Use of bauxite for enhanced removal of bacteria in slow sand filters

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ABSTRACT

Slow sand filtration (SSF) is a well-known process for drinking water treatment and is widely used for the production of biologically stable drinking water and particle removal. The removal process of particles and microorganisms is highly dependent on the buildup of the schmutzdecke at the filter surface. During the ripening period and especially for cold waters, the buildup of the schmutzdecke may take several months until such filters are biologically mature and at steady-state regarding their removal performance for particles and microorganisms. In order to improve the performance of SSF in terms of the removal of bacteria, e.g. *Escherichia coli* and *Enterococcus,* pilot tests using natural bauxite as a filter media have been performed. The results showed a significant improvement in bacteria retention within the filter bed of a second-stage slow sand filter containing different depths of bauxite.

Key words | bauxite, *E. coli*, *Enterococcus*, slow sand filtration, zeta potential

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INTRODUCTION

Slow sand filtration (SSF) is a well-known process for drinking water treatment and was first introduced in the city of Paisley in Scotland to provide water to a local industry (Graham & Collins 2014). Thanks to the overall efficiency of SSF for the removal of waterborne pathogens, epidemics such as cholera almost totally disappeared from central Europe by the beginning of the 20th century (WHO 1975).

During recent decades, the removal of pathogenic microorganisms such as bacteria, viruses, *Giardia lamblia* and *Cryptosporidium parvum* in SSF has been widely documented (e.g. WHO 1975; Bellamy *et al.* 1985; IRC 1998; Hijnen *et al.* 2004; Hijnen *et al.* 2007). Presently, numerous utilities in Europe, Asia and North America continue to use SSF either for particle removal or as a polishing step at the end of multi-barrier treatment chains (e.g. Gimbel *et al.* 2006).

At the beginning of the 20th century, many utilities started to progressively replace SSF by rapid sand filtration (RSF), which has become today the most common filtration process used for drinking water treatment. Rapid filters are generally operated at hydraulic loadings of 5–10 m/h and require the dosage of a coagulant in order to destabilize the particles which are to be retained in the filter bed. RSFs also require automatic backwash with water and air, requiring electric pumps and pressured air.

Slow sand filters are commonly designed with hydraulic loadings of 0.1–0.2 m/h and media depths of about 1 metre (WHO 1975). Quartz sand is usually used as filter media but pumice, anthracite and granular activated carbon (GAC) are also used in some cases. It is generally admitted that the removal of particles and microorganisms is strongly related to the buildup of the schmutzdecke (filter ripening) at the filter surface (e.g. WHO 1975; IRC 1998; Unger & Collins 2008). The duration of the ripening period is largely dependent on the SSF-influent water quality, particularly in terms of the concentration of particles and biodegradable organic matter as well as temperature.

In general, the different filter media used for drinking water filters all have negative surface charges at the pH values of natural waters (e.g. Sharma *et al.* 1987). The surface charge of filter media is commonly measured as zeta

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potential. Because the particles and microorganisms suspended in the raw water are also negatively charged (e.g. Schinner *et al.* 2010), their retention within the filter bed is compromised by the repulsive forces between the media grains (the collectors) and the particles/microorganisms suspended in the water to be treated. Yao *et al.* (1971) have provided a fundamental understanding of the retention mechanisms of particles and the relevant parameters in packed bed filters.

The overall fraction of particles removed in a packed bed filter at steady-state can be calculated using the following equation (Yao *et al.* 1971):

$$\frac{N}{N_0} = \exp\left\{\frac{-3(1-\varepsilon_0)}{2}\alpha\eta\frac{L}{d_m}\right\} \quad (1)$$

where N_0: initial concentration of particles, N: final concentration of particles, ε_0: initial bed porosity, α: collision efficiency factor, η: single collector efficiency, L: filter bed depth, d_m: average media diameter (the filter media is assumed to be spherical).

