

Effect of Bicarbonate on Arsenic Removal by Coagulation

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In Bangladesh, groundwater is the primary source of potable water. But it contains high concentration of arsenic, as well as iron and manganese in many regions. Arsenic and iron removal plants are used in many such areas. Previous studies have demonstrated effectiveness of coagulation (e.g., using alum and ferric chloride) process for removal of arsenic from groundwater; effects of pH and phosphate on arsenic removal by coagulation are also well understood. This study evaluates removal of both As(III) and As(V) in the presence of bicarbonate/alkalinity. Results of laboratory batch experiments showed that during alum coagulation, removal of As(V) decreased significantly with the increase of alkalinity/bicarbonate due to competitive adsorption of As(V) and bicarbonate. On the other hand, alkalinity/bicarbonate did not have any significant effect on As(III) removal. At lower alkalinity/bicarbonate concentration, removal of As(V) has been found to be much higher than As(III) for similar alum doses; but as bicarbonate concentration increased, As(III) removal overtakes As(V) removal due to poor removal of As(V) in the presence of high bicarbonate concentration. During ferric chloride coagulation, very low alkalinity/bicarbonate (e.g., less than 50 mg/l as CaCO₃) resulted in poor removal of both As(III) and As(V) due to formation of insufficient ferric hydroxide flocs. Bicarbonate/alkalinity of groundwater should therefore be carefully considered while designing coagulation based arsenic removal system.

Field of Research: Water Treatment

Keywords: Arsenic; Alkalinity; Bicarbonate; Coagulation

1. Introduction

In Bangladesh, groundwater extracted through tube wells are the major source of water supply throughout the country. However, large numbers of wells exceed permissible limits for As, as well as manganese, and iron (BGS & DPHE 2000; Hasan & Ali 2010; Unicef & BBS 2009). On a national scale, 25.5 percent of water sources have As concentration exceeding the WHO guideline value of 10 ppb, and 12.5 percent have As concentration exceeding national standard of 50 ppb (Unicef & BBS 2015). Among the common As removal processes (Maji et al. 2007), coagulation using iron salts (e.g., naturally occurring iron, ferric chloride, ferrous sulfate) or aluminum salts (e.g., alum) is one of the promising methods (Edwards 1994; Maji et al. 2007). The adsorption of arsenic onto iron/aluminum floc can be affected by the commonly found ions in water, such as phosphate, silicate,

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sulphate, nitrate and bicarbonate (Guan et al. 2009; Hering et al. 1996; Laky & Lickso 2011; Roberts et al. 2011; Ruiping et al. 2007; Ware 2013,), which compete with As for adsorption onto iron/aluminum solids. Although effects of a number of anions (especially phosphate) on As adsorption have been well-studied, there is limited data on effect of bicarbonate on As adsorption/removal. Some field observations (ITN-BUET 2011) and studies (Habib 2013), however, suggest possible effect of bicarbonate on As adsorption/removal.

Among various technologies used for removal of arsenic from water, the most common are adsorption and co-precipitation with iron (e.g., in arsenic-iron removal plants, AIRPs) and coagulation with iron salt (e.g., ferric chloride) or alum. Arsenic Iron Removal Plants (AIRPs) are typically based on the principles of aeration (which promotes oxidation of dissolved ferrous iron into ferric hydroxide precipitates), adsorption and co-precipitation of arsenic, and filtration (to remove arsenic-rich ferric hydroxide flocs from water)(Concern Worldwide & ITN-BUET 2016).

Two parameters are considered important for effective As removal in AIRPs. One is the presence of sufficient iron in groundwater (e.g., > 5 mg/l) and the other is relatively high alkalinity, which promotes oxidation of Fe(II) (Concern Worldwide & ITN-BUET 2016). However, bicarbonate ions could compete with As for adsorption sites, thereby affecting arsenic removal. This study evaluates the effect of bicarbonate on removal of arsenic during coagulation by alum and ferric chloride.

Most treatment processes (e.g., coagulation, adsorptive filtration) are more capable of removing ionic contaminants. Therefore, the non-ionized trivalent arsenic (in neutral pH range) is more difficult to remove from the water than the ionized pentavalent form of arsenic (Kartinen & Martin 1995). Batch experiments have been conducted before to investigate the combined effects of phosphate, silicate, and bicarbonate on the removal of arsenic from Bangladesh groundwater and simulated groundwater by iron hydroxides. The apparent adsorption constants indicated that the affinity of anions for iron hydroxide sites decreased in the following order - arsenate> phosphate>arsenite> silicate> bicarbonate. Phosphate, silicate, and bicarbonate decreased the removal of As even at relatively low concentrations and low surface site coverage (Meng et al. 2002). However, there is lack of study on effect of bicarbonate alone on removal of arsenic. From the limited research which has been conducted, it has been observed that bicarbonate lowers the adsorption of As(V) on iron oxide mineral but may not have prominent effect on As(III)(Suzuki 2011).

