



## **TREATMENT OF HOUSEHOLD GREY WATER WITH A SUBMERGED UF MEMBRANE FILTRATION SYSTEM**

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### **ABSTRACT**

In an ecological sanitation system, the household wastewater is separated on a household level into three streams: grey water, urine and faeces. Grey water with low contamination of pathogens and nitrogen represents 50-80% of total household wastewater and can be reused as a valuable water resource for non-potable purposes after proper treatments. This paper studies a resource and nutrient oriented decentralized or semi-centralized grey water treatment system, which uses a submerged spiral wound ultra-filtration (UF) membrane module. The UF Membrane acts as an absolute barrier to suspended particles and pathogens greater than the size of membrane and is not subjected to the variation of grey water compositions and chemical shock, which could adversely affect the performance of the biological treatment processes. This membrane based grey water treatment system is aimed to recover, treat and reuse the resources present in grey water. The resources to be recovered from the system include water, nutrients and organic substances. The permeate, which is recycled from this system is low in organic substances and turbidity and is free of microorganisms and suspended solids. In addition, soluble nutrients like ammonia and phosphorous can pass through the UF membrane and remain in the permeate. The permeate from this membrane filtration system can then for example be used in gardening and agriculture for irrigation and fertilisation or alternatively for toilet flushing after chlorine disinfection. The retentate generated in this system is suggested to be treated with black water and kitchen waste in an anaerobic digester for producing biogas or compost. The paper finally discusses the filtration performance of the submerged UF membrane module.

**KEY WORDS:** Ecological sanitation; grey water; ultra-filtration membrane filtration

### **INTRODUCTION**

Population growth, coupled with ever-increasing urbanization, and in many cases a parallel rise in specific water demand, results in a continuously increasing demand for urban water in many regions around the world (Friedler and Hadari, 2006). Ongoing droughts and global climate change accentuate this worldwide situation and specifically in the arid and semiarid regions. 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. The volume of grey water produced varies according to lifestyles, living standards, population structures (age, gender), customs and habits, and water installations. Mehlhart *et al.* (2007)

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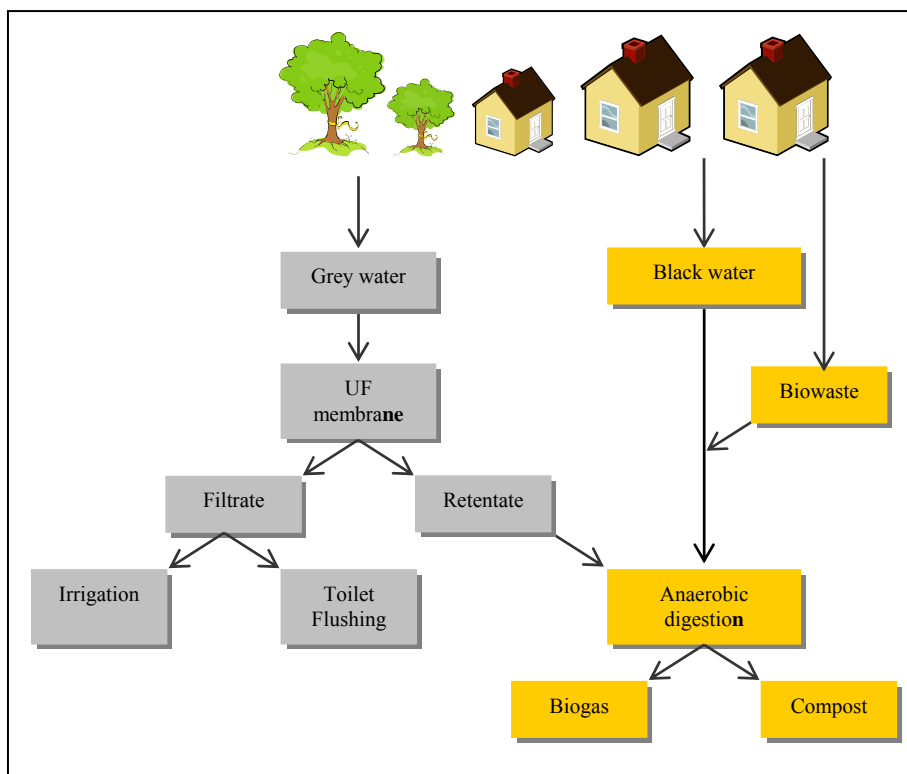


