of As on plant metabolism is complicated (Bolan et al., 2013). The root to shoot As(V) transport occurs through different PO_4 transporter (PHT) proteins. The regulators of PO_4 transport i.e., OsPHF1 (PO_4 transporter traffic facilitator 1) and PHR2 (PO_4 starvation response 2) have been reported to affect As (V) uptake by rice plants (Awasthi et al., 2017a). Increased PO_4 concentration may impact the steady state As(V) uptake by rice or As(V) could moderately inhibit PO_4 translocation in rice plants (Anawar et al., 2018).

Irrigation of As-contaminated water can alter As and PO_4 balance in soil solution, resulting in PO_4 mobilization and loss due to leaching or high PO_4 utilization by rice plants (Anawar et al., 2018; Samal et al., 2020; Talukder et al., 2014). The development of aerobic/partial-aerobic conditions around the plant roots, due to high respiration rate and excretion of oxygen containing root exudates, oxidation of As(III) to As(V) could occur increasing As(V) competition with PO_4 in plants (Bakhat et al., 2017).

Phosphorus can decrease As concentration in rice shoot and grain by increasing displacement of As from root adsorption sites, thus leading to enhance As concentration in the rhizosphere (Ahmad et al., 2020a; Jain and Loeppert, 2000). It has been indicated that As uptake in rice grain was significantly enhanced from soils with initially sufficient P. Addition of P tends to decrease As accumulation in rice grains (Islam et al., 2019). Various studies have shown that applying P modifiers can increase the mobility of As in rhizosphere, thus increasing As uptake by plants (Talukder et al., 2011; Wang et al., 2014a). Like excessive use of P in some cases can desorb 80% of soil adsorbed As into the soil solution-phase (water soluble As pool) that can leach deeper into the soil profile depending on rainfall, and soil physical and chemical properties. Notably, the mobility and availability of P pool in soil can also control the formation of Fe plaque on rice plant roots (Farias et al., 2017).

There are indications that rice plants with enough P concentration have increased their ability to tolerate As which may be due to large biomass and reduced retention of As by rice plants (van Genuchten et al., 2012). Therefore, rice varieties that have improved the ability to extract P from soil can provide a means of reducing As uptake and minimizing the risks associated with P-induced migration and As absorption. Lee et al. (2016) suggested three important factors controlling As transport in soil and rice plants: (i) the competition between As and P for exchange sites; (ii) the antagonistic effect of PO_4; and (iii) the role of P in translocating As from root to shoot. The toxicity of As in plants depends on the As/P ratio in the soil, not the actual As concentration. In a survey report on Chinese rice fields it was found that changing the P content in shoot can reduce the accumulation of As in rice grains (Lu et al., 2010). In As-contaminated soils, the use of calcium forms Ca-P-As complex that leads to a decrease in As mobility (Neupane and Donahoe, 2013). A series of experiments were carried out using water management regimes and P application to minimize As translocation in different rice genotypes in Bangladesh. All the experiments indicated that application of P (optimum levels or 0, 12.5 and 25 mg kg^{-1}) reduced As content in grain, root, and shoot up to the permissible limits (Talukder et al., 2012; Talukder et al., 2014). Future research should be directed toward understanding cellular mechanisms controlling the relationship between As and P absorption and competition in rice plants in contrasting rice genotypes.

5.2. Silicon (Si)

Silicon supplementation of rice could help in reducing uptake and transfer of As to rice plant shoot and grain that is ascribed to silicic acid (Si) competition with As(III) species for absorption via similar (aquaporins) channels in rice plants (Fig. 5) (Ahmad et al., 2013). The similar geochemistry of both As(III) and Si can create competition for the adsorption sites in paddy soil on colloid surface, and with increasing Si application As concentration can also increase in soil solution (Limmer et al., 2018). Therefore, the complex interaction in soil (antagonism) and rice plants (synergism) between these two elements is important to understand in biogeochemical As cycling under the impact of Si fertilization. Silicon is absorbed by plants in the form of orthosilicate (H_4SiO_4) and its concentration in rice tissues is higher than some other crop plant species (Sun et al., 2020; Zhang et al., 2020). Due to the similarity between As(III) and H_4SiO_4, similar absorption pathways have been reported in plant roots (Suriyagoda et al., 2018). Arsenite is taken up in rice plants by two Si transport channels: (i) low Si-1 (Lsi1), that is known as a Si influx carrier located at the exterior part of exodermis and endodermis; and (ii) low Si-2 (Lsi2) that is a Si efflux transporter/carrier at the interior proximal side of exodermis and endodermis. It facilitates in discharge of Si ions into the xylem tissue of rice plants (Islam et al., 2019; Seyfferth et al., 2018).

