Column Experiments on Arsenic Removal Through Adsorption From Water Using Different Natural and Synthetic Adsorbents

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Research Article

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Abstract

With the aim of exploring a best adsorbent from locally available sands for removing arsenic from water, eight different adsorbents are tested through column experiments using those materials as filter bed. Based on earlier batch experimental results five locally available sands (Scoria, Skye, Iron ore, NT red and TGS), one commercial sand (GFH) and two synthetic sands (IOCS and IOCS-AOCS) were selected for the column experiments. Target was to treat arsenic from water up to WHO standard level of 10 µg/L for a considerable period. It is found that Skye sand is capable to treat arsenic-contaminated water to the WHO standard for the longest period, followed by TGS, Iron ore and NT red sands. Scoria sand is unable to treat water up to the WHO standard. Although, GFH, IOCS and IOCS-AOCS are capable to remove arsenic to an excellent level, however practically not suitable as they get clogged due to accumulation of finer particles in the filter bed. Also, it is found that artificial coatings enhance the arsenic removal capabilities, however susceptible to clogging.

Introduction

Arsenic contaminations in drinking water has become a global issue and there are several natural weathering reactions and anthropogenic activities behind it. According to World Health Organisation (WHO), arsenic contamination above 10 µg/L is not acceptable in the drinking water (Ahmad et al., 2020). It is evident that long-term exposure and consumption of water having higher concentration of arsenic causes several health hazards (skin diseases, pigmentation and neurological disorders) and even cancer (Maity et al. 2012; Islam and Islam 2010). Although arsenic in the natural water present in both the organic and inorganic forms, its inorganic form is more toxic as compared to the organic form. Most common forms of arsenic in the natural water are As(III) and As(V) depending on the redox potential and pH of water (Mohan and Pittman, 2007). Due to its character of high reactivity with oxygen, arsenic in the groundwater converts to inorganic arsenic such as pentavalent arsenate, As(V) and trivalent arsenite, As(III) (Pizarro et al., 2021). In the surface water, arsenic is mainly present as oxyanions (H_2AsO_4^- , HAsO_4^{2-}), whereas in the groundwater the As(III) is stably present as arsenious acid (H_3AsO_3) (Gu et al., 2005).

Both the commonly found inorganic species, i.e. As(III) and As(V) are potential hazards to the environment and humanity. Being a major threat to the community, over the years various physicochemical techniques have been proposed and developed for the removal of arsenic from drinking water. Among the reported methods, oxidation method, coagulation method, membrane technologies and adsorption methods are widely studied and reported in the literature, and Murugesan et al. (2006), Amrose et al. (2013), Bora et al. (2016), Choong et al. (2007), Ioannis et al. (2003), Goren and Kobya (2021) and Boussouga et al. (2021) are few examples. Most of the approaches or techniques utilised are effective in removing higher concentrations of arsenic, i.e. through treatment arsenic concentration in the treated water becomes lower, however in most cases is not removed completely. Moreover, most of those techniques are associated with high operational cost. In the contrary, the adsorption techniques are

effective even on the lower concentration of the arsenic and is less expensive. The adsorption process involves passing the contaminated water through a column having packed/compacted bed of solid adsorbent material, which are preferably taken from different natural sources (Shakoor et al., 2017).

Callegari et al. (2018) investigated effectiveness of column filter with iron oxide coated sand (IOCS) to remove arsenic (As[V] and As[III]) from groundwater and reported that arsenic removal rates of 99% are achievable with an easy-to-operate process. Majumder (2018) conducted experimental and mathematical modelling study on use of activated alumina for removing As[V] from water and investigated effects of contact time, adsorbent dose, pH level, and initial arsenic(V) concentration on the removal efficiencies. Recently researchers are focusing on the variation and modification of adsorbent material for the high removal of arsenic from drinking water. Pizarro et al. (2021) demonstrated the magnetite-nanoparticle-supported modification on micrometric-zeolite enhances the properties of zeolite as an arsenic adsorbent. Gu et al. (2005) investigated granular activated carbon is an exceptionally good carrier substrate for iron impregnation to increase the adsorption capacity for arsenic removal. Yao et al. (2014) synthesized a magnetic composite adsorbent with activated carbon and iron oxide for the removal of As(V). They have reported that a synthesized magnetic composite as compare to pure activated carbon provides high adsorption capacities.

