Limited effectiveness of household sand filters for removal of arsenic from well water in North Vietnam

Short title: Limited effectiveness of sand filters for arsenic removal from well water

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ABSTRACT

Since well water utilized for domestic purposes in Red River Delta of North Vietnam has been reported to be polluted by arsenic, barium, iron and manganese, household sand filters consisting of various components are used. However, information about the effectiveness of various sand filters for removal of the four toxic elements in well water is limited. In this study, arsenic levels in 13/20 of well water samples and 1/7 of tap water samples exceeded WHO health-based guideline value for drinking water. Moreover, 2/20, 6/20 and 4/20 of well water samples had levels exceeding the present and previous guideline levels for barium, iron and manganese, respectively. Levels of iron and manganese, but not arsenic, in well water treated by sand filters were lower than those in untreated water, although previous studies showed that sand filters removed all of those elements from water. A low ratio of iron/arsenic in well water may not be sufficient for efficient removal of arsenic from household sand filters. The levels of barium in well water treated by sand filters, especially a filter composed of sand and charcoal, were significantly lower than those in untreated water. Thus, we demonstrated characteristics of sand filters in North Vietnam.

Keywords: arsenic, barium, iron, manganese, sand filter, well water

INTRODUCTION

Well water derived from groundwater is important as a domestic water source including drinking water in Asian countries. However, well water may have a problem of naturally occurring arsenic (As) due to As-enriched aquifer. In fact, arsenic pollution of drinking water from wells has been found in many countries including Bangladesh, India, Cambodia, Myanmar, and Vietnam (Argos *et al*. 2012; Kumasaka *et al*. 2013). Previous studies showed that chronic exposure to As via drinking water can cause various diseases including cancers, cutaneous pigmented disorders and diabetes (Nizam *et al*. 2013; Yajima *et al*. 2015).

The World Health Organization (WHO) has provided guideline values for the quality of drinking water for barium (Ba), iron (Fe) and manganese (Mn) in addition to As in the present and/or previous editions (WHO 1984; WHO 2008; WHO 2011). Barium naturally coexists with other toxic elements in arsenic-polluted well water in Bangladesh and Vietnam (Frisbie *et al*. 2009; Kato *et al*. 2013). Our previous fieldwork studies showed a correlation between As and Ba in well drinking water and in human samples (urine, nails and hair) in Bangladesh (Kato *et al*. 2013). Our previous experimental studies *in vitro* showed that Ba promotes a malignant characteristic of human non-tumorigenic keratinocytes (Thang *et al*. 2015) and that Ba suppresses As-mediated anti-cancer effects (Yajima *et al*. 2012). Moreover, Ba was shown to promote hearing loss in mice (Ohgami *et al*. 2012) and humans (Ohgami *et al*. 2016) in our experimental study and epidemiological study, respectively. Previous studies showed that Fe promotes all steps of carcinogenesis (Weinberg 1996). In fact, previous studies have shown effects of iron on tumor initiation and tumor growth (Kumasaka *et al*. 2013; Torti & Torti 2013). A previous epidemiological study also showed that an elevated level of serum iron increases the risk of several carcinomas in humans (Wen *et al*. 2014). Exposure to Mn in drinking water has been reported to be associated with neurobehavioral disorders in mice (Krishna *et al*. 2014; Kumasaka *et al*. 2014). Exposure to Mn from well drinking water is also associated with an increase in infant mortality, cognitive deficit, memory deficit, and lower intelligence scores in humans (O'Neal & Zheng 2015). Thus, high uptake is a problem, which necessitates WHO to provide guidelines for the quality of drinking water, though some of the four elements are needed at trace levels. In addition, high levels of As, Ba, Fe and Mn are harmful to aquatic life, biota and the environment as well as human health (Wang 1988; Korte & Fernando 1991; Ventura-Lima *et al*. 2011; Kalantzi *et al*.

2013).

