Physicochemical Treatment of Light Grey Water: An Experimental Study Using Membrane Filtration

E. Giresunlu*, E. Soyer** and B. Beler-Baykal*

*Istanbul Technical University, Department of Environmental Engineering, TURKEY
(E-mail: giresunlue@itu.edu.tr; baykalb@itu.edu.tr)
** Marmara University, Department of Environmental Engineering, TURKEY
(E-mail: elif.soyer@marmara.edu.tr)

Abstract
Light grey water is a renewable source of water which may be returned to almost any point of the water cycle after proper treatment. Most of the work in the literature focuses on treatment of organics and biological treatment processes for grey water reclamation. However, grey water from different sources exhibit considerable variations in quality and treatment schemes are dependent upon grey water characterization. This work investigates the treatment of light grey water from a five-star hotel with a low BOD/COD ratio and high particulate organics, through physicochemical treatment employing membrane filtration, with and without pretreatment in a sand filter unit. Lab scale experiments were conducted using laboratory-made membranes. The results revealed that membrane filtration without a biological unit, with or without sand filter pretreatment was successful in providing effluent quality required of reclaimed grey water, with 90% BOD; 70-79% COD, 100% SS and 100% pathogenic indicator removals. Although the presence of a sand filter as a pretreatment unit did not change effluent quality immensely, it indeed had a positive contribution in terms of retarding immediate clogging of the membrane, to lead to longer filtration times and higher water volumes to be processed. All in all, this work provides support for the need of characterization of grey water prior to the selection of the treatment scheme and the possible use of physicochemical treatment only for this purpose.

Keywords
Light grey water, wastewater characterization, physicochemical treatment, membrane filtration, sand filtration as pretreatment

INTRODUCTION

Alternative management concepts are being proposed to be able to use domestic wastewater in the most efficient way as a source. One of those concepts, stream segregation, enables the recycling of valuable materials available in domestic wastewater, like nutrients and water (Beler Baykal, 2015). Grey water is that part of domestic wastewater which excludes wastewater coming from toilet bowls and consists of wastewater from different washing functions in the household. It is usually wash-water generated in baths, showers, wash basins, washing machines, sinks and dish washers etc. It constitutes about 75% of domestic wastewater by volume, and with low quantities of nutrients and about 40% of organics, it has a lower pollution potential as compared to the conventional mixed one. At this time, grey water is mostly reused for irrigation and toilet flushing, however it is a source that can be returned to almost any point of the water cycle after proper treatment.

Grey water itself can be segregated into two fractions as light and dark grey water (Birks and Hills, 2007). Light grey water refers to the portion coming from baths/showers and wash basins, while dark grey water is that part which originates from washing machines, kitchen sinks and dish washers. As such, light grey water has a lower pollution potential and can be treated easier as compared to the dark one.

Characteristics of grey water may change depending upon various factors such as source of generation, personal habits, water usage and geographical location (Jefferson et al., 2000). Even if the sources where the grey water was collected are the same, the quality may change significantly in buildings with different functions (Giresunlu et al., 2016). Generally, grey water quality is comparable to weak domestic wastewater or better, when values given by Metcalf & Eddy (2003) are taken as the basis. In the literature, the primary concern with grey water is considered to be organic matter, while earlier work showed that microbiological indicators should also be examined prior to reuse. Although in general, grey water is analyzed for organic matter mostly for its possibly high organics content, microbiological indicator concentrations can go as high as conventional domestic wastewater as given in Table 1 and elsewhere (Giresunlu and Beler-Baykal, 2015). Nutrients are not considered as problematic pollutants in grey water, as their concentrations are generally low (Giresunlu et al., 2016) to be compared to other domestic wastewater streams, or conventional domestic wastewater itself.

Currently, reusing grey water is a trending application, especially in settlements and hotels, specifically for those which seek green building certification, as green building certification systems require reduction in the amount of water consumed in the building through water reuse. Examples of literature work devoted to characterization of light hotel grey water are given in Table 1, showing that regardless of the fact that all are hotel light grey water, variations in wastewater characteristics may be observed. COD, BOD, TSS and VSS concentrations reported by Metcalf & Eddy, 2003 for weak conventional domestic wastewater are 250, 110, 120 and 95 mg/L respectively. All grey water concentrations given in Table 1 show lower pollution potential as compared to typical conventional domestic wastewater in terms of organic matter and nitrogen as well as phosphorus (Metcalf & Eddy, 2003). On the other hand, total suspended solid concentrations of weak grey water samples are similar to each other, while they too are lower as compared to the typical TSS concentration of conventional weak domestic wastewater.