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reported that in Germany a grey water daily production of up to 70 liters per person may be taken as a calculation basis for new buildings or buildings where sanitary equipments have been refurbished. This constitutes 50-80% of total household wastewater (Eriksson *et al.*, 2003; Roesner *et al.*, 2006; Friedler and Hadari, 2006). In terms of organic strength, grey water shows similar characteristics to that of the entire municipal wastewater. As urine is not included in grey water, it has a limited percentage of nitrogen content, which mainly derives from organic nitrogen in particle form (80%-90%) (Elmitwalli and Otterpohl, 2007). The phosphorus concentration in grey water is similar to that in the entire municipal wastewater and ranges typically from 3 to 7 mg/l.

Due to the low contamination level of pathogen and nitrogen, reusing grey water for non-potable purposes is receiving more and more attention. A number of technologies have been applied for grey water treatment worldwide varying in both complexity and performance. Organic pollutants in grey water can be removed efficiently with biological treatment, as has been proven from yearlong experience gained with constructed wetland. The remaining issue was that the remaining micro-organisms and suspended solids are not low enough for the grey water to be directly reused for non-potable purpose. In order to meet these requirements, a slow sand filtration step and a disinfection step have to be applied after passing the treatment in constructed wetland. By applying the Membrane Biological Reactor (MBR) grey water treatment process the produced effluent meets the most stringent standards for non-potable water reuses. One disadvantage of both the constructed wetland and the MBR using biodegradation 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 addition, variation of grey water compositions coupled with a deficiency of macronutrients (nitrogen) adversely influence the treatment efficiencies of the biological treatment processes (Jefferson *et al.*, 1999).

In this study, a grey water treatment system equipped with a submerged UF membrane module is used to recover the resources present in grey water. The permeate generated from this system is not only free of micro-organisms and suspended solids but also rich in soluble nutrients like ammonia and phosphorous. It is therefore can be reused for irrigation or alternatively for toilet flushing after disinfection. The retentate generated from this system contains both organics and nutrients and can be later treated together with black water and kitchen waste in an anaerobic digester for producing biogas and compost. The concept of this decentralised wastewater management system is shown in Figure 1.



**Figure 1 Resources and nutrients oriented wastewater management system**

## MATERIALS AND METHODS

### Grey water

The grey water used in this study was collected from the ecological settlement in Lübeck/Flintenbreite, Germany, where grey water and black water are separately collected at its source. A semi-centralised sanitation concept is operated in a peri-urban area since the beginning of the year 2000. At present, 103 inhabitants are connected to the semi-central sanitation system. It includes wastewater from baths, showers, hand wash basins, washing machines, dishwashers and kitchen sinks. The grey water is drained by gravity to a double-step septic tank, which was applied for pre-treatment to remove larger particles, hair, oil and grease. The effluent from the septic tanks was then fed into the submerged spiral wound UF membrane filtration system.

### Experimental set up and procedure

A schematic diagram of the grey water treatment system is shown as in Figure 2. The submerged membrane grey water treatment unit consists of three compartments, each with an effective volume of 0.5 cubic meters. The first, second and third compartment act as the balance tank, membrane filtration tank and permeate collection tank. Air is introduced intermittently (40 second on/20second off, 3 Nm<sup>3</sup>/h) through a fine bubble membrane diffuser installed directly below the membrane module in order to provide an uplifting flow of bubbles which scour the membrane surface to prevent the membrane from fouling. A vacuum pump is applied to pump the permeate through the permeate collection channel to the permeate collection tank. A submerged pump placed in the permeate collection tank is used for backwashing the membrane. A ROCHEM open channel spiral wound membrane module with a total membrane surface of 8.2 square meters and a normalized pore size of 0.0062 µm is submerged into the membrane filtration tank. Once the concentration of suspended solids in the membrane filtration tank reaches 3000 mg/l, the filtration cycle is terminated and the retentate can be discharged and treated together with black water and kitchen waste in an anaerobic digestion system. This membrane filtration system was run at a constant trans-membrane pressure (TMP) of 0.12 bars. The submerged membrane filtration system was operated under a filtration mode of 10 minutes filtration followed by 30 seconds backwashing with permeate.

