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Iron hydroxide-coated sand filter for household drinking water from roof-harvested rainwater

M. Mansoor Ahammed and V. Meera

ABSTRACT

This paper reports the performance of iron hydroxide-coated sand medium in removing bacteria and heavy metals from roof-harvested rainwater. In long-duration column tests (15 cm × 2.5 cm ID columns; empty bed contact time 15 min) employing roof-harvested rainwater, total coliform and faecal coliform removal ranged from 97% to >99%, and effluent turbidity remained below 2.0 NTU up to 500 bed volumes (BV). In comparison, bacterial removal by uncoated sand medium was always below 21%. Further column tests duplicating the use pattern of household water purification devices (daily input of 10 BV) indicated that intermittent use of the medium did not affect its performance, with bacterial removal remaining in the same range of 97–99% for 500 BV (50 d). Long-duration column tests conducted using a roof runoff (galvanised iron roof) with elevated levels of heavy metals, lead and zinc, showed that the medium is capable of effectively removing bacteria and heavy metals simultaneously for 600 BV, with effluent lead and zinc levels below the WHO guideline values for drinking water. Influent pH did not affect the bacterial removal in the pH range studied (pH 6.0–9.0). No leaching of iron from the sand was noticed for this pH range. The study showed the potential of iron hydroxide-coated sand as a sorptive filter medium for use in simple home water purification devices for treatment of roof-harvested rainwater which is generally contaminated by microorganisms and heavy metals.

Key words | bacteria removal, coated sand, rainwater harvesting, water purification, water quality

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INTRODUCTION

Water-borne diseases are an important public health problem in both developed and developing countries. Consumption of unsafe water continues to be one of the major causes of 2.2 million diarrheal disease deaths occurring annually, mostly in children in developing countries (WHO/UNICEF 2000). In developing countries, people drinking unsafe water include those living in rural as well as urban areas. One approach to ‘safe water for all’ is household water treatment and safe storage, which is promoted globally by the WHO to reduce the global burden of water-borne diseases (Sobsey 2006). There is considerable evidence that simple, low-cost methods at the household level are capable of drastically improving the microbial quality of household stored water and reducing the risks of diarrheal diseases in the population of all ages in developed and developing countries.

In this context, domestic rooftop rainwater harvesting is receiving increased attention worldwide as an alternative source of drinking water. Traditionally, collection and storage of rainwater from individual household roof catchments is the major option to the people in water-scarce regions in rural areas of developing countries. Due to ubiquitous contamination of surface and groundwater resources by microbial and chemical contaminants, rainwater harvesting has become more relevant now even in areas which enjoy high rainfall. While rainwater is generally considered as nonpolluted, or at least not significantly polluted, contamination occurs when it falls on the roof,

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collects dirt, dissolves some heavy metals in the case of metal surfaces, and then flows into storage. Many studies have shown that roof-harvested rainwater is heavily contaminated microbiologically by a variety of indicator and pathogenic organisms unless special care is taken during collection and storage of water (Yaziz *et al.* 1989; Pinfold *et al.* 1993; Appan 1997; Simmons *et al.* 2001; Lye 2002). Elevated levels of toxic metals are also reported in roof-collected rainwater (Förster 1996; Ghanayem 2001; Chang *et al.* 2004). This shows the need for proper treatment of rainwater if it is to be used for drinking. Boiling and disinfection have been suggested as possible methods for this. Application of these methods, however, is often limited in rural households due to practical difficulties. This indicates the need for developing simple, easy to use household methods to remove various contaminants in rainwater.

