

## A Review: Effect of Physicochemical and Hydro geochemical Factors on Arsenic Sedimentation and Mobilization

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**ABSTRACT:** The adsorption behavior of arsenic was studied experimentally for a wide pH range from 2 to 13.7. It was found that adsorption of  $As^{3+}$  occurs at pH 8, while adsorption of  $As^{5+}$  occurs at pH 4. At low pH like 2, the adsorption of  $As^{5+}$  and  $As^{3+}$  is very low, while at high pH like 13.5 the adsorption rate increases. The order of adsorption of different constituents of As and aqueous media is  $AsO_3^- > AsO_3^{3-} > H_2AsO_3^- > H_2O > H_3AsO_3$  for As(III) and  $AsO_4^{3-} > HAsO_4^- > H_2AsO_4^- > H_3AsO_4 > H_2O$  for As(V). Arsenic acid ( $H_3AsO_3$ ) adsorbed at pH 8, while arsenic acid ( $H_3AsO_4$ ) adsorbed at pH 4. Arsenic forms in  $H_3AsO_3$  and  $H_3AsO_4$  are  $As^{3+}$  and  $As^{5+}$ , respectively. Conjugate bases of  $H_3AsO_3$  and  $H_3AsO_4$  are  $AsO_3^-$  and  $AsO_4^{3-}$ , respectively, which show increasing rates of adsorption at high pH like 13.5. Arsenic adsorption in aqueous media is aided by ions such as  $Cl^-$ ,  $HCO_3^-$ ,  $H_2PO_4^-$ ,  $SiO_3^{2-}$ , and  $PO_4^{3-}$ . Cations such as  $Na^+$ ,  $Mg^{2+}$ , and  $Ca^{2+}$  increase the rate of adsorption of  $As^{3+}$  and  $As^{5+}$  contents in groundwater, and this rate would be further increased with an increase in the concentration of these ions. The reason for the great adsorption of As contents is due to the electrostatic forces created on adsorbents by the origin of charge on the adsorbent surface after the adsorption of cations.

**Keywords:** Arsenic adsorption, Arsenate ( $As^{5+}$ ) and Arsenite ( $As^{3+}$ ), Co-existing ions, Electro positivity, Electrostatic force.

### INTRODUCTION

At present, arsenic toxin and its widespread distribution are a major concern throughout the world, including in India. The present study reveals a high concentration of As (range 50–986 g/l) in ground water affected by vast areas of the north-eastern states of India. This high concentration of As in ground water becomes a significant topic when compared to the admissible limits quoted by the WHO (World Health Organization) and BIS (Bureau of Indian Norms) (10 g/l and 50 g/l, respectively). In this paper, we are going to explain the effects of physicochemical and hydrogeochemical factors on arsenic sedimentation and mobilization in the Ganga-Meghna-Brahmaputra Basin. Arsenate ( $As^{5+}$ )

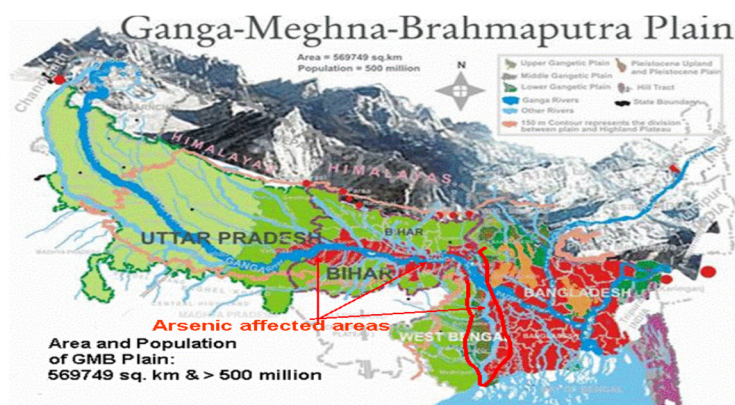
and Arsenite ( $As^{3+}$ ) are two types of oxidation states of arsenic in the groundwater of affected areas. These two oxidation reactions are interconverted by the oxidation of  $As^{3+}$  into  $As^{5+}$  and the reduction of  $As^{5+}$  into  $As^{3+}$ . Arsenic also exists in another form, i.e., organic form, and it's formed by the biomethylation of arsenic.

