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SLUDGE TREATMENT OF ARSENIC REMOVAL UNIT IN BANGLADESH AND REMOVAL OF ARSENIC USING MICROORGANISMS

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ABSTRACT: Arsenic removal unit was constructed by Miyazaki University research group at Marua village in Bangladesh. The sludge drained from the unit was directed to the sludge tank and settled down. Then the supernatant water in the tank was released to the artificial pond. The values of ORP in the sludge tank measured in 2006 had increased considerably compared with the one measured in 2005. Only arsenic concentration of the filtrate of the sludge in the sludge tank had decreased considerably compared with one year ago. The numbers of living cells in the sludge tank and the artificial pond showed similar result on the each arsenic concentration and estimated to be more than 10^2 cells/ml using 10g As/L arsenic medium by plate culture methods. These bacteria might be useful for the development of the sludge disposal methodology in the future.

KEY WORDS: Arsenic, Sludge, Gravel Sand Filter (GSF), Bangladesh

INTRODUCTION

Jessore district is one of the worst arsenic affected regions in Bangladesh. Miyazaki University research group had been constructed pond sand filters (PSF) in some areas as an alternative safe water source for the arsenic affected people (H. Yokota, *et al.*, 2001). In these PSFs' horizontal roughing filter (HRF), developed by All Indian Institute of Hygiene and Public Health (AIHH&PH), has been used as a pretreatment unit for the slow sand filter (SSF). The HRF works as an alternative process of coagulation-sedimentation. Therefore, it was assumed that such a coagulation-sedimentation process could be used to remove arsenic from groundwater of Bangladesh, which includes high amount of naturally iron. The arsenic removal unit is named Gravel Sand Filter (GSF) and is expected to overcome the dry season's low flow problem encountered by the existing PSFs. The GSF, which is perfectly same as an existing PSF, was constructed at Marua village of Chaugacha upazilla at Jessore district in Bangladesh and users started using this unit since January 2003 (M. M. Hussainuzzaman, *et al.*, 2003).

When water is being poured into the unit through the inlet, it gets aeration for oxidizing Fe(II) and As(III) to Fe(III) and As(V) respectively. Naturally occurring iron in the groundwater is being utilized to remove arsenic here. Arsenic is being adsorbed on the iron oxide

particulates' surface and then being filtered along the gravel tanks, acting as HRF.

Prolonged accumulation of iron sludge containing very high concentrations of arsenic has been occurred by the long-term operation of the unit. In order to eliminate the clogging problem for the sludge accumulation, the valve drainage in the gravel chambers has been performed regularly (H. Yokota, *et al.*, 2003).

A sludge disposal methodology becomes an important problem currently. In order to solve this problem, a sludge tank has been constructed for settling the sludge after being drained out of the gravel chambers. The drained water containing sludge is directed to the tank where they get time to settle down and then the supernatant water has been released to the artificial pond. Precipitated arsenic sludge has been collected periodically from the bottom of the sludge tank. The arsenic sludge has been solidified with cement. Tests have been performed to check the arsenic leaching characteristics of cement sludge mixture. Even though the arsenic concentration of the supernatant water flowed to the artificial pond was low, the arsenic concentration in the pond would be increased over time. For the purpose of elucidating arsenic cycle in the pond, we have carried out water and sediment analysis of the pond. This paper is reported result of water and sediment analysis of the pond and the arsenic resistant of bacteria in the pond.

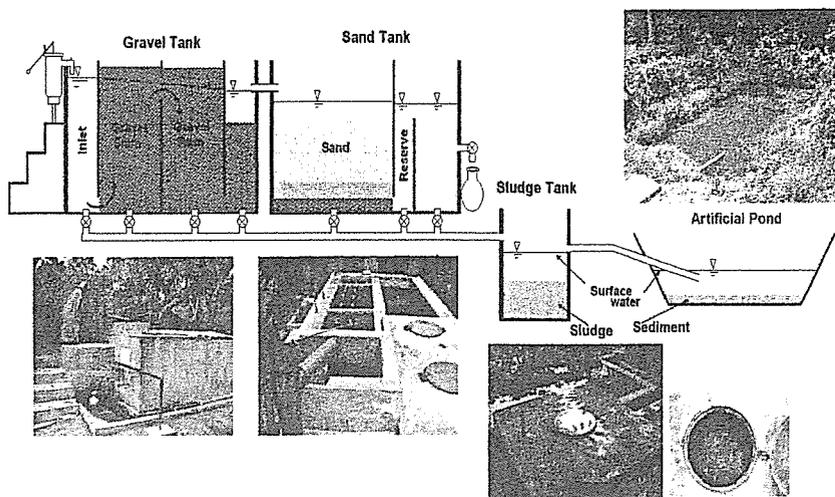


Fig.1 Schematic diagram of GSF

RESULTS AND DISCUSSIONS

Water qualities of the sludge tank and the artificial pond

The values of ORP measured in 2005 showed lower than -200mV with the top and the bottom of the tank and the bottom of the pond, and revealed that reduction state prevailed on their points (Table 1). The values of ORP measured in 2006 increased in the all points compared with the ones measured in 2005. Especially, the values of ORP in the tank measured in 2006 showed more than 200mV.

METHOD

Sample collection and preparation

In September 2005 and August 2006, samples of surface water, sludge and sediment was collected from the sludge tank and the artificial pond on the GSF constructed at Marua village in Bagladesh, respectively. The samples of sludge and sediment were separated into filtrate and residue using aspirating filtration. The residues were evaporated to dryness. The dried residues were measured on contents of arsenic, iron and protein, and attempted elemental analysis by SEM-EDX. The samples of surface water and the filtrates were measured on concentrations of arsenic and iron. The numbers of living cells in their samples were estimated.

