

# A Low-Tech Management Option to Remove Arsenic-Rich Wastes Derived from Arsenic-Iron Removal Plants in Rural Bangladesh

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**Abstract:-** Exposure to naturally occurring arsenic in groundwater is a serious concern for many residents of rural Bangladesh both in their drinking water and their foodstuffs. Arsenic-Iron Removal Plants (AIRPs) have provided a simple and relatively inexpensive method to remove arsenic from groundwater using AIRPs, a decentralized water treatment system; however, the arsenic-rich wastes removed from the drinking water then involve the resulting arsenic wastes being released back to the environment from which they came, and hence, not well managed. Measurements of representative wastewaters from AIRPs indicate arsenic concentrations between 210 µg/L and 5900 µg/L, all of which exceed the Bangladeshi standard of 200 µg/L for discharge to inland waters. Results from a novel and simple soakaway pit design containing a layer of 5 cm of brick chips, followed by a layer of 10 cm of sand, are shown as able to remove more than 95 % of the arsenic from the arsenic flocculant collected during cleaning of the AIRPs. Simple is good, as this low-tech technology will provide an effective opportunity to avoid having the arsenic re-enter the ambient environment from which it came.

**Keywords:** Arsenic, Arsenic-Iron Removal Plant (AIRP), wastewater, Bangladesh

## 1. INTRODUCTION

Significant concentrations of naturally occurring arsenic have been reported in the groundwater in many countries, particularly Bangladesh, India, Cambodia, Vietnam, and Nepal. As an indication of the extent of the problem with natural arsenic, it is estimated that 42 – 60 million Bangladeshi people now consume water at arsenic levels greater than 10 µg/L [1, 2]. Bhattacharya et al. [3] referred to the arsenic contamination in the Bengal basin as ‘possibly one of the worst environmental catastrophes in the history of human civilization’. Smith et al. [4] described it as the ‘worst case of mass chemical poisoning in the world’ and the situation of arsenic being a major public-health problem is continuing [5]. There are some areas in Bangladesh where the arsenic concentration has been observed to approach 2 mg/L [6].

In response to the exposures from naturally occurring arsenic in shallow groundwater in Bangladesh, a technology now being fairly widely utilized is the Arsenic-Iron Removal Plant (AIRP). The AIRP is composed of two circular concrete tanks on a platform and functions on the principles of oxidation and co-precipitation (see Figure 1 and Photo 1). Ferrous iron is oxidized by using an aeration tray in Tank 1, forming insoluble iron oxide flocs, to which arsenic adsorbs [7,8]. Filtration media composed of brick chips, charcoal, and coarse sand removes the flocs in the up-flow tank (in Tank 2). Nevertheless, while the AIRP has been installed extensively in arsenic-prone regions of Bangladesh, there has been minimal attention given to management strategies for the arsenic-rich wastes that are generated by the AIRPs in the flocculant; these arsenic-enriched wastes are mostly just released back to the environment during the necessary periodic cleaning of the AIRPs.

The co-precipitated flocs of iron and arsenic form a fine sludge, (i) at the bottom of Tank 1 and Tank 2 of the AIRP and (ii) within the filtration media in Tank 2. Regular cleaning removes these sludges by opening the valves at the bottom of the two tanks and physical removal of the sand and charcoal media by hand washing. During the cleaning, both Tanks 1 and 2 are flushed until the water runs clear, out of the drain valves (i.e. no sludge/flocculant visibly remains). A drainpipe moves the wastewaters arising from these cleaning processes away from the AIRP platform and in most cases, the pipe drains toward a pond, river, or back to the adjacent agricultural soil or drainage ditches [7, 9].

A number of studies have dismissed the hazards of the waste produced from the AIRP [10, 11, 12, 13]. These studies are based on the toxicity characteristic leaching procedure (TCLP) developed by the USEPA and generally suggest that the leachate derived from the arsenic-rich wastes of AIRPs does not contain high levels of arsenic. While scientifically valid re the TCLP, these studies are misleading, as the conditions surrounding disposal of arsenic-rich wastes in rural Bangladesh are not reflected by the TCLP since the TCLP procedure was designed for use as a leaching test simulating the acidic-forming conditions inside municipal solid waste landfills, instead of focusing on the issues relevant to evaluating leaching from arsenic-rich wastes in the context of rural Bangladesh [11,14]. For example, [10] found that use of distilled water, groundwater, rainwater, or pond water in place of the TCLP extraction fluid caused the AIRP wastes to leach 26 – 33% of the arsenic present, in contrast to the 0.69 – 3.3 % leached under the TCLP. The leaching behavior of arsenic-rich wastes is complex and a function of both pH and other compounds in the surrounding environment.

