


## Modification of slow sand filtration of secondary effluents by electric fields: electrokinetics and influence factors

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### ABSTRACT

In this study, the effect of electric fields applied to a quartz sand bed on electrokinetics and the disinfection performance of an E-slow sand filter (reduction of *Escherichia coli*) were investigated. Results of electrokinetic experiments have shown that coliforms are electroosmotically transported to the cathode with a velocity of  $0.6 \text{ cm h}^{-1}$  ( $E = 4 \text{ V cm}^{-1}$ ). In the absence of an electric field and at electric field strengths of  $<4 \text{ V cm}^{-1}$ , a diffuse random movement to the cathode and the anode was the only translocation mechanism. Hence, the electroosmotic transport may represent an additional component of motion during E-slow sand filtration influencing its disinfection performance. Based on this, comparative experimental investigations on the disinfection performance of a slow sand filter without and combined with an electric field were carried out. In the absence of an electric field, the adsorptive filter effect of quartz sand contributes to an initial bacterial retention by 0.8 log levels. Applying an electric field strength of  $4 \text{ V cm}^{-1}$  caused an increase of *E. coli* reduction up to 4 log levels. The electrokinetically enhanced accumulation of *E. coli* in the filter bed is assumed to be the major effect mechanism in the E-slow sand filter.

**Key words:** electric field, electrokinetically enhanced accumulation of *E. coli*, electroosmotic transport, E-slow sand filtration, wastewater reclamation

### HIGHLIGHTS

- E-slow sand filtration seems to be very promising to overcome the limits/drawbacks of common slow sand filters, e.g., the insufficient disinfection performance, long ripening periods, high space requirements, etc.
- The accumulation of *Escherichia coli* in a filter bed of quartz sand can be electrokinetically enhanced.
- A high disinfection performance (4 log levels) is immediately achieved ( $E = 4 \text{ V cm}^{-1}$ ).

### INTRODUCTION

According to estimates of the United Nations Environment Programme (UNEP), more than 1.8 billion people will be living in countries or regions with absolute water scarcity by 2025 (UNEP 2007). The pressure on water resources is increased not only in arid and semiarid regions but also in fast-growing megacities around the world. Since the beginning of the 20th century, the planned/controlled reuse of wastewater (WW) has developed into a central task of the sustainable water resource management. Activities in research and development currently focus on decentralized and semi-centralized concepts since their structures offer better conditions for the establishment of recycling systems, and innovations in WW technology can be implemented more easily.

The hygienic quality requirements for WW reuse are predominantly oriented towards the planned usage. These are, in turn, regulated by thresholds and guidance values, e.g., for faecal indicator bacteria (e.g., faecal coliforms: *Escherichia coli*), in widely differing norms and legal provisions specific to the respective countries. Due to the infectious risk caused by a multitude of pathogens that are still contained in WW after mechanical–biological treatment, specific disinfection methods are indispensable for their satisfactory reduction.

In general, slow filtration represents a simple and low-maintenance process (low operating and investment costs, and low energy consumption) for improving the hygienic parameters of reclaimed WW. As the most common form of slow filtration,

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slow sand filtration effectively separates particles and pathogens by combining biological, physical, and chemical processes (Logsdon *et al.* 2002). It represents an essential part of drinking water treatment since it is the only effective method for eliminating pathogenic protozoa, e.g., *Giardia* spp. and *Cryptosporidium* spp. Furthermore, pathogenic bacteria can be adsorbed and eliminated by predation. In studies on disinfection of surface water and WW (sand depth: 0.38–1.35 m; effective sand size: 0.3–0.6 mm; hydraulic loading rate: 0.12–0.4 m h<sup>-1</sup>), the reduction of total coliforms ranged from 0.3 to 3.5 log levels, and the reduction of enterococci was up to 2.6 log levels (Bellamy *et al.* 1985; Ellis 1987; Farooq & Alyousef 1993; Sadiq *et al.* 2003). However, the insufficient disinfection performance of slow sand filters often results in exceeding the thresholds for process and irrigation water. Downstream slow sand filters are permanently water-saturated and a level of supernatant water rests above the filter bed. However, a ripening period of several weeks to several months is required to ensure a constant disinfection performance (Stevik *et al.* 1999; Campos *et al.* 2002; Sánchez *et al.* 2006; Bauer *et al.* 2011). This is a substantial disadvantage for decentralized WW reclamation since filtered WW is strongly limited in use within this time due to the residual risk of infection. To ensure a constant maximum disinfection performance over a long period of time, and thus a minimum of maintenance efforts, a supernatant level of up to 2 m is permitted (Amy *et al.* 2006). The filter bed height can amount up to 1.5 m. The resulting high place requirement for slow sand filters represents a further drawback concerning its implementation into an extended decentralized WW treatment.

Nevertheless, knowing that slow sand filtration is a simple, comfortable, robust, and low-cost/low-maintenance method for WW reclamation, an innovative modification of this process is very promising for overcoming the limits/drawbacks discussed above. Thereby, the combined use of electric fields and slow sand filtration might be a solution in regard to the insufficient disinfection performance and the high space requirement of common slow sand filters. Most bacteria possess a small surface charge due to ionizable groups on the bacterial cell surface resulting in an amphoteric character (De Flaun & Condee 1997; Desai & Armstrong 2003). Therefore, the cell surface carries a net negative charge under weak and strong basic conditions. Under strong acidic conditions, the cell surface is charged positively. Thus, bacteria can migrate to the oppositely charged electrode (electrophoretic movement). Moreover, bacteria can also be transported by electroosmosis, the creation of pore water flow along charged surfaces (Wick *et al.* 2004; Shi *et al.* 2008). Thus, the electrophoretic movement or the electroosmotic transport of coliforms may represent an additional component of motion that influences their probability of deposition on collector surfaces in the filter media. As a result of this, the efficiency of the separation process could be enhanced.