Overall, from previous studies it can be inferred that bicarbonate ions in groundwater inhibit iron oxide, aluminum hydroxide etc. to act as arsenic adsorbents. On the other hand, bicarbonate has been found to promote formation of ferric hydroxide flocs, thereby promoting arsenic removal by adsorption onto iron flocs. However, knowledge about the effects of bicarbonate is limited and needs to be investigated systematically. Therefore, the main objective of this study was to evaluate the effect of alkalinity (bicarbonate) on removal of arsenic during alum and ferric chloride coagulation. It has been found that higher concentration of bicarbonate could significantly reduce removal of arsenate onto aluminum hydroxide flocs during alum coagulation; on the other hand, low bicarbonate concentration could reduce removal of both arsenite and arsenate during ferric chloride coagulation due to poor formation

of iron flocs. Section 1 of this paper describes the background and objectives of this research, and Section 2 describes the methodology followed in the research. Section 3 presents the important results obtained from this research. Finally, Section 4 presents the major conclusions.

2. Methodology

2.1 Chemicals and Stock Solutions

Reagent grade chemicals were used for all experiments and distilled water was used to prepare stock solutions. Arsenic trioxide (As_2O_3) was used to prepare As(III) stock solution and sodium hydrogen arsenate heptahydrate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) was used to prepare As(V) stock solution. Aluminum sulphate ($\text{Al}_2(\text{SO}_4)_3 \cdot 16\text{H}_2\text{O}$) was used for preparing alum stock solution, and ferric chloride (FeCl_3) salt was used for preparing ferric chloride stock solution. Bicarbonate stock solution was prepared by dissolving sodium bicarbonate (NaHCO_3) salt in distilled water.

2.2 Batch Experiments for Arsenic Removal

Sample preparation: Batch experiments in the laboratory were carried out by using water at 0.01 M and 0.10 M ionic strengths for avoiding the possible interference of other ions except bicarbonate; it was therefore possible to analyze effect of only bicarbonate ion on removal/adsorption of arsenic. Sodium chloride (NaCl) was used as background electrolyte. Water was spiked with arsenic (both arsenite and arsenate) stock solutions to attain desired level (300 $\mu\text{g/l}$) of initial arsenic concentration. Bicarbonate concentration was varied by using NaHCO_3 stock solution. Batch experiments were carried out to assess removal of both arsenite and arsenate in the presence of bicarbonate by alum coagulation and ferric chloride coagulation.

For alum coagulation bicarbonate concentration was varied from 100 mg/l to 500 mg/l; for ferric chloride coagulation bicarbonate concentration was varied from 50 mg/l to 500 mg/l. Alum dose was varied from 20 to 100 mg/l, and ferric chloride dose was varied from 0 to 15 mg/l. Trial experiments were carried out to determine the required dose of acid/base (0.5N sulfuric acid or 0.5N sodium hydroxide) to attain desired pH of the solution (pH 6.5 to 7.5) under the experimental conditions. Then the determined doses of acid/base were added to the solution prior to the addition of coagulant in the batch experiments. For alum coagulation experiments, the final pH of the solution varied from 6.5-7.5 and for ferric chloride coagulation experiments, it varied from 7.0-8.0.

Coagulation experiments in Jar Test Apparatus: After addition of required quantity of acid/base, bicarbonate and coagulant (alum or ferric chloride) were added to beakers containing 0.01 M or 0.10 M NaCl solutions. The samples were then subjected to 1 minute of rapid mixing at 45 rpm followed by 14 minutes of slow mixing at 25 rpm in a jar test apparatus. The flocs were allowed to settle down for 15 minutes. Then the final pH was measured and samples were collected for analysis of arsenic and alkalinity. Arsenic concentration was measured with an Atomic Absorption Spectrophotometer (Shimadzu, AA 6800). The concentration of bicarbonate was calculated from measured pH and alkalinity. Arsenic removal by

alum/ferric chloride was estimated by subtracting final As concentration from initial As concentration and expressing it as percentage of initial As concentration.