Sand filtration is a commonly used method for removing particles from water. This process may not be effective for removal of microorganisms and relies either on addition of coagulants to the influent water or the development of biofilm on the surface of sand media. On the other hand, metallic hydroxides have been used in water treatment for many years and are known to be good adsorbents of bacteria and viruses. This has led to the development of metal hydroxide-coated sand as a filtration/adsorption medium in water treatment. Sand coated with iron and aluminium hydroxides has shown to be a good filter media for removal of bacteria, viruses and turbidity (Ahammed & Chaudhuri 1996; Lukasik *et al.* 1999) and heavy metals (Bailey *et al.* 1992; Theis *et al.* 1992; Liu *et al.* 2005) from water. The potential of metal hydroxide-coated sand has been evaluated using sewage-spiked tap water (Lukasik *et al.* 1999) and surface water with high turbidity (Ahammed & Chaudhuri 1996). Since the characteristics of rainwater are different from other natural waters, tests need to be conducted to assess the suitability of this medium for treatment of roof-harvested rainwater. No studies have been reported in the literature on the suitability of this medium for treatment of roof-harvested rainwater. Thus the objective of the present study was to assess the effectiveness of iron hydroxide-coated sand as a filtration/adsorption medium for removal of various contaminants in rainwater so that it can be incorporated into simple household water purification devices for use in developing countries. The contaminants studied were bacteria, lead and zinc. Bacteria removal was evaluated using total coliforms, faecal coliforms, and heterotrophic plate counts (HPC). Short- and long-duration column tests were conducted using rainwater collected from different roof surfaces with varying characteristics.

MATERIALS AND METHODS

Coated sand medium

The medium was prepared using river sand, passing through an 800 μm sieve and retained on a 300 μm sieve. The sand was washed and dried at 105°C before the hydroxide coating was applied. Several methods are available in the literature for coating granular media with iron oxide/ hydroxide (Anderson *et al.* 1982; Bailey *et al.* 1992; Lukasik *et al.* 1996; Liu *et al.* 2005). In the present study the method suggested by Lukasik *et al.* (1996) was employed for coating sand with iron hydroxide. A 1,000 g batch of sand was rinsed with deionised water until the supernatant was clear, air dried, and then spread out to form a 3-cm thick layer in a plastic tray. A volume of 1,000 mL of 1.0 M ferric chloride was added to the tray. After 30 min of contact the excess solution was poured off and the sand was allowed to dry for 24 h. The dried sand was transferred slowly into a 2-litre glass beaker containing 1,000 mL of 3.0 M ammonium hydroxide and was allowed to soak for 10 min. The excess liquid was poured off, and the sand was air dried in the plastic tray with occasional stirring. The dried sand was rinsed vigorously with deionised water to remove loose precipitates, then air dried in the plastic tray. The coated sand was stored in plastic bottles at room temperature until used.

Rainwater samples

Rainwater collected from two different roofs (cement concrete roof and galvanised iron (GI) roof) with varying characteristics was used in different tests. The roofs were located in a residential area adjacent (about 300 m) to a busy highway. No 'first-flush' device was attached to the roofs. Roof runoff was collected for a complete rain event. The physico-chemical and microbiological characteristics of the collected rainwater are presented in Table 1. The water was stored in the dark at 4°C in plastic bottles. The required

Table 1 | Quality of roof–harvested rainwater

Parameter	Cement concrete roof	Galvanised iron (GI) roof
pH	6.5	6.9
Turbidity, NTU	8.2	6.5
Conductivity, μS/cm	96	128
Alkalinity, mg/l as $CaCO_3$	14	18
Total coliforms, MPN/100 mL	1,700 ± 420	300 ± 65
Faecal coliforms, MPN/100 mL	170 ± 27	14 ± 6
Heterotrophic plate count, CFU/mL	3,500 ± 600	1,500 ± 450
Iron, mg/l	0.05	0.11
Lead, mg/l	0.04	0.11
Zinc, mg/l	0.04	3.6

Values represent mean of 3 samples.

amount of stored rainwater was withdrawn daily from the bottles after thorough mixing and was brought to room temperature before column tests. Roof-collected rainwater was used in all column tests except in the tests to determine the effect of pH on microbiological removal. In this test, sewage-spiked rainwater, adjusted to the desired pH, was used so as to increase the influent bacteria levels.