In recent decades, extensive studies were conducted to investigate new applications of electric energy in the field of mass transfer and phase separation processes. The use of electric fields for the separation of particulate contaminations (aerosols) to clean exhaust gases has already been the state of the art for many years (Kraemer & Johnstone 1955; Nielsen & Hill 1976; Shapiro *et al.* 1983). The transfer of this technology to aqueous systems (solid–liquid separation) is mainly limited to sludge dewatering and special types of surface filtration technologies (Moulik 1971; Lockhart 1983; Bollinger & Adams 1984; Ptasinski & Kerkhof 1992; Hofmann & Posten 2003). The removal of particles from aqueous systems by electrofiltration was studied by Judd & Solt (1991) and Zhang *et al.* (2000). Judd & Solt (1991) examined the electrophoretically supported depth filtration using different fibre materials. Thereby, the collector efficiency was increased linearly with the electric field strength. However, a quantitative agreement with the classical theory for electrostatically augmented aerosol deposition was poor. Zhang *et al.* (2000) demonstrated the important role of an electric field applied to a conductive and non-conductive granular filter bed to increase the particle collection. Li *et al.* (2009) studied the removal efficiency of turbidity from drinking water by electrofiltration on a pilot scale. In general, an improved removal of particles was observed in the presence of an electric field. At an electric field strength of 4.8 V cm<sup>-1</sup>, the particle removal was increased by nearly 0.4 log levels. This effect was mainly observed for particles ≤4 μm corresponding to the size range of pathogens (mainly bacteria). Thus, the results of this study illustrate the potential removability of microorganisms by electrofiltration. However, the modification of depth filtration by electric fields to disinfect WW has hardly been investigated, previously. Very few studies were performed using model solutions. Only Kulkarni *et al.* (2005) have explicitly shown that the removal of the water-borne pathogen *Cryptosporidium parvum* increased from 10 to 70% due to the electric field applied to fine sand media and from 30 to 96% using magnesium oxide. Wick *et al.* (2010) has further observed that the composition and physiology of microbial communities were not influenced by unpulsed electric fields. Until today, the disinfection mechanism of E-depth filtration has not been understood well. It has become apparent that systematic investigations on the modification of slow sand filtration by electric fields are required to assess its applicability to the disinfection of secondary effluents.

Against this background, the subject of this study was to achieve a fundamental understanding of how electric fields applied to a quartz sand bed affect the filter performance in view of hygienic parameters (coliforms, *E. coli*). Thereby, possible mechanisms of action, in particular,

- an electrokinetically improved accumulation of coliforms in the filter medium (an increase of the collector efficiency),
- an irreversible inactivation of coliforms by electrochemically produced free chlorine, and
- an irreversible inactivation of coliforms under acidic or basic pH conditions as a result of the electrolytic water decomposition

were examined. Derived from this, influence factors on the disinfection performance of an E-slow sand filter (reduction of *E. coli*) were analyzed and assessed. For this purpose, two experimental set-ups – an electrokinetic apparatus and a downstream E-slow sand filter – were used. Thereby, the electrokinetic apparatus was utilized to investigate which electrokinetic phenomenon occurs if an electric field is applied to a quartz sand bed. This information is essential to understand and to assess how electrokinetics may enhance the filtration performance. The *E. coli* reduction was examined using a downstream slow sand filter without and combined with an electric field.

The results of this study provide essential information to assess the combined application of electric fields and slow sand filtration for the disinfection of secondary effluents as a part of WW reclamation.

## MATERIALS AND METHODS

### Reagents, chemical, and microbiological analyses

All chemicals used were of reagent grade and could be applied without further purification.

Anion concentrations were measured by an ion chromatograph DX 500 (DIONEX Co., USA) equipped with a conductivity detector and an IonPac AS 19 column ( $2 \times 250$  mm, eluent 20 mM KOH, flow rate  $0.25 \text{ mL min}^{-1}$ ).

The dissolved organic carbon (DOC) concentration was measured with the device TOC-V<sub>CPN</sub> (Shimadzu, Japan).

The photometric determination of free chlorine and total oxidants was carried out by means of *N,N*-diethyl-*p*-phenylenediamine (DPD) method (EN ISO 7393-2) utilizing the UV/VIS photometer Cary 50 Series (Varian Inc., USA). The EN ISO 7393-2 describes the determination of free chlorine and total chlorine at pH values ranging from 6.2 to 6.5. In the case of total chlorine, iodide ions were added to the DPD reagent. These ions reduce chlorine substitution products to free chlorine, so that they are also captured. However, this method proved to be unselective in the presence of other oxidants, such as hydrogen peroxide or peroxodisulfate, being also able to oxidize iodide ions to iodine. Besides chlorine, iodine can also react with the DPD reagent to a red dye. As these and other by-products can occur during the electrochemical disinfection of water, the results of this photometric method were designated as the concentration of total oxidants.

The zeta potential was determined with a Zetaziser Nano ZS (Malvern Instruments, UK) using the Laser Doppler Measurement and the Light Phase Analysis (M3-PALS).

The standard deviations of the analytical methods applied were found to be 5% for the DPD method, 5% for the determination of DOC, 10% for the anion analyses (e.g., chloride, etc.) by ion chromatography, and 5% for the determination of the zeta potential.

The quantitative determinations of *E. coli* and total coliforms were conducted by means of the Colilert-18/Quanti-Tray procedure (IDEXX, USA).

Triplicate measurements of disinfection and reactivation experiments exhibited a good reproducibility of the microbiological methods within a 95% confidence interval.

### Preparation of the coliform (*E. coli*) stock suspension and spiking biologically treated WW or model water

In order to adjust a stable initial bacterial concentration in each assay, it was essential to cultivate coliforms from the sewage and then spike the water with them. The proportion of *E. coli* within the total of coliforms amounted to approximately 50%.

