Removal of arsenic from contaminated groundwater using laterite, sand and ash: a case study in Son Dong commune, Hoai Duc district, Ha Noi

Loại bỏ Asen trong nước ngầm bằng cách sử dụng đá ong, cát và tro tại xã Sơn Đồng, huyện Hoài Đức, Hà Nội

Research article

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Arsenic (As) contaminated groundwater has been a major concern due to the negative impacts to exposed people. This research was conducted to assess and compare the removal efficiency of As from groundwater by laterite, sand, and ash. The experiment was carried out in 14 days in a household scale in Son Dong commune, Hoai Duc district, Ha Noi. Groundwater was pumped directly from a well and flowed through 20 cm (diameter) x 80 cm (length) columns. The initial As concentration in groundwater was 526 µg/L decreasing to an average of 189, 192 and 154 µg/L after being filtrated using sand, ash, and laterite, respectively. Average removal efficiency of sand, ash and laterite during the experiment was 63.3, 63.9, and 70.5%, respectively. Laterite had higher As removal efficiency may be due to higher content of goethite and kaolinite in this sorbent which resulted in better adsorption of As. The concentrations of As in the outflow water were higher than the allowable limit set by the national technical regulation on drinking water quality (QCVN 01:2009/BYT). Therefore, it requires higher volume of sorbents or additional treatment technologies for removal of As from groundwater.

Ô nhiễm asen (As) trong nước ngầm hiện nay là một vấn đề cấp bách đối với xã hội bởi những rủi ro tiềm ẩn với sức khoẻ con người. Nghiên cứu được thực hiện nhằm đánh giá và so sánh khả năng hấp phụ As trong nước ngầm của đá ong, cát và tro. Thí nghiệm được tiến hành trong vòng 14 ngày đặt tại một hộ gia đình có nguồn nước ngầm bị ô nhiễm As thuộc xã Sơn Đồng, huyện Hoài Đức, Hà Nội. Nước ngầm được bơm từ giếng và chảy qua các cột đựng vật liệu có đường kính và chiều dài lần lượt là 20 cm và 80 cm. Nồng độ As ban đầu là 526 µg/L đã giảm xuống còn 189, 192 và 154 µg/L sau khi lọc bằng cát, tro và đá ong. Hiệu suất xử lý As trung bình của cát, tro và đá ong lần lượt là 63,3, 63,9 và 70,5 µg/L. Đá ong xử lý As tốt hơn có thể do hàm lượng goethit và kaolinit cao hơn trong vật liệu hấp phụ này dẫn đến khả năng hấp phụ As tốt hơn. Tuy nhiên hàm lượng As trong nước đầu ra vẫn chưa đạt quy chuẩn cho nước uống (QCVN 01:2009/BYT). Do đó, cần tăng thêm lượng vật liệu hoặc kết hợp với các phương pháp khác để xử lý As hiệu quả hơn.

Keywords: adsorption, arsenic, ash, laterite, precipitation, sand

1. Introduction

Arsenic (As) is a ubiquitous element found in the atmosphere, soils, rocks, natural waters and organisms. Of the various As sources in the environment, drinking water probably poses the greatest threat to human health [22]. A variety of treatment methods were developed for the removal of As from water, including coagulation [19], adsorption [5], ion exchange [2], electrocoagulation [10] and biological processes [8]. There is a variety of materials in treatment of arsenic contaminated groundwater such as bentonite [27], red mud [7], kaolinite [17], laterite [1] and raw laterite [23]. Raw laterite is an effective material

that is environmental-friendly, affordable and available in some areas of Vietnam such as Ha Noi, Vinh Phuc, and Thai Nguyen provinces.

Vietnam is one of the countries suffering serious As contamination in groundwater. The As concentrations in the Red River and Mekong aquifers vary within 1– 845 μg/L, and 1–3050 μg/L (average 159μg/L), respectively [4]. The increasing demand of groundwater for domestic and drinking water has posed high risks for exposed community. Ha Noi is considered one of the most affected areas in northern Vietnam. There are eight major well fields supplying water to this city which process 500.000 m^3 of water per day. However, the high As concentrations found in 48% of the tube-wells greater than 50 μg/L and in 20% of the tube-wells greater than 150 μg/L indicated that several million people consuming untreated groundwater might be at a considerable risk of chronic arsenic poisoning [3].

