

# Arsenite Removal from Drinking Water using Naturally available Laterite in Sri Lanka

R.M.S.C.Ranasinghe, D.R.I.B.Werellagama and R. Weerasooriya

**Abstract:** Arsenite, As(III) is the most soluble form of arsenic species. Arsenic removal efficiency by laterite (commonly found in Sri Lanka) was examined as a function of pH, initial arsenite concentration, laterite dosage, contact time and mixing rate. More than 90% arsenite removal could be achieved within 5 minutes when pH is around 10. By treating the water at this pH range, the current USEPA standard for arsenic in drinking water (10 ppb) can be maintained when the arsenite / laterite ratio is less than 10( $\mu\text{g/g}$ ). Results of the study showed that naturally available laterite in Sri Lanka can be used as an effective adsorbent to treat arsenic contaminated water.

**Keywords:** Arsenite removal, laterite, arsenic in Sri Lanka, drinking water treatment, well water

## 1. Introduction

Arsenic in natural water originates from natural and anthropogenic sources. Naturally it is released into groundwater from naturally occurring minerals [1]. It has been reported that arsenic occurs naturally in about 245 minerals, which when subjected to weathering will release soluble arsenic into natural waters [2]. It is commonly found in rocks and soils with high sulfur content. For example Arsenopyrite ( $\text{FeAsS}$ ), pyrite ( $\text{FeS}_2$ ), orpiment ( $\text{As}_2\text{S}_3$ ), realger ( $\text{AsS}$ ), and chalcopyrite are some of the sulfide minerals, which contain high levels of arsenic [3]. Among them arsenopyrite is the commonly available mineral.  $\text{As}_2\text{O}_5$ , scorodite ( $\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$ ), gypsum, Fe-smectite, claudetite ( $\text{As}_2\text{O}_3$ ) are some other kinds of minerals containing arsenic. Therefore from the dissolution of these arsenic-bearing minerals arsenic can be released to groundwater systems. In the surface water, arsenic can be derived from weathering of geological materials, through mixing with high arsenic geothermal waters and by mixing of waste stream from a variety of industrial processes. Petroleum refining, glass melting, paper production, cement manufacturing, paint manufacturing, production of semiconductors and smelting of ores are some of the examples for those industrial processes. Also this is released into the environment by the dispersion of arsenic containing fertilizers, pesticides, and herbicides [2, 4-6].

The aqueous chemistry of arsenic is complicated. Speciation and solubility of arsenic is influenced both by redox status and

pH. Arsenic occurs in the environment mainly as the inorganic arsenic oxides of arsenite(III) and arsenate(V). Arsenates predominate in well oxidized waters while arsenites occur in reduced environments [7-9]. Depending on the pH, in aqueous solutions As(III) occurs in different forms such as  $\text{H}_3\text{AsO}_3$ ,  $\text{H}_2\text{AsO}_3^-$ ,  $\text{HAsO}_3^{2-}$  and  $\text{AsO}_3^{3-}$  [10]. Similarly mole fraction distribution of arsenate species are  $\text{H}_3\text{AsO}_4$ ,  $\text{H}_2\text{AsO}_4^-$ ,  $\text{HASO}_4^{2-}$ , and  $\text{AsO}_4^{3-}$  [11].

Arsenite, As(III) is much more toxic, soluble and mobile than As(V) [8,12]. It has been reported that arsenite is 25-60 times more toxic than arsenate and inorganic arsenic compounds are more toxic than organic [13]. The acute and chronic toxicity of arsenic in humans has been documented especially in countries like Argentina, Bangladesh, China, Chile, Ghana, Hungary, West Bengal (India), Mexico, Thailand, Taiwan Vietnam and USA [1,14]. In Sri Lanka also; questions were raised recently whether arsenic is one of the causative agents of chronic kidney diseases [4-6]. Arsenic causes cancers and tumors in the skin, bladder, genital organs and eyes [1]. Conjunctivitis, melanosis, hyperpigmentation, hyperkeratosis and peripheral vascular disorders are the most

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commonly reported symptoms of chronic arsenic exposure. In severe cases gangrene in the limbs and malignant neoplasm have also been reported. Acute short-term exposure to high doses of arsenic also can cause adverse health effects. In addition to these, arsenic in drinking water can cause diabetes, anaemia as well as reproductive & developmental, immunological, and neurological effects [14].

