

Effects of Anions on Arsenic Adsorption with Iron Hydroxide-A Review

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In this review paper interaction of arsenic with iron hydroxide and effects of anions, such as silicate, phosphate and bicarbonate on arsenic adsorption by iron hydroxides, are discussed. Arsenic is strongly adsorbed on the surface sites of iron hydroxides through the formation of inner-sphere complexes. Interaction of arsenic along with silicate or phosphate in the suspension of iron hydroxides affects arsenic adsorption to iron hydroxides and decreases due to competitive adsorption of the anions. Especially phosphate and arsenic exhibit similar chemical behaviour and compete for the adsorption sites. Carbonate decreases the sorption capacity of arsenite As(III) on iron hydroxides. The affinity of the anions for the sorption on iron hydroxides decreases in the following order; arsenate As(V) > phosphate > As(III) > silicate > bicarbonate. The effect of phosphate, silicate and bicarbonate in separate solutions reduce As(III) adsorption at relatively low concentrations and low surface site coverage. While the anions have none or moderated effects on As(V) adsorption but As(V) adsorption is considerable reduced in multi-anions solution.

KEYWORD

Arsenite, Arsenate, Adsorption, Anion effect, Iron hydroxide, Water.

INTRODUCTION

Nine districts in West Bengal, India and 42 districts in Bangladesh have arsenic levels in groundwater above the World Health Organization maximum permissible limit of 50 µg /L. The area and population of the 42 districts in Bangladesh and the 9 districts in West Bengal are 92,106 km² and 79.9 million and 38,865 km² and 42.7 million, respectively (Chowdhury *et al.*, 2000). In Bangladesh and India, drinking water drawn from underground sources has been responsible for widespread arsenic poisoning affecting nearly 100 million people (Mandal *et al.*, 1996; Nickson *et al.*, 1998). In many remote villages, arsenic contaminated groundwater is the only viable source of drinking water and cost-effective arsenic removal technology is a bare necessity to provide safe drinking water. Besides the presence of an unacceptable level of arsenic,

the groundwater is otherwise quite fit for drinking (Sarkar *et al.*, 2005). Arsenic contamination of the ground water occurs by both natural processes, such as weathering of arsenic containing minerals and anthropogenic activities, such as uncontrolled industrial discharge from mining and metallurgical industries and application of organo-arsenical pesticides (Krishna *et al.*, 2001). Inorganic arsenic is predominantly present in natural waters. Arsenate As (V) and arsenite As (III) are primary forms of arsenic, as well as monomethylarsonic acid (MMAA) and dimethylarsinic acid (DMAA) in soils and natural waters (Ferguson and Gavis, 1972). As (III) is more mobile in groundwater and 25 to 60 times more toxic than As (V). As(V) is the predominate species in oxygenated waters (Christen, 2000)

High concentration of arsenic in water has caused symptoms of chronic arsenic poisoning in local populations of many countries, like India, Bangladesh, Taiwan, Mongolia, China, Japan, Poland, Hungary, Belgium, Chile, Argentina and North Mexico. Arsenic

in water is found above the maximum contaminant limit (MCL) set by the corresponding nations (Nemade *et al.*, 2007a; Jain and Ali, 2000). Manifestation of higher doses of inorganic arsenic compounds in the human body leads to the disease called arsenocosis. Arsenic is a carcinogen and its ingestion may deleteriously affect the gastrointestinal tract, cardiovascular system, central nervous system and diseases, like skin lesions, hyperkeratosis, and hyperpigmentation (Farrel *et al.*, 2001). Due to its toxic effects on human health, recently the USEPA has lowered the maximum contaminant level for arsenic in drinking water from 50 to 10 µg/L. Since As(III) removal is more difficult than As(V) removal and As(III) has a lower affinity than As(V) for iron hydroxides, the oxidation of As(III) to As(V) is required prior to treatment in order to enhance the extent of arsenic removal (Lenoble *et al.*, 2002). Oxidants can be used to oxidize As(III) to As(V). The oxidation of As(III) by oxygen is kinetically very slow, whereas the oxidation of As(III) is rapid when oxidants, such as free chlorine, permanganate, hypochlorite, ozone, and hydrogen peroxide, are used (Scott *et al.*, 1995). As(III) can also be oxidized both abiotically and biologically. Wilkie and Hering (1996) have studied hydrous Fe oxide, Raven *et al.* (1998) and Jessen *et al.* (2005) ferrihydrite, Sun and Doner (1996) goethite, Nikolaidis *et al.* (2003) zerovalent iron, Zhang and Itoh (2005) Fe oxide-loaded slag, Zeng (2003) silica-containing Fe oxide, electrocoagulation (Nemade and Chaudhari, 2006). These synthetic Fe-containing adsorbents had good affinities for both As(III) and As(V) due to their high specific surface area.