Current disposal practices of AIRPs warrant attention to the manner in which arsenic-rich wastes are of increasing concern among Non-Government Organizations (NGOs) and AIRP beneficiaries. Even decades after the problem was identified, millions of people are still exposed to arsenic through their drinking water and food [15]. Poor management of arsenic-rich waste is both an environmental and human health concern [16] and the issue of arsenic poisoning and the need for mitigation strategies have been widely described (e.g. [17]). Disposal to land may cause phyto-accumulation in grains and vegetables, particularly rice, resulting

in foodborne exposure. Others have reported [e.g. 18, 19] that the body burden of arsenic in food consumed in rural Bangladesh is approximately twice the arsenic body burden from water. Hence, issues of waste management of the arsenic are of great importance to address, and the trends of arsenic exposure continue [20]. Moreover, arsenic concentrations as low as 10 mg/kg soil are capable of causing phytotoxic effects, such as significantly reduced crop yields [21]. Similarly, there is evidence that fish reared in ponds containing arsenic are capable of bioaccumulation of arsenic [22]. In response to these issues, the Government of Bangladesh created a standard stating that liquid wastes with more than 200 µg/L arsenic may not be discharged to inland surface waters or to irrigated lands.

Few studies have examined waste management strategies arising from cleaning activities of the AIRPs. Ahmed [23] recommended the use of a soakaway pit for capturing arsenic-rich wastes from the AIRP, but the effectiveness of such a device has not been tested, nor has the idea been widely implemented. The objective of this research was to characterize the wastewater arising from different aspects of the cleaning of the AIRP, to test the efficacy of a soakaway pit, and to evaluate methods for safe disposal of arsenic-rich wastes in rural Bangladesh. This study was accomplished using samples of wastewater from AIRPs in the village of Mohadevpur in rural Bangladesh, and by subsequently field-testing a soakaway pit methodology.

## 2. CURRENT WASTE MANAGEMENT APPROACHES

Capturing arsenic-rich wastes from the cleaning of the AIRPs requires that a long-term waste management strategy be developed to address the ultimate fate of the arsenic. A field sampling program was conducted wherein wastewaters developed during the cleaning of household AIRPs (350 L capacity) were collected in the village of Mohadevpur, ~ 75 km from Dhaka. These were wastewaters obtained as the water discharged from the AIRP cleaning process, which carries suspended arsenic-iron flocs from the AIRPs. Wastewaters were obtained from two different AIRPs. One was cleaned two months prior to our sampling, while the other had been cleaned three weeks prior to our sampling. To characterize the wastewaters in terms of concentration of arsenic and iron, samples were collected during routine AIRP cleaning by residents. The samples represented four stages of the AIRP cleaning:

- (i) first flush (see Figure 1 for locations here and below) - collected at the drain when the aeration tank was initially emptied and the wastewater was highly turbid, as the water level inside the tank had fallen to approximately 25 cm;
- (ii) second flush - captured at the drain when residents clean the aeration tank;
- (iii) sand cleaning - as the sand layer was physically removed for cleaning, these samples were collected from the pail containing the washed sand during the initial rinse; and
- (iv) brick chip cleaning - the sample was collected from the initial rinse of the bricks.

### 2.1 Waste Characterization

Wastewater samples resulted in arsenic concentrations ranging from 210 µg/L to 5900 µg/L. Concentrations of iron in wastewater samples ranged from 28 mg/L to 840 mg/L. The highest concentrations of both arsenic and iron were found in wastewater originating from washing the brick chips, followed by wastewater from the second flush, and sand washing. The lowest concentrations of arsenic and iron in the wastewater were obtained from the first flush. Figure 2 shows the average concentrations of arsenic and iron in wastewater originating from the four points in the cleaning process. All values exceeded the Bangladeshi standard for disposal to inland waters of 200 µg/L arsenic. These values provide a range of concentrations found in wastewater from the different aspects of the AIRP cleaning.

Additional insights in the arsenic levels from the AIRP were determined from back-calculation from average influent and removal efficiency values obtained from [24] found that 0.09 g arsenic, 17 g iron, and 4 g phosphate are released to the environment during the cleaning, assuming a cleaning cycle of four weeks and 50 L water use per day from the AIRP. This amounts to 1.2 g arsenic, 220 g iron, and 50 g phosphate generated from AIRP wastewater per year for the Mohadevpur region. In other areas of Bangladesh, it is not unusual for influent concentrations to exceed 300 µg/L arsenic, and in some cases 500 µg/L [4]. In addition, AIRPs that are community-based as opposed to the household-based AIRPs may have much greater volume throughputs than the household AIRPs characterized in this study.

These findings indicate that current practices of arsenic release back to the ambient environment of the AIRPs are highly undesirable. The following sections explore potential options for long-term management of arsenic-rich wastes originating from AIRPs.

### 2.2 Disposal to ponds with cow dung

The disposal of arsenic-rich waste to ponds with cow dung is often recommended under the premise that inorganic arsenic will undergo a biochemical reaction in the presence of microorganisms, creating a gas that is released to the atmosphere [10]. Studies examining the bacterial transformation of inorganic arsenic to volatile compounds show that arsine (AsH<sub>3</sub>), and mono-, di-, and trimethylarsines may be produced in variable proportions depending on the strains of bacteria present [25,26]. Studies suggest that a maximum of 35% of arsenic present may be volatilized [10, 26].

