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Effects of a sidestream concentrated oxygen supply system on the membrane filtration performance of a high-loaded membrane bioreactor

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ABSTRACT

To investigate the influence of high-pressure and shear effects introduced by a concentrated oxygen supply system on the membrane filtration performance, a laboratory-scale membrane bioreactor (MBR) fed artificial municipal wastewater was operated continuously for 80 days in four phases equipped with different aerations systems: (P1) bubble diffusers (days 0–40), (P2) concentrated oxygen supply system, the supersaturated dissolved oxygen (SDOX) (days 41–56), (P3) bubble diffusers (days 57–74), and (P4) SDOX (days 75–80). Various sludge physical-chemical parameters, visual inspection of the membrane, and permeability evaluations were performed. Results showed that the high-pressure effects contributed to fouling of the membranes compared to the bubble diffuser aeration system. Biofouling by microorganisms appeared to be the main contributor to the cake layer when bubble diffusers were used, while fouling by organic matter seemed to be the main contributor to the cake layer when SDOX was used. Small particle size distribution (PSD) (ranging from 1 to 10 and 1–50 μm in size) fractions are a main parameter affecting the intense fouling of membranes (e.g., formation of a dense and thin cake layer). However, PSD alone cannot explain the worsened membrane fouling tendency. Therefore, it can be assumed that a combination of several factors (which certainly includes PSD) led to the severe membrane fouling caused by the high-pressure and shear.

1. Introduction

The advantages of membrane bioreactors (MBR) combine the biological treatment with membrane filtration over conventional activated sludge systems (CAS) include: (i) producing a reliable and high-quality effluent; (ii) operating at much higher mixed liquor suspended solids concentrations (MLSS), reducing the system footprint; (iii) working at much higher solids retention times (SRTs); and (iv) handling unexpectedly high organic loadings and shock loads without degrading system performance (Kim et al., 2019).

MBRs are typically equipped with conventional diffused aeration systems, fine and/or coarse bubble diffusers, to provide the dissolved oxygen (DO) required for the biological process and membrane scouring. Poor oxygen transfer performance of diffused aeration systems has been widely reported (Duran et al., 2016; Germain et al., 2007; Henkel et al., 2011; Kim et al., 2019; Kim et al., 2020; Kim et al., 2021; Krampe and Krauth, 2003; Muller et al., 1995). The oxygen transfer performance is strongly influenced by the MLSS concentration in the reactor. Conventional MBRs operate at a higher MLSS concentration (approximately 10 g/L) compared to CAS systems (approximately 3 g/L). Therefore, the inefficiency caused by the conventional diffused aeration system is more pronounced in MBRs than in CAS systems. Operating MBRs at higher than usual MLSS concentrations (i.e., MLSS concentrations above 10 g/L) is highly desirable to increase the treatment capacity of such systems and/or to further reduce the system footprint. One such concept of an MBR operating at MLSS concentrations greater than 10 g/L was presented by Kim et al. (2019) as a high-loaded MBR (HL-MBR). Significantly, the negative effects of high MLSS concentration on oxygen

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transfer are more noticeable when diffused aeration systems are used in such systems. Therefore, operation of conventional MBRs beyond MLSS concentrations of 10 g/L is either technically infeasible or extremely energy inefficient (Kim et al., 2019).

