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Arsenic removal from groundwater by a combination of chlorination and consecutive sand filtration

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Abstract

The efficiency of arsenic removal from groundwater by chlorination coupled with adsorption on ferric hydroxide and sand filtration was evaluated in this study. Experiments were conducted at field using 5 groundwater resources containing total arsenic concentration of about 50 µg/L. The results showed that As (III) can be completely oxidized by chlorine in real waters containing low ammonia content (about 1 - 3 mgN/L). In cases of high ammonia concentration (8 - 20 mgN/L), As(III) oxidation was ineffective due to fast competitive reactions between ammonia and chlorine to form chloramines which reacts with As(III) much slower compared to chlorine. Following sand filtration can remove arsenic to some extent owing to As (V) adsorption on ferric hydroxide.

1. Introduction

Arsenic is a naturally occurring element in the earth's crust. Its existence in groundwater was explained by oxidative weathering, anoxic dissolution of As - bearing minerals and anthropogenic activities [1, 2]. Generally, arsenic occurs in groundwater at concentration below 10 µg/L and mainly in inorganic forms, arsenous acid (As (III)) and arsenic acid (As (V)). Under anoxic condition of groundwater, arsenous acid is predominant. Between the inorganic forms As (III) is more toxic than As (V). Groundwater is being used as one of supplying water resources. In Some areas in the world, including Banglades, India, Taiwan, Chile, Nepal, western United State and Vietnam [3, 4, 5, 6, 7], groundwater was found to be heavily contaminated by arsenic. Long term consumption of water containing elevated arsenic content can lead to internal cancer, electroencephalographic abnormalities, vascular diseases, severe hearing loss, disturbances of the haematopoietic system, peripheral neurological damage [8]. Due to its potential severe effect to human health, WHO revised the maximum concentration limit for arsenic in drinking water by decreasing it from 50 µg/L to 10 µg/L in 1995. This value was adopted as a regulatory limit by Vietnam in 2002. As the results of this revision, more groundwater resources in Vietnam have arsenic concentration exceeding new limit. Therefore simple arsenic treatment procedures are urgently needed not only for Vietnam but also for other countries.

Several techniques have been developed to remove arsenic from water such as oxidation combined with adsorption, coagulation, precipitation [9], ionic exchange, membrane filtration [10, 11] and biological method [12]. Because application of these methods is costly in Vietnam, it is infeasible to carry out in large plant scale. In groundwater, iron (II) is a common contaminant. It readily is oxidized by oxygen to form ferric hydroxide which has high adsorption capacity toward arsenic. Therefore using existing iron to remove arsenic is a promising option. This method could change water treatment facilities toward arsenic removal with minimum investment and chemical addition. Numerous studies reported that As (V) is more strongly adsorbed on ferric hydroxide than As (III) does. Therefore, firstly As (III) is needed to be oxidized for more effective removal. Among oxidants chlorine, which is widely used in Vietnam as a disinfectant, is a potential agent for As (III) oxidation. Kinetic of arsenite oxidation by chlorine has been studied recently [13]. The report showed that in distilled water extremely fast As(III) oxidation had been observed with half live time of 95 ms at 2 mg/L chlorine dose, pH = 7. However it took hours to completely oxidize As (III) in real water containing high amount of ammonia. In addition to ammonia, groundwater is contaminated by other reductants, which can effect on efficiency of arsenic removal such as ferrous iron, manganese, sulphur and organic matters. Field experiment should be done to evaluate effect of these contaminants on arsenic removal process. In this research five Hanoi groundwater containing a wide range of ammonia concentration from 1 to 20 mg N/L had been used.

2. Experiments

Materials and chemicals

All chemicals used for treatment were markedly available, all they were technical grade and used without further purification. NaOCl (8 – 10 %) was used as the source of chlorine. Stock solutions of chlorine (100 and 200 mg/L) were prepared daily and standardized by iodometric titration [14]. Deionized water was used for preparing chemical solutions for analysis. 1-mL polyethylene tip packed with 0.8 g of selective aluminosilicate adsorbent was used for selectively removal of As (V) from As (III) samples. Filtering column was made from PVC tube with the inner diameter of 39 mm, the height of 1200 mm. The column was filled by a 1 m layer of quartz sand, which has the size of 0.6 – 1.2 mm and is widely used in Hanoi water plants for conventional iron filtration. As (III) and total As (As_t) samples were preserved in HCl solution (1 %) and stored at 4°C until analysis.