Following volatilization, the fate of arsine and methylarsine species is not well understood. [27] found that arsine gas resulting from semiconductor facilities in Taiwan oxidized to  $As_4O_6$  in the atmosphere.  $As_4O_6$  is thought to sorb to atmospheric particles or condense to arsenolite, which may dissolve in rainwater to produce arsenous acid ( $H_3AsO_3$ ). By this route, arsenic is returned to the earth's surface, or inhaled on airborne particles. The fate of methylarsines is also not well known, although it has been reported that demethylation may occur as a result of exposure to ultraviolet radiation [28]. As well, ponds are likely to create reducing conditions, which will cause the arsenic to desorb and mobilize back to the aqueous phase.

Since arsine and methylarsines are denser than air, disposal to ponds should be carefully weighed pertinent to the possible health hazards of these volatilized arsenic species. Arsine gas is considered highly toxic, causing hemolysis, subsequent hypoxia, and ultimately renal failure in acute cases [29]. Blackwell and Robins [30] report that exposure to 25-50 ppm of arsine gas for a half-hour duration can be fatal. In contrast, the toxicity of methylarsine species is not clear. Acute inhalation studies estimate a relatively low toxicity of  $LC_{50}$  of >200 000 ppm for trimethylarsine [31]. However, recent studies *in vitro* report that both dimethylarsine and trimethylarsine are strong genotoxins, exhibiting toxicity 100 times more potent than any other arsenic species [32].

Given the above, this method of disposal is considered undesirable as it creates new and uncertain routes of exposure which represent a potential health hazard.

For these reasons, this method of disposal is not recommended.

### 2.3 Solidification and Stabilization

Solidification and stabilization is a treatment technology used to restrict the mobility of arsenic in waste by encapsulation into cement or other materials. In many ways, solidification and stabilization presents an attractive option, as all of the materials required are affordable, locally available, and regularly used in rural Bangladesh. Moreover, the waste from a soakaway pit used to capture the arsenic would not need to be dewatered or processed before incorporation with cement.

Nevertheless, the disadvantages of solidification and stabilization include the occupational hazards associated with use of cement containing arsenic, and the uncertainty of long term leaching of arsenic in the cement. Significant health risk has been associated with the inhalation of dust containing arsenic, particularly during construction of the cement blocks [14].

While inhalation of dust containing arsenic may lead to the same cancers as ingestion of water containing arsenic, the inhalation can be mitigated by educating construction laborers on the hazards of arsenic-rich waste and by requiring that the cement be used in a context that will limit exposure by inhalation (for example, in the construction of latrines). However, the effectiveness of solidification and stabilization for immobilizing arsenic is uncertain over extended periods. Cement contains highly alkaline pore water (pH 13) that may cause desorption of arsenic from iron in the encapsulated waste [33]. The mobilized arsenic is thought to react with calcium to form calcium arsenates [33] and [12]. Although calcium arsenate is not readily soluble, some studies suggest the compound dissolves over the long term in the presence of carbonate, bicarbonate, carbon dioxide, or acidic conditions [34, 33]. Nonetheless, short-term studies show that solidification and stabilization is highly effective in capturing arsenic. A 24-hour study utilizing distilled water as extraction fluid showed that solidification and stabilization of arsenic-rich wastes with Portland cement and lime reduce leaching by three orders-of-magnitude when compared to leaching from untreated waste [35].

Some existing studies [e.g. 36, 34, 33] on leaching of cementous -wastes utilize leaching tests but future studies are needed to determine the leachability of arsenic wastewaters encapsulated in cement in a local environment. To mimic the leaching effects for rural Bangladesh, the standardized USEPA Synthetic Precipitation Leaching Procedure (SPLP) represents the best available option of existing leaching tests. The SPLP simulates the leaching that will occur from a material in the presence of slightly acidic rainwater (i.e. post-encapsulation in the bricks, once in use as a 'brick').

### 2.4 Vitrification

Vitrification, the process of capturing metals in a durable and leach-resistant mass through high temperature treatment [37], in this case referring to the incorporation of the arsenic-rich wastes into clay bricks. As with solidification and stabilization using cement, vitrification is desirable in that it is inexpensive and requires only locally available materials.

The disadvantages of vitrification are similar to those of solidification and stabilization, but few studies have investigated the effectiveness of vitrification for capturing arsenic-iron wastes, particularly not in Bangladesh. One study reports bricks with 10% arsenic-rich waste by volume (arsenic concentration of  $6.10 \text{ kg/m}^3$ ) fired for five hours at  $800 - 900^\circ\text{C}$  leached  $< 200 \text{ }\mu\text{g/L}$  [36]. Another study reports leachate values as low as  $10 \text{ }\mu\text{g/L}$  for bricks with 15 – 25% arsenic-rich waste (arsenic concentration of  $2400 \text{ mg/kg}$ ) fired at  $1000^\circ\text{C}$  for six hours [38]. The leaching capability of the bricks is cited to be strongly dependent on waste content, brick composition, and firing temperature and duration.