Methods of analysis

Values of oxidation-reduction potential (ORP), pH, dissolved oxygen (DO) were measured on top and bottom of the sludge tank and the artificial pond on site, respectively. Value of ORP (TOA DKK RM-20P), pH (TOA DKK HM-20P) and DO (TOA DKK DO-24P) were determined by glass electrode method. To measured contents of arsenic and iron, samples were pretreated by wet ashing method. To measured protein contents, samples were pretreated by alkaline degradation method. Arsenic concentration was determined by atomic absorption spectrometry (Shimadzu AA-6650) with arsenic speciation pretreatment system (Shimadzu ASA-2SP). Total Fe and Fe(II) were determined by phenanthroline absorption spectrophotometry (Shimadzu UV-1200). Elemental analysis was carried out by EDX (Horiba EMAX-5770) attached to SEM (Hitachi S-4100M). The number of living cells was estimated by plate culture method using standard agar medium containing arsenic (0-10g As/L).

Table 1 Water qualities of the sludge tank and the artificial pond (A) Sep. 2005

Source		pH	ORP(mV)	DO(mg/L)
Sludge tank	Top	8.0	-227	1.1
	Bottom	7.2	-231	0.9
Artificial pond	Top	7.9	-6	3.7
	Bottom	7.0	-208	0.4

(B) Aug. 2006

Source		pH	ORP(mV)	DO(mg/L)
Sludge tank	Top	7.6	201	1.5
	Bottom	7.6	206	1.1
Artificial pond	Top	7.1	-1	0.6
	Bottom	6.6	-109	0.6

Arsenic and iron concentration of the sludge tank and the artificial pond

Only the arsenic concentration of the filtrate of the sludge in the tank showed more than 1mg/L in the four samples measured in 2005 (Table 2). But the arsenic concentration of the filtrate of the sludge in the tank measured in 2006 was 0.320mg/L. Though the arsenic concentration in the sludge of the pond decreased considerably, the arsenic concentration in the surface water of the pond increased a little, compared with one year ago. The arsenic concentrations in the pond hardly changed in 2005 and 2006. The ratios of As(III) in the total As measured in 2005 showed more than 79% with three samples except the surface water of the pond. The ratio of As(III) in the total As showed more than 90% with the filtrate of the sludge in the pond and ones showed less than 25% with other samples, when they were measured in 2006. Organic arsenic compounds were slightly confirmed with all samples.

Iron concentrations measured in 2005 indicated almost the same value on the tank and the pond. The ratios of Fe(II) in total Fe showed more than 94% with

their samples at that time. Iron concentrations of all the samples measured in 2006 were considerably low. Iron concentrations on the tank and the pond decreased considerably in one year.

Number of living cells in the sludge tank and the artificial pond

The number of living cells in the tank and the pond was similar and estimated to be more than 10^5 cells/ml using medium containing 0-0.1g As/L arsenic (Table 3). The numbers of living cells using 10g As/L arsenic medium in both places were estimated to be more than 10^2 cells/ml. The colonies in the surface water and the filtrate were apparently different. These results hardly changed in 2005 and 2006. The number of living cells was hardly changed by the environment. The results on numbers of living cells using 10g As/L arsenic medium

indicate that the arsenic resistant microorganisms have a place in the tank and the pond. Microorganisms, that were apparently different from each other, were isolated from the plates containing 10g As/L arsenic.

Analysis of dried residues from the sludge tank and the artificial pond

Total arsenic contents in dried residues of the sludge from the tank and the sediment from the pond were 41mg/kg and 12mg/kg, when they were measured in 2005 (Table 4). Their values hardly changed in 2006. Total iron contents in dried residues measured in 2006 hardly changed on the pond and increased a little on the tank compared with the one measured in 2005. Contents of protein increased a little on the tank and the pond compared with 2005 and 2006. It was indicated that there were arsenic in the particles of dried residues by SEM-

Table 2 As and Fe concentrations on the sludge tank and the artificial pond

(A) Sep. 2005

Source		As(III) (mg/L)	As(V) (mg/L)	Total As (mg/L)	As(III)/T-As (%)	Fe(II) (mg/L)	Fe(III) (mg/L)	Total Fe (mg/L)	Fe(II) /T-Fe (%)
Sludge tank	Surface water	0.098	0.025	0.123	79.7	2.3	0.1	2.4	95.2
	Filtrate of sludge	1.206	0.141	1.347	89.5	9.8	0.3	10.1	97.0
Artificial pond	Surface water	0.009	0.045	0.054	16.7	2.1	0.1	2.2	94.3
	Filtrate of sediment	0.068	0.018	0.086	79.1	11.3	0.4	11.7	96.2

(B) Aug. 2006

Source		As(III) (mg/L)	As(V) (mg/L)	Total As (mg/L)	As(III)/T-As (%)	Fe(II) (mg/L)	Fe(III) (mg/L)	Total Fe (mg/L)	Fe(II) /T-Fe (%)
Sludge tank	Surface water	0.027	0.151	0.178	15.2	0.2	0	0.2	100
	Filtrate of sludge	0.080	0.240	0.320	24.9	0	0	0	-
Artificial pond	Surface water	0.008	0.050	0.057	13.3	0.1	0	0.1	100
	Filtrate of sediment	0.039	0.004	0.043	90.7	0	0	0	-

Table 3 Number of living cells in the sludge tank and the artificial pond

(A) Sep. 2005

Source		Colony forming unit (cells/ml)				
		-	0.01g As/L	0.1g As/L	1g As/L	10g As/L
Sludge tank	Surface water	3.0×10^6	8.1×10^5	8.0×10^5	6.5×10^4	5.0×10^2
	Filtrate of sludge	6.7×10^5	6.5×10^5	6.6×10^5	1.0×10^3	4.6×10^2
Artificial pond	Surface water	2.3×10^5	2.3×10^5	2.3×10^5	5.2×10^4	4.8×10^2
	Filtrate of sediment	5.5×10^5	5.5×10^5	5.5×10^5	1.1×10^3	5.1×10^2

