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Removal of anionic detergents from water and treatment of gray water by micelle–clay composites

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ABSTRACT

Shortage of water has resulted in attempts to purify contaminated water. This study demonstrates removal of anionic detergents from synthetic water, well water, and gray water (GW), and the removal of pathogenic microorganisms from GW. The complex of montmorillonite with micelles of the organic cation, octadecyltrimethylammonium, was expected to yield efficient adsorption of anionic detergents and microorganisms on account of its large surface area, positive charge, and existence of large hydrophobic domains. The removal of anionic detergents, total suspended solids, and bacteria was tested by column filters packed with micelle/montmorillonite complexes mixed with excess sand. Efficient removal of anionic detergents was demonstrated for synthetic water (130 and 30 ppm), well water (from 1.35 to below 0.3 ppm) and from two domestic laundries. A model that considers convection, adsorption, and desorption yielded adequate simulations and predictions for the above kinetics of filtration. GW from showers and sinks included several million of fecal coli per 100 mL, indicating health hazard in using the water without proper treatment. The second column of two in series removed all pathogenic bacteria after the passage of 30 L, whereas activated carbon yielded poor removal. The micelle–clay filter can adequately purify such GW for reuse in irrigation and toilet flushing.

Keywords: Micelle–clay complex; Gray water; Anionic detergents; Water purification

1. Introduction

An *anionic detergent* includes a hydrophilic negatively charged head group, which can be carboxylate, sulfate, phosphate, or sulfonate, and a hydrophobic long chain of hydrocarbons. Anionic detergents or surfactants are used by laundries, car washes, dry cleaning, soaps, and dishwashing. As a result of extensive use, considerable amounts of anionic surfactants are released into the environment, causing pollution of surface and ground water, rivers, and seawater [1]. The US EPA has set 0.5 ppm as the maximum contamination level (MCL) of anionic detergents in drinking water, whereas recently the Israeli Ministry of Health set the MCL as 0.3 ppm. Currently, the main treatment of detergent contamination in well water and groundwater is by filtration employing activated carbon. This treatment, that is highly effective in filtering many other pollutants, is relatively less efficient in the removal of anionic surfactants. Membrane bioreactor (MBR) technology was used for the removal of anionic surfactants in wastewater treatment and especially for reuse in decentralized applications; 97% removal was reported for gray water (GW) treatment in Morocco [2].

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1.1. Gray water

Shortage of freshwater resources is an increasing concern, particularly in arid and semi-arid regions. There is a demand to develop alternative water resources and to maximize water usage, e.g. to reclaim domestic and industrial wastewater [3]. Many countries have initiated wastewater reclamation as part of their overall water management plan. For example, in Israel, more than 75% of its treated wastewater is reclaimed for agricultural irrigation [4]. GW amenable for treatment is the domestic wastewater from sinks, showers, baths, and laundries. GW recycling is often undertaken with little or no treatment, but this practice could be a potential health and environmental hazard [5,6].

Proper treatment of GW is essential prior to its reuse. Decentralized wastewater systems, often called "on-site" systems, treat wastewater close to the source, typically providing treatment for individual homes, several homes, or businesses. These systems rely, at least in part, on biological treatment, such as constructed wetlands and biofilters [7]. If appropriately managed, decentralized domestic waste water-treatment systems can effectively treat wastewater, safeguard human health and the environment and can be an integrated component of the wastewater infrastructure [7,8]. Treatment of limited sources can be challenging, because significant variations in both influent quantity and quality can occur over short periods of time [9]. Variations occur in pH, temperature, and concentrations of organic matter, and pathogenic microorganisms and can adversely affect the efficiency of treatment processes. Meeting the standards for unlimited reuse of treated GW requires disinfection [10]. Yet, disinfection of treated water from such systems is often a challenge as it has to be efficient, reliable, low cost, low risk, and easy to operate and maintain. Recently chlorine and UV radiation are used [11]. Chlorination is usually efficient, reliable for most pathogens but being a strong oxidizer, it should be handled with care. Moreover, the application rate changes according to water quality that may be costly to control in on-site systems and may produce toxic byproducts such as halomethanes. Disinfection by UV is safer to handle but requires maintenance and does not have residual effect as chlorine. Therefore, there is a need for the development of disinfection alternatives for onsite treatment.

