



## Review

# An overview of sustainable techniques used for arsenic removal from drinking water in rural areas of the Indo-Pak subcontinent

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## Abstract

Arsenic contamination in drinking water is a widespread issue in developing countries, especially in the Indo-Pak subcontinent. Modern arsenic removal technologies may not be feasible in this part of the world due to poor economic conditions and remoteness of many rural areas. Some cost effective techniques like oxidation and co-precipitation can be used as an alternative in these situations. Oxidation techniques widely used in sub-continent include passive sedimentation, solar oxidation and in-situ oxidation. Co-precipitation techniques include bucket treatment unit, three pitcher methods, BCSIR filter unit, fill and draw unit, arsenic removal unit attached to tube well and a five step purification system. The objective of the present synthesis is to compare these cost effective techniques for arsenic hit rural areas of Pakistan. Though modifications and improvements may be needed to get better results in our local conditions but it will open field for research in this particular area, which is being neglected so far and is necessary for sustainable development in drinking water sector in Pakistan. These methods can be highly effective as these are adjustable over a wide range of arsenic contamination. These techniques are best suited to Pakistan's rural conditions as these neither need electricity nor pressure. Moreover, the raw material for these techniques is cheaper and can be made easily available at the domestic level even in far flung areas of Pakistan. It is recommended that these sustainable techniques should be introduced in arsenic hit rural areas of Pakistan.

**Keywords:** Arsenic, removal, water, techniques, subcontinent

Arsenic (As) is an omnipresent hazardous metal mainly existing as an oxyanion compound in ground water (Smedley and Kinniburgh 2002) and ranks 20th in natural abundance, 14th in the seawater, 12th in the human body and consisting of 0.00005% of the earth crust (Mandal and Suzuki, 2002). Concerns over the presence of arsenic in ground water has increased a great deal in recent years as it is posing a serious threat to the millions of people across the globe encompassing from America to Asia in countries including Argentina, Bangladesh, Hungary, India, United States, China, Vietnam and Pakistan. Though this problem is also being faced by developed countries like America but it is even more worse in populous areas of Bangladesh and West Bengal (India), where millions of people are prone to the chronic effects of this carcinogenic element (Nickson *et al.*, 1998; Choudhary *et al.*, 2000). Currently the limit of As concentration in drinking water is 10  $\mu\text{g L}^{-1}$  according to WHO and USEPA standards, which was 50  $\mu\text{g L}^{-1}$  earlier (WHO 1993, USEPA 2000). But all the developing countries having this problem are still struggling to achieve the standards of 50  $\mu\text{g L}^{-1}$  as an interim target before

achieving the goal of 10  $\mu\text{g L}^{-1}$  ultimately in their drinking water. Chronic exposure to As in drinking water with greater concentration than 50  $\mu\text{g L}^{-1}$  can result in severe health troubles with the symptoms of skin, cardiovascular, renal, hematological and respiratory disorders (Smedley and Kinniburgh, 2002). More than 27% of the shallow ground water aquifers in Bangladesh are having the As concentration more than 50  $\mu\text{g L}^{-1}$  (Khan *et al.*, 2000) posing a serious threat to 90% of rural population consuming the water from 4-5 million tube wells anchored in these shallow water aquifers (Ahmed, 2001). This emerging global problem is becoming severe in India, Bangladesh and Nepal where millions of people are potentially at risk of arsenic contamination (Smith *et al.*, 2000).

The above-mentioned serious threats to the inhabitants in Bangladesh and India convinced Pakistan to recognize the need to assess drinking water quality for arsenic contamination in its ground waters. Since 1999, Pakistan has taken various initiatives in collaboration with UNICEF. As a result of these initiatives, the presence of arsenic

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contamination has been recognized in Pakistan, particularly in the central and southern parts of the country and consequently an arsenic mitigation programme, at national level was launched by the Government of Pakistan in assistance of UNICEF (Ahmed *et al.*, 2004; Nickson *et al.*, 2005; Sharma, 2006). Shocking levels of ground water arsenic concentration has been observed during the course of water quality surveys conducted by PCRWR during 2000, 2001, 2003 and 2004 (PCRWR, 2000, 2003a, 2003b, 2004). District Rahim Yar Khan has been declared as most worst arsenic contaminated area. Other contaminated areas include the city of Multan, Bahawalpur, Sheikhupura, Lahore, Kasur, and Gujranwala. Similar conditions are observed in Muzaffargarh (Nickson *et al.*, 2005). In Punjab, 20% population and in Sindh 36% of the population is exposed to the arsenic limits greater than WHO standards (Islam-ul-Haq *et al.*, 2007). Higher arsenic levels in ground water have also been observed in KPK by UNICEF.