(B) Aug. 2006

Source		Colony forming unit (cells/ml)				
		-	0.01g As/L	0.1g As/L	1g As/L	10g As/L
Sludge tank	Surface water	6.9×10^6	6.8×10^5	3.8×10^5	5.3×10^4	4.7×10^2
	Filtrate of sludge	1.1×10^7	1.3×10^6	6.8×10^5	1.2×10^3	4.8×10^2
Artificial pond	Surface water	1.3×10^7	9.9×10^5	4.0×10^5	5.2×10^4	5.3×10^2
	Filtrate of sediment	1.3×10^7	1.4×10^6	5.5×10^5	1.0×10^3	5.2×10^2

Table 4 Contents in dried residues from the sludge tank and the artificial pond

(A) Sep. 2005

Source	As(mg/kg)	Fe(g/kg)	Protein(g/kg)
Sludge tank	41	24.4	242
Artificial pond	12	37.6	157

(B) Aug. 2006

	As(mg/kg)	Fe(g/kg)	Protein(g/kg)
Sludge tank	40	37.5	263
Artificial pond	10	38.5	170

EDX (Fig.2). Contents of arsenic in dried residues from the tank and from the pond were up to 3.64wt.% and 2.99wt.% on elemental analysis by SEM-EDX, respectively (Table 5).

CONCLUSION

The values of ORP in the sludge tank measured in 2006 increased considerably compared with the one measured in 2005, and revealed that oxidation state prevailed in the tank. The arsenic concentration of the filtrate of the sludge in the tank decreased considerably and one of the surface water in the tank increased a little,

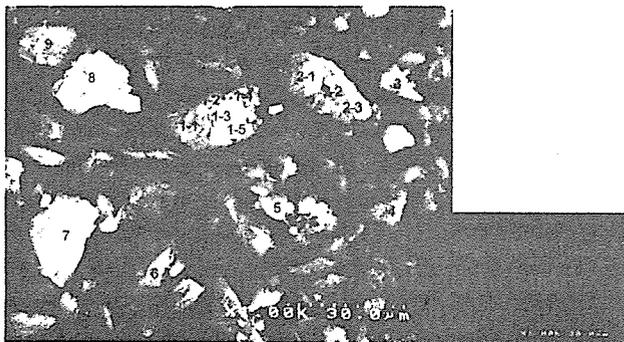


Fig.2 SEM image of dried residues from the artificial pond (Sep. 2005). Inset shows the EDX pattern of As enrichment at the granules.

Table 5 Elemental analysis of dried residues from the artificial pond (Sep. 2005)

No.	Content(wt.%)								
	Na	Mg	Al	Si	S	K	Ca	Fe	As
1-1	0	10.25	16.74	35.66	0.67	2.26	0	19.03	2.99
1-2	0	6.68	16.71	35.66	0.00	3.75	2.52	14.91	1.59
1-3	0	5.23	16.76	36.57	0.95	4.25	2.25	16.51	1.31
1-4	0	7.08	16.08	34.02	0.68	3.92	1.65	18.90	1.53
1-5	0	4.58	16.51	38.43	1.08	5.34	1.54	14.64	0.98
2-1	0	4.18	20.94	39.04	0.93	3.39	1.22	16.61	0
2-2	0	6.18	17.67	30.11	0	1.72	1.00	29.83	0.93
2-3	0	6.74	17.55	29.28	0.57	2.05	1.08	30.40	0
3	3.46	0	15.13	61.61	0	1.93	2.01	4.14	0
4	7.57	0	15.34	58.80	0.83	1.23	0.88	1.64	0
5	0	0.90	17.85	43.76	0.58	8.62	1.93	9.28	0.14
6	0	0	2.41	75.61	0.75	2.00	0.82	5.05	0
7	2.42	0.17	14.79	53.25	0.84	3.15	0	5.72	0
8	0	0.28	2.44	74.64	0.38	1.53	0	1.26	0
9	0	0.25	23.19	40.87	0.36	17.69	0	5.70	0

compared with one year ago. The ratios of As(III) in the total As measured in 2006 decreased considerably in the tank compared with the one measured in 2005. Oxidation state in the tank might have caused these results.

The results on the water qualities, the arsenic concentrations and total arsenic contents in the artificial pond indicated that the environment of the pond had hardly been changing from 2005 to 2006.

Organic arsenic compounds were slightly confirmed with samples from the tank and the pond. Therefore, it seems that arsenic on the tank and the pond has been converted into organic arsenic by biological methylation of arsenic.

The number of living cells was hardly changed by the environment. The arsenic resistant microorganisms were isolated from the plates containing 10g As/L arsenic. We are currently checking activity to arsenic methylation and capability to arsenic removal of their microorganisms. These microorganisms might be useful for the development of the sludge disposal methodology in the future.

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Efficiency of Arsenic Removal Unit Working in Bangladesh and Cement Stabilization of Its Sludge

ABSTRACT: To supply safe water to the arsenic affected community, an arsenic removal unit, gravel sand filter (GSF), was developed by modifying the pond sand filter (PSF) so that it can be used to overcome the dry season low flow problem of it. Performance data of that arsenic removal unit shows that it can be the most suitable option to solve that problem of the PSF. GSF uses the naturally occurring iron to remove arsenic by coprecipitation. In this process it produces arsenic rich iron sludge. Disposal of this sludge to the nature may pose a risk of secondary contamination to the stratum. Leaching tests of the arsenic rich iron sludge and the solidified sludge with cement indicates that arsenic leaching ratio of sludge ranges between 3 and 13 % and that of cement stabilized sludge is between 1 and 6 %. This means that the arsenic has a strong bond with the iron sludge and its treatment may not be difficult.