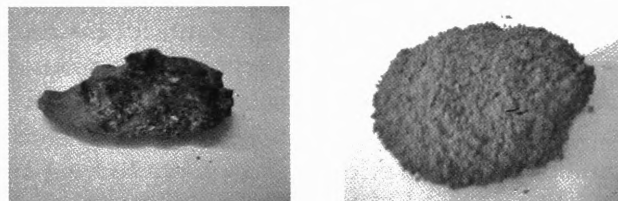
The maximum permissible limit of arsenic in drinking water in Sri Lanka is 50 ppb (SLS 614, 1983), while USEPA and WHO (1993) guideline is 10 ppb. New Zealand drinking water guideline (2008) is also 10 ppb. According to the USEPA, the maximum contaminant level goal (MCLG) of arsenic in drinking water is zero. MCLG is the level below which there is no known or expected risk to health. Even for Sri Lanka a lower guideline which is reasonably achievable considering the treatment performance and rapidly improving analytical capability is needed to achieve public health protection goals.

Several studies have been carried out on arsenic removal and those technologies generally fall into three major classes. They are chemical precipitation, adsorption and membrane separation. Each technology has its own merits and demerits [15]. Most of the removal methodologies were based on adsorption where the removal efficiency and the cost effectiveness depend on the type of adsorbent used. Many natural & artificial adsorbents are used and it is understood that materials containing Fe, Al and SiO<sub>2</sub> can remove arsenic from drinking water efficiently.

Laterite locally known as “cabook” in Sri Lanka is a reddish colour highly weathered clayey rock material which is rich in iron or aluminum oxides. The constituents of laterite are minerals such as goethite, gibbsite, hematite, kaolinite etc. Due to the cellular or vesicular nature of laterite it has a high specific surface area, porosity and permeability [16,17].

Results of a past study on Sri Lankan laterite [16] indicate a dominant presence of the three oxides Fe<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub>. According to those findings, the Fe-rich layer is well developed in the laterite from lowlands; where the laterization process is believed to be active. Both hydrous iron and aluminum oxide components in laterite have a pH of the point zero charge (commonly called as pH<sub>zpc</sub>) of 8.5-8.6 [18,19]. This is the pH where the net surface

charge is zero. Under natural conditions (typical pH value of naturally available laterite varies between 4-7) they are characterized by net positive surface charge and hence have the capacity to absorb anionic contaminants. Presence of Fe and Al, high porosity and the availability of anion exchange sites are advantageous for using laterite as an adsorbent for removal of arsenic.



**Fig.1- Laterite rock and lateritic soil**

Therefore in this research arsenite, As(III) removal efficiency using laterite, a naturally available material in Sri Lanka was studied. As(III) was selected primarily due to its high mobility, solubility, and toxicity. This paper presents the findings of this study and the results of a survey on arsenic in Sri lankan ground water as of 2001/2002 [15]. More recent data (2012) are available in reference [20, 21].

## 2. Materials and Methods

### 2.1 Arsenic Analysis

After reviewing available analytical methodologies carefully, a method based on the Hydride Generation-Atomic Absorption Spectrometry (HG-AAS), was used for arsenic analysis due to its high sensitivity. Analysis of arsenic in the experiments was carried out using this method at the Department of Chemical Engineering, Faculty of Engineering, University of Peradeniya, Sri Lanka. A Hydride Generation Atomic Absorption Spectrophotometer with a hollow cathode lamp as the radiation source (AAnalyst 300, Perkin Elmer) was used for this purpose [22]. The detection limit of the instrument was 1 ppb. Flow Injection Analysis System (FIAS 100) was used for continuous hydride (AsH<sub>3</sub>) generation. The process involves the reaction of the acidified solution (sample mixed with hydrochloric acid) with sodium borohydride as the reducing agent. Readings were verified by spiking the standard solutions (by checking the results of a standard arsenic solution, by spiking with different concentrations of arsenic. An alternative approach is to analyse a proportion of samples in duplicate).