Grey water may be reclaimed through the use of different biological treatment schemes including low tech / high foot print ones like constructed wetlands, or high tech / compact ones like

Table 1. Examples of light grey water characteristics from hotels (std dev ları nasıl buluyorsunuz?)

Parameter	March et al., 2004		Gual et al., 2008		Giresunlu et al., 2016	
	C	n	C	n	C	n
COD	171 mg/L	12	72.7±50 mg/L	34	120±114 mg/L	41
TOC	58 mg/L	18	41±10 mg/L	34		
sCOD					41±18 mg/L	37
BOD					31±18 mg/L	35
TSS	44 mg/L	12	32.2±7 mg/L	34	72 ±124 mg/L	41
VSS					39±57 mg/L	40
NH_3-N					2.48±1.23 mg/L	36
TN	11.4 mg/L	12	4.1±2 mg/L	34		
TKN					6.97±5.43 mg/L	36
PO_4-P					1.01±0.98 mg/L	36
TP					2.21±2.29 mg/L	36
Turbidity	20 mg/L		38.8±20 mg/L	34		
Total coliform			6	34	7.15 log_{10} cfu/100 mL	3
Fecal coliform					6.95 log_{10} cfu/100 mL	3
E.coli					6.92 log_{10} cfu/100 mL	3
Enterococci					0	3

sequencing biological reactors (SBR), rotating biological contactor (RBC) and membrane bioreactors (MBR), with the advantage of MBR being effective both for organic matter and pathogen removal. Additionally, physicochemical treatment processes may also be used for grey water reclamation. Among these, membrane filtration will have additional benefits for providing pathogenic safety. The choice of treatment scheme will be dictated by grey water characteristics (Giresunlu et al., 2016). However, most frequently, treatment of grey water prior to reuse is done with a biological treatment step to remove organics, mostly through MBR, while a limited number of work examined physicochemical treatment systems for grey water treatment. Examples of literature work addressing removal efficiencies for various grey water systems with a filtration unit are summarized in Table 2. Holden and Ward (1999) have done physical treatment trials with mixed grey water from apartments using membrane filtration to show that removal efficiencies were better as compared to sand filtration with 86% BOD and 84% COD. Filtration was employed with different pore sizes on different fractions of light grey water from university changing rooms and the maximum removal was reported as 82%, 56% and 35% in terms of BOD, COD and TSS respectively, for shower grey water filtered through 0.025 mm pore size (Santos et al., 2012). Ultrafiltration, nanofiltration and reverse osmosis were applied to laundry grey water, which is considered as dark grey water with higher organic matter content, and while ultrafiltration removed 54% COD and 49% TSS, reverse osmosis treatment efficiencies were about 100% (Sostar-Turk et al., 2005). The hotel grey water which was collected from bathtubs/showers and washbasins were treated through sand filtration and sedimentation and the reported removal efficiencies were 54% and 58% in terms of COD and TSS respectively (March et al., 2004).

Literature work on treatment of grey water through biological systems is also given in Table 2. Light grey water collected from a house was treated through screen, RBC and sand filtration, a combination of physical and biological treatment systems, ended up with 94%, 75% and 82% removal efficiencies in terms of BOD, COD and TSS respectively (Friedler et al., 2005). MBR, the treatment system that combines the benefits of biological treatment and membrane filtration, was applied to grey water from baths/showers and wash basins of a student residence hall and 95% BOD, 84% COD and 96% TSS removal efficiencies were observed (Giresunlu and Beler-Baykal, 2016), while the same treatment system resulted with 93% BOD and 85% COD removal for shower grey water of a sports complex (Merz et al., 2007). In these examples, organics removal was higher as expected upon the addition of biological treatment. However, BOD/COD sCOD/COD ratios are important in terms of efficiency of the treatment system selected.