Recently, innovative oxygen delivery technologies have been developed to improve oxygen transfer in biological wastewater treatment plants (WWTP). Concentrated oxygen delivery systems such as the supersaturated dissolved oxygen (SDOX) system are presented as promising technologies for improving the oxygen transfer performance. The SDOX system consists of a pressurized chamber operated at a pressure of approximately 8 bar. A high purity oxygen (HPO) source is introduced into the pressurized chamber. A stream of the mixed liquor from the biological reactor of the wastewater treatment system is recirculated through the pressurized chamber where it gets in contact with the HPO source at high-pressure conditions. Consequently, concentrated DO concentrations can be achieved, for instance, DO concentrations of up to 350 mg/L in clean water can be reached (Kim et al., 2020). The concentrated and/or supersaturated mixed liquor is released back to the biological reactor introducing in such way large amounts of concentrated DO. Kim et al. (2020) assessed the oxygen transfer performance of the SDOX system at MLSS concentrations ranging from approximately 4 to 45 g/L, and it was compared to the oxygen transfer performance of conventional diffused aeration systems. The SDOX showed much higher oxygen transfer rates (OTRs) (14 g O_2/L/d) compared to diffused aeration (fine-bubble diffusers) (2.4 g O_2/L/d). Moreover, the SDOX system reached oxygen transfer efficiencies (OTEs) of approximately 100% in clean water. Fine-bubble diffusers exhibited an OTE of maximum 5% per meter of submergence (Kim et al., 2019). Besides, the authors reported much higher alpha factors (mass transfer ratio of process-water to clean-water) for the SDOX system than for the fine-bubble diffusers. The SDOX system demonstrated a much better oxygen transfer performance compared to fine-bubble diffusers, particularly, when working at MLSS higher than 10 g/L (i.e., in the HL-MBR operational range). Thus, the SDOX system overcomes the oxygen transfer limitations imposed by conventional diffused aeration systems for MBRs working at high MLSS concentrations.

However, when using the SDOX technology, the mixed liquid is subjected to high-pressure conditions and shear forces. Hence, the biological activity of the sludge in the biological systems might be impaired. Kim et al. (2020) evaluated the influence of the SDOX system on the biological performance of an MBR system. The authors concluded that the biological performance of the system was not affected by the introduction of the SDOX unit (i.e., high-pressure conditions and shear forces).

Notwithstanding the advantages of using SDOX in terms of oxygen transfer and biological performance, the shear effects and high-pressure conditions exerted by the SDOX technology may modify the properties of the sludge. Such changes on the sludge properties may eventually influence the performance of downstream solid-liquid separation processes commonly observed in wastewater treatment processes such as gravity settlers and/or membrane filtration. For instance, Zhang et al. (2015) reported that shear forces generated by conventional diffused aeration systems already contributed considerably to the modification of particle size distribution (PSD) of sludge negatively affecting the sludge settleability. In addition, changes in the PSD of the sludge significantly affect the membrane filtration performance in membrane filtration processes (Fortunato et al., 2018; Shen et al., 2015); the sludge flocs are attracted and accumulated onto the membrane surface contributing to the formation of a cake layer that contributes to reduce the membrane filtration performance. The smaller the sludge particle size, the more compact the cake layer, so the larger the negative effects on the membrane filtration performance (Hennemann et al., 2021; Meng et al., 2006; Meng et al., 2007a; Meng et al., 2007b; De Temmerman et al., 2015). The shear forces and high-pressure conditions introduced by the SDOX technology can contribute to shifting the PSD of the sludge to larger amounts of smaller particles, eventually worsening down-stream membrane filtration separation processes. Moreover, the breakage of sludge-flocs due to the shear forces and high-pressure condition may release compounds such as extracellular polymeric substances (EPS) and soluble microbial products (SMP). These compounds have been reported to exert a negative effect on the membrane filtration process (Chang et al., 2002; Le-Clech et al., 2006).

The SDOX technology is presented as a promising alternative for supplying DO in biological wastewater treatment process; particularly, when working at high MLSS concentrations. Particularly, MBR systems seem to be the proper niche for the SDOX technology when working at higher than usual MLSS concentrations as in the HL-MBR concept (i.e., MLSS concentrations higher than 10 g/L). However, the potential impact of the SDOX technology on the down-stream solid-liquid separation processes as well as membrane filtration performance has not been yet investigated. The SDOX technology is an innovative process for supplying DO and the technology has been mostly evaluated in the context of clean water including bioremediation processes, aquaculture applications, river restorations, and odor control processes, among others. Only a few studies have been reported utilizing the SDOX system in the context of biological wastewater treatment (Kim et al., 2019, 2020, 2021).

To the best of the authors' knowledge, the impact of the SDOX technology on the membrane filtration process of MBR systems working at high MLSS concentrations has not been assessed. This study will therefore address such needs directly. The specific objectives of this research are as follows: (i) evaluating the potential impacts of the SDOX technology on the membrane filtration performance of an MBR system operated at high MLSS concentrations, (ii) sludge properties including total suspended solids (TSS), volatile suspended solids (VSS), PSD, SMP, and EPS were determined and related to the membrane filtration performance, and (iii) scanning electron microscope (SEM) images of the membranes were evaluated and energy dispersive X-ray (EDX) analysis were also carried out for characterizing the surfaces of membranes to better determine the causes affecting the membrane filtration process due to the presence of the SDOX technology.