The single collector efficiency η is a ratio, i.e. the rate at which particles (and microorganisms) strike the collector (the media grain) divided by the rate at which particles (and microorganisms) flow toward the collector (Yao *et al.* 1971). These authors have shown that the single collector efficiency η is lowest for particles in the range of 1 μm diameter. This is of particular interest because many pathogenic bacteria and viruses contained in natural waters are about 1 μm in size. The removal of such microorganisms in packed bed filters is therefore particularly challenging.

The collision efficiency factor α reflects the chemistry of the system and is defined as a ratio, i.e. the number of contacts which succeed in producing adhesion divided by the number of collisions which occur between suspended particles (and microorganisms) and the filter media (Yao *et al.* 1971). In a completely destabilized system, i.e. with optimized dosage of a coagulant, α is ideally equal to 1 (Yao *et al.* 1971). However, for SSF where no chemicals are added for particle destabilization, α is much smaller than 1 and largely dependent on the surface charge of the particles (microorganisms) and the filter media (e.g. Tufenkji & Elimelech 2004; Schijven *et al.* 2014). Consequently, for slow sand filters, α can be increased by using media with a less negative surface charge. As mentioned earlier, the commonly used filter media for SSF, e.g. quartz sand and GAC, have relatively low collision efficiency factors for particle and microorganism removal, because of their overall negative surface charge (e.g. Sharma *et al.* 1987; Kim *et al.* 2012).

Different authors have shown the effects of media surface charge on the removal of particles and microorganisms in filter beds (Prasad & Chaudhuri 1986; Truesdail *et al.* 1998; Elimelech *et al.* 2000; You *et al.* 2005; Pal *et al.* 2006). These studies generally used some artificially treated filter media in order to obtain less negatively charged filter media, e.g. quartz sand or GAC coated with aluminum and/or ferric oxides. These studies have shown the tremendous effect of the surface charge of the filter media on the removal of suspended particles and microorganisms in packed filter beds.

Other researchers have shown the advantages of adding zerovalent iron particles to biofilters in order to generate positively charged iron oxides (rust) to which the microorganisms (and particles) may adsorb (Noubactep 2010; Bradley *et al.* 2011).

In order to overcome the drawbacks of the negatively charged filter media commonly used for SSF, the use of natural bauxite as a filter media has been tested in the present study. Bauxite, an aluminum ore, contains a large fraction (>50%) of aluminum oxides (Al_2O_3) and is the raw material used for the production of aluminum. Bauxite is readily available in large quantities worldwide (e.g. West Africa, Australia, China, and Brazil). Different studies have shown that the zeta potential of natural bauxite at the pHs of natural waters is close to zero (e.g. Barbato *et al.* 2011). It was therefore expected that natural bauxite would be more efficient for the removal of microorganisms and particles in slow sand filters compared to filter media such as quartz sand.

MATERIALS AND METHODS

The principal objectives of the study were the following:

- Investigate the performance of natural bauxite as a filter medium at pilot scale over an extended period of time (>1 year) using a highly variable natural water.

- Evaluate the effects of different depths of bauxite in slow sand filters on the performance of the filters.
- Evaluate the potential leaching of dissolved aluminum in the effluent of a slow sand filter containing 100% bauxite.

The pilot tests were conducted during a period of 18 months using raw water from a small creek in Porrentruy, Switzerland. Figure 1 shows the layout of the multistage filtration (MSF) pilot plant. The treatment chain consisted of two upflow roughing filters (URF) in series followed by two stages of SSF. The hydraulic loadings of the URFs and the SSF followed the general recommendations of WHO (1975) and Sandec (1996): 0.45 m/h for the URFs, respectively, 0.15 m/h for SSF. Different authors have shown the performance and robustness of MSF for the treatment of poor quality raw waters in terms of particle and microorganism removal (Sandec 1996; IRC 1998).

The raw water quality of the karstic creek (Bacavoine) is shown in Table 1. Raw water quality is highly variable as a function of the meteorological conditions in the watershed.