Volatilization of arsenic species during high temperature firing is an important consideration of the vitrification process. Clay bricks are generally fired at a temperature of  $700^\circ\text{C}$  to  $1100^\circ\text{C}$  [39]. Thurnau & Fournier [40] examined volatilization of arsenic during hazardous waste incineration and found that for an artificial soil matrix containing arsenic trioxide as 0.6 – 8.0 % of the arsenic present, volatilized over a temperature range of  $816 - 927^\circ\text{C}$ . For actual soil from a superfund site, the authors found that 5 to 19% volatilized. Similarly, Gray et al. [41] found that 40% of arsenic present in contaminated clay volatilized at  $1125^\circ\text{C}$ .

The behavior of arsenic adsorbed to iron oxides in clay firing has not been well documented and its behavior is largely unknown. Wang and Tomita [42] found that arsenic formed compounds with iron that were thermally stable up to  $1300^\circ\text{C}$  during coal combustion; however, this has not been confirmed for the arsenic-iron compounds present in arsenic wastes from AIRPs.

Moreover, it is important to emphasize that many of the facilities that currently fire bricks in rural areas are unlikely to have precise temperature control to manage volatilization or methods to capture volatilized species. As with the cement blocks, the clay bricks must be utilized appropriately to ensure that any exposure to arsenic (e.g. sorbed to dust) in the bricks will be minimized.

In addition to issues associated with volatilization, vitrification suffers from some logistical concerns. Vitrification of arsenic-rich waste in rural Bangladesh would require collection and transport of wastes to a central firing facility, whereas solidification and stabilization with cement may occur within villages. For this reason, vitrification would necessitate the development of a community waste collection program. While Johnston et al.[9] suggest that a community collection program would minimize resident exposure by utilizing trained technicians in waste collection, such a program would require financial motives to be successful. Technicians are unlikely to volunteer without some form of compensation.

### **2.5 Pits or Latrines**

In the absence of an alternative, arsenic-rich waste may be disposed to brick-lined pits or latrines. Although both pits and latrines may create reducing conditions that would allow arsenic to become mobile and to leach into the surrounding soil, lining the hole with a layer of clay could prevent this. Johnston et al. [9] suggest that sanitary latrines are normally isolated from shallow tube wells and, as a result, possible leaching of arsenic from latrines does not pose an immediate health concern. While disposal to brick-lined pits or latrines is not an appropriate long-term waste management strategy, this approach offers an improvement over disposal to ponds or fields.

### **2.6 Collection and Dispose to a Landfill**

The most straightforward strategy would involve establishment of disposal locations where the arsenic wastes could be transported to a centralized handling facility such as a regional landfill. Clearly, this approach would necessarily require some degree of funding but would have great benefit in terms of containment/removal of the arsenic.

## **3. FIELD-EXPERIMENTS USING A SOAKAWAY PIT METHODOLOGY**

Two novel and inexpensive model soakaway pits were constructed to investigate the potential use of a soakaway pit for capturing arsenic-rich wastes from AIRPs. Each model soakaway pit was constructed in a 15 L pail. Each pail was filled with 5 cm of brick chips, followed by 10 cm of sand, with the two media separated by nylon net (Figure 3). One pail contained fine sand ( $< 595 \mu\text{m}$ ) while the other contained coarse sand ( $850 \mu\text{m}$ ), locally known as “Sylhet sand.” Prior to assembly of the soakaway pit model, the sand used in the pails was washed until the water ran clear. Five holes of 2 cm diameter were added to the bottom of the pails to ensure that flow of water through the media was not restricted. An additional pail was placed under the model soakaway pits to capture post-filtration samples for analysis of performance.

To test the performance of the model soakaway pits, wastewaters as per above during the cleaning of AIRPs were collected in the field in 1.25 L quantities from five different AIRPs, where each AGRP was being cleaned 1 – 8 weeks prior to sampling. These wastewater samples were collected from periodic sampling during routine cleaning of the various AIRPs. This process occurred in 13 trials (7 trials using the fine sand, 6 trials using the coarse sand). Statistical significance tests were performed using the student's t-test [43] to determine if performance differed between the coarse sand and fine sand model soakaway pits.

Each model soakaway pit was cored after all testing had been completed with a PVC pipe of 2.5 cm diameter to determine where arsenic had proportionally been captured in the model soakaway pits. The core was divided into upper and lower 5 cm segments and solubilized in 200 mL acidified water ( $\text{pH} < 2$ ), agitated by hand for 10 minutes before the acidified solution prior to decanting to a sample bottle for analytical testing.