Experimental procedure

The study was carried out at 5 Hanoi's water plants (denoted as S1, S2, S3, S4, S5) where conventional groundwater treatment is being employed. This method comprises of 4 steps as represented in figure 1.

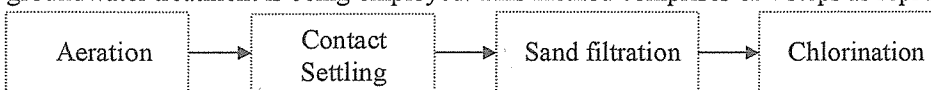


Figure 1. Scheme of conventional technology for iron removal from groundwater in Hanoi

For all experiments, water after settling step was collected and used as a target for As removal trial. Two liters of settled water was taken and mixed vigorously by magnetic stirring plate. A given volume of chlorine stock solution was dosed into the solution by syringe to get required chlorine concentration. In these experiments chlorine doses were applied as following: 0.0; 1.0; 1.5; 3.0; 5.0 mg/L. Then chlorinated samples were filtered through sand column simulating plant's rapid sand filter at the rate of about 5 m/h. This procedure of arsenic treatment is described in figure 2. After 15 minute filtering, the filtrate was collected for As (III), As_t analysis. For As (III) analysis samples were filtered through aluminosilicate cartridge to remove As (V). The experiment was duplicate for each chlorine dose to get average value. Experiments for each groundwater were conducted in the same day.



Figure 2. Schematic representation of proposed arsenic treatment method

3. Results and discussion

Two types of groundwater containing low (1-3 mg N/L) and high ammonia (8-20 mg N/L) amounts have been used in this study. Characteristics of this raw groundwater are given in table 1. The data showed that arsenic concentrations in raw groundwater are 5 - 8 times higher than Vietnamese standard (10 µg/L). Ferrous iron contents are relative high, which is expected to have an advantage for removing arsenic. Manganese concentrations are relative low except for S3 sample. Otherwise, manganese reacts slowly with chlorine [15], hence its effect on As (III) oxidation can be neglected in this study.

Table 1. Characteristics of raw groundwater

Factors	S1	S2	S3	S4	S5
pH	7.03	6.75	6.6	6.8	6.6
Fe(II), mg/L	3.7	15.4	3	10.2	12.7
Fe _t , mg/L	4	15.8	3.4	10.5	13.3
NH ₄ ⁺ , mg/L	1.7	9.8	2.6	19.7	8
Mn ²⁺ , mg/L	0.2	0	1.2	0.13	0.17
As _t , µg/L	45	50.5	41.5	72	80.5

Fe_t: total iron

Aeration and settling steps gave some change in water quality as given in table 2. pH value of water was elevated above 7.0 that is favourable for Fe (II) oxidation. Most of Fe (II) was oxidized in settling step to form ferric hydroxide except for S2 sample. During this step arsenic was adsorbed on newly formed ferric hydroxide. But less than 50% As_t was adsorbed. In settled water arsenic was mainly occurs in As (III) form. Ammonia was not removed during settling step because of negligible ammonia adsorption on ferric hydroxide. This factor is expected to be the main factor having effect on arsenic treatment efficiency.

Table 2. Characteristics of settled waters

<i>Factors</i>	<i>S1</i>	<i>S2</i>	<i>S3</i>	<i>S4</i>	<i>S5</i>
pH	7.4	7	7.3	7.6	7.3
Fe(II), mg/L	0.6	7.2	0.3	0.6	1.7
Fe _t , mg/L	2.7	15.1	1.1	7.5	10.2
NH ₄ ⁺ , mg/L	1.4	10.5	1.1	20	7.9
Mn ²⁺ , mg/L	0.2	0	1.1	0.1	0.1
As(III), µg/L	19	32	22.5	46.5	52
As _t , µg/L	23	34	22.5	48.5	55

By chlorination, it is expected that As (III) will be oxidized to As (V) that can lead to better arsenic removal in consecutive sand filtration. As (III) concentration in filtrates are given in table 3 and plotted in figure 3. In water containing low ammonia content (S1), chlorine dose of 1.5 mg/L is enough for complete As (III) oxidation. However, in case of S3 sample having 1.1 mg/L N-NH₄⁺, As (III) residual was relative high after chlorination and filtration. The reason for this phenomenon is still unknown. In water containing high ammonia (S2, S4, S5), low efficiency of As (III) oxidation was observed. This result is expected to be due to extremely fast reaction between ammonia and chlorine to form chloramines that is much less active in As (III) oxidation than chlorine [13].