Groundwater Arsenic contamination in Eastern Gangetic River Basin reported worldwide in recent studies which causes many adverse effects not only for drinking but also for irrigation purpose (Azam, 2013). During monsoon time arsenic goes into groundwater by disintegration of Fe(III)-oxy hydroxides and in pre monsoon time arsenic reversibly adsorb on Fe(III)-oxyhydroxides (Azam, 2020). A definite relationship

exists between the behavior of arsenic and rainfall intensity (Azam, 2020) Arsenic mostly exists in two types of oxidation states which are arsenate ( $\text{As}^{5+}$ ) and Arsenite ( $\text{As}^{3+}$ ) (Azam and Kunwar 2018). Many factors control arsenic concentration and transport in groundwater, which include: Redox potential (Eh), adsorption/ desorption, precipitation/dissolution, Arsenic speciation, pH, presence and concentration of

competing ions, biological transformation, etc (Azam and Kunwar 2018).

Studies on As contamination in groundwater in different states of India, such as West Bengal, Jharkhand, Bihar, and Uttar Pradesh, show the flood tide plain of the Ganga River, while Assam and Manipur show the flood tide plains of the Brahmaputra and Imphal gutters.



**Fig. 1.** Brahmaputra-Ganga-Meghna Basin showing Arsenic affected groundwater areas.

Arsenic enters the food chain through plants and animals and harms the environment and humans (Cullen and Reimer 1989). There are two types of arsenic contamination:

i. Spontaneous releases of arsenic from arsenic-rich groundwater (Smedley *et al.*, 2003) and ii. Human emissions (Zhang *et al.*, 2014) include mining and mine concentration (Wei and Zhou 1992), irrigation (Norra *et al.*, 2005), and pesticide use (Alam *et al.*, 2015), etc.

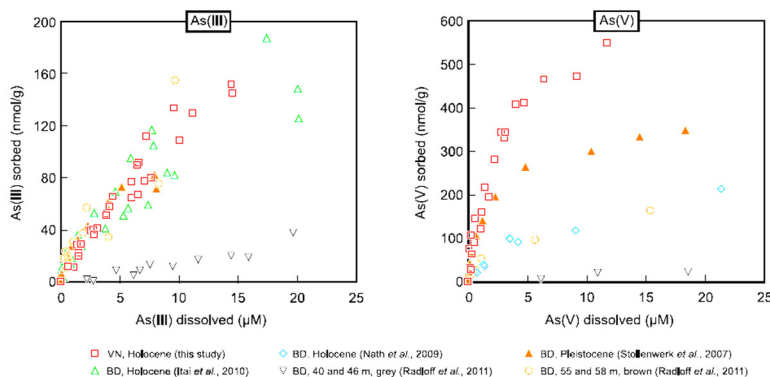
Local arsenic contamination is due to, as environmental conditions change, released as from mines and desorbed from minerals. Severe arsenic contamination occurs due to the discharge of arsenic-containing wastewater into underground rivers through pipes, fissures, gullies, and sinkholes (Zhang *et al.*, 2014). With the increase in population and industrial development, the seriousness of the drinking water problem becomes very important. It has been reported that a large number of water bodies in various parts of the world have AS concentrations above 50 g/L and have been identified as problematic (Smedley and Kinniburgh 2002). It has also been observed that the groundwater and rivers that spread across the Gangetic Plain in India are highly contaminated by arsenic. This condition of high AS causes a serious problem for the safety of aquatic ecosystems and the health of local populations (Ma *et al.*, 2003). Sprinkler irrigation has demonstrated to be a useful tool in minimizing the bioaccumulation of As and

Cd in rice grain of Carnise genotype cultivated in a soil heavily polluted by both elements (elemental concentration of 50 mg kg<sup>-1</sup> each) (Spanu, *et al.*, 2021). A significant release of As and NO<sub>3</sub> from sediments at shear stress levels typically measured in estuaries, Substantial release of these contaminants due to sediment resuspension does not hence require large storm events but occurs at shear stresses with magnitudes that occur regularly (Izaditame *et al.*, 2022).

**Mobilization Mechanisms of As.** It has been found that arsenic-contaminated groundwater was not mainly associated with arsenic-rich areas in source rocks in most of the regions studied. Two important factors were identified.

First, mobilizing arsenic from the solid or adsorbed phase to the groundwater occurs due to highly specific biogeochemical processes, and second, arsenic contents are retained in aquifers because mobilized arsenic accumulates and is not washed away (Smedley and Kinniburgh 2002). In other words, it can be said that the rate of arsenic released from the source must be rapid compared to the groundwater flushing rate. Arsenic can be transported in groundwater in a variety of ways.