A graphite furnace atomic absorption spectrophotometer (Shimadzu AA6800) at the Bangladesh University of Engineering and Technology (BUET) was used to quantify arsenic and iron concentrations. USEPA Methods 206.2 and Method 200.9, both without heated digestion, were used to determine arsenic and iron concentrations respectively. Wastewater samples containing fine sediment were decanted in preparation for the atomic absorption spectrophotometer. All samples were collected in HDPE bottles.

To ensure that the materials used in the model soakaway pits did not contribute arsenic or iron to the effluent samples, blank samples of distilled water were passed through the model soakaway pits prior to all other samples. As QA/QC for the atomic absorption spectrophotometer, matrix spikes, blank spikes and blanks for both arsenic and iron were intermittently analyzed with groups of other samples.

## **4 RESULTS AND DISCUSSION**

### **4.1 Waste Capture**

The fine sand and coarse sand model soakaway pits showed arsenic capture of 97% and 96% respectively. Average iron capture was 97% and 91% for fine and coarse sand respectively. No significant difference ( $\alpha = 0.05$ ) was found between the capture rates of the fine and coarse sand for either arsenic or iron. Figure 4 shows the influent and effluent concentrations of arsenic before and after movement through the model soakaway pits. Effluent leaving the soakaway pits reached a maximum of 71  $\mu\text{g/L}$  arsenic and 10  $\text{mg/L}$  iron.



Coring the model soakaway pits revealed that 88% and 95% of the captured arsenic was held in the top 5 cm of the fine and coarse sand, respectively. Similarly, 86% and 73% of the captured iron was sorbed in the top 5 cm of the fine and coarse sand model soakaway pits, respectively. Since only one core was possible, these values should not be considered precise but they indicate that the upper layer of sand was responsible for the substantial majority of arsenic and iron capture, and that a large amount of sand remains to capture a high proportion of the waste for continuing use during successive periodic cleaning of the AIRPs in the field. The model soakaway pit concept may be easily scaled up to capture the volume of the household AIRP. Figure 5 shows one such possibility. As a management alternative, the design presented in Figure 5 directs wastewater towards the soakaway pit. Coarse sand (of the two options within these analyses) was selected because it allows rapid drainage without a significant decrease in arsenic capture.

## 5. CONCLUSION

With increasing awareness of alternative routes of exposure to arsenic and the identified high arsenic levels in both drinking water and vegetables irrigated with arsenic-lade irrigation water, there is a growing need to develop a waste management strategy for decentralized AIRPs in rural Bangladesh. The study presented here examined wastewater collected during routine cleaning and found that it contained 210 µg/L to 5900 µg/L arsenic and 28 mg/L to 840 mg/L iron. Use of simple but effective soakaway pits composed of 5 cm brick chips and 10 cm sand showed > 95% arsenic removal and > 90% iron removal from wastewater. These results indicate that a scaled up soakaway pit, adapted to the volume of an AIRP, would capture and retain the arsenic-rich waste present in wastewater in rural Bangladesh. Transport of this captured arsenic to a reasonably secure landfill represents an important opportunity to remove arsenic generated from the AIRPs, and thereby prevent the flocculant arsenic from re-entering the foodchain and drinking water, show potential to fill this long term management role using this simple approach.

## 6. ACKNOWLEDGEMENTS

The authors gratefully acknowledge the research funding provided by the Natural Sciences Engineering Research Council and scholarship funding from Ontario Graduate Studies. Participation by Mujibur Rahman of Bangladesh University of Engineering Technology is greatly appreciated.