Table 3. As (III) concentration in filtrate at various chlorine doses

<i>chlorine doses, mg/L</i>	<i>S1</i>	<i>S2</i>	<i>S3</i>	<i>S4</i>	<i>S5</i>
0	19	19	48.5	22	36
1	2.3	13.5	35	17.5	29
1.5	1	11	31	20.5	26
3	1	9.3	29.6	17	25.5
5	1	7	30.5	19	23.5

Results of iron analysis illustrated that iron concentration in water after sand filtration were below 0.4 mg/L, it means that As adsorbed on solid FeOOH was retained in the filtering media, therefore total arsenic in filtered water is soluble form. Results for total arsenic content in filtered water are given in table 4 and plotted in figure 4. The data reveals that arsenic was removed to some extent by sand column filtration. Incomplete as (V) adsorption may be due to slow kinetic of the process.

Table 4. As_t concentration in filtrate at various chlorine doses

<i>chlorine doses, mg/L</i>	<i>S1</i>	<i>S2</i>	<i>S3</i>	<i>S4</i>	<i>S5</i>
0	21	19.5	48	36.5	51
1	9.3	16.3	46		45.5
1.5	11.6	11.6	45	34	42
3	9.6	11.6	41	31	44
5	10	7.6	41	31	43

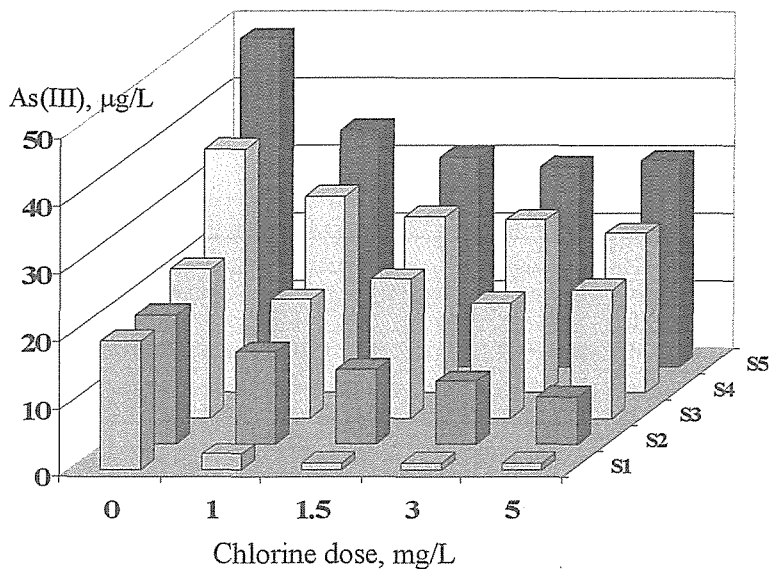


Figure 3. As (III) residuals after chlorination and sand filtration

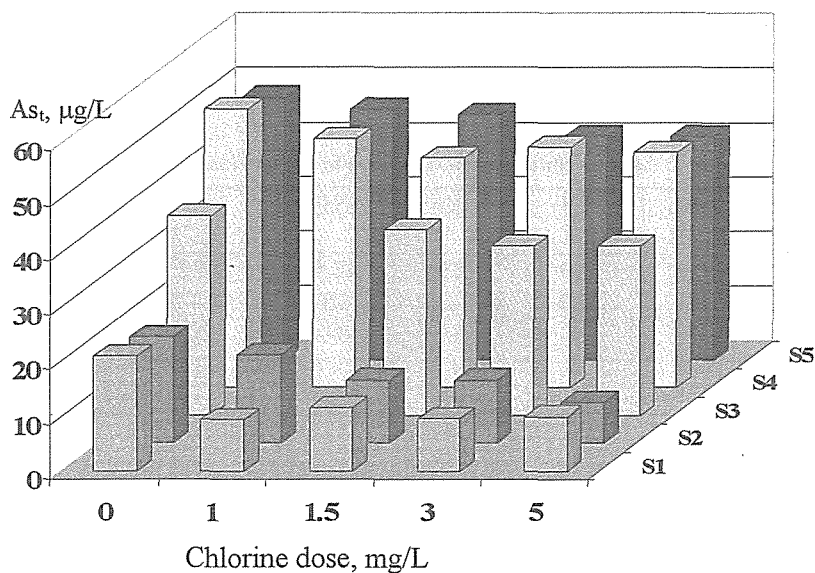


Figure 4. As(t) residuals after chlorination and sand filtration

4. Conclusions

Various doses of chlorine have been used to treat arsenic in groundwater in the field trials. Two types of groundwater containing low (1-3 mg N/L) and high (8-20 mg N/L) ammonia content were examined. It revealed that complete As (III) oxidation can be achieved in real water containing low ammonia content. However, low efficiency of As (III) oxidation can be also observed because of unknown reason. In groundwater containing high ammonia concentration, As (III) was inefficiently oxidized by chlorine due to relatively fast reaction between chlorine and ammonia to form chloramines which are much less active toward As (III). In this case other oxidants such as potassium permanganate, ozone, hydro peroxide can be alternative options. Following sand filtration can remove more arsenic owing to arsenic adsorption on ferric hydroxide