Earlier work devoted to long-term monitoring of light grey water characteristics in a five-star hotel revealed that organics content was low and mostly in particulate form (Giresunlu et al., 2016) implying that the “bio” part of the MBR may actually not be needed. The aim of this work was to investigate the reclamation of light grey water from baths/showers and wash basins in a five-star hotel, specifically focusing upon its treatment through membrane filtration, focusing on whether membrane filtration only would be sufficient. In lab scale experiments, the effectiveness of membrane filtration and membrane filtration following sand filtration as a pretreatment was investigated.

MATERIAL AND METHODS

A long-term monitoring of light grey water, collected from washbasins, baths and showers, in a five-star hotel, located in Istanbul, Turkey was carried out for one full year. Characterization was done in terms of organic matter (total, soluble, particulate), solids and nutrients (Giresunlu et al., 2016), and some of the samples were tested for microbiological indicators elsewhere (Giresunlu and Beler-Baykal, 2015). The results of year-round characterization are summarized in Table 3.

Table 2. Removal efficiencies for various grey water systems with a filtration unit

Source	Fraction	Treatment process	Parameter	Removal	References
Apartment	Mixed	Sand filtration	BOD COD Turb.	63% 75% 27%	Holden & Ward (1999)*
Apartment	Mixed	Membrane	BOD COD Turb.	86% 84% 99%	Holden & Ward (1999)*
University changing room	Wash basin	Filtration (0.130 mm)	BOD COD TP TSS	65% 47% 43% 59%	Santos et al. (2012)
University changing room	Wash basin	Filtration (0.025 mm)	BOD COD TP TSS	61% 72% 66% 82%	Santos et al. (2012)
University changing room	Shower	Filtration (0.025 mm)	BOD COD TP TN TSS	82% 56% 70% 90% 35%	Santos et al. (2012)
Restaurant	Wash basin	Filtration (0.130 mm)	BOD COD TP TSS	59% 50% 28% 46%	Santos et al. (2012)
House	Laundry	UF membrane	COD TN TSS Turb.	54% 38% 49% 15%	Sostar-Turk et al. (2005)
House	Laundry	NF membrane	BOD COD TSS	56% 93% 100%	Sostar-Turk et al. (2005)
House	Laundry	RO membrane	BOD COD TSS Turb.	98% 98% 100% 97%	Sostar-Turk et al. (2005)
Hotel	Bathtub, wash basin	Filtration, sedimentation	COD TN TSS Tur.	54% 39% 58% 18%	March et al. (2004)*
House	Shower/ bath, wash basin	Screen, RBC, sand filter	BOD COD TP TSS Turb.	94% 75% 58% 82% 98%	Friedler et al. (2005)
Student residence hall	Shower/ bath, wash basin	MBR	BOD COD TP TKN TSS	95% 84% 46% 80% 96%	Giresunlu and Beler-Baykal (2016)*
Sports complex	Shower	MBR	BOD COD TP TKN Turb.	94% 85% 19% 63% 98%	Merz et al. (2007)

*calculated using the concentrations reported in original references

Table 3. Characteristics of raw light grey water (Giresunlu et al., 2016)

	COD	sCOD	BOD	TSS	VSS	NH_3-N	TKN	PO_4-P	TP
Min (mg/L)	30	19	8	0	0	0.58	2	0	0.26
Max (mg/L)	710	152	82	838	376	5.55	50.17	4.01	8.35
Mean (mg/L)	120	41	31	72	39	2.48	6.97	1.01	2.21
Number of samples	41	37	35	41	40	36	36	36	36

The raw grey water investigated in this work was representative of all the characteristics given in Table 3 and its specific composition is presented in Table 4. All analyses were carried out according to Standard Methods (Eaton et al., 2005) except for COD and sCOD, which were analyzed using ISO 6060 method.

