

Adsorbent-based point-of-use system for arsenic removal in rural areas

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ABSTRACT

Arsenic in groundwater is recognised as a threat to public health worldwide and specifically in rural areas of several developing countries (e.g. Bangladesh). Different treatment technologies are available or under investigation that, when properly operated, can reduce arsenic concentrations to a sufficiently low level. These technologies are inappropriate for rural communities in developing countries where arsenic-related problems are most pronounced, and where centralised water treatment facilities are frequently absent. Arsenic-related health hazards are associated with the use of arsenic-contaminated water for drinking and food preparation, corresponding to only 2–4 litres per capita per day. Arsenic removal in rural areas, where centralised water treatment and distribution facilities are unavailable, should consequently be carried out at household level and the system applied should be simple and affordable. Arsenic adsorption capacities of different virgin and iron-coated materials were first examined in batch adsorption experiments. Both iron-coated sand (ICS) and iron-impregnated granular activated carbon (IGAC) show high As(III) and As(V) adsorption capacities that decrease at high pH values. Three simple point-of-use systems, namely, a 'tea bag', a 'coffee filter' and a simple 'family filter', all with iron-coated sand or iron-impregnated granular activated carbon were tested. The 'tea bag' system was not successful because too long an adsorption time was required. The 'coffee filter' performed better, however, results obtained with the simple 'family filter' were most promising. The simple 'family filter' containing 2 litres of IGAC is expected to supply a family in rural Bangladesh with arsenic-free water for drinking and cooking for, on average, 18 months without adsorbent replacement.

Key words | adsorption, arsenic removal, iron-coated sand, iron-impregnated granular activated carbon, point-of-use systems

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INTRODUCTION

Major arsenic contamination of groundwater has been found in Bangladesh, India, Taiwan, Mongolia, several countries in South, Central and North America, New Zealand, Hungary, the Baltic States and many other countries (BGS 1999; Bruton-Ward 2000; Hering & Chu 2000). Arsenic is known as an extremely toxic compound (Pontius *et al.* 1994). The provisional guideline of WHO is 10 µg/l, and the estimated excess lifetime skin cancer risk associated with exposure to this concentration is 6×10^{-4} (WHO 1996).

There are several effective water treatment techniques such as coagulation-sedimentation-filtration, nanofiltration, reverse osmosis, etc. (Shen 1973; Fox 1989; Scott *et al.* 1995; McNeil & Edwards 1997; Jekel & Seith 1999; Madiec *et al.* 1999) that, under properly optimised process conditions, can reduce arsenic concentrations below the permissible level in drinking water (WHO and EC: 10 µg/l; USA, Bangladesh, India and some other countries: 50 µg/l) (EC 1998). In addition, the effectiveness of several other advanced treatment techniques is currently under

investigation (e.g. fluidised-bed sand reactor, subsurface groundwater treatment, etc., see Rott & Friedle 1999; Stamer & Nielsen 1999). These techniques, however, are inappropriate for rural communities in developing countries where arsenic-related problems are most pronounced, and where centralised water treatment facilities are frequently absent.

The arsenic adsorption capacity of activated alumina, activated bauxite, conditioned zeolites, granular ferric hydroxide, iron-oxide coated sand (Fe/Cu impregnated), granular activated carbon, hematite, iron-manganese ore, and many other materials have been investigated in various studies (Gupta & Chen 1978; Huang & Fu 1984; Fox 1989; Joshi & Chaudhuri 1996; Driehaus *et al.* 1998; Simeonova 1999; Reed *et al.* 2000). Materials with high metal (Fe, Al, Cu) content or metal-coated materials demonstrated the highest arsenic adsorption potential. Solution pH and arsenic speciation were reported to have a strong effect on adsorption efficiency (Gupta & Chen 1978; Huang & Fu 1984; Fox 1989; Reed *et al.* 2000).

Arsenic-related health hazards are associated only with the use of arsenic-contaminated water for drinking and food preparation. For these purposes only ~2–4 litres of water per capita per day are used. This strongly suggests that in rural areas where central water supply systems are absent, it is feasible to remove arsenic only from the water used for drinking and food preparation. Arsenic removal should consequently be done at the household level and the applied point-of-use (POU) system(s) should be appropriate for conditions prevailing in rural communities of developing countries.