2. Materials and methods

2.1. Experimental setup

A bench-scale MBR was equipped with either conventional fine bubble diffusers (Fig. 1a), or the SDOX unit (Fig. 1b) for introducing DO. The MBR was made of transparent acrylic glass with a total volume of 30.6 L (16 × 25.5 × 75 cm), and it was operated at a working volume of 6.5 L. A flat-sheet membrane (XJ3 module by Kubota) made of chlorinated polyethylene was submerged in the middle and lower part of the MBR basin. The membrane had an effective filtration area of 0.11 m^2 with a nominal pore size of 0.4 μm. A coarse bubble diffuser (Uxcell, model number: US-SA-AJD-231698, Hong Kong) was placed at the bottom of the MBR basin to continuously scour the membrane. Air was supplied by a blower (HIBLOW HP 80, Techno Takatsuki, Japan) at an air flow rate of 2 $m^3/m^2/h$ to satisfy the specific membrane scouring aeration needs (i.e., the membrane was continuously scoured with a tangential air flowrate of 0.22 m^3/h). A piston fluid metering pump (FMI PM6014 RHV, Fluid Metering, Inc, USA) was provided to extract the permeate out from the MBR.

During the operational phases P1 and P3, DO was supplied by conventional fine bubble diffusers (Hydrofarm, Inc, USA). The diffusers were placed at the bottom of the MBR, and they were operated at an air flowrate of approximately 0.5 m^3/h; the fine bubble diffusers served as the primary source of oxygen (Fig. 1a). In addition, two baffles were placed at both sides of the immersed membrane to secure a uniform distribution of the airflow. The membrane surface was continually air-scoured. The transmembrane pressure (TMP) of membrane was continuously monitored by a digital gauge (Ashcroft 2274, USA) fitted to

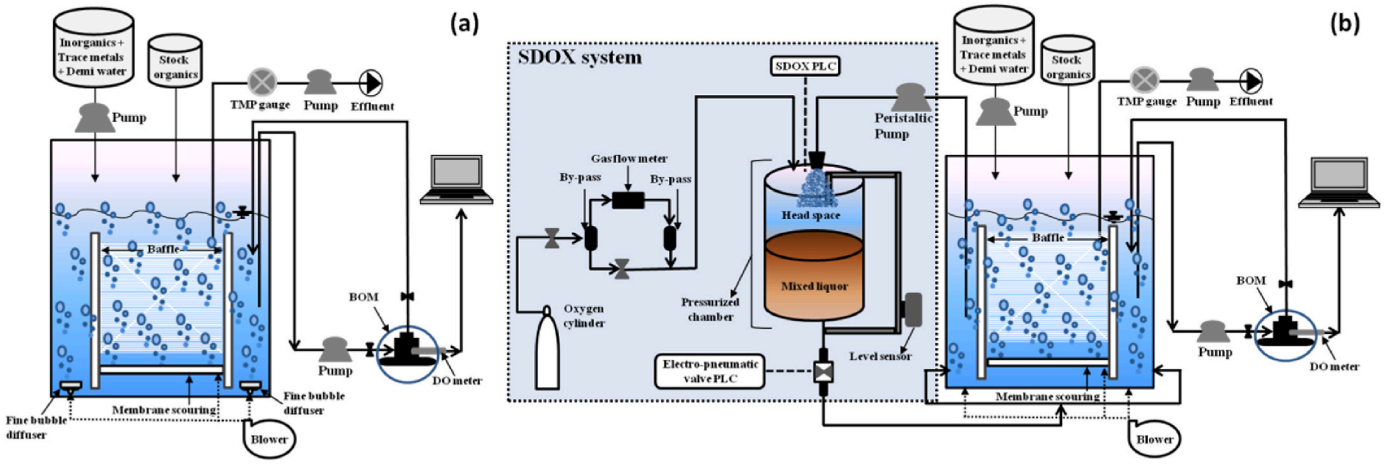

Fig. 1. Experimental setup of the membrane bioreactor system provided with (a) bubble diffusers and (b) supersaturated dissolved oxygen system.