Mohan and Pittman (2007) provided a detailed review of different adsorption techniques used for arsenic removals. Though different researchers investigated effectiveness of using different synthetic materials for adsorption processes, as most of the communities who need using such treatment method(s) are unable to afford expensive treatment options, demand for using low-cost, locally available materials as adsorbent is quite high. Saldaña-Robles et al. (2018) reported effectiveness of granular ferric hydroxide as adsorbent in removing As(V) from water. With the emergence of sustainability awareness, recently researchers are focusing on different types of natural sorbents, as well as some by-products and/or waste materials. A few researchers worked on the modification of natural adsorbents to enhance the removal efficiency of arsenic. Asere et al. (2019) presented a detailed review of chemically modified natural adsorbents used for arsenic treatment. Zhang et al. (2004) used natural iron ores for the removal of arsenic from water. Shaikh et al. (2020) studied arsenic removal efficiency by biochar-based iron nanocomposites. Zeng et al. (2021) investigated use of a novel core-shell magnetic nanoparticles prepared from iron-containing water treatment residuals as arsenic removing adsorbent. Villela-Martínez et al. (2020) investigated use of bone char as adsorbent in removing arsenic from water. Byambaa et al. (2021) derived adsorbent material from acid mine drainage sludge for the removal of arsenic from water. However, uses of any such recycled material or waste by-products are subject to stringent environmental regulations, which often require thorough investigations for any potential pollutants present in the recycled/waste material. As such, considering strict regulations and cost-effectiveness, filter media made from sand or ceramic is commonly used. Also, it is an objective to secure/make these filters based on locally available material. Consequently, several studies focused on sand/ceramic made filter media. However, most of the past studies on arsenic removal with natural adsorbents were investigated through batch experiments (Pizarro et al., 2021; Khan and Imteaz, 2021; Zeng et al., 2021; Shaikh et al., 2020; Villela-Martínez et al., 2020; Khan and Imteaz, 2020; Majumder, 2018; Saldaña-Robles et al., 2018; Yao et

al., 2014). In many cases under a practical scenario the adsorbents exhibit different removal efficiency compared to an efficiency obtained through batch experiment. Moreover, under continuous operation the removal efficiency deteriorates, which can only be investigated through column experiments or full-scale filter. Khan and Imteaz (2021) reported a detailed study involving batch experiments with six different types of natural adsorbents and concluded that Skye sand is having highest arsenic removal capacity. To replicate the Skye sand, Khan and Imteaz (2020) studied the effectiveness of synthetic adsorbents such as iron oxide-coated sand (IOCS), aluminium oxide-coated sand (AOCS) and different mixtures of these synthetic adsorbents. They have reported that IOCS and mix of IOCS-AOCS(50%-50%) are having even higher arsenic removal capacities compared to Skye sand. To study the arsenic removal efficiencies of these (natural and synthetic) adsorbents under practical scenario, this study describes detailed column experiments with the same adsorbents.

Materials And Methods

Five different types of natural materials were collected from different parts of Australia for the current study. Among the studied materials, Skye, TGS and Scoria sands were collected from Melbourne, Victoria. Red sand was collected from Alice Springs, Northern Territory and iron ore sample was collected from Pilbara, Western Australia. Selection of these sands were based on the criteria of the easy availability within Australia, where the experiments were conducted. To assess the locally available materials' removal efficiency with a benchmarked material, a sixth type of sand was selected. This additional sand sample is commercially available and known as granular ferric hydroxide (GFH). GFH sand was obtained from GEH Wasserchemie GmbH & Co., Osnabrueck, Germany. Detailed physical characteristics and arsenic removal mechanisms of these samples were discussed by Khan and Imteaz (2021). The synthetic adsorbents, IOCS and AOCS were prepared in the laboratory using finer fractions (150–300 micron) of quartz sand. Details of the preparation of the synthetic sands and their effectiveness are outlined in Khan and Imteaz (2020).