### Oxidation and co-precipitation techniques

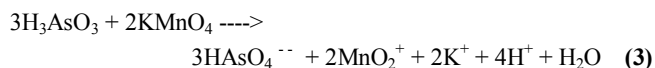
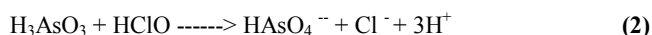
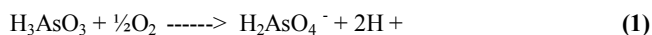
There are several methods available for removal of arsenic from water from small to large scale. Most arsenic removing technologies include oxidation, adsorption, co-precipitation, ion exchange and membrane filtration. Ion exchange and membrane filtration are not much suited for the rural and less developed areas of the subcontinent. Reason being that ion exchange is not as effective because several other cations present in ground water do compete with arsenic. So its ability to remove arsenic is limited. Membrane filtration is an effective technology for arsenic removal but high tech operation and high costs make it unsuitable for the masses in subcontinent. So oxidation and co-precipitation are most cost effective measures, frequently adopted in the world (Cheng *et al.*, 1994; Kartinen and Martin, 1995; Hering and Elimelech, 1996; Hering *et al.*, 1996; Joshi and Chaudhuri, 1996; Hering *et al.*, 1997; Hering and Chiu, 2000; Ravenscroft *et al.*, 2001).

During recent years, numerous small-scale methods for arsenic removal have been developed and many improvements have been made in already existing conventional methods keeping in view the needs and suitability to rural areas.

### Oxidation

In ground water, arsenic is present in the forms of As(III) and As(V) in varying degrees. As(III) is difficult to remove, so most of the methods involve the conversion of As(III) to As(V). So, this oxidation process of conversion from arsenite to arsenate is used as a pretreatment in most cases and is carried out by oxygen, ozone, free chlorine, birmessite, hypochlorite, hydrogen peroxide, permanganate, and Fulton's reagent. Though ozone has a very high

oxidation potential but it can react with organic matter and can cause side reactions (Wolfgang *et al.*, 1995). Using chlorine poses the risk of introducing harmful tri-chloromethane into the drinking water. Atmospheric oxygen, hypo-chloride and permanganate are usually used in developing countries (Ahmed 2001):



Atmospheric oxygen takes lot more time to oxidize certain amount of volume but oxidants like permanganate and hypo-chloride can convert arsenite to arsenate very quickly. Permanganate is a very effective and stable oxidizing agent, but gives water an unattractive color, which can be removed by an adsorbing media such as sand (Jalil and Ahmed, 2001).

### Passive sedimentation

If the water is stored for certain period of time before it is used, the atmospheric oxygen causes the passive sedimentation of the arsenic from zero to high reductions. As in Bangladesh rural community is used to use "basha pani" stored in pitchers, the passive sedimentation has taken attention in recent years. But this depends on the quality of the water. Passive sedimentation depends on the precipitating iron contents of the water. More than 50% decrease in As contents can be achieved by passive sedimentation if the ground water contains 380-480 mg L<sup>-1</sup> of alkalinity as CaCO<sub>3</sub> and 8-12 mg L<sup>-1</sup> of iron (Ahmed and Rehman, 2000). Other studies have shown 25% reduction of As contents using passive sedimentation. So, it is not a reliable method. In rapid assessment of technologies, passive sedimentation stood unsuccessful to decrease arsenic contents to the required level (BAMWSP-DFID-Water Aid 2001).