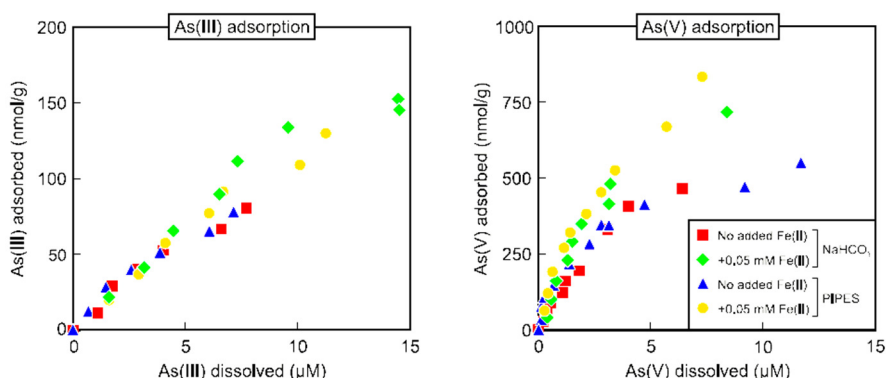
1. Alkaline and oxidizing conditions carrying out desorption of arsenic and reducing conditions results in the dissolution of arsenic
2. Reduction of the bond strength between arsenic and mineral surfaces (Smedley and Kinniburgh 2002).



**Fig. 2.** Distribution of As (III) & As (V) shown by various studies.

The main cause of groundwater contamination by arsenic in West Bengal is due to the oxidation of sulfide minerals (pyrite-FeS<sub>2</sub>); this has been strongly discussed by many researchers (Das *et al.*, 1995). This hypothesis explains that arsenic is released from sulfide minerals (arsenopyrite) in shallow aquifers by oxidation (Mandal *et al.*, 1998). Another reason is the lowering of

groundwater tables due to overexploitation of groundwater for irrigation purposes, which causes the release of arsenic. Present studies explain that desorption or dissolution of arsenic from iron oxide is a possible process for the regional distribution of arsenic in ground water (Smedley, 2004).



**Fig. 3.** Mobilization of As (III) & As (V) with Fe- redox process.

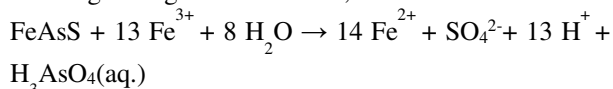
Another potentially important process to increase A/C in water is the dissolution of iron oxide itself under reducing conditions. Research also reveals that excessive water use for irrigation and fertilizer use caused the mobilization of phosphate from fertilizers below shallow aquifers and the desorption of As through anion exchange at reactive mineral surfaces.

To explain its occurrence, release, dissolution, sorption, mechanism, etc. in soil-water phases at local scale and to understand the chemistry of arsenic, many studies on groundwater arsenic modeling were carried out (NIH and CGWB 2001; Majumdar *et al.*, 2002; Michael and Voss 2008). However, there is still a lack of understanding of the chemistry of arsenic in geo-environmental and geochemical processes. Most of the modeling studies explain the following points:

(i) Groundwater arsenic sources are located in situ and in local patches, and their mobilization is determined by the use of groundwater regimes.

(ii) Careful management of aquifers allows long-term availability of arsenic-free groundwater without the risk of disturbing arsenic-contaminated zones.

(iii) development of unpolluted deep aquifers and freshwater bodies in combination with surface water sources to ensure arsenic-free water supplies for both drinking and agricultural needs;



Upon initiation of reducing conditions in the subsoil, this causes the dissolution of as-rich iron oxyhydroxide (FeOOH). Inorganic As species are mainly retained in the solid phase through interactions with the FeOOH coating on soil particles under oxidizing conditions, and in the presence of Fe, FeOOH coatings can dissolve when reducing conditions are initiated in such environments. Fermentation of subsoil peat releases organic molecules (such as acetate) to reduce FeOOH

dissolution, resulting in the release of  $\text{As}^{3+}$ ,  $\text{As}^{5+}$ , and  $\text{Fe}^{2+}$  present on such coatings. The chemical reaction is:  $8 \text{FeOOH-As(s)} + \text{CH}_3\text{COOH} + 14\text{H}_2\text{CO}_3 \rightarrow 8\text{Fe}^{2+} + \text{As(d)} + 16\text{HCO}_3^- + 12\text{H}_2\text{O}$

where  $\text{As(s)}$  is sorbed As and  $\text{As(d)}$  is dissolved As.

