

Low-cost Technique of Arsenic Removal from Water and Its Removal Mechanism

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Abstract

This study examines the potential of removing arsenic from water by coprecipitation with naturally occurring iron. The experimental study examined the sensitivity of removal of arsenic in response to manual mixing and prolonged settlement. It was found that about 88% arsenic removal could be achieved after 24 h settlement. It has also demonstrated that provided the iron levels are sufficiently high (say 1.2 mg/l), simple shaking of a container and allowing the iron-arsenic complex to settle out for 3 days could reduce the concentration of arsenic from 0.10 mg/l to Bangladesh standard (0.05 mg/l). There was evidence that adsorption may be the dominating trapping mechanism when Fe/As weight ratio was ³ 10.

INTRODUCTION

In recent years there has been widespread coverage in the media about the problems of arsenic in Bangladesh's drinking water. This has been an unforeseen consequence of a large-scale programme to replace contaminated surface water sources by 'safe' groundwater. The programme led to the installation of about 4 million tubewells, never suspecting the presence of arsenic in the aquifers carrying the groundwater.

Arsenic in groundwater was first detected in 1993 following reports of many people suffering from arsenical skin diseases. Further investigations showed the extent of the problem with large areas of the country's water supply being affected and millions of people are at serious risk of arsenic poisoning.

Technology for arsenic removal from water already exists (Kartinen and Martin, 1995). However, the socio-economic conditions which prevail in Bangladesh, do not permit implementation of this type of technology on grounds of cost. The task was to develop technology, which could be implemented at household level and at virtually zero cost.

A related problem in the groundwater in Bangladesh and some other parts of the world is the presence of iron. While this not a health hazard, it is usually removed because of taste and staining problems. It is well known that iron hydroxide adsorbs arsenic (Ferguson and Gavis, 1972). In this study an attempt was made to exploit the naturally occurring iron as a means of arsenic removal through the development of simple practices based on adsorption-coprecipitation and settlement.

BACKGROUND

Groundwater in Bangladesh: Arsenic and Iron

The presence of arsenic in groundwater in Bangladesh is the most serious health hazard the country has ever faced. A recent study carried out by BGS and MotMacdonald (1999) shows many areas within Bangladesh with arsenic concentration greater than 0.05 mg/l implying that millions of people are at serious risk of arsenic poisoning. According to EGIS (1997) report the concentration of arsenic generally varies from 0.02 to 0.5 mg/l (exceeding the WHO standard of 0.01 mg/l and Bangladesh Standard of 0.05 mg/l). Regarding the iron concentration, about 65% of the area of Bangladesh contain dissolved iron in excess of 2 mg/l and in many areas concentration is as high as 15 mg/l (Ahmed et al, 1998). While arsenic and iron pose individual problems, their association in groundwater has the potential of providing a simple means of removing arsenic by coprecipitation and adsorption.

Arsenic Chemistry and Its Removal

Arsenic occurs in waters in several different forms depending on the pH and redox potential E_h . Arsenate (As(V)) and arsenite (As(III)) are the primary forms of arsenic found in natural waters (Ferguson and Anderson, 1974). The thermodynamically stable forms are As(V) in oxygenated surface water and As(III) in reducing groundwater. The latter is of the concern in this study.

However, both forms can occur together in both environments due to the slow oxidation and reduction kinetics (Edwards, 1994).

From previous studies (Jekel 1994, Shen, 1973; Sorg and Logsdon, 1978; Edwards, 1994; Cheng et al, 1994; Hering et al, 1996), it is already known that coagulation-precipitation is a very effective and most frequently applied technique in arsenic removal. In this technique, addition of coagulant facilitates the conversion of soluble arsenic species into insoluble products through coprecipitation and adsorption

Adsorption of arsenic onto preformed $\text{Fe}(\text{OH})_3$ has also been shown to be an effective method for removing arsenic (Hering et al, 1996; Driehaus et al, 1998). Like coagulation, removal of As(III) by adsorption is less than that of As(V). However arsenic removal during coagulation with FeCl_3 is more efficient than arsenic adsorption onto preformed hydrous ferric oxide (Hering et al, 1997).