In rice plants, Si inflow transporter Lsi1 (rice with low Si content 1; OsNIP2) is responsible for absorption of As(III), while Si outflow transporter Lsi2 (rice with low Si content 2) mediates outflow of As(III) (Chen et al., 2017). Therefore, the coordinated function of these two transporters can create effective flow of Si and As(III) through endodermis and toward xylem tissue. Ma et al. (2017) showed that Lsi2 plays more important role than Lsi1 in transporting As to rice grains, but switching off Lsi2 may also disrupt Si absorption that can inhibit rice growth and reduce grain yield by 60%. Song et al. (2014) observed that a C-type ATP-binding-cassette (ABC) transporter (OsABCC) family, OsABCC1 (i.e., a Si transporter in rice), was responsible for the reduction and detoxification of As in rice plants. Their findings supported that As translocation in rice grain can be reduced significantly by OsABCC1 transporter via As sequestration into vacuoles of phloem cells of rice.

Silicon supplementation under As stress can upregulate the vascular sequestration of As in rice plants and decrease metabolic damage to plant tissue. Various mechanisms are related to the use of Si in soil or Si as foliar dressing on leaf to reduce bioaccumulation of As from roots and transfer to rice shoot and grain (Begum et al., 2016). Such pathways can involve: (i) the antagonism between Si and As at the root absorption sites as As(III) divides Si transport pathway into rice root cells; (ii) Si can enhance the development of plaque on the surface rice roots, that is due to establishment of Fe-oxides-silicate complexes on the root surface, thus decreasing As uptake by roots and increasing tolerance; and (iii) Si also help in the upregulation and expression of those genes which are thought to be involved during absorption and transport of As in rice (Begum et al., 2016).

Arsenic concentration in soil solution and plant tissue has been observed to be relatively lower in response to Si application in soils dominated by Fe(III) oxides, rather than Ca-bearing mineral phases (e.g., $CaSO_4$, $CaCO_3$). These data show that the Si-induced development of plaque at the rice root under flooded situations may be the main mechanism for reducing As accumulation in rice after introduction of Si into soil (Islam et al., 2019). This indicates that Si applied to low-Fe soils can only effectively minimize the accumulation of As in rice, if Si is used at a high dose (Li et al., 2017; Limmer et al., 2018; Seyfferth et al., 2018).

5.3. Sulfur (S)

Sulfur content in paddy soil shows a large spatial variability due to human activities such as SO_2 emission and SO_4 fertilizers application to soil. The behavior of S is closely related to E_h of the paddy soil that controls the valence state of S varying from −2 to +6, and the oxidation of organic sulfur compounds (Norton et al., 2017). High concentration of Fe(III) and SO_4 in paddy soils and with activity of SO_4-reducing bacteria, sulfide (S^{2-}) and Fe(II) are produced which reacts to form nanoparticles of FeS (e.g., mackinawite) (Niazi and Burton, 2016). Microorganisms can trigger the oxidation of elemental sulfur (S) to SO_4, thereby limiting the reduction of MnO_2 or $Fe(OH)_3$ in anaerobic paddy fields (Herath et al., 2018).

Sulfur is also a component of thiol-rich compounds such as phytochelatin (PC), which can produce stable complexes that later on can decrease the bioavailability and toxicity of As in rice tissues (Hussain et al., 2019b). The negative correlation between PC concentration and As in cereals confirms that PC may have a possible role in minimizing As movement from root-shoot- grain by making these stable complexes or increasing the drainage of roots from the external medium (Hussain et al., 2019b; Norton et al., 2017; Zhang et al., 2011). Arsenic translocation from root to shoot can be reduced by these two mechanisms: (i) reduction of OsLsi1, OsLsi2 and S-induced expression in rice as an active transporter, and (ii) induced S-thiol synthesis thus increasing complexation of As (Dixit et al., 2016; Hashimoto and Kanke, 2018). Therefore, S-fertilizer application coupled with microbial SO_4 reduction can precipitate As either with FeS or as As(III)-S^{2-} stable precipitates. The previous bulk of research has demonstrated that the bacterial populations in rhizosphere region of rice roots might have the key role to biologically transform SO_4 to S^{2-} form that can stabilize As as As(III)-S like phases, as well as via binding on the surface of Fe(II) sulfide minerals (e.g., mackinawite (FeS), As_2S_3) (Dixit et al., 2016; Fisher et al., 2007; Wang et al., 2018a).

The above discussion suggests that some long-term field trials are needed to determine the possible role and merits of using S-containing fertilizers for improvement in rice growth and limiting the transfer of As to rice grain in contrasting paddy soils. Therefore, a thorough and fundamental understanding is required on the type, stability and formation of As(III)-S^{2-} or FeS-As like phases under flooded soil environments.