The application of fertilizers to subsurface soil causes the release of As sorbed to aquifer minerals by competitive exchange with phosphate ( $\text{H}_2\text{PO}_4^-$ ) ions that migrate into aquifers.

Excessive accumulation of in groundwater is the second mechanism involving the dissolution of  $\text{FeOOH}$  under reducing conditions and is considered to be the most probable reason for.

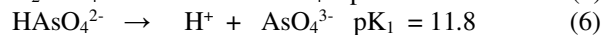
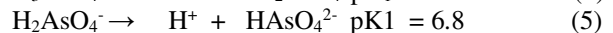
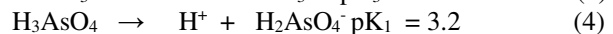
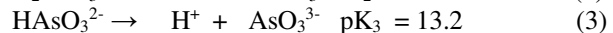
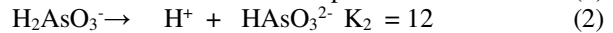
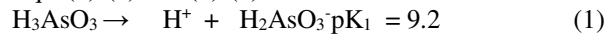
**Adsorption-desorption process of arsenic in sediment-water interface.** For water contamination, sediment is considered a buffer zone. Many different types of complex biological, chemical, and physical processes, such as adsorption, desorption, and/or aggregation or precipitation, occur at water interfaces in sediments (Zou, 2004). Under certain conditions, sediments thus accumulate pollutants and become sources of pollutants (Jin *et al.*, 2001). Arsenic aggregates with positive colloids in water, as it generally occurs as negative colloids ( $\text{Sb}_2\text{S}_3$ ,  $\text{H}_2\text{SiO}_3$ , etc.). Due to the adsorption process, arsenic deposits as sediments in water beds due to the rapid transformation from the aqueous to the solid phase (Anawar *et al.*, 2004; Rahman *et al.* 2006). Therefore, the sediment-water interface has a significant impact on arsenic migration through the adsorption-desorption process within. Through laboratory experiments and field observations, Chen and Li (1998); Gong *et al.* (2006) studied how, under hydraulic disturbance, the adsorption-desorption of arsenic at the sediment-water interface occurs. Research has explained that sediment and water are closely correlated.

The potential for arsenic contamination in surface waters is due to the increased release of arsenic into water from sediments. During the dry season, arsenic in sediments has a poor ability to resuspension. Therefore, little arsenic is transported downstream. During the wet season, flooding upsets the sediment and water balance, allowing arsenic to resuspension and downstream transport of pollution.

**Arsenic adsorption affected by hydrogeochemical characteristics and physicochemical properties of sediment.** Previous studies have found that adsorption capacity is affected by variations in pH and the charge of colloids. Existence of arsenic in sediments as suspended colloids and light behavior because the colloidal charge changes with changes in pH (Sadiq, 1997), it is extracted by aqueous alkaline solutions (Smedley *et al.*, 2002). According to Manning and Goldberg (1997), the adsorption capacity of arsenic in kaolinite reaches a maximum at pH 5.0 but decreases sharply above pH 6.5.

Anawar *et al.*, (2004) tested the release rate of As from sediment pore water and found that arsenic begins to desorb when pH reaches 9. Release capacity peaks when pH reaches 12. Johnston and Sarker (2007) rely on a double-diffusion model to calculate the pH dependence of the adsorption of arsenic acid ( $\text{As}^{5+}$ ) and arsenic acid ( $\text{As}^{3+}$ ) by hydrated iron oxide (HFO) and the Dzombak and Morel (1986). It is known that when the pH exceeds 8.5, the amount of arsenic adsorption drops sharply.

Because the distributions of As(III) and As(V) in water solution depend on the pH of the medium, the adsorption mechanism of Arsenic is more complex, as shown by Eqs. (1)-(3) and (4)-(6) at 25°C.