The objective of the study was to assess the applicability of different materials as adsorbents for arsenic removal in POU systems suitable at the household level in rural areas. The effectiveness and suitability of three types of POU treatment systems, namely, a 'tea bag', a 'coffee filter' and a simple 'family filter' were investigated. As a preliminary step the adsorption capacities of virgin (olivine, basalt and sand) and iron-coated materials (iron-coated sand and iron-impregnated granular activated carbon) were assessed. The effect of arsenic speciation (As[III] versus As[V]), pH, contact time, adsorbent dosage and initial arsenic concentration on the arsenic removal efficiency of selected adsorbents was then evaluated in

a series of batch adsorption experiments and/or filter column runs.

MATERIALS AND METHODS

Batch adsorption experiments

Laboratory scale batch adsorption equilibrium experiments were conducted to assess the arsenic removal potential of different adsorbents and to study the effect of pH, initial arsenic concentration and adsorbent dosage on arsenic removal efficiency. A fixed amount of adsorbent was put into 0.56 litre capacity polyethylene bottles. The bottles were filled with the model water, capped and mixed continuously with the mechanical shaker at a room temperature of 25°C. Before arsenic analysis, samples were filtered through a 0.2 µm pore size cellulose acetate membrane filter to separate finely dispersed adsorbent particles.

Point-of-use experimental systems

The arsenic removal potential of three very simple systems, i.e. a 'tea-bag', a 'coffee filter' and a simple 'family filter', all based on selected adsorbents, was examined in a series of laboratory experiments. These systems should be used as POU systems and should be suitable for use in rural communities of developing countries (e.g. Bangladesh).

The 'tea-bag' POU system comprised a small porous paper bag, similar to tea bags, filled with 16 g of selected adsorbent (iron-coated sand, ICS, or iron-impregnated granular activated carbon, IGAC). The bag was placed in a 2-litre capacity polyethylene bottle filled with model water. The arsenic concentration in the solution was measured at regular time intervals. The 'coffee filter', a simple and easy to use system at the household level, consisted of a PVC filter holder with a standard coffee paper filter containing 30 ml of adsorbent (~40 g ICS or 15 g IGAC). Five litres of model water (400 µg As[V]/l) were double-filtered through the filter filled with adsorbent, and the arsenic concentration in the filtrate was

Table 1 | Composition of model water

Parameter	O ₂	Ca	Mg	Fe	Mn	SO ₄	Cl	Na	PO ₄
mg/l	10.5	53	7.9	0.03	0.005	81	125	134	0.03

analysed. Applied experimental conditions allowed very intensive contact between the model water and the adsorbent, however contact time was restricted to ~1 minute.

The simple ‘family filter’ consisted of two parallel operating PVC filter columns, 69 mm in diameter and 500 mm high, with associated flow control valves, containers for preparation and storage of model water, and filtrate storage vessels. In the filters 300 ml or 450 ml of adsorbent (ICS or IGAC) were placed above 250 ml of coarse sand. The filters were operated at a constant filtration rate of 0.23 m/hr thus allowing adsorption times of 21 or 33 minutes. The capacity of the system (approx. 20 l/day) is sufficient to cover the daily water consumption (drinking and cooking) of a typical family in rural Bangladesh.

Model water

Due to the relevance of the study specifically for Bangladesh, model water was used that mirrored the typical groundwater composition in this country, characterised by high bicarbonate and carbon dioxide concentrations, and neutral to slightly acidic pH. Model water was prepared from tap water by increasing the HCO₃⁻ concentration to 275 mg/l, decreasing the pH to 6.8 and consequently increasing the carbon dioxide concentration to 80 mg/l. Concentrations of other model water quality parameters are given in Table 1. An average arsenic concentration in groundwater from contaminated wells in Bangladesh is 180 µg/l, and both As(III) and As(V) are present (BGS 1999). In the majority of experiments, however, model water with a much higher initial arsenic concentration (~500 µg/l in both As(III) and As(V) form) was prepared. Consequently, the arsenic range in more than 94% of wells in Bangladesh was covered (<6% of

contaminated wells in Bangladesh have >500 µg As/l; BGS 1999). A few experiments were also carried out with model water with arsenic concentrations corresponding to low (100 µg/l) and maximum (1,600 µg/l) concentrations found in Bangladesh (BGS 1999). The primary standard of 10 mg/l for As(V) was prepared from the standard 1000 mg/l stock solution (Merck). The primary standard for As(III) was prepared by dissolving arsenic trioxide in 1 M NaOH and diluting the solution with de-mineralised water.