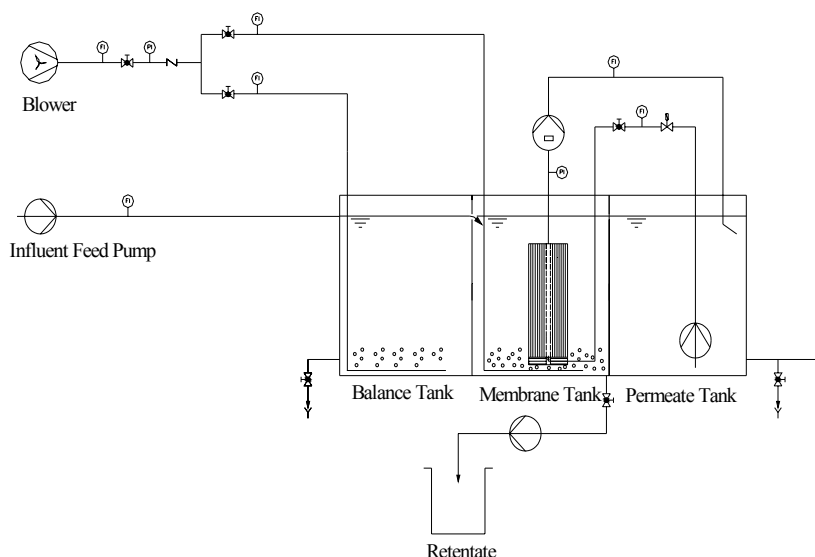


Figure 2 Schematic diagram of the grey water treatment system

### Analytical methods

The Total Organic Carbon (TOC) and Total Nitrogen (TN) levels were analyzed with a TOC/TN analyser (multi N/C3000, Analytic/Jena Company). NH<sub>4</sub>-N, NO<sub>2</sub>-N, NO<sub>3</sub>-N were measured using a reflectometer (Merck). The Total phosphorous (TP) and ortho phosphate levels were measured with 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 following the German Standard Method DIN38414. Chromocult Coliform Agar developed by German Firm Merck was used to determine the amount of *E. coli* in this experiment. Colonies of *E. coli* were characterised by dark-blue to purple spots after the sample had been incubated for 24 hours at a temperature of 36°C. The effluent of the septic tanks was centrifuged for 10 minutes at 3000 rpm to separate suspended solids from supernatant. The supernatant was filtered through a 0.45µm membrane filter to separate colloidal TOC from dissolved TOC. The total TOC (suspended TOC + colloidal TOC + dissolved TOC), supernatant TOC (colloidal TOC + dissolved TOC) and the filtered sample (dissolved TOC) were measured using the TOC analyser. The suspended TOC and colloidal TOC were calculated from the difference between the total TOC and supernatant TOC, and between the supernatant TOC and dissolved TOC. The dissolved organic carbon in permeate was characterized according to a Liquid Chromatography Organic Carbon Detection (LC-OCD) method (DOC-LABOR DR. HUBER). A HW50S column was used in the study due to its good resolution for organic substances with low molecule weight.

## RESULTS AND DISCUSSIONS

The analysis of the major pollutants, including TOC, TN, NH<sub>4</sub>-N, NO<sub>2</sub>-N, NO<sub>3</sub>-N, TP, pH, electrical conductivity, turbidity in feed and permeate was performed between March and May 2007. The characteristics of the influent and effluent are summarized in Table 1.