KEYWORDS: arsenic, cement stabilization, leaching, sludge

Introduction

Arsenic contamination in the groundwater of Bangladesh is the most catastrophic arsenic poisoning in the history of water supply. According to the survey report conducted by the government and British Geological Survey, an estimated 35 million people are exposed to the risk of arsenic greater than 50 ppb concentration (Bangladesh standard for arsenic in potable water) in the drinking water (BGS, 2001) (Ahmed 2003). The arsenic containing sediments that formed this deltaic plain causes this pollution (Islam and Uddin 2003; Kinniburgh et al. 2003). The majority of the rural population of Bangladesh depend on groundwater for domestic water requirement (Ahmed 2003). Therefore, it is very important to find a way out to get safe water with arsenic below the acceptable concentration for drinking water. One alternative may be the use of surface water, but surface water is polluted by microbial contaminants, which also needs treatment. Besides, due to the increase of pisciculture in the ponds and other water bodies it is difficult to find a suitable source of surface water, because, those water are chemically and biologically loaded with fish feed. To find a sustainable solution to this problem, many government and nongovernment organizations are presently working in Bangladesh.

Background

Several laboratory researches have been conducted by the researchers aiming the use of naturally occurring iron for arsenic removal from iron-rich groundwater of Bangladesh. Mamtaz and Bache (2001) demonstrated that adsorption and coprecipitation with iron followed by settlement could remove significant amounts of arsenic provided that there is sufficient iron and a sufficient settling time (≥ 3 d). They mentioned this method having considerable promise as a low-cost technique because of the total absence of added chemicals. Leupin and Hug (2005) showed that a few cycles of aeration of synthetic groundwater and subsequent filtration through quartz sands containing zero-valent iron fillings could bring arsenic down from 0.5 mg/L to below 0.05 mg/L. Along with arsenic, most of the added phosphate in the synthetic water was also removed by this process without an added oxidant.

The university of Miyazaki has been working on the cause and remediation of arsenic poisoning in

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Bangladesh since 1997. Pond sand filter (PSF) has been developed and constructed in some severely arsenic affected villages to provide safe water to the villagers. Those PSFs were supplying safe water to the villagers with an excellent performance (Yokota et al. 2001). But in dry season these units cannot work properly due to the lack of water in the ponds. This situation compelled the users of those units to use the arsenic contaminated tube well (TW) water. Reexcavation of the ponds is expensive and might not be sustainable. So, it was necessary to find an alternative solution to this problem of the PSF.

The PSFs had a horizontal roughing filter attached to it as a pretreatment unit for the high turbid pond water. The interparticular spaces of the gravels in that roughing filters worked as a mechanical strainer as well as a bunch of mini settling basins to remove the turbidity. It was thought that settling basin property of the roughing filter could be utilized to remove the settleable arsenic rich iron flocs, which can be produced as a result of oxidizing the iron rich ground water of Bangladesh. This hypothesis was tested in laboratory; later, batch tests in the rural Bangladesh showed positive results regarding arsenic removal from iron rich ground water by oxidizing it and passing it through the similar setting of the roughing filters (Hamabe et al. 2004). Based on those test results an arsenic removal unit, structurally similar to the PSF, has been developed to overcome the low flow problem encountered by it during dry season. It was constructed at Marua village of Chougacha upazila of Jessore district in eastern Bangladesh (near 23° 15.25'N, 89° 05'E as observed on the "small area atlas of Bangladesh" published by Bangladesh Bureau of Statistics, 1989).

The developed unit has operated about two years solely with groundwater to evaluate its long term performance as well as to adjust its setting for the finest performance. The groundwater contained arsenic conc.=0.217 mg/L, (Std. Dev.=0.038, averaged from 30 measurements, by atomic absorption spectrophotometer, Shimadzu AA 680) and total iron conc.=4.68 mg/L, (Std. Dev.=1.03, averaged from seven measurements by spectrophotometer Hach DR2010, Shimadzu UV-1600) but there is temporal variation of concentrations of these parameters (BGS 2001). In this paper, performance of GSF has been discussed briefly. The unit produces arsenic rich iron sludge in the process of arsenic removal, which is washed out periodically. The washed sludge is allowed to settle in an underground reservoir where other natural debris, i.e., washed soil particles, leaf, etc., also mixes up naturally. That sludge has been collected and tested for its arsenic leaching characteristics. The leaching test results have also been presented in this paper. But before going into that discussion, the working principle of the GSF will be discussed.

Gravel Sand Filter

The gravel sand filter (GSF) consists of four major units: inlet, gravel chambers, slow sand filter (SSF), and reservoir (Fig. 1). Water from the tubewell passes through gravel roughing filters and the sand filter before reaching the supply tap. Water flows from the pump to the inlet chamber and then flows to the gravel tank/chamber 1 through the bottom holes of the partition wall between them. Average diameter of gravel in the gravel chambers and outlet are about 5 mm but drain section at the bottom of the chambers contain larger gravels up to an average diameter of 20 mm. Water flows diagonally along the chamber to reach the holes, located near the upper part of the partition wall between gravel tank 1 and gravel tank 2. In gravel tank 2, water flows diagonally downward and enters into the outlet chamber of the roughing filters through the bottom holes in the partition wall. Then water spills over to the sand filter through a groove in the partition wall between them. In the sand filter water flows vertically down and is collected through the bottom drainage section consisting gravels and goes to the reservoir section. The users collect water by opening the tap of the reservoir/supply tank.