Adsorbents

Different types of virgin materials, iron-coated sands and iron-impregnated granular activated carbons were used in preliminary batch adsorption experiments designed to assess their arsenic adsorption capacity and to select suitable adsorbents for further experiments. Olivine (0.56–0.8 mm), basalt (1.1–1.4 mm) and quartz sand (0.8–1.25 mm) were used as representatives of virgin materials. Seven types of iron-coated sands (ICS-1 to ICS-7) were prepared in the laboratory: ICS-1 was prepared according to the procedure proposed by Benjamin *et al.* (1996). Iron-coated sands ICS-2 to ICS-7 were prepared in the laboratory by conducting filtration runs at different pH and feed water iron concentrations (Sharma *et al.* 2000). Two types of iron-coated sand (ICS-8 and ICS-9) were obtained from groundwater treatment plants Noord Bargeres and Gilze, respectively, both in the Netherlands. Two types of iron-impregnated granular activated carbons (IGAC-1 and IGAC- 2) were made available by Norit B.V.

Arsenic analysis

An atomic absorption spectrometer (Perkin Elmer 1100 B), equipped with graphite furnace (GF) and HGA 300

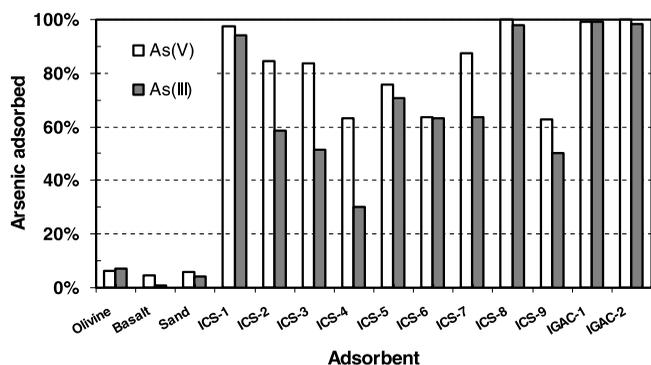


Figure 1 | Adsorption of arsenic onto different adsorbents; arsenic concentration in model water 470 $\mu\text{g/l}$; adsorbent dosage 8 g/l; pH 6.8; adsorption time 8 hours under continuous shaking.

programmer, was used for arsenic analysis. The arsenic analyses were carried out according to the Dutch Standard Method NEN 6457, with an arsenic detection limit of $\sim 3 \mu\text{g/l}$ and an accuracy of $\pm 1 \mu\text{g/l}$.

RESULTS AND DISCUSSION

Arsenic removal potential of different adsorbents

In batch experiments at a dosage of 8 g/l, all investigated iron-coated materials demonstrated high to very high arsenic removal efficiency for both As(III) and As(V) (Figure 1). Different removal efficiencies were, however, achieved with different ICS types (50–100%). In general higher removal efficiencies of As(V) were observed in comparison to As(III). A dosage of 8 g/l of ICS-1, ICS-8 and both types of IGAC was sufficient to reduce both As(III) and As(V) concentrations close to or below 10 $\mu\text{g/l}$ within 8 hours. In contrast the virgin materials tested (olivine, basalt and quartz sand) demonstrated very low arsenic adsorption capacity (<10%).

Results obtained in this set of adsorption batch experiments strongly suggest that the arsenic adsorption potential of the materials tested could be exclusively attributed to the adsorptive properties of iron coating present on the surface of the materials.

ICS-8 and IGAC-2 (further on referred to as ICS and IGAC respectively) were selected for further experiments due to their high adsorption capacities. The characteristics of these adsorbents are given in Table 2.