Dalahmeh et al. [12] tested four different filter materials for GW treatment, pine bark, activated charcoal, polyurethane foam, and sand. The bark and activated charcoal proved to be the most efficient at removal of anionic detergents and bacteria. The effectiveness of bark and charcoal filters was largely attributed to biofilm development and biological degradation. Bani-Melhem and Smith [13] used a submerged MBR to treat GW, which removed efficiently anionic surfactants and coliforms.

The current study investigates the possibility to purify drinking water and GW from anionic detergents and bacteria using column filters based on the micelle–clay system, which have been effective in filtering pollutants. The micelle–clay complex is formed by interacting clay particles with micelles of an organic cation, which is characterized by low critical micelle concentration and spontaneously creates nano-scale micelles in water. These micelles have a structure similar to detergents, in which the outer part contains the positively charged cation head group in direct contact with water, and a hydrophobic core. This structure provides preferential adsorption medium for many organic pollutants, both anionic and neutral. The positively charged micelles adsorb well on a clay that has a negative charge, resulting in a spontaneous formation of a complex consisting of a large number of micelles and clay layers, which in our preparation were loaded at 150% of the cation exchange capacity of montmorillonite and therefore were specifically designed to interact with anions [14]. We present a novel means for the purification of drinking water from anionic detergents and treatment of GW by using micelle–bentonite complexes [15]. We have employed two complexes based on the clay bentonite and the organic cations Octadecyltrimethylammonium (ODTMA) and Benzyldimethylhexadecylammonium (BDMHDA). We hypothesized that these complexes, being cationic detergents, could be powerful in the removal of anionic detergents from water, and also in the removal of bacteria, which carry a negative surface charge. Efficient removal of anionic detergents and bacteria by the micelle–clay complex makes it an ideal system for the purification of GW.

2. Materials and methods

2.1. Materials

Bentonite was purchased from Tolsa—Steetley UK. The organic cation ODTMA was purchased as a bromide salt from Sigma-Aldrich (Sigma Chemical Co., St. Louis, MO). Quartz sand (0.8–1.5 mm particle size) was provided by Negev Minerals Ltd. Israel. The organic cation BDMHDA (Benzyl-dimethyl hexadecyl ammonium) was purchased as a chloride salt from Fluka Chemie (Buchs, Switzerland). The anionic detergent sodium dodecyl sulfate (SDS 95%) was purchased from Sigma, Israel. Nonwoven polypropylene geo textile filter was from Markham Culverts Ltd, Papua-New Guinea.

Structural formulas of ODTMA, BDMHDA, and SDS are shown in Fig. 1.

Calgon-type granular-activated carbon was purchased from Calgon-Chemviron Carbon Corporation, Pittsburgh, Pennsylvania, USA.

2.2. Micelle–clay complex preparation

The complex was prepared by dissolving 4.7 g of the salt of organic cations ODTMA, or BDMHDA in 1 L of deionized water for 24 h by stirring 12 mol of organic cation per liter; 10 g of bentonite clay per liter was added to the solution and stirred for another 72 h [14,15]. In later experiments (with GW), concentrations of ODTMA-Br and clay were increased by 50% to 7.1 g and 15 g per 1 L, and the mixing time was reduced to 24 h. Incubation was in a 10 L bucket with Heidolf laboratory mixer. The complex was partially dried with a vacuum pump in a Buchner funnel as a "cake" with moisture content of 70–80% w/w. The resulting product was frozen for 24 h and later freeze-dried in a lyophilizer. The dry complex was ground to powder and sieved through 250-μm mesh-size sieves (Ari Levi, Israel).

2.3. Filtration experiments

Filtration experiments were performed using glass or Perspex columns, (5 cm diameter, 25 cm long) with active layer length of 20 cm, which included the micelle–clay complex mixed with sand in excess at 1:100 ratio (w/w). For a 1:100 ratio, 6.5 g of dry ODTMA or BDMHDA complex were mixed with 644 g of sand (thoroughly washed with deionized water and dried at 105˚C for 24 h). A geotextile filter was put at the bottom of the column and covered by 2 cm layer of sand.

The column was connected to a peristaltic pump (Cole-Palmer MasterFlex), saturated initially with distilled water at a flow rate of 5 mL/min from the bottom. The volume of pores in the column was determined by passing known water quantities at slow rate through the column; for a 1:100 ratio, it was 28% of the volume of the active layer. Filtration was carried out at flow rates of 20 up to 60 mL/min, and the volumes passed varied from 1 to 300 L.

2.4. GW collection

Twenty-liters plastic containers were filled with GW from showers and sinks from a drain outlet at student dormitories. An initial solution was taken from each container for analysis. The water was filtered as previously described.