In this process, water gets aeration in the inlet chamber which helps chemically (Deutsch 1997) turning divalent iron (Fe^{2+}) and trivalent arsenic (As^{3+}) to their oxidized trivalent iron (Fe^{3+}) and pentavalent arsenic (As^{5+}) compounds respectively. In the prevailing pH range of the water, Fe^{3+} forms relatively insoluble ferric hydroxide [$\text{Fe}(\text{OH})_3$] (Peavy et al. 1985). Arsenic ions have strong adhesion toward the $\text{Fe}(\text{OH})_3$ particle surface and hence are adsorbed there (Ali and Ahmed 2003). Then, while flowing through the gravels in the roughing filters, the particles experience sudden constrictions, leading to flocculation, and then wider areas, leading to slower flow and hence, settling. Thus the arsenic containing iron sludge is being deposited in the inter-particular space of the gravels and relatively arsenic free water passes through the next sections. Thus arsenic is being removed from groundwater. In this process the gravel chambers gradually get clogged with accumulated arsenic rich iron sludge and eventually stops the flow. In such situations when flow rate falls, bottom drain valves of the gravel chambers are opened for some time to allow the sludge being washed out. The sludge is accumulated all over the gravels in the gravel

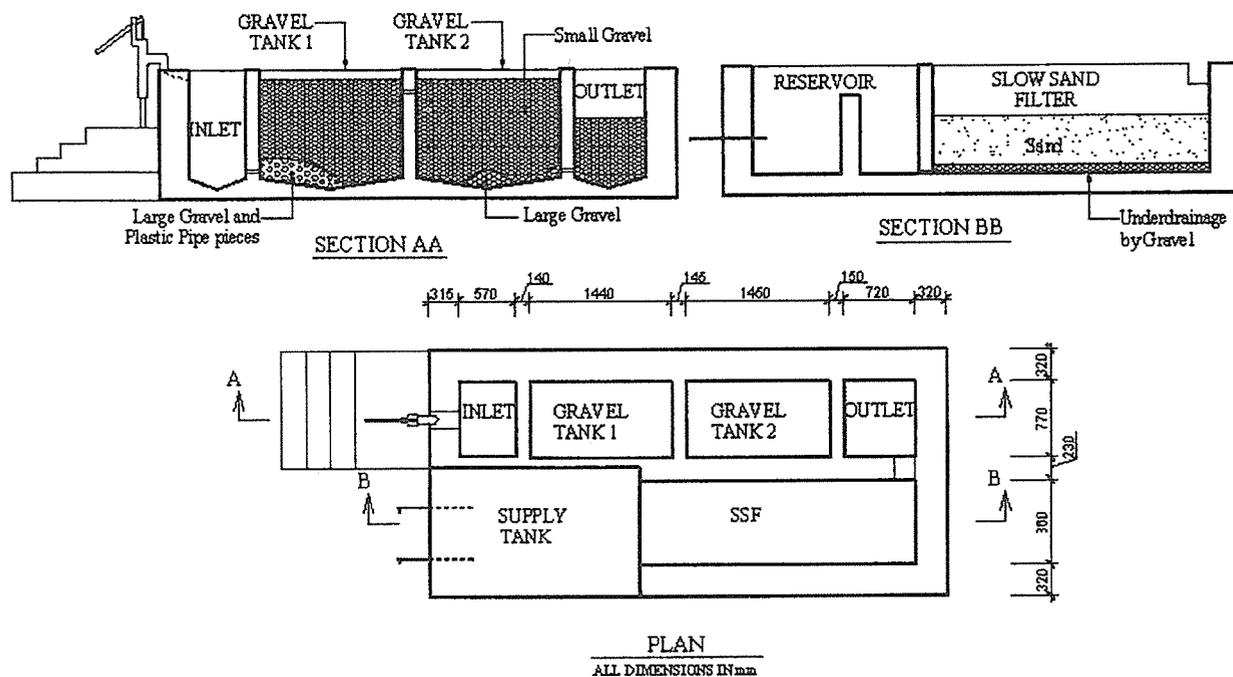


FIG. 1—Diagram of the modified gravel sand filter (GSF).

tank. Cleaning by backwashing is not enough to bring out all the sludge from all the parts of the gravel tank. Only sludge accumulated near the drain zone is being washed out. Majority of the sludge being accumulated near the bottom due to gravity force, so, small amount of sludge remains accumulated in the gravel after the backwashing. But after several backwashing the accumulated sludge in the upper part of the gravel tank becomes large enough to clog the flow. Therefore, thorough cleaning of the gravel, by taking them out of the tank and placing them inside after cleaning becomes necessary. In the field such severe clogging occurred almost once in every three months.

The washed sludge flows to an underground reservoir, where it settles down along with other debris that may fall in the drains and the reservoir from the surroundings. The supernatant water from that underground reservoir flows to a lined pond for expected further bioremediation (Stolz et al. 2002) before flowing to a natural pond. The method of monitoring and maintaining along with the data analysis has been provided in the following sections.

Methods

Field and laboratory measurements were performed for several parameters, i.e., dissolved oxygen content (DOC), oxidation-reduction potential (ORP), arsenic (As) and iron (Fe), from different sampling points throughout the unit. Sampling points in the gravel chambers were pre-installed vertical PVC tubes of 38 mm diameter reaching at different depths into the gravels. Before the measurements, tubes were purged to allow fresh water from the gravel inside the tube. The analysis of the collected data gave a better understanding of the processes going on inside the newly built unit during December 2003 to January 2004 period. During another field visit and measurements in January 2004, after one year of operation, some modification/rearrangements were made to solve some maintenance problems encountered by the users.

The modification is done because the users reported about clogging problem of the unit. To reduce clogging the pore volume of the bottom large gravels in the drain section is increased by introducing small pieces of polyvinyl chloride (PVC) plastic pipes (Fig. 1). PVC pipe of 18 mm diameter has been cut into 25 mm long pipe pieces; to make such cutting locally available cutter used for cutting PVC pipe strainer openings was used. Therefore, the 20–30 mm gravels, used at the bottom drain zone of the gravel tank, had similar sizes of those small pipe pieces. It was expected that a pipe piece would occupy almost same volume as large gravel, but would leave a considerable amount of empty space inside the hollow cylindrical piece helping an increase of pore volume of the drain zone. Any ratio of mix would increase the porosity by some degree and this presumption has been checked out by a simple test where equal volume

of cut pieces of PVC pipes has been mixed with gravel, all measured by buckets. The volume of water required to overflow a bucket filled with (a) only large gravels and (b) large gravel and pipe piece mix has been measured. After five trials the measured average water volumes required were 37.2 and 56.3 % of the bucket volume, respectively. A comparison of the porosity of coarse gravels (Todd 1980), which shows 28 % porosity for 16–32 mm sized gravels, ensures the justification of this measurement as the samples used here were more uniform in particle size. This test verified the expectation of porosity increase, which was hoped to solve the frequent clogging problem of the gravel filter. A longer clogging cycle was expected by this modification.