Approximately 13 million people in Vietnam (16.5% of the population), who mostly reside at the Red River Delta, are drinking water from wells (Nguyen *et al*. 2009; Winkel *et al*. 2011). There have been many reports on pollution of arsenic and other toxic elements in well drinking water in North Vietnam (Berg *et al*. 2006; Agusa *et al*. 2006, 2014; Nitzsche *et al*. 2015). Levels of As in the well water are more than 1,000 μg/L (Berg *et al*. 2001), more than 100-fold higher than the value in WHO health-based guidelines for drinking water. Previous studies also showed increased levels of Fe and Mn in well water in North Vietnam (Agusa *et al*. 2006; Berg *et al*. 2006; Nitzsche *et al*. 2015). However, there is little information on Ba levels in well water in North Vietnam compared to the information on As, Fe and Mn levels.

Another source of drinking water in Vietnam is harvested rainwater. The quality of harvested rainwater in Mekong Delta was investigated in a previous study (Wilbers *et al*. 2013). The quality of well water and that of rainwater were compared in another study in Hanoi (Agusa *et al*. 2006). However, there has been no report comparing the quality of well water, rainwater and tap water in North Vietnam.

Household sand filters are commonly used to remediate well water in North Vietnam in order to obtain drinkable water by removing Fe, which causes discoloration and a metallic taste (Berg *et al*. 2006). Previous studies showed that sand filters are effective for removing As, Fe, and Mn from well water (Berg *et al*. 2006; Nitzsche *et al*. 2015). However, information on the effectiveness of sand filters for removal of Ba in well water is limited despite the fact that previous studies showed high levels of Ba in well water in North Vietnam (Agusa *et al*. 2006; Winkel *et al*. 2011). To our knowledge, there is also limited information on the effectiveness of sand filters consisting of different components for removal of toxic elements in well water.

In order to assess the health risk of domestic water in North Vietnam, levels of four toxic elements (As, Ba, Fe and Mn) were compared among well water, rainwater and tap water in this study. The effectiveness of household sand filters consisting of different components for removal of toxic elements was also investigated.

METHODS

Water sampling

Water samples were collected at Vinh Tru Town (6 hamlets), Nhan Khang commune (3 hamlets) and Dong Ly commune (1 hamlet) in Ly Nhan District of Ha Nam Province, where pollution of As in well water was previously reported (Nguyen et al., 2009). A total of 20 untreated and 77 sand filter-treated well water samples were collected. In addition, 105 harvested rainwater and 7 tap water samples were collected. All samples were collected in polyethylene bottles. Each bottle was filled with sampled water after rinsing out the inside of the bottle with sampled water. Tightly capped bottles were promptly sent from Vietnam to Japan by airplane. Samples were kept at 4°C and measurements of total levels of As, Ba, Fe and Mn were completed within two weeks after arrival in Nagoya University. Free and informed consent of the participants or their legal representatives was obtained, and the study protocol was approved by the Ethical Committee of Chubu University, Aichi, Japan (approval no. 20008 on July 9, 2008; approval no. 260019 on July 8, 2014) and the Ethical Committee of Nagoya University, Aichi, Japan (approval number 2013-0070 on July 22, 2013) by following the regulations of the Japanese government.

Analytical method

Levels of As, Ba, Fe and Mn in water samples were quantified by using an inductively coupled plasma-mass spectrometer (ICP-MS; 7500cx, Agilent Technologies Inc, CA) according to the method previously described (Ohgami *et al*. 2012; Kato *et al*. 2013). The limits of detection for As, Ba, Fe and Mn by the ICP-MS are 0.1, 0.1, 3.0 and 0.1 μg/L, respectively.

Statistical analyses

The Mann-Whitney *U* test and chi-square test were used for statistical analyses to compare differences between groups. The JMP Pro software package (version 11.0.0; SAS Institute, Cary, NC, USA) was used for all statistical analyses.

RESULTS

Levels of the 4 elements in various sources of water in Red River Delta of North Vietnam

Median levels of As, Ba, Fe and Mn in well water samples (n=20), rainwater samples (n=105) and tap water samples (n=7) collected in Ha Nam Province, Vietnam are shown in Figure 1. Levels (mean ± SD) of As, Ba, Fe and Mn in well water samples (n=20) collected in Ha Nam Province, Vietnam are shown in Table 1. Boxplots (Figure 1) show that the levels of As, Ba, Fe and Mn in rainwater and tap water were lower than those in well water. However, there was one tap water sample containing 27.8 μg/L of As (Figure 1a).

