LAND POLLUTION (G HETTIARACHCHI, SECTION EDITOR)

# Arsenic Contamination in Soil and Sediment in India: Sources, Effects, and Remediation

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Abstract Arsenic contamination is turning out to be a major problem these days with its area coverage and the number of people affected directly or indirectly. Now, the level of the contaminant has spread over the soil and sediments from groundwater and other natural sources. Arsenic poisoning in groundwater events is familiar to the world, but the consequences of soil contamination are still unrevealed to the community, specially the people of contaminated counties. Arsenic is a serious instantaneous concern for the people and other life forms regarding the poisoning through crops and vegetables. Many remediation technologies that mainly include physical, chemical, and a few biological methods have been evolved with time to check its effects. The physical and chemical methods for this purpose are often inefficient and/or very expensive, mainly limited to application in aqueous systems, and produce toxic sludge, which again becomes a matter of concern. But bioremediation relies on the fact that biological

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organisms have the ability to degrade, detoxify, and even accumulate harmful chemicals and offers attractive perspectives for biomonitoring (via biosensors), treatment of wastewater, and the recycling of polluted soils.

Keywords Arsenic  $\cdot$  Soil contamination  $\cdot$  Toxicity  $\cdot$  Bioremediation

# Introduction

Arsenic (As) is one of the toxic compounds which pose a high risk to large human populations. Although it had been historically used as a drug to treat skin infections, and beautification, it was also used for human murder [1, 34]. The contamination of arsenic in South Asian groundwater aquifers was first reported in the mid-1990s [13], and since then, a lot of work had been done for the past two decades. The utilization of these groundwater sources for irrigation and for drinking affected the various forms of life including humans, cattle, and crops [108••, 109••, 111], even causing death [61]. Earlier reports had shown that more than 200 million people across 70 countries are affected from As-contaminated groundwater [117, 118]. Yu et al. [135] found that per year there will be 1.2 million cases of health manifestations in Bangladesh only due to arsenic poisoning.

The riverine deposits of Bengal Delta Plains (BDP) formed the fertile plains of the three rivers Ganges Brahmaputra, and Mehgna. From the western to the northeastern borders of BDP, it have the Indian Plate, Chota Nagpur Plateau, Shilong Plateau, and Naga Lusai orogenic belt, making them intensely neotectonic [110]. The sediment deposition in these regions is also influenced by the regional vegetation and climate. Thus, the sources and distribution of As in these plains are largely controlled by the distribution of organic matter, oxic-anoxic conditions, and indigenous microbial flora [9, 36, 48••, 53, 62]. The organic content of the soil had always been considered as an important component of the global carbon pool [71]. Two different types of sedimentary depositions are found in the BDP region. The first type is the gray micaceous Holocene sand depositions which are mainly linked with shallow aquifers, contaminated with high levels of As, known as gray sand aquifers (GSA) [5]. The second type is the brown sand aquifers. These are deep Pleistocene depositions, which, though vulnerable to As contamination, are considered safe for the time as the As levels are  $<50 \mu g/1$  [18, 19, 100, 101]. The release of As is, however, a more complex process and involves many different parameters. Thus, a combined multidisciplinary study is needed including microbiological, geochemical, hydrological, and mineralogical approaches to identify the major factors controlling these processes in the BDP regions. However, many such large-scale studies had been done till date by many research groups across the globe, in finding out the source, distribution, and remediation ways to solve this problem, resulting in a large and growing literature. This review gives a brief overview of these works, covering most of the studies reported in the past 5 years, focusing on characteristics, As toxicity, sources and distribution of As, factors affecting As availability and mobility, factors affecting As transformation, effect of As on living forms, awareness, role of NGOs, and remediation.

# Characteristics

# Physical Characteristics

Arsenic (As) as a metalloid has three allotropes, gray, yellow, and black [89]. Naturally found arsenic has one stable isotope  $^{75}$ As [47]. However, many other isotopes have been synthesized. The compounds of As are similar to those of phosphorus (P), and they belong to the same group in the periodic table. Arsenic occurs primarily in two oxidation states, the trivalent state arsenite As(III) and the pentavalent state arsenate As(V). Other than these, As is also present in -3 state in arsenides which are alloy-like intermetallic compounds.

## Forms of Arsenic in Soil

Arsenic is present in soil in inorganic and organic forms. The inorganic forms of arsenic are more prevalent than the organic forms. The inorganic components majorly include the mineral forms, whereas the organic arsenic form is mostly found in living organisms due to arsenic consumption.