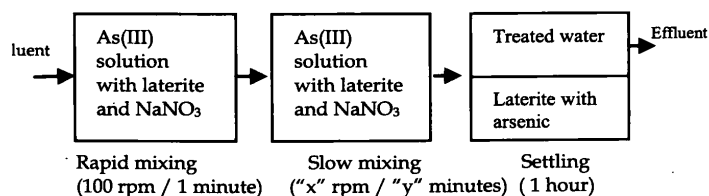
## 2.2 Preparation of adsorbent

Laterite used for this study was obtained from Pasyala, a town in South-Western in Sri Lanka. Initial pH of the 2g/L laterite - water suspension was 6.37. According to the literature both hydrous iron and aluminum oxide components in laterite have a  $pH_{zpc}$  of 8.5-8.6 [18,19]. Laterite sample was crushed, powdered (ground with a mortar and pestle) and sieved to separate the powder passing through 0.075 mm sieve. Smaller particle size was selected due to its high specific surface area, which is an advantage in the adsorption process.

## 2.3 Procedure

The experiments were conducted as batch studies at room temperature. 1000 ppm standard arsenite solution was used as the stock arsenite solution and subsequent dilutions were carried out using distilled water, when necessary. Acid washed glass beakers were used for the experiments. NaOH and HNO<sub>3</sub> were used to adjust the pH value while NaNO<sub>3</sub> was used to maintain the ionic strength.

Jar test apparatus was used and all the experiments were carried out in duplicate. The procedure used was as follows. Initial arsenite solutions were prepared diluting the stock solution and adding NaNO<sub>3</sub> to maintain the ionic strength. Then laterite (prepared as described above) was added to the sample followed by rapid mixing to ensure complete mix in the solution. A variable speed electrical stirrer, inserted into the solution carried out this mixing. Then reducing the speed, slow mixing was performed, allowing reactions to take place and sorption to occur. Then the sample was allowed to settle about 1 hour. After settling the supernatant was taken to another bottle and preserved using concentrated HCl until analysis. Every sample was analyzed on the following day.



**Fig.2 - Treatment train employed**

### 2.3.1 Effect of pH and Initial arsenite concentration

The effect of pH on the removal of arsenite was studied by equilibrating the reaction mixture of 200 ppb arsenite solution in 0.1M ionic strength and 2 g/L laterite; at different initial pH values. 100 rpm rapid mixing was performed for 1 minute which is followed by 30 rpm slow mixing rate for 15 minutes. Next the samples were allowed to settle about 1 hour and then the supernatant was preserved until analysis. Similar experiments were performed using 1200 ppb initial arsenite concentration (with 30g/L and 2 g/L laterite concentrations), and the effect of pH variation was studied.

### 2.3.2 Effect of laterite dosage

For finding out the optimum laterite dosage, different laterite concentrations such as 2, 5, 10, 15, 20, 30, 50 g/L were used. The experiment was performed similar to the procedure given in section 2.3.1 with an initial arsenite concentration of 200 ppb. The pH of the samples were maintained around 10.

### 2.3.3 Effect of contact time and mixing rate

To study the effect of contact time, a similar experiment was performed with initial arsenite concentration of 200 ppb and laterite concentration of 15 g/L at pH 10. The slow mixing time was varied as 5,10,15,30, 45 and 60 minutes and the slow mixing rate was maintained at 30 rpm. Rapid mixing was maintained at 100 rpm for 1 minute.

The effect of slow mixing rate on arsenite removal was observed by performing a similar experiment but varying the slow mixing rate from 10 to 50 rpm. Here initial arsenite concentration was 200 ppb while initial laterite concentration was 20 g/L. The rapid mixing rate was 100 rpm within 1 minute and slow mixing rate was maintained about 10 minutes. pH was maintained around 10.

### 2.3.4 Iron concentration of treated water

Since laterite consists of Fe<sub>2</sub>O<sub>3</sub>, whether it increases the iron concentration in the treated effluent was verified. The total iron concentration of treated water was also tested using a HACH DR/2010 Spectrophotometer. Silver diethyl dithiocarbamate method was used in this analysis and the range of detection is 0 - 0.200 ppm. Water with 200 ppb initial arsenite concentration at 0.1M ionic strength was treated using 20 g/L laterite at pH 10 and the final total iron concentration in the treated water was analysed.



## 2.4 Arsenic in Sri Lankan aquifers

In the survey carried out in 2001/2002 [15] most of the samples were collected from the available dug wells and tube wells, which were used for drinking purposes (sampling locations are given in Fig.3). Some of the samples (15% of the

total samples) were collected from the areas where arsenic-bearing minerals are found (but only as minor accessory minerals) [17]. About 110 samples were analysed.