3. Results and Discussions

3.1 Effect of Bicarbonate on Arsenic Removal by Alum Coagulation

As(V) removal: Table 1 shows experimental data on removal of As(V), present at an initial concentration of 300 µg/l, by alum coagulation from 0.10 M NaCl solution for different alum doses (20, 50 and 100 mg/l) and at different alkalinities. It clearly shows that as alkalinity increases As(V) removal decreases. For example, As(V) removal decreased from 56.2% to 38.9% as alkalinity increased from 84 to 400 mg/l (as CaCO₃). The effect of alkalinity appears to be more pronounced at lower alum dose. The decrease in arsenic removal appears to be due to competitive adsorption of As(V), present mainly as HAsO₄²⁻ and H₂AsO₄⁻ in the pH range at which the experiments were carried out, and bicarbonate (HCO₃⁻) which is the principal contributor of alkalinity in the pH range of the experiments.

Figure 1 shows removal of As(V), present at an initial concentration of 300 µg/l, as a function of calculated bicarbonate concentration. It shows that As(V) removal decreases significantly as bicarbonate concentration increases.

Table 1: Removal of As(V) (Initial Concentration 300 µg/l) by Alum Coagulation from 0.10 M NaCl Solution

(a) Alum Dose = 20 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
84	7.5	131.3	56.2
220	7.3	144.4	51.8
400	7.3	183.2	38.9
(b) Alum Dose = 50 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
105	7.7	52.8	82.4
269	8.0	71.5	76.2
418	7.9	270.5	9.8
(c) Alum Dose = 100 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
63	7.0	22.6	92.4
166	7.3	24.6	91.8
291	7.9	75	75

Table 2 shows experimental data on removal of As(V), present at an initial concentration of 300 µg/l, by alum coagulation from 0.01 M NaCl solution for two

different alum doses (20 and 100 mg/l) and at different alkalinities. It shows that at 0.01 M NaCl, alkalinity did not have a significant effect on As(V) removal. Figure 2 shows As(V) removal from 0.01 M NaCl solution as a function of bicarbonate concentration. It also shows no significant effect of bicarbonate concentration on As(V) removal at 0.01 M ionic strength.

Figure 1: Removal of As (V) (Initial Concentration 300 µg/L) by Alum Coagulation from 0.10 M NaCl Solution as a Function of Bicarbonate Concentration

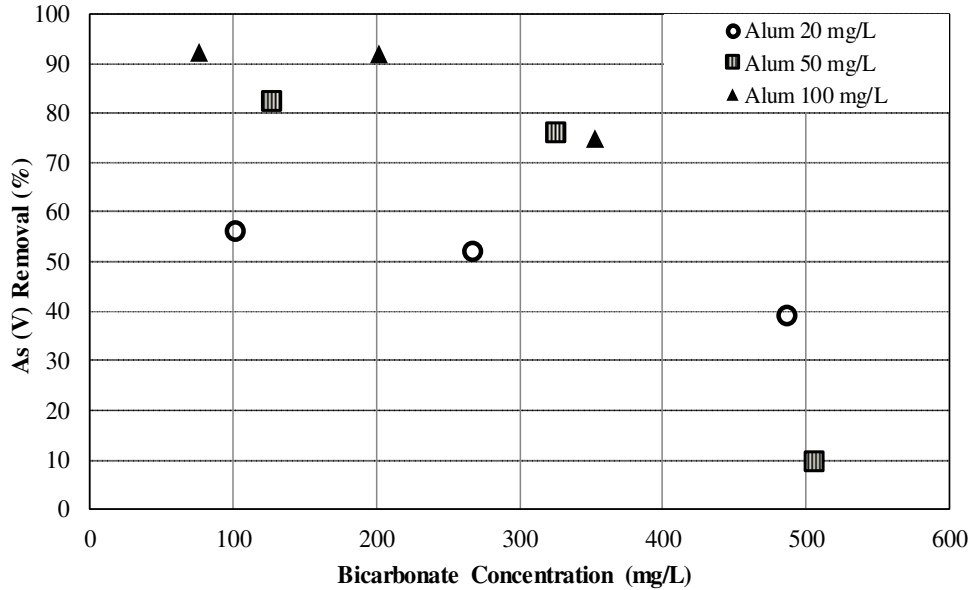
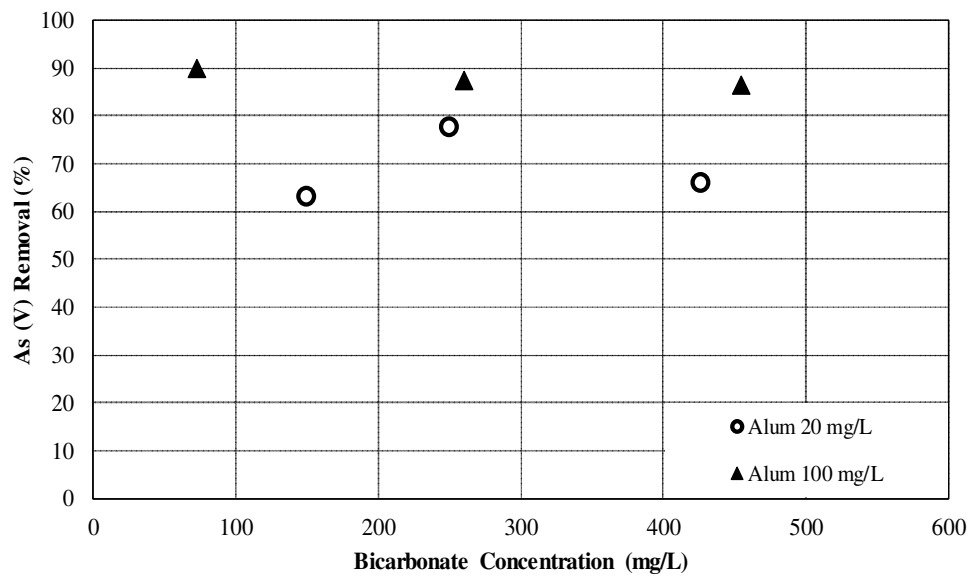