the permeate line and connected to a data acquisition system. During the operational phases P2 and P4, DO was supplied by a bench-scale SDOX unit (Fig. 1b). The bench-scale SDOX unit consisted of a pressurized chamber made of stainless steel connected to an HPO source (pure oxygen cylinder (MESSER, Croatia)). The pressurized chamber had a total volume of 2.75 L. Approximately 20% of the total volume (0.55 L) was occupied by the mixed liquor sludge solution to be oxygenated, while the 80% remaining (2.20 L) consisted of the headspace. The pressure in the SDOX pressurized chamber was set at 6.9 bar. The pressurized chamber was provided with two analogic pressure gauges (McDaniel Controls, USA), a digital pressure sensor (SICK AG, Germany), and a level digital sensor (Setra Systems, USA). An electro-pneumatic valve (NVF3-MOH-5/2-K-1/4-EX, FESTO, Germany) was introduced at the effluent drainage of the pressurized chamber. The pressure sensors, level sensors, and the electro-pneumatic valve were used to monitor and control the level and pressure of the pressurized chamber by the aid of a program logic controller system (SIMATIC S7-1200, Siemens, Germany). The HPO was supplied into the SDOX system through a gas flowmeter provided with mass totalizer capacities (Model # 32,908–59, Cole-Palmer, USA). The sludge stream was introduced into the SDOX system through a 6 mm orifice by a high-pressure peristaltic pump (EW-74203-24, Cole-Palmer, USA) at a flowrate of 0.3 L/min. The supersaturated sludge stream was released back into the MBR introducing in such way DO into the MBR. Such stream was released at the bottom of the MBR basin providing also mixing to the MBR system. In this study, the DO set point was 2.0 mg/L.

2.2. Experimental procedure

The MBR was inoculated with fresh activated sludge from the aerobic basin of the municipal WWTP located in the city of Zagreb in Croatia. The WWTP was operated as a CAS process and designed only for carbon removal. The plant was operated at an SRT of approximately 5 day at an average MLSS concentration of approximately 4.0 g/L. The sludge was filtered through a 500 μm sieve and thickened up to an MLSS concentration relevant of approximately 15 g/L first by gravity settling at the WWTP facility followed by membrane filtration thickening.

After inoculating the MBR system, it was then operated with the fine bubble diffusers for 40 days (P1). On P1, the biomass was acclimating to the synthetic influent wastewater and operational conditions in the MBR. As such, the baseline conditions regarding membrane fouling in the MBR could be established. On day 41, the aeration system was switched to the SDOX unit and the MBR was operated under identical operational conditions as in P1 for 16 days (P2). P2 aimed at investigating the potential effects of the SDOX system (high-pressure conditions, shear effects, among others) on membrane fouling. Then, the SDOX unit was replaced again by the fine bubble diffusers, and the system was operated for 18 additional days (P3). P3 was included to monitor the changes in membrane fouling when the system was exposed again to bubble diffusers. Finally, the aeration system was again replaced by the SDOX unit, and the evaluation was continued for six additional days (P4) to confirm the effects of the high-pressure conditions on membrane fouling. The fouled membranes during the entire evaluated period were replaced by an unused membrane once the TMP exceeded the value of 0.45 bar. In this study, membrane relaxation and/or membrane backwashing were not applied. That is, the membrane was continuously operated to maximize the effects of membrane fouling (i.e., to test the limits of the membrane in terms of membrane fouling and membrane filtration performance).

The MBR was fed with synthetic wastewater. The organic constituents included glucose, acetate, peptone, and yeast, were added to the MBR by gravity drips using a gravity medical infusion at a flowrate of 1 L/d. A second solution containing the inorganic components of the synthetic wastewater was added through a piston fluid metering pump (FMI PM6014 RHV, Fluid Metering, Inc, USA) at a flowrate of 39.6 L/d. That is, the total influent flowrate to the MBR was set at 40.6 L/d delivering the wastewater composition of the synthetic wastewater to the MBR system as describe in Table 1. Such a flowrate established a total hydraulic retention time of approximately 4 h and a membrane flux (i.e., the amount of permeate produced per membrane surface area per time) of 15 L/m^2/h (generally abbreviated as LMH). The SRT was set to 10 days by withdrawing 0.65 L/d of sludge from the MBR.