Since the levels of the 4 elements in well water were higher than those in rainwater and tap water, pollution of the toxic elements in well water was then focused on in this study. The median level (14.6 μg/L) of As in well water exceeded value in the current WHO guidelines (10 μg/L) for drinking water, while the median level of Ba in well water was below the value in the current guidelines (700 μg/L). Although there are no guideline values of Fe and Mn in the present edition, the median levels of Fe and Mn in well water were below the guideline values in the previous editions (300 μg/L for Fe and 400 μg/L for Mn). The percentages of unsafe wells with levels of As, Ba, Fe and Mn exceeding the values of WHO health-based guidelines for drinking water in the present and/or previous editions were 65%, 10%, 30% and 20%, respectively (Table 1).

Levels of the 4 elements in untreated well water and well water treated by sand filters

In our fieldwork study, 79.4% (77/97) of randomly selected wells water were treated by sand filters (Figure 2), suggesting that the use of sand filters is common in Ha Nam Province. Boxplots of the levels of As, Ba, Fe and Mn in untreated well water samples (n=20) and well water samples treated by sand filters (n=77) (Figure 2) show that the levels of As in untreated well water and well water treated by sand filters are comparable (Figure 2a). Correspondingly, the percentages of unsafe wells with As-polluted untreated water and water treated by sand filters were comparable (Table 1). These results suggest very limited effectiveness of sand filters for removal of As from well water in North Vietnam. Levels of Ba, Fe and Mn in well water treated by sand filters were significantly lower than those in untreated well water (Figure 2b-d), though the difference in Ba levels is smaller than the differences in Fe and Mn levels in untreated well water and well water treated by sand filters. Furthermore, the percentages of wells with Fe-polluted and Mn-polluted but not Ba-polluted well water treated by sand filters were significantly lower than those with water not treated by sand filters (Table 1). These results suggest that the effectiveness of sand filters for removal of Fe and Mn is greater than their effectiveness for removal of Ba in well water in North Vietnam.

Levels of As, Ba, Fe and Mn in untreated well water and in well water treated by three kinds of sand filters consisting of sand only, sand plus charcoal and sand plus gravel (Figure 3) were compared. The levels of As in untreated well water and in well water treated by the three kinds of sand filters were comparable (Figure 4a), while levels of Fe and Mn in well water treated by the three kinds of sand filters were lower than those in untreated well water (Figure 4c, d). These results suggest that all kinds of sand filters can remove Fe and Mn but not As. Moreover, levels of Ba in well water treated by a sand filter consisting of sand and charcoal were lower than the levels in untreated well water (Figure 4b). Levels of Ba in well water treated by a sand filter consisting of sand only and in well water treated by a sand filter consisting of sand plus gravel were comparable to the levels in untreated well water. These results suggest that a sand filter consisting of sand and charcoal may effectively remove Ba from well water.

DISCUSSION

We first compared levels of As, Ba, Fe and Mn in well water, rainwater and tap water in North Vietnam. Since the level of As in 65% of the well water samples exceeded the value in WHO health-based guidelines for drinking water, well water is not suitable for drinking water as was shown in previous studies (Berg *et al*. 2007; Agusa *et al*. 2014). Rainwater and tap water seem to be suitable for drinking from the viewpoint of the four toxic elements. Unexpectedly, however, As at a level of 27.8 μg/L was detected in one of seven tap water samples. Our results suggest that continuous monitoring for various kinds of water including tap water is important in the future in North Vietnam.