Effect of contact time, initial arsenic concentration and applied adsorbent dose

In the batch experiments conducted, a relatively long adsorption time of between 24 and 48 hours was, under the conditions applied, required to achieve complete removal of As(III) or As(V) (Figure 2). During the initial 8 hours very efficient removal ($\geq 85\%$) was achieved with both adsorbents. Complete removal of As(V) was achieved with ICS within 24 hours, while approximately 48 hours were required to remove As(III) completely with both ICS and IGAC as well as As(V) with IGAC. Such long adsorption times required could be prohibitive for application of adsorbents tested for arsenic removal with POU systems at the household level.

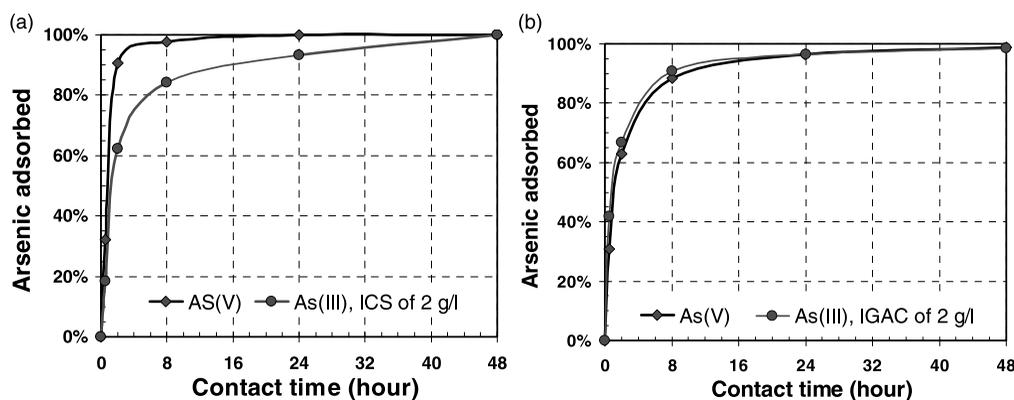
Arsenic adsorption kinetics are related to initial arsenic concentration (Figure 3). Results obtained demonstrate that even very high initial arsenic concentrations in the model water (1,600 $\mu\text{g/l}$, corresponding to the highest arsenic level observed in groundwater in Bangladesh), can be efficiently removed by applying somewhat higher ICS or IGAC dosages (8 g/l), coupled with sufficient contact time. Similarly Figure 4 demonstrates that relatively low IGAC or ICS dosages (0.4–2 g/l) are sufficient to remove arsenic below WHO guideline values from groundwater with relatively lower arsenic concentrations (53% of wells in Bangladesh have arsenic concentrations between 50 and 100 $\mu\text{g/l}$).

Effect of pH

As(V) removal efficiency of both ICS and IGAC decreases with an increase in pH. Decrease of As(V) removal efficiency for both adsorbents tested was pronounced at pH values > 7.5 (see Figure 5). At the same time, As(III) removal efficiency of both ICS and IGAC was only slightly affected by an increase in pH.

Table 2 | Characteristics of selected adsorbents

Adsorbent	Size (mm)	Fe content (mg/g)	Apparent density (g/l)	Pore volume (ml/g)	Remarks
ICS-8	1.8–4.0	330	1,358		9.3 years in use
IGAC-2	~1.07 (d_{50})	81	500	0.732	Norit code: HD 4000 Fe

**Figure 2** | Arsenic removal in batch system as a function of contact time; model water with $500 \mu\text{g As}^{3+}/\text{As}^{5+}/\text{l}$; adsorbent: (a) ICS, (b) IGAC, adsorbent dosage of 2 g/l; pH=6.8, continuous shaking.

As(V) or As(III) adsorption on hydrous ferric oxides takes place through formation of surface complexes. In general, cation adsorption increases with an increase in pH, while anion adsorption is highest at low pH and decreases gradually as pH increases. Within the range of tested pH, As(V) species (H_2AsO_4^- , HAsO_4^{2-}) are anionic and hence the trend observed. In addition, the percentage of HAsO_4^{2-} , a divalent As(V) species, increases with an increase in pH (Gupta & Chen 1978). The point of zero charge of hydrous iron oxide is around pH 7.8–8.2 (Dzombak & Morel 1990). Consequently, anion adsorption on hydrous ferric oxides is highest at low pH and decreases gradually as pH increases. At pH > 7.8 adsorption of As(V) species takes place against repulsive electrostatic forces, resulting in a strongly reduced adsorption efficiency.

