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Treatment of household grey water with a UF membrane filtration system

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ABSTRACT

As water is becoming a rare resource, the onsite reuse and recycling of grey water are practiced in many countries as a sustainable solution to reduce the overall urban water demand. In this paper, a decentralized grey water treatment system, which used a submerged spiral-wound ultrafiltration (UF) membrane module, was studied. This grey water treatment system aimed to treat and recover the resources present in the grey water. The study revealed that the UF membrane filtration system was able to maintain a permeate flux between 6 and 10 $L/m^2/h$. TOC can be reduced from the influent value of 161 to 28.6 mg/L in the permeate, meaning an average elimination rate of 83.4%. In addition, soluble nutrients such as ammonia and phosphorus can pass through the UF membrane and remain in the permeate. The total nitrogen and total phosphorus in the permeate were 16.7 and 6.7 mg/L respectively. The permeate was low in turbidity (below 1 NTU) and free of suspended solids and had an excellent physical appearance. Around 40 organic substances were tentatively detected by the non-target GC/MS analysis in the permeate. Phthalates, flame-retardants, and several other unknown substances were identified as the major detected trace organics. The concentrations of trace organic substances in the permeate were found to be comparable with those of the secondary municipal effluents. The permeate can be used in gardening and agriculture for irrigation and soil fertilization or alternatively for toilet flushing after disinfection. The retentate generated in this system can be treated with blackwater and kitchen waste in an anaerobic digester at a later stage for producing biogas or compost.

Keywords: Ecological sanitation; Grey water; Wastewater reuse; Ultrafiltration membrane; Trace organics

1. Introduction

Access to enough water of sufficient quality is a must for all human, animal, and plant life as well as for most economic activities. However, unprecedented population growth, coupled with ever-increasing urbanization and in many cases a parallel rise in specific water demand, has resulted in water scarcity in many regions around the world [1]. Ongoing drought conditions and global climate changes worsen this situation worldwide, especially in the arid and semiarid regions. In addition, water pollution caused by uncontrolled discharge of human wastes has exceeded the self-purification ability of the natural water cycle. As a result, the quality of many potable water resources has constantly deteriorated. Water scarcity, declining water quality and water related disasters are the three main concerns related to current and future water resources. One solution to these problems is to treat wastewater to a sufficient quality so that it could be put to

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beneficial use rather than simply pollute surface and ground water.

Grey water is defined as urban wastewater including sources from baths, showers, hand wash basins, washing machines, dishwashers and kitchen sinks, but excluding input from toilets [2–5]. The volume of grey water produced varies according to lifestyles, living standards, population structures (age, gender), customs and habits, and water installations. Mehlhart [6] reported that in Germany a grey water daily production of up to 70 L per person may be taken as a calculation basis for new buildings or buildings where sanitary equipment has been refurbished. Grey water constitutes 50–80% of total household wastewater [1,7,8]. In terms of organic contents, grey water shows similar characteristics to that of all municipal wastewater [4,9,10]. As urine is not included in grey water, it has a limited amount of nitrogen, mainly deriving from organic nitrogen in particulate form (80%–90%) [11]. The phosphorus concentration in grey water is similar to that in the entire municipal wastewater [11,12]. Due to the low contamination level of pathogens and nitrogen, reusing grey water for non-potable purposes is receiving more and more attention [12].

A number of technologies have been applied for grey water treatment worldwide, varying in both complexity and performance. Year-long operating experiences have proven that organic pollutants in grey water can be removed efficiently through biological treatments [12–15]. One disadvantage of biological treatments however is that nutrients, which are beneficial for plant growth, are partially lost. Organic materials, which can be applied for biogas production, are also eliminated.

In this study, a grey water treatment system equipped with a submerged UF membrane module was used to recover the resources present in grey water. The permeate generated from this system was not only low in microorganisms and free of suspended solids but also rich in soluble nutrients like ammonia and phosphorous. It therefore can be reused for irrigation or alternatively for toilet flushing after disinfection. The retentate generated from this system contained both organics and nutrients and can be later treated together with black water and kitchen waste in an anaerobic digester for producing biogas and soil conditioner.

