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Supplementary Information

Removal of *As(III)* using a natural laterite fixed-bed column intercalated with activated carbon: solving the clogging problem to achieve better performance

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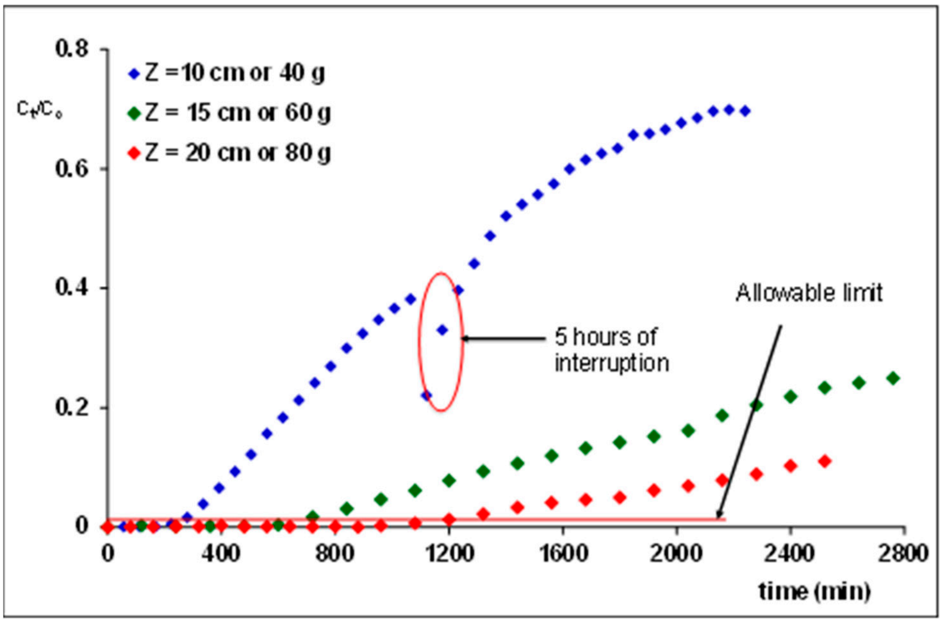
Supplementary information on the previous literature data (SI1)

The fixed-bed column adsorption of As(III) on laterite soil was investigated for the first time by Maji et al. **[1]**. The authors utilized a small laboratory column (diameter of 2 cm; bed heights of 10 cm, 20 cm, and 30 cm; and an initial concentration of arsenic of 0.5 mg/L) configuration consisted of a single layer of laterite soil placed between two layers of glass wool. However, no further tests were carried out using fixed-bed columns with large diameters and small-sized particles of laterite. Maiti et al. investigated a household column filter (diameter: 15 cm) for arsenic adsorption **[2]**. The filter was based on two laterite layers (raw laterite (35 cm) and treated laterite (34 cm)) placed between two layers of sand (15 cm and 10 cm, respectively). Recently, on the basis of their results obtained with a small laboratory column (diameter: 0.35 cm, height: 4 cm), Sarthak et al. proposed a new field filter system (internal diameter: 35 cm, height: 97 cm) by integrating a single layer of acid-activated laterite (size: 0.165 mm) placed above a fine sand layer and below a diffusion chamber containing iron nails, which improved the removal capacity of the arsenic and other heavy metals (manganese, cadmium, lead, antimony, mercury, nickel, and copper) **[3]**. Moreover, they integrated a bed of activated carbon (10 cm) in between layers of fine sand and coarse sand, which enhanced the removal of pathogens and the adsorption of iron from the water sample, as well as a gravel bed under the coarse sand **[3]**. It is worth noting that their filter system was a proposed one, and field tests were not conducted. The characteristics of the filter were only given in terms of technical, as well as socioeconomic, feasibility. In 2020 and 2021, Nguyen et al. investigated two column configurations with successive layers, including a single laterite layer **[4-5]**. In the first study, a field adsorption column with a diameter of 0.76 m comprised two ordering layers, as follows: a sand layer (0.5 m) at the bottom followed by natural laterite (0.4 m) and commercial granular activated carbon (0.3 m) **[4]**. In the second study, they experimented first with a small laboratory column with a 30 mm inner diameter and a 0.50 m height. To prevent any migration and to allow for the uniform distribution of the solution through the column, the natural laterite layer (41 cm) was placed between two layers of 1.0 mm acrylic beads and column balls. On the basis of the laboratory column adsorption study, they set up a household filter with an inner diameter of 14 cm and a height of 65 cm, and the natural laterite was placed between two layers of sand **[5]**. However, in all of the above studies, no permeability data were provided to explain why these configurations improved the performance of the percolation treatment.