Table 2: Removal of As(V) (Initial concentration 300 µg/l) by Alum Coagulation from 0.01 M NaCl Solution

(a) Alum Dose = 20 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
124	7.7	110.6	63.1
206	7.4	67.4	77.5
351	7.5	101.9	66.0
(b) Alum Dose = 100 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
60	7.3	31	89.7
214	7.5	38	87.3
376	7.9	40.6	86.5

Figure 2: Removal of As (V) (Initial Concentration 300 µg/l) by Alum Coagulation from 0.01 M NaCl Solution as a Function of Bicarbonate Concentration.



As(III) removal: Table 3 shows experimental data on removal of As(III), present at an initial concentration of 300 µg/l, by alum coagulation from 0.10 M NaCl solution at different alkalinities and for different alum doses (20, 50 and 100 mg/l). It shows that unlike As(V) (see Table 1), alkalinity did not have any effect on removal of As(III) from water. Removal of As(III), as a function of bicarbonate concentration for different alum doses is shown in Figure 3. From this figure, it is clear that bicarbonate (HCO_3^-) did not compete with As(III) which was present primarily as H_3AsO_3 in the pH range at which the experiments were carried out.

Table 3: Removal of As(III) (Initial Concentration 300 µg/l) by Alum Coagulation from 0.10 M NaCl

(a) Alum Dose = 20 mg/l			
Alkalinity (mg/l as CaCO ₃)	pH	As Residual (µg/l)	As Removal (%)
90	7.1	171.8	42.7
248	7.3	171.8	42.7
385	7.5	177.9	40.7
(b) Alum Dose = 50 mg/l			
Alkalinity (mg/l as CaCO ₃)	pH	As Residual (µg/l)	As Removal (%)
91	7.6	174	42.0
260	7.7	167.6	44.1
388	7.6	167.7	44.1
(c) Alum Dose = 100 mg/l			
Alkalinity (mg/l as CaCO ₃)	pH	As Residual (µg/l)	As Removal (%)
84	6.9	167.7	44.1
236	7.3	165.6	44.8
422	7.6	167.7	44.1

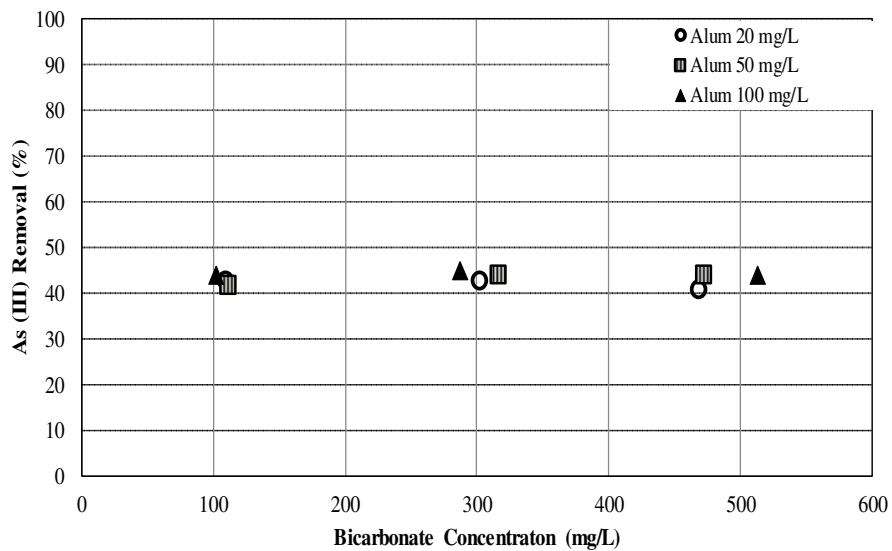
Figure 3: Removal of As (III) (Initial Concentration 300 µg/l) by Alum Coagulation from 0.10 M NaCl Solution as a Function of Bicarbonate Concentration.