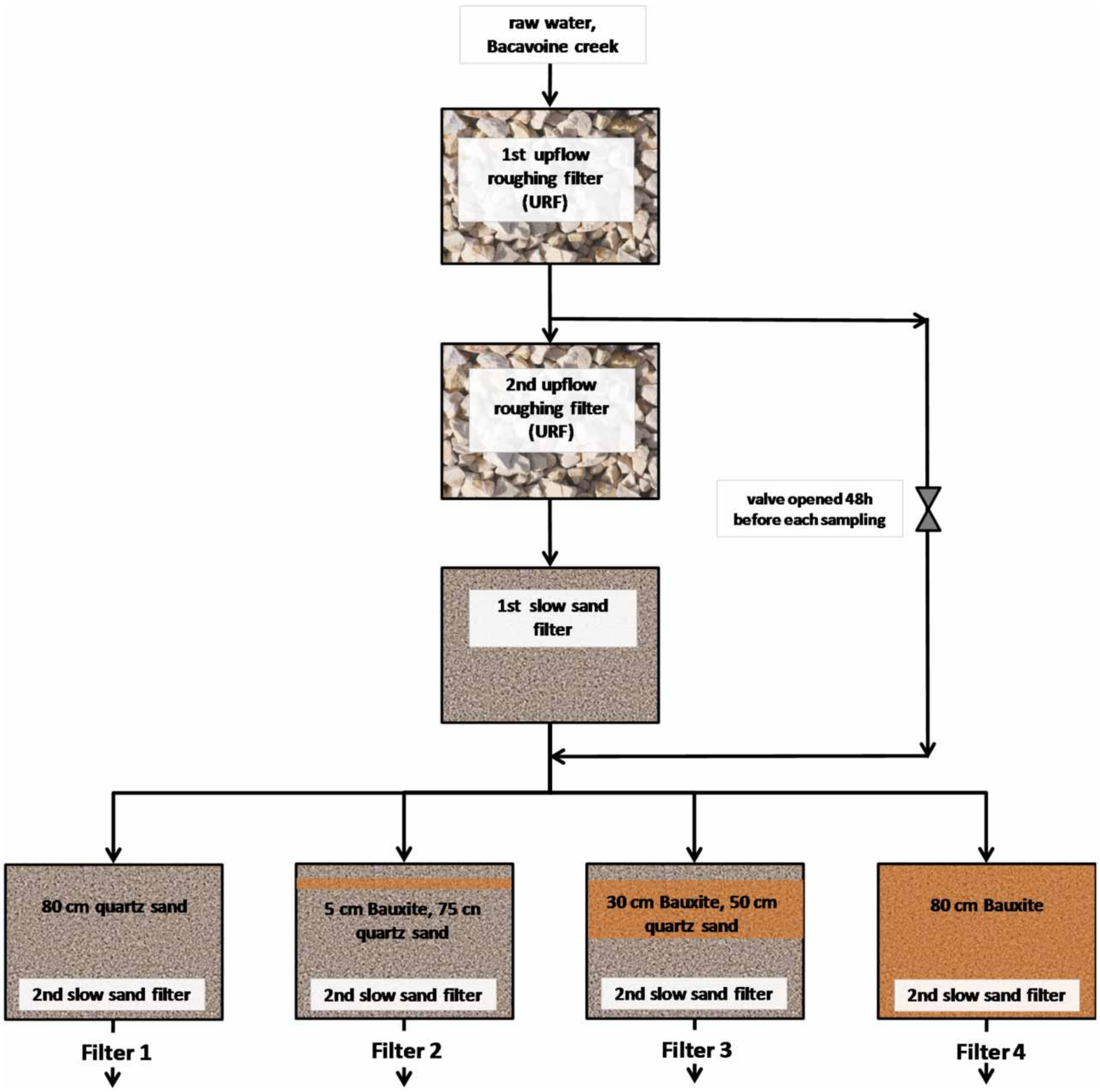

Figure 1 | Layout of the MSF pilot plant.