Supplementary information on the origin of clogging (SI2)

Studies carried out in our laboratory on arsenic removal by column percolation using lateritic materials have shown its effectiveness in treating arsenic-polluted water **[6].** The characteristics of the fixed-bed columns were the following: 10, 15 and 20 cm as bed height, 2, 3 and 7 cm as diameter. To study the effect of bed height on breakthrough curves, a flow rate of 3 mL/min and a particle size between 0.595 and 0.71 mm and a column diameter of 2.5 cm were maintained. The bed heights were 10 cm (40 g), 15 cm (60 g) and 20 cm (80 g). Figure 1 shows the breakthrough curves for different bed heights. The breakthrough times at the column outlet corresponding to bed heights of 10 cm, 15 cm and 20 cm are 224 min, 600 min and 960 min, respectively. The volumes of treated solutions corresponding to 10 cm, 15 cm and 20 cm bed depths at the breakthrough points were 672 mL, 1800 mL and 2880 mL, respectively. The breakthrough time increased from 224 min to 600 min for a height of 10 cm to 15 cm, i.e. an increase of 6h 16 min. For a height of 10 cm to 20 cm, the breakthrough time increased from 224 min to 960 min, i.e. an increase in breakthrough time of 12h 16 min. These results clearly showed that for a better performance of the laterite-filled column i.e. a longer breakthrough time, a higher bed depth was desirable. These results indicated that an increase in breakthrough time and volume of solution treated as a function of bed depth was linked to the greater availability of the adsorbent surface, which provided more binding sites for arsenic adsorption **[7]**. However, a thorough understanding of column behavior required knowing what might happen with higher bed height and larger diameter. As a result, starting from a bed height of 20 cm, we increased the column diameter from 2.5 to 7 cm; this choice was based on ground field column dimensions, which are generally larger than those used at laboratory scale **[6]**. This research into arsenic removal using a fixed-bed column filled with laterite revealed limitations in its application with 7 cm diameter columns and bed height higher than 20 cm **[6]**. We found that the effluent stopped percolating through the fixed-bed column after a certain flow period. This issue, which is linked to the slowing down of the passage of water over the long term, has been identified as clogging by laterite particles. These findings constituted the entry point of the present submitted work, which extended the investigations on the following parameters: fixed-bed column height, laterite particles size, and arsenic (III) initial concentration.



Figure

Figure 1. Breakthrough curve of As(III) for a fixed bed filled with the laterite adsorbent at different depths (C_0 = 1,5 mg/L and Q = 3 mL/min)

It may be noted that the clogging issue is already known to exist in other systems **[8-10]**, but it has not yet been demonstrated for fixed-bed columns filled with natural laterite. In the case of percolation through a laterite-lined column, small-sized laterite particles exhibit low permeability to water, which leads to the clogging of the adsorptive porous system. As part of our investigations, we carried out experimental tests with other types of swelling materials such as local bentonite and smectite. A fixed-bed column (20 cm as bed height and 7 cm as diameter) was filled with either local bentonite or local smectite and water was allowed to percolate in the column. As can be seen in the video, we did not get any water flow out of the column **(Video_ Clogging with natural local materials_mp4)**. These results confirm the difficulty faced when the percolation material is a swelling material in a fixed-bed column.

Supplementary information on sand (SI3)

Experimental methods of the sand sample characterization

The BD sample was analysed for particle size. After washing and drying, a 100 g sample was sieved using a series of 7 AFNOR standardised sieves. The mesh sizes in mm were: 1; 0.5; 0.4; 0.35; 0.25; 0.20; 0.16; 0.125. The calculation of the weight percentage of each refusal and that of each pass are given by the expressions**.**

$$\% \textbf{ refusal} = \left(\frac{\mathbf{A}}{\mathbf{B}}\right) \times \mathbf{100} \qquad (1)$$

$$\% \textbf{ exceeding} = \left(\frac{\mathbf{B-A}}{\mathbf{B}}\right) \times \mathbf{100} \qquad (2)$$

A is the mass of reject in g and B is the initial mass BD sample.