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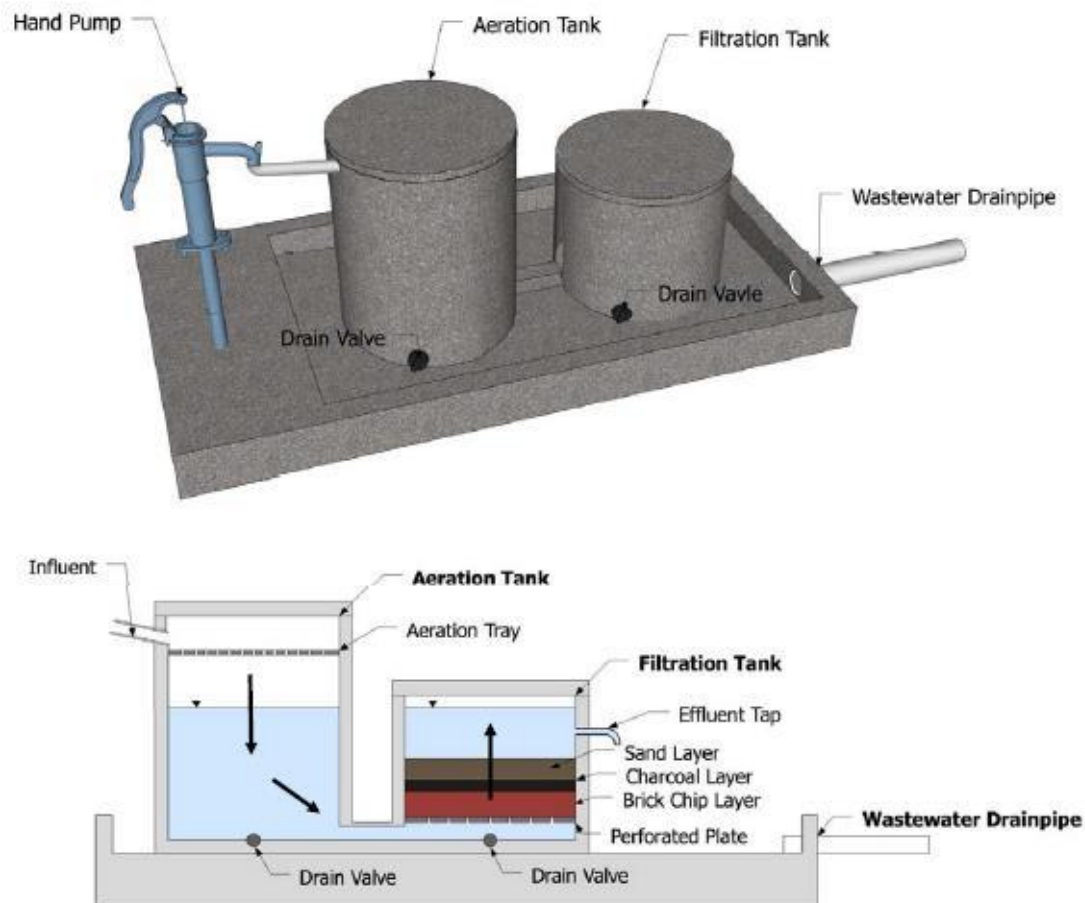


Figure 1. External and internal schematic of AIRP

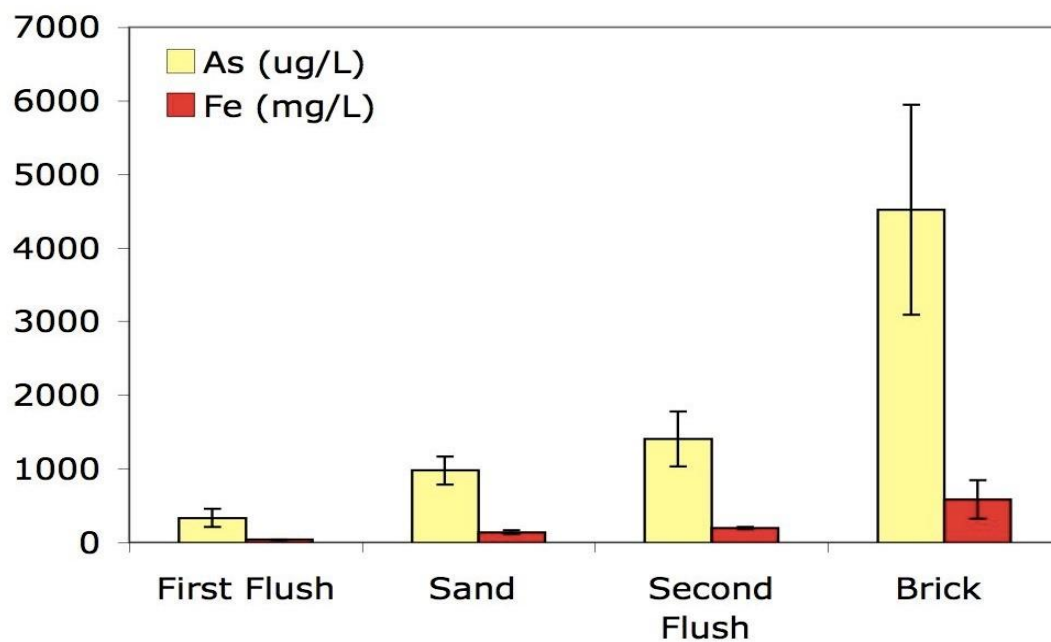


Figure 2. Average arsenic ( $\mu\text{g/L}$ ) and iron ( $\text{mg/L}$ ) concentrations in wastewater resulting from different steps in the cleaning process. The error bars show the range of values found.