## Inorganic Forms

The major concentration of soil As is present in mineral forms. Arsenic has more than 300 mineral forms, including arsenates, sulfides, sulfosalts, arsenites, arsenides, native elements, and metal alloys. The major mineral forms of As are detailed in Table 1. Among these, sulfide (e.g., arsenopyrite, pyrite, loellingite, realgar) and arsenate minerals (e.g., scorodite, beudantite, yukonite) are the most common soil-bound mineral forms [72, 112]. The other minerals are generated during weathering.

## Organoarsenic Compounds

Inorganic arsenic, when it enters the food chain, gets methylated and forms less toxic organic forms like monomethylarsine (MMA), dimethylarsine (DMA), and trimethylarsine (TMA; Fig. 1; [72]). Other than these, many other organic forms were developed during World War I like lewisite, adamsite, and cacodylic acid [41, 50, 115]. Some forms of organic arsenic, such as arsenobetaine or arsenocholine, are found in seafood [61].

## Arsenic Toxicity

The toxicity of various forms of arsenic strongly depends on their oxidative states and chemical structures [83] The inorganic forms of As present in soil, when taken up and transported through the food chain, turn out to be toxic, affecting various life forms. Among the two oxidation states As(III) and As(V), the arsenite form As(V) is less toxic and mostly

Table 1 Major mineral forms of Arsenic

Mineral group	Mineral name	Formula
Oxide of arsenite	Arsenolite	As <sub>2</sub> O <sub>3</sub>
	Claudetite	As <sub>2</sub> O <sub>3</sub>
Oxide of arsenate	Arsenic pentoxide	As <sub>2</sub> O <sub>5</sub>
Fe-arsenate	Arseniosiderite	Ca <sub>2</sub> Fe <sub>3</sub> O <sub>2</sub> (AsO <sub>4</sub> ) <sub>3</sub> ·3H <sub>2</sub> O
	Parasymplesite	Fe <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O
	Pharmacosiderite	K[Fe <sub>4</sub> (OH) <sub>4</sub> (AsO <sub>4</sub> ) <sub>3</sub> ]·6.5H <sub>2</sub> O
	Scorodite	FeAsO <sub>4</sub> ·2H <sub>2</sub> O
	Symplesite	Fe <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O
	Yukonite	Ca <sub>7</sub> Fe <sub>12</sub> (AsO <sub>4</sub> ) <sub>10</sub> (OH) <sub>20</sub> ·15H <sub>2</sub> O
Fe sulfoarsenates	Beudantite	PbFe <sub>3</sub> (AsO <sub>4</sub> )(SO <sub>4</sub> )(OH) <sub>6</sub>
	Tooeleite	Fe <sub>6</sub> (AsO <sub>4</sub> ) <sub>4</sub> (SO <sub>4</sub> )(OH) <sub>4</sub> ·4H <sub>2</sub> O
	Zýkaite	Fe4(AsO4)3(SO4)(OH)·15H2O
Ca-Mg arsenates	Hörnesite	Mg <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O
	Pharmacolite	Ca(HAsO <sub>4</sub> )·2H <sub>2</sub> O
Other metal arsenates	Annabergite	Ni <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O
	Erythrite	Co <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O
	Köttigite	Zn <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O
	Mimetite	Pb <sub>5</sub> (AsO <sub>4</sub> ) <sub>3</sub> Cl

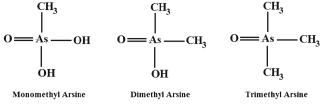


Fig. 1 Major organoarsenic compounds

present in immobile mineral forms, whereas the As(III) form is more toxic and gets mobilized into water and enters living cells. Thus, the lethal dose ( $LD_{50}$ ) of As(III) is also very less (15–42 mg/kg) compared to As(V) (20–800 mg/kg) [69]. As(III) binds with the sulfahydryl group of proteins, causing various clinical manifestations discussed later.

Arsenic, unlike other heavy metals like mercury (Hg) and lead (Pb), shows greater toxicity in inorganic forms than in organic forms. Among the organic arsenic forms, MMA and DMA are more toxic than TMA. Thus, the  $LD_{50}$  value for MMA (1.8 g/kg) and DMA (1.2 g/kg) is lower than that of TMA (10 g/kg; [83]).

## Sources and Distribution of Arsenic

The prevalent form of arsenic, which is found to contaminate aquifers around the globe, is the one that is present naturally in the environment [104, 111]. A variety of studies have been done over the past few decades to track factors which control As mobilization which ends up in groundwater, but these studies are debated till date [31, 54, 84, 97, 104]. One of the well-accepted mechanisms states that release of As occurs when Fe oxides are reductively dissolved, an effect prevalent in sedimentary environments [14–16, 31, 39, 87, 88, 97, 104, 114].