**Table 1 Compositions of influent (effluent of the double-step septic tanks) grey water and permeate**

| Parameter                          | Influent | Effluent |
|------------------------------------|----------|----------|
| Temperature (°C)                   | 20       | 21       |
| pH (-)                             | 7.5      | 7.2      |
| Turbidity (NTU)                    | 140      | <1       |
| Electro conductivity (µS/cm)       | 1115     | 1200     |
| TOC (mg/l)                         | 161      | 28.6     |
| TN (mg/l)                          | 16.5     | 16.7     |
| NH <sub>4</sub> -N                 | 10.1     | 11.8     |
| Organic N                          | 8.6      | 4.5      |
| NO <sub>3</sub> -N                 | <0.01    | <0.1     |
| NO <sub>2</sub> -N                 | <0.01    | <0.1     |
| TP (mg/l)                          | 9.7      | 6.7      |
| Ortho PO <sub>4</sub> -P (mg/l)    | 7.5      | 5.9      |
| Particle PO <sub>4</sub> -P (mg/l) | 2.2      | 0.6      |

### TOC fractions and TOC removal

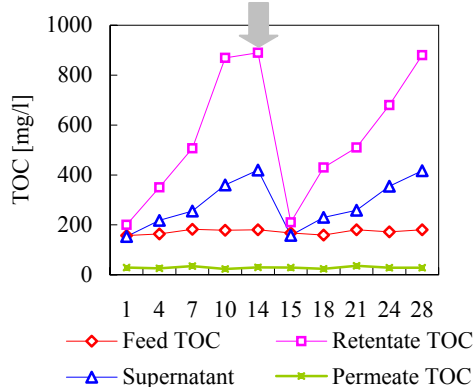
Our results showed that the suspended TOC, colloidal TOC and soluble TOC constitute 30%, 34% and 36% of the total TOC. Compared with the results of Elmitwalli and Otterpohl (2007), the particulate TOC (suspended + colloidal) was 16% lower. As an observation of the septic tanks showed, this was mainly a result of the suspended solids settling in the septic tanks. Figure 3 shows that the TOC was reduced by 83.4% and an average permeate TOC concentration of 28.6 mg/l was obtained during the whole observation period. The TOC of the sample 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, leading to a higher rejection rate. After two weeks of filtration, the retentate TOC in the membrane tank increased continuously from the initial value of 157 mg/l to around 890 mg/l at the end of each filtration cycle. An increasing trend in the membrane filtration tank was found also for the supernatant TOC. Despite that the retentate TOC and supernatant TOC levels kept rising, the level of the TOC in permeate remained relatively constant. A satisfactory and above-average removal of organics by direct UF membrane filtration was achieved, suggesting that the suspended TOC and colloidal TOC constitute the majority of total TOC in grey

water. Our results are contradictory to Jefferson's findings (1999), 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 from the initial value of 35 mg/l 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.

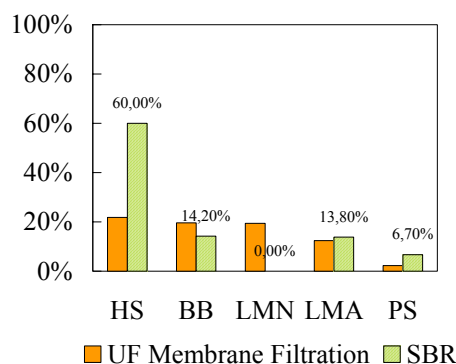
### LC-OCD chromatography

Liquid chromatography with on-line organic carbon detection (LC-OCD) was used to characterize the different organic fractions in the permeate. As shown in Figure 4, humic substance (HS) (21.8%), building blocks (BB) (19.6%), low molecular neutrals (LMN) (19.4%) and low molecular acids (LMA) (12.4%) are the major organic substances in the permeate. Polysaccharide (PS) represents only 2.3%, indicating that the biodegradability of the permeate is quite low. Other organic substances accounting for the remaining 24.5% cannot be detected by the LC-OCD method. Humic substances in the effluent from a Sequencing Batch Reactor (SBR) grey water treatment system covered 60% of the total chromatographable organic carbon (Li, 2004a), showing that biological treatment causes the formation of humic substances (Figure 4). Due to the low biodegradation level in the membrane filtration tank the transformation of easily biodegradable organics to humic substances, which negatively influences the colour of reclaimed water, was limited and their concentration in the permeate was very low. Figure 4 also shows that LMN which represent low molecular weight trace organics in the permeate are higher than that in the effluent of the SBR grey water treatment system.