### Microbial oxidation

Two types of metal-microbiological interactions can be used for arsenic removal. i) Microbial oxidation of As (III) to As (V) and its subsequent precipitation. ii) Bio-accumulation of arsenic by microbial biomass. The oxidation method can be operated in an immobilized reactor reservoir. A cheap source of organic substrate like sugarcane juice can be added along with iron fillings (Parknikar, 1998). Iron fillings promote development of iron – oxidizing bacteria that oxidize iron at a rate 50-103 times faster than chemical oxidation of iron. Arsenic is then

adsorbed on the ferric iron. Treated overflow of water typically contain arsenic  $< 0.05 \text{ mg L}^{-1}$  for initial concentration up to  $4.0 \text{ mg L}^{-1}$  (Parknikar, 1998).

### Solar oxidation and removal of As (SORAS)

This easy method involves the reduction of As contents of water in transparent pet or other U-VA transparent bottles (Wegelin *et al.*, 2000). SORAS method is based on photochemical oxidation of As (III) to As (V) and then precipitation of As (V) absorbed on Fe(III) oxides. Ultraviolet radiation can catalyze the process of oxidation of arsenite in presence of other oxidants like oxygen (Young, 1996). As removal efficiency completely depends upon the intensity of UV-A and duration of irradiation. SORAS will be more efficient with increasing duration. Addition of citric acid also enhances the photochemical oxidation of As (III). Field tests in Bangladesh showed removal efficiency between 45-78% with an average of 67%. Concerning the Bangladesh guideline value of  $50 \mu\text{g L}^{-1}$ , SORAS can treat raw water having an arsenic concentration below  $100\text{-}150 \mu\text{g L}^{-1}$  (Weling *et al.*, 2007). Addition of potassium permanganate (if necessary together with aluminium sulphate), attains a higher arsenic removal efficiency, approx. 80% and 90%, respectively. Hence, SORAS would allow treatment of raw water containing a higher arsenic concentration. The procedure involves filling up of four fifth of a transparent plastic bottle with water, addition of few drops of lemon juice and keeping in the sunlight in horizontal position for whole day after vigorous shaking for thirty minutes. Then the bottle is turned in upright position for whole night and filtered by a piece of cloth.

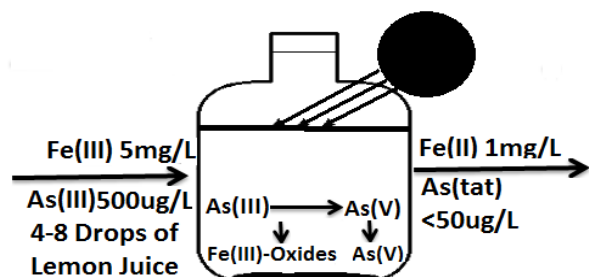
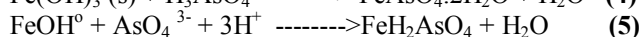
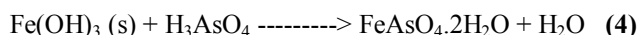


Figure 1: Solar Oxidation and Removal of As (SORAS)

### In-situ oxidation

Stored, aerated tube well water is released back into the ground water aquifer just below the head into the pipe of tube well to cause in situ oxidation of iron and As (Van 2008; Van *et al.*, 2008a, 2008b, 2008c). The intermittent injection of stored aerated water into the aquifer oxidizes adsorbed Fe(II) to Fe(III) and forms new adsorption areas for Fe(II) and Arsenic. The dissolved oxygen converts

ferrous to ferric and Arsenite to arsenate. The probable reactions of arsenate to ferric hydroxide are shown in Equations 4 to 5. Experiments show that this in situ precipitation and adsorption onto ferric iron reduces the As contents in ground water to about 50%. In-situ arsenic removal can be a trustworthy option, as arsenic is held in the aquifer and no waste flow is produced.