The amount of arsenic in the groundwater of Bengal Delta Plain (West Bengal, India and Bangladesh) ranges from 50 to 3200  $\mu$ g/l, a value much greater than national drinking water standards which is 50  $\mu$ g/l for India and Bangladesh and also surpasses the level set by WHO which is 10  $\mu$ g/l [14–16, 130].

Arsenic can be introduced in the environment either by natural processes (such as during atmospheric emissions or when naturally occurring minerals rich in arsenic are desorbed and dissolved) or by anthropogenic actions (such as mining, combustion of fossil fuels, metal extraction processes, timber preservatives, etc.) [14–16]. Broken-down grains rich in iron and arsenic adsorbed authigenic secondary oxide/hydroxide precipitates, termed as arsenic traps, might be one of the reasons of high arsenic occurrence in groundwater of BDP [14–16, 37, 80, 81]. Fe(III) present on the surface of BDP sediments may adsorb Fe(II) during iron oxide reduction. Fe(II), thus adsorbed, reduces As(V) to As(III). This reaction is responsible for the entry of toxic and labile As(III) in groundwater of BDP [14, 15].

Arsine is a very toxic gaseous As-containing compound [32, 72] formed in a highly reducing environment [72, 106]. A decrease in pH favors arsine formation. In anoxic conditions, arsine is liberated from marshy soil and swampy surfaces along with monomethylarsine, dimethylarsine, and trimethylarsine [38, 72]. Oxidation leads to conversion of such gases to aqueous phases of As(V)-bearing compounds [72, 122, 123].

In the atmosphere, As is prominently present as particles. Volcanic eruptions, wind mobilization, marine aerosols, and industrial exhausts give rise to As in the atmosphere. These particles settle on the ground when fossil fuels are burnt or when smelters are used and are termed as wet or dry deposition [72, 96]. Wet deposition is that portion of atmospheric As which is dissolved in rainwater [72].

A low concentration of As is seen in alluvial sands of Aspolluted aquifers in BDP (less than 10 mg/kg; [13, 72]). Weathering of As-bearing minerals leads to deposition of As in peaty, bog, acid sulfate, and various other natural soils. Average As content in such soils is less than 100 mg/kg. Industrial contaminants lead to the highest levels of As deposition in sediments and soils which may soar up to several thousands of milligrams per kilogram [72]. Fly ash produced by coal combustion leads to As deposition in the Indian subcontinent [72, 91].

# Geogenic Arsenic Distribution Across India

Weathering of the Himalayas leads to development of aquifers in sands settled on Pleistocene sediments. Sand aquifers are separated from overlying silts rich in organic matter by a palaeosol rich in clay. Arsenic-contaminated groundwater intersects boreholes containing gray sands which have undergone reductive dissolution. Sands present at greater depths become As deficient as clay-rich palaeosol separates organic matter necessary for reductive dissolution from the sands [103]. Flow of groundwater leads to frontal movement of redox and dissolved organic matter zones which in turn is thought to cause As discharge from deeper sands contained in such boreholes [62, 63, 85]. This model proposes the likelihood of As pollution caused by the presence of organic matter and sedimentary structure of aquifers [27, 29, 72].

In India, parts of many states have been identified as As contaminated in the case of soil and sediments, which are shown in Fig. 2 [25, 26, 91, 97, 106, 115, 116]. Arsenic mobilization in the Bengal Basin can occur by discharge of arsenic into alluvial sediments by oxidation of arsenic-containing pyrite [74, 75, 99], displacement of anions of As present in aquifer sedimentary minerals by phosphate anions used in fertilizers which are applied on the soil surface [5, 6, 100, 101], and discharge of arsenic in anoxic conditions by reduction of iron oxyhydroxide during sediment burial [14–16, 80, 81, 87, 88, 100, 101].