Table 1 | Raw water quality of Bacavoine creek[a]

Parameter	Temperature [°C]	pH [–]	Conductivity [µS/cm at 20 °C]	Turbidity [FTU]	DOC [mg C/L]	Ammonium [mg NH_4^+/L]	Nitrate [mg NO_3^-/L]	Total hardness [°f]	*Escherichia coli* [CFU/100 mL]	*Enterococcus* [CFU/100 mL]	HPC [CFU/mL]
Minimum	6.9	6.25	354	0.3	0.5	0.00	10.0	21.0	0	0	30
Maximum	14.1	7.89	530	39.0	4.1	1.96	47.0	29.0	10,300	4,500	21,000
Average	9.7	7.29	439	3.5	1.3	0.06	16.1	26.1	604	339	3,149
Std Dev	0.9	0.23	27	5.9	0.7	0.2	4.6	1.3	1,370	710	3,901

[a]n = 120, grab samples between 1980 and 2009, 1 °f = 10 mg $CaCO_3$/l.

The second-stage slow sand filter step was subdivided into four parallel compartments with a surface area of 0.2 m^2 containing different filter media. In order to challenge the second-stage slow sand filters with water of relatively poor quality, the filters were fed with the effluent of the first roughing filter for a period of 48 h before each sampling campaign (Figure 1). Following the sampling, the operational mode of the pilot plant was switched back to normal. A total of 22 campaigns were realized over a period of 18 months. The composition of the media of the four second-stage filters is shown in Table 2.

The bauxite used for our experiments was raw bauxite from southern France provided by SIBELCO Europe, (34540 Balaruc-les-Bains, France) with a media diameter of 0.2–0.5 mm. The zeta potential of the ground bauxite (d < 0.063 mm) was measured by EMPA, Dübendorf, Switzerland using the electro-acoustic method and an apparatus from Colloid Dynamics. The standard quartz sand in filters 1 to 3 had a media diameter of 0.1–0.3 mm and was provided by Carlo Bernasconi Ltd, Bärschwil, Switzerland.

Escherichia coli and *Enterococcus* were analyzed using the standard plate-count method according to *Standard Methods* (APHA AWWA WEF 2014) and were quantified as colony forming units (CFU/100 mL). Instant turbidity was measured with a portable turbidity meter (Hach Company, Loveland, Colorado 80539, USA) at the same time as the microbiological samples were taken. Dissolved aluminum and other metals were analyzed using the inductively coupled plasma mass spectrometry (ICP-MS) method and a Varian ICP-MS apparatus.

The pilot investigation was started at the beginning of June 2013 and lasted for over 500 days.

Table 2 | Composition of the filter media of the four second-stage slow sand filters

Filter 1	Filter 2	Filter 3	Filter 4
Top of filter			
80 cm quartz sand	5 cm quartz sand 5 cm bauxite 70 cm quartz sand	5 cm quartz sand 30 cm bauxite 45 cm quartz sand	80 cm bauxite
Bottom of filter			

RESULTS AND DISCUSSION

Zeta potential of bauxite

Figure 2 shows the zeta potential of the natural bauxite used for the pilot tests. The measured isoelectric point was between 6.8 and 6.9. At the pH of the natural creek water (pH =7.0–7.3) the zeta potential of the media was between −1 and −3 mV.

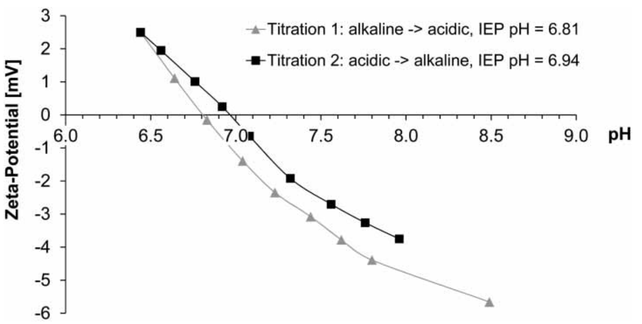

Figure 2 | Zeta potential of the natural bauxite used in the pilot tests.