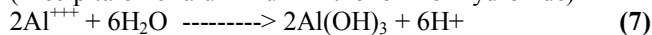
### Co-precipitation, coagulation and flocculation

This is widely used method of As removal in Bangladesh and India. In co-precipitation and subsequent coagulation process the chemicals (like Aluminium alum, ferric chloride, ferric sulphate) is added to the water in a container and forcefully stirred for few minutes. The micro-flocs (Aluminium hydroxide or ferric hydroxide) are formed immediately. Gentle stirring is continued for few more minutes to allow these micro-flocs to grow larger for efficient settling. During this flocculation process negatively charged As(V) and other anions are adsorbed onto the settling flocs electro statically. As(III) being neutral is hard to adsorb onto flocs, so pre-treatment of ground water converting As(III) to As(V) by using any suitable oxidant like ozone, bleaching powder or manganese dioxide is required for efficient removal of As. Bleaching powder is commonly used in Bangladesh and India for this purpose. Sedimentation and some times, filtration is required to completely remove the flocs (having As adsorbed onto them in the form of Al-As complex or Fe-As complex) from the water. Alum coagulation is best within the narrow pH range of 7.2-7.5 and iron coagulation does work at wide range of temperature from 6.0-8.5 (Ahmed and Rahman, 2000). Apart from pre-treatment, Fe/As ratio of the water does also play an important role in As removal efficiency by flocculation (Meng *et al.*, 2001).

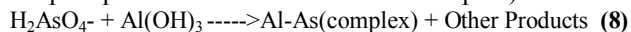
Chemical reactions involved in alum coagulation are as followed (Alum dissociation into aluminium):



(Precipitation of aluminium in the form of hydroxide)



Co-precipitation in the form of Al-As complex)



Similar sort of reactions do occur in the case of iron sulphate and iron chloride

### Bucket treatment Unit (BTU)

The BTU is composed of two buckets over one another (Figure 2). Upper bucket is of red colour while lower

bucket is green. Certain amount of suitable chemicals like Aluminium sulphate and permanganate are added to the water in red bucket and stirred strongly for 25 minutes with the help of a wooden spoon and then allowed to settle for 3 hours. After three hours the supernatant is allowed to flow through a pipe with valve from just above the bottom of the red bucket into the green bucket fitted with sand filter. Actually the process of arsenic removal takes place in red bucket in various steps like oxidation, flocculation, co-precipitation and sedimentation. Aluminium sulphate is used as flocculent or permanganate is used as an oxidant. This household process is developed by DPHE-Danida for domestic use in rural areas. Thousands of such units are distributed and installed in rural areas of Bangladesh. Efficiency of these units is variable from very low to high As removal. The reason being narrow pH required for the Aluminium coagulation. This problem can be removed by using iron salts instead of aluminium sulphate which works at wide pH range. These units work well in areas where Iron contents of water are high as it helps to improve the efficiency of co-precipitation. BTU is an excellent As removing domestic unit (Kohnhorst and Paul, 2000) to the desired levels in the areas with As contents less than 500 ppb (Tahura *et al.*, 2000). The use of this unit should not be encouraged in areas having As contents higher than 500 ppb as it may not remove As upto desired levels. This device can be made easily adopted among masses in rural areas of Bangladesh and India by keeping the chemicals involved and raw material for buckets locally available to keep its cost low enough for the poor.



Figure 2: Two bucket unit (Ali *et al.*, 2001)

Rapid assessment tests for BTU has failed in rural conditions of many areas of Bangladesh due to poor mixing and pH variations at different localities (BAMWSP, DFID, WaterAid, 2001) and could not remove As to  $0.05 \text{ mg L}^{-1}$ . Bangladesh. University of Engineering and Technology (BUET) has made little modifications to the BTU and got better results by using  $100 \text{ mg L}^{-1}$  of ferric chloride and  $1.4 \text{ mg L}^{-1}$  of potassium permanganate in modified BTU units. In their results, the feed tube well water was having As up

to 640 ppb but the treated water mostly showed less As concentration ( $< 20 \text{ ppb}$ ).

### Stevens institute technology

This technology also uses pair of buckets (Figure 3). One bucket is used for mixing flocculent and oxidant, which are iron sulphate and calcium hypo-chloride in this case and the other bucket is used for sedimentation and filtration. The second bucket has a sand filter bed at the bottom. Above the sand filter bed another bucket is placed inside the second one. The inner buckets have slits on the bottom to allow the water percolation, sedimentation and keeping the sand bed intact. Just above the bottom a plastic pipe out let is placed with cloth filter at the origin. Maintenance involves only the replacement of the sand bed filter after 3-4 days. Rapid Assessment test has passed this technology as it has shown to remove As from 80-95% samples to reduce the contents less than  $0.05 \text{ mg L}^{-1}$  (BAMWSP, DFID and Water Aid, 2001).