2. Materials and methods

2.1. Grey water

The grey water used in this study was collected from the ecological settlement in Flintenbreite, Germany, where grey water and black water are separately collected at the source. This system includes wastewater from baths, showers, hand washbasins, washing machines, dishwashers, and kitchen sinks. The grey water was drained by gravity to a two-stage septic tank with an average retention time of 2 days. This was carried out as a pretreatment step, in order to remove larger particles, hair, oil and grease. The effluent from the septic tank was then fed into the submerged spiral-wound UF membrane filtration system. Samples of the effluents of the two-stage septic tank and the UF membrane filtration system were taken from March to May 2007. All samples were taken in glass bottles and transported immediately at 4°C to the laboratories.

2.2. Experimental set-up and procedure

A schematic diagram of the grey water treatment system is shown in Fig. 1. This grey water treatment consists of three compartments, each with an effective volume of 0.5 m^3. The first, second and third compartments acted as the balance tank, the membrane filtration tank and the permeate collection tank respectively. An open channel spiral-wound membrane module (Rochem UF System) with a total membrane surface of 8.2 m^2 and a normalized pore size of 0.0062 μm was submerged into the membrane filtration tank. This membrane filtration system was run at a constant transmembrane pressure (TMP) of 0.12 bars. The submerged membrane filtration system was operated under a filtration mode of 10 min filtration followed by 30 s backwashing with permeate. Once the concentration of suspended solids in the membrane filtration tank reaches around 3000 mg/l, the filtration cycle is stopped and the retentate is discharged and can be treated in an anaerobic digestion system.

2.3. Analytical methods

The total organic carbon (TOC) and total nitrogen (TN) levels were analyzed with a TOC/TN analyser (multi N/C3000, Analytic/Jena). NH4-N, NO2-N, NO3-N were measured using a reflectometer (Merck). The total phosphorous (TP) and orthophosphate levels were measured using cuvette tests (Dr. Lange). Electrical conductivity, temperature and the pH level were measured using probes. The turbidity was measured with a Hach 2100P portable turbidity meter. The suspended solids were analyzed using the German Standard Method DIN38414. For the analysis of trace organics, a volume of 220 ml of each sample was stirred and extracted three times with dichloromethane (analytical grade). The combined extracts were dried with sodium sulphate and concentrated to 1 ml by means of vacuum evaporation. Extracts were analyzed with a gas chromatograph Agilent 6890N equipped with a mass selective detector 5975B; injector: Gerstel KAS 4, 300°C; column: HP-5 ms, 30 m, I.D. 0.25 mm, film thickness 0.25 μm; carrier gas: helium,

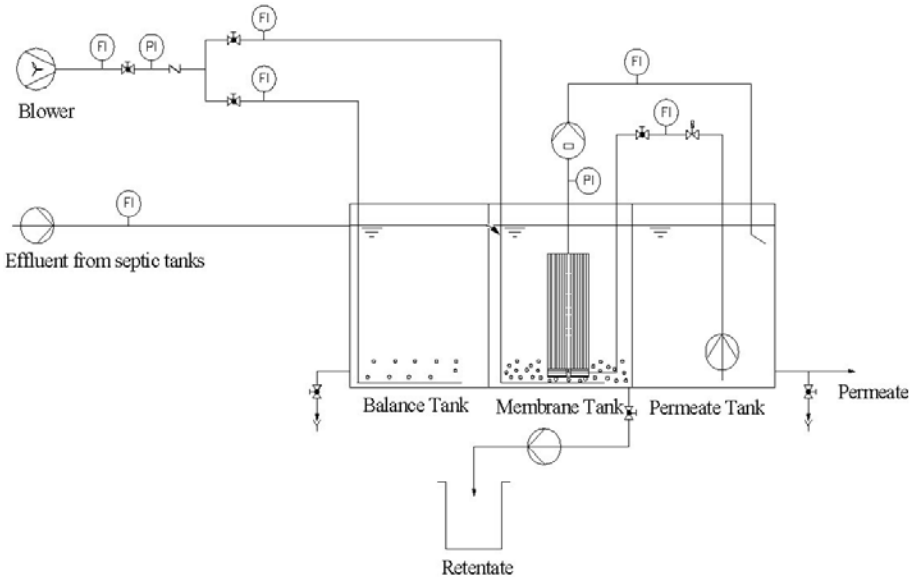

Fig. 1. Schematic diagram of the grey water treatment system.