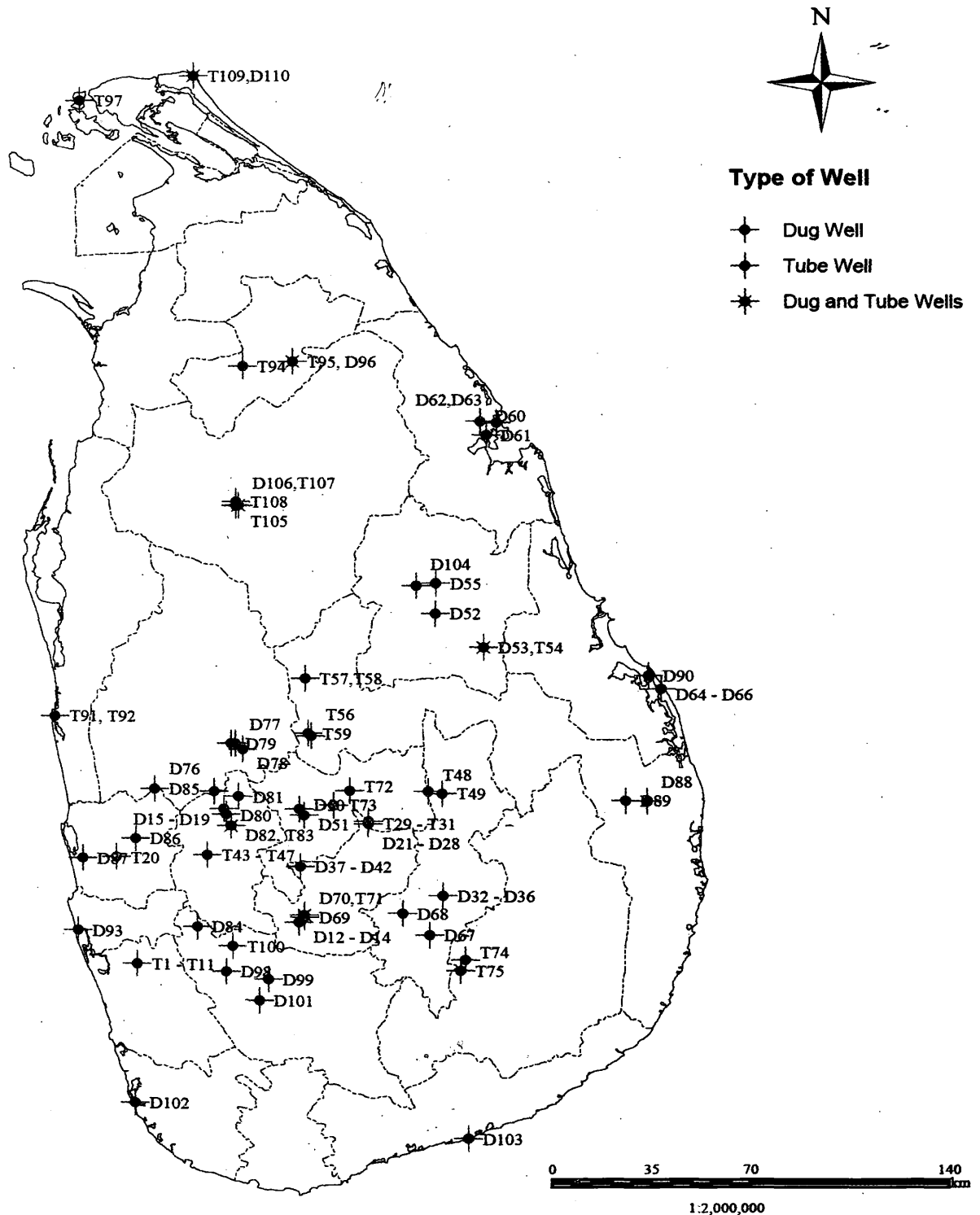


Fig.3 - Sampling locations for arsenic level determination in Sri Lankan ground water (Survey carried out in 2001/2002 [15])

