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Impacts of NF concentrate recirculation on membrane performance in an integrated MBR and NF membrane process for wastewater treatment

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ABSTRACT

As water shortages are increasing, the need for sustainable water treatment and the reuse of water is essential. Water reuse from wastewater can be accomplished in a membrane bioreactor (MBR) in the secondary activated sludge stage of a wastewater treatment plant. To remove viruses, dissolved organics and inorganics still present in the MBR permeate, nanofiltration (NF) can be applied. Nevertheless, the major drawback of nanofiltration membranes is the production of a concentrate stream that cannot be discharged to the environment. In this research we investigate the concept of a combined MBR and NF system with NF concentrate recirculation back to the MBR to produce reusable water in a sustainable way. Long-term continuous operation (1 year) shows that the NF permeate quality is not impacted by the recirculation. Fouling on the NF membrane is mostly the result of inorganics, while organics (e.g. humic acids) do not have a major impact on NF fouling. In fact, the flux of the NF was enhanced by the presence of humic acids due to recirculation. However, the MBR showed increased fouling and consequently more frequent membrane cleaning. The results presented show that the continuous production of reusable water from wastewater in a combined MBR and NF process with NF concentrate recirculation can be successful.

1. Introduction

As fresh water shortages are increasing, the need for sustainable water treatment processes has become essential. For example, water reuse from municipal wastewater could be accomplished by a membrane bioreactor (MBR) for combined biological and membrane treatment. Commonly, micro- (MF) or ultrafiltration (UF) membranes are applied to separate solids (biological sludge) from the treated water phase and in this manner deliver a high quality and solids free permeate. However, MBR permeate can still contain viruses and dissolved organic pollutants that have to be removed before the water can be reused. This can be accomplished by dense membrane processes such as nanofiltration (NF) or reverse osmosis (RO). NF produces a very high quality permeate that is ready for reuse for instance for irrigation or industrial applications such as cooling water [1]. Although RO membranes would give the highest permeate quality, their high salt retention compared to NF would result in a strong increase in salinity in the MBR. This could be harmful to the bacteria because increased osmotic pressure damages bacterial cell walls. Also from an energetic point of view, RO generally would be less favorable than NF as RO requires higher feed pressures. However, the major drawback of nanofiltration is the production of a concentrate stream, which can have a volume up to 10–20% of the wastewater volume.

NF concentrate can also be considered a resource for the recovery of valuable compounds like phosphates, although this would require additional technology [2]. Depending on the wastewater constituents, which to a certain extent are rejected by NF, the concentrate cannot be simply discharged to the environment because it contains heavy metals and organic micropollutants, which are harmful for aquatic organisms [3]. A possible strategy to avoid this, is to recirculate the NF concentrate to the preceding MBR system [4,5].

List of abbreviations: COD, Chemical oxygen demand; EPS, Extracellular polymeric substances; HRT, Hydraulic retention time; IC, Inorganic carbon; LMH, L m^{-2} h^{-1}; MBR, Membrane bioreactor; MF, Microfiltration; NF, Nanofiltration; PBS, Phosphate buffer saline; RO, Reverse osmosis; SEM, Scanning electron microscope; SRT, Sludge retention time; TC, Total carbon; TMP, Transmembrane pressure; TOC, Total organic carbon; UF, Ultrafiltration

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Combined MBR and NF treatment of wastewaters is already described in the literature [4]. However, the impacts of (full) recirculation of untreated NF concentrate on the biological MBR processes and on MBR and NF membrane performance have not yet been investigated. The prolonged (biological) contact time caused by the concentrate recirculation may stimulate the biodegradation of some of the organic micropollutants such as pharmaceuticals and personal care products [6]. However, as organics are known to foul NF membranes [7], the recirculation of non-biodegradable organic matter could also directly or indirectly contribute to membrane fouling.

Since rejection of micropollutants by NF is high, NF concentrate recirculation will yield higher multivalent cation concentrations in the MBR. Especially increased calcium, but also magnesium and iron concentrations, are known to reduce the fouling potential in MBRs [8,9] because they promote bioflocculation. However, when concentrations of multivalent ions become too high, this may induce scaling of the membrane.

The aim of this study was to investigate the impact of NF concentrate recirculation in a combined MBR and NF process (MBR NF) on the performance of the MBR and NF membranes in more detail, and to evaluate the feasibility of this integrated concept for production of reusable water. For this purpose two MBR systems were operated in parallel for 364 days. The permeate of one MBR was treated by nanofiltration (NF) and the NF concentrate was recirculated to this MBR. The other MBR without NF concentrate recirculation served as a blank system. Differences between the two systems in terms of transmembrane pressure (TMP), critical flux, membrane resistances, membrane rejections of specific constituents, contributions to fouling by scaling and organic fouling were all investigated, and the effect of the pH on these was determined as well. Also NF membrane autopsies in an early and a later stage of the operation were carried out. The main foulants were identified by dedicated dead-end filtration tests with MBR sludge and MBR supernatants.