Column tests were carried out in a glass tube attached to a 500 mL water reservoir. The diameter of the tube was 3 cm and the filter bed was around 15 cm in depth. The column was stabilized with clamps and stands. The filter bed was made with the selected natural adsorbent for each type of selected sample. The volume of each filter bed was 100 mL. However, the mass of the filter bed was different as the different selected materials were having different specific gravities. The specific gravity of Skye sand, TGS sand, NT red sand, Scoria and Iron ore were 2.58, 2.57, 2.52, 1.32 and 4.3, respectively. With a 100 mL volume, the corresponding masses of Skye sand column, TGS sand column, NT red sand column, Scoria column and Iron ore column were 258, 257, 252, 132 and 430 g, respectively. For each of the adsorbents, three similar filters were prepared to obtain a representative adsorption capacity of the adsorbents. The initial concentration of arsenic in the feed water of filters was 100 µg/L. However, to assess the removal efficiency of some highly efficient samples, in the later stage for some of the filters, the arsenic concentration in feed water was increased to 500 and 1000 µg/L. The hydraulic capacity of the filters was fixed to 500 mL per hour by a control valve. The pH of feed water was adjusted to 7.0. All

the column experiments were carried at 22° C. The filtrate from the column was collected with a beaker of 1L capacity. Special care was taken to prevent air-trapping in the filter bed by maintaining a water head of (5 cm) on filter bed at the intermission of flow. For arsenic analysis, samples were taken at regular intervals from the filtrate in the beaker.

Arsenic concentrations were measured with an atomic absorption spectrometer (GBC, Model no. 906/AA) fitted with a hydride generation system (HG-AAS 3000). The parameters used for analysis of arsenic by HG-AAS are: wavelength 193.7 nm, band pass 1.0 nm, lamp current 8.0 mA and fuel (acetylene) 1.3 L/min. Arsenic standard solution (1000 mg/L) was used for the calibration of the spectrometer. Sodium arsenite (NaAsO_2) was used for the preparation of arsenic contaminated stock solutions for the absorption experiments. Stock solutions having arsenic concentration of 1000 mg/L were prepared by dissolving 0.867 g of NaAsO_2 in to 500 mL of Deionised (DI) water. Arsenic was in the trivalent form both in the standard solution and NaAsO_2 . Feed water of different arsenic concentrations were prepared by diluting this stock solution with DI water by appropriate ratio.

Achieved arsenic removal efficiencies through column experiments for different sands were graded from highest to lowest. Also, column test results were compared with the earlier batch experiments conducted with the same samples as reported in Khan and Imteaz (2021).

Results

As mentioned earlier for each material, three columns were prepared and tested with the same arsenic contaminated water. To replicate the continuous operation in the real-life, the contaminated water was filtered continuously through these columns having beds of selected adsorbent. Accumulated filtered water was measured as total filtered water in terms of bed volume (100 mL). Arsenic concentrations at the filtered water were measured at regular interval after each 100 bed volume of filtration and each experiment was continued until the arsenic concentration in the filtered water reached to the breakthrough limit, which is 10 µg/L defined by the WHO as safe limit for human consumption. Figure 1 shows the relationship of accumulated water flows through the columns and corresponding arsenic concentrations in the filtered water for all the three columns using Skye sand. From the figure it is clear that all the three experiments provided identical patterns of diminishing arsenic removal capacity after a threshold treated water volume. It is to be noted that this sort of diminishing arsenic removal capacity after repetitive use of filter media is likely, mainly due to clogging of the filter media. The initial arsenic concentration in the feed water was 100 µg/L (ppb). Considering the high adsorption capacity of Skye sand and as with the low initial concentration of arsenic (100 µg/L), filtered water samples were having no trace of arsenic, the arsenic concentration in the feed water was increased to 500 µg/L after 400 bed volumes and further increased to 1000 µg/L after 1100 bed volumes. It is found that all the three samples started showing trace (1 µg/L) of arsenic after almost same bed volumes (1800–1900). The bed volumes of filtered water which could be achieved before the breakthrough concentration (10 µg/L) for columns 1, 2 and 3 were 3200 (320 litres), 3100 (310 litres) and 3300 (330 litres), respectively. The amounts of arsenic adsorbed in