As(III) species up to a pH of approximately 7.0 are exclusively non-ionic (H_3AsO_3^0). As such their adsorption onto hydrous ferric oxides was not strongly affected by a

change in pH. A further increase in pH up to 7.8 resulted in a slight increase in adsorption (10%), probably due to the formation of anionic As(III) species (H_2AsO_3^-) and related attractive electrostatic forces with the positively charged hydrous iron surface of ICS and IGAC. At higher pH, above 7.8, anionic As(III) species are formed (H_2AsO_3^- , HAsO_3^{2-}) and at the same time hydrous iron oxide has a negative charge resulting in repulsive electrostatic forces and a related decrease of As(III) removal efficiency.

At pH of 6.8, a common value for groundwater in Bangladesh, both ICS and IGAC are very efficient in adsorption of both As(V) and As(III). ICS adsorbs As(V) more efficiently while IGAC is more efficient in As(III) removal. Aeration of groundwater, commonly applied in groundwater treatment, should, however, be minimised prior to adsorptive arsenic removal with ICS or IGAC given the increase in pH associated with aeration and the resulting negative effect on arsenic adsorption efficiency.

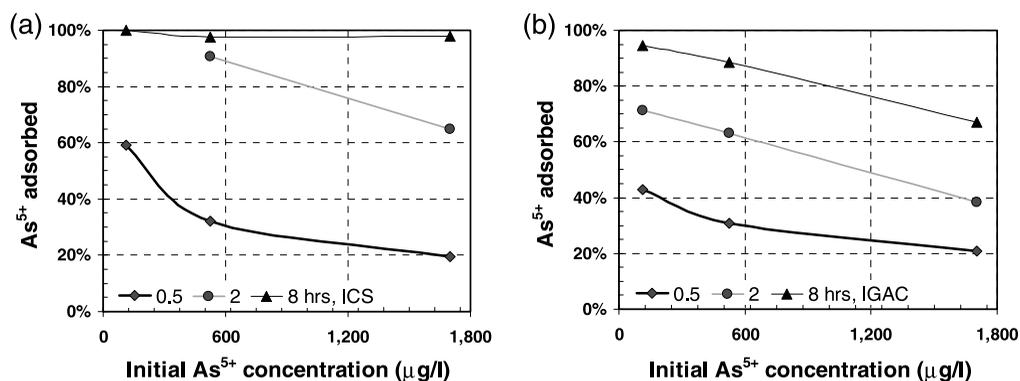


Figure 3 | Effect of initial arsenic concentration and contact time on arsenic removal; adsorbent: (a) ICS, (b) IGAC, adsorbent dosage 2 g/l, pH=6.8.

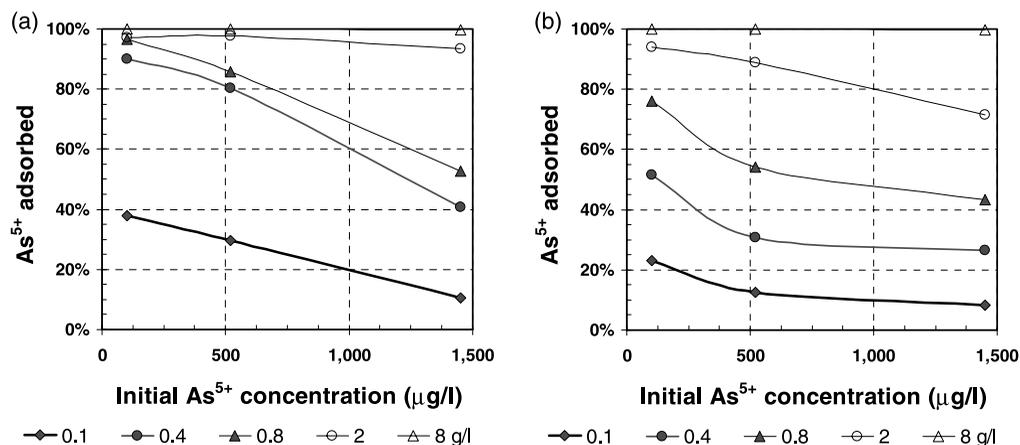


Figure 4 | Effect of initial arsenic concentration and adsorbent dosage on As(V) removal; adsorbent: (a) ICS, (b) IGAC, contact time 2 hours; pH=6.8.