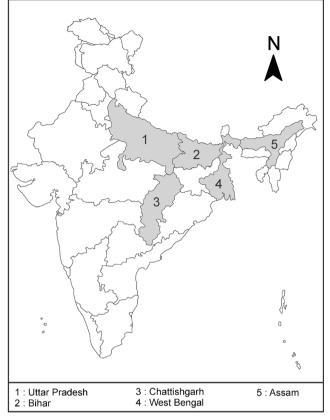


Fig. 2 Map of India showing states with As contamination in soil and sediments

## Factors Affecting Arsenic Availability and Mobility

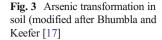
Bioavailability of As in soil is powerfully influenced by the chemical and physical characteristics of soils together with the character of minerals and clay content, organic matter, texture, pH and Eh, cation-exchange capability (CEC), and presence and concentration of oxides and hydroxides of metals, Al, Mn, etc. [62, 63, 79, 107••]. Both As(III) and As(V) are powerfully adsorbed to hydrous oxides of metal, Mn, and Al in acid soils whereas Ca oxides in alkaline soils to a lesser extent sorb anionic As species [132, 133].

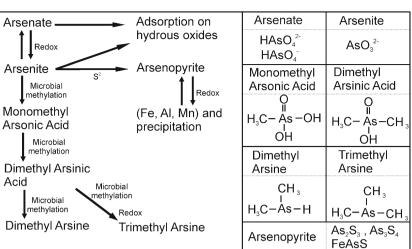
Generally, fine-grained soils limit the quality of each As species than coarse-grained soils due to the presence of many minerals and organic constituents in them [30]. However, considering other aspects, such as if the concentrations of iron and aluminum hydrous oxides within the soil are low, As tends to be mobile. Additionally, the presence of a less-soluble mineral part and ionic forms that are powerfully adsorbed to soil particles or co-precipitated with different minerals are responsible for the reduction in bioavailability of arsenic from soil [79]. Apart from these factors, studies also recommend that bioavailability of arsenic in contaminated soils is also influenced by aging and sequestration [122].

The main sorbent for As(III) and As(V) is iron oxyhydroxides (FeOOH). As(V) gets more strongly attached

to iron oxyhydroxides as compared to As(III). Clay or clayey soil contains more FeOOH when compared to sandy soil, and therefore, clayey soil has more arsenic. Clayey soils are less toxic than sandy soils because in clayey soils arsenic is strongly adsorbed. In an anaerobic condition under microbial action or reduction conditions, arsenic bound to iron oxyhydroxides is freely released. There are bacteria that are capable of reducing Fe(III) as well as oxidizing Fe(II). It is also reported that reduction of arsenic from V to III state has taken place without desorption from iron oxyhydroxides [118, 119]. In aerobic conditions, iron oxyhydroxides become insoluble and therefore there is lesser release of arsenic. The arsenic and iron relation is dynamic in nature and plays an important role in aging of the paddy field. Amorphous iron oxides have strong affinity for arsenic.

Also in soils, as in water, the chemical behavior of As is in many ways similar to that of phosphorus (P), especially in aerated systems, where As(V) ions generally resemble the orthophosphate ion closely [128]. However, under conditions normally encountered in soils, As is more mobile than P and unlike P can undergo changes in its valency [113-115]. Generally, As does not follow the typical behavior of other metal contaminants. For instance, it is highly soluble in neutral to alkaline pH (6.6-7.8) whereas most of the heavy metals are more mobile under acidic conditions. However, As can also be moderately soluble under acidic conditions. For this reason, its chemistry is more complex in soils than many other pollutants. Arsenic forms a variety of inorganic and organic compounds in soils [127] and is present as an inorganic species, either As(V) or As(III) [79]. The forms present in soils depend on the type and amounts of sorbing components of the soil, pH, and redox potential. Under oxic soil conditions (Eh> 200 mV; pH 5–8), As is commonly present in the +5 oxidation state. However, As(III) is the predominant form under reducing conditions [77-79]. Both As(V) and As(III) species can undergo chemical and or microbial oxidation-reduction and methylation reactions in soils and sediments and can adsorb on hydrous oxides of Fe, Al, and Mn ([17]; Fig. 3). The most important natural attenuation process known for As(III) compounds is precipitation as As sulfide  $(As_2S_3)$ . As(III) is more toxic and mobile in soils than As(V), and methylated species such as monomethylarsonic acid [MMAA, CH<sub>3</sub>AsO(OH)<sub>2</sub>] and dimethylarsinic acid [DMAA, [(CH<sub>3</sub>)<sub>2</sub>AsO(OH)] are also mobile [14, 15]. However, these methylated forms are volatile and unstable under oxidizing conditions and are cycled back into the soil environment in inorganic forms [34]. Ferric hydroxide [Fe(OH)<sub>3</sub>] plays an important role in controlling the As concentration in soils. Both As(V) and As(III) are adsorbed onto the surface of Fe(OH)<sub>3</sub>, but the adsorption of As(V) is much higher than that of As(III) [14, 15]. In general, highly oxic soils sorb three times more As(V) than soils containing small amounts of oxic minerals [113, 114]. The presence of iron, aluminum, and calcium compounds is the most





important factor in controlling the fixation in soil [18]. The sorptive capacity of a soil for an ion is a function of its surface area and hence its clay content; this explains why As is more mobile in sandy soil than fine-textured soils [128].