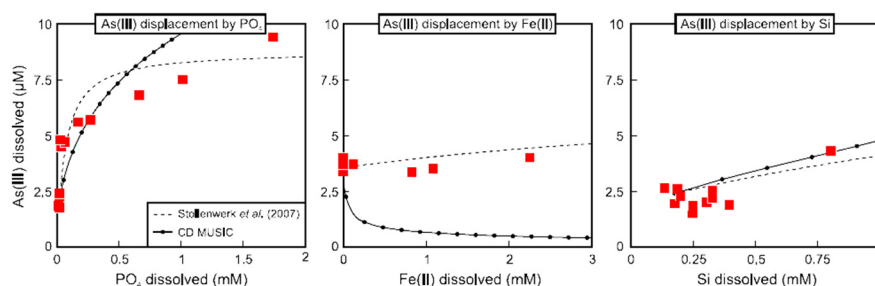


Therefore, different ways of arsenic species distribution on the surface due to different pH ranges

A study of the effect of pH change on the rate of adsorption shows that large amounts of As(V) are adsorbed on iron hydroxide when the pH is between 4 and 10, and more As(V) is released into the water as the pH increases. In addition, it was observed by Gupta and Chen (1978) that As(V) is readily adsorbed to aluminum hydroxide at pH 4–7, with strong adsorption occurring above pH 7.0. Between pH 4 and 9, adsorption kinetic behavior does not show any relevant change. The displayed result shows that acidic conditions promote the adsorption of As(V), while alkaline conditions promote the adsorption of As(III).

**Coexisting ions.** The presence of coexisting ions in water either suppresses or promotes the adsorption behavior of arsenic. Arsenic content in groundwater (Smith *et al.*, 2003; Smedley *et al.*, 2005). As a result, As(III) adsorption is inhibited by competitive effects between anions in water such as  $\text{SO}_4^{2-}$ ,  $\text{SiO}_3^{2-}$ ,  $\text{PO}_4^{3-}$ ,  $\text{HCO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{F}^-$ , and  $\text{HPO}_4^{2-}$ , and they could increase the adsorption of As(III) anions to varying degrees. With increased concentration of anions in water, the inhibitory effect becomes stronger, and it was observed that anions such as  $\text{Cl}^-$ ,  $\text{H}_2\text{PO}_4^-$ , and  $\text{SO}_4^{2-}$  have particularly strong inhibitory effects in this adsorption behavior, the adsorption of As(III) (Livesey and Huang 1981). Adsorption of As(V) (Goh and Lim 2005), However, for As(V), new adsorption sites are formed when ions such as  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ ,  $\text{F}^-$ , and  $\text{HCO}_3^-$  combine with the sediment. Adsorption of As(III) and As(V) is related to the presence of cations such as  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{Na}^+$  in water, and they increase adsorption rate with increasing cation concentration. This is due to the accumulation of cations on the surface of the adsorbent.





**Fig. 4.** Arsenic adsorption affected by the presence of Coexisting ions in water.

The presence of arsenic enhances the electropositivity behavior of an adsorbent surface and directly enhances the electrostatic force of a cation. This enhancement of electrostatic force has the effect of attracting and binding more arsenic anions on the surface of the adsorbent; therefore (Smith *et al.*, 2002), binding more arsenic anions on the surface of the adsorbent increases with increasing cation concentration. Schmedley *et al.* (2003) As a result, high As concentrations in groundwater are due to high concentrations of NH<sub>4</sub><sup>+</sup>, HCO<sub>3</sub><sup>-</sup>, Fe, Mn, and P. The fact that the highest As concentrations are likely to be found in groundwater indicates that SO<sub>4</sub><sup>2-</sup>-reduction occurs under strongly reducing conditions and As mobilization occurs with low SO<sub>4</sub><sup>2-</sup>-concentrations. In addition, Seddique *et al.* (2008) examined major aquatic species with high arsenic Groundwater is Ca-Mg-HCO<sub>3</sub>-chemicals Characteristics and hydrological conditions of arsenic-rich groundwater in Sonargaon (Bangladesh, Middle East)

**Physico-chemical properties of sediments.** The adsorption of As(III) and As(V) by sediments indicates the presence of oxides (iron, aluminum, manganese, etc.) and organics in water bodies. As(III) and As(V) in the particles form insoluble arsenic compounds, which enhance the ability of sediments to adsorb arsenic. Which are influenced by complex surface reactions that can adsorb oxides of iron, aluminum, and manganese, and 80% of the arsenic in water precipitates in sediments containing high arsenic ferrate, as observed by Moenke (1956). At pH = 5.1, Oscarson *et al.* (1983); Wang *et al.* (2010) observed strong adsorption to As(V) and As(III) with interaction of iron manganese compounds.