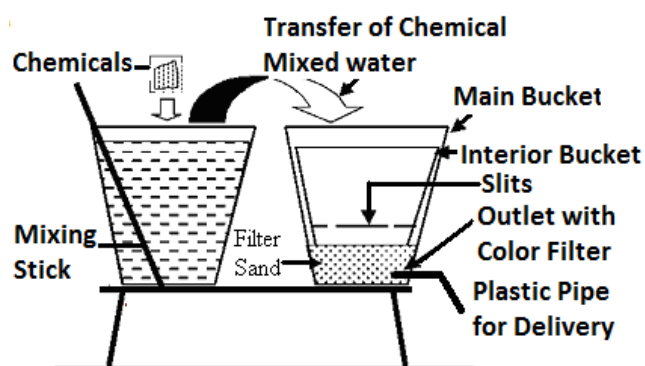


Figure 3: Stevens Institute Technology (Ahmed, 2003)

### Three pitcher method (Sono-3 Kolshi filter)

Three pitcher method is commonly used in Bangladesh for arsenic removal. System consists of three earthen pitchers over one another supported with the help of a steel or bamboo rack or stand (Figure 4). More than eighty percent of Bangladesh rural population use earthen pitchers for water storage and local people call it as kolshi, so method called as 3-kolshi filtration system. Top most and middle pitchers have small holes at the bottom covered with polyester cloth. Above the cloth, a bed of brickettes is placed in both upper and middle kolshis. Then in upper most pitcher above the brickettes a bed of coarse sand and then bed of cast iron turnings is placed. In the middle one above the brickettes a bed of fine sand and then a charcoal bed is placed. The ground water is poured into the top most kolshi and arsenic free water is collected in the lowest one. As all the raw material is locally provided this is a cost effective system to be used at domestic level in rural areas

of India and Bangladesh. Experiments have shown that up to 96% of As removal can be achieved using this method (Hussain *et al.*, 2006). The cost of this filter is 5 US \$ (Munir *et al.*, 2001).

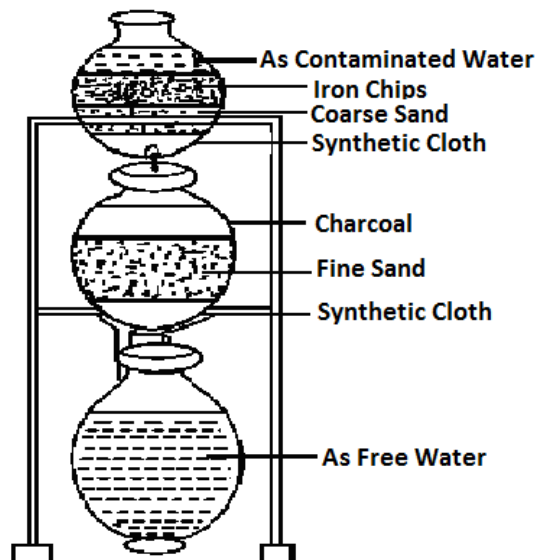


Figure 4: Three pitcher method (Khan *et al.*, 2000)

#### BCSIR filter unit

Bangladesh Council of Scientific and Industrial Research (BCSIR) have developed an arsenic removal system, which uses the process of coagulation/coprecipitation with an iron based chemical followed by sand filtration. BCSIR has developed a low cost arsenic filter. The technology consists of adding a floc forming composition to the arsenic contaminated water followed by stirring and settling. The chemicals are composed of iron oxide, alum, activated charcoal and calcium carbonate, which are to be mixed in definite proportions, homogenized and micronized. After settling, the water is passed through a filter bed composed of sand and some iron bearing minerals of definite particle size range, which are to be activated by suitable chemical and heat treatment. The dose of the floc forming composition depends on the extent of arsenic contamination. Water containing up to 2.7 ppm arsenic could be purified below safe limit set by WHO (BCSIR 1999).