2. Experimental setups

2.1. Lab scale MBR and NF setup

To determine the effect of NF concentrate recirculation on the MBR process, two 7-L lab scale MBR units were operated for 364 days. Both reactors were aerated with coarse bubble aeration plates at an average airflow rate of 0.6 $m^3\,m^{-2}\,h^{-1}$ for biological as well as membrane aeration. Municipal wastewater from a nearby sewer system was pre-settled and fed to both reactors at a flow rate of 36 L day^{-1} (Fig. 1). Wastewater characteristics are shown in Table 1.

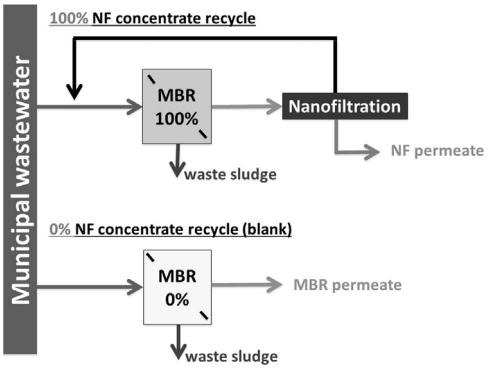

Fig. 1. Lab scale MBR systems without (R0%) and with full (R100%) NF concentrate recycling.

Table 1
Wastewater characteristics (mg L^{-1}) in terms of COD and its fractions as well as elements and anions measured in the dissolved phase.

	Unit			Unit	
COD total	mg O_2 L^{-1}	403 ± 107	**TC**	mg L^{-1}	137 ± 22
COD suspended	mg O_2 L^{-1}	209 ± 35	**TIC**	mg L^{-1}	93.3 ± 9.6
COD colloidal	mg O_2 L^{-1}	85 ± 24	**TOC**	mg L^{-1}	42.4 ± 17.8
COD dissolved	mg O_2 L^{-1}	109 ± 48	**Calcium**	mg L^{-1}	68.2 ± 7.5
NH_4–N	mg L^{-1}	38.1 ± 9.8	**Magnesium**	mg L^{-1}	18.0 ± 4.7
NO_3–N	mg L^{-1}	< 0.3	**Silicon**	mg L^{-1}	15.3 ± 1.4
Copper	mg L^{-1}	0.55 ± 0.22	**Sulfur**	mg L^{-1}	2.3 ± 8.0
Potassium	mg L^{-1}	21.6 ± 3.6	**Chloride**	mg L^{-1}	177 ± 62
Phosphorous	mg L^{-1}	6.0 ± 1.8	**Sodium**	mg L^{-1}	137 ± 28

Each MBR contained two submerged MF flat sheet membranes (Kubota, Japan) with a surface area of 0.116 m^2 per plate and 0.4 µm pore size. The membranes were operated at a flux of 6.4–7.5 L $m^{-2}\,h^{-1}$ (LMH). Membrane relaxation steps of 2 min were performed every 8 min.

One MBR system (R0%) served as a control system without NF concentrate recirculation and the permeate was discharged into the sewer system. The other MBR (R100%) was also fed with the pre-settled wastewater and additionally with NF concentrate. For this purpose 600 L R100% MBR permeate was collected in time during two weeks, stored at room temperature, and every 2 weeks batch wise concentrated down to 100 L. For this, a lab scale nanofiltration setup containing a 2.5″ spiral-wound NF270 module (DOW) was used. A maximum permeate recovery of around 85% was achieved. This concentrate production was carried out at 11 bar. After each cycle of concentrate production, a chemical cleaning of the 2.5″ module was performed at a pressure of 2 bar. The module was rinsed with citric acid at pH 4 for 10 min, followed by a rinse with tap water for 10 min.

The 100 L of concentrate was fed continuously at a flow rate of 6.48 L day^{-1} to the R100% MBR, additionally to the pre-settled wastewater. The NF concentrate represented 15% of the total inflow (wastewater+concentrate) and the wastewater the other 85%.

Temperature, pH, hydraulic retention time (HRT), solids retention time (SRT) as well as the dissolved oxygen concentration in both MBR systems were equal. The temperature and pH in the MBRs were not controlled but remained constant around 16.7 ± 2.1 °C and 7.5 ± 0.2, respectively. HRT and SRT were set to 4.7 h and 16 days, respectively. Dissolved oxygen concentration in the reactors was kept around 1–4 mg L^{-1} by manual control of the airflow rate. The airflow rate was varying in time (also depending on the strength of the wastewater) but was always kept equal in both reactors to assure equal shear. The TMP was recorded with pressure sensors (Endress+Hauser, Cerabar M). When the TMP of one of the MBR membranes exceeded 400 mbar, all membranes in the two reactors were cleaned. As this was determined by the R100% MBR in this research (which showed the most severe fouling), cleaning needed to be done twice a month. The membranes were taken out of the reactors, cleaned physically with a sponge, followed by chemical cleaning for 4 h: soaking in 0.2 g L^{-1} sodium-hypochlorite (NaOCl) for 2 h, followed by intermediate rinsing with tap water and by acid cleaning with citric acid (2 h immersion in 1.25 g L^{-1} in citric acid). The membranes were rinsed with tap water before they were replaced in the reactors.

2.2. Dead-end filtration cell setup

Several filtrations described in the following section were carried out in stainless steel stirred dead-end filtration cells, operated at different pressures, with different feed volumes and