Figure 4: Comparison of Removal of As (V) and As(III) Present at an Initial Concentration of 300 µg/L from 0.10 M NaCl Solution in the Presence of Bicarbonate Ions.(Alum dose = 50 mg/L)

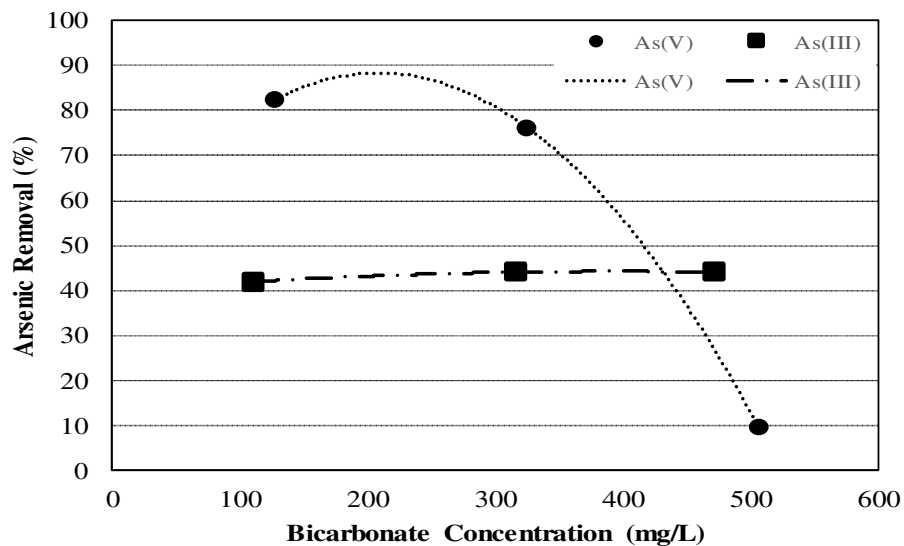


Figure 4 shows removal of As(V) and As(III), both present at an initial concentration of 300 µg/l, by alum coagulation from a 0.10 M NaCl solution as a function of bicarbonate concentration. As noted earlier, bicarbonate is the principal contributor of alkalinity in the pH range at which the experiments were carried out. It shows that at lower bicarbonate concentration, removal of As(V) is much higher compared to that of As(III). But as bicarbonate concentration increases, it competes with As(V) significantly lowering its removal, while As(III) removal remains virtually unaffected. Thus, this study suggests that at higher bicarbonate concentration, As(III) removal could become higher than As(V) removal during alum coagulation.

3.2 Effect of Bicarbonate on Arsenic Removal by Ferric Chloride (FeCl₃) Coagulation

As(V) removal: Table 4 shows experimental data on removal of As(V) present at an initial concentration of 300 µg/l from 0.10 M NaCl solution at different alkalinities and at different ferric chloride doses(5, 10 and 15 mg/l). It shows that in general at very low alkalinity (e.g., 35 mg/l as CaCO₃), As(V) removal is poor, and removal increases as alkalinity increases. But as alkalinity increases further (e.g., exceeding 300 mg/l), As(V) removal drops slightly.

This phenomenon is most likely due to the effect of alkalinity on formation of ferric hydroxide flocs and competitive adsorption of As(V) and bicarbonate onto ferric hydroxide flocs. Alkalinity promotes oxidation of ferrous ion to ferric ion and the formation of ferric hydroxide flocs, which act as adsorbent. At low alkalinity, the poor removal of As(V) appears to be due to lack of sufficient ferric hydroxide flocs. This was confirmed by visual observation of flocs at different alkalinities during the coagulation experiments, as shown in Figure 5 (flocs appear black in Fig. 5). As alkalinity increased beyond 50 mg/l, ferric hydroxide flocs became visible in the experimental beakers, and As(V) removal increased. As alkalinity increased further,

it did not have much influence on As(V) removal except for very high concentrations exceeding about 400 mg/l; at such high alkalinities, As(V) removal decreased slightly. This is most likely due to competitive adsorption of As(V) (present mostly as HAsO_4^{2-} and H_2AsO_4^-) and bicarbonate (HCO_3^-) onto ferric hydroxide flocs.