To this end, 0.1 mL of a fresh sewage sample (WW treatment plant, Dresden, Germany) was spread on selective agar for coliform germs (Mac Conkey Agar, Merck, Germany) and incubated for 20 h at 37 °C. Some colonies were collected through an inoculating loop and inserted into a nutrient solution consisting of pancreatic peptone and meat (SIFIN, Germany). This solution was kept in an incubator for 5 h at 37 °C. The test culture thus created was mixed with glycerine, filled in Greiner tubes, and stored at  $-70$  °C. The working culture was produced by adding 1 mL of the test culture to 50 mL of the nutrient broth and incubating it for 5 h at 37 °C. Thereafter, 0.1 mL of this solution was spread to an agar plate and grown to a

bacterial layer at 36 °C for 24 h (DEV nutrient agar). This layer was washed with a 5 mL solution prepared with 8.5 g of NaCl and 20 mL of phosphate buffer (pH = 7.3). This final suspension had a durability of 7 days and contained from 1 to  $9 \times 10^9$  CFU/mL.

For the experimental investigations on slow sand filtration without and combined with electric fields, the bacteria concentrate was spiked to the biologically treated WW or to the model water (MW) to obtain an initial concentration of approximately  $1\text{--}3 \times 10^5$  *E. coli* per 100 mL and  $2\text{--}5 \times 10^5$  total coliforms per 100 mL representing the maximum (faecal) coliform concentration in biologically treated WW.

Investigations on the electrokinetic transport of coliforms were conducted with an initial concentration of approximately  $1\text{--}2 \times 10^8$  *E. coli* per mL and  $2\text{--}4 \times 10^8$  total coliforms per mL, respectively.

## Experimental procedures

### Electrokinetic transport of coliforms

The electrokinetic apparatus was used to examine which electrokinetic phenomenon occurs if an electric field is applied to a quartz sand bed. In this way, the electrokinetic transport of coliforms in the pore water of a quartz sand bed could be studied. The electrokinetic apparatus consisted of two electrode chambers ( $V = 0.0753$  L), a lid-covered compartment ( $V = 0.405$  L, lid with sample ports in distances of 3, 4.5, 6, 9, and 12 cm from the centre (0 cm) towards the cathode and the anode) filled with quartz sand, and a bypass channel below the compartment with quartz sand having a hydraulic contact with both electrode compartments to exclude bacterial transport by advective hydraulic water flow (Figure 1).

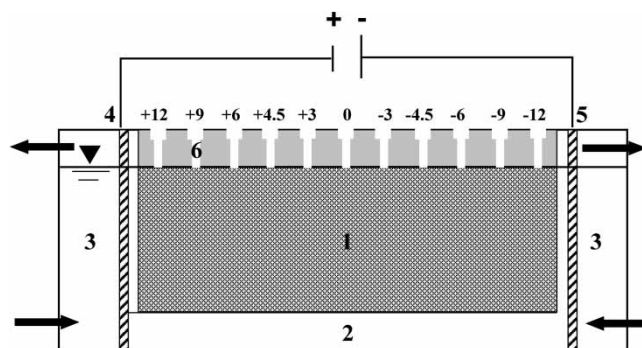
Stainless steel electrodes (De Nora GmbH, Germany) with an effective area of  $77.2$  cm<sup>2</sup> (a lattice factor of 1.8) were integrated into the electrode chambers. Furthermore, Sefar Tetex fabrics (SEFAR AG, Germany) were placed 1.7 cm from the electrodes to separate the quartz sand from the electrode chambers.

Before starting each electrokinetic experiment, the sterilized quartz sand was saturated with the liquid to be examined for 24 h. Then, 2 mL of bacterial inoculum with a concentration of  $2\text{--}4 \times 10^8$  total coliforms per mL was slowly injected for 1 min at half of the bed depth at an equal distance from the two electrodes in the centre of the compartment with quartz sand. A constant electric field was applied for 6 h. The electrolyte in both electrode chambers was recirculated to a 1 L reservoir at a flow rate of  $6.5$  mL min<sup>-1</sup>. The inflow and the outflow rates were controlled by a pump to keep the water table at a constant height throughout the experiment.

The influence of the electric field strength on the electrokinetic transport of coliforms was studied at 0, 2, 4, and  $6$  V cm<sup>-1</sup> ( $t = 6$  h) in quartz sand (0.7–1.25 mm; uniformity coefficient = 1.4) saturated with 0.05 M Tris acetate buffer solution (TA, pH = 7.6).

Sterilized TA buffer solution (free of chloride) was used (recirculated) to avoid any pH changes in the electrode chambers and in the electrokinetic chamber during the experiments. Moreover, the anodic oxidation of chloride to the germicide-free chlorine should be prevented.

The influence of the water matrix on the electrokinetic transport of coliforms was determined by using WW (pH = 7.6) and TA buffer solution ( $c = 0.05$  mol L<sup>-1</sup>; pH = 7.6) at an electric field strength of  $4$  V cm<sup>-1</sup> ( $t = 6$  h). Thereby, the absence of



**Figure 1** | Set-up for the electrokinetic experiments (after Wick *et al.* 2004). 1: quartz sand; 2: hydraulic bypass; 3: recirculation of the electrolyte (TA buffer solution or WW); 4: anode; 5: cathode; 6: sample ports (positions in cm from the injection point 0).

electrochemically produced germicides in the sand saturated with biologically treated WW was also required. Therefore, a compartment directly behind the anode chamber was filled with activated carbon (F 300, grain size: 0.3–0.4 mm) to reduce free chlorine and other oxidants. The biologically treated WW was taken from a municipal sewage treatment plant in Dresden. Selected chemical parameters are summarized in Table 1.