### 3. Results and Discussion

#### 3.1 Effect of pH and initial arsenite concentration.

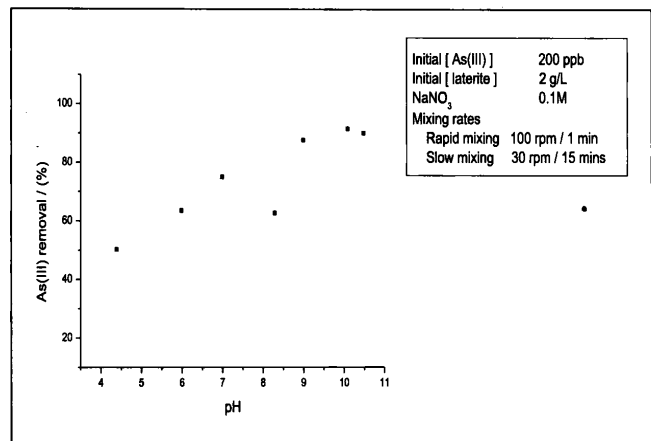
Figures 4, 5, 6 and 7 represent the removal of arsenite anions by laterite at different pH values with 200 ppb and 1200 ppb initial arsenic concentrations. In these experiments 0.1M ionic strength was maintained. According to the results more than 50% arsenite removal could be obtained throughout the whole pH range. Removed amount depends on the used laterite concentration. In the acidic and neutral pH ranges the removal of arsenite by laterite is less than that in the basic pH range. According to the results, arsenic concentration of final effluent in the acidic pH range is 3-5 times higher than that in the basic pH range. Highest removal percentage was obtained within basic pH range mainly when pH is around 10 (more than 90% removal of arsenite). This may be explained by the different species of As(III) oxy-anions present in different pH ranges. According to literature [10,23],  $\text{H}_3\text{AsO}_3$ , which is the neutral form of arsenite, is the predominant species in the acidic and neutral pH ranges. In basic pH ranges when  $\text{pH} > 8$   $\text{H}_2\text{AsO}_3^-$ ,  $\text{HAsO}_3^{2-}$ , and  $\text{AsO}_3^{3-}$  are dominant and therefore As(III) is non reactive below this pH. Therefore high arsenite removal efficiency can be obtained in the basic pH range.

As described above, the hydrous oxides of iron and aluminum in laterite can have anion exchange sites, which is important for arsenite removal. Also when stirring the sample, due to the aeration, dissolved iron (Fe(II)) will be oxidized to Fe(III) and precipitated as iron (Fe(III)) oxyhydroxide. At high pH levels formation of smaller Fe(III) hydroxide precipitates get increased. These smaller precipitates provide a higher effective surface area for arsenite adsorption. This will enhance the process of flocculation at the slow mixing stage [24]. Also due to the formation of aluminum hydroxide the flocculation process may be increased.

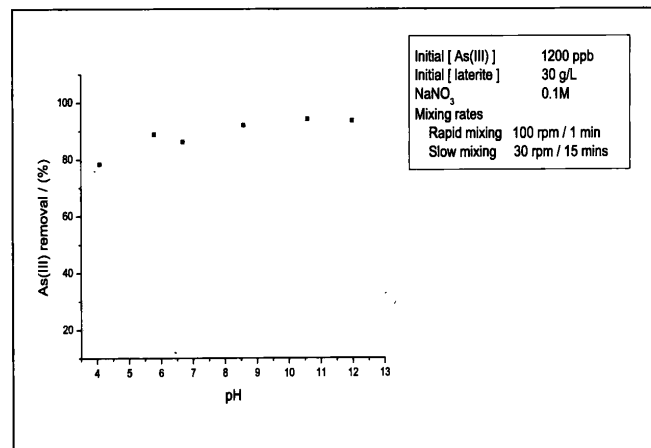
If the removal of arsenite from water by laterite is only by adsorption, removal efficiency should be high when  $\text{pH} < \text{pH}_{\text{zpc}}$  (pH of the point zero charge). But according to the findings of the current research, removal efficiency was high when  $\text{pH} > \text{pH}_{\text{zpc}}$ . Therefore removal of arsenite is not only by adsorption, but may be due to both sorption mechanisms. (May be by absorption to laterite particles and

by adsorption as described above). The actual removal process may be characterized by analysing the arsenic-laterite sludge. If an XRD analysis is carried out for both raw and used (for water treatment) laterite; it will give more insights in to the removal process.