Hoai Duc is a district in Hanoi which suffers from As contaminated groundwater. The average As concentration in groundwater in Hoai Duc was approximately 261 μg/L [24]. High awareness of As problem in groundwater was observed in Son Dong commune, Hoai Duc district. Local community use both rainwater and filtered water in daily life. Some natural materials (e.g., sand, gravel, ash, pebbles) have been widely using for filtering groundwater at household scale. However, the effectiveness of these materials for As removal to meet the required regulation has not been determined. In addition, the application of natural laterite to filtrate groundwater at household scale has not been reported.

The objective of study is to assess and compare the efficiency of removing As from contaminated groundwater using laterite, sand and ash through an experiment at one household in Son Dong commune, Hoai Duc district, Ha Noi.

2. Materials and methods

2.1. Field survey

The first survey was conducted in six households in Son Dong commune, Hoai Duc district, Ha Noi in February, 2017. In these sites, sand and ash are two most common materials that local inhabitants use to filter groundwater. The outflow water after filtration is used for both drinking and domestic purposes.

A total of 12 groundwater samples at 6 households were collected. The household with highest As concentrations will be selected for further experiment.

2.2. Experiment setup

Laterite was collected from Thach That district of Ha Noi. Figure 1 shows the experiment which was located in a household at 21°02'50.3"N; 105°41'59.2"E in Son Dong commune, Hoai Duc district, Ha Noi.

Figure 1. Experiment setup

The pilot experiment was designed three replicated with three kinds of columns containing sand, ash, laterite (Figure 1). Each material column is 80cm in length and 20cm in diameter to make the ratio of diameter-to-length of 1:4 [13]. The weight of sand, ash, and laterite in each column was approximately 25, 2.2, and 20 kg, respectively. The inflow water was pumped directly from groundwater in the selected household water with a velocity of 300 ml/min. The experiment was carried out continuously in 14 days, given the constant As concentrations in the outflow water. Inflow and outflow water samples were collected after 1 hour, 3 hours, 6 hours, 1 day, 2 days, 4 days, 6 days, 8 days, 11 days, and 14 days after setting up. A total of 70 water samples were collected for determination of As concentrations.

2.3. Sample treatment and analysis

Water samples were first filtered through 0.45μm before As and Fe determination by Atomic Absorption Spectroscopy (AAS, 240FS, VGA77, Agilent) at the VNU Key Laboratory of Geo-environment and Climate change Response.

Samples of laterite, sand and ash were dried using the NIIVE OVER KD200 oven at the temperature of 80 – 105°C until the sample weight was constant. After the samples were dried, samples were crushed into fine powder using the MRC laboratory Equipment Manufac Urer. Mineral compositions of the material samples were determined using the X-ray Diffraction (XRD - Siemens D5000) at VNU University of Science.

2.4. As removal efficiency calculation

The As removal efficiency (%) of different materials is calculated by the equation:

$$\% = \frac{C_o - C_e}{C_o} \times 100\%$$

Where: C_o: Initial As concentration (μg/L)

C_e: As concentration at equilibrium state (μg/L)

3. Results and discussions

3.1. Mineral compositions of materials

The results of this study demonstrated that the main minerals of sand and ash were quartz with the proportions of 90% and 8.2%, respectively. The main components of laterite were goethite (42%) and kaolinite (40%). Other minor components of laterite were quartz (9%), hemantite (3%), titanite, azurite, magnetite, pyrite.

3.2. As concentrations (µg/L) and Fe/As ratios in study sites

The initial As concentrations measured in six study sites from HD1 to HD6, in Son Dong commune, Hoai Duc district varied from 82 µg/L in HD1 to 575 µg/L in HD4 (Figure 2). The As concentrations of HD2, HD3, HD5 and HD6 were 290, 302, 286 and 328 µg/L, respectively (Figure 2). The average As concentration in 6 sites was 311 µg/L. The site HD4 was selected for further experiment due to the highest concentration of As among the 6 investigating sites. At low horizontal hydraulic gradients and under reducing conditions, As was released in groundwater by microbial activity, causing widespread contamination in the low-lying deltaic and floodplain areas [21]. The weathering of As-bearing minerals released this element into water flows. Under the influence of tectonic, geodynamic, those As-bearing materials were transported from the high-altitude areas to low-altitude areas. The relationship is non-linear between As content in groundwater and surface elevation of study sites. This indicated that the surface topography is inversely related with As concentration in the study area [9].
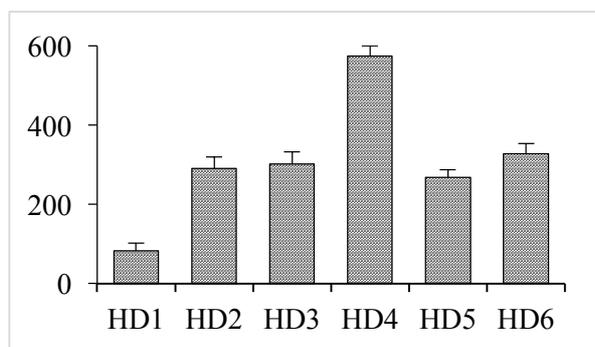
Figure 2. Initial As concentrations in study sites