At the end of each experiment, pore water samples were taken simultaneously at the sample ports. The pH value, the coli-form concentration, and the concentration of electrochemically produced free chlorine and total oxidants were determined.

### Slow sand filtration without and combined with electric fields

The reduction of *E. coli* in biologically treated WW (pH = 7.6;  $c_{\text{chloride}} = 110 \text{ mg L}^{-1}$ ) and in a MW (free of chloride; pH = 7.0;  $\text{NaNO}_3$  and  $\text{Na}_2\text{SO}_4$  were added to distilled water to adjust the conductivity) was examined using a slow sand filter without and combined with an electric field (laboratory scale, Figure 2).

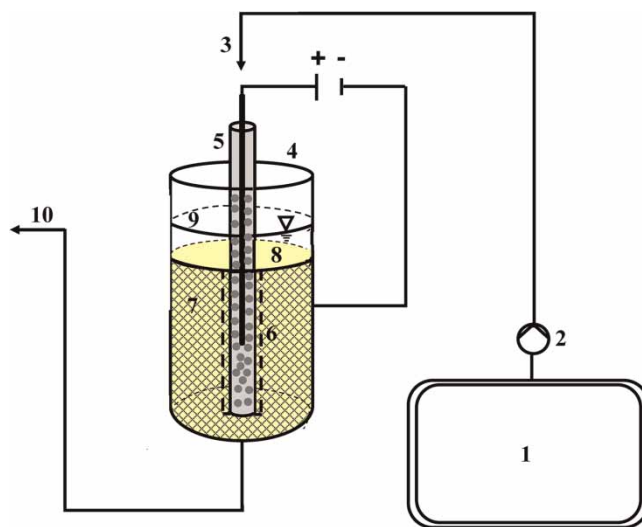
Selected chemical parameters of the WW and the MW used in this study are summarized in Table 1.

The water passed the filter column from the top (supernatant) to the bottom. The supernatant water level was almost constant (4–5 cm). The anode consisted of a graphite rod that was integrated into granular activated carbon (F300; 0.3–0.4 mm; Aquatec Jünger GmbH, Germany). A stainless steel mesh represented the cathode material. Thereby, the granular activated carbon as a part of the anode was surrounded by a Sefar Tetex fabric (SEFAR AG, Germany) being permeable to water, but impermeable to particles in order to avoid its release to the quartz sand bed. To prevent the electrolysis of the supernatant

**Table 1** | Chemical parameters of the applied secondary effluent and MW

Chemical parameters	WW	MW
Conductivity in $\mu\text{S cm}^{-1}$	1,626	1,007
pH	7.6	7.0
DOC in $\text{mg L}^{-1}$	8.9	< 0.1
$c_{\text{chloride}}$ in $\text{mg L}^{-1}$	110	< 0.1
$c_{\text{nitrate}}$ in $\text{mg L}^{-1}$	136	40
$c_{\text{nitrite}}$ in $\text{mg L}^{-1}$	< 0.1	< 0.1
$c_{\text{phosphate}}$ in $\text{mg L}^{-1}$	0.3	< 0.1
$c_{\text{sulfate}}$ in $\text{mg L}^{-1}$	100	400

DOC, dissolved organic carbon; WW, biologically treated wastewater; MW, model water.



**Figure 2** | Experimental set-up for the E-slow sand filtration. 1: reservoir; 2: pump; 3: inlet; 4: E-slow sand filter; 5: anode (graphite rod integrated in activated carbon); 6: Sefar Tetex fabric; 7: cathode (stainless steel mesh); 8: filter bed; 9: supernatant water; 10: outlet.

water, the height of the Sefar Tetex fabric (SEFAR AG, Germany) was equal to the height of the filter bed. The part of the anode, which rose up to the supernatant water, was closed, so that no electric field could be produced there (Figure 2). The adjustment of a constant electric field strength was realized by setting a constant voltage on the filter bed using a power supply (EA PS 8360-10 DT, Elektro-Automatik GmbH & Co. KG, Germany). The electric field was applied perpendicular to the flow direction. The height of the filter column was 32.5 cm. The diameter amounted to 9 cm. The height of the filter bed, which consisted of quartz sand (0.7–1.25 mm; uniformity coefficient = 1.4), was 18 cm.

The *E. coli* reduction in biologically treated WW (pH = 7.6) was examined by slow sand filtration without and combined with an electric field. Experimental examinations with an electric field were conducted at  $2\text{--}4\text{ V cm}^{-1}$ , whereby the effective retention time in the filter bed was 3.9 h.

To gain a fundamental understanding of the effective mechanism for *E. coli* reduction by slow sand filtration combined with electric fields, the pH values of the E-filter inflow and outflow were measured. Furthermore, the survival rate of *E. coli* and total coliforms in biologically treated WW at different pH values ranging from 2 to 12 (adjusted by 1 M NaOH or 1 M HCl) was determined within an experiment duration of 1–7 h. Moreover, the concentrations of free chlorine and total oxidants were determined after each assay. A possible temporary local germicidal effect of electrochemically produced free chlorine before having been reduced on activated carbon (part of the anode) was further examined. For this purpose, the chloride-free MW and chloride-containing WW ( $c_{\text{chloride}} = 110\text{ mg L}^{-1}$ ) spiked with *E. coli* were filtrated by E-slow sand filtration at an electric field strength of  $4\text{ V cm}^{-1}$  ( $t_{\text{effective}} = 3.9\text{ h}$ ).

During each assay, the electric current was measured and the corresponding electric charge input  $Q_V$  (normalized to the pore water volume of the filter bed) was calculated.