Removal of *E. coli* and *Enterococcus*

The results for *E. coli* and *Enterococcus* in the influent and the effluents of the four parallel filters are shown in Tables 3 and 4.

The data in Tables 3 and 4 show the positive effect of the use of bauxite on the removal of fecal indicator bacteria. In the filter containing 100% bauxite (filter 4), *E. coli* was never detected in the effluent during the entire experiment, whereas the removals in the control filter containing quartz sand (filter 1) were substantially lower (Table 3). For *Enterococcus* the results were similar with no detectable

Table 3 | *E. coli* in the influent and the effluents of the four parallel, second-stage slow sand filters

		***E. coli* [CFU/100 mL] in effluent**				
Date	**Time since startup (days)**	**Influent**	**Filter 1, 80 cm q.**	**Filter 2, 5 cm b., 75 q.**[a]	**Filter 3, 30 cm b., 50 cm q.**	**Filter 4, 80 cm b.**
05.06.2013	0	Startup of experiment				
11.06.2013	5	1,300	600	580	0	0
24.06.2013	19	178	38	13	0	0
19.08.2013	75	79	12	2	0	0
04.09.2013	91		Introduction of 5 cm of bauxite in filter 2			
16.09.2013	103	37	23	4	1	0
14.10.2013	131	240	22	81	0	0
18.11.2013	167	150	1	2	0	0
09.12.2013	188	16	2	1	0	0
16.01.2014	226		Removal of the schmutzdecke from the four filters			
03.02.2014	244	96	0	3	0	0
03.03.2014	272	90	1	0	0	0
31.03.2014	300	5	1	0	0	0
28.04.2014	328	87	8	1	0	0
02.06.2014	363	21	4	2	0	0
30.06.2014	391	270	133	27	4	0
18.08.2014	440	118	43	3	1	0
08.09.2014	461		Removal of the schmutzdecke from the four filters			
15.09.2014	468	25	8	0	0	0
13.10.2014	496	220	72	51	2	0
17.11.2014	531	370	147	125	35	0
08.12.2014	551	210	50	37	5	0

[a]5 cm bauxite, 75 cm quartz sand.

Table 4 | *Enterococcus* in the influent and the effluents of the four parallel, second-stage slow sand filters

		Enterococcus [CFU/100 mL] in effluent				
Date	**Time since startup (days)**	**Influent**	**Filter 1, 80 cm q.**	**Filter 2, 5 cm b., 75 cm q.[a]**	**Filter 3, 30 cm b., 50 cm q.**	**Filter 4, 80 cm b.**
05.06.2013	0	Startup of experiment				
11.06.2013	5	420	240	200	0	0
24.06.2013	19	170	28	2	0	0
19.08.2013	75	32	8	3	0	0
04.09.2013	91	Introduction of 5 cm of bauxite in filter 2				
16.09.2013	103	14	16	3	0	0
14.10.2013	131	90	8	21	0	0
18.11.2013	167	18	1	0	0	0
09.12.2013	188	26	0	0	0	0
16.01.2014	226	Removal of the schmutzdecke from the four filters				
03.02.2014	244	44	19	1	0	0
03.03.2014	272	14	0	0	0	0
31.03.2014	300	0	0	0	0	0
28.04.2014	328	12	1	0	0	0
02.06.2014	363	0	3	3	0	0
30.06.2014	391	230	97	36	4	0
18.08.2014	440	61	23	4	1	0
08.09.2014	461	Removal of the schmutzdecke from the four filters				
15.09.2014	468	23	5	0	1	0
13.10.2014	496	250	44	40	1	0
17.11.2014	531	200	52	43	16	1
08.12.2014	551	80	15	10	1	0

[a]5 cm bauxite, 75 cm quartz sand.

bacteria in the effluent of filter 4 except for one value of 1/100 mL on day 531 after startup, whereas filter 1 showed substantially higher values in the effluent during the entire experiment (Table 4).