Adsorption isotherm

As(III) and As(V) adsorption on both ICS and IGAC can be well characterised by the Freundlich isotherm (Figure 6). Both adsorbents demonstrated high arsenic adsorption capacity. At an equilibrium arsenic concentration of 10 µg/l, the arsenic adsorption capacities of ICS and IGAC were 2.4 mg As/g adsorbent and 0.7 mg As/g adsorbent respectively. ICS demonstrated higher arsenic adsorption capacity per unit weight in comparison with IGAC, probably due to a higher percentage of iron (330 and 81 mg/g for ICS and IGAC respectively). Both adsorbents showed higher As(III) than As(V) adsorption capacity, in contrast to results obtained in other batch experiments, characterised by relatively short adsorption

times. Prolonged adsorption time (12 days) and oxygen-rich water resulted in (partial) oxidation and transformation of As(III) to As(V) that probably caused better utilisation of adsorption sites available for both As(III) and As(V).

'Tea bag'

The 'tea bag' POU system, under the conditions tested (16 g of ICS or IGAC in 2 litres model water without shaking), demonstrated very limited arsenic removal capacity within 8 hours, which is the adsorption time considered acceptable for practical reasons (i.e. adsorption to take place during the night hours). Much longer

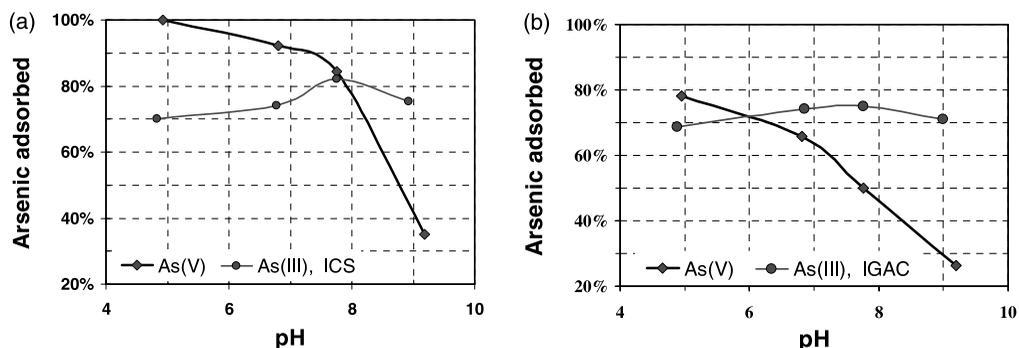


Figure 5 | Effect of pH on arsenic removal efficiency; initial arsenic concentration ~500 µg/l; adsorbent dosage 1 g/l; contact time 8 hours.

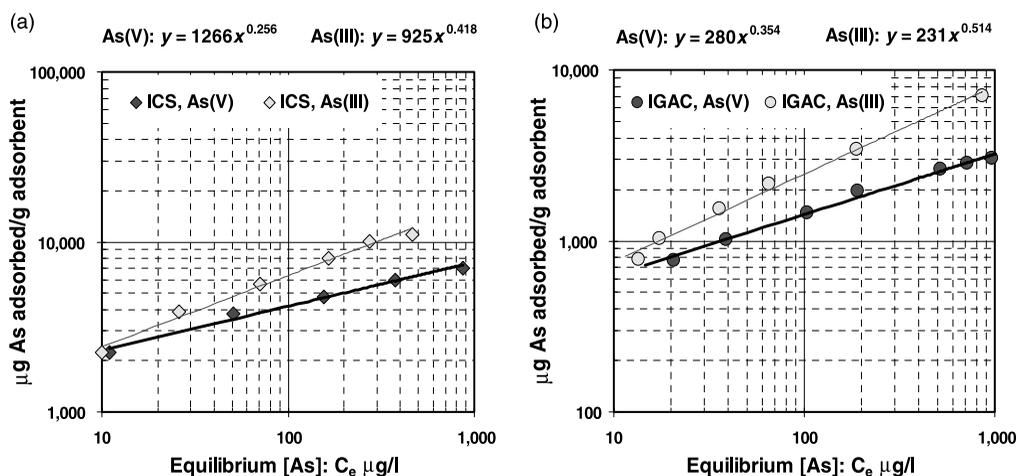


Figure 6 | Freundlich adsorption isotherm for arsenic adsorption onto (a) ICS and (b) IGAC; initial arsenic concentration 1,600 µg/l; adsorption time 12 days; pH=7.2.

contact time (>9 days) was needed to achieve an acceptable arsenic removal efficiency (Figure 7a). Such a long contact time is, however, not acceptable.