Table 4: Removal of As (V) (Initial Concentration 300 $\mu\text{g/l}$) by Ferric Chloride Coagulation from 0.10 M NaCl Solution

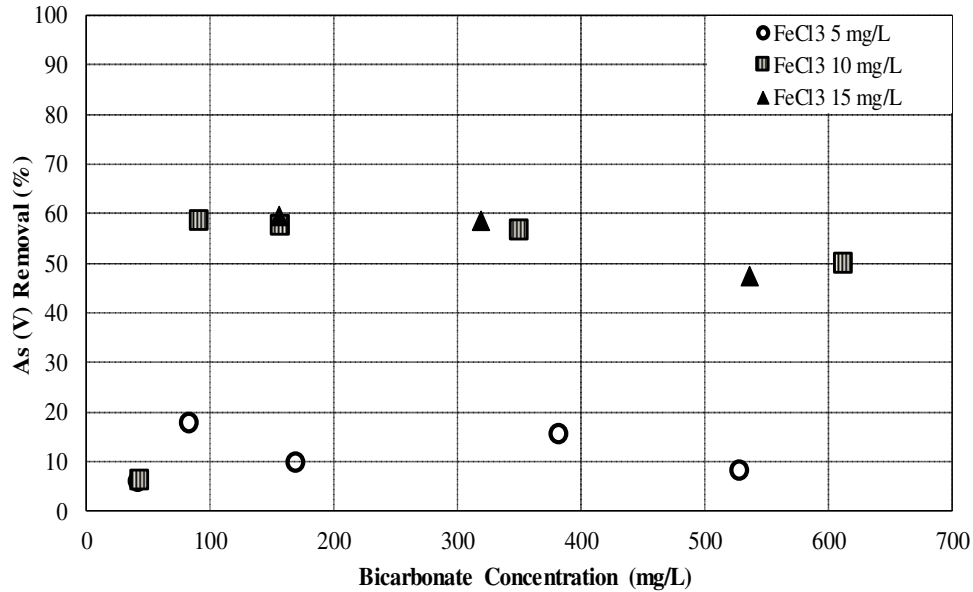
(a) FeCl_3 Dose = 5 mg/l			
Alkalinity (mg/l as CaCO_3)	pH	As Residual ($\mu\text{g/l}$)	As Removal (%)
35	7.3	282.1	6.0
69	7.4	246.8	17.7
140	7.4	271.1	9.6
317	8.0	253.4	15.5
438	8.0	275.7	8.1
(b) FeCl_3 Dose = 10 mg/l			
Alkalinity (mg/l as CaCO_3)	pH	As Residual ($\mu\text{g/l}$)	As Removal (%)
35	7.3	280.8	6.4
75	7.4	123.9	58.7
129	7.7	126.7	57.8
288	7.6	129.4	56.9
506	7.9	150	50.0
(c) FeCl_3 Dose = 15 mg/l			
Alkalinity (mg/l as CaCO_3)	pH	As Residual ($\mu\text{g/l}$)	As Removal (%)
128	7.5	121.6	59.5
262	7.3	124.2	58.6
443	7.8	158.3	47.2

Figure 5: Effect of Alkalinity on Formation of Ferric Hydroxide Flocs during Coagulation with FeCl_3



Figure 6 shows removal of As(V) from 0.10 M NaCl solution as a function of bicarbonate concentration. It shows very poor removal of As(V) at very low bicarbonate concentration due to absence of iron flocs, and slight reduction of As(V) removal at very high bicarbonate concentration due to competitive adsorption of arsenic and bicarbonate.

Figure 6: Removal of As (V) by FeCl₃ Coagulation from 0.10 M NaCl Solution as a Function of Bicarbonate Concentration



As(III) removal: Table 5 shows removal of As(III), present at an initial concentration of 300 µg/l, from 0.10 M NaCl solution at different ferric chloride doses (5, 10, 15 mg/l) and at different alkalinities. Similar to the case for As(V) it shows relatively poor removal of As(III) at very low alkalinity. As explained earlier, this is most likely due to poor formation of flocs at low alkalinity. As alkalinity is increased up to about 100 mg/l, As(III) removal increased. Further increase of alkalinity did not have a significant effect on As(III) removal. This probably reflects lack of competition between unionized As(III) (present as H₃AsO₃) and bicarbonate (HCO₃⁻).