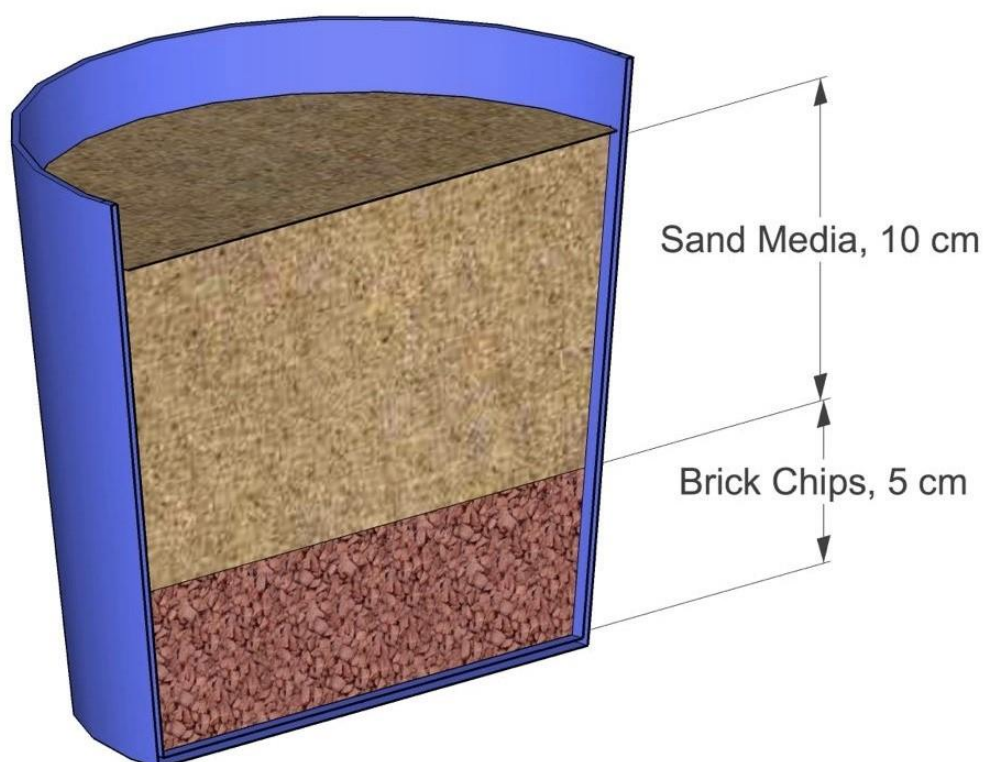


Figure 3. Cross-section of the model soakaway pits



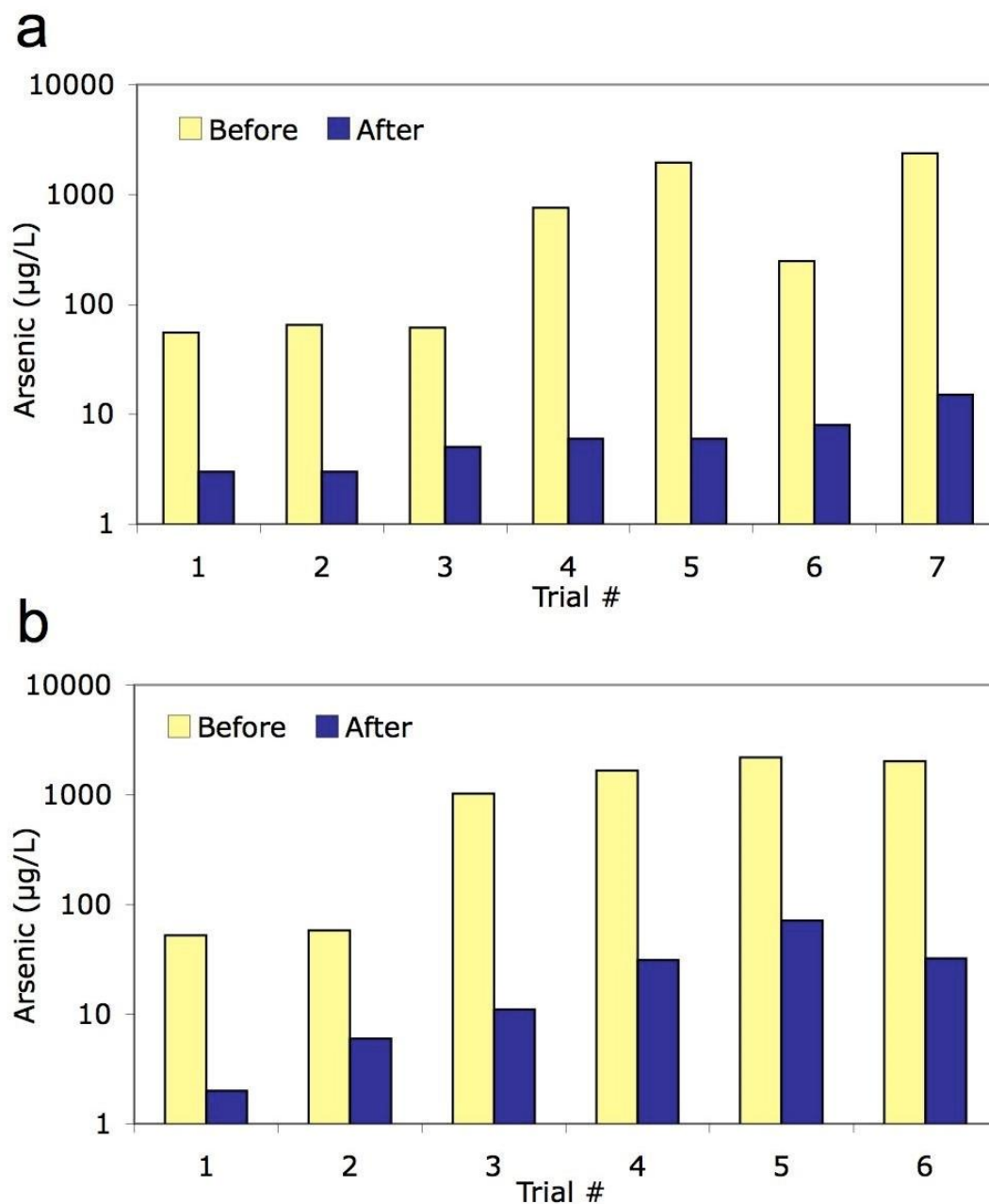


Figure 4. Arsenic concentrations in wastewater before and after it has been passed through the fine sand (a) and coarse sand (b) soakaway pits