Periodical (twice a month) water samples were also collected from different parts of the unit and arsenic concentrations was checked to track the malfunctioning part as well as the efficiency of the unit. Data showed a malfunction of the unit causing increased arsenic concentration in the supply than the concentration of arsenic in water before it passed the sand filter. Investigation showed that it was due to the clogging of the sand bed with finer particles. Along with problems of lower flow it created a reducing situation inside the sand causing arsenic leaching back to the water. This problem was solved by periodical scraping of the sand surface (Hussainuzzaman et al. 2004). Among other parameters, total daily water consumption by the users is recorded from an attached water meter at the supply tap. During field measurements with the unit, a model pumping pattern, consisting of a two minute pumping followed by a three minute rest, was maintained with a view to mimic the actual stop-go-stop type flow experienced by the unit.

Besides these water samples for arsenic and flow data, samples were also collected from the accumulated sludge in the underground reservoir, once a month. The sludge tank is cleaned twice a year hence the collected sludge sample comprises of sludge from different months washouts. Sludge samples were then allowed to settle for 2/3 days in glass containers and the supernatant water had been spilled out. Then the semisolid compound has been stored in laboratory grade airtight plastic containers and brought back to the laboratory of the university of Miyazaki at Japan. The semisolid materials were air-dried and then oven dried for about 24 h in 105 °C. The dried material is then crushed in sieved to take out particles larger than 2 mm. Samples were then tested for total arsenic determination, leaching tests with and without cement solidification.

Cement stabilized sludge samples were prepared by hand mixing the dry sieved sludge with ordinary Portland cement at three different cement to sludge ratios (wt. cement/wt. dry sludge): 0.02, 0.1, and 0.2 respectively. Then distilled water was used to provide hydration water at a water to sludge ratio (wt. water/wt. dried sludge) of 0.2. They were mixed thoroughly with steel spatula and stored in airtight plastic containers for 28 days and then pulverized before leaching test. The leaching tests were done according to the Japanese standard procedure for leaching test (JLT 46³), which includes: oven drying sludge and smash it to take only that part having a maximum particle size of 2 mm; then put distilled water at a solid/water mass ratio of 0.1 and then put it into a shaker with 200 cycle/s, 4–5 cm shaking for 6 h; then the water is separated from the solid by centrifuging it at 3000 rpm for 20 min; then the liquid part is filtered by 0.45 µm filter paper to get the leachate; the leachate was tested with a Hydride-generation atomic absorption spectrophotometer (AAS, Shimadzu AA-6650).

Determination of total arsenic content of the sludge was performed following Japanese standard process of solid extraction (JLT 46). In this process digestion of the soil sample (2 g) was performed with concentrated nitric acid (15 ml) and 1:1 sulfuric acid (15 ml) on a hot plate (below 300 °C). More nitric acid (10 ml) was added when the liquid is decreased approximately to 15 ml. This process was repeated until white smoke came out following brown gas. Then for two or three times distilled water (15 ml) was added with the cooled liquid and heated again. The mix was then cooled at room temperature and was being filtered with a glass fiber filter (0.45 µm), volume adjusted to 100 ml with distilled water and stored for testing with AAS.

Sludge samples were tested for total arsenic content as well as for the leaching properties of arsenic. Leaching test data for sludge along with its stabilization experiments with cement has been presented in the following sections. Chronological arsenic removal performance data before and after increasing the gravel pore volume by applying PVC pipe pieces (as mentioned before) has also been presented and compared.

³Association for Environmental Measurement & Analysis in Japan. *The bottom sediment test methods* (as listed in the environmental agency notification no. 46; published in 2001, pp 1511–1514, in Japanese)

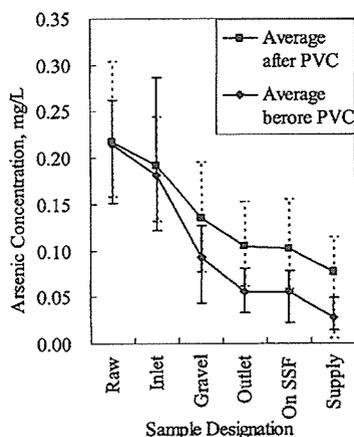


FIG. 2—Comparison of arsenic removal performance before and after putting PVC pipe pieces at the drain section of the gravel tank 1; error bars indicate data range.

Results and Discussions

Arsenic Removal Performance

Arsenic removal performance data is obtained from the periodical samples' test results by hydride-generation atomic absorption spectrophotometer (AAS; Shimadzu AA-660). Results show that the arsenic removal performance met the Bangladesh standard (concentration below 0.05 mg/L) before the application of the PVC pipe pieces. But after this modification to reduce clogging problem, the arsenic removal performance deteriorated (Fig. 2).

In Fig. 2, Raw indicates the raw water sample; Inlet indicates the sample obtained from the mid-depth section of the inlet chamber; Gravel indicates the sample obtained from gravel tank 1, near the holes through which water passes from gravel tank 1 to gravel tank 2; Outlet indicates the sample from the surface of the outlet section, just before overflowing to the SSF; and, Supply indicates samples collected from the supply tap. Average after PVC indicates the average arsenic concentration data of the samples collected after applying PVC pipe pieces, it comprises of 18 data points from February 14, 2004 to November 21, 2004. On the other hand, Average before PVC is developed from the average data before that modification with PVC pipe pieces; it comprises of 11 data from July 12, 2003 to January 13, 2004. The figure shows that the slope of the lines from outlet to supply remains the same before and after the modification. The deterioration is concentrated around gravel chambers, which have some influence over inlet and outlet sections.