2.5. Laundry effluent collection

The GW from laundries was collected at two locations. (i) Two 40 L containers were arranged to collect the laundry machine effluent of one household in Jabel Mukaber, which was one of 7 units involved in a project of Engineers without Borders, Israel. The washing machine discharge hose was inserted into the container until it was full. The containers were filled within six days and collected two days later. After the containers were collected from the site, they were kept at −4˚C for 4 days until the beginning of the experiment. In the first stage, all effluents were filtered through a sand column to remove coarse suspended colloids. (ii)The second laundry effluent was the first wash of a domestic laundry, which used powder and fabric softener, 5 L was collected and kept at −4˚C for two days until the filtration experiment.

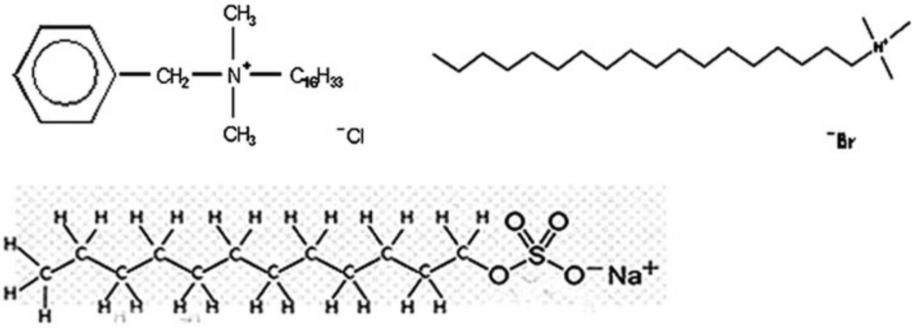

Fig. 1. Structural formulas of organic cations: Benzyl-dimethyl hexadecyl ammonium (BDMHDA, left), Octadecyl-N,N,N, trimethyl ammonium (ODTMA, right) and structural formula of anionic detergent: SDS.

2.6. Analytical determinations

The anionic detergent content was determined by Standard Method 5540C at "Aminolab" laboratory in Nes Ziona, Israel. Accuracy of the SM 5540C method is 0.1 ppm. Turbidity and TSS content were determined at the interdepartmental laboratory of The R.H. Smith Faculty of Agriculture, Food and Environment of the Hebrew University, Rehovot, Israel.

2.7. Bacteria counting

Bacterial analysis was performed at "Bactochem" laboratory, Nes Ziona, Israel, and consisted of counts of total bacteria (method 0-1-2-203), Coliforms (method 0-1-2-201), Fecal Coliforms (same method) and Fecal Streptococcus (method 0-1-2-204).

2.8. Theoretical: analysis of kinetics of filtration

Adsorption and convection are described by Eq. (1) below; numerical solution was executed by a FORTRAN program [16]. A column of length L is filled with material whose initial molar concentration of adsorbing sites is R_o, whose concentration changes later to $R(X, t)$. The beginning and end of the filter are at the coordinates $X = 0$ and L, respectively. We consider that the pollutant concentration at the inlet, C_o, is constant, i.e. $C(X, t) = C_o$, $X \leq 0$, where t denotes time.

The kinetic parameters are C_1 (M^{-1} min^{-1}, rate constant of forward adsorption), D_1 (min^{-1}, rate constant of desorption), v (flow velocity); α (≤1) denotes the degree of hysteresis, which was not considered in this case.

$$dC(X,t)/dt = -v\,\partial_C/\partial_X - C_1 \cdot C(X,t)\,R(X,t) + \alpha \cdot D_1(R_o - R(X,t)) \tag{1}$$

2.9. Statistics

The statistical criteria employed for simulation and prediction of certain experimental results of filtration by the calculations according to Eq. (1) were the values of R^2 and RMSE, the Root Mean Square Error, defined by

$$\text{RMSE} = \left(\sum (Y_{\text{ci}} - Y_{\text{expi}})^2/(n-2)\right)^{0.5} \tag{2}$$

In which n is the number of data points (we used averages of triplicates), and Y_{Ci} and $Y\text{expi}$, are the calculated and experimental values of percent removal. The term (n–2) in Eq. (2) is due to using two adjustable parameters.