A previous study showed that the level of As in well water from Ly Nhan District, North Vietnam was 420 μg/L and that the level in sand-filtered water was 23 μg/L, with 80% of the filtered water still containing As at a level higher than 10 μg/L (Agusa *et al*. 2014). Other studies in Vietnam also showed that sand filters could remove 80% (Berg *et al*. 2006) to 95% of As (Nitzsche *et al*. 2015). Sand filters have been reported to be effective for removal of Mn and Fe with 86.2-99.6% efficiency and nearly 100% efficiency, respectively (Nitzsche *et al*. 2015). Correspondingly, our results showed that levels of Fe and Mn in well water treated by sand filters were lower than those in untreated well water, suggesting that sand filters are effective for removal of Fe and Mn in well water. In contrast to previous studies (Agusa *et al*. 2014; Nitzsche *et al*. 2015), however, levels of As in untreated well water and in well water treated by sand filters were comparable in our study. A previous study showed that Fe/As ratios of ≥50 and >250 are required to reduce As concentrations to levels below 50 μg/L and 10 μg/L, respectively, indicating that the level of Fe in water affects removal of As by sand filters (Berg *et al*. 2006). The median of Fe/As ratio in well water was about 5 in this study (Supplemental Figure S1) and this might be a reason why sand filters could not remove As from well water. Our results suggest that the Fe/As ratio in well water should be examined to determine the effectiveness of sand filters for removal of As. Since a sand filter composed of sand and zero-valent iron filings was shown to be effective for arsenic removal (Leupin & Hug 2005; Leupin *et al*. 2005; Mehta & Chaudhari 2015), the use of such a sand filter may be one way to overcome the low performance of a sand filter based on the Fe/As ratio. Proper design of a sand filter may also improve the effectiveness for As removal (Leupin *et al*. 2005). Our results showing a decrease of 24% in Ba in well water treated with sand filters suggest that sand filters can partially remove Ba from well water. To our knowledge, however, there has been no study showing the effectiveness of charcoal and a charcoal-containing sand filter for Ba removal. Further study on their effectiveness for Ba removal is needed.

CONCLUSIONS

After confirming pollution of As, Ba, Fe and Mn in well water in North Vietnam, we found that sand filters could remove Fe and Mn but not As, despite the fact that previous studies showed that sand filters could remove As (Berg *et al*. 2006; Nitzsche *et al*. 2015). We then showed that a low ratio of Fe/As in well water in North Vietnam might be one of the reasons for the poor performance of sand filters. Thus, this fieldwork study clarified characteristics of various sand filters being used in North Vietnam.

Statement of conflict of interest

All authors declare that there is no conflict of interest.

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Figure legends

Figure 1. Boxplots of levels of 4 elements in well water, rainwater and tap water in North Vietnam. Concentrations of As, Ba, Fe, and Mn in well water, harvested rainwater and tap water in Ha Nam Province, Vietnam are presented. Boxplots are used to present the first quartile, median, third quartile (each presented by a horizontal line of the box) and minimum (lower whisker) and maximum (upper whisker) values. Broken lines show previous (Fe and

Mn) and present (As and Ba) guideline values for drinking water by WHO. * and **, significantly different (*, $p<0.05$; **, $p<0.01$) from well water by the Mann-Whitney *U* test.

Figure 2. Boxplots of levels of 4 elements in untreated well water and well water treated by sand filters. Concentrations of As, Ba, Fe, and Mn in untreated well water (control) and well water treated by sand filters in Ha Nam Province, Vietnam are presented. Boxplots are used to present the first quartile, median, third quartile (each presented by a horizontal line of the box) and minimum (lower whisker) and maximum (upper whisker) values. Broken lines show previous (Fe and Mn) and present (As and Ba) guideline values for drinking water by WHO. * and **, significantly different (*, $p<0.05$; **, $p<0.01$) from the untreated control by the Mann-Whitney *U* test.

Figure 3. Cross-sectional graphs of household sand filters used in Ha Nam, North Vietnam. Schemas for 3 kinds of sand filters composed of sand (a), sand and charcoal (b) and sand and gravel (c) are presented.

Figure 4. Boxplots of levels of 4 elements in untreated well water and well water treated by 3 kinds of sand filters. Concentrations of As, Ba, Fe, and Mn in untreated well water and well water treated by various sand filters composed of sand only, sand plus charcoal and sand plus gravel in Ha Nam Province, Vietnam. Boxplots are used to present the first quartile, median, third quartile (each presented by a horizontal line of the box) and minimum (lower whisker) and maximum (upper whisker) values. * and **, significantly different (*, $p<0.05$; **, $p<0.01$) from the untreated control by the Mann-Whitney *U* test.