during filtration process. However, slow kinetic of the adsorption may be a limitation for effective arsenic removal by this process.

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Reference

1. Badal Kumar Mandal, Kazuo T. Suzuki (2002), Arsenic round the world: a review, *Talanta*, 58, 201–235.
2. P.L. Smedley, D.G. Kinniburgh (2002), A review of the source, behavior and distribution of arsenic in natural waters. *Applied Geochemistry*, 17, 517–568.
3. Pham Hung Viet, Pham Thi Kim Trang, Michael Berg, Nguyen Thi Minh Hue, Bui Hong Nhat, Vi Thi Mai Lan, Tran Thi Hao, Pham Thi Dau, Vu Thi Mai, Nguyen Van Mui (2004), Risk of arsenic contamination at tube wells in Red river delta, VNU's 3th Scientific workshop, Hanoi.
4. Nguyen Khac Hai, Dang Minh ngoc et al., Chander, Nguyen Quy Hoa (2004), Arsenic contamination in groundwater in Hoa Hau, Vinh Tru, Bo De communes of Hanam province, VNU's 3th Scientific workshop, Hanoi.
5. Ngo Ngoc cat, Le Van Cat, Ngo Viet Dung, Tran Thanh Tung (2004), Arsenic contamination in groundwater in Ha Nam, Phu Tho: situation and treatment technologies, VNU's 3th Scientific workshop, Hanoi.
6. J.M. McArthur, P. Ravenscroft, S. Safiullah, M.F. Thirlwall. Arsenic in groundwater: testing pollution mechanisms for sedimentary aquifers in Bangladesh. *Water Resources Research*, 37 (1), 109-117.
7. Mohammad Mahmudur Rahman, Mrinal Kumar Sengupta, Sad Ahamed, Uttam Kumar Chowdhury, Md. Amir Hossain, Bhaskar Das, Dilip Lodh, Kshitish Chandra Saha, Shymapada Pati, Imrul Kaies, Ajoy Kishore Barua, Dipankar Chakraborti (2004), The magnitude of arsenic contamination in groundwater and its health effects to the inhabitants of the Jalangi—one of the 85 arsenic affected blocks in West Bengal, India, *Science of the Total Environment*.
8. World health organization (1981), *ENVIRONMENTAL HEALTH CRITERIA 18: ARSENIC*, Geneva.
9. M. Clara F. Magalhães (2002), Arsenic: An environmental problem limited by solubility, *Pure Appl. Chem.*, Vol. 74, No. 10, pp. 1843–1850.
10. Judit Floch, Miklos Hideg (2004), Application of ZW-1000 membranes for arsenic removal from water sources, *Desalination* 162, 75-83.
11. Svetlozar Velizarov, Joao G. Crespo & Maria A. Reis (2004), Removal of inorganic anions from drinking water supplies by membrane bio/processes, *Environmental Science & Bio/Technology* 3: 361–380.
12. I. Katsoyiannis, A. Zouboulis, H. Althoff, H. Bartel (2002), As(III) removal from groundwaters using fixed-bed upflow bioreactors, *Chemosphere* 47, 325–332.
13. Michael C. Dodd, Ngoc Duy Vu, Adrian Ammann, Van Chieu Le, Reinhard Kissner, Hung Viet Pham, The Ha Cao, Michael Berg and Urs von Gunten (2006), Kinetics and Mechanistic Aspects of As(III) Oxidation by Aqueous Chlorine, Chloramines, and Ozone: relevance to drinking Water treatment, *Environ. Sci. Technol.*, 40, 3285 – 3292.
14. *Standard Methods for the Examination of Water and Wastewater*, 20th ed.; APHA, AWWA, WPCF: Washington DC, 1998.
15. Oliver J. HaoAllen P. Davis, Peter H. Chang (1991), Kinetics of manganese(II) oxidation with chlorine. *Journal of environmental engineering*, 117(3), 359 – 373.