Arsenic adsorption kinetics can be enhanced if continuous mixing is introduced. Under the condition tested, the 'tea bag' POU system with continuous mixing achieved almost complete (>99%) arsenic removal within 8 hours (Figure 7b). However continuous shaking is not practicable under conditions that prevail in rural Bangladesh. Comparison of the results obtained in experiments with and without shaking strongly suggests that (external) mass transfer plays a critical role in adsorptive arsenic removal.

In summary, the 'tea bag' POU system with ICS or IGAC is not suitable for arsenic removal because of the

need for either a very long contact time or continuous shaking.

'Coffee filter'

The 'coffee filter' POU system equipped with either ICS or IGAC demonstrated very high arsenic removal potential even under non-optimised conditions (e.g. a very short contact time). An initial arsenic concentration of 400 µg/l was, after the second filtration step, reduced to 168 µg/l and 62 µg/l for ICS and IGAC respectively. The greater efficiency of the system with IGAC can be explained by the difference in pore size distribution of IGAC and ICS. IGAC is characterised by larger pores that are likely to

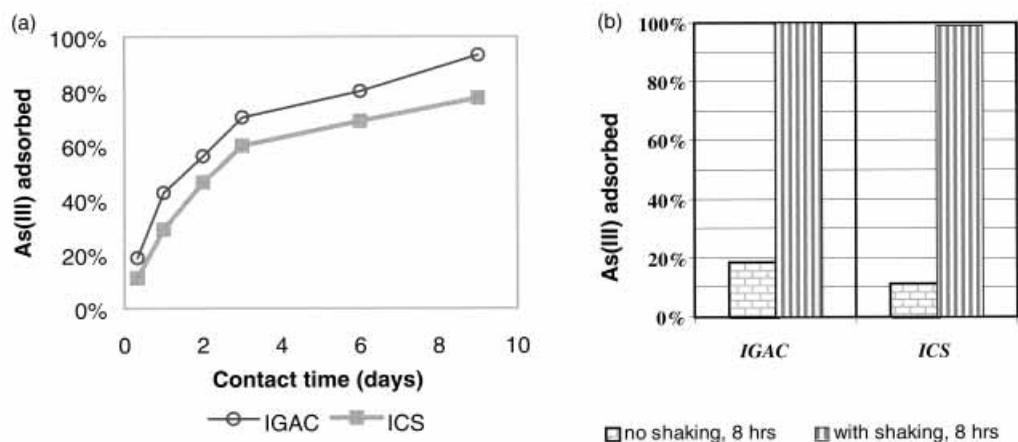


Figure 7 | Effect of (a) contact time and (b) mixing conditions on arsenic removal with 'tea bag' POU system; initial As(III) concentration 560 $\mu\text{g/l}$, adsorbent 8 g/l.

allow much faster pore diffusion and subsequent arsenic adsorption. Arsenic concentrations in the filtrate were, however, still above WHO and Bangladesh standards. In order to produce a successful system the contact time should be much longer and short-circuiting should be excluded. Alteration of adsorbent properties (e.g. pore size distribution), that will allow more efficient and faster pore diffusion and adsorption, can further enhance the arsenic adsorption kinetics and improve the removal efficiency of the system.

Filtration POU system

A simple 'family filter' equipped with either ICS or IGAC demonstrated very high removal potentials for both As(III) and As(V). The filter equipped with IGAC had noticeably higher As(V) removal efficiency (Figure 8) under the non-optimised conditions applied (high initial arsenic concentration of $\sim 500 \mu\text{g/l}$, filtration rate of 0.23 m/h and ICS/IGAC filter bed volume of 300 ml corresponding to an adsorption time of approximately 21 minutes). The arsenic concentration in the filtrate exceeded the WHO guideline level of $10 \mu\text{g/l}$ after 80 and 1,300 filter bed volumes were filtered with filters equipped with ICS and IGAC respectively.