## **Factors Affecting the Arsenic Transformation**

## Soil Factor

Within the soil, the nature and proportion of the constituents determine the arsenic behavior. It is seen that inorganic arsenic species have high affinity for hydrous oxides of Fe, Mn, and Al in acid soils whereas calcium oxides in alkaline soils to a lesser extent adsorb anionic As species [132, 133]. And thus, the anionic species are more bioavailable to crops grown in alkaline soils [44]. However, if the concentrations of iron and aluminum hydrous oxides in the soil are low, As tends to be mobile. Reduced bioavailability in soil is thought to be primarily a function of the presence of a less-soluble mineral phase and ionic forms that are strongly adsorbed to soil particles or co-precipitated with other minerals. Factors that influence the adsorption capacity of soils influence the bioavailability and subsequent mobility of As in soils. The studies suggest that bioavailability in contaminated soils is also influenced by aging and sequestration [122, 123]. Soil pH was the most important soil property affecting the decrease in bioaccessibility on aging [134].

## Adsorption and Desorption

Adsorption and desorption processes are the principal factors affecting the transport, degradation, and biological availability of compounds in soil [113]. Hayes and Traina [56] reported that As(V) was strongly adsorbed on metal oxides and formed relatively insoluble precipitates with Fe and therefore became

less bioavailable. They also reported that, at higher pH, As(III) sorbed more weakly than As(V) to metal oxides and therefore became bioavailable [79, 85]. In summary, soil conditions that promote precipitation or adsorption also tend to reduce the mobility and bioavailability of As. Numerous studies have demonstrated that the plant uptake of As is greatly influenced by its form in soil [22, 24, 77, 78, 121]. The presence of other ions such as Fe and P also affects As availability and phytotoxicity [45].

# Microbial Processes

The microbial flora of a niche majorly influences the oxidation status and availability of As. Though microbes are widely reported for oxidation, reduction, methylation, and demethylation of As, the oxidation of reduced As species has been less widely studied except a few studies [23, 48••, 90] than the processes of microbial reduction of As.

Microbial methylation of As by common bacterial species in soil has been reported by a number of authors [12]. Methanogenic bacterial genera had been widely reported for methylation of As to different forms such as MMA and DMA [82, 98]. However, DMA is very unstable and quickly oxidizes under aerobic conditions [34]. The methyl forms of As undergo volatilization and thus have more toxicity than the nonmethylated forms [12].

## Effect of Arsenic on Living Forms

Arsenic(V) is a chemical analog of phosphate that can disrupt at least some phosphate-dependent aspects of metabolism in all living forms. Arsenic(V) can be translocated across cellular membranes by phosphate transport proteins, leading to imbalances in phosphate supply [42]. In general, during phosphorylation reactions, arsenic(V) competes with phosphate and forms unstable adducts. For example, during the formation of ATP, arsenic binds to ADP forming As(V)-ADP, and hence, the cycle will be a futile one; this decreases the ability of the cell to produce ATP and carry out their metabolism. Oxidative carbon metabolism, amino acid and protein relationships, and nitrogen and sulfur assimilation pathways are also impacted by arsenic exposure [42]. The effects of arsenic on different living forms are discussed in the following paragraphs.

## Effects of Arsenic on Plants

Arsenic, being a non-essential element, is not required for the growth of living organisms, with the exception of a newly discovered bacterium that replaces phosphorus with As for many cellular functions [131]. There are cases where the terrestrial plants may accumulate As by root uptake from soil or by absorption of airborne As deposited on the leaves [125]. In general, plants growing in natural soil contain low levels of As (<3.6 mg/kg [46]). At a higher concentration, arsenic is toxic to most plants. Arsenate is the dominant species in soils, and its similarity to phosphate allows it to compete for the same uptake carriers in the root plasmalemma. It interferes with metabolic processes and inhibits plant growth and development through arsenic-induced phytotoxicity [77, 78].