3. Results and discussion

3.1. Removal of anionic detergents

Table 1 and Fig. 2 show the results of filtration of concentrated solutions (130 and 30 ppm, respectively) of the anionic detergent SDS by two filters, each including 6.5 g of a micelle–clay complex, ODTMA, or BDMHDA mixed with excess sand (at a ratio of 1:100 w/w). The removal was efficient and essentially similar for both filters, i.e. almost complete removal of SDS for the first 5 L. Table 1 indicates that more than 7 mmol of SDS can be adsorbed by each filter, which includes less than 2 mmol of excess positive charges. This result implies that the adsorption of SDS does not merely occur by charge neutralization, but also by other interactions, such as Van der Waals interactions at the external surface and interlayers of the micelle–clay complex.

The results in Fig. 3 describe filtration of well water (well #8, Bat-yam, Israel) at an average initial concentration of 1.33 ppm of anionic detergents and a flow rate of 51 mL/min, which corresponds to a flow velocity of 1.56 m/h. It should be noted that a flow velocity of 10 m/h in a large filter, e.g. of length of 1.6 m, would yield a larger contact time than in our experiment and consequently would yield a larger capacity per mass of the active ingredient than in the current experiment.

Table 1
Removal of the anionic detergent SDS from a 130 ppm solution[a] by a filter[b], which included micelle (ODTMA/BDMHDA)—montmorillonite complex

Volume passed (L)	Emerging concentration of SDS (ppm)	Removal (%)
ODTMA		
2	<0.1	~100
5	0.3	99.8
10	23.4	82
15	29.5	77.3
20	50	61.5
BDMHDA		
2	<0.1	~100
5	2.1	98.4
10	6.7	94.8
15	44.1	66.1
20	38.9	70.1

[a] A relative error of 10% originated mainly due to the dilution of 130 ppm SDS solution.

[b] The length of the active layer was 20 cm; the diameter of filter column was 5 cm. The micelle-clay complex was mixed with sand at 1:100 ratio (w/w). The flow rate was 51 mL/min.

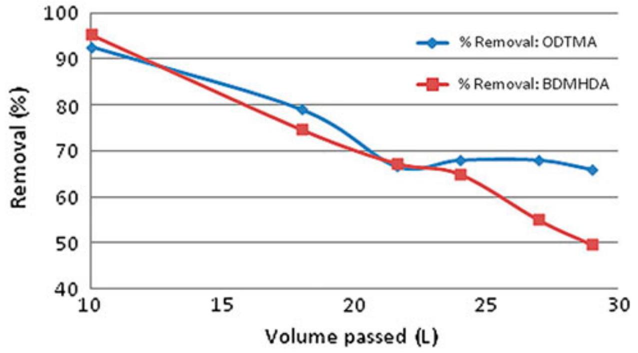

Fig. 2. SDS removal from 30 ppm solution by filters, which included an ODTMA-based or BDMHDA-based micelle-clay complex, mixed with sand at a 1:100 ratio (w/w). The relative error of 10% originated mainly due to the dilution of 30 ppm SDS solution.

After the passage of 90 L through the column, the percent removal was 97%. Results are similar to removal of anionic surfactants with a rotation biological contractor (RBC) which also removed 97% [17]. In another experiment (results not shown), complete removal (less than 0.1 ppm in the emerging water) was observed from the first 100 L taken from the well. The error in determination of anionic detergents was about 0.1 ppm. The estimated relative error in the experimental quantities is less than 10%.

The results in Fig. 3 were adequately simulated by calculations according to Eq. (1), the value of R^2 being 0.92. The results indicate that 220 L passed through the filter with a final concentration of anionic detergents below 0.3 ppm, which is the MCL in Israel (the EPA limit is 0.5 ppm).

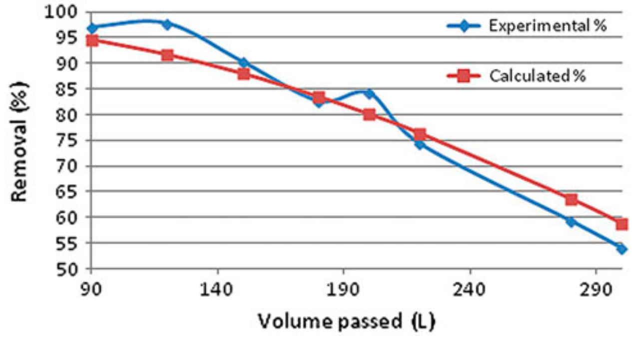

Fig. 3. Removal of anionic detergents from well water (1.33 ppm initial concentration): experimental and calculated values. The filter included a micelle-clay complex ODTMA-montmorillonite mixed with sand at a 1:100 ratio (w/w). The calculations were performed according to Eq. (1) and employed the following values of parameters: R_0 = 0.03 M; C_1 = 80 M^{-1} min^{-1}; D_1 = 0.0004 min^{-1}, where R_0 is the molar concentration of adsorbing sites, C_1 and D_1 are the kinetic constants of adsorption and desorption, respectively. The flow rate was 50 mL/min. The fits yielded RMSE = 4.3; R^2 = 0.92.