Table 5: Removal of As (III) (Initial Concentration 300 µg/l) by Ferric Chloride Coagulation from 0.10 M NaCl Solution

(a) FeCl₃ Dose = 5 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
69	7.9	247.4	17.5
93	8.0	229.8	23.4
115	7.9	224.3	25.2
261	8.0	224.8	25.1
415	8.0	242.1	19.3

(b) FeCl₃ Dose = 10 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
94	7.1	223.5	25.5
135	7.3	222.4	25.9
329	7.9	212.3	29.2
472	8.0	203.5	32.2

(c) FeCl₃ Dose = 15 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
59	7.2	235.5	21.5
124	7.4	162	46.0
269	7.9	162.5	45.8
413	7.8	148	50.7

Figure 7 shows removal of As(III) as a function of bicarbonate concentration. This also shows negligible effect of bicarbonate on removal of As(III). A comparison of Table 4 and Table 5 suggest more effective removal of As(V) by ferric chloride coagulation compared to As(III).

Figure 7: Removal of As (III) by FeCl₃ Coagulation from 0.10 M NaCl Solution as a Function of Bicarbonate Concentration.

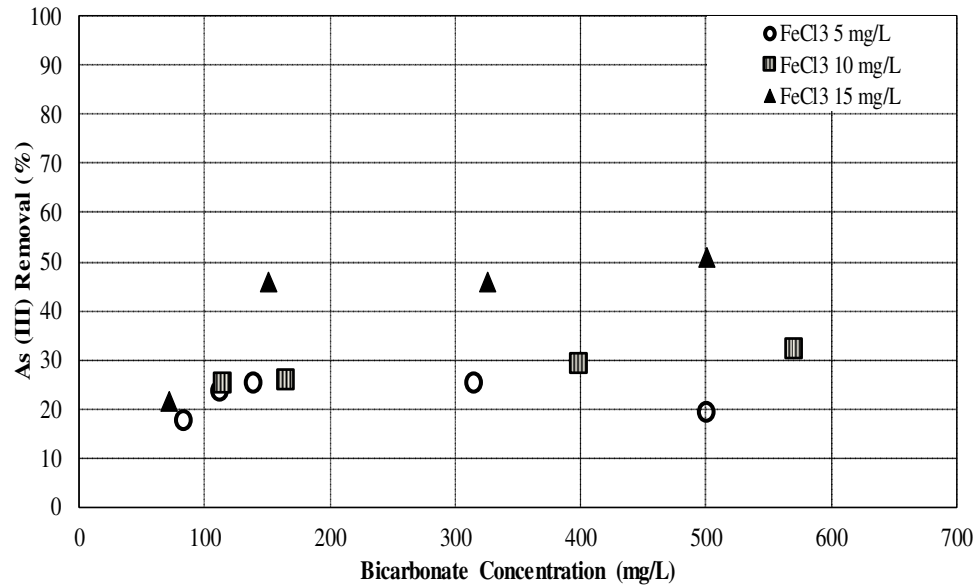


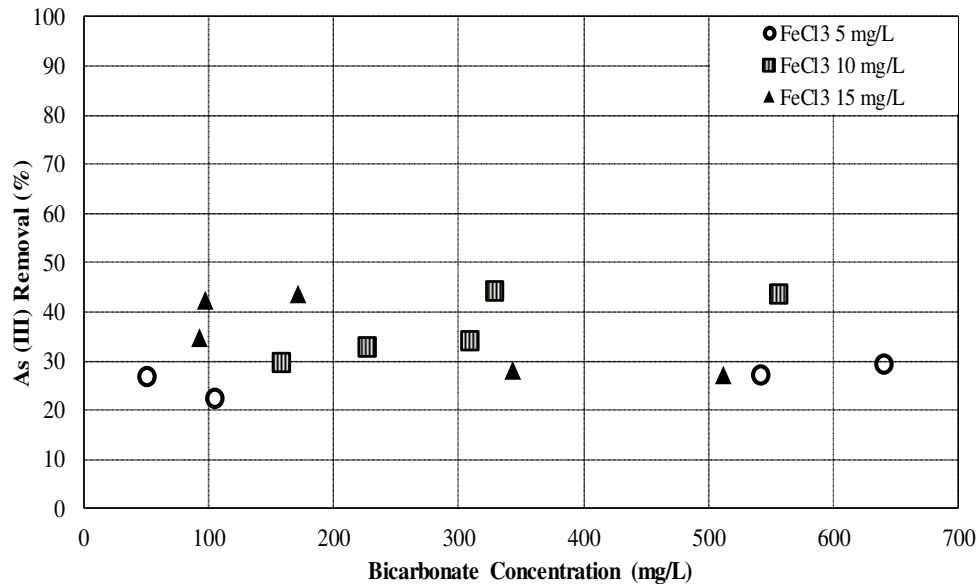
Table 6 shows removal of As(III), present at an initial concentration of 300 µg/l, from 0.01 M NaCl solution at different alkalinities and at different ferric chloride doses (5, 10, 15 mg/l). Although there are limited data on As(III) removal at very low alkalinities, in general As(III) removal appears to be relatively low at lower alkalinities; this appears to be due to poor iron floc formation. As alkalinity increased, As(III) removal increased.