Settlement is a well-known mechanism in the context of iron precipitation and iron removal. Depending on the initial iron concentration in the raw water, Ghosh and O'Connor (1966) indicated that a 1 h design period was 'inadequate for complete iron precipitation'. It suggests that where arsenic removal depends on iron precipitation, the settling time must exert a major influence on the removal process. A study carried out by Shen (1973) with just arsenic-containing water showed that sedimentation could only remove 8.7% arsenic after 24 h settlement. A drawback of Ghosh and O'Connor (1966) is the lack of settling time. In the case of Shen's (1973) sedimentation, the study is restricted to arsenic and provides no insight into the potential influence of iron and the removal efficiency.

Materials and Methods

Laboratory nano-pure water was used for the preparation of the standard and sample waters. The chemicals employed for the experiments were general purpose grade (GPR) and used without any purification. Solutions of arsenic (As(III)) were prepared from concentrated stock solution of arsenic trioxide, As_2O_3 , (1 mg/ml) in 0.5 M/l HCl supplied by BDH, UK for use in the tests. In common with other studies (Shen, 1973; Edwards, 1994; Cheng et al, 1994) iron presence was based on preparing FeCl_3 solution.

All experiments were performed with a constant ionic strength of 0.01 M/l NaNO_3 and 0.1 g/l NaHCO_3 to provide necessary alkalinity. pH was maintained by adding 0.1 M NaOH. All glassware was cleaned by soaking 10% HNO_3 and rinsed three times with nano-pure water. Blank tests (without Fe) confirmed that no arsenic was lost through adsorption onto the glassware. Each type of test was carried out in (at least) triplicate.

Arsenic was measured by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) method. In the ICP-AES method, all samples and

standards were acidified according to the standard methods (APHA, AWWA and WEF, 1995).

Values of the parameters such as arsenic, pH and iron/arsenic ratios were chosen to be representative of the range found in Bangladesh (EGIS, 1997). The As(III) form of arsenic was selected for the experimental investigation, as it is the form of arsenic likely to be found in groundwater (DFID et al, 2000). Experiments were carried out with initial As(III) concentration of 0.2 mg/l, Fe dosages of 4.0 mg/l and pH 7.5.

Additional detail on methods included in the subsequent text to support specific parts of the experimental programme.

EXPERIMENTS AND RESULTS

Mixing, Filtration and Settlement

Mixing is a necessary stage in order to ensure aeration and to induce flocculation as well as assuring dispersion of the chemicals. At village level, shaking is a straightforward means of achieving mixing. This was compared with mechanical mixing, the latter being included for reference purposes. Similarly filtration was introduced for reference purposes, but the prime focus of the study was to exploit sedimentation as a means of achieving solid-liquid separation following the initial As-Fe interaction.

Mechanical mixing was applied to 1 litre sample water (0.01M/l NaNO₃ and 0.1 g/l NaHCO₃) containing 0.2 mg/l As(III), 4.0 mg/l Fe at pH 7.5. Sample containing in a 2-litre capacity conical glass flask was mixed in an orbital shaker (KL2) at a rapid rate (410 rpm) for 5 min, at a slow rate (100 rpm) for 25 min and allowed to settle. In the series based on manual mixing, samples were shaken vigorously for periods in the range of 15 s to 5 min and then allowed to settle. After 2 h settlement, two sets of supernatant were collected at a depth of 20 mm from the top surface from each type of samples (both mechanical and manually mixed samples). One set of sample was analysed for residual As(III) concentration with filtration through 0.45 µm filter papers and another set was analysed for the same conditions but without filtration. Table 1 presents the effects of mixing condition and filtration on As(III) removal at different mixing time.

For the filtered samples, shown in Table 1, it is observed that the removal efficiency is insensitive to the mixing regime, whereas for the unfiltered samples, removal depends on mixing type and time. In the latter case, the duration of mixing probably enhances flocculation, because it seems likely that the larger removal rates are associated with larger particle sizes. It is seen that 5 min manual mixing is almost as effective as mechanical mixing.