## RESULTS AND DISCUSSION

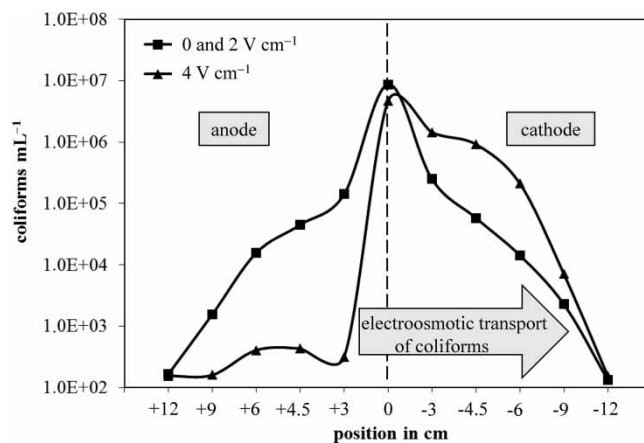
### Electrokinetic phenomena

#### Influence of the electric field strength

In order to examine the influence of the electric field strength on the electrokinetic transport of coliforms in the pore water of quartz sand, it was necessary to keep the pH value constant (constant zeta potential) and to ensure the absence of electrochemically produced free chlorine. Hence, the experiments were conducted with a chloride-free TA buffer solution ( $c = 0.05\text{ mol L}^{-1}$ ) at pH = 7.6 (typical pH of biologically treated WW).

In the absence of an electric field and at electric field strengths of  $<4\text{ V cm}^{-1}$ , no directed movement of coliforms occurred. In this case, a diffuse random movement to the cathode and the anode was the only translocation mechanism.

In the presence of an electric field ( $E \geq 4\text{ V cm}^{-1}$ ), the transport of coliforms to the cathode is clearly dominated by electroosmosis at  $E \geq 4\text{ V cm}^{-1}$  (Figure 3). No electrophoretic movement of negatively charged coliforms to the anode was observed.



**Figure 3** | Electrokinetic distribution of coliforms in quartz sand ( $d = 0.7\text{--}1.25\text{ mm}$ ,  $d_{10} = 0.83\text{ mm}$ , uniformity coefficient = 1.4) saturated with 0.05 M TA buffer solution (pH = 7.6) ( $E = 0, 2, 4\text{ V cm}^{-1}$ ,  $t = 6\text{ h}$ ; results at  $E = 0$  and  $2\text{ V cm}^{-1}$  showed no difference, all experiments were conducted separately).

This is in contrast to the findings by De Flaun & Condee (1997) showing a unidirectional movement of four different motile and non-motile strains to the anode only. Our observations, however, are in good agreement with the findings by Wick *et al.* (2004) and Shi *et al.* (2008) and can be explained by the zeta potential of the coliforms and the quartz sand affecting the electrokinetic mobility of bacteria. The overall electrokinetic mobility of bacteria is the resultant of their electrophoretic and electroosmotic mobility (Shi *et al.* 2008). The electrophoretic mobility is directly proportional to the zeta potential of the coliforms. The electroosmotic mobility is directly proportional to the zeta potential of the quartz sand used in this experiment. Both showed a negative zeta potential at pH = 7.6. For coliforms, one signal was observed at  $-20.4$  mV and one at  $-3.8$  mV (Table 2). The quartz sand showed a zeta potential signal at  $-47.7$  mV. Thus, the doubled to nine-fold higher fixed negative surface charge of the sand, together with the positively charged ions in the Stern double layer, caused an electroosmotic flow of the solution to the cathode. The electroosmotic mobility superimposes the electrophoretic mobility. Therefore, the mean electrokinetic transport velocity of  $0.6$  cm h<sup>-1</sup> was dominated by electroosmosis. This electroosmotic velocity was also observed for polycyclic aromatic hydrocarbon-degrading bacteria examined by Shi *et al.* (2008) and Suni & Romantschuk (2004). The mean electroosmotic mobility amounted to  $0.15$  cm<sup>2</sup> (Vh)<sup>-1</sup>.

Increasing the electric field strength from  $4$  to  $6$  V cm<sup>-1</sup> resulted in a strong temperature rise to  $60$  °C. This effect can be explained by the electrical resistance of the quartz sand. As such high temperatures are undesirable for the operation of the E-slow sand filter in practice, the electroosmotic transport of coliforms at  $\geq 6$  V cm<sup>-1</sup> was not examined any further.

### Influence of the water matrix

The water matrix is an important influence factor on the electrokinetic transport of bacteria. The more complex it is, the more likely is the fact that electrochemical reactions may cause changes influencing the electrokinetic transport of coliforms. Investigations on the influence of the water matrix also required the absence of electrochemically produced germicides in the sand saturated with biologically treated WW. No free chlorine and a marginal concentration of total oxidants of  $\leq 0.03$  mg L<sup>-1</sup> were observed directly behind the compartment filled with activated carbon and in the pore water of the whole sand bed. The results of the experimental examinations in the chloride-free TA buffer solution (pH = 7.6) were used for comparison.

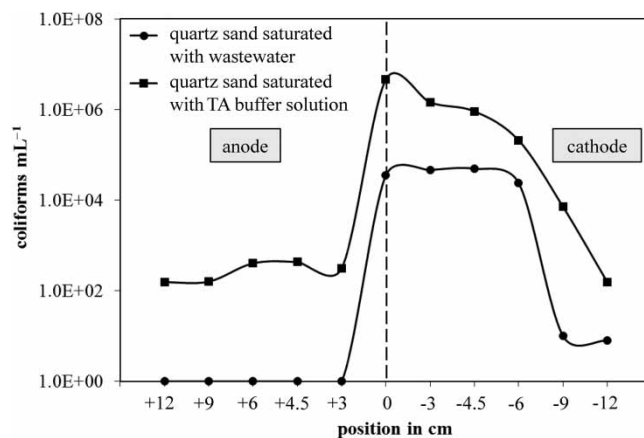
As can be seen from Figure 4, coliforms were transported with a mean electroosmotic velocity of  $0.6$  cm h<sup>-1</sup> towards the cathode, regardless of the water matrix.