Raw grey water from the hotel was treated in lab-scale units directly using membrane filtration and membrane filtration following sand filtration. For sand filtration, a plexiglas column with 4.4 cm diameter and 1 m length was used. Diameter of the sand used was chosen as 0.8-1.0 cm, which is 40 times smaller than the column diameter, to prevent wall effects (Fand and Thinakaran, 1990). The column was filled with 84.5 cm of sand and operated in down-flow mode with a filtration rate of 6 m/h. The column effluent, filtrate, was not taken into the clean water reservoir during the first 10 min of operation since this initial stage of filtration is known as filter ripening and the quality of the filtrate is lower than what would normally be expected.

Ultrafiltration membranes evaluated in this study were prepared using polysulfone, PSf, and polyvinylpyrrolidone, PVP (Sigma-Aldrich, molecular weight Mn=26,000 and 40,000 Da, respectively). N-methyl-2-pyrrolidone, NMP, and deionized water were used as solvent and coagulant, respectively. Membranes were fabricated by immersion-precipitation process. Membrane casting solution was composed of 18% PSf, 15% PVP, and 67% NMP by weight. Mixtures were homogenized with continuous stirring of several hours, deposited onto glass plates using a casting device (Universal Blade Applicator, Paul N. Gardner Company, Inc.) with a preset height of 254 µm and transferred into a deionized water bath at room temperature to start phase-inversion process. Membrane film peeled off from the glass plate by itself within 1 min was washed thoroughly with deionized water. Resulting membranes were soaked in deionized water and stored at 4°C prior to use.

Filtration experiments were carried out using a 300 mL dead-end stirred cell (Sterlitech HP4750). Cell was connected to a reservoir that was pressurized by nitrogen gas cylinder. Sample in the cell was stirred at 625 rpm to produce turbulence and prevent clogging of the membrane by accumulated solids. Active area of the membrane was 14.6 cm^2. Operating pressure during experiments was 1.5 bar and the volume of permeate was recorded every 30 seconds over a filtration period. Pure water flux of the membrane used was determined filtering deionized water (and is referred to as J_0) before starting filtration of grey water. Permeate fluxes at the end of one filtration cycle (J_1) and at the beginning of a subsequent filtration cycle following the surface wash of the membrane (J_2) were normalized using J_0 since there were slight variations in pure water fluxes of lab-made membranes.

Decline in permeate flux, and reversible and irreversible components of this decline were estimated using the equations below:

$$F = \left[1 - \frac{J_1}{J_0}\right] x100 \qquad \text{Flux decline at the end of one filtration cycle} \qquad (1)$$

$F_I = \left[1 - \frac{J_2}{J_0}\right] x100$	Irreversible flux decline or flux decline that is not recovered with membrane wash	(2)
$F_R = \left[\frac{J_2 - J_1}{J_0}\right] x100$	Reversible flux decline or flux decline that is recovered with membrane wash	(3)

The sum of reversible and irreversible flux decline is equal to the overall flux decline ($F = F_I + F_R$).

Raw and effluent water quality, variation in permeate flux and extent of fouling as well as its reversibility through the use of Eqs 1-3 were determined as a measure of success of the treatment scheme.

RESULTS AND DISCUSSION

Characteristics of the grey water sample used in this study are given in Table 4. The COD concentration of raw grey water sample was 159 mg/L, while the sCOD concentration was 54 mg/L, which are representative of those given in Table 3 as typical characteristics of this light grey water. The BOD concentration of the influent was 30 mg/l, which is almost the mean value of the 35 samples. TSS and VSS concentrations in raw grey water were 214 and 52 mg/L respectively, which are higher than the average given in Table 3 as typical concentrations of this particular grey water.

The sample used in this study showed lower organic matter concentrations as compared to the weak conventional domestic wastewater (Metcalf & Eddy, 2003), while these values stand between the two literature work done with weak hotel grey water (March et al., 2004; Gual et al., 2004). Average TSS concentration in this work was lower as compared to weak domestic wastewater, while it was slightly higher than the literature data focusing on the same type of grey water. All average nutrient concentrations were under typical values for weak conventional domestic wastewater. On the other hand, as literature work on light hotel grey water analyzed total nitrogen only, the characteristics of grey water in this study was not comparable.

The average microbiological indicator concentrations in terms of total coliforms, fecal coliforms, E.coli and enterococci were 1.42E+07, 8.91E+06, 8.37E+06 and 0 cfu/100 mL (Giresunlu et al., 2016), which are comparable to conventional domestic wastewater microbiological indicator concentrations.