Table 1: As(III) removal at different mixing conditions of filtered and unfiltered samples after 2 h settlement (Initial As(III) 0.2 mg/l, Fe 4.0 mg/l and pH 7.5)

Test condition	Mean As(III) removal, %	
	Filtered	Unfiltered
Manual mixing:		
15 s manual mixing	63.7	32.2
30 s manual mixing	63.6	45.4
1 min manual mixing	64.3	44.5
3 min manual mixing	64.0	49.3
5 min manual mixing	63.6	53.5
Mechanical mixing	68.5	60.4

To investigate the effects of settlement on As(III) removal, tests were carried out following the same procedure in manual mixing method, the samples being allowed to settle for 24 h. Supernatant was collected at specified time intervals, 2, 4, 6 and 24 h and analysed for residual As(III) concentration without filtration. Fig. 1 shows the effects of settling time on As(III) removal for manually mixed unfiltered samples at varying mixing times. In Figure 1 it is evident that at shorter settling times say 2-6 h, the removal is fairly sensitive to the mixing time. In contrast, at longer settling time (24 h) the removal is less sensitive to the initial stage of mixing.

Insight into the Removal Mechanism

From the experiments described above, it was evident that manual mixing followed by settlement was reasonably effective for reducing the levels of arsenic in the presence of iron. Having gained this experience, further experiments were conducted to discover more about the interaction between As(III) and Fe(III). Experiments were carried out with 1.0 litre sample water in a 2 litre capacity conical glass flask at Fe dosage of 4.0 mg/l, As(III) concentration ranging from 0.1–7.5 mg/l and pH 7.5. The flask was shaken manually for 1 min and then transferred to a 1.0 litre capacity cylinder (432 mm height) to settle for 1 day. After 24 h, the supernatant was collected from the mid depth of the cylinder and the residual arsenic was measured. The removal of As(III) was determined from the difference between As(III) added and the measured residual.

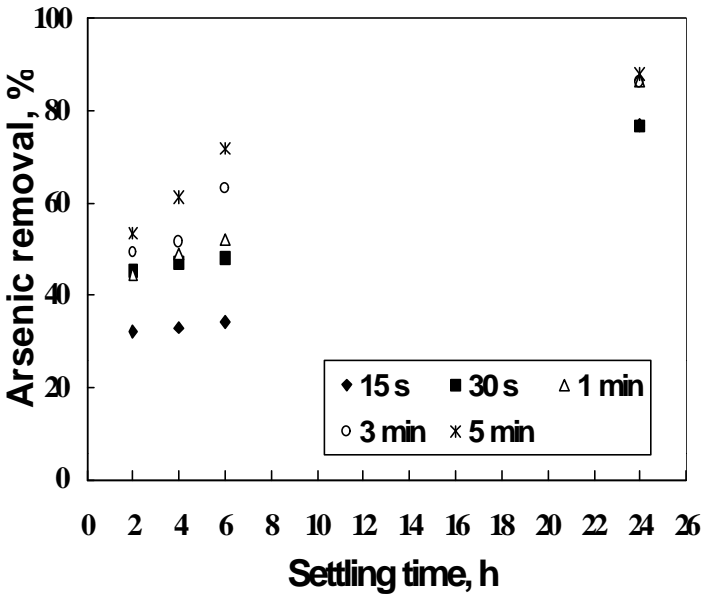


Figure 1 : Effects of settlement on arsenic removal for manually mixed unfiltered samples at varying mixing time with initial As(III) concentration of 0.2 mg/l, Fe 4.0 mg/l and pH 7.5.

Results of this test are plotted in Figure 2. When the initial As(III) is between 1.6 to 2.8 corresponding to As/Fe ratio (by weight) in the range of 0.4 to 0.7, it is seen that removal becomes less sensitive to initial arsenic concentration with a removal density ~ 0.15 mg As/mg Fe over this range. However further increase of As/Fe ratio lead to higher removals.

In this type of test, the removal depends on a series of mechanisms i.e. contact between Fe and As (influenced by molecular diffusion), floc formation (influenced by shaking) and solid-liquid separation by sedimentation. In order to gain an insight into the data corresponding to removal at 24 h, the results were plotted in the same form as an adsorption isotherm (Figures 3 and 4). Here it is recognised that the resultant trend refers the combination of processes rather than just adsorption. This form of plot has the benefit of identifying the removal ratio (mg(As)/g(Fe)) corresponding to the residual level of arsenic $\frac{3}{4}$ the latter needing to comply with target water quality standards. Figures 3 and 4 also show adsorption data from Pierce and Moore (1982) study. Pierce and Moore (1982) investigated the behaviour of As(III) removal by purely adsorption onto pre-

prepared amorphous $\text{Fe}(\text{OH})_3$. They found that for a certain range of initial As(III) concentration (0.05–1.0 mg/l), the adsorption reaches a saturation point. At higher initial As(III) concentration (2.5–50 mg/l), the adsorption of As(III) per unit mass of adsorbent increased linearly with increasing equilibrium concentration.