In addition, the distribution of medium and medium-high arsenic levels outside the estuary was studied by Li *et al.* (2011). Adsorption amounts of As(III) and As(V) in extra-estuary sediments occur due to high levels of organic matter and metals such as iron, manganese, and aluminum in estuary sediments. in the meantime, and also observed that the adsorption amount of As(III) to the sediments is larger than that of As(V), Aside from that, Uddin and Lundberg (1998) performed the correlation analysis. There is a positive correlation with both Al and Fe and the content of the main chemical elements and arsenic in the sediment.

Functional groups include carboxyl, phenolic hydroxyls,

alcohol extract hydroxyls, ethers, amides, and organic particles that cooperate with arsenic, giving the sediment a greater adsorption capacity so it can adsorb large amounts of arsenic (Wang and Mulligan 2006). In addition (Ding *et al.*, 2007), arsenic migration and accumulation on colloidal iron oxide occur because the organic acid colloid has a large specific surface area, adsorption capacity, and a protective colloidal effect against colloidal iron oxide (Wang and Mulligan 2006). Most studies have found that the amount of arsenic adsorbed on sediment is directly proportional to the organic matter content of the sediment, and some studies suggest that the release of arsenic is carried out by the presence of organic matter on the sediment surface Yang *et al.* (2008). In the groundwater of the Hetao Plain migration, concentration and transformation of arsenic are facilitated by the presence of a reducing environment rich in high pH and organic matter. Because microorganisms produce a reducing environment by decomposing a substance that consumes oxygen organically, As a result, groundwater's high arsenic content is increased by the release of As from the sediments. Through geochemical studies of the arsenic environment in Hetao Plains of Inner Mongolia, organic matter is an important contributor to arsenic-rich groundwater and showed rich sedimentary environments, Tang *et al.* (1996).

## CONCLUSIONS

We have obtained many results for understanding the effects of physicochemical and hydrogeochemical factors on arsenic sedimentation and mobilization properties, and researchers have conducted numerous studies related to morphology. adsorption laws of arsenic in water. Promising conclusions:

1. The morphology and adsorption of arsenic are affected by the pH of aqueous media, and with increasing pH, arsenic adsorption decreases. but the optimum pH for arsenic adsorption gives conflicting results regarding it. Under optimal environmental conditions, the adsorption of As(III) and As(V) as a function of pH gives even more different and conflicting conclusions regarding it.
2. It was observed that cations facilitate the process while anions coexisting in water interfere with the arsenic adsorption process. The presence of coexisting ions in

water either suppresses or promotes the adsorption behavior of arsenic. Arsenic content in groundwater As a result, As(III) adsorption is inhibited by competitive effects between anions in water such as  $\text{SO}_4^{2-}$ ,  $\text{SiO}_3^{2-}$ ,  $\text{PO}_4^{3-}$ ,  $\text{HCO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{F}^-$ , and  $\text{HPO}_4^{2-}$ , and they could increase the adsorption of As(III) anions to varying degrees.

3. Minerals are positively correlated with arsenic content, and the Mn, Fe, and Al contents of the sediments facilitate the adsorption of arsenic. However, the adsorption of arsenic by organic matter remains a controversial issue.

4. Functional groups include carboxyl, phenolic hydroxyls, alcohol extract hydroxyls, ethers, and amides. Organic particles cooperate with arsenic, giving the sediment a greater adsorption capacity so it can adsorb large amounts of arsenic.

5. Presence of Arsenic enhances the electropositivity behavior of an adsorbent surface and directly enhances the electrostatic force of a cation. This enhancement of electrostatic force has the effect of attracting and binding more arsenic anions on the surface of the adsorbent.

6. Another potentially important process to increase As in water is the dissolution of iron oxide itself under reducing conditions. Research also reveals that excessive water use for irrigation and fertilizer use caused the mobilization of phosphate from fertilizers below shallow aquifers and the desorption of As through anion exchange at reactive mineral surfaces.

## FUTURE SCOPE

1. The presence of arsenic enhances the electropositivity behavior of an adsorbent surface and directly enhances the electrostatic force of a cation. This increase in electrostatic force has the effect of attracting and binding more arsenic anions on the adsorbent's surface.

2. Minerals are positively correlated with arsenic content, and the Mn, Fe, and Al contents of the sediments facilitate the adsorption of arsenic. However, the adsorption of arsenic by organic matter remains a controversial issue for further research..

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**Conflict of Interest.** None.

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