However, the coliform concentration in biologically treated WW was about 1–2 log levels lower than in the TA buffer solution. This effect is the result of a strong pH gradient that developed in the pore water of sand saturated with WW (Figure 5) within the experiment duration of 6 h. Due to the electrolytic water decomposition, the pH value decreased to 3.2 near the anode and increased up to 10.3 near the cathode. A low pH value of 2.7 was also observed at the injection point. This pH value remained constant at 3 and 12 cm from the injection point towards the anode and increased from 4 to 11.3 towards the cathode. Compared to that, the pH value remained almost constant in the pore water of sand saturated with the chloride-free TA buffer solution (Figure 5).

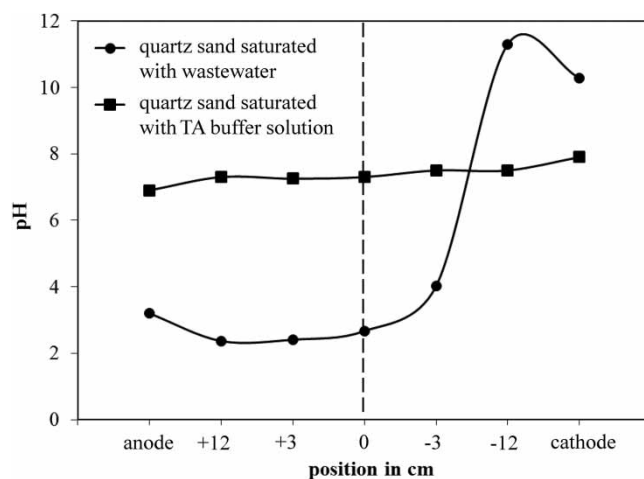
The investigations on the viability of coliforms at the observed pH values of 2.7 and 10 showed that at a pH value of 2.7, coliforms are reduced by 1.5 log levels within the experiment duration of 6 h. At a pH value of 10, however, no considerable change in the coliform concentration was observed (data not shown). The pH value influences not only the viability, but also the zeta potential and thus the surface charge of coliforms. Charged polymers on the bacterial cell surface contain ionizable groups resulting in an amphoteric character. Therefore, the cell surface carries a net negative charge under weak and strong basic conditions. Under strong acidic conditions, the cell surface is charged positively. This fact was confirmed by the zeta potentials determined at different pH conditions (Table 2).

**Table 2** | Zeta potentials of coliforms in biologically treated WW in dependence on the pH value (pH = 2.7, 4.0, 7.6, 11)

pH	Zeta potential of coliforms in mV
2.7	+2.9; +1.0
4.0	-16; -1.8
7.6	-20.4; -3.8
11	-21.0; -4.9



**Figure 4** | Electrokinetic distribution of coliforms in quartz sand ( $d = 0.7\text{--}1.25$  mm,  $d_{10} = 0.83$  mm, uniformity coefficient = 1.4) in dependence on the pore water matrix ( $\text{pH} = 7.6$ ,  $E = 4$  V  $\text{cm}^{-1}$ ,  $t = 6$  h).



**Figure 5** | pH of different pore waters in quartz sand ( $d = 0.7\text{--}1.25$  mm,  $d_{10} = 0.83$  mm, uniformity coefficient = 1.4) in dependence on the distance towards the anode and the cathode ( $E = 4$  V  $\text{cm}^{-1}$ ,  $t = 6$  h).

The changing surface charge of the coliforms in WW at varying pH conditions showed no influence on their electrokinetic transport since it is dominated by electroosmosis. However, pH values of  $<3$  have a lethal effect on coliforms. Although the electroosmotic mobility can be influenced by the pH in general, the former result indicates that the electroosmotic transport of coliforms in WW takes place in a wide pH range during the E-slow sand filtration of biologically treated WW. Thus, it could represent a possible additional component of motion in the E-slow sand filter influencing its disinfection performance.

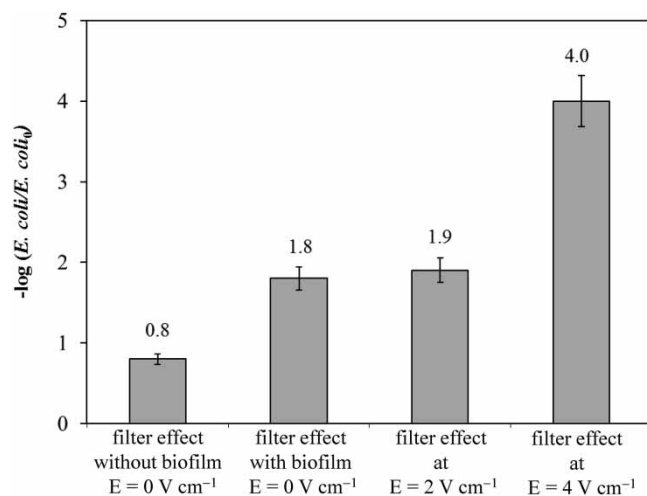
## E-slow sand filtration

### *E. coli* reduction

The contact of coliforms with a sand grain and their accumulation on it requires a bacterial transport close to the collector surface that can be influenced in the presence of an electric field. Figure 6 shows that the *E. coli* reduction is increased with increasing electric field strength.

In the absence of an electric field, the adsorptive filter effect of quartz sand contributes to an initial bacterial retention by 0.8 log levels. After a 1-month ripening period of the slow sand filter, the *E. coli* reduction has increased and stabilized to 1.8 log levels. This is due to the growing importance of the biofilm on the top and in the upper layer of the filter bed which develops with the increasing operating time of the slow sand filter without an electric field. It is well known that its high biological activity supports the removal of pathogens (Langenbach *et al.* 2010). At an electric field strength of





**Figure 6** | Log reduction of *E. coli* in biologically treated WW disinfected by slow sand filtration without and with combined electric fields ( $E = 0, 2, 4 \text{ V cm}^{-1}$ ,  $v_{\text{hydraulic}} = 0.019 \text{ m h}^{-1}$ ,  $t_{\text{eff}} = 3.9 \text{ h}$ ,  $d = 0.7\text{--}1.25 \text{ mm}$ ,  $d_{10} = 0.83 \text{ mm}$ , uniformity coefficient = 1.4).