Retentate discharge and membrane cleaning



**Figure 3 Reduction of TOC by the submerged UF membrane filtration system**



**Figure 4 LC-OCD chromatography of the permeate and the effluent from a SBR grey water treatment system (Li, 2004a)**

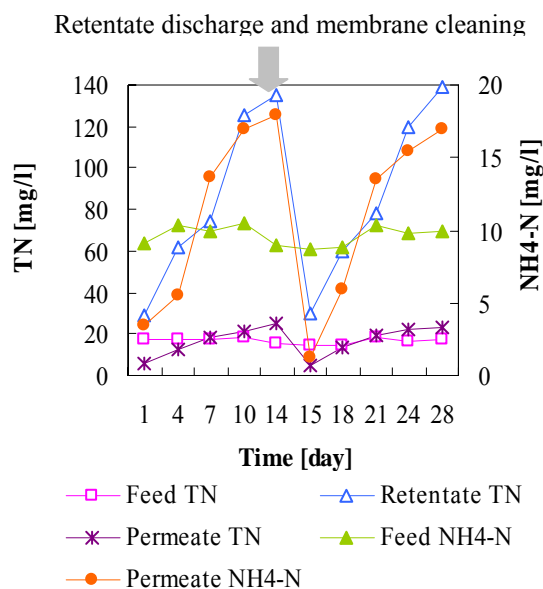
### Nitrogen removal

Elmitwalli and Otterpohl (2007) analyzed the raw grey water before it flowed into the septic tanks at the same source in Lübeck, Flintenbreite. The average TN concentration they found was 27.2 mg/l and the NH<sub>4</sub>-N concentration was 4.2 mg/l. Their study indicated that grey water had a limited amount of nitrogen, which mainly was in particle form (80–90%), and that ammonia nitrogen constituted just 10-20% of the total nitrogen. Particle nitrogen originates from protein contained in household cleaning products, personal care products and from kitchen sink. 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 (Figure 5). In comparison with Elmitwalli and Otterpohl's study (2007), the TN was reduced by 40% owing to the sedimentation of particle organic nitrogen in the septic tanks. The increased ammonia concentration can be explained by the fact that part of the particle organic nitrogen was reduced to ammonia nitrogen under the anaerobic condition in the septic tanks. As for this membrane filtration system however, there was very little effect regarding the removal of nitrogen. The TN in permeate was around 16.69 mg/l. As seen in Figure 5, the

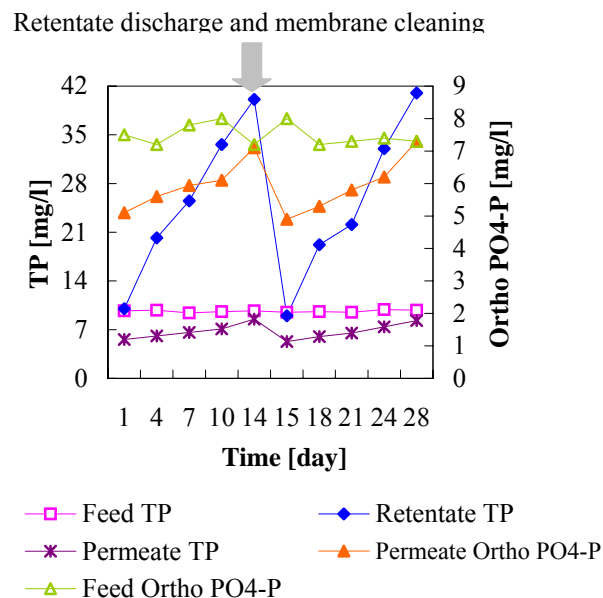
particle organic nitrogen was rejected by the membrane, letting the TN concentration in the retentate rise continuously. Parallel to the particle organic nitrogen accumulating in the retentate, the ammonia concentration in the permeate also increased. This is because the particle organic nitrogen, which was rejected by the membrane, was hydrolyzed to soluble ammonia nitrogen under aerobic condition in the membrane filtration tank. Since nitrifier bacteria cannot acclimatize in the membrane filtration tank in short time (two weeks), no significant nitrification was observed. To avoid the formation of nitrite and nitrate in the membrane tank, a fine bubble diffuser was directly located under the spiral wound module, plus the upper part of the membrane module was kept above water level in the membrane filtration tank. Therefore most of the supplied air flowed through the feed side channel in the membrane module to the atmosphere, which kept the dissolved oxygen concentration in the membrane tank at a maintained low level. However, nitrite was still found in the permeate during the observation period.