#### Fill and draw units

Educational institutions and bunches of families in Bangladesh use this relatively large plant for arsenic removal installed under DPHE-Danida Arsenic Mitigation Pilot Project (DPHE-Danida 2000). It consists of large tank fitted with the manually operating mixer as shown in the Figure 5. The tank is tapering at the bottom to allow the

flocs to gather in the depression. The outlet tap is fitted just above the tapering edge, which passes the water into an up flow sand bed filtration unit, from where the As free water is collected near the top. From the center of the tapered bottom, a sludge withdrawal pipe emerges. The tank is filled and chemicals (Coagulant and oxidant) are added and then are mixed with the help of the manual mixer fitted with gear and repeller in order to make the mixing forcible for 30 seconds with 60 rpm. Then the sedimentation is allowed to occur for whole night. The treated water is drawn next day from the filtration unit as shown in the Figure 5. High As removal efficiencies are observed (Ahmed, 2001).

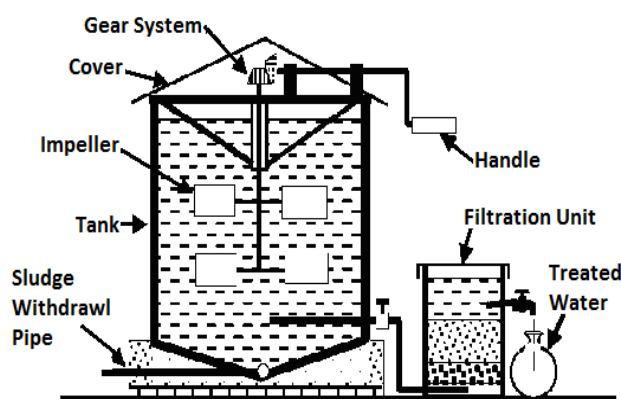


Figure 5: DPHE-danida fill and draw As removal unit (Ahmed, 2003)

#### Arsenic removal unit attached to tubewell

This compact arsenic removal plant is used in west Bangal-India fitted with hand pumps. Basic principle of this plant is similar to the above-mentioned "Fill and Draw Unit" except that mixing and flocculation steps are separated. The construction of the sedimentation tank is similar to the Fill and Draw Unit with tapering bottom and sludge withdrawal pipe. The filtration system is also similar to that of Fill and Draw Unit with up flow filtration. The coagulant used in this method is Aluminium alum and oxidant is hypo-chloride. Both chemicals are used in diluted form at step-A as shown in figure 6. It can remove up to 90% of As from the ground water containing  $300 \text{ mg L}^{-1}$  As contents.

#### Use of naturally occurring iron

It is well known fact that As is adsorbed electrostatically onto the flocs of  $\text{FeOH}_3$  and can be removed by sedimentation and filtration afterwards. Iron is present in the form of  $\text{FeOH}_2$  in ground water. Its oxidation leads to  $\text{FeOH}_3$  forming flocs and attracting As(V). In



Bangladesh iron and arsenic co-exist in most parts of the country. Three fourth samples having Fe contents more than 5 mg/L have shown that they are also unfit due to co-existence of iron. So in Bangladesh iron removing plant are potentially used for the As removal as well. Only the effective aeration of iron-contaminated water can remove As from ground waters. Most of the iron removing plants based on the principle of aeration, flocculation, sedimentation and filtration are removing the As to desired levels. In order to increase the efficiency of the As removal in Iron removal plants (IRPs) some oxidants are used to convert As (III) to As (V) to easily co-precipitate with  $\text{FeOH}_3$  flocs. So IRPs are used as ARPs as well in many iron contaminated areas of Bangladesh. On this principle large IRPs are constructed in towns of Bangladesh. Evaluation of many existing community IRPs have shown that they can remove As from 50 to 80% Greater is the As/Fe ratio of the water; greater will be efficiency of the plant (Mamtaz and Bache, 2001; Mamtaz 2008).