In the filter containing 30 cm of bauxite (filter 3), the data in Tables 3 and 4 suggest that *E. coli* and *Enterococcus* started to break through after about 1 year of continuous filter operation or about 4,600 bed volumes (BV) of filtered water (BV calculated on the basis of 30 cm of bauxite). Until day 363, both indicator bacteria were always below the detection limit in the effluent of filter 3, except for one value of 1 *E. coli*/100 mL on day 103 following startup (Table 3).

In filter 2, where 5 cm of bauxite was added on day 91, the effluent concentrations of *E. coli* and *Enterococcus* were in general slightly lower compared to filter 1 (control), however, the difference was relatively low and somewhat inconsistent (Tables 3 and 4). Based on these results it appears that the addition of 5 cm of bauxite was insufficient in order to substantially increase the filter performance.

The results obtained for turbidity are shown in Figure 3. The results showed a similar trend compared to the results for bacteria, i.e. an improved performance of the bauxite-containing filters (filters 3 and 4) compared to the control filter. The entire dataset for turbidity is shown in the Supplementary Material (available with the online version of this paper).

The effluent turbidity in filters 3 and 4 was continuously below 0.02 NTU during the entire experiment, independently of the influent turbidity (cf. Table S1). In the control filter, the turbidity was more variable and substantially higher (Figure 3 and Table S1).

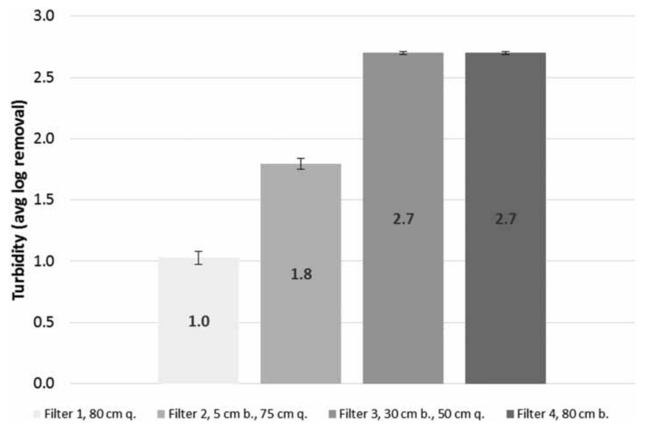

Figure 3 | Calculated turbidity removals in the four parallel, second-stage slow sand filters. Error bars: +/− 1 SDEV, log removals calculated assuming a limit of detection <0.001 NTU.

The improved particle removal in filters 3 and 4 was indirectly confirmed by a more rapid headloss buildup in these filters compared to the control filter (data not shown).

The results shown in Tables 3 and 4 and Figure 3 also show that the positive effect of bauxite on the retention of bacteria and particles lasted for a long period of time. For packed bed filters receiving raw waters with a high concentration of particles, Yao *et al.* (1971) have shown that the initial positive effect of the use of positively charged filter media on the retention of particles only lasts for a relatively short period of time. In such systems the collector (media grains) is in fact rapidly covered with the negatively charged particles and the ability of such filters to remove particles (and microorganisms) from the water therefore rapidly decreases.

The observed breakthrough of *E. coli* and *Enterococcus* in filter 3 was likely the result of the continuous covering of the surface of the bauxite particles with negatively charged particles. The duration until the bacteria breakthrough occurs in a given bauxite-containing filter is site-specific and a function of the overall particle load entering the filter, the bauxite layer depth, the hydraulic loading, the specific surface charge of the natural bauxite and potentially the content of natural organic matter as well as water temperature. For a given treatment chain, the optimized removal of particles ahead of the bauxite filter layer is therefore of critical importance in terms of the bauxite layer's capacity for microorganism removal. The use of bauxite in slow sand filters is therefore particularly adequate for second-stage SSFs.