The reason for this deterioration might be explained by the changed sludge accumulation pattern as observed during cleaning of gravel tank due to low flow situation. A thick, sticky mud like sludge cluster was observed in the gravels near the inlet holes, spreading about 20–50 cm from the holes as the gravel chambers were cleaned with the initial setting. In that situation, the passing water and oxidized iron flocs and arsenic were forced to pass through that sludge-packed region, which resulted high removal efficiency. On the other hand, when the clogged gravel tank was cleaned after applying PVC pipes, absence of such sticky mud like sludge cluster was noticed within the initial 0–15 cm from the inlet holes. Instead, thick sludge cluster was observed about 15 cm away from the inlet holes spreading along the drainage zone. This change of sludge accumulation pattern might have resulted from the difference of porosity between the small gravels and the large gravels in drain zone. Before applying the PVC pipes, the pore fraction of the large and small gravels were almost the same (measured average value, 35.2 % in small gravels and 37.2 % in large gravels). Therefore, sludge tended to settle at the vicinity of the entrance holes of the gravel tank 1. But with the new setting the drainage section near the bottom had more empty spaces for sludge accumulation. Therefore, most of the sludge bulk was flown to and being accumulated in that region leaving the entrance hole vicinity relatively clean. Water was not forced to pass through the gravels and thus chance of contact between arsenic and iron sludge became low resulting reduced removal and efficiency. Besides, channeling within the accumulated sludge in relatively large pore space might have played a role in this case (Ali et al. 2001).

TABLE 1—Arsenic content test result for sludge.

Sample ID	Source GSF No.	Collection date	pH	Arsenic content, mg/kg
1-6	GSF#1	28-Jun-2003	6.9	66.47
1-7	GSF#1	27-Jul-2003	6.7	48.16
1-8	GSF#1	30-Aug-2003	6.8	68.49
2-6	GSF#2	28-Jun-2003	7.0	10.19
2-7	GSF#2	27-Jul-2003	7.1	11.80
2-8	GSF#2	30-Aug-2003	6.9	18.97

Besides arsenic, total iron concentration of the treated water was below 0.01 mg/L (measured by Hach DR2010). In fact, almost all the iron is removed before water reaches the second gravel tank (in Fig. 1). Measured oxidation-reduction potential (ORP) of groundwater always was negative ($ORP < -100$) indicating a reducing state but treated water had positive ORP all the time. The ORP shifts from negative to positive indicating reducing to oxidizing state in the gravel tank 1 and continues rising along the water flow path at gravel tank 2. Dissolved oxygen content ($DOC = 1.3$ mg/L) showed a fall of its concentration along the water path through the gravel tanks as the DOC being used up by iron and arsenic as well (Hussainuzzaman et al. 2004). During these field measurements (December 02–January 03 and January 04) model pumping flow rate was approximately 720 litres/hour. The flow meter data (October 31, 2004 to November 21, 2004) reveals that average daily water consumption was about 1500 liters.

Arsenic Leaching Test from Sludge

The iron sludge accumulates arsenic and hence it may pose a risk of secondary contamination if not disposed properly. Generally the arsenic concentration of the soil stratum in Marua village is very small. Most of the arsenic is contained in a clay layer, where the highest arsenic concentration is about 20 mg/kg (Tanabe et al. 2001). Therefore, delivering this arsenic bearing iron sludge to the surrounding soil may not be a good option of disposal because of probable leaching of arsenic. Ali et al. (2003) checked the leaching of arsenic from some common arsenic treatment units operating in Bangladesh and found by the TCLP test that, in general, leaching of arsenic from the wastes was not significant and that none of the waste samples were “hazardous” as defined by the USEPA. But leaching with groundwater showed higher leachability of arsenic from some samples, by which, they suggested that TCLP test did not appear to be appropriate for assessing long-term leaching of arsenic from treatment wastes. Badruzzaman (2003) showed that in the field disposal conditions, as practiced by the users of arsenic removal systems in rural Bangladesh, a significant amount of arsenic is lost from sludge through biomethylation induced by the presence of organic matters.

Therefore the arsenic bearing sludge should be stabilized by some means. Jing et al. (2005) stated that cement treated arsenic sludge showed reduced leachability of As(III) in TCLP tests. They also showed that longer curing time of cement stabilized waste material reduces the As(III) leachability further. Phenrat et al. (2005) showed that arsenic can be chemically fixed into cementitious environment of solidified/stabilized matrices by three important immobilization mechanism; sorption onto C-S-H surface, replacing SO_4^{2-} or ettringite, and reaction with cement components to form calcium-arsenic compounds, the solubility limiting phases. Dermatas et al. (2004) checked the arsenic leachability of quicklime-fly ash stabilized field and artificial soil samples in a semidynamic leaching test by measuring the effective diffusion coefficient. They found that solidified/stabilized samples had lower leachability of arsenic. They also found that precipitation is the dominant mechanism of arsenic immobilization. Mollah et al. (2004) showed that the early hydration of cement inhibited by the presence of AsO_4^{3-} , and that the inhibition is mainly caused by the formation of highly insoluble $Ca_3(AsO_4)_2$ on the surface of hydrating cement particles. They concluded that Portland cement may be considered as a potential matrix to immobilize As(V) bearing wastes.

Therefore it can be expected that stabilizing the arsenic rich sludge with cement can reduce leaching of arsenic from it to the environment. To evaluate the possibility of arsenic leaching and its stabilization with the prevailing situation, leaching tests were performed with collected sludge and cement stabilized sludge. Test results are presented in Tables 1–3.