column 1, 2 and 3 were 0.96, 1.04 and 0.92 mg of As (per g of sand) respectively at equilibrium concentration of 10 µg/L.

Figure 2 shows the performance of TGS sand columns in regard to arsenic removal capacities through the column beds. Like the case of Skye sand, all the three experiments provided identical patterns of diminishing arsenic removal capacities after a threshold treated water volume. The initial arsenic concentration in the feed water was 100 µg/L. The concentration was increased to 500 µg/L after 400 bed volumes and further increased to 1000 µg/L after 1100 bed volumes. The volumes of water filtered through the columns 1, 2 and 3 were 2700 bed volumes (270 litres), 2600 bed volumes (260 litres) and 2500 bed volumes (250 litres) respectively, until breakthrough of the WHO standard of 10 µg/L was reached. Amount of arsenic adsorbed in column 1, 2 and 3 were 0.77, 0.73 and 0.70 mg of As (per g of sand) respectively at equilibrium concentration of 10 µg/L. For all the three samples the threshold bed volume after which the arsenic concentration in the filtered water was traceable (1µg/L) after almost same bed volumes (1400–1500). These threshold bed volumes are lower than the threshold bed volumes for Skye sand. This lower threshold bed volume conforms with the lower bed volumes which can be achieved before the breakthrough level, which were also lower compared to the Skye sand. It is to be noted that in the batch experiments with the same samples as presented by Khan and Imteaz (2021), performance of TGS sand was also lower than the performance of Skye sand.

Figure 3 shows performance of Iron ore sand in regard to arsenic removal capacities through the column beds. The initial arsenic concentration in feed water was 100 µg/L. The concentration was increased to 500 µg/L after 800 bed volumes and to 1000 µg/L after 1400 bed volumes. All the three samples showed similar diminishing arsenic removal efficiency patterns. The volumes of water filtered through columns 1, 2 and 3 were 2000 bed volumes (200 litres), 2300 bed volumes (230 litres) and 2200 bed volumes (220 litres), respectively, until breakthrough of the WHO standard of 10 µg/L was reached. Amounts of arsenic adsorbed in columns 1, 2 and 3 were 0.23, 0.29 and 0.27 mg of As (per g of iron ore), respectively. Also, bed volumes at which arsenic concentration of 1µg/L found in the treated water were very close (1100–1300). These amounts are lower compared to the bed volumes at which arsenic presence in the filtered water were traceable for both the Skye and TGS sands. Also, bed volumes achieved before reaching to the breakthrough arsenic concentration were lower compared to both the Skye and TGS sands. This grading of arsenic removal capacities are similar to what were observed in the case of batch experiments presented by Khan and Imteaz (2021).