1 ml/min; temperature program: 70°C (2 min), 8°C/min, 290°C (15 min); ionization: EI 70 eV, MS source 230°C. The mass spectra obtained were compared with a computerized spectra library NIST 05a. Identification of detected compounds was performed through comparison of the recorded mass spectra with a computerized mass library. The compounds were considered qualitatively identified if their spectra and retention time correlated with the spectra from the computerized mass library (overlap larger than 90%).

3. Results and discussion

The analysis of the major parameters, including temperature, pH, turbidity, electrical conductivity, TOC, TN, NH_4-N, organic N, NO_3-N, NO_2-N, TP, ortho PO_4-P and particulate PO_4-P in feed and permeate, was performed and the characteristics of the influent and effluent are summarized in Table 1.

Table 1
Compositions of the influent grey water and the permeate

Parameter	Influent	Permeate
Temperature (ºC)	20	21
pH	7.5	7.2
Turbidity (NTU)	140	0.5
Electrocal conductivity (μS/cm)	1115	1200
TOC (mg/l)	161	28.6
TN (mg/l)	16.5	16.7
NH_4-N	10.1	11.8
Organic N	6.4	4.5
NO_3-N	<0.01	<0.1
NO_2-N	<0.01	<0.1
TP (mg/l)	9.7	6.7
Ortho PO_4-P (mg/l)	7.5	5.9
Particulate PO_4-P (mg/l)	2.2	0.6

3.1. TOC removal

Our results showed that the TOC was reduced by 83.4% during the observation period. Fig. 2 shows that the TOC was reduced from the influent value of 161 mg/l to 28.6 mg/l in the permeate. The soluble TOC (samples filtered through the 0.45 μm) was 55 mg/l, which equals a reduction of 66% compared to the total TOC. As the median membrane pore size selected in this study was smaller than 0.45 μm, all suspended TOC, most of the colloidal TOC and part of the soluble TOC were rejected by the membrane. Ramon et al. [16] reported a low-strength grey water treatment system with direct nanofiltration membrane. In this study, the membrane with a molecular weight cut off of 0.2 kDa achieved an organic removal rate of 93%, indicating that the pore sizes of the membrane used have an important impact on the organic removal efficiency.

After every 2 weeks of filtration, the retentate TOC in the membrane tank increased continuously to around 890 mg/l at the end of each filtration cycle. The TOC in the supernatant, obtained by centrifuging the mixed liquor at 4000 rpm for 10 min also showed an increasing trend in the membrane filtration tank (Fig. 2). Despite that the retentate TOC and supernatant TOC levels kept rising, the level of the TOC in the permeate remained relatively constant as shown in Fig. 2. A satisfactory and above-average removal of organics by direct UF membrane

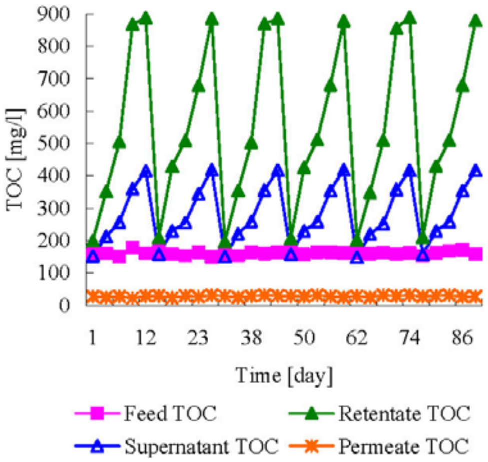

Fig. 2. Reduction of TOC by the submerged UF membrane filtration system.