Table 1 shows the total arsenic content in the sludge along with its pH. Table 2 shows leaching of arsenic from the sludge, cement stabilized sludge (containing 2 and 10 % cement), pH of the leachate, and

TABLE 2—*Elution test result for arsenic sludge.*

Sample ID	Arsenic leaching							
	Sludge Only		sludge+2 % Cement			Sludge+10 % Cement		
	mg/kg	%	pH	mg/kg	%	pH	mg/kg	%
1-6	2.04	3.08	10.5	1.09	1.64	11.8	0.80	1.20
1-7	2.04	4.28	10.7	0.93	1.94	11.8	0.82	1.71
1-8	2.72	3.97	10.7	1.25	1.83	11.8	0.86	1.25
2-6	0.91	8.92	10.9	0.59	5.83	11.8	0.35	3.42
2-7	1.51	12.82	10.8	0.51	4.30	11.9	0.41	3.45
2-8	1.18	6.21	10.9	0.78	4.09	11.8	0.44	2.31

the percentage of total arsenic as shown in Table 1. Table 3 contains data of tests with a different set of samples. It contains the total arsenic content of the sample along with the leached arsenic from sludge without any cement and with 20 % cement.

GSF 1 refers to the GSF of this discussion and GSF 2 is another GSF located in the same village but with different design, input water quality and number of users than GSF 1. Results show wide variation of total arsenic content of the collected sludge. This variation is not easy to explain because in reality the sludge settles in the underground reservoir in an uncontrolled environment. Therefore, the amount of foreign materials, carried by runoff from its surrounding, varies with time and level of maintenance of the unit. Besides, the settled sludge may have spatial variation in concentration along the reservoir bed because of the point source with respect to the reservoir. Therefore two grab samples, collected simultaneously from different locations at the reservoir bed, may result in different concentrations of total constituents as well as leaching.

Result of the test (Table 2) shows that 3.8 to 12.82 % of the arsenic leaches out of the sludge. Therefore 85–95 % of the arsenic is confined into the sludge. The leaching figures ranging from 0.12 to 0.27 mg/L meet the criteria for industrial sludge disposal site in Japan but exceed the World Health Organization (WHO) guideline value for leaching (0.01 mg/L) by a wide margin.

The decreasing tendency of arsenic leaching with increasing amount of cement (Table 2 and Fig. 3) associates higher pH. In this condition calcium (source=Portland cement) arsenate solids may have formed, which are stable under this condition and hence prevent the release of arsenic (Khoe et al. 1997; Mollah et al. 2004). With a different batch of samples, an increased amount of cement has been used to further verify the stabilization effect. The results are very much in line with the previous results as shown in Table 3. Therefore, cement stabilization of sludge might be a very effective way for safe sludge disposal in the prevailing field condition. Carbonation of the cement-stabilized sludge in humid conditions might reduce this high pH associated with the use of Portland cement (Gervais et al. 2004).

Conclusions

This study shows the long term performance data of a community based arsenic removal unit which was developed to solve the low flow problem encountered by the existing PSFs during the dry season. The long-term data shows that the unit performs efficiently to remove arsenic from tubewell water (raw water arsenic=0.217 mg/L) and produces safe drinking water (arsenic concentration below 0.05 mg/L). This process is very cost efficient as no additional chemical is needed and it uses the naturally occurring iron of groundwater. The only problem associated with the system is maintenance, which includes clogging of the

 TABLE 3—*Leaching test results for cement stabilized and nonstabilized arsenic sludge.*

Sample ID	Collection date	Total arsenic content, mg/kg	Leaching, mg/kg		Percent leaching		pH condition	
			Without cement	With 20 % cement	Without cement	With 20 % cement	Without cement	With 20 % cement
GSF-1	26-Jun-03	269	1.08	0.04	0.40	0.01	7.53	12.26
GSF-1	31-Jan-04	81	6.43	0.12	7.94	0.14	7.75	12.29
GSF-1	27-Mar-04	19	1.36	0.10	7.17	0.53	8.13	12.31
GSF-2	26-Jun-03	24	0.46	0.02	1.89	0.10	7.65	12.19
GSF-2	31-Jan-04	228	1.49	0.47	0.65	0.20	7.80	12.24
GSF-2	29-May-04	55	4.60	0.21	8.42	0.39	7.85	12.09

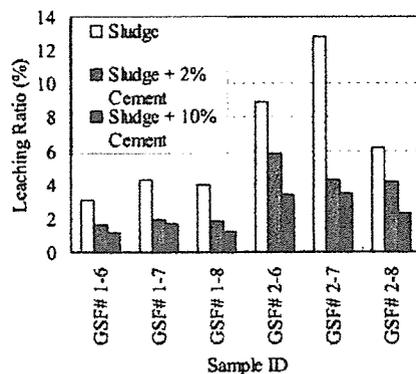


FIG. 3—Comparison of arsenic leaching from stabilized sludge with different cement content.

gravel filter requiring thorough cleaning of the gravels once about every three months. Therefore, to make the system more efficient and to use it solely as a good arsenic removal unit, this clogging problem should be solved or an easy maintenance system should be developed.

The unit produces arsenic rich iron sludge, which may pose a risk of secondary contamination of the environment by the leached arsenic if not disposed properly. This study also checked the possibility of cement stabilization of the sludge in the prevailing condition. Data shows that arsenic leaching has been reduced by solidification/stabilization with cement. An increase of cement amount in the stabilization process increased the stabilization efficiency as less arsenic is leached out in that case. The first batch experiments showed arsenic leaching between 3 and 13 % and for raw sludge while stabilization with 2 % cement reduced that to 1.5–6 % (Table 2). A further increase of cement to 10% has decreased leaching to 1–3.5 %. Use of 20 % cement for stabilization with a different sample set showed a decrease of leaching from the 0.4–8.5 % range to the 0.01–0.55 % range. Higher pH associated with the increase of cement for stabilization seems to be a common problem like any other material containing cement. Therefore it appears that safe disposal of arsenic rich sludge may not be a problem.

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