These percentages are used to establish the grading curve of the sand sample and to determine the Uniformity Coefficient (UC):

$$\mathbf{CU} = \frac{\mathbf{d_{60}}}{\mathbf{d_{10}}} = \frac{\mathbf{d_{40}}}{\mathbf{d_{90}}} \quad (3)$$

d_{60} (d_{40}) is the diameter allowing 60% of the sand to pass through (retaining 40% of the sand) and d_{10} (d_{90}), for 10% of the sand to pass through (90% refusal). Knowing the CU enables us to determine whether the sand is homogeneous and therefore suitable for filtration. Homogeneity is respected if $1.2 \leq CU \leq 1.8$ .The diameter allowing the passage of 10% sand is called the effective size, TE = d10**.**

Sand characterization results

The curve describing the particle size is shown in **Figure 2.** From the particle size analysis curve, we determined the main particle size parameters of BD sand and its uniformity coefficient (Table 1). The results obtained showed that the BD sand has a compliance coefficient (**CU = 1.8**) which is within the required homogeneity range. BD sand is therefore suitable for filtration. As an inert material, BD sand was used to carry out a comparative performance study, in the adsorption of As(III), between the laterite column intercalated with sand layers and the laterite column intercalated with activated carbon layers.

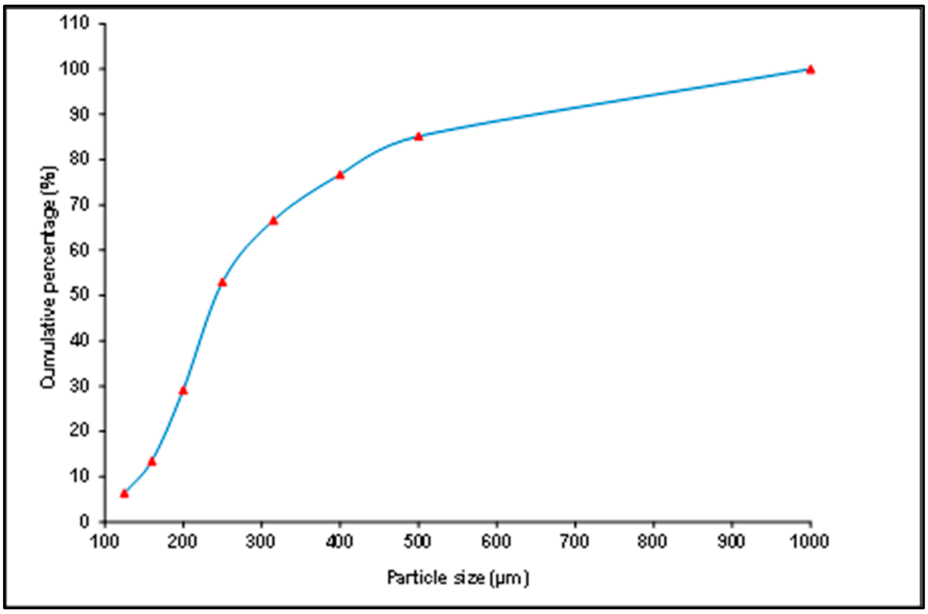


Figure 2. Size distribution of BD sand

Table 1: Main particle size parameters and uniformity coefficient for BD sand.

d_{10} (µm)	d_{50} (µm)	d_{60} (µm)	d_{90} (µm)	CU
150	240	275	650	1.8

Supplementary information on the choice of flow rate 50 mL/min (SI4)

The flow rate in this study was set to 50 mL/min. Indeed we experimented the fact that a 50% lower flow rate makes it possible to treat a larger volume of solution. But as we demonstrated in our previous investigations **[6]**, an operation at a lower flow rate of 20 mL/min lasted 120 min and we treated a volume of water of 2.400 mL. However, in our context, we had to make a compromise between a short time (with a flow rate of 50 mL/min) needed to treat a daily amount of water acceptable for the rural population and a long time (with a lower flow rate smaller than 50 mL/min) needed to achieve a larger volume of solution. We found that a 50 mL/min flow rate was the optimal flow rate in our study at real sites working conditions in villages.

Supplementary information (SI5): Laterite fixed-bed column intercalated with gravels layers.