The toxicity symptoms may include inhibition of seed germination [2], decrease in plant height [2, 24, 67, 77, 78], depressed tillering [70], reduction in root growth and some necrosis (unprogrammed cell/living tissue death; [2]), decrease in shoot growth [33], and lower fruit and grain yield [2, 3, 24, 70] and sometimes lead to death [10, 77, 78, 136] discolored and stunted roots, withered and yellow leaves [74], and reductions in chlorophyll and protein contents, and in photosynthetic capacity [77, 78]. However, little is known about the effect of arsenic on photosynthesis, the basis of the plant bio-chemical system. As almost all of the abovementioned adverse physiological and agronomical effects of arsenic are related to the basic photochemical reaction in rice plants [135, 136], photosynthesis, it is important to measure chlorophyll a and b, the major photosynthetic pigments, contents in rice leaves to justify their correlations with rice growth and yield.

Paddy rice is more susceptible to As accumulation than other cereals because of the high mobility of arsenic under a flooded condition [73]. A previous study performed in wheat plants reported a decrease in the amylolytic activity due to arsenic toxicity. *Pteris vittata* commonly known as brake fern is the first species known as a hyperaccumulator for As. Brake fern can also hyperaccumulate arsenic, deriving from insoluble forms up to three to six times greater than the As concentration in soil, whereas the bioaccumulation of arsenic in aquatic organisms occurs primarily in algae and lower invertebrates [58, 73].

#### Effect on Human Health

The risk of human exposure to soil As has greatly increased in the last two decades mainly due to the expansion of residential areas into former agricultural land [76]. This has affected human health very adversely. Chronic exposure to As causes many clinical manifestations of which cutaneous lesions are the highest reported [35, 130]. These arsenic-associated clinical manifestations include melanosis (hyperpigmentation), keratosis, and leukomelanosis (hypopigmentation). Other than causing superficial cuticular diseases, As is also a well-known carcinogen, causing skin, lung, bladder, liver, and kidney cancers [66, 86]. The other health manifestations include carotid atherosclerosis [65, 75], ischemic heart disease [4], and impaired cognitive abilities and motor functions [52]. Arsenic also affects the hormonal regulations via the retinoic acid, thyroid hormone, and estrogen receptors [11].

#### Effect of Arsenic on Animals

Arsenic-contaminated food and water are the channels through which this metalloid enters a mammalian body. There are instances where derivatives of arsonic acid are fed to domestic livestock to enhance body growth and inhibit any oncoming disease [40]. Arsenic poisoning in homeothermal animals is implicated by high mortality in the course of 2-3 days [40, 84, 105]. Other symptoms of arsenic poisoning include abdominal pain, weakness, salivation, nausea, and death in the ultimate scenario. Autopsy of such affected individuals show a reddened edematous gastric and intestinal mucosa, yellowing of the liver, and edema in the lungs. Other effects include necrosis of mucosal epithelia and degeneration of the renal tube and gastrointestinal capillaries. Prolonged exposure to arsenic contamination leads to depression, dehydration, anorexia, frequent urination, and imbalance in body temperature [40, 84, 105]. Cutaneous arsenic exposure leads to drying up and deadening of the skin [40, 84]. Arsenic inhalation in rodents led to nasal flow discharge and eye irritation [40, 60].

Lab animals showed symptoms of PNS abnormalities, anemia, leukopenia, melanosis, cardiac malfunction, and liver damage. Surprisingly, these symptoms vanished when exposure got refrained [40, 94, 95]. Acute and sub-acute symptoms demarcate arsenic poisoning whereas chronic poisoning is rare [40, 84, 132]. This is because of immediate follow-up of detoxification and excretion processes [40, 132]. External uptaking of arsenic compounds affects the spleen, kidney, liver, and lungs; these compounds can then mobilize to ectodermal tissues such as nails and hair as these tissues are rich in sulfur-containing proteins [40].

Domestic animals are also widely affected by arsenic poisoning. A classic example is the death of around a hundred cattle due to overdose of arsenic trioxide which was applied externally to prevent lice infestation. Autopsy reports suggested subcutaneous edema and hemorrhage in the intestine, kidney, and epidermis [40, 102]. In a study in Bangladesh, cattle poisoned with arsenicals showed symptoms of restlessness, diarrhea, instability in walking, convulsions, panting, and salivation [40]. Such symptoms were followed by their death within the next 12–36 h. Autopsy reports showed hemorrhage in the submucosa of the gastrointestinal tract [40] and tissue deposits greater than 10 mg/kg fresh weight of the liver and kidney [40, 120]. It is presumed that cattle have an enhanced attraction towards weeds sprayed with an arsenic-containing weed killer because of the salty taste of arsenic compounds [40, 105].