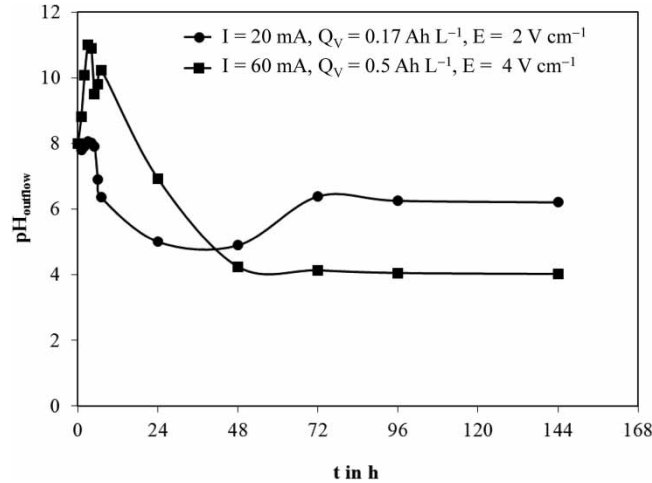
$2 \text{ V cm}^{-1}$  applied to the filter bed, the *E. coli* reduction of 1.9 log levels corresponds to the disinfection performance of a slow sand filter with an established biofilm on the filter bed (without an electric field). This result indicates that a constant bacterial reduction can be ensured immediately without any ripening period of the filter. This represents a major advantage compared to the slow sand filter without an electric field. Doubling the electric field strength to  $4 \text{ V cm}^{-1}$  also doubled the log reduction of *E. coli* up to 4 log levels corresponding to an overall enhancement of the collector efficiency by the factor of 5 (compared to the slow sand filter without a biofilm). However, the question arises if the increased *E. coli* reduction is, in fact, caused by the electrokinetically improved accumulation of the bacteria in the filter bed or rather by a temporary local germicidal effect of electrochemically produced free chlorine before having been reduced on the surface of activated carbon. Investigations on E-filtration of chloride-free MW and chloride-containing WW ( $c_{\text{chloride}} = 110 \text{ mg L}^{-1}$ ) have shown that *E. coli* bacteria were reduced to the same extent in both waters (3.8–4 log levels, data not shown). This illustrates that the enhanced disinfection performance of the E-filter is not the result of a local germicidal effect of electrochemically produced free chlorine in chloride-containing WW. Thus, these findings confirmed our hypothesis that the accumulation of *E. coli* is electrokinetically enhanced in the presence of an electric field applied to the filter bed.

### pH of the E-filter outflow

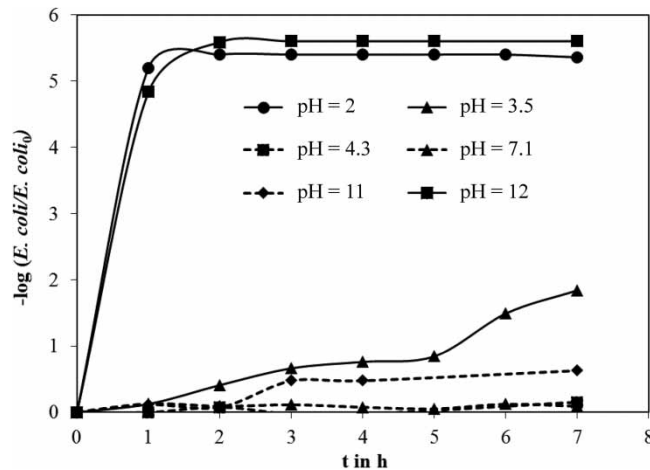
Figure 7 shows the pH value of the E-filter outflow for different electric currents/electric charge inputs (as a result of the adjusted electric field strength at 2 and  $4 \text{ V cm}^{-1}$ ) in dependence on the operation time of the E-filter.

A considerable increase in the pH value was observed within the first 5 h of E-filtration, so that basic pH conditions first dominated in the E-filter outflow. The pH value increased from 7.9 to 10.5 with a tripling of the electric current from 20 to 60 mA corresponding to an increase of the electric charge input ( $Q_V$ ) from 0.17 to  $0.5 \text{ Ah L}^{-1}$ . After 7 h, the pH value has decreased to 6.3 at an electric current of 20 mA ( $Q_V = 0.17 \text{ Ah L}^{-1}$ ). However, at an electric current of 60 mA ( $Q_V = 0.5 \text{ Ah L}^{-1}$ ), a pH value of 10 was observed. After 24 h, the pH value has decreased and stabilized after 48–72 h depending on the electric current/electric charge input. At 20 mA ( $Q_V = 0.17 \text{ Ah L}^{-1}$ ), a constant pH value of 5.8 was observed, whereas at 60 mA, the pH value was 4.3 ( $Q_V = 0.5 \text{ Ah L}^{-1}$ ). The mobility of protons (produced at the anode) under an electric field is higher than the mobility of hydroxyl ions (produced at the cathode) resulting in an acid front migrating from the anode to the cathode. The electroosmotic flow to the cathode can promote this effect. This may change the properties of the filter material with increasing running time of the E-filter resulting in a decrease of the pH value of the E-filter outflow. From the examined bacterial survival under the observed acidic and basic pH conditions (Figure 8) can be concluded that *E. coli* are viable for 1–7 h in a pH range from 4 to 11. Only at a pH value of 11, a low *E. coli* reduction was observed to a maximum of 0.4 log levels.