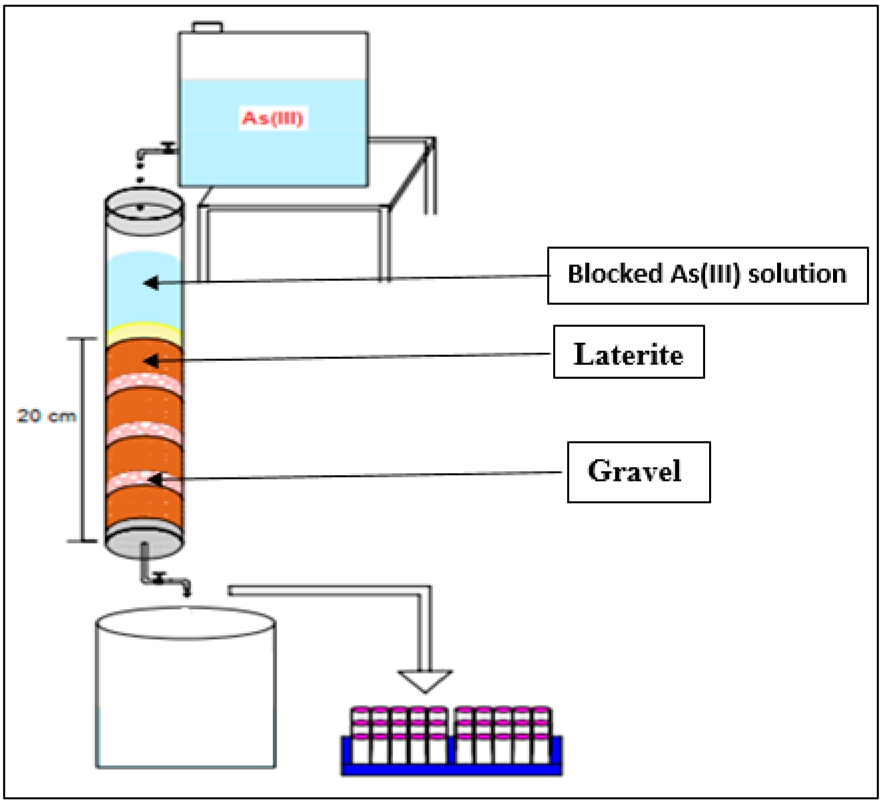


Figure 3. Laterite fixed-bed column intercalated with gravels layers.

Supplementary information (SI6): Fixed-bed column cost analysis: laterite column intercalated with activated carbon (BA-AC) layers and fixed-bed column only filled with activated carbon

Table 2. Cost of fixed-bed column configurations

Column configuration	Cost of treatment (USD)

laterite fixed-bed column intercalated with activated carbon layers	48.92
fixed-bed column filled with only activated carbon	223.87

Supplementary information on the saturation point of the column (SI7)

In the literature, experiments carried out with field-scale columns (column diameter greater than 4 cm) generally do not allow full saturation to be achieved in less than one year **[2,5].** In our context, given that we are using a field-scale column with a diameter of 7 cm, saturation is not expected to be reached within a few months. Indeed, due to the mass used in such columns, it can take months to reach full saturation ($C_t/C_0 = 1$). In our case, as we do not reach the point of complete saturation, a good approximation will be to consider that the column is (i) exhausted when we reach a breakthrough point, which is set at a value of 10 µg/l by the WHO, and (ii) saturated when the concentration of As(III), at a time t, is greater than 80%C_0 (C_t/C_0>0.8). Indeed, this consideration takes into account current working conditions on contaminated sites.

Supplementary information on the proposed domestic filter model (SI8)

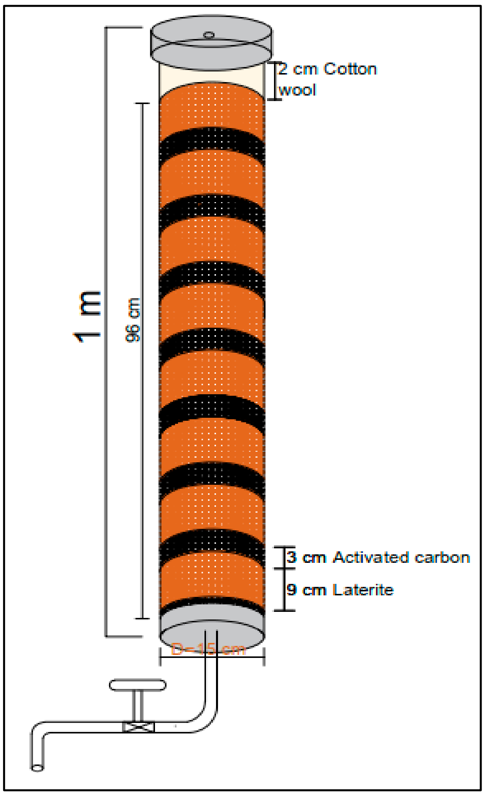


Figure 4. Proposal for a household filter based on DA laterite alternated with BA-AC activated carbon

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