Table legend

Table 1. Levels of 4 elements in untreated well water and well water treated by sand filters. Levels (mean ± SD) of As, Ba, Fe and Mn in untreated well water and well water treated by sand filters in North Vietnam are presented. Unsafe wells (%) are percentages of wells with levels of As, Ba, Fe and Mn exceeding the values in WHO health-based guidelines in untreated well water and well water treated by sand filters. #, ## and ###, WHO guideline values in the previous (###, 1984; ##, 2008) and present (#, 2011) editions. **, statistically different ($p<0.01$) from the untreated control by the chi-square test. ns, not significant.

Supplemental Figure

Figure S1. Boxplot of Fe/As ratio in well water from Ha Nam Province, Vietnam. Fe/As ratios of untreated well water (n=20) in Ha Nam Province, Vietnam. Boxplots are used to present the first quartile, median, third quartile (each presented by a horizontal line of the box) and minimum (lower whisker) and maximum (upper whisker) values.

Figure 1

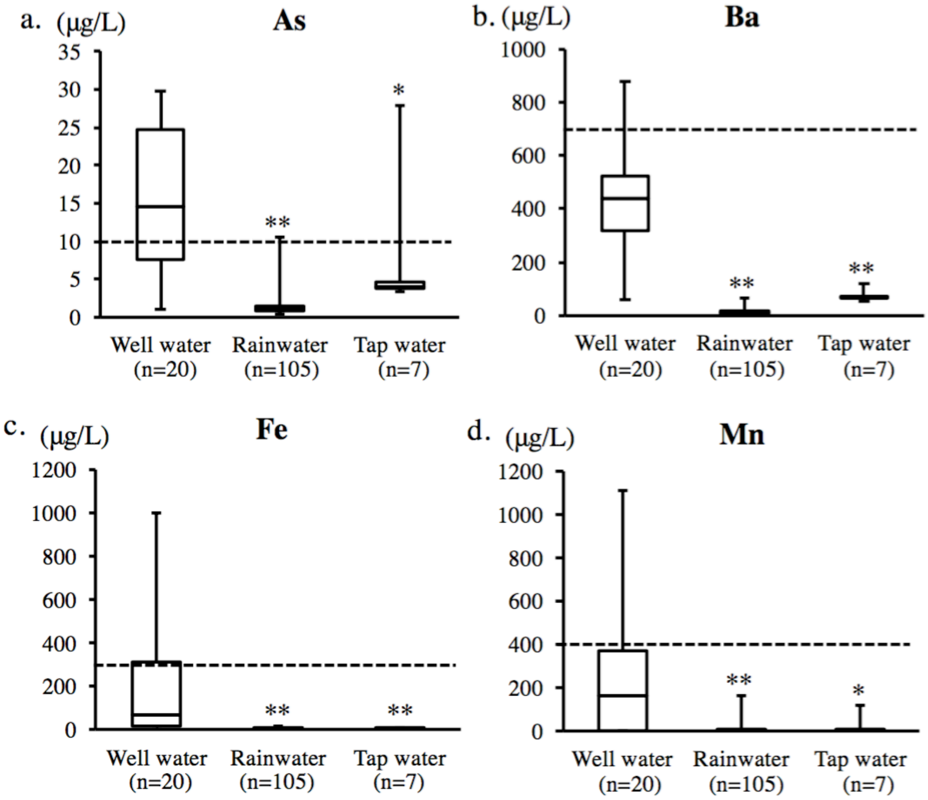
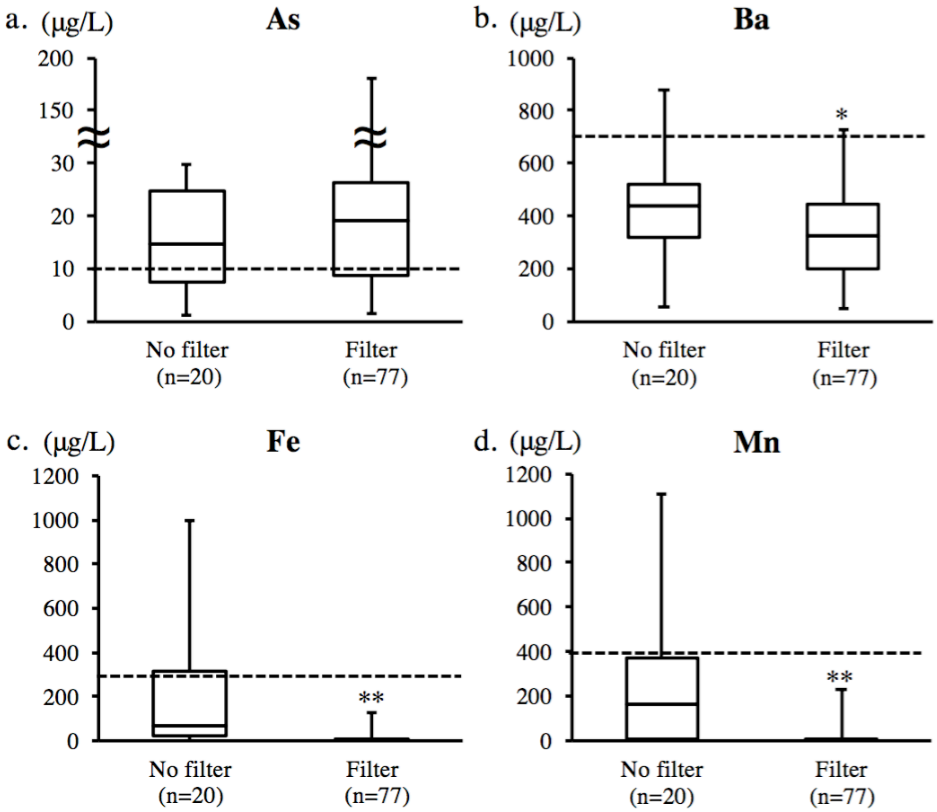