As represented in Fig.6 and Fig.7, arsenite removal by laterite get decreased with the increasing initial arsenite concentration and with the decreasing laterite concentration. This is due to the limited amount of sorption sites in a particular laterite dosage. Also iron oxyhydroxide formation gets decreased with decreasing laterite concentration.

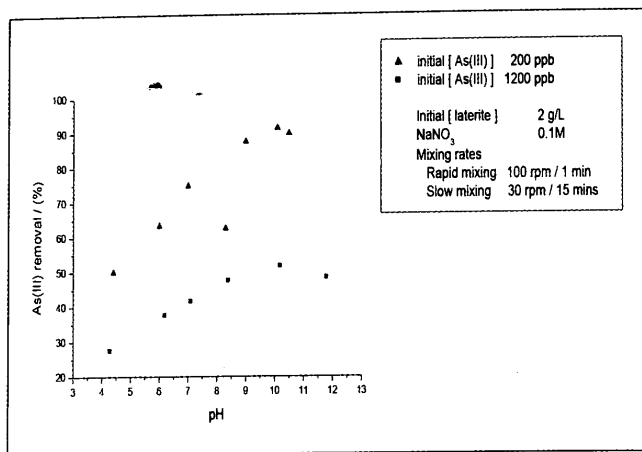


**Fig.4- Arsenite removal percentage using laterite, as a function of pH with the initial arsenite concentration of 200 ppb.**

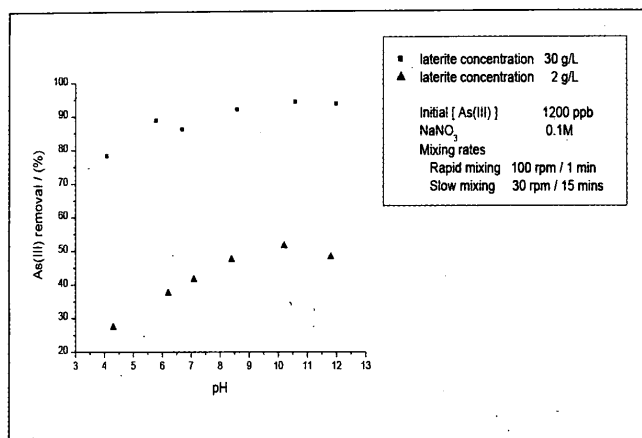


**Fig.5- Arsenite removal percentage using laterite, as a function of pH with the initial arsenite concentration of 1200 ppb.**





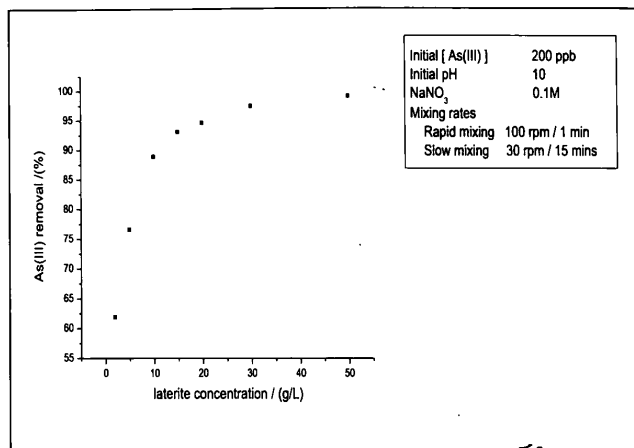
**Fig.6- Arsenite removal percentage using laterite, as a function of pH and the initial arsenite concentration**