Calculations for the case of a filter of a length of 1.6 m filled with a mixture of the powdered complex ODTMA–clay mixed with excess sand at a 1:19 w/w ratio indicate that 1 kg of ODTMA can purify 150 m^3 of the water of this well at a flow velocity of 9 m/h.

3.2. Treatment of GW

As described in the Methods section, GW taken from showers and sinks of a student dormitory was filtered through sand and then through two columns in series including each 6.5 g of the micelle–clay complex based on ODTMA. The results in Table 2 indicate that the untreated water included a very large concentration of Coliforms and Fecal coliforms (7.2 and 2.4 million, respectively) per 100 mL. It was established that untreated GW irrigation was found to contaminate the soil, as levels of fecal coliforms were greater at GW irrigated sites than at sites irrigated with potable water. A risk of infection may result from contact with soil after irrigation if enteric pathogens are present in the GW supply [18].

After the passage of 15 L, none of these bacteria were detected in the emerging water. This amounts to a reduction in the numbers of these bacteria by more than six orders of magnitude. The total count of bacteria was reduced by five orders of magnitude in the water after passing through the second filter. In

Table 2
Filtration of grey water by two columns[a] in series including micelle (ODTMA)—montmorillonite mixed with sand (1:100 w/w ratio): bacterial count

Sample	Analysis	Microbial count (CFU/100 mL)
Raw water	Total count	3.1×10^7
	coliforms	7.2×10^6
	Fecal coliforms	2.4×10^6
Filtered water after 15 L; Column I	Total count	6.9×10^4
	coliforms	<1
	Fecal coliforms	<1
Filtered water after 15 L; Column II	Total count	1×10^2
	coliforms	<1
	Fecal coliforms	<1

[a] Each column was as in Table 1. The organic cation used was ODTMA. The flow rate was 50 mL/min.

Table 3
Filtration of grey water from showers/sinks (as in Table 2) and domestic laundries effluent (first wash) by two columns[a] in series, which include micelle (ODTMA)—montmorillonite mixed with sand (1:100 w/w ratio)

Sample	Before filtration	Filtered water after 4 L; Column I	Filtered water after 4 L; Column II
Anionic detergents			
Showers/sinks	0.4	<0.1	<0.1
Laundry #1	51	6.3	<0.1
Laundry #2	151	100	35
TSS			
Showers/sinks	26.3	1	0
Laundry #1	375	220	148
Laundry #2	560	300	236
Turbidity from showers/sinks (NTU)	3.05	0.14	0.1

[a] Each column was as in Table 1. Units of anionic detergents and TSS were mg/L.

comparison, the RBC was capable of reducing fecal coliform concentrations from 3.0×10^5 to 1.7×10^2 [17].

The results in Table 3 (for water from showers and sinks) show a reduction of Total suspended solids (TSS) from 26 to 1 ppm after the passage of 4 L through the first filter and to below detection limit after the passage through the second one. The turbidity was reduced significantly to 0.1 NTU from an initial level of 3 NTU. The anionic detergents in the water were reduced to below the detection limit.

The results demonstrate that the values of TSS in the water of the first wash from domestic laundries were larger by an order of magnitude than the value found for GW from sinks and showers. The values of TSS in another study on GW [19] (80 ppm) are between our values for sinks and showers and laundries. The values of TSS from laundries were reduced by 2.4- to 2.5-fold upon emergence from the second filter, but were too high with respect to the limits allowed for irrigation. It can be projected that doubling the amount of the micelle–clay complex in each filter to 13 g would have solved this problem. The values of anionic detergents in the water from the first wash from the laundries were two orders higher than those from sinks and showers. The second filter reduced the concentration of anionic detergents from one of the domestic laundries from 51 ppm to below detection limit. We note that the water from laundries included a significantly smaller number of bacteria, about 1,000-fold less fecal coliforms than from sinks and showers and no fecal streptococci.