## Remediation

Arsenic present in different forms in soil and sediments is the major contaminant of groundwater in the Indo-Gangetic plain. Since a large population is dependent on groundwater as source of potable drinking water, remediation or treatment of this water before consumption is one of the major requirements. Many remediation technologies have been developed for the removing of arsenic from groundwater mainly. These mostly include the physical- or chemical-based methods and a few bioremediation methods as described below.

Physical or Chemical Remediation

## Oxidation

Addition of chemical oxidants causes conversion of toxic mobile As(III) to a form less toxic and immobile As(V). As(III) can be oxidized by a number of chemicals such as gaseous chlorine, permanganate, hydrogen peroxide, and Fenton's reagent ( $H_2O_2/Fe^{2+}$ ) [68]. Direct ultraviolet (UV) radiationbased oxidation of arsenite in the presence of sulfite as catalyte [49], or ferric iron [42, 43] or citrate (EAWAG, 1999), is also done.

#### Precipitation

The dissolved arsenic can be removed by forming lowsolubility solid minerals such as calcium arsenate [59]. The precipitate of such low-solubility solid minerals can be removed through sedimentation and filtration. Addition of coagulants causing floc formation can also be utilized to remove arsenic by co-precipitation. This is accompanied by filtration.

## Adsorption and Ion Exchange

Minerals having a strong affinity for arsenic, such as iron and aluminum hydroxide, strongly adsorb arsenic ions onto their solid surfaces [49]. After sorption, water can be purified by gravity settling and filtration.

#### **Biological Remediation**

Although biological remediation is a slower process, it is more environment friendly [126]. Recent studies have shown that bioremediation of arsenic in soil can be accomplished using plants and different microbes [7, 8, 129].

## Bioremediation by Plants

Phytoremediation is accomplished by using different types of green plants [36, 51]. The various mechanisms of phytoremediation are given below:

*Phytoextraction* Few species of plants, while growing on contaminated sediments, accumulate metal arsenic into their roots [20] and translocate them to the surface. The main limiting factors for phytoextraction are the following:

- · Bioavailability of arsenic around the rhizosphere
- Rate of uptake of arsenic by roots
- Rate of loading of arsenic into xylem/translocation to shoots
- Cellular tolerance to arsenic

*Phytostabilization* Some plants decrease the volume of water percolating through the sediment matrix and thus act as a barrier, preventing contamination of soil [99]. Phytostabilization can also occur through sorption, complexation, or metal valence reduction and serves as a useful tool in mining land areas [99].

*Rhizofiltration* While capillary water is being absorbed through the root xylem, the heavy metals, if present in the water, are also being taken up. This process is called rhizofiltration. Rhizofiltration mainly functions in remediation of extracted groundwater, subsurface or surface water, and wastewater with low concentrations of contaminant.

*Phytovolatilization* Plants take up the contaminants from groundwater through their roots, pass them through xylem vessels and volatilize them, and release them during transpiration. This process is called phytovolatilization [57].

# Microbial Bioremediation

Cost-effective and environment-friendly ways can be found using various bioremediation processes involving microbes. Microbial processes such as As oxidation and reduction, mobilizes and immobilizes through sorption, biomethylation, complexation, and can also bioremediate different forms of arsenic [21], among which microbial oxidation of arsenite As(III) to arsenate As(V) is predicted to be one of the most promising ways to immobilize the dissolved arsenic [48••, 90]. These processes are supported by in situ available organic carbon and other inorganic components like Fe, Mn, and S concentrations. The various methods of microbial bioremediation of arsenic are discussed in the following paragraphs.

*Biostimulation* The process of biostimulation includes introduction of stimulants into a system to induce the indigenous microbial flora to enhance their bioremediation mechanism [33]. These stimulants can be water, nutrients, oxygen, S, etc. [1]. Many other stimulants like nitrogen (N), phosphorus (P), sodium acetate (Na-CH<sub>3</sub>COOH), and ethanol had also been tested.

*Bioaugmentation* The process of bioaugmentation includes introduction of laboratory-tested microbial strains into an artificial system. In such process, small-scale laboratory-based researches of rates of bioremediation are tested, and then, the efficient groups of microbes are introduced to a large-scale system. Genetically engineered, highly efficient stains can also be developed and used. Recent studies on arseniteoxidizing bacteria in the Indian subcontinent give evidences of the presence of such microbes that can be isolated and utilized for bioaugmentation [48••].