As(III) can also be oxidized biologically by bacteria isolated from soil. Recently As(III) is oxidized to As(V) by constructed soil filter (CSF) and subsequently co-precipitation of As(V) by iron complex (Nemade *et al.*, 2007b; Nemade, 2007). The most common technique for the removal of arsenic is co-precipitation with ferric salts. However, the removal of arsenic from Bangladesh groundwater is more difficult than the removal of

arsenic from US groundwater due to the elevated concentration of silicate and phosphate in Bangladesh groundwater (Meng *et al.*, 2001). The objective of the present paper is to recapitulate the interactions between arsenic and anions, such as silicate, phosphate and carbonate with iron oxyhydroxides and the combined effects of anions for arsenic adsorption onto iron hydroxides.

ADSORPTION MECHANISM OF ARSENIC AND IRON HYDROXIDES

Arsenic has a high adsorption affinity on iron oxides, such as amorphous iron hydroxide (Goldberg, 2002; Jain and Loeppert, 2000), ferrihydrite (Raven *et al.*, 1998), magnetite (Gimenez *et al.*, 2007) and goethite (Manning *et al.*, 1998; Sun and Doner, 1996). As(V) adsorption increases at low pH in the pH range of 4 to 9 while the adsorption of As(III) increases at high pH (Holm, 2002; Goldberg and Johnson, 2001). As(V) is more strongly adsorbed to iron hydroxides than As(III) (Bowell, 1994). On the other hand As(III) adsorption is greater than As(V) adsorption on iron hydroxides when the solution pH is more than 8. Raven *et al.* (1998) also found that ferrihydrite adsorbed greater amounts of As(III) than As(V) at pH 4.6 and 9.2 with an initial arsenic addition of 13.3 mol As/kg Fe. Oscarson *et al.* (1983) reported that As(III) adsorption was greater than As(V) adsorption on iron hydroxides after 12 hr of reaction at pH 7.

Roberts *et al.* (2004) investigated the application of Fe(II) instead of the usually applied Fe(III) which was shown to be advantageous, as oxidation of Fe(II) by dissolved oxygen causes partial oxidation of As(III) and iron(III) (hydr)oxides formed from Fe(II) have higher sorption capacities. In the presence of P and Si, Fe(II) leads to precipitation with more sorption sites for phosphate and arsenate than Fe(III).

Goldberg and Johnson (2001) showed that As(V) adsorption on iron hydroxides increases with increasing ionic strength, indicating that inner-sphere surface complexes are formed. They also found that As(III)

adsorption decreases on iron hydroxides when the ionic strength increases, indicating an outer-sphere adsorption mechanism. Based on microscopic experiments, these researchers concluded that As (III) adsorption on iron hydroxides formed both inner-sphere and outer-sphere surface complexes. Adsorption mechanism can be explained using spectroscopic techniques. Extended X-ray absorption fine structure (EXAFS) studies have provided evidence that As(V) forms an inner-sphere bidentate surface complexes on goethite (Fendorf *et al.*, 1997) and ferrihydrite (Waychunas *et al.*, 1993). Fourier transformed infrared (FTIR) spectroscopy (Sun and Doner, 1996) has investigated that As (V) forms inner-sphere surface complex with iron hydroxide sites in co-precipitated and adsorbed solids. Goldberg and Johnson (2001) observed inner-sphere surface complexes between As (V) and goethite with Raman spectroscopy and inner-sphere adsorption mechanism to goethite for As(III) with EXAFS and FTIR spectroscopy.

EFFECT OF SILICATE ON ARSENIC ADSORPTION WITH IRON HYDROXIDES

Silicon is a semi-metallic element, is the second most common element on earth after oxygen and can be found in nearly all natural waters (Holm, 2002). Silica has proved to be the most common and problematic interferant for arsenic removal. There is some debate as to the relationship, but it is clear that high silica levels, especially those above 20 ppm, result in dramatically reduced capacity for arsenic. All natural waters contain aqueous silica at typical concentrations of 1-20 mg/L as SiO₂ for surface water and 7-45 mg/L as SiO₂ for groundwater. Aqueous silica can sorb to the surfaces of iron oxides; therefore, it is expected to influence practical aspects of chemistry in environmental systems. The mechanisms by which aqueous silica can influence natural and engineered processes have not been completely defined but may include competition for iron sorption sites and colloid stabilization (Davis *et al.*, 2001). The iron oxide surface has a strong affinity for silicate.