The results shown in Tables 4–7 were obtained in an additional experiment. These results reiterate the large number of indicator and pathogenic bacteria per 100 mL, e.g. 10 million, whereas the allowed numbers for irrigation vary between 10 and 100 per 100 mL. The results demonstrate that the water passing through the second filter (i.e. through a system which included 13 g of the micelle–clay complex) after 30 L included no bacteria at all, whereas after 15 L no fecal

Table 4
Filtration of grey water by two columns in series including micelle (ODTMA)—montmorillonite mixed with sand (1:100 w/w ratio), compared to filtration by two columns in series with activated carbon mixed with sand (1:100 w/w ratio): Bacterial count[a]

Description of sample	Volume passed (L);	Total bacterial count in 1 mL	Fraction emerging[b]
Initial grey water 1	–	1.6×10^6	–
Initial grey water 2	–	1.3×10^6	–
1-st column:			
Activated carbon	5	9.1×10^5	0.63 [(0.2–1)]
Complex	5	8.7×10^2	6×10^{-4} [$(0.2–1) \times 10^{-3}$]
Activated carbon	15	1.4×10^6	0.97 [(0.5–1)]
Complex	15	8.1×10^2	5.6×10^{-4} [$(0.2–1) \times 10^{-3}$]
Complex	30	5.2×10^4	0.04 [(0.01–0.07)]
2-nd column:			
Activated carbon	15	9.1×10^5	0.63 [(0.2–1)]
Complex	15	<1	0
Complex	30	1	6.9×10^{-7} [$(0.3–1) \times 10^{-6}$]

[a] The estimated relative error in the number of bacteria in initial solution is about 50%. The estimated relative error in the number of emerging bacteria is less than 20%.

[b] The numbers in parentheses indicate the range of possible values.

Table 5
Filtration of grey water from showers/sinks (as in Table 4): coliforms[a]

Description of sample volume passed (L)	Coliforms in 100 mL	Fraction emerging[b]
Initial grey water (Average)	2.15×10^7	–
5 L; 1-st column		
Activated carbon	9.6×10^6	0.45 [(0.2–0.75)]
Complex	<1	0
15 L; 1-st column		
Activated carbon	2.1×10^7	0.98 [(0.5–1)]
Complex	<1	0
30 L; 1-st column		
Complex	7×10^1	3.3×10^{-6} [$(2–5) \times 10^{-6}$]
15 L; 2-nd column		
Activated carbon	1.1×10^7	0.51 [(0.1–0.9)]
Complex	<1	0
30 L; 2-nd column		
Complex	<1	0

[a] The estimated relative error in the number of bacteria in initial solution is about 50%. The estimated relative error in the number of emerging bacteria is less than 20%.

[b] The numbers in parentheses indicate the range of possible values.

coliforms or fecal streptococci were detected even in the water emerging from the first filter. In contrast, an activated carbon filter yielded poor removal of bacteria even after the passage of 5 L.

The results (Tables 2 and 4–7) demonstrate an extremely large number of pathogenic bacteria in GW collected from showers and sinks. These values are in the same range as in Travis et al. [19]. The use of such water for irrigation without treatment poses a health hazard. On the other hand, a filter that included just 13 g and in certain cases 6.5 g of a micelle–clay complex mixed with excess sand at a ratio of 1:100 w/w was shown to remove all pathogenic bacteria from 30 L of the GW.

A comparison of our results with those of several recent studies that employed a variety of technologies for treatment of GW [19, 13, and 12] demonstrates the advantage of the micelle–clay complex in reducing significantly the number of pathogenic bacteria, in fact below detection limit, despite the relatively small amounts of the complex. This efficiency seemingly arises from the characteristics of the complex, which has large hydrophobic domains and is positively charged, whereas the bacteria had an excess of negative charge on their external surface. Laboratory studies [20] demonstrated that the release of bacteria from the complex was very slow, i.e. the adsorption was almost irreversible.

The question of costs will not be elaborated here, but we foresee several feasible developments. One is the use of a granulated complex, which will avoid the need to mix the complex with sand and will increase the amount of the complex in the filter by almost 50-fold in comparison with the amount used in the current study. The second development will be to design a GW reactor, which in the first stage consists of a biological treatment by moving bed biofilm reactor [21,22]. This will reduce the amount of TSS

Table 6
Filtration of grey water from showers/sinks (as in Table 4): Fecal coliforms[a]

Description of sample and volume passed (L)	Fecal coli in 100 mL	Fraction emerging[b]
Initial grey water (Average)	1.14×10^7	–
5 L; 1-st column		
Activated carbon	8.2×10^6	0.72 [(0.3–1)]
Complex	<1	0
15 L; 1-st column		
Activated carbon	1.1×10^7	0.97 [(0.6–1)]
Complex	<1	0
30 L; 1-st column		
Complex	1.2×10^1	1.1×10^{-6} [$(0.6–1.6) \times 10^{-6}$]
15 L; 2-nd column		
Activated carbon	8.7×10^6	0.77 [(0.3–1)]
Complex	<1	0
30 L; 2-nd column		
Complex	<1	0

[a] The estimated relative error in the number of bacteria in initial solution is about 50%. The estimated relative error in the number of emerging bacteria is less than 20%.