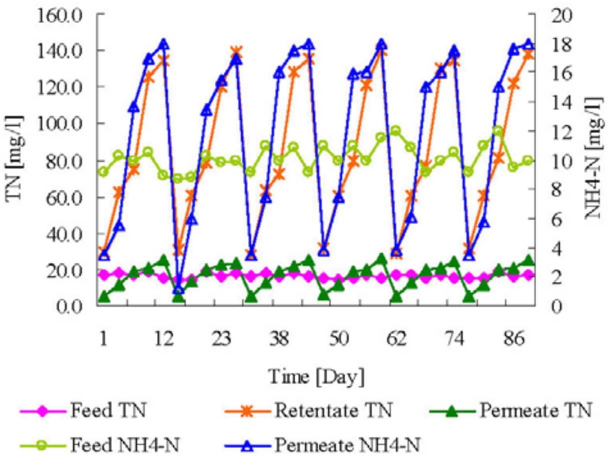

Fig. 3. Reduction of nitrogen by the submerged UF membrane filtration system.

filtration was achieved, confirming that the suspended TOC and the colloidal TOC constitute the majority of total TOC in grey water. The results obtained in this study were contradictory to Jefferson's findings [2], which claimed that grey water is relatively low in suspended solids, and a greater proportion of the contaminants are dissolved. At the end of each filtration cycle, the concentration of suspended solids in the membrane filtration tank increased to around 3000 mg/l. The concentrations of TOC and suspended solids in the retentate were lower than the theoretical values calculated by mass balance, which indicate that a partial biodegradation of the organic substances occurred as a result of introducing aeration.

3.2. Nitrogen removal

The TN concentration in our study varied from 14.2 mg/l to 18.8 mg/l with an average value of 16.5 mg/l, whereby ammonia nitrogen constituted more than 50% of the total nitrogen (Fig. 3). The study led by Elmitwalli and Otterpohl [11] revealed that the average TN concentration in grey water was 27.2 mg/l and the NH_4-N concentration was 4.2 mg/l, showing that grey water had a limited amount of nitrogen, which mainly was in particulate form (80–90%), and that ammonia nitrogen constituted only 10–20% of the total nitrogen. The concentration of the nitrogen contents is lower in grey water compared to household wastewater since no urine is present [17]. The nitrogen in grey water mainly originates from protein contained in food residues, household cleaning products, and personal care products [18]. In comparison with Elmitwalli and Otterpohl's study [11], the TN was reduced by 40% owing to the sedimentation of particulate organic nitrogen in the septic tanks. The increased ammonia concentration can be explained by the fact that part of the particulate organic nitrogen was reduced to ammonia nitrogen under the anoxic condition in the septic tanks. As for this membrane system, there was very little effect on removal of nitrogen. The average TN in permeate was 16.7 mg/l. As seen in Fig. 3, the particulate organic nitrogen was rejected by the membrane, letting the TN concentration in the retentate rise continuously. As the dissolved oxygen (DO) concentration in the membrane tank was maintained at a low level (DO <1 mg/l), the particulate organic nitrogen might be reduced to ammonia nitrogen, causing a higher ammonia concentration in the permeate.

3.3. Phosphorus removal

The mean total phosphorus (TP) and orthophosphate concentrations found in the feed water were 9.65 mg/l and 7.49 mg/l (see Fig. 4). Due to the continued use of phosphate-containing detergents, the total phosphorus concentration in grey water is equal to the whole household wastewater. The average TP concentration in Elmitwalli and Otterpohl's study [11] was 9.8 mg/l, of which the orthophosphate concentration and particulate phosphorus concentration constituted 8.0 mg/l and 1.8 mg/l, respectively. Compared with the study of Elmitwalli and Otterpohl [11], the septic tanks had no influence on the removal of phosphorus. This can be explained by the fact that particulate phosphorous in grey water constitutes less than 30% of the TP, and is largely in colloidal form which means that it cannot be removed effectively by sedimentation in the septic tanks. Due to the use of phosphorus in detergents, the phosphorus concentration in grey water is present at similar level compared to all municipal wastewater. Similar to nitrogen, membrane filtration also had limited effect on removing phosphorous since orthophosphate passes through the membrane and was found in the permeate. The average