Sigg and Stumm (1981) have reported that approximately 0.7 mmol/L of aqueous silicate can be adsorbed to 6 g/L goethite at an initial concentration of 0.8 mmol/L of silicate. Silicate can also affect the chemistry of iron hydroxides. Anderson and Benjamin (1985) found that ferrihydrite crystallized to goethite in less than 24 in the absence of silicate, while the presence of silicate inhibited crystallization of ferrihydrite for 1-2 week. Silicate inhibits coagulation and crystallite growth of iron hydroxides. The surface chemistry of Si-containing iron hydroxides also depends on the Si content. When the silicate: iron molar ratio increases from 0 to 0.35, the Pzc of the iron hydroxides containing silicate decreased from pH 8 to pH 4 approximately (Swedland *et al.*, 1999). In the presence of silicate, the crystallization and particle size of iron oxides can be impacted (Rushing *et al.*, 2003). Silicate sorption to iron hydroxides produces a highly negative surface charge, which leads to the creation of smaller iron particles from large particles. The iron concentration is less than 0.1 mg/L in water in contact with fresh iron hydroxides after passing through a 0.45 pore size filter, whereas a significant increase in iron concentration is observed after filtration of water in contact with aged iron hydroxides containing silicate (Davis *et al.*, 2001).

Silicate competes with arsenic for the sorption sites of iron hydroxides (Meng *et al.*, 1993). The adsorption of As(V) decreases with increasing pH and silicate concentration. Silicate increases iron mobilization and interferes with arsenic removal which is due to a decrease in the surface potential of the iron hydroxides due to adsorption of silicate. Meng *et al.* (2000) showed that the soluble iron concentration increased significantly from 47 to 2040 mg/L when the silicate concentration was 5 mg/L as Si and the pH was increased from 8.6 to 9.4. The rate of oxidation increases as the concentration of silica increases between pH range of 6.6 to 7.1. Rushing *et al.* (2003) have found that silica slows Fe(II) oxidation to Fe(III) at pH 6.5. More than 95% of the Fe(II)

is oxidized after 30 min in the absence of silicate, however, only 66% of the Fe(II) is oxidized after 30 min in the presence of silicate.

COMPETITION OF PHOSPHATE AND ARSENIC ON IRON HYDROXIDES

Phosphate occurs in some groundwater and concentration varies from 0.02 to 2.7 mg/L (Meng *et al.*, 2002). Also phosphate and AsO_4^{3-} has similar sorption behaviour. According to Manning and Goldberg (1996), phosphate can seriously interfere the arsenic sorption onto hydrous ferric oxide (HFO). The presence of 6.5 mg/L of phosphate can lower the arsenic adsorption capacity of iron oxide ore by 30-50% at pH 7.0 (Zhang *et al.*, 2005). Phosphate adsorption on iron hydroxides has been investigated using modeling and FTIR spectroscopy (Arai and Sparks, 2001). FTIR spectroscopy has demonstrated that phosphate forms an inner-sphere bidentate binuclear complex on iron hydroxides. The structure and particle size of the iron hydroxides can be affected by phosphate. Crystallite growth is slower in the presence of phosphate and reported the reduction of approximately 10% and 30% of phosphate sorption on iron hydroxides in the presence of 5 mg/L and 14 mg/L of silicate, respectively. The particle size of iron oxide suspension using photon correlation spectroscopy found that phosphate decreases particle size of iron hydroxides. He *et al.* (1996) used transmission electron microscopy (TEM) to investigate the phosphate effect on the colloid structure of iron hydroxides and found that aggregated iron hydroxides had a three-dimensional branched chain structure; however, the formation of the chain structure of iron hydroxides was inhibited in the presence of phosphate.

The interaction between phosphate and citrate on iron hydroxides and found that the presence of citrate decreased phosphate adsorption on iron hydroxides at pH below 7, whereas citrate adsorption decreases in the presence of phosphate on iron hydroxides over a wide pH range. The literature reported the competitive sorption of As(V)

and phosphate on iron oxide and found that the sorption of As(V) is reduced by increasing the initial P/As (V) ratio. At an initial P/As(V) ratio molar ratio of 1.5, approximately 72 % of P is adsorbed at an initial As(V)/P molar ratio of 1.5. The phosphate adsorption on iron hydroxides decreases in the presence of humic acid, magnesium, sulphate and fluoride at low pH whereas calcium increased phosphate adsorption at high pH. Phosphate and As(V) exhibit similar chemical behaviour and compete for specific adsorption sites. Many researchers have reported desorption of arsenic on iron hydroxides in the presence of phosphate (Jackson and Miller, 2000).