The treatment applied to this grey water in the hotel was an MBR, which combines the benefits of both biological and physical treatment. Based on the information provided by Morel and Diener, 2006), BOD/COD ratio higher than 0.4-0.5 indicates high biodegradability. BOD/COD ratio and particulate form of organics for the sample used in this study are calculated as 19% and 66% respectively based on the values presented in Table 4, indicating low biodegradability and makes physicochemical treatment/membrane filtration an option.

In this study, direct membrane filtration and membrane filtration following sand filtration were applied to the grey water sample to evaluate the contribution of a pretreatment step on effluent quality as well as on the performance of membrane filtration.

A survey of Table 4, where the treatment efficiencies are shown, reveals that direct membrane filtration removal efficiencies in terms of COD and sCOD were calculated as 70% and 13% respectively. This shows that membrane filtration was successful in removal of particulate organic matter, however removal of soluble organic matter was limited as expected. The results also indicate that only 1 mg/L corresponding to 2% particulate organic matter was not removed with direct membrane filtration. Direct membrane filtration ended up with an effluent quality of 3 mg/l

Table 4 Results of treatment trials

Parameter	Raw grey water quality	Effluent quality after membrane filtration	Removal efficiency for membrane filtration	Effluent quality after sand filtration	Removal efficiency for sand filtration	Effluent quality after sand filtration + membrane filtration	Removal efficiency for sand filtration + membrane filtration
COD[1]	159	48	70%	56	65%	34	79%
sCOD[1]	54	47	13%	54	-	33	43%
pCOD[1*]	105	1	99%	2	98%	1	99%
BOD[1]	30	3	90%	9	70%	3	90%
sBOD[1]	9	2	78%	9	-	2	78%
TOC[1]	66	20	70%	23	65%	14	79%
TSS[1]	214	0	100%	82	62%	0	100%
VSS[1]	52	0	100%	1	98%	0	100%
NH_3-N[1]	1.61	0.85	47%	1.57	2%	0.8	50%
PO_4-P[1]	0.58	0.48	17%	0.58	-	0.48	17%
TP[1]	1.15	1.1	4%	1.15	-	1.09	5%
Total Coliform[2]	8.70E+06	0		2.10E+06		0	
Fecal Coliform[2]	7.50E+06	0		6.20E+05		0	
E.Coli[2]	2.60E+05	0		3.00E+04		0	
Enterococci[2]	0	0		0		0	

[1]mg/L [2]cfu/100 mL [*]calculated

BOD and 2 mg/l sBOD, which achieves the target grey water reuse limit of 5 mg/l BOD as given by Nolde (1999), and provides 90% BOD removal. The unit provided 100% of TSS and VSS removal, as expected, and the effluent was free of solid matter.

The raw grey water sample tested had appreciable amount of microbiological indicators. Although these concentrations were comparable to conventional domestic wastewater, the treatment unit was capable of removing all the microbiological indicators tested.

When membrane filtration was employed after sand filtration, the effluent quality did not change appreciably as can be observed from Table 4, however, the advantage of sand filtration was obvious with fluxes. Treated water flux immediately declined when the filter received raw grey water indicating an immediate pore blocking. Permeate production as soon as filtration started was recorded as 40% of the pure water flux of the same membrane. This corresponds to a decrease of 60% of pure water flux as shown in Figure 1. As a result of this severe pore blocking, only 5% of the flux decline was recovered by membrane washing and irreversible flux decline was calculated as 69% of the pure water flux at the end of 30 minutes of filtration.

On the other hand, feed water that was pretreated using a sand filter column showed almost no appreciable initial decline in water flux, as initial flux was around 99% of the pure water flux. This was due to the removal of suspended solids, which included particulate fractions of COD and BOD, during sand filtration. As shown in Table 4, removal efficiencies in terms of COD, BOD and SS were 65%, 70% and 62%, respectively.