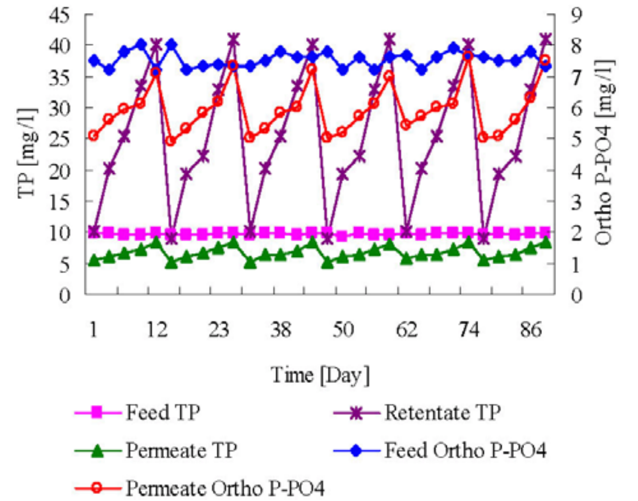

Fig. 4. Reduction of phosphorus by the submerged UF membrane filtration system.

TP in permeate was 6.7 mg/l with an average orthophosphate concentration of 5.9 mg/l. At the end of each filtration cycle, the retentate TP increased continuously to around 40 mg/l.

3.4. Membrane flux

As shown in Fig. 5, the submerged membrane filtration system was able to maintain an initial permeate flux of 10 $l/m^2/h$. The flux decreased from the initial value of 10 l/m^2h to around 6 $l/m^2/h$ after 2 weeks operation. This system was run at a constant TMP of 0.12 bars.

Rather than suspended solids, dissolved organic matter was found to be a major cause for membrane fouling. Soluble and colloidal matter are assumed to be responsible for membrane pore blockage, whilst suspended solids account mainly for cake layer resistance, which can be removed by the cross flow effect introduced by the intermittent aeration. It was observed that the dissolved organic carbon (DOC) (filtrated through a *0.45 μm* membrane) in the membrane filtration tank increased from the initial value of 55 mg/l to nearly 200 mg/l at the end of each filtration cycle (not shown). This study further showed that the decrease in the membrane flux corresponded very well with the rise in the DOC concentration in the membrane filtration tank. A backwashing cycle with permeate was carried out for 30 s after each 10-min filtration cycle to remove the reversible membrane fouling caused by gross solids attached to the membrane surface and by colloids in the membrane pores. At the end of each filtration cycle, the membrane was maintained by alkaline chemical cleaning in order to remove any irreversible membrane fouling. After the maintenance the membrane flux recovered to the initial value.

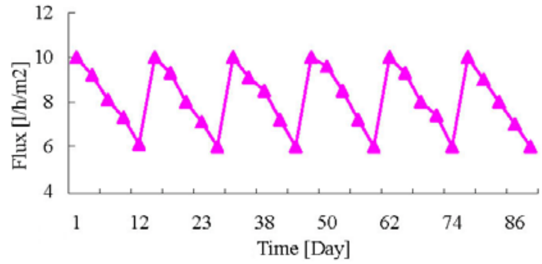

Fig. 5. Variation of the membrane flux in two filtration cycles.

In the study from Lesjean and Gnirss [14], a submerged plate and frame membrane bioreactor (MBR) grey water treatment unit was operated under low SRT and low HRT condition and achieved a stable permeate flux of 7 $L/m^2/h$. The membrane flux achieved in this study was comparable to that from Lesjean and Gnirss [14]. However, the duration of each filtration cycle was relatively short. This study was in agreement with the results from Ahn et al. [19], who demonstrated that the MBR system performed better than the direct membrane filtration system in terms of filtration characteristics.

3.5. Removal of conductivity, turbidity and colour

Many soluble salts, deriving from the drinking water supply, human diet and household chemicals, have been found in grey water. Most of the soluble salts are sodium compounds, such as sodium chloride used in the human diet, sodium nitrate used as meat preservatives and in food preparation, and sodium sulfate, sodium tripolyphosphates and sodium carbonates used in laundry products [20,21]. The concentrations of soluble salts stand in close correlation with both electrical conductivity and sodium adsorption ratio (SAR). The conductivity of the feed water, retentate and the permeate in our study showed insignificant variation, ranging from 1100 to 1200 μS/cm. Application of irrigation water with excessive sodium in relation to calcium and magnesium concentrations can destroy the soil structure and may be toxic to some crops. The metal elements in the feed and the permeate were not analyzed in our study. However, based on the study conducted by Palmquist and Hanæus [22], the SAR of the separately collected raw grey water from ordinary Swedish households is 3.37, which poses no risks for irrigation use. Turbidity was reduced from 140 NTU in the feed water to less than 1 NTU in the permeate. The removal of up to 100% of turbidity by membranes has been observed [16,23]. Due to the exclusion of urine in grey water, the permeate was colourless.