Figure 4 shows the performance of NT red sand columns in regard to arsenic removal efficiency. Again, almost similar diminishing trends of removal efficiencies were observed with the passage increase in filtered water flow (bed volume). The arsenic concentration in the feed water was 100 µg/L. Unlike all the previously mentioned samples, feed water arsenic concentration was not increased for the NT red sand, due to its lower arsenic removal capacity. The bed volumes at which arsenic concentration of 1µg/L found in the treated water were very close (300–310), which is much earlier compared to Skye sand, TGS sand and Iron ore sand. The volumes of water filtered through the columns 1, 2 and 3 were 390 bed volumes, 410 bed volumes and 400 bed volumes respectively, until breakthrough of the WHO standard of

10 µg/L was reached. Amounts of arsenic adsorbed in columns 1, 2 and 3 were 0.015, 0.016 and 0.016 mg of As (per g of sand), respectively. These amounts are much lower compared to all the previously mentioned amounts of arsenic adsorptions by Skye sand, TGS sand and Iron ore sand.

Figure 5 shows the performance of the scoria sand filter columns in regard to arsenic removal capacity. The arsenic concentration in feed water was 100 µg/L. The volume of water filtered through each column was 80 bed volumes (8 litres). However, none of the filters was able to remove the arsenic to a concentration below the WHO standard of 10 µg/L. The arsenic concentration in the filtrate was above 40 µg/L from the beginning in each filter. This is mainly due to the limited capacity of scoria sand in removing arsenic, which was also established by batch experiments as reported in Khan and Imteaz (2021).

In regard to the testing of the GFH filter columns, contaminated water with the same initial arsenic concentration (100 µg/L) was fed. Being a commercial grade sand considering the higher capacity of GFH, the arsenic concentration in the feed water was increased to 500 µg/L after 300 bed volumes. The performance of all the GFH sand columns were excellent in removing arsenic and residual arsenic concentration in the filtrate was always under the detection limit. However, all the filters clogged due to formation of a layer of fine particles in the filter bed, which occurred after 320 (column 1), 400 (column 2) and 430 (column 3) bed volumes. Similar phenomena were observed with the experiments with the IOCS and IOCS-AOCS columns. With IOCS columns, 100% arsenic removals were observed up to certain flows, after which all the columns were clogged due to formation of a layer of fine particles in the filter bed. Clogging occurred after 80 (column 1), 70 (column 2) and 50 (column 3) bed volumes. With the IOCS-AOCS (50%-50%) same behaviour was observed, i.e. excellent (100%) arsenic removal until the filter was clogged. In this case the clogging occurred earlier; after 25 (column 1), 40 (column 2) and 60 (column 3) bed volumes. Such clogging phenomena is a common nuisance for any filter media. Imteaz et al. (2016, 2019) have presented detailed modelling studies on such clogging for arsenic filtration.

Table 1
Column experiments with different adsorbents, Temperature 22° C, pH 7.0

Adsorbent in the column	Volume treated at WHO standard (Bed volume)	Average arsenic removal capacity at WHO standard (mg of As/ per g of sand)
Skye sand	3100 ~ 3300	0.97 ± 0.05
TGS sand	2500 ~ 2700	0.73 ± 0.04
Iron ore sand	2000 ~ 2300	0.26 ± 0.03
NT red sand	360 ~ 390	0.016 ± 0.010
Scoria sand	N/A	N/A

Discussions

The results presented in the earlier section are summarised in Table 1, where studied adsorbents are graded as per arsenic removal capacity from highest to lowest. From Table 1 it is clear that Skye sand has got the highest arsenic removal capacity. Among the tested samples, the removing capacities from highest to lowest are Skye, TGS, Iron ore, NT red and Scoria sands. Scoria sand was never able to remove arsenic to the WHO standard level. It is to be noted that the batch experimental results with the same samples showed the same grading of arsenic removal efficiencies, except for GFH sand. Also, it is found that the performances of Scoria and NT red sands are much inferior compared to Skye, TGS and Iron ore sands. Similar trend on the differences of performances were also observed with the batch experiments. Although, GFH, IOCS and IOCS-AOCS filters were having very good arsenic removal capacities, they were excluded from the comparison and grading as all the three columns with each of the samples were clogged after repeated operations. GFH, IOCS and IOCS-AOCS filters failed after accumulated flows of 320 ~ 430 bed volumes, 50 ~ 80 bed volumes and 25 ~ 60 bed volumes respectively. It is to be noted that in the batch experiments IOCS and IOCS-AOCS (50–50) showed superior arsenic removal capacities than Skye sand (Khan and Imteaz, 2020). Also, GFH sand showed second highest (slightly below the Skye sand) arsenic removing capacity (Khan and Imteaz, 2021).