### 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 Figure 6). The average TP concentration in Elmitwalli and Otterpohl's study (2007) was 9.8 mg/l, of which the orthophosphate concentration and particle phosphorus concentration constituted 8.0 mg/l and 1.8 mg/l, representatively. Compared with the study of Elmitwalli and Otterpohl (2007), the septic tanks had no influence on the removal of phosphorus. This can be explained by the fact that particle phosphorous in grey water constitutes less than 30% of the TP, and is largely in colloidal form which results that it cannot be removed effectively by settlement in the septic tanks. 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 TP in permeate was 6.7 mg/l with an average orthophosphate concentration of 5.9 mg/l. The slightly risen concentration of orthophosphate in the permeate is explained by the rejected particle phosphorus which accumulated in the membrane filtration tank, plus part of particle phosphorous was hydrolyzed to orthophosphate under aerobic condition.



**Figure 5 Reduction of nitrogen by the submerged UF membrane filtration system**



**Figure 6 Reduction of phosphorus by the submerged UF membrane filtration system**

### Removal of conductivity, turbidity, color and pathogens

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 molecules, such as sodium chloride used in the



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human diet, sodium nitrate used as meat preservatives and in food preparation, and sodium sulfate, sodium tri-polyphosphates and sodium carbonates used in laundry products. The concentrations of soluble salts stand in close correlation with electrical conductivity. The conductivity of the feed water, retentate and the permeate in our study showed insignificant variation, ranging from 1100 to 1200  $\mu\text{S}/\text{cm}$ . Turbidity was reduced from 140 NTU in the feed water to less than 1 NTU in the permeate. Due to the low concentration of humic substances and exclusion of urine in grey water, the permeate was colourless. Chiemchaisri (1992) reported that a MBR installed with two types of membranes (pore size 0.1 and 0.03  $\mu\text{m}$ ) was able to achieve the same 4 to 6 log removal of the seeded Q $\beta$  coliphage at a stable stage although the membrane pore sizes are larger than the size of viruses (25 nm). Due to the smaller membrane pore size (0.0062  $\mu\text{m}$ ) used in this study, it is therefore not surprising that the permeate was free of color and SS, and that the existence of *E. coli*. could not be determined.