[b] The numbers in parentheses indicate the range of possible values.

Table 7
Filtration of grey water from showers/sinks (as in Table 4): Fecal Streptococci[a]

Description of sample and volume passed (L)	Fecal streptococci in 100 mL	Fraction emerging[b]
Initial grey water (Average)	2.05×10^4	–
5 L; 1-st column		
Activated carbon	5.8×10^3	0.28 [(0.08–0.48)]
Complex	<1	0
15 L; 1-st column		
Activated carbon	2×10^4	0.98 [(0.5–1)]
Complex	<1	0
30 L; 1-st column		
Complex	<1	0
15 L; 2-nd column		
Activated carbon	1×10^4	0.49 [(0.2–0.8)]
Complex	<1	0
30 L; 2-nd column		
Complex	<1	0

[a] The estimated relative error in the number of bacteria in initial solution is less than 50%. The estimated relative error in the number of emerging bacteria is less than 20%.

[b] The numbers in parentheses indicate the range of possible values.

and COD whose adsorption by the filter material may reduce the number of available sites for adsorption of bacteria. In view of this intended development, we avoided focusing on removal of COD in the current study. In a recent study [23], the same set of filters as in the current study reduced 4-fold, the values of COD and BOD of partially treated wastewater (from initial COD value of 80 ppm) after the passage of 14 L. The removal of organic material in the first stage will also be instrumental in facilitating regeneration of the granulated complex, which will sorb smaller amounts of organic material and larger numbers of bacteria, since preliminary results of regeneration of the complex after adsorption of bacteria are promising.

The results in Tables 4–7 indicate that 13 g of a powdered complex in the filters removed all pathogenic bacteria from more than 30 L of the grey water used. Hence, if the granulated complex would be similar in its efficiency to the powdered one, then filling a column filter exclusively with 2,600 g of granulated complex (less than 3.5 L) is estimated to suffice for purification of 9,000 L, since having the active material more concentrated will increase its capacity per g of complex by 50%. This amounts to three months of operation in a single family house. If the two-stage reactor will double the capacity, then a grey water reactor of a volume of 300 L will suffice for six months of operation before replacement of the filter material.

4. Conclusions

(a) Efficient removal of anionic detergents from water was achieved by a filter that included micelle–montmorillonite complexes in which the organic cation was ODTMA, or BDMHDA.
(b) A model that considered convection, adsorption, and desorption could adequately simulate removal of anionic detergents from water by filtration.
(c) GW from showers and sinks included several million of pathogenic bacteria per 100 mL, indicating a health hazard using this water for irrigation without proper treatment. Filters including a micelle– clay complex were shown to remove efficiently the pathogenic bacteria from this GW, thus enabling to use the treated water safely for irrigation, or toilet flushing. In comparison, activated carbon was inadequate for the removal of bacteria from GW.

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Abbreviations

BDMHDA	—	benzyldimethyl hexadecylammonium
BTC	—	breakthrough curve
CMC	—	critical micelle concentration
DW	—	deionized water
EPA	—	Environmental Pollution Agency
MCL	—	maximum contamination level
ODTMA	—	octadecyltrimethylammonium
SDS	—	sodium dodecyl sulfate
BF	—	biofilter
CEC	—	cation exchange capacity
CW	—	constructed wasteland
DWW	—	domestic waste water
GW	—	greywater
NTU	—	Nephelometric Turbidity Units
OM	—	organic matter

References

[1] T. Cserhati, E. Forgacs, G. Oros, Biological activity and environmental impact of anionic surfactants, Environ. Int. 28 (2002) 337–348.

[2] C. Merz, R. Scheumann, B. El Hamouri, M. Kraume, Membrane bioreactor technology for the treatment of greywater from a sports and leisure club, Desalination 215 (2007) 37–43.

[3] W.A. Jury, H.J. Vaux Jr., The emerging global water crisis: Managing scarcity and conflict between water users, Adv Agron 95 (2005) 1–76.