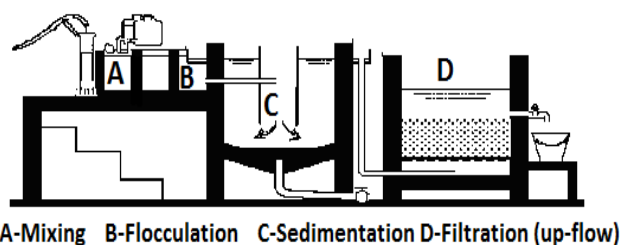


Figure 6: Arsenic removal plants attached to tube well (Ahmad and Rehman, 2000)

#### A five-step low-cost purification system

This cost effective system involves the principle of air/light oxidation, precipitation and adsorption. It consists of inlet tank, precipitation tank, adsorption tank, water lock and reservoir (Figure 7). Inlet tank is a brick made tank just adjacent to the tube well for the contaminated water. From inlet tank via a tap water comes in precipitation tank in spreaded form on the troughs to increase the surface area exposed to light and air. A series of baffles in the precipitation tank push the water to the surface repeatedly exposing it to light and air. Here the co-precipitation of arsenic occurs and the flocs are trapped by sediment traps. Then the remaining arsenic is adsorbed in in the adsorption tank having mixture of charcoal and ash added with spent brine and the fine rust. Then arsenic free water comes in the water lock tank whose primary function is to check the system from being dried out. Then the water comes in reservoir tank from where it is consumed via a tap. Different chemicals were used to achieve the optimal results and ultimately 200 ppb of As concentration in feed water was reduced to bellow detection limits i.e. < 5 ppb, so

As reving efficiency remained greater than 97.5% (Philip and Chowdhury, 2004).

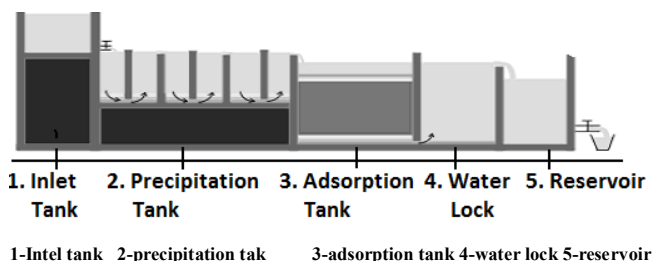


Figure 7: Five step filtration system (Philip and Chowdhury, 2004)

#### Summary of the techniques and their relative efficiency

Technology	Efficiency	Reference
<b>Oxidation</b>		
Passive	More than	BAMWSP, DFID, Water Aid
Sedimentation	50%	2001; Ahmad <i>et al.</i> , 2000
Microbial Oxidation	More than	Parknikar 1998
	98.75%	
SORAS	45-78%;	Wegelin <i>et al.</i> , 2000
	80-90%	
In-situ Oxidation	50%	Ahmed and Raham, 2000
<b>Co-precipitation, coagulation and flocculation</b>		
Bucket Treatment Unit	More than	Tahura <i>et al.</i> , 2000;
	96.9%	BAMWSP, DFID,
		WaterAid 2001; BUET
Stevens Institute Technology		BAMWSP, DFID,
Three Pitcher Method (Sono-3 Kolshi filter)	upto 96%	WaterAid, 2001
BCSIR Filter Unit	99.5	Hussain <i>et al.</i> , 2006
Fill and Draw Units	-	
		BCSIR 1999
Arsenic Removal Unit Attached to Tubewell	90%	DPHE-Danida, 2000;
Use of Naturally Occurring Iron	50-80%	Ahmed, 2001; Ahmed, 2003
A Five-Step Low-Cost Purification system	More than	Ahmed, 2001; Ahmed and Rehman, 2000
	97.5%	
		Mumtaz, 2008
		Philip and Chowdhury, 2004

#### Conclusion

The above mentioned techniques are successful in India and Bangladesh to varying extent depending upon the water quality, local conditions, operational issues and inadequacy of proper training among the rural populations. These techniques can be successfully applied to the As hit rural areas of Pakistan with necessary modifications according to our local conditions and As contents in drinking water. The cost of the As removal techniques

would play an important role in their adoption and sustainable use in rural areas of Pakistan. The discussed techniques can be fruitfully kept at affordable cost to our poors by fabricating with local materials and usage of common and cheaper chemicals where necessary. Though all of these methods have weaknesses and strengths but can be polished to local conditions of Pakistan with initiated research in this particular sector after being introduced in pilot phase.

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