Figure 2a displays the variation in flux (J/J_0) versus time for the filtration of sand-filtered sample. Treated water flux at the end of four subsequent filtration cycles was 44, 56, 62 and 68% less than

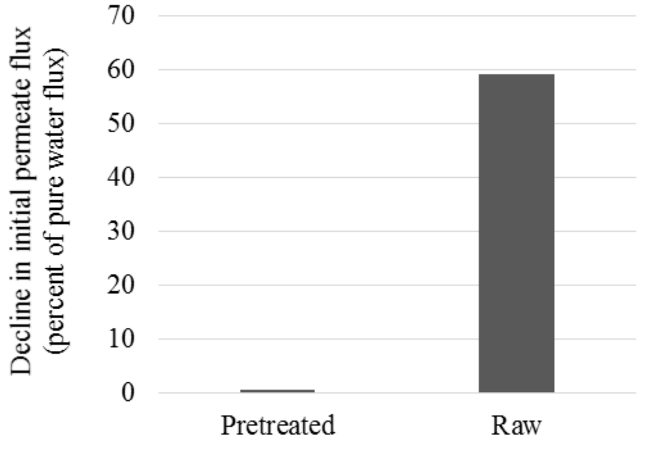

Figure 1. Initial flux decline during membrane filtration of grey water with and without of pretreatment

the pure water flux obtained with clean, unused membrane. The ability of the membrane to recover after fouling during each period was assessed as shown in Figure 2b. Membrane cleaning resulted in a 26% recovery of flux (as percentage of pure water flux of the same membrane) at the end of first filtration cycle. As the membrane pores were getting clogged, flux recovery after simple surface washes of the membrane dropped to 21 and 18% of the pure water flux (2nd and 3rd filtration cycles). Irreversible loss of membrane performance was around 18% of the pure water flux at the end of first cleaning cycle and this value increased to 35 and 44% for the following 2nd and 3rd stages of filtration, respectively.

The amount of water that can be treated during one hour of filtration was measured for both cases. Figure 3 shows the fractions of treated water and the capacity loss of both cases assuming that they could produce 100 units of water if fed with deionized water. In other words, a clean membrane produces 100 units of water operated at its pure water flux, J_0. The membrane receiving raw grey water suffered from immediate pore blocking and this resulted in a production of treated water of only 28% of the overall capacity, or 28 units. Thus, the remaining 72% of the capacity was lost. On the other hand, when a sand filtration step was carried out as a pretreatment before membrane filtration, membrane performed better and was able to produce 52 units of treated water, which is almost twice as much.

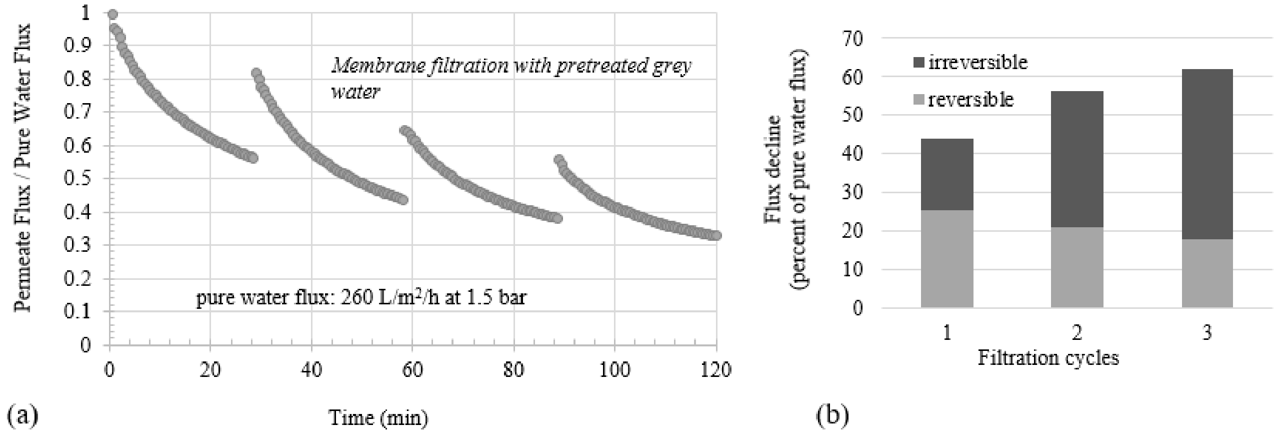

Figure 2. Flux decline during filtration of pretreated grey water (a) and recovery during membrane cleaning (b)