Column tests

Iron hydroxide-coated sand or uncoated sand was packed in columns of 15 cm × 2.5 cm ID. Glass wool was used at the bottom of each column to prevent loss of media. Before being used in the experiments, deionised water was passed through the column to remove any unbound metallic hydroxide. Samples were passed through the columns by using gravity flow. Two types of column tests were conducted. In continuous column tests, roof-collected rainwater (concrete roof) was passed continuously through the columns at an empty bed contact time (EBCT) of 15 min. The test was continued for 600 bed volumes (BV). In intermittent column tests, 10 BV of rainwater was allowed to pass through the columns once a day with the same EBCT of 15 min, and this test was continued for 60 d (corresponding to a total BV of 600). Influent and effluent were analysed for total coliforms, faecal coliforms, heterotrophic plate count (HPC) and turbidity after every 100 or 50 BV.

To study simultaneous removal of bacteria and heavy metals, similar column tests were conducted using rainwater collected from a GI roof which had elevated levels of heavy metals. The effect of pH on bacteria removal was studied in the pH range of 6.0–9.0 using sewage-spiked rainwater. For this, 20 BV of rainwater was allowed to pass through a similar column at an EBCT of 5 min. The entire effluent was collected and analysed for different indicator organisms.

Bacterial enumeration

The concentration of total coliforms was estimated by a multiple-tube fermentation method (most probable number method) by employing lactose–tryptose broth. The inoculated samples were incubated at 37°C for 48 h. The result is expressed as most probable number per 100 mL (MPN/100 mL). The concentration of faecal coliforms was estimated again by a multiple-tube fermentation method using A-1 medium (Himedia Laboratories, Mumbai, India). The samples were incubated at 35°C for 3 h and at 44.5°C for 21 h, and the result is expressed as MPN/100 mL. Heterotrophic bacteria were enumerated using the pour plate method. In this method, 0.5 mL of water sample (or diluted water sample) was poured into a petridish. 5–6 mL of plate count agar was poured over this and was mixed well. The petridishes were incubated at 37°C for two days for the development of colonies. The result is expressed as colony-forming units per mL (CFU/mL). All the tests were conducted in accordance with the techniques described by American Public Health Association (APHA 1995).

Analysis of metals

Iron was analysed using the phenanthrolene method (APHA 1995). Lead and zinc were determined using flame atomic absorption (atomic absorption spectrophotometer Varian).

RESULTS AND DISCUSSION

Results of the column tests using roof-collected rainwater (cement concrete roof) are shown in Figures 1 and 2 and

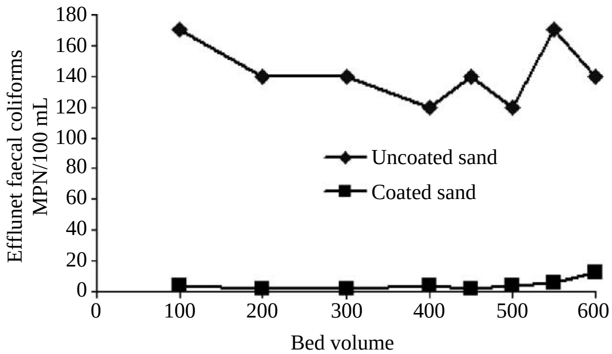

Figure 1 | Faecal coliform removal in long-duration column tests.

Table 2 | Summary of the results of continuous long-duration column tests

Parameter	Coated sand	Uncoated sand
Effluent turbidity, NTU	0.5–1.8	0.8–2.2
Effluent total coliforms, MPN/100 mL	14–40	1,100–1,700
Effluent faecal coliforms, MPN/100 mL	<2–4	120–170
Effluent HPC, CFU/mL	110–190	2,200–3,600

Influent for the test was rainwater collected from cement concrete roof (Table 1). Values indicate the range observed during the first 500 BV.

summarised in Table 2. The data show that coated sand media removed 97 to >99% of the total and faecal coliforms from the rainwater. After about 500 bed volumes (BV), the effluent quality deteriorated, and percent removal decreased to about 90% at 600 BV. The column operation was terminated after 600 BV. Effluent turbidity was generally below 2.0 NTU. Heterotrophic bacteria levels were below 200 CFU/mL. Compared to this, the total and faecal removal in the uncoated sand was only 11–21% throughout the filter run and heterotrophic plate counts were always >2,000 CFU/mL.