[4] Mekorot, Wastewater treatment and reclamation, in: Watec, 2007, Tel-Aviv, Israel

[5] A.J. Hamilton, F. Stagnitti, X. Xiong, S.L. Kreidl, K.K. Benke, P. Maher, Wastewater irrigation: The state of play, Vadose Zone J. 6 (2007) 823–840.

[6] A. Maimon, A. Tal, E. Friedler, A. Gross, Safe on-site reuse of greywater for irrigation—A critical review of current guidelines, Environ. Sci. Technol. 44 (2010) 3213–3220.

[7] US EPA, Decentralized Wastewater Treatment Systems: A Program Strategy (EPA 832-R-05-002), EPA, Washington, DC, 2005.

[8] B. Hood, E. Gardner, R. Barton, R. Gardiner, C. Beal, R. Hyde, C. Walton, Decentralized development: The Ecovillage at Currumbin, Water J. of Aust. Water Assoc. 37 (2010) 58–66.

[9] E. Friedler, D. Butler, Quantifying the inherent uncertainty in the quantity and quality of domestic wastewater, Water Sci. Technol. 33 (1996) 65–78.

[10] A. Gross, A. Wiel-Shafran, N. Bondarenko, Z. Ronen, Reliability of small scale greywater treatment systems and the impact of its effluent on soil properties, Int. J. Environ. Stud. 65 (2008) 41–50.

[11] L. Metcalf, H.P. Eddy, G. Tchobanoglous, F.L. Burton, H.D. Stensel, Wastewater Engineering, Treatment and Reuse, 4th ed., McGraw Hill Education, Columbus, OH, 2003.

[12] S. Dalahmeh, M. Pell, B. Vinnerås, L. Hylander, I. Öborn, H. Jönsson, Efficiency of bark, activated charcoal, foam and sand filters in reducing pollutants from greywater, Water, Air Soil Pollut. 223 (2012) 3657–3671.

[13] K. Bani-Melhem, E. Smith, Greywater treatment by a continuous process of an electrocoagulation unit and a submerged membrane bioreactor system, Chem. Eng. J. 198–199 (2012) 201–210.

[14] Y.G. Mishael, T. Undabeyita, G. Rytwo, B. Papahadjopoulos-Sternberg, B. RubinS. Nir, Sulfometuron adsorption via alkylammonium cations adsorption as monomers and micelles on montmorillonite, J. Agric., Food Chem. 50 (2002) 2856–2863.

[15] T. Polubesova, S. Nir, D. Zadaka, O. Rabinovitz, C. Serban, L. Groisman, B. Rubin, Water purification from organic pollutants by optimized micelle-clay systems, Environ. Sci. Technol. 39 (2005) 2343–2348.

[16] S. Nir, D. Zadaka-Amir, A. Kartaginer, Y. Gonen, Simulation of adsorption and flow of pollutants in a column filter: Application to micelle-clay mixtures with sand, Appl Clay Sci. 67–68 (2012) 134–140.

[17] Y. Alfiya, O. Damti, A. Stoler-Katz, A. Zoubi, A. Shaviv, E. Friedler, Potential impacts of on-site greywater reuse in landscape irrigation, Water Sci. Technol. 65 (2012) 757–764.

[18] L.M. Casanova, V. Little, R.J. Frye, C.P. Gerba, A survey of the microbial quality of recycled household greywater, Am. Water Res. Assoc. 37 (2001) 1313–1319.

[19] M.J. Travis, A. Wiel-Shafran, N. Weisbrod, E. Adar, A. Gross, Greywater reuse for irrigation: Effect on soil properties, Sci. Total Environ. 408 (2010) 2501–2508.

[20] A. Shtarker-Sasi, A. Castro Sowinski, O. Matan, T. Kagan, S. Nir, Y. Okon, M.N. Nasser, Removal of bacteria and Cryptosporidium from water by Micelle—Montmorillonite Complexes, Desalin. Water Treat. (2013) 1–9, doi: 10.1080/19443994.2013.776993.

[21] H. Odegaard, B. Rusten, T. Westrum, A new moving bed biofilm reactor-applications and results, Water Sci. Technol. 29 (1994) 157–165.

[22] H. Odegaard, Innovations in wastewater treatment: The moving bed biofilm process, Water Sci. Technol. 53 (2006) 17–33.

[23] M. Khamis, R. Karaman, M. Qurie, J. Abbadi, S. Nusseibeh, A. Manassra, S. Nir, Performance of micelle-clay filters for removing pollutants and bacteria from tertiary treated wastewater, J. Environ. Sci. Eng. 1 (2012) 160–168.