EFFECT OF CARBONATE AND SULPHATE ON ARSENIC ADSORPTION

Inorganic carbon is found in all natural waters. Meng *et al.* (2000) found negligible effect of carbonate on arsenic by ferric chloride in pH range of 4 to 9.5. Fuller *et al.* (1993) also reported that the presence of carbonate species had no effect on the adsorption of arsenic by ferric hydroxide. Carbonate can be encountered in all natural waters. Total carbonate is naturally present at concentration greater than 120 mg/L as C in some ground water (Holm *et al.*, 2002). Some researchers have measured carbonate adsorption on iron hydroxides. Van Geen *et al.* (1994) observed that the maximum adsorption of carbonate on iron hydroxides occurred at approximately pH 6. Increasing ionic strength decreases carbonate adsorption on iron. These results indicate the formation of weak carbonate surface complexes at the interface between water and iron hydroxides.

The adsorption mechanism of carbonate on iron oxide can be determined utilizing microscopic tests. Attenuate total reflectance Fourier Transformed Infrared (ATR-FTIR) data has shown that carbonate forms monodentate inner-sphere complexes in iron oxide. Van Gree *et al.* (2001) have observed that the presence of carbonate decreases chromate adsorption on iron hydroxides. In contrast, carbonate promotes the adsorp-

tion of selenate and sulphate in the pH range of 6 to 8 and also forms ternary surface complexes with U(VI) and Pb(II). Fuller *et al.* (1993) reported that the presence of carbonate had little effect on As(V) sorption in co-precipitation experiments. Wilkie and Hering (1996) reported that the addition of carbonate had a slight effect on As(V) adsorption at pH 9 and no effect on As(III) adsorption at pH 6. High arsenic concentrations in groundwater are correlated with high bicarbonate concentrations (Nickson *et al.*, 2000; Kim *et al.*, 2000). Appelo *et al.* (2002) have demonstrated using model calculations that carbonate in soil and groundwater reduces the sorption capacity of arsenic on iron hydroxides.

According to Meng *et al.* (2000), the removal of As(III) and As(V) by ferric chloride was not affected in pH range of 4 to 10 in presence of sulphate and no apparent change in arsenic removal was observed when concentration increased from 0 to 300 mg/L. According to Hering *et al.* (1996), the presence of 960 mg/L of sulphate has decreased the As removal efficiency to some extent but did not give any effect on As(V) removal efficiency by ferric chloride. The binding affinity of anions on the surface site of iron hydroxides can be used to describe the interactions between anion, and iron oxide. Liu *et al.* (2001) reported that As(V) had a stronger affinity than phosphate for iron hydroxides. Meng *et al.* (2002) calculated the binding affinity of the anions, such as phosphate, silicate, As(III), and As(V) on iron hydroxides. The affinity of the anions for iron oxide sites decreased in the following order arsenate > phosphate > arsenite > silicate at pH 6.8.

Meng *et al.* (2002) showed the combined effects of anions on As(III) and As(V) adsorption in multi-anions solutions. Simulated Bangladesh and West Bengal groundwater containing the same arsenic, iron, cation, and anion was used in their study. Overall As(III), adsorption decreased except in chloride solution. Silicate and bicarbonate concentrations were 160 and 550 times greater than As(III), respectively whereas the

binding affinity of As(III) was higher than silicate and bicarbonate, while bicarbonate, phosphate and silicate had none or moderate effects on As(V) adsorption. This was attributed to the strong affinity of As(V) on iron hydroxides. However, As(V) adsorption decreased dramatically in multi-anions solution containing phosphate. As a result of the increased surface site coverage, the As(V) adsorption decreased when phosphate coexisted with silicate and bicarbonate. The presence of silicate and phosphate in the suspension of iron hydroxides decreased the As(V) adsorption. Genc and Tjell (2003) studied the order of anion suppression on arsenate removal which was found to be phosphate > silicate > sulphate > bicarbonate on a molar basis.

CONCLUSION

Several researchers have studied the fundamental mechanisms of arsenic removal and the effects of anions for arsenic removal, by iron oxides. Recently combined effects as multi-anions have been documented. The main objective of this paper is to elucidate the mechanisms of arsenic adsorption on the surface of iron hydroxides and the effects of anions, such as silicate, phosphate and carbonate on the arsenic adsorption by iron hydroxides and benefit the design of more effective arsenic treatment processes and the development of more accurate models for predicting the transport of arsenic in aquifers. Combined effects of phosphate, silicate, and bicarbonate caused the high mobility of arsenic in natural water.

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