Therefore, acidic pH values in a range of 4–6 and basic pH conditions ranging from 8 to 11 do not contribute substantially to the enhanced disinfection performance of the E-slow sand filter at an optimum effective retention time of almost 4 h.



**Figure 7** | Time course of pH (E-filter outflow) in dependence on the electric field strength and the resulting current/electric charge input (WW,  $E = 2, 4 \text{ V cm}^{-1}$ ,  $I = 20, 60 \text{ mA}$ ,  $Q_V = 0.17, 0.5 \text{ Ah L}^{-1}$ ,  $v_{\text{hydraulic}} = 0.019 \text{ m h}^{-1}$ ,  $t_{\text{eff}} = 3.9 \text{ h}$ ,  $d = 0.7\text{--}1.25 \text{ mm}$ ,  $d_{10} = 0.83 \text{ mm}$ , uniformity coefficient = 1.4).



**Figure 8** | Time course of log *E. coli* reduction in dependence on the pH of biologically treated WW.

**CONCLUSIONS**

Slow sand filtration combined with electric fields could be an alternative to common disinfection methods applied for an extended decentralized WW treatment. In this study, valuable results were achieved for gaining a fundamental mechanistic understanding of how an electric field applied to a quartz sand filter bed affects the accumulation of coliforms, in particular *E. coli*, in the filter bed (disinfection performance of the E-filter). Cell surface of coliforms carries a net negative charge under neutral and weak basic conditions being typical for biologically treated WW. In the presence of an electric field, bacteria experience a force proportional to their net charge and the electric field strength. Thus, an electrophoretic movement of coliforms towards the anode is to be expected. However, our investigations clearly show that coliforms are electroosmotically transported to the cathode with a velocity of  $0.6 \text{ cm h}^{-1}$ . This is a result of the doubled to nine-fold higher negative surface charge of quartz sand compared to the surface charge of coliforms. The electroosmotic mobility superimposes the electrophoretic mobility. Assuming that bacteria move on fluid streamlines in the E-slow sand filter, the electroosmotic transport is an additional component of motion relative to them resulting in an increased probability of deposition on the collector surface (enhancement of the collector efficiency). Thus, the accumulation of coliforms (including *E. coli*) in the filter bed is increased significantly. This effect does not occur without an electric field. The streamlines are close to the collector

surface, but they will not be changed. Thereby, the disinfection performance of almost 2 log levels of a common slow sand filter with a ripening period can be increased immediately up to 4 log levels by an electric field applied to the filter bed. The achieved disinfection performance corresponds to common disinfection methods such as chlorination, UV, or membrane processes. A germicidal effect of electrochemically produced germicides (mainly free chlorine) can be excluded due to the special constructive design of the E-slow sand filter. Moreover, the observed acidic and basic pH values do not contribute to the *E. coli* reduction in the E-slow sand filter. Hence, the electrokinetically enhanced accumulation of faecal coliforms in the filter bed can be seen as the major effect mechanism in the E-slow sand filtration of secondary effluents. The maximum disinfection performance is achieved by an electric field (perpendicular to the water flow direction) with a field strength of  $4 \text{ V cm}^{-1}$  on a filter bed of quartz sand with a grain size range of 0.7–1.25 mm and an effective retention time of almost 4 h. However, this disinfection method is limited by the voltage that has to be applied and the distance between the anode and the cathode. These aspects have to be considered for up-scaling of the E-filter. The higher the distance between the anode and the cathode, the higher the voltage to adjust the electric field strength. This results in an increase of the electric resistance that causes rising temperatures of the WW inside the filter column. To avoid this, the distance between the anode and the cathode should be  $\leq 4 \text{ cm}$  and the applied voltage should be  $\leq 16 \text{ V}$ . Alternatively, this concept could be implemented by a cuboid-shaped E-filter, in which several rectangular mesh electrodes integrated in the quartz sand filter bed are connected in parallel. An advantage of this set-up is that the total current is split into the respective compartments, in which the electric field is created, and thus minimized. Moreover, a compromise between the disinfection performance and the resulting pH of the E-filter outflow should be made for the E-slow sand filter operation in practice since not only the microbial contamination but also the pH value represents an important WW specific parameter being defined in legal provisions. The pH value of reclaimed WW is recommended to be in a range of 6–9. Due to the resulting pH value of 4 at  $E = 4 \text{ V cm}^{-1}$ , an additional neutralization step might be required. If an electric field strength of  $< 4 \text{ V cm}^{-1}$  is applied, germicides have to be electrochemically produced to achieve a complete disinfection at moderate pH values which meet legal regulations. The E-slow sand filter should be operated at low maintenance by using a backwash system that has to be adapted to the special requirements. Moreover, the high integration capability in existing decentralized WW treatment plants is advantageous since the required space can be minimized.

In general, not only the bacterial accumulation in the filter bed but also the removal of other colloids may be enhanced by an electric field resulting in an increased quality of reclaimed WW. Long ripening periods for the stabilization of the disinfection performance of the filter are not required.

The modification of slow sand filtration by an electric field makes an important contribution to the further development of alternative disinfection methods required for WW reuse. Thereby, the greatest challenge is to realize a technically simple, robust, and easily implementable system with a reliably high disinfection performance (without the use of harmful chemicals). In contrast to the cost-intensive and technically extensive membrane filtration, the new E-slow sand filter meets all these requirements.

However, long-term field studies are indispensable to ensure low-maintenance long-term operation in practice and to finally assess this technology, especially in view of its economic efficiency.

## ACKNOWLEDGEMENTS

The authors thank the German Federation of Industrial Research Associations (AiF) for the financial support of this project.

## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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First received 29 September 2021; accepted in revised form 8 May 2022. Available online 8 June 2022