## Awareness and Role of NGOs

The presence of arsenic in groundwater was first detected in 1993, but the depth of the magnitude and the extent of the problem were not taken cared of till 1997. The detailed study for this was not performed thoroughly by many agencies involved due to the lack of testing [25, 26, 113]. In India many states including West Bengal, Jharkhand, Bihar, and Uttar Pradesh in the flood plain of Ganga River; Assam and Manipur in the flood plain of Brahamaputra and Imphal; and Rajnandgaon village in Chhattisgarh state have so far been identified in 2008 to be exposed to arsenic through drinking arsenic-contaminated groundwater [92, 93]. Many initiatives were taken that led to the development of strategies to reduce As exposure from drinking water. In this regard, both national and international agencies started working to provide safe drinking water to the affected rural population, by remediation of the As-contaminated groundwater, changing the sources of drinking water by targeting deeper safe aquifers, or supplying treated surface water [7, 8]. In the course, awareness-building measures suggested mainly include conducting sensitization workshops at state, district, block, and Gram Panchayat levels by the Public Health Engineering Department (PHED) of the state and imparting of training to local clubs, NGOs, etc.

Noticeably, all the efforts are being put on the groundwater arsenic contamination but not on the soil contamination that is proving to turn out to be the major source now.

Governmental organizations, NGOs, and research and international organizations are trying to mitigate the problem by providing various options like dug wells, pond sand filters, household As-removal filters, and rainwater harvesting. None of these options are found suitable and accepted by the people because of their limitations such as cost, maintenance, disposal of sludge, non-availability, etc. [53]. Use of surface water is also not suitable because it is highly polluted with pathogens, agrochemicals, and industrial wastes. Few community-based As-removal plants are installed by NGOs and international organizations in some highly As-affected areas. Supplies of drinking water from such large-scale As-removal or water treatment plants through pipelines in rural areas are costly and difficult.

#### **Measures to Reduce As Exposure**

In order to check the effects of arsenic, long-term actions are required from mining, metal smelting and refining, combustion of low-grade coal, pesticide use, and timber treatment. Most importantly, in these As affected with high groundwater arsenic, action is needed to reduce the intake of arsenic from drinking water and food [64, 130], which may include provision of safe drinking water, regular monitoring of the arsenic level in groundwater, awareness among dependents, designing and developing arsenic removal technologies, discriminating between high-arsenic and low-arsenic water sources by painting hand pumps in different colors (e.g., red and green) [124], or adapting sediment-coloring tools [63, 64]. In addition to these, monitor high-risk populations for early signs of arsenic poisoning, usually skin problems and other effects [55, 56].

# Conclusions

Arsenic (As) is one of the toxic compounds present in soil in inorganic and organic forms and has been identified to pose a high risk to large human populations. The sources and distribution of As are largely controlled by many factors like the distribution of organic matter, oxic-anoxic conditions, indigenous microbial flora, etc. Arsenic can be introduced in the environment either by natural processes or by anthropogenic actions. The toxicity of various forms of arsenic strongly depends on their oxidative states and chemical structures.

The introduction of arsenic to the deeper sands of boreholes can be due to the frontal movement of redox and dissolved organic matter in the groundwater from shallow aquifers [28]. In soil, the bioavailability of As is mainly influenced by the chemical and physical characteristics of soils together with the character of minerals and clay content, organic matter, texture, pH and Eh, cation-exchange capability (CEC), and presence and concentration of oxides and hydroxides of metals, Al, Mn, etc.

The effects of arsenic(V) can be seen by its analogy with phosphate that can disrupt at least some phosphate-dependent aspects of metabolism in all living forms. It can also be translocated across cellular membranes by phosphate transport proteins, leading to imbalances in phosphate supply. In the case of plants, As is toxic at higher concentrations. It mainly interferes with metabolic processes and inhibits plant growth and development through arsenic-induced phytotoxicity. In the case of animals, the chronic exposure causes many clinical manifestations of which cutaneous lesions are the highest reported; arsenic is also a well-known carcinogen, causing skin, lung, bladder, liver, and kidney cancers.

Currently, the As present in soil and sediments in different forms is the major contaminant in the Indo-Gangetic plain due to a large population residing in the area. Thus, there is a need for a routine assessment of arsenic on a regular basis. Apart from that, the remediation or treatment measures in these areas are of major concern at this point. For this, governmental organizations, NGOs, and research and international organizations along with the participation of the local people are required considering the present scenario.

#### **Compliance with Ethics Guidelines**

**Conflict of Interest** Anamika Shrivastava, Devanita Ghosh, Ayusman Das, and Suatapa Bose declare that they have no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

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