**Fig.7- Arsenite removal percentage using laterite, as a function of pH and the laterite dosage.**

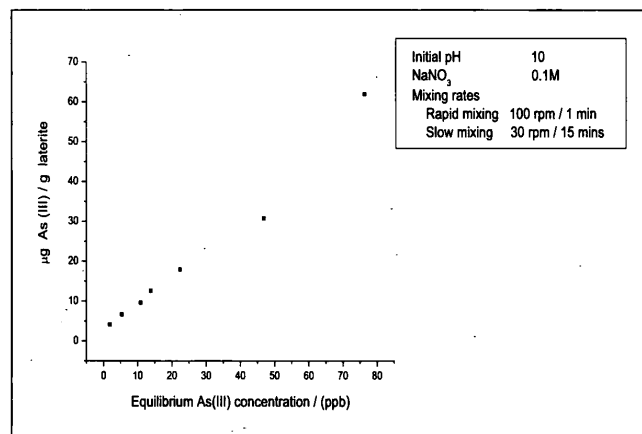
### 3.2 Effect of laterite dosage

The study of removal percentage as a function of sorbent dosage is important in establishing the optimum use of sorbent for any sorption process. Fig 8 shows the effect of laterite dosage on the removal percentage. According to these results, by increasing the sorbent dosage from 2 to 50 g/L (i.e. increasing 25 times) the removal percentage increased from 62% to 99% (i.e. increased by 37%). The increase in removal percentage can be explained due to the greater number of sites available to arsenite with increasing laterite dosage. Therefore when the initial arsenite concentration is 200 ppb, to obtain the 10 ppb standard in final effluent, laterite dosage of 20 g/L or higher has to be used. This implies that arsenite / laterite ratio needs to be less than 10 ( $\mu\text{g/g}$ ) to reach the drinking water standard of 10 ppb. Laterite dosage should be further increased to treat water within neutral pH levels since the arsenite removal efficiency is less within this range (refer Fig.4 - Fig.7).



**Fig.8- Arsenite removal percentage using laterite, as a function of the laterite dosage.**

Fig.9 shows the adsorption isotherm for arsenite removal by laterite. This can be used to calculate the required laterite dosage to obtain a particular equilibrium arsenite concentration. Furthermore the results obtained are well fitted in the linear form of Freundlich isotherm.

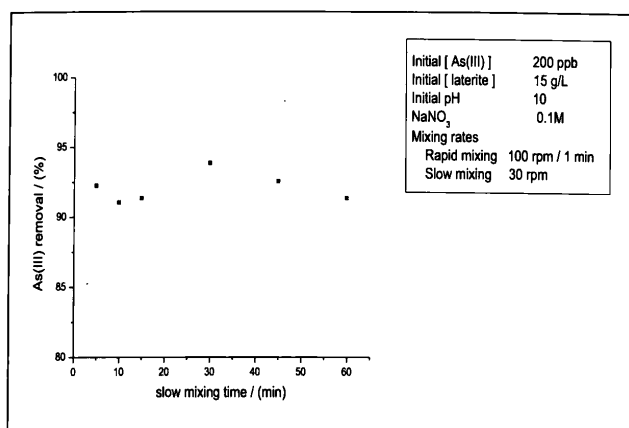


**Fig.9- Adsorption isotherm for the removal of arsenite by laterite.**

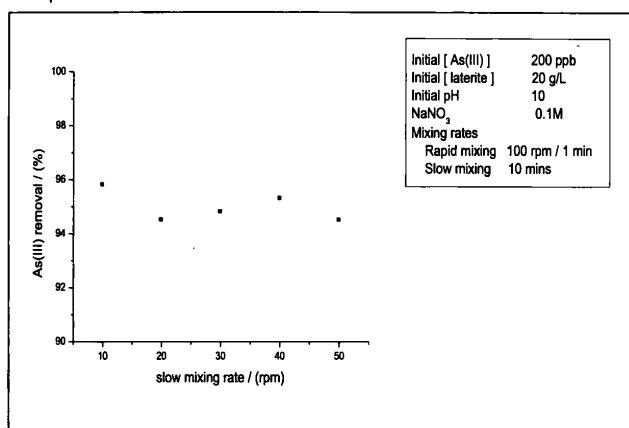
### 3.3 Effect of contact time and mixing rate

Fig.10 shows the effect of contact time with laterite for the removal efficiency; using a solution of initial arsenite concentration of 200 ppb. Sorption was very rapid reaching the equilibrium state within 5 minutes. This rapid removal is a great advantage for using this as an efficient arsenic removal method in drinking water treatment.

Fig.11 shows the arsenite removal percentage using laterite as a function of slow mixing rate (agitation rate). Varying the agitation rate from 10 rpm to 50 rpm does not have any significant effect on the removal process. This supports the earlier observation that the removal of arsenite by laterite is a rapid process.