Table 1. Fe/As ratios in sampling sites

Sampling sites	Fe/As ratio
HD1	51
HD2	58
HD3	49
HD4	21
HD5	45
HD6	57

The proportion of dissolved iron (Fe) and As present in groundwater is a suitable parameter for estimating the As removal potential. The concentration of dissolved Fe is a key parameter for As removal from groundwater [4]. Table 2 showed that Fe/As ratios in groundwater collected at six study sites from HD1 to HD6 were 51, 58, 49, 21, 45 and 57, respectively. The different concentrations of Fe may be one of the reasons causing the change of As concentrations in locations because As removal is directly related to the concentration of Fe added to the system. The high content of Fe available in groundwater may combine with As via co-precipitation which results in the removal of As. For example, to reduce the As concentration from 100 µg/L to reach the value less than 5 µg/L, it requires the Fe/As ratio as 40 [18]. Another study indicated that the Fe/As ratio of 50 or more was needed to reduce As concentrations to levels below 50 µg/L. To reach the WHO drinking water guideline of 10 µg As/L, Fe/As ratios of >250 were required [4]. Fe/As ratio measured in the experiment site HD4 – the selected experiment site – was 21 which may result in high concentrations of As in this area. Low Fe/As ratio in this site also highlights the need for using Fe-rich materials for treatment of As contaminated water to regulated level.

3.3. Removal of As by laterite, sand, and ash

3.3.1. pH

Figure 3 showed pH values of the inflow and outflow water samples. The pH values of water passing through laterite columns slightly decreased while those of sand and ash increased during the experiment. However, the pH values always remains within the allowance limit of pH in drinking water (QCVN01:2009/BYT).
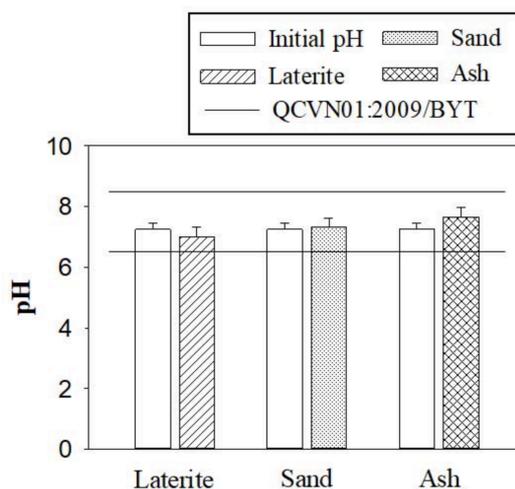
Figure 3. pH values in inflow and outflow water samples

3.3.2. Removal of As in groundwater by laterite, sand, and ash

Average As concentration in inflow water was 524 µg/L which decreased to 73, 80, and 82 µg/L by laterite, sand, and ash, respectively in one hour of treatment (Figure 4). After 14 days of the experiment, the average As concentrations decreased to 189, 192 and 154 µg/L by sand, ash and laterite, respectively. The removal of As by laterite, sand, and ash was due to the oxidation of iron and arsenite and precipitation of iron(III) arsenate [12] as follow:

$$Fe^{3+} + HAsO_4^{2-} \rightarrow FeAsO_{4\ (precipitation)} + H^+$$

Therefore, laterite, sand, and ash using for filtration of groundwater may provide a surface network to mechanically keep the precipitation of As and Fe compounds. Fe/As ratio in groundwater is thus the important factor affecting the As removal capacity [4].

The results of this study showed the significant decrease of As concentrations by laterite in comparison with those by sand and ash ($p<0.05$). However, no significant difference of As concentrations in outflow water between using sand and ash was obtained. Higher content of goethite (42%) and kaolinite (40%) in laterite may cause the adsorption of As to these minerals and thus resulting in better removal of As from contaminated groundwater [23][14].