a. (µg/L)
As
35
30
25
20
15
10
5
0
*
**
Well water (n=20)
Rainwater (n=105)
Tap water (n=7)
b. (µg/L)
Ba
1000
800
600
400
200
0
**
**
Well water (n=20)
Rainwater (n=105)
Tap water (n=7)
c. (µg/L)
Fe
1200
1000
800
600
400
200
0
**
**
Well water (n=20)
Rainwater (n=105)
Tap water (n=7)
d. (µg/L)
Mn
1200
1000
800
600
400
200
0
**
*
Well water (n=20)
Rainwater (n=105)
Tap water (n=7)

Figure 2



a. (µg/L)
As
200
150
30
20
10
0
No filter
(n=20)
Filter
(n=77)
b. (µg/L)
Ba
1000
800
600
400
200
0
*
No filter
(n=20)
Filter
(n=77)
c. (µg/L)
Fe
1200
1000
800
600
400
200
0
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No filter
(n=20)
Filter
(n=77)
d. (µg/L)
Mn
1200
1000
800
600
400
200
0
**
No filter
(n=20)
Filter
(n=77)

Figure 3

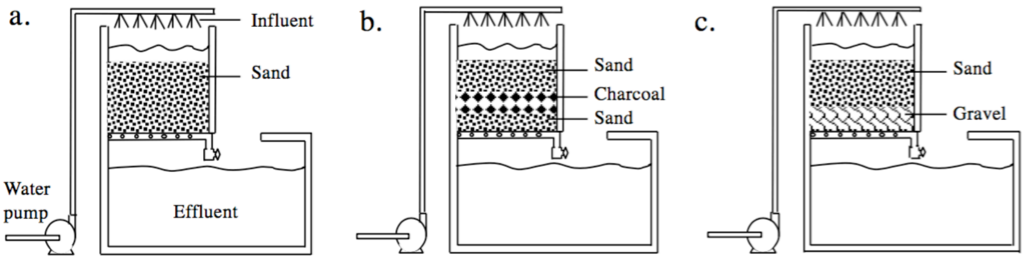
a.
Influent
Sand
Water pump
Effluent
b.
Sand
Charcoal
Sand
c.
Sand
Gravel