A slow increase in the arsenic concentration in the filtrate after the WHO guideline level was reached,

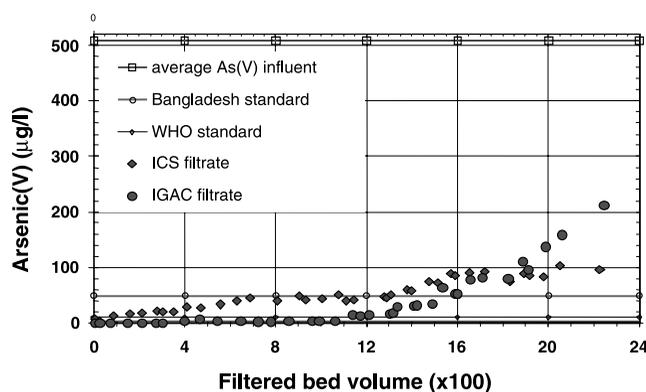


Figure 8 | As(V) concentration in the filtration POU system with ICS or IGAC; filtration rate 0.23 m/h; empty bed contact time 21 minutes; feed water pH=6.8.

strongly suggests that the arsenic adsorption capacity of both adsorbents tested was not exhausted, and that the performance of the system can be further improved by optimising filter design parameters.

Additional runs with filters that allow 50% longer adsorption time (50% increase in adsorbent volume) demonstrated that considerably higher filtrate volumes, with arsenic concentrations below $10 \mu\text{g/l}$, can be produced (2,000 and 750 empty bed volumes in the case of IGAC and ICS respectively).

Further optimisation experiments with the filtration POU system are needed and should result in more efficient

utilisation of the adsorption capacities of ICS and IGAC. In particular, improved filter design (e.g. adsorbent volume, contact time, filtration rate, etc.) is likely to result in better performance of the system equipped with ICS, which could be obtained from existing groundwater treatment facilities or produced locally.

Applications of the filtration POU system

Preliminary experiments performed with a non-optimised filtration POU system with IGAC or ICS, demonstrated the very high arsenic removal potential of this very simple and easily affordable treatment system. The system does not require electricity and, with the exception of adsorbent, could be produced locally. Extrapolation of existing results suggests that the POU system containing 2 litres of IGAC can supply a family in rural Bangladesh with arsenic-free water for drinking and cooking for an average of 18 months (assuming an average raw water arsenic concentration of $\sim 200 \mu\text{g/l}$). Exhausted adsorbent can be easily replaced. Further laboratory and field tests are required to establish a robust design of the unit. Additionally, the effect of other groundwater components (e.g. iron) on the performance of such a unit, and the operational and maintenance requirements (e.g. periodic cleaning of the unit) should be examined.

CONCLUSIONS

- Iron-coated materials tested, i.e. iron-coated sand (ICS) and iron-impregnated granular activated carbon (IGAC), demonstrated high As(III) and As(V) adsorption capacities. At an equilibrium arsenic concentration of $10 \mu\text{g/l}$ arsenic adsorption capacities of ICS and IGAC were 2.4 and 0.7 mg As per gram adsorbent respectively. ICS and IGAC arsenic adsorption potentials can be mainly attributed to the adsorptive properties of hydrous ferric oxides on their surfaces.
- In batch experiments ICS and IGAC arsenic adsorption efficiencies increased not only with an

increase in adsorbent dosage (0.1–8 g/l), but also with an increase in adsorption time, as well as with a decrease in pH and/or initial arsenic concentration.

- As(V) removal efficiencies of both ICS and IGAC are significantly reduced with an increase in pH above ~ 7.5 . In contrast As(III) removal efficiencies are only marginally affected by solution pH. Under pH values common for groundwater in Bangladesh (± 6.8), high arsenic removal efficiencies with both ICS and IGAC can be expected.
- The tested ‘tea bag’ system is not suitable because of the requirement for a very long contact time or continuous shaking. The ‘coffee filter’ system performs better, however, the simple ‘family filter’ is most promising. The simple ‘family filter’ containing 2 litres of IGAC is expected to supply a family in rural Bangladesh with arsenic-free water for drinking and cooking for 18 months on average.

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