Table 6: Removal of As(III) (Initial Concentration 300 µg/l) by Ferric Chloride Coagulation from 0.01 M NaCl Solution

(a) FeCl₃ Dose = 5 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
42	7.8	219.5	26.8
87	7.8	233	22.3
447	7.7	219	27.0
527	7.6	212	29.3
(b) FeCl₃ Dose = 10 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
130	7.2	211	29.7
186	7.1	201.5	32.8
254	7.3	198	34.0
270	7.4	167.5	44.2
458	7.7	169.5	43.5
(c) FeCl₃ Dose = 15 mg/l			
Alkalinity (mg/l as CaCO₃)	pH	As Residual (µg/l)	As Removal (%)
76	7.0	196	34.7
80	7.1	172.5	42.5
142	7.8	169	43.7
282	7.2	216	28.0
420	7.2	218	27.3

Figure 8 shows As(III) removal as a function of bicarbonate concentration, indicating insignificant effect of bicarbonate on As(III) removal. Comparison of Table 5 and Table 6 suggests limited effect of ionic strength on As(III) removal during ferric chloride coagulation.

Figure 8: Removal of As (III) by FeCl₃ Coagulation from 0.01 M NaCl Solution as a Function of Bicarbonate Concentration.



4. Conclusion

From previous studies it can be inferred that bicarbonate ions compete with arsenic for adsorption onto metal oxides/hydroxides. On the other hand, bicarbonate has been found to promote formation of ferric hydroxide flocs, thereby promoting arsenic removal by adsorption onto iron flocs. But data about the effects of bicarbonate on arsenic removal is limited. In this study, effect of bicarbonate/ alkalinity on removal of both As(V) and As(III) by alum and ferric chloride coagulation from 0.10 M and 0.01 M NaCl solutions have been investigated. Increasing bicarbonate/alkalinity significantly decreased the removal of As(V) during alum coagulation (from 0.10 M NaCl solution) due to competitive adsorption of anionic As(V) species and bicarbonate. Whereas alkalinity/bicarbonate had a negligible effect on the removal of As(III). At lower alkalinity/bicarbonate concentration, removal of As(V) has been found to be much higher than As(III) under similar alum doses; but as bicarbonate concentration increases, As(III) removal overtakes As(V) removal due to poor removal of As(V) in the presence of high concentration of bicarbonate. This study revealed that bicarbonate has a significant effect on formation of ferric hydroxide floc, and this in turn has a significant impact on arsenic removal during ferric chloride coagulation. During ferric chloride coagulation, removal of both As(III) and As(V) have been found to be very poor at very low (e.g., < 50 mg/l) alkalinity/ bicarbonate; this is due to poor/no iron-floc formation at such low alkalinities. As alkalinity/bicarbonate concentration increases, removal of both As(III) and As(V) increase. Further increase in alkalinity did not have very significant influence on arsenic removal. In Bangladesh, alkalinity of groundwater varies significantly among regions, with very high concentrations reported for south and south-western regions; many of these regions also suffer from high concentration of arsenic. Therefore, effect of alkalinity/bicarbonate on arsenic removal should be carefully considered while designing arsenic removal systems that are based on adsorption. This study has a few limitations which need to be addressed in future studies. For example, in

this study, effect of other anions (e.g. phosphate, silicate, sulphate, etc.), which may also compete with arsenic for adsorption, have not been studied. Besides, no effort was made to vary pH and initial arsenic concentration in batch experiments.

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Aziz, Jahin, Tasnim & Ali

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