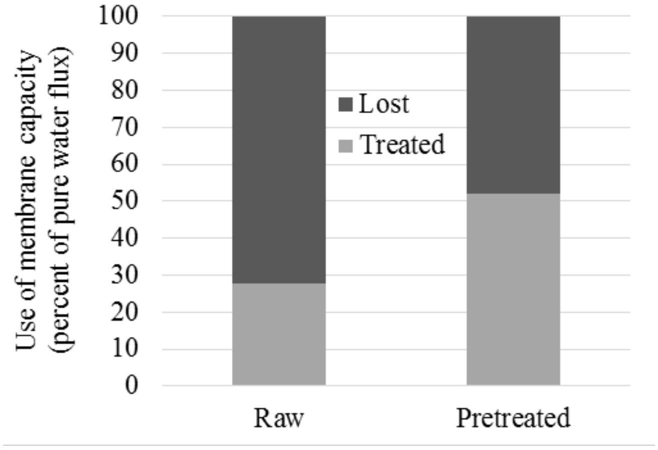

Figure 3. Use of membrane capacity during membrane filtration of grey water with and without pretreatment

The results of this study reveal that, depending on the characteristics of grey water, biological treatment may not be necessary if the organic matter is mostly in particulate form. This study gave 90% and 70% removal efficiencies in terms of BOD and COD respectively when membrane filtration was applied directly, while the removal efficiencies were 90% and 79% when sand filtration was applied prior to membrane filtration. These efficiencies are lower as compared to one of the work that applied membrane filtration to apartment mixed grey water giving 84% COD removal (Holden and Ward, 1999), and are higher as compared to other ultrafiltration work in the literature, which states 54% COD removal for laundry grey water (Sostar-Turk et al., 2005). Filtration of shower grey water from a sports complex through 0.025 mm ended up with 82% BOD, 56% COD and 35% TSS removal efficiencies (Santos et al., 2012), which are lower as compared to this work. Moreover, application of sand filtration and sedimentation (March et al., 2004) gave lower efficiencies in terms of COD and TSS.

On the other hand, literature work on treatment of light grey water collected from a house through screen, RBC and sand filtration (Friedler et al., 2005) ended up with removal rates similar to those mentioned above. Treatment efficiencies for an MBR treating shower grey water originating from a sports complex were reported as 93% BOD and 85% COD (Merz et al., 2007) which is compatible with the results obtained in this present work.

The differences in various sources mainly originate from variations in the characteristics of grey water samples that the studies were carried out with, which may have implications on whether a biological unit is needed for the treatment as opposed to physicochemical treatment only, and the treatment systems selected for that specific grey water.

Overall, the results show that the choice of the treatment scheme and its success is highly dependent on the characteristics of the grey water especially the ratio of particulate form of organic matter, and its biodegradability.

CONCLUSION

Reclamation of light grey water from baths/showers and wash basins in a five-star hotel through physicochemical treatment via membrane filtration was investigated in this work, specifically focusing on the efficacy of this physicochemical treatment method employing membrane filtration, and the effect of pretreatment using sand filtration.

Major conclusions from experimental results are as follows:

- Membrane filtration as a physicochemical process, with or without a sand filter is successful in providing effluent quality meeting organic matter, suspended solids and coliform targets for grey water reclamation with treatment levels of 70-79 for COD, 90% for BOD, and 100% for both suspended solids and coliform.
- Evaluation of the performance of the laboratory-made ultrafiltration membrane unit revealed that the use of a rapid sand filter pretreatment step prevents immediate clogging of membrane pores although sand filtration did not have a major contribution regarding effluent quality. Retardation of early clogging of the membrane through the use of a sand filter pretreatment unit provided nearly two times higher treated water volume at the end of the first hour as compared to the one without the sand filter pretreatment.

The results obtained point out further that the success of treatment is highly related to the characteristic of grey water to be treated. Moreover, biodegradability and particulate fraction of organic matter are two important factors to determine the proper treatment method. These results also point at the significance of characterization of raw grey water quality in terms of the selection of the best treatment option.

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