Figure 4

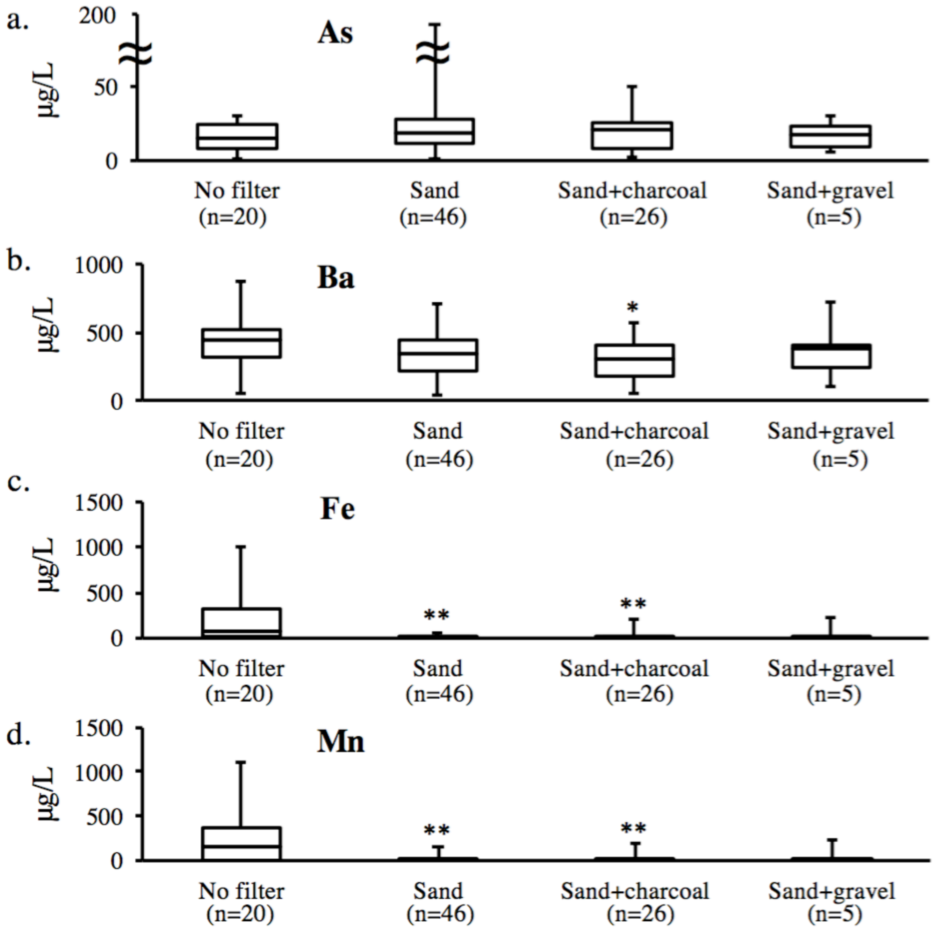
a.
As
200
50
0
μg/L
No filter
(n=20)
Sand
(n=46)
Sand+charcoal
(n=26)
Sand+gravel
(n=5)
b.
Ba
1000
500
0
μg/L
*
No filter
(n=20)
Sand
(n=46)
Sand+charcoal
(n=26)
Sand+gravel
(n=5)
c.
Fe
1500
1000
500
0
μg/L
**
**
No filter
(n=20)
Sand
(n=46)
Sand+charcoal
(n=26)
Sand+gravel
(n=5)
d.
Mn
1500
1000
500
0
μg/L
**
**
No filter
(n=20)
Sand
(n=46)
Sand+charcoal
(n=26)
Sand+gravel
(n=5)

Table 1

Element	WHO guideline value (µg/L)	Element concentration (µg/L)		Filter	Unsafe wells (%)	
		Mean	SD			
As	10 #	15.7	20	No	65	ns
		23.7	28.0	Yes	74	
Ba	700 #	422.4	240.4	No	10	ns
		322.9	168.6	Yes	2.6	
Fe	300 ###	186.3	267.2	No	30	**
		10.2	22.3	Yes	0	
Mn	400 ##	236.0	287.7	No	20	**
		16.2	46.4	Yes	0	

Figure S1