5.4. Nitrogen (N)

The major nutrient, N, is essential for biosynthesis of different biomolecules (e.g., nucleic acids, chlorophyll, proteins, plant hormones), and its availability in soil also assists in regulating the growth of rice plants in paddy fields (Suriyagoda et al., 2018). In the soil, there are two most important N forms available: i.e., nitrate-N (NO_3-N), which is present under aerobic conditions and ammonium-N (NH_4-N), that is prevalent in flooded (paddy) soils (Ma et al., 2020b). In flooded rice fields, the main form of N is NH_4-N, while NO_3-N content is found to be <10 μM. Rice plants can utilize both forms of NH_4 and NO_3 as a N source (Fan et al., 2009). Nitrate is a strong oxidant, therefore, the addition of NO_3 may decrease aqueous concentration of Fe(II) by oxidizing Fe(II) to Fe(III) in soil solution, and as such reducing the formation of Fe plaques in vicinity of rice roots (rhizosphere) (Carrijo et al., 2018; Saleque et al., 2004). However, increasing concentration of NH_4 can enhance reduction of Fe(III) to Fe(II) that facilitates diffusion of Fe(II) into the rhizosphere zone and development of Fe plaques on the root surface, thus decreasing As accumulation by rice plant. In addition, NH_4 in the rhizosphere can decrease pH, while NO_3 has an opposite effect (Srivastava et al., 2019). The effect of N supply on the response of rice seedlings to As stress (25 μM) was monitored for 7 days. It was observed that, compared to the 7-day treatment alone, low N + As led to improvement in root length of lateral and adventitious roots as well as their root density in significantly, while the average length of high nitrogen (HN) + As continued to decrease. Treatment with low nitrogen (LN) + As caused a significant decrease in root As concentration (0.848 mg kg^{-1} DW), while with As alone treatment root As content (1.434 mg kg^{-1} DW) did not decrease significantly. In contrast, the shoot As content (0.0068 mg kg^{-1} DW) treated with HN + As increased significantly compared to As alone (0.0034 μg g^{-1} DW) (Srivastava et al., 2019). Therefore, fertilizing plants with N source can play an important role in the migration of As in soil rhizosphere and its absorption in rice plants. Under As stress, the influence of N supplementation to rice plants remains relatively unexplored, and further experimentation is needed.

6. Conclusions and future outlook

This review covered a broad spectrum of factors and processes playing key role in As biogeochemical cycling and uptake by rice in paddy soils under flooded conditions. Many factors affect As bioavailability and translocation in rice shoot and grain. Arsenic enters the rice plants via phosphate transport proteins as As(V) and through aquaporins (Si) channels as As(III). The oxidative stress due to reactive oxygen species (ROS) production after exposure to As in rice plants can be counteracted by the production and complexation with compounds rich in thiol such as glutathione and PC. Although thiol complexation leads to the sequestration of As in vacuoles within root cells, while depending on the genotype and soil conditions a considerable amount of As can be transported in rice tissues.

Use of optimal amount of nutrients can effectively decrease As translocation to rice grain and shoot. Under neutral to acidic soil conditions, Fe can immobilize and sequester As in more efficient way, while in alkaline to neutral conditions Mn has a greater impact in immobilizing As. The above discussion indicates that use of organic amendments might be preferable over Fe-based materials because of their ability to supply nutrients for rice crop growth, improve soil physical, biological and chemical health, eco-friendly nature, cost-effectiveness, and abundant availability.

High application of P fertilizers may increase As uptake by rice grain due to the fact that phosphate supplementation could increase competition for soil minerals adsorption sites between As(V) and phosphate. Arsenic and Fe plaques are promoted by the application of S on the root surface that may decrease the As uptake through formation of FeS nanoparticles or As(III)-sulfide stable precipitates. Importantly, it is imperative to differentiate the effect of natural processes and anthropogenic activities on the release of As, and their impacts on accumulation in rice growing in and surrounding these areas. The following aspects should be considered in future work:

- Genotypic differences in rice are crucial to determine against As tolerance and accumulation by rice plants. Thus, fundamental knowledge on effect of phosphate, Si and Se as amendment to reduce As uptake by rice in contrasting rice varieties, i.e., coarse and fine varieties.
- Since, As concentrations in pore water and rice plant rhizosphere may change with rice varieties under the influence of various organic and inorganic materials, and phosphate, Fe-based, biochar, nano-gypsum and $Si(OH)_4$ generalizations should not be made.
- The role of magnetic biochar, nano-biochar and biomaterials needs to be explored in paddy soils along with the effect on As uptake and nutritional status of plants.
- It is important to quantify As losses and role of microbial processes in biomethylation and volatilization of As species and its possible effect on micro-fauna and flora.
- Arsenic contaminated coupled climate change stresses must be evaluated using various climate change attributes in laboratory scale experiments and field trials. This will allow us to identify potential rice varieties for implementation in food security, climate change, and environmental contamination remediation mitigation programs.
- In addition, extreme weather events can change the dynamics of As in paddy fields. The environmental conditions must be examined to understand the As mobility and release under paddy environments.
- By providing the nutrients needed for the growth of microorganisms, biostimulation can reduce As toxicity in the rice field environment due to the improvement of microorganism-mediated processes including As immobilization as As(III)-sulfide stable phases, As(III) oxidation, biomethylation and volatilization.
- The use of unexplored organic modifiers such as vermicompost, biosorbents on agricultural paddy soils, and how they can improve the physical, chemical and biological fertility of the soil. Root exudates affect the structure and function of microbial combinations, and then mediate various biochemical transformations of As in the root zone, including redox reactions and chemical forms.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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