Before commenting on the behavioural features shown in Figures 3 and 4, it is necessary to provide some of the background behind adsorption plots. Pierce

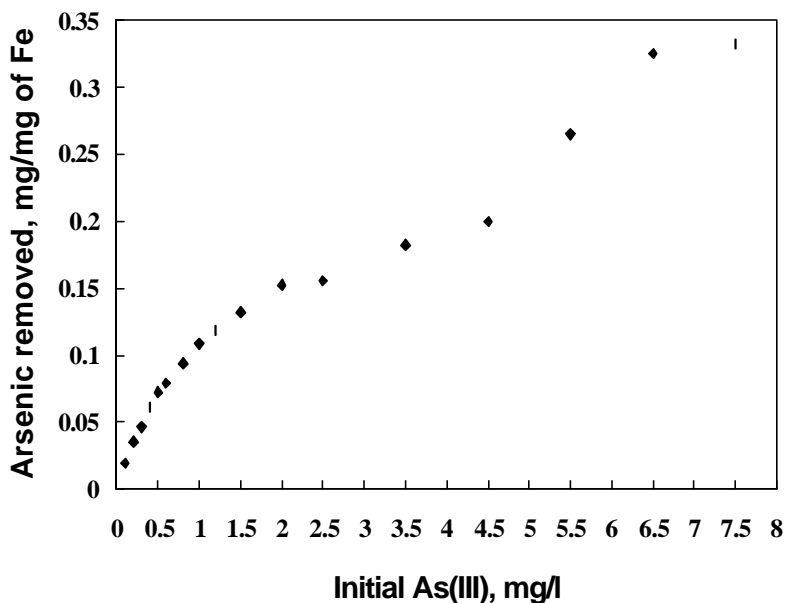


Figure 2 : Removal density of arsenic using FeCl_3 with 1 min manual mixing and 1 day settlement with initial As(III) concentration over the range of 0.1 – 7.5 mg/l, Fe 4.0 mg/l and pH 7.5.

and Moore (1982) data refers to adsorption at pH 7.0 whereas the pH in the current study was 7.5. The differences in between the separate studies are not considered to be significant as it is known that around neutral pH range, the As(III) removal by coagulation is largely unaffected by pH (Edwards, 1994). The adsorption tests reported by Pierce and Moore (1982) were carried out for 24 h continuous mixing by magnetic stirrers and then supernatants were centrifuged and analysed for arsenic concentration. The present work was also carried out for 24 h settlement, the supernatant being analysed for residual arsenic concentration without filtration.

In Figure 3 there is a remarkable similarity between the trends (removal/adsorbed) in the domain $As(mg)/g Fe < 60$. This may be a coincidence or alternatively may point to the role of adsorption as the dominating removal mechanism when Fe/As weight ratio is ≥ 10 . In the case of the adsorption-centrifugation combination (Pierce and Moore, 1982), saturation occurs at an adsorption density of around $70\text{ mg As(III)/g Fe}$ whereas higher levels of removal (about $130\text{ mg As(III)/g Fe}$) are achieved by the coprecipitation-sedimentation technique for the stated range of initial $As(III)$ concentration.