The intermittent column tests, duplicating the use pattern of household point of use water purification devices, with 10 BV per day also showed similar performance. Summary of the results is presented in Table 3. A gradual improvement in the performance of the uncoated sand was noted after about 200 BV (that is, 20 d) (data not shown). However, its performance never matched that of the coated sand medium. This improvement in the performance of uncoated sand might be explained based on the development of a microbial biofilm on the sand surface as in the case of slow sand filters.

In order to assess the performance of the coated sand medium in simultaneously removing bacteria and heavy metals, column tests were conducted with runoff from a galvanised iron (GI) roof which contained elevated levels of heavy metals, zinc and lead (Table 1). Results of the test presented in Table 4 shows that high metal levels did not influence the bacterial removal, and total coliform and faecal coliform removals were similar to that found in runoff from the cement concrete roof which had a low level of heavy metals. Further, the bacterial removal did not show any deterioration even after 600 BV. This can be explained on the basis of low levels of bacteria in the runoff from a GI roof (Table 1). The column run was terminated after 600 BV. Effluent heavy metal levels remained below 0.10 mg/l for zinc and 0.01 mg/l for lead throughout the column run.

Results of the short-duration column tests employing sewage-spiked rainwater to study the effect of pH on bacteria removal are presented in Figure 3. The data show no influence of pH on the removal of faecal coliforms by coated sand in the pH range studied (6.0–9.0). Several effluent samples were analysed for iron during this test, and the concentration of iron (used for coating the sand) was always less than 0.1 mg/l, much lower than the WHO guideline value

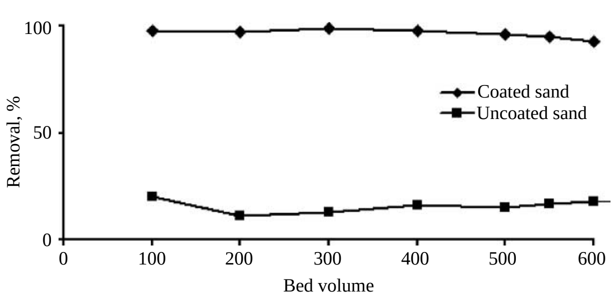

Figure 2 | Total coiform removal in long-duration column test.

Table 3 | Summary of the results of intermittent long-duration column tests

Parameter	Coated sand	Uncoated sand
Effluent turbidity, NTU	0.5–1.8	0.5–2.4
Effluent total coliforms, MPN/100 mL	17–30	600–1,700
Effluent faecal coliforms, MPN/100 mL	<2–7	60–160
Effluent HPC, CFU/mL	80–210	900–3,500

Influent for the test was rainwater collected from cement concrete roof (Table 1). Values indicate the range observed during 500 BV.

Table 4 | Summary of the results of the column tests for simultaneous removal of bacteria and heavy metals

Parameter	Value
Effluent total coliforms, MPN/100 mL	4–17
Effluent faecal coliforms, MPN/100 mL	<2–4
Effluent zinc, mg/l	<0.10
Effluent lead, mg/l	<0.01

Influent for the test was rainwater collected from GI roof (Table 1). Values indicate the range observed during 600 BV.

of 0.3 mg/l for drinking water (WHO 2003), which indicates that the coatings were stable at the pH range of 6.0–9.0.

Comparison of results to literature

A comparison of the results of this study with that reported in the literature on the use of coated sand for bacteria removal indicates a lower total and faecal coliform removal in the present study. For example, Lukasik *et al.* (1999) observed up to 5 log (that is, 99.999%) removal of *Escherichia coli* in column tests. This is presumably due to the high *E. coli* levels (10^6–10^7 CFU/mL) used in their studies. Also, while Lukasik *et al.* (1999) used sewage-spiked tap water, the present study employed rainwater which had a lower ionic strength. It is reported that ionic strength greatly affects the adsorption of microorganisms onto surfaces (Rutter & Vincent 1984). It should also be noted that, while in the present study bacterial removal decreased after 500 BV, Lukasik *et al.* (1999) reported a decreased *E. coli* removal after just 50 BV.