3.6. Trace organic substances in the permeate

The non-specific parameters such as TOC are insufficient to perform a valid health risk evaluation. Therefore,

Table 2
Organics tentatively identified in the permeate

Compounds (retention time)	Semi-quantitative characterisation	Group (possible sources)
Ethyl citrate (16.43)	Trace	Fragrance
3-Oxo-2-pentyl- methylester cyclopentyacetic acid (16.34)	Minor	Fragrance
Methylbenzylalcohol (6.71)	Trace	Flavour (food)
3,7-Dihydro-1,3,7-trimethyl-1H-purin-2,6-dione (18.99)	Trace	Flavour (caffeine)
Piperine (29.90)	Minor	Flavour (food)
Dipropylene glycol (9.83, 9.92)	Trace	Personal care product
Benzophenone (16.16)	Trace	Cosmetic UV filter
Squalene (29.12)	Trace	Cosmetic
Dodecanoic acid (14.92)	Minor	Surfactant
Tetradecanoic acid (17.7)	Minor	Surfactant
Hexadecanoic acid (20.25)	Minor	Surfactant
2-Phenoxyethanol (9.47)	Trace	Preservative in personal care product
2-Methylthio-benzothiazole (15.69)	Trace	Preservative (intermediate degradation product)
1-Chloro-2-propanol phosphate (3:1) (18.35)	Major	Flame retardant
2-Butoxyethanol phosphate (3:1) (25.28)	Major	Flame retardant
Dimethyl phthalate (13.41)	Trace	Softeners and plasticizers
Diethyl phthalate (15.51)	Trace	Softeners and plasticizers
N-Butylbenzol- sulfonamid (NBBS) (18.14)	Minor	Softeners and plasticizers neurotoxic
Phthalate (19.19, 20.37, 26,61)	Major	Softeners and plasticizers
Benzylbutyl phthalate (24.70)	Trace	Softeners and plasticizers
1H-Benzotriazole (13.58)	Trace	Anticorrosive chemical (carcinogenic)
Hexahydro-methano-1H-indol (10.52)	Minor	Medicine (intermediate degradation product)
Ibuprofen (16.76)	Trace	Medicine (anti-inflammatory)

Note: Ratio of peak area to area of largest peak (“main”) in the respective chromatogram below 5%: “trace”; 5–20%: “minor”; 20–100%: “major.

there is a need to screen the organic compounds in the permeate. The identification of the organics by their mass spectra was tentative in this study and results were not verified by re-analyzing reference solutions of the pure compounds. However, the identification results were very plausible, because many of the identified substances are contained in materials like household chemicals, personal care products and foodstuffs, which partially end up in grey water.

Around 40 organic substances were detected in the permeate of the direct membrane filtration system. Table 2 shows that phthalates, flame-retardants, and several other unidentified substances (at retention time of 22.50 min, 23.10 min, 23.42 min, 30.99 min, and 38.18 min) were found to comprise the majority of the organic substances in the permeate. Comparison with the quantitative GC/MS analyses performed with municipal effluents [24] leads to the assumption that the concentrations of organics detected by GC/MS in the permeate are in the range of μg/l. The removal of organic pollutants in the submerged UF membrane filtration system occurs mainly by means of physical filtration and air stripping while biochemical transformation plays a minor role.

Phthalates are extensively used as plasticizers in a wide array of household plastic materials and were discovered in the Swedish grey water [22]. The presence of non-volatile lipophilic phthalates in the permeate indicates that phthalates are ubiquitous and are difficult to remove. There are concerns that human exposure to some phthalates might adversely affect male reproductive health [25].