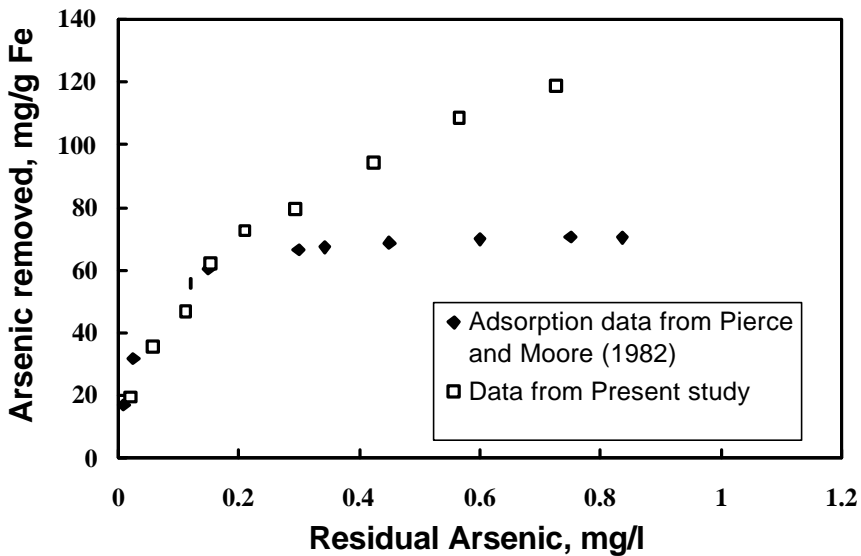


Figure 3 : A comparison of arsenic removal density as a function of residual arsenic concentration by adsorption-coprecipitation and sedimentation with $FeCl_3$ at pH 7.5 and adsorption onto pre-formed $Fe(OH)_3$ at pH 7.0 for initial $As(III)$ concentration of 0.05-1.0 mg/l and contact time 24 h.

Figure 4 shows the comparison at higher initial $As(III)$ concentration and displays a very different form of behaviour. In contrast to Figure 3, the removal density obtained by adsorption study (Pierce and Moore, 1982) is higher than those obtained by the present study for the stated range of initial $As(III)$ concentration (Figure 4). The different nature of the removal density of the present study might be due to the differences in experimental procedure. Pierce’s work was carried out by introducing preformed ferric hydroxide to the solution and not by neutralising an acid solution that contains both Fe and As, whereas the

latter procedure was adopted in the present study. Further, at higher initial dosage of As(III) (≥ 2.5 mg/l), a large amount of 0.1M NaOH was required to maintain pH 7.5 constant. Here, it is conceivable that the NaOH might change or reorder the internal properties of adsorption surface sites.

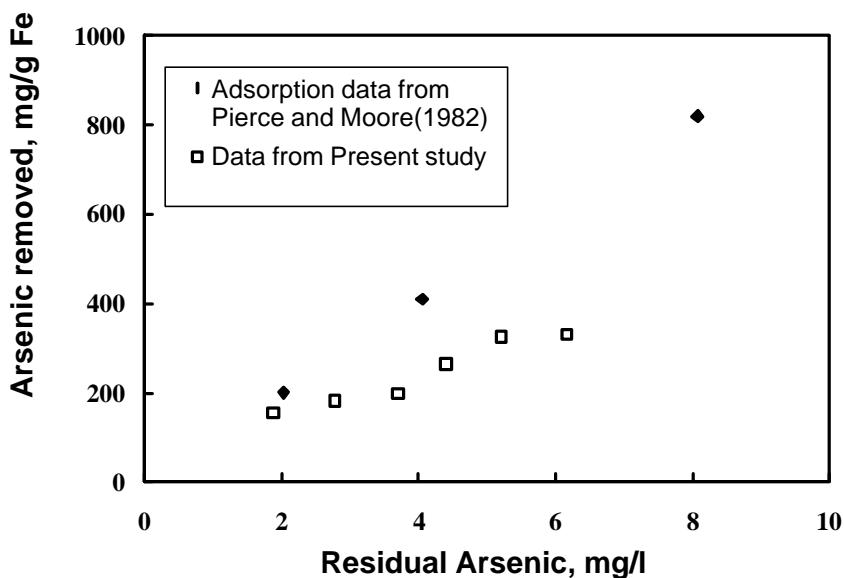


Figure 4 : A comparison of arsenic removal density as a function of residual arsenic concentration by adsorption-coprecipitation and sedimentation with FeCl_3 at pH 7.5 and adsorption onto preformed $\text{Fe}(\text{OH})_3$ at pH 7.0 for initial As(III) concentration of 2.5 - 10 mg/l and contact time 24 h

Implication of Iron Concentration on Reducing the Level of Arsenic

A series of tests was conducted with varying As(III) concentration ranging from 0.1 mg/l to 0.5 mg/l and Fe/As ratios (by weight) ranging from 5 to 40 at pH 7.5. The sample was mixed for 1.0 min manually and transferred to a measuring glass cylinder (432-mm height) to settle for 3 days. After 3 day settlement, the supernatant was collected and the residual arsenic was measured.