Conclusions

Numerous materials were investigated for the assessment of arsenic removal capacity through adsorption. Most of the studies were based on batch experiments, outcomes of which may not be same in the real-life scenario. To overcome this drawback and to ascertain more reliable arsenic removal capacity through certain material, column experiments are performed. This paper presented arsenic removal efficiencies through column experiments using several natural and synthetic adsorbents. Five locally available natural sand samples and one commercial sand sample (GFH) were selected for further assessment following on their earlier batch experimental results with the same samples. Also, two synthetic samples; IOCS and mix of IOCS-AOCS, which were found to be very effective in removing arsenic were selected. These materials were selected for further assessment with the view of their practical application. Each sample was tested with three separate columns having bed material of the same sample. It is found that for all the samples results of three separate columns were similar and were following the same patterns confirming the representativeness of results for each sample.

From the column experimental results, it is found that Scoria sand is not able to treat arsenic to the maximum level in drinking water set by WHO. Among other tested samples, it is found that the Skye sand is having highest arsenic removal capacity, followed by TGS sand, Iron ore sand and NT red sand. At WHO standard, average arsenic removal capacities (per g of sand) of these samples are: 0.97 ± 0.05 mg for Skye sand, 0.73 ± 0.04 mg for TGS sand, 0.26 ± 0.03 mg for Iron ore sand and 0.016 ± 0.010 mg for NT red sand. Among the other tested samples, GFH sand and the two synthetic sands (IOCS and mix of IOCS-AOCS) showed very good arsenic removal efficiencies for some periods (GFH sand: 320 ~ 430 bed volumes, IOCS: 50 ~ 80 bed volumes and IOCS-AOCS: 25 ~ 60 bed volumes). After which all these column filters failed due to clogging of the filter beds through accumulation of fine particles in the filters. It is to be noted that in the real-life such filters are often backwashed to remove such clogging. Nonetheless,

usually through such backwashing the filters do not come back to their original state and clogging increases with the passage of time (i.e. flow through the filter). Mentioned findings warrant testing with such columns and/or real size prototype, as only batch experiments do not reveal these sorts of practical implications. Another inference which can be made through above findings is that artificially coated sands may render high effectiveness in removing arsenic, however are susceptible to earlier clogging making those not suitable for real life application.

Declarations

Ethics approval and consent to participate: The study did not involve any human or animal

Consent for publication: No data or material used from third person or somewhere else

Availability of data and material: All data generated or analysed during this study are included in this published article

Competing interests: There is no competing interest to be reported

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Figures

Figure 1

Arsenic removal efficiencies with time with Skye sand column filter (Feed water arsenic concentration: 100 µg/L for 0-400 BV, 500 µg/L for 400-1000 BV, 1000 µg/L for ≥ 1100 BV)

Figure 2

Arsenic removal efficiencies with time with TGS sand column filter (Feed water arsenic concentration: 100 µg/L for 0-400 BV, 500 µg/L for 400-1000 BV, 1000 µg/L for ≥1100 BV)

Figure 3

Arsenic removal efficiencies with time with Iron ore sand column filter (Feed water arsenic concentration: 100 µg/L for 0-800 BV, 500 µg/L for 800-1400 BV, 1000 µg/L for >1400 BV)

Figure 4

Arsenic removal efficiencies with time with NT red sand column filter (Feed water arsenic concentration: 100 µg/L)

Figure 5

Arsenic removal efficiencies with time with Scoria sand column filter (Feed water arsenic concentration: 100 µg/L)