The less volatile flame retardants (1-Chloro-2-propanol phosphate (3:1) and 2-Butoxyethanol phosphate (3:1)) were present in the permeate in major concentrations. Around 95% of 1-Chloro-2-propanol phosphate (3:1) is used in rigid polyurethane foam plates in the walls of buildings for thermal insulation. 1-Chloro-2-propanol phosphate (3:1) is of low to moderate toxicity for oral, dermal, and inhalation routes. 2-Butoxyethanol phosphate (3:1) is applied as a wood floor polish agent.

Fatty acids and fatty acid esters were found in the permeate of the UF membrane filtration system. Being relatively hydrophobic, these compounds are attached to particles and can be partially retained by the UF membrane as well. Many fatty acids and fatty acid esters are used in the manufacture of detergents, personal care products and food oils.

Squalenes, present at higher levels in the influent, were removed partially in the permeate. Squalenes are similar to hormones like cortisone, estradiol and progesterone. Consequently, the substances may have hormonal effects.

Due to the introduction of air in the membrane filtration tank, the stripping process eliminated many volatile fragrances. The persistent fragrances, such as 3-Oxo-2-pentyl- methylester cyclopentyacetic acid and ethyl citrate were identified in the permeate at trace levels.

The polar and less volatile benzophenone, which is used as a UV filter was identified in the permeate at trace concentrations. Though the grey water samples were taken from March to May 2007, benzophenone was found in the permeate. To avoid sunburns and skin cancer, a large number of sunscreen creams have come into use. In addition, sunscreen creams are also used in many personal products such as beauty creams, hair sprays, shampoos, and shower gels for product stability as well as in products such as plastics, clothing, and varnish [25]. UV filters are washed off directly during bathing, or are transferred to towels or clothes that will eventually be washed, releasing these chemicals into the grey water. It has been reported that some UV filters exhibit estrogenic or anti-androgenic activity [26,27].

As urine and faeces are not included in the grey water stream, the introduction of unchanged pharmaceuticals and metabolites through urine and faeces is avoided. Therefore, pharmaceuticals present in grey water should be lower than in the mixed wastewater. However, the quantitative analysis showed that pharmaceuticals, such as Hexahydro-methano-1H-indol and Ibuprofen (anti-inflammatory) were identified in the permeate at low levels. The presence of pharmaceuticals can be explained by body excretions that take place either during a shower, brushing of teeth, or during general washing (as present in the mouth or on the skin or excreted as urine). Dermatological creams can also be washed during bathing and, as such, may end up in the grey water.

4. Conclusions

The study shows that direct membrane filtration with a submerged spiral-wound module is able to achieve satisfactory results in terms of water quality, process stability and membrane flux. The following conclusions can be drawn from this study:

1. The TOC was reduced from the influent value of 161 mg/L to 28.6 mg/l in the permeate with an average reduction rate of 83.4%. In addition, soluble nutrients like ammonia and phosphorous can pass through the UF membrane and remain in the permeate. The total nitrogen content and total phosphate content in the permeate were 16.7 mg/l and 6.7 mg/l respectively. The permeate was low in turbidity (below 1 NTU), free of suspended solids and had an excellent physical appearance.

2. The quality of the permeate obtained in this study did not meet EU guidelines for bathing water due to the slightly higher concentration in organic substances. The permeate however was rich in nutrients and free of suspended solids. The retentate generated from this system can later be treated together with black water and kitchen waste in an anaerobic treatment system to produce biogas and soil conditioner.

3. One of the main advantages of using this permeate is that it can enhance the fertility of the land to which it is applied. The reclaimed grey water can also be used for toilet flushing after disinfection. This would reduce the total household consumption of potable water by approximately 25% and allow the nutrient contents in grey water to be added to urine.

4. In the permeate, phthalates, flame-retardants, and several other unidentified substances were identified to be the majorities of the detected organics.

5. The concentrations of trace organics in the permeate are found to be comparable to those of the secondary municipal effluents. With respect to trace organics, reuse of the permeate for restricted non-potable purposes, such as toilet flushing and irrigation should not be a problem.

6. In order to meet the non-restricted water reuse standard, advanced treatment methods such as reverse osmosis and advanced oxidation can be applied to eliminate the residual trace organics in the reclaimed grey water.

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