The influence of Fe/As ratio on the As(III) removal is presented in Fig. 5. The trend in Figure 5 shows that the effectiveness of As(III) removal is strongly influenced by the Fe/As ratio and is also sensitive to initial As(III) concentration. The trends are reasonably similar for the different values of the Fe/As ratios. In

Mamtaz (2000), it was shown that the data was adequately represented by the empirical equation:

$$\text{As(III) removal (\%)} = 3.98(\text{Initial As(III)})^{0.22} (\text{Fe/As})^{0.39} (\text{pH})^{0.24} \quad (1)$$

where As(III) is expressed in $\mu\text{g/l}$. This equation is valid for As(III) concentration of 0.1 to 0.5 mg/l, Fe/As ratio (by weight) 5 to 40 and pH 5 to 8 and a settling time of 3 d.

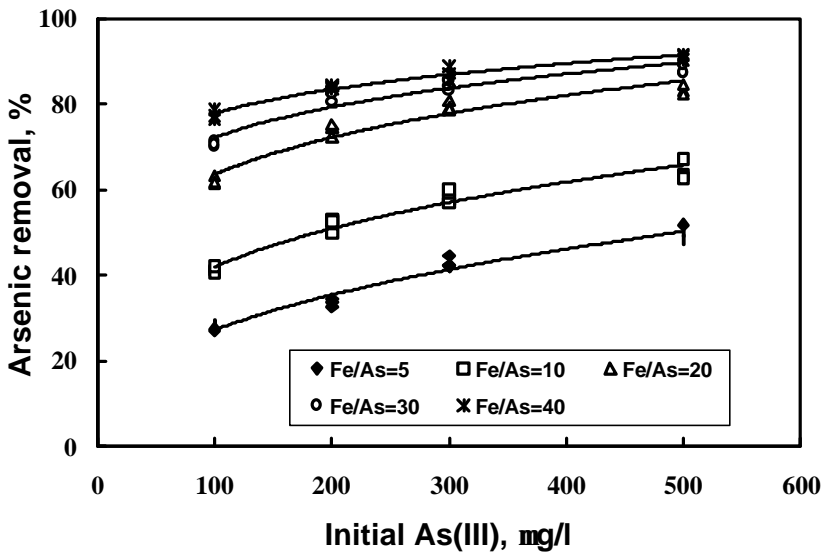


Figure 5 : Effects of arsenic removal at different Fe/As weight ratios and initial As(III) concentration at pH 7.5 after 3 day settlement.

The present paper focuses on a different aspect of the data shown in Figure 5. From the experimental results of Fe/As ratio (by weight) tests at different initial As(III) concentration at pH 7.5 and 3 day settling (Figure 5), it is possible to calculate the minimum amount of Fe required for reducing the arsenic level to the Bangladesh standard (0.05 mg/l) for a given As(III) concentration. A contour analysis allows one to identify the 50 $\mu\text{g/l}$ isoconcentration line of residual arsenic for the data shown in Figure 6. This was well represented by the empirical relationship as follows (Figure7):

$$\text{Fe} = 66 \text{ As}^{1.75} \quad (2)$$

where, Fe and As concentrations are in mg/l.

In essence, eqn. 2 shows the necessary iron concentration to allow reduction of the arsenic to achieve the Bangladesh standard (50 µg/l) for a given level of arsenic. For example, when the arsenic level is 0.10 mg/l, the minimum amount of iron requirement is 1.2 mg/l at pH 7.5 and with 3 day settlement to attain the Bangladesh limit (0.05 mg/l). It is noted that Figure 6 and 7 and Equation 2 are tied to 3 days settlement. Had a different time scale been used, the trends would have been different because of the dependence of the removal rate on the settling time.