Many studies showed that higher As removal efficiencies can be generally achieved with increased Fe concentrations. As adsorbed onto ferric hydroxide was investigated for arsenite and arsenate in a pH range of 4-9 at varying Fe to As ratios. The extent of arsenite adsorption at pH 4 and 7 demonstrated strongly dependent on total arsenite concentration, and increased adsorption was observed with increasing Fe/As ratios [26]. It was also observed from this study that As concentrations in outflow water were higher than the limit set by national technical regulation on drinking water quality (QCVN 01:2009/BYT) [19]. This fact highlights the need to add more sorbents, to use modified materials for better adsorption, or to combine with other technologies in order to meet the regulation of As in drinking water. In addition, the sand columns started to stuck after 2 days of experiments, possibly due to the precipitation filling the porosity of these sand columns.

The highest As removal efficiency was obtained on the first day of the experiment. From the second to the end of the experiment, As removal efficiencies by the 3 materials were almost constant. The average As removal efficiency

of laterite, sand, and ash during 14 days of the experiment was 70.5, 63.9, and 63.3%, respectively (Figure. 5). As removal by laterite was significantly higher than that by sand and ash ($p<0.05$). The results of this study also demonstrated lower removal efficiency of As than those reported in the previous studies (Table 2). The differences were possibly due to the differences in characteristics of sorbents, initial concentrations of As in groundwater, composition of groundwater and experimental designs.
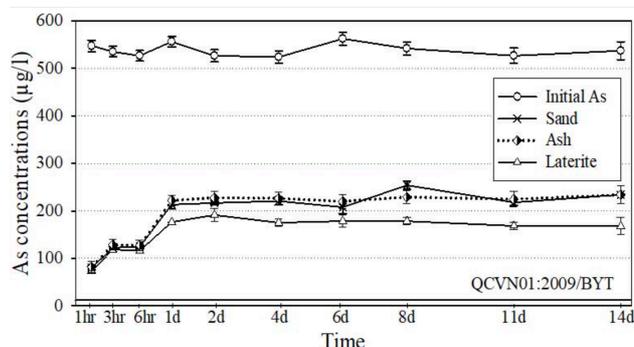
Figure 4. As concentrations (µg/L) in inflow and outflow waters
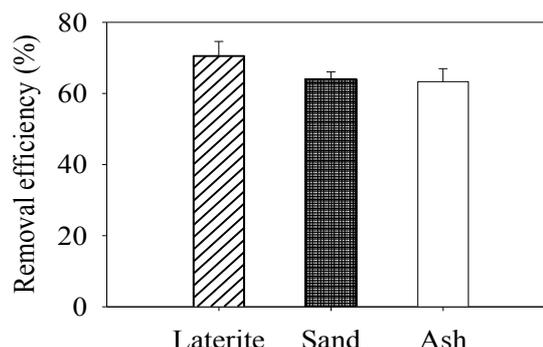
Figure 5. Average As removal efficiency of materials during 14 days of the experiment

Table 2. Comparison of different As sorbents with similar column filter (*Note: - No data*)

Sorbents	As initial concentration (µg/L)	Column diameter (cm)	Flow rate	As removal efficiency (%)	As outflow concentration (µg/L)	Reference
Laterite	526	20 x 80	300 ml/min	70.5	154	This study
Laterite soil	330	2 x 10	7.75 ml/min	98	10	[15]
Manganese greensand	100	-	1.5 l/min	81	-	[25]
Iron oxide-coated cement (IOCC)	2,000	2 x 10; 2 x 20; 2 x 30	8.5 ml/min	-	10	[11]
Siderite–hematite	500	0.3 x 15	0.51 ml/min	99	< 10	[6]
Calcined bauxite ore	2,000	6 x 30	200 ml/h	-	< 10	[16]

4. Conclusions

The initial As concentration is 526 µg/L decreased to an average of 189, 192 and 154 µg/L by sand, ash and laterite during 14 days of the experiment. As removal efficiency of sand, ash and laterite was 63.3, 63.9 and 70.5%, respectively. As removal from groundwater by laterite, ash, and sand may be due to precipitation. Higher As removal efficiency of laterite than ash and sand was possibly due to higher content of goethite and kaolinite in laterite. The materials should be modified or combined

with other technologies for treatment of As in groundwater to meet the regulation for drinking water (QCVN01:2009/BYT).

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