In the system which was tested in the present study where two slow sand filters were operated in series, the bauxite-containing slow sand filter (second stage) received only a relatively low load of particles and the positive effect on the retention of microorganisms thus lasted for a relatively long time. In the filter containing 80 cm of bauxite, the bacteria breakthrough did not occur during the entire experiment, which lasted for over 500 days, representing over 2,400 BVs of filtered water.

For naturally occurring aluminum in certain waters, Weber-Shirk & Chan (2007) have shown the positive effect of the presence of aluminum in the influent of slow sand filters on their performance in bacteria removal and buildup of the schmutzdecke. This is likely the result of the favorable surface properties of the accumulated aluminum particles in the top part of the slow sand filters leading to enhanced removal of bacteria.

A potential drawback of the use of bauxite for SSF might be the leaching of dissolved aluminum. Therefore, dissolved aluminum was analyzed in several effluent samples of filter 4. The US EPA has set a secondary (non-mandatory) MCL for aluminum of 0.05–0.20 mg/L for drinking water.

During one sampling campaign (September 3, 2013), different heavy metals were also measured in the effluent of filter 4. The results are shown in Tables 5 and 6.

Dissolved aluminum in the effluent of the filter containing 100% natural bauxite was always below the detection limit (<5 μg/L) and the different heavy metals could not be quantified (below the detection limit) or only at very low concentrations (cobalt). Consequently, the presence of dissolved aluminum in the effluent of the filter containing

Table 5 | Dissolved aluminum concentrations in the effluent of filter 4 (100% bauxite)

Date	Time since startup (days)	Concentration [μg/L]
11.06.2013	5	<5.0
03.09.2013	90	<5.0
09.12.2013	188	<5.0
17.06.2014	378	<5.0

Table 6 | Concentrations of aluminum and heavy metals in the effluent of filter 4 (September 3, 2013 – Day 90 since startup)

Substances (dissolved)	Concentration [μg/L]
Aluminum	<5.0
Chrome	<0.5
Manganese	<1.0
Cobalt	0.7 ± 0.07
Nickel	<0.5
Copper	<0.5
Zinc	<5.0
Arsenic	<1.0
Cadmium	<0.1
Lead	<0.5

natural bauxite was not an issue of concern under the conditions tested in the present study.

CONCLUSIONS

Based on the experimental results presented and the theoretical considerations on particle retention in packed bed filters, the following conclusions can be drawn:

1. The use of natural bauxite represents a considerable improvement of the performance of slow sand filters under certain circumstances compared to filter media such as quartz sand. The use of such filter media in a second-stage slow sand filter substantially improved the removal of bacteria, i.e. *E. coli* and *Enterococcus*, and inorganic particles measured as turbidity over a long period of time (>500 days).
2. The better performance of bauxite is likely the result of the lesser negative surface charge of natural bauxite compared to quartz sand leading to an increase in the collision efficiency factor, α.
3. As a consequence of the improved retention of particles (and microorganisms), headloss buildup may increase in such filters and this aspect needs further investigations and has to be considered carefully for practical applications.
4. Dissolved aluminum in the effluent of the filter containing natural bauxite was not of concern under the tested conditions. However, this issue has to be taken into account in any potential application, in particular because of the pH-dependency of dissolved aluminum.
5. For over 500 days of continuous operation, the MSF pilot unit used in this study (two roughing filters followed by one standard SSF and one SSF containing 100% bauxite) allowed production of drinking water using a raw water of poor quality and a treatment process using no electrical energy and no chemicals. This is of particular relevance for small system applications and in situations where robust treatment systems are required.
6. The improved removal of *E. coli* and *Enterococcus* in slow sand filters containing natural bauxite is of fundamental importance regarding the role of microbiologically safe drinking water for public health worldwide. Bauxite is readily available in many parts of the world and relatively inexpensive. Because MSF at large scale or at point-of-use level is a particularly adapted treatment process for remote locations and developing countries, the use of natural bauxite in slow sand filters may lead to a general improvement of the sanitary conditions in such circumstances.

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