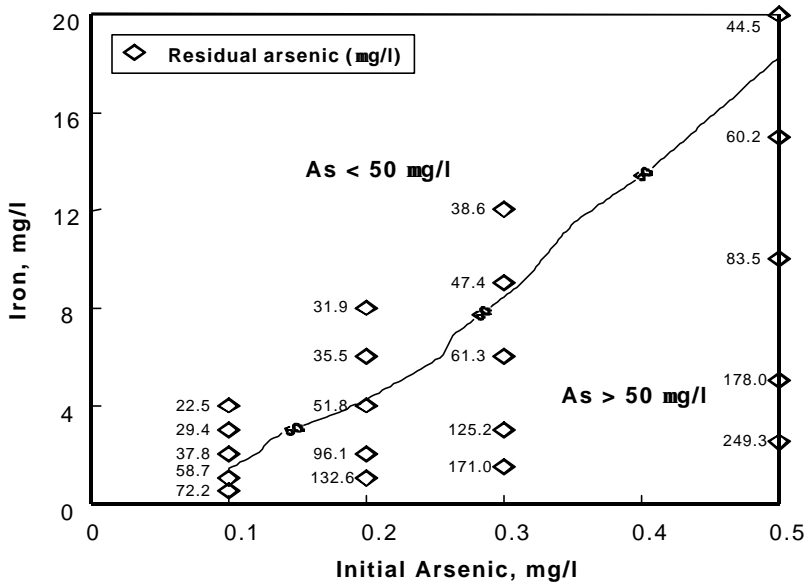


Figure 6 : Contour line showing the boundary of the zone complying with Bangladesh standard (50 mg/l) for residual arsenic concentration at pH 7.5 after 3 day settlement.

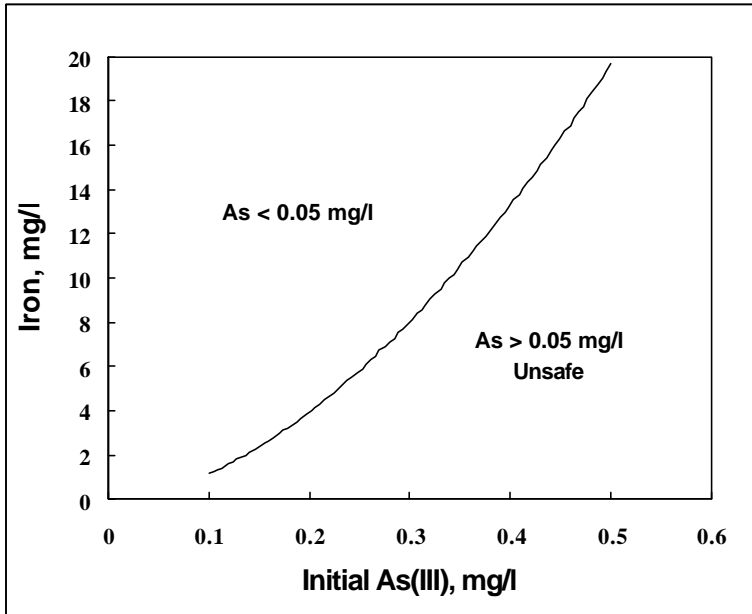


Figure 7 : Amount of minimum Fe required for a given arsenic concentration at pH 7.5 after 3 days settlement to comply with the Bangladesh Standard for arsenic (0.05 mg/l).

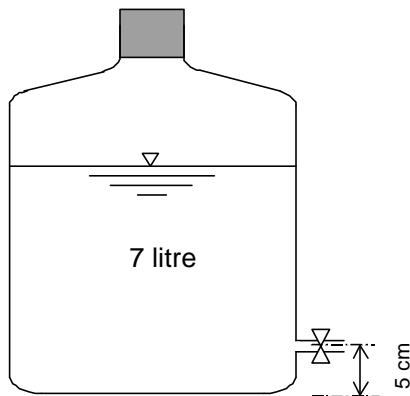


Figure 8 : Container to be used for arsenic removal

DISCUSSION

The principal achievement in this study has been the demonstration that adsorption and coprecipitation with iron followed by settlement can remove significant amounts of arsenic provided that there is sufficient iron and a sufficient settling time (≥ 3 d). As such the method has considerable promise as a low-cost technique because of the total absence of added chemicals.

A surprising aspect of the study (see Table 1 and Figure 1) was that manual mixing and sedimentation was capable of achieving higher rates of removal than mechanical mixing and filtration through a 0.45 μm filter paper. The reasons for this are not clear and require further investigation. At early time (< 6 h) manual shaking was shown to influence the separation process presumably arising from the enhancement of the size of the microflocs which were just visible to the naked eye. Simple increases in size are not necessarily responsible for increased settling because experience shows that flocs are fractal structures, larger sizes tending to lower effective density (Gregory, 1989).