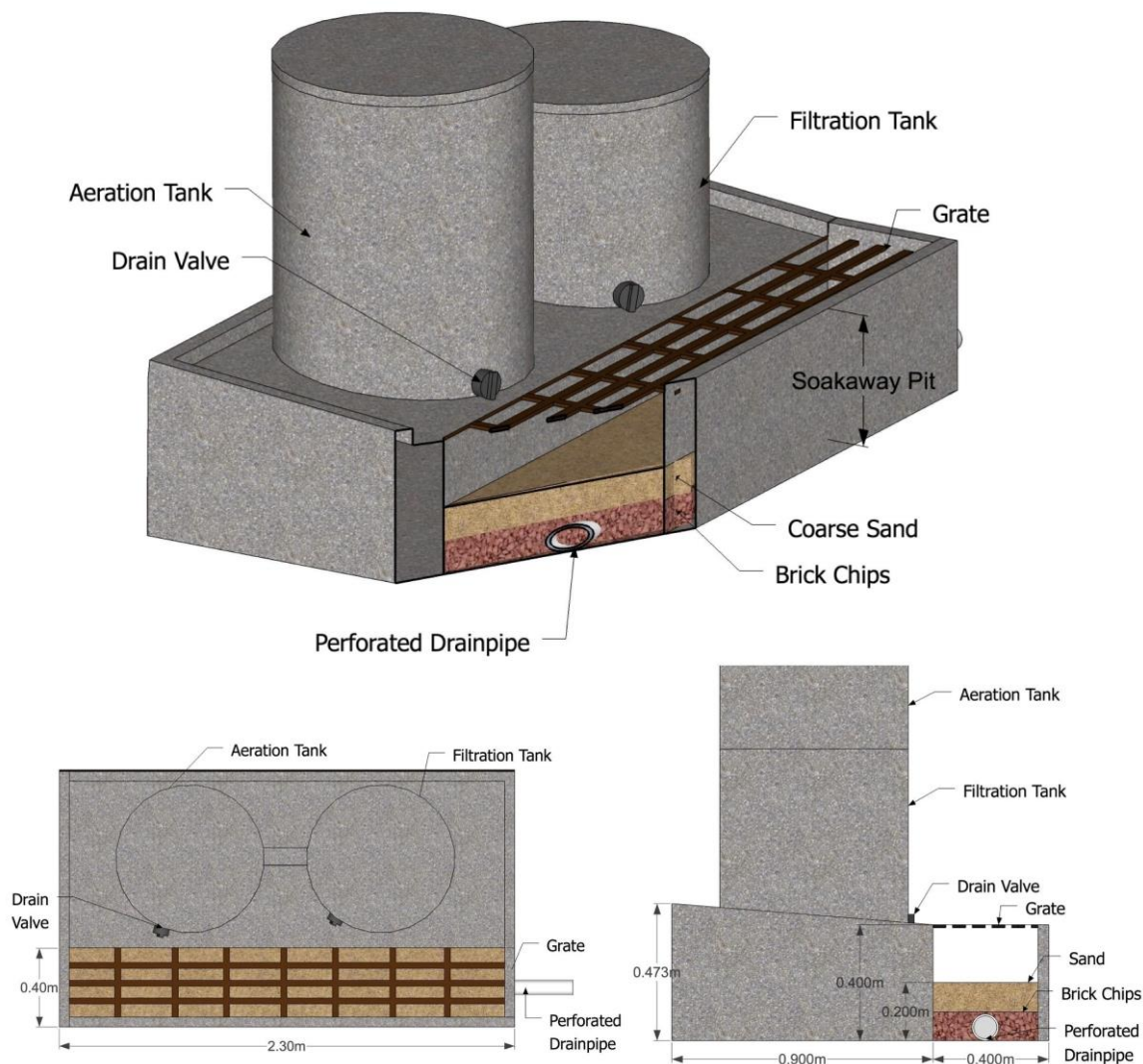


Figure 5. A possible design for a household soakaway pit, with a section plane cut at the left corner to show the layers of media. Below left and right is an aerial view and side profile.



Photo 1 Photo of AIRP from Manikganj