### Comparison of the system performance with Membrane Bioreactor (MBR) and constructed wetland

The MBR was proved to be the most advanced and appropriate technology for grey water recycling and reuse due to its ability to remove pathogens and its process stability (Pidou, 2006). Constructed wetland has been considered as one of the most environmentally friendly and cheapest technologies for decentralized grey water recycling. In the study led by Lesjean and Gnirss (2006), a MBR grey water treatment unit was operated under low SRT and low HRT conditions. The COD was reduced from the influent value of 493 mg/l to 24 mg/l in the permeate with an elimination rate of greater than 85%. The nitrogen concentration was lowered from 21 mg/l to 10 mg/l, however with an inconstant elimination rate which ranged from 20 to 80%. Phosphorus was removed by around 50%, decreasing from 7.4 mg/l in the influent to 3.5 mg/l in the effluent. The SS in the permeate was beneath 1 mg/l throughout the entire observation period. In parallel, the effluent from the septic tanks was fed into an intermittently operated vertical constructed wetland in Lübeck, Flintenbreite. The effluent of the constructed wetland had a comparably low TOC concentration (5-28 mg/l). Similarly, the TN concentration was below 5 mg/l and the TP concentration varied from 5.55-6.80 mg/l. *E. coli* bacteria concentrations ranged from 300-26200 /100ml (Li *et al.*, 2004b). Due to the higher organic load in the influent, the effluent of the constructed wetland in Lübeck, Flintenbreite was not able to meet a 10 mg/l BOD<sub>5</sub> standard constantly (Oldenburg, 2007). As biological degradation is insignificant in the direct UF membrane filtration tank, the removal rates for pollutants obtained in this submerged membrane filtration system were lower than in the results from both Lesjean and Gnirss (2006) and Li *et al* (2004b). The organic load of the reclaimed water however was low enough for the water to be used for irrigation. *E. coli* bacteria and suspended solids were not detectable in the effluent from the direct UF membrane filtration system. Instead of removing nutrients from wastewater, which requires additional costs and also increases the complexity of the treatment process, both the valuable nutrients and the reclaimed water could be recycled. The permeate from the submerged membrane filtration system was rich in organic and nutrients and was therefore more appropriate to be used for irrigation purposes. In addition, the nutrients concentration captured in the retentate was higher than in the sludge produced in the MBR system, so that the retentate could be later treated with black water and kitchen waste for producing biogas or compost.

### Membrane Flux

The submerged membrane filtration system was able to maintain an initial permeate flux of 10 l/m<sup>2</sup>/h. The flux decreased from the initial value of 10 l/m<sup>2</sup>/h to around 6 l/m<sup>2</sup>/h after two weeks operation. 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  $\mu\text{m}$  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. 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 seconds after each 10 minute 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

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each filtration cycle, the membrane was maintained in form of chemical cleaning in order to remove any irreversible membrane fouling. After the maintenance the membrane flux recovered to the initial value.

### **Conclusions and future work**

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 obtained quality of the permeate quality in this study did not meet EU guidelines for bathing water due to the exceeding concentrations in organic substances and nutrients. The permeate however was rich in nutrients and free of bacteria and 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 compost. The submerged membrane filtration system can be applied for grey water reuse in decentralized regions and areas with a low population density where the use of centralised wastewater treatment systems becomes cost inefficient due to the high investment costs for constructing, operating and maintaining a long pipe network to collect and transport wastewater. Thanks to the membrane filtration system, the majority of wastewater can be recycled by capturing essential nutrients for plant growth and agricultural production. Unless the water quality requirements regarding the concentration of organic material is obligatory ( $BOD_7 < 5$  mg/l) (Mehlhart *et al.*, 2007), the reclaimed grey water in the study can also be used for toilet flushing after chlorine disinfection. This is because the residual organic substances in the reclaimed grey water and the optimal temperature in the water transportation and storage system can create a milieu for pathogens to grow. To avoid secondary pathogen contamination, a minimum residual chloride concentration of 0.25 mg/l should be maintained in the water transportation and storage system. This would reduce the total household consumption of potable water by around 25%, and allow for the nutrient contents in grey water to be added to urine. This however needs to be evaluated on a pilot scale test, as the formation of disinfection by products and unpleasant odour might occur.

Hazardous substances like heavy metals and trace organic substances derived from household chemicals, personal care products (PCPs), and pipe corrosion have been detected in grey water at very low concentrations (Palmquist and Hanæus, 2005). These substances however may pose a potential health and environment risk if the reclaimed grey water is used for garden and agricultural irrigation, therefore there is need to study the fates of these hazardous substances as they undergo this direct UF membrane filtration. To avoid the formation of nitrite in the membrane tank, the submerged membrane filtration system, in which air is collected from the top of membrane filtration tank and recycled for further membrane scouring, is closed. This largely reduces the oxygen content in the supplied air and the formations of nitrite and nitrate are avoided in the permeate. Due to the reduced dissolved oxygen concentration in membrane filtration tank, degradation of the organic carbon is reduced. Finally, the use of recycled air also saves on costs for energy consumption. This concept however is still to be tested on a pilot scale, where water quality can be set against operating costs.

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