In contrast to the experiences with settlement in this study, Shen (1973) regarded sedimentation as an ineffective method for removing arsenic. The major difference between the current study and Shen's study is that Shen (1973) carried out sedimentation test *without* the presence of iron or any adsorbent. This emphasises the advantage that is gained by coprecipitation to enhance arsenic removal.

When comparing the removal mechanism of the present study with other studies, the main limitation is that preparation techniques such as manual shaking and settlement have no direct equivalents. An additional complication is the time-dependence of the removal process. Hence, it is difficult to make direct comparisons with other studies. In Figure 2, it was evident that the removal density did not reach a saturation point of the range of As(III) concentration tested (0.1-7.5 mg/l). Ferguson and Anderson (1974) also reported that arsenite adsorption did not reach a state of saturation; they observed a maximum adsorption density at 0.4 mole As /mole Fe. Pierce and Moore (1982) suggested that a multisite adsorption occurred in arsenic adsorption onto $\text{Fe}(\text{OH})_3$ at higher initial arsenic concentration i.e. arsenic can penetrate into oxide surface which is theoretically possible for an open permeable structure of amorphous $\text{Fe}(\text{OH})_3$.

The coincidence of the trends plotted in Fig. 3 is intriguing. At first sight it suggests that adsorption is responsible for the removal of arsenic up to the initial arsenic concentration of 0.4 mg/l (corresponding residual concentration is ~ 0.15 mg/l). However without further scrutiny to examine the time-dependence (in our study), this interpretation must remain speculative. Nevertheless it should be pointed out that just as our own data is linked to a time-dependent process, this

aspect is also true of the Pierce and Moore (1982) study wherein centrifugation after 24 h adsorption is used as the separation technique. According to Hering et al (1996) adsorption is a dominant mechanism in the arsenic removal by coagulation. Some comparative studies of As(V) adsorption on preformed $\text{Fe}(\text{OH})_3$ and coprecipitation showed that higher removal densities (mole As/mole Fe) could be achieved by coprecipitation when compared with adsorption, this effect being most pronounced at high As-Fe ratios (Fuller et al, 1993; Edwards, 1994). Clearly this subject requires further investigation.

The prime objective of this work was to ascertain whether the As/Fe interaction offered sufficient promise for arsenic removal (a practical system being described in the Appendix). It is believed that in spite of a lack of understanding of the underlying mechanisms, the method is potentially very useful. A necessary condition is the sufficiency of iron. Relationship of the type shown by Equation 2 is likely to be useful for discerning the minimum iron concentration.

ARSENIC REMOVAL AT VILLAGE LEVEL

A simple and practical method that can be adopted at village level for reducing the level of arsenic in contaminated groundwater in Bangladesh is described below. This method applies to water in, which there is a sufficient iron concentration to form iron-arsenic precipitates.

For iron concentration between 1.0 and 20.0 mg/l and arsenic concentration between 0.1 to 0.5 mg/l, there is a reasonable expectation that after following the procedure described below, the water will comply with the Bangladesh Water Quality Standard for arsenic (0.05 mg/l).

- Fill a clean container of 10 litre capacity (Fig. 8) with about 7 litre water to be treated leaving an air space
- Shake the container for 1 minute
- Allow 3 days for particle settling
- Take out treated water through the tap, not exceeding a flow-rate of 0.5 litre/min (Mamtaz and Bache, 2000).

CONCLUSIONS

Manual flocculation followed by sedimentation appears to be a promising method for removing arsenic from groundwater containing naturally occurring iron. Over prolonged periods of settlement (≥ 24 h), removal of up to 88% was achieved (Fig. 1). Provided the iron levels are sufficiently high (say ≥ 1.2 mg/l),

it has been demonstrated that simple shaking of a container and allowing the arsenic-iron complex to settle out for 3 days could reduce the concentration of arsenic from 0.10 mg/l to the Bangladesh standard (0.05 mg/l).

Evidence (Fig. 3) suggests that adsorption may be the primary mechanism controlling the As(III) removal by the techniques employed in this study and when the Fe/As weight ratio is ≥ 10 . However this aspect requires further scrutiny.

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