It is reported in the literature that iron coating increases the point of zero charge (pH_{zpc}) of the sand to about 8.6 against 6.8 of silica sand (Liu *et al.* 2005). The coating also increases the specific surface area of the medium (from 0.22 m^2/g for silica sand to 15.4 m^2/g for coated sand) (Liu *et al.* 2005). This indicates that adsorption, and not mechanical straining, is the main mechanism of bacterial removal by the coated sand. This is further supported by the increased effluent bacterial levels after 500 BV similar to a breakthrough in an adsorption operation. Lukasik *et al.* (1999) also observed similar trends.

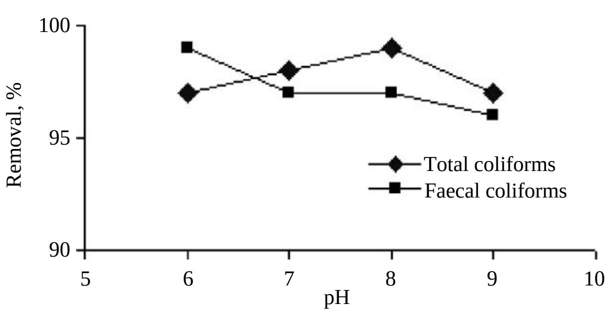

Figure 3 | Effect of pH on bacterial removal.

The observation that pH does not influence the bacterial removal in the range studied (pH 6.0–9.0) is of great practical significance in the application of the coated medium in point of use purification devices. It is reported by several authors that, while roof-harvested rainwater generally has a pH below 7.0, its storage in some materials, notably ferrocement, which is the most common material for storage tanks in developing countries, increases the pH of rainwater. Stored rainwater pH values as high as 9.9 have been reported in the literature (Handia 2005). Effectiveness of the coated sand in removing heavy metals without affecting the bacterial removal further enhances its potential for use in home water purification devices for treating roof-harvested rainwater in developing countries as elevated levels of metals are reported in some roof runoff.

It may be noted that bacteria levels used in the tests are very high compared to the levels that are generally found in domestic roof water harvesting systems. All roof water harvesting systems generally employ a first-flush device that eliminates the first few millimetres of greatly polluted roof runoff. In the present study, no first-flush device was used so as to get a higher influent bacterial concentration for column tests. This indicates that the effluent faecal coliform and total coliform levels from the coated sand columns would also be correspondingly lower. It should, however, be emphasised that the coated medium would not guarantee a zero faecal coliform level, and zero faecal coliform can be achieved only by some form of disinfection.

While the column studies indicated the potential of the coated media for use in a household filter, it is necessary for the household user to know when the bed is exhausted. This can be based on the volume of the water purified by a filter. It is also necessary for the user to know what to do after the bed is exhausted. There is evidence in the literature to suggest that bacteria removal takes place only in the top few centimetres of the coated sand bed. This suggests that, by replacing this layer by fresh media, bacterial removal can be restored to the original levels. Further studies should be carried out to find out how well the filter works in a range of situations and how long it can produce acceptable water. Based on this, a strategy can be worked out for the user to renew/replenish the media when it loses its effectiveness.

CONCLUSIONS

Column experiments were conducted with iron hydroxide-coated sand for removing bacteria and heavy metals from roof-harvested rainwater. Comparing plain sand, coated sand performed much better for removing bacteria and heavy metals. Long-duration column tests indicated that coated sand has significant treatment capacities for total and faecal coliforms with removal greater than 97% for 500 BV. Further long-duration column tests with elevated levels of heavy metals, lead and zinc showed that the medium is capable of effectively removing bacteria and heavy metals simultaneously for 600 BV, with effluent lead and zinc levels below the WHO guideline values for drinking water. Influent pH did not affect the bacterial removal in the pH range generally found in the rainwater harvesting systems. The results of the study showed the potential of iron hydroxide-coated sand as a sorptive filter medium for use in simple home water purification devices in developing countries for treatment of roof-harvested rain-water which is generally contaminated by microorganisms and heavy metals. Further studies are underway to use the coated sand medium in rooftop rainwater harvesting systems.

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