**Fig.10- Arsenite removal percentage using laterite, as a function of slow mixing time.**



**Fig.11- Arsenite removal percentage using laterite, as a function of slow mixing rate.**

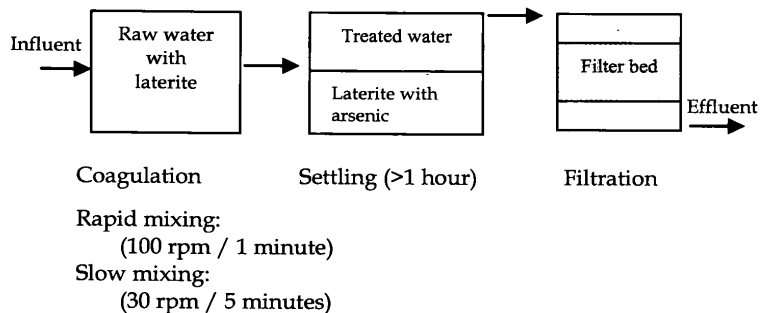
### 3.4 Iron concentration in the water treated using laterite

No iron was detected in the treated water. The iron concentration in the same sample without adding arsenic was found to be 0.22 mg/L. This supports the earlier observations (discussed in section 3.1), that iron oxyhydroxides will form, adsorb arsenic and precipitate.

Therefore considering these results, the arsenic removal method studied in the current research, by using naturally available laterite, can be recommended as a low-cost method for water treatment applications.

The efficiency of this method may be increased by converting arsenite to arsenate or by using laterite with higher iron concentration. Then the optimum laterite dosage can be reduced than the currently obtained value. For this, laterite from various places in Sri Lanka has to be

analyzed for the availability of high iron concentration and that can be used in this process.



**Fig.12- Proposed treatment train**

This method can be improved for direct application in the existing water treatment plants to supply water at a larger scale. Laterite should be mixed with raw water (contaminated with arsenic) at the coagulation stage, where alum is added to remove the turbidity of water. The treated water can be separated from the laterite sludge (which contains arsenic) at the settling tank. The efficiency of this treatment can be further increased by passing the treated water through a conventional sand filter. As described in literature [25,26], the accumulated sludge can be used to construct bricks. But a well defined method for the disposal of arsenic sludge should be researched.

This method can also be used at household level to treat small quantities of drinking water, especially within the rural community. For this, a column filter with granules of laterite, can be used. A similar type of filter with pieces of bricks is used in the dry zone of Sri Lanka to remove fluoride from ground water. Arsenic removal efficiency of this filter system with laterite has to be further researched.

### 3.5 Arsenic in Sri Lankan aquifers

As mentioned above the USEPA standard for arsenic in drinking water is 10 ppb and the Sri Lankan standard is 50 ppb. According to the survey carried out in 2001/2002 [15], Very low arsenic levels ( $\leq 1$  ppb) were reported in the samples analysed. In another study to analyse well water quality, less than 10 ppb average arsenic levels have been reported in 5 districts in Sri Lanka [20]. Furthermore no arsenic has been detected in another study conducted to check the availability of heavy metals in 40 water samples collected from different water sources in 5 districts in Sri Lanka [21].



## 4. Conclusions

Efficiency of arsenite removal using naturally available laterite in Sri Lanka is presented in this paper. More than 90% arsenite removal could be achieved within 5 minutes when pH is around 10. Therefore by treating the water at this pH range, the current USEPA standard for arsenic in drinking water (10 ppb) can be maintained when the arsenite / laterite ratio is less than 10( $\mu\text{g/g}$ ). This method can be improved for direct application in the existing water treatment plants to supply arsenic free water at a larger scale. For this further studies can be carried out to finalize an optimum dosage at natural pH range. Laterite pieces in column filters can also be used at household level to treat small quantities of drinking water, especially within affected communities. Therefore as a further study it is recommended to research the arsenite removal efficiency of this filter system in order to develop a low-cost water treatment methodology for the field application. The feasibility of this method in affected areas, with the presence of other constituents such as fluoride, sulfate, phosphate, nitrate and chloride has to be investigated.

According to the survey carried out in 2001/2002 [15], high arsenic levels were not detected in the surveyed ground waters and the current USEPA standard for arsenic in drinking water (10 ppb) was not exceeded in the ground water in the areas where samples were analysed (in shallow water, less than 15m depth). Furthermore, it is suggested to implement a 10 ppb maximum permissible limit as the new arsenic standard in drinking water for Sri Lanka. This may help to achieve public health protection goals.

## Acknowledgement

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## **SECTION II**