2.3. Samples collection and analytical methods

2.3.1. Membrane fouling evaluation (transmembrane pressure and membrane permeability determination)

The membrane permeability was measured twice in demineralized

Table 1
Characterization of the synthetic wastewater reaching the membrane bioreactor system.

Chemical compounds	Concentration (mg/L)	Chemical compounds	Concentration (mg/L)
$C_6H_{12}O_6$	421.88	$FeCl_3 \cdot 6H_2O$	19.36
$C_2H_3NaO_2$	571.28	$C_{10}H_{14}N_2Na_2O_8 \cdot 2H_2O$	30.00
Peptone	260.00	$MnCl_2 \cdot 4H_2O$	0.74
Yeast	40.00	$ZnSO_4 \cdot 7H_2O$	2.50
NH_4Cl	65.69	$CuSO_4 \cdot 5H_2O$	0.61
KH_2PO_4	48.33	$CoCl_2 \cdot 6H_2O$	2.09
$NaHCO_3$	251.95	$Na_2MoO_4 \cdot 2H_2O$	0.26
$CaCl_2$	40.37	H_3BO_3	0.13
$MgSO_4$	65.65	$NiSO_4 \cdot 7H_2O$	0.29

water: (i) before exposing/submerging each new membrane into the MBR; and (ii) after the membrane was completely fouled (i.e., when TMP of membrane reached a value of 0.45 bar). The membranes were submerged in a separate tank with a working volume of 31.7 L (14 × 22 × 103 cm) filled with demineralized water. The permeability was determined by measuring the stabilized TMP at different permeate fluxes. The permeate was extracted by a piston fluid metering pump (FMI PM6014 RHV, Fluid Metering, Inc, USA). The TMP was monitored by a digital gauge (Ashcroft 2274, USA) fitted to the permeate line. The flowrate was controlled by a flowmeter controller (FMI V200, Fluid Metering, Inc, USA) connected to the piston fluid metering pump. The flux was calculated by dividing the measured flowrate by the membrane filtration area, and the permeability was obtained by dividing the calculated flux by the TMP. The permeability was determined using no less than four data points – fluxes vs stabilized TMPs. The flux was gradually increased and the corresponding stabilized TMP values were recorded. Flowrates ranging from approximately 0.7 to 2.2 L/h were selected providing fluxes ranging from approximately 6.6 to 22.0 LMH. The permeability was calculated determining the slope when plotting the flux as a function of the stabilized TMP. The slow, fast, and total membrane fouling rates were calculated based on the instantaneous changes of the TMP as a function of time (Yoon, 2016). The slow membrane-fouling rate was determined at low TMP values (i.e., at TMP values lower than 0.06 bar), while the fast fouling rate was determined at large TMP values (i.e., at TMP values larger than 0.06 bar and until reaching the maximum allowed TMP value of 0.45 bar).

2.3.2. Total suspended solids and volatile suspended solids determination

The TSS and VSS concentrations were determined following the standard methods for the examination of water and wastewater (APHA, 2017). The determination of TSS and VSS was carried out between 2 and 3 times a week.

2.3.3. Particle size distribution and diluted sludge volume index

The PSD of the sludge was determined using a Malvern Mastersizer 2000 that can detect the particle size ranging from 0.02 to 2000 μm (Malvern Instruments Ltd, Malvern, UK) with laser diffraction functionalities. The PSD results are reported as the values of the particle size at 10% (D10), 50% (D50), and 90% (D90) cumulative distribution. The D50 is also reported as the median particle size (MPS). That is, D50 indicates that 50% of the total sludge particles are less than or equal to the D50 value. Similarly, D10 indicates that 10% of the total sludge particles are less than or equal to the D10 value. The PSD determination was carried out between 2 and 3 times a week. The PSD determinations were carried out in triplicate, and the average value of PSD was reported.

Settleability tests were performed to determine the sedimentation of the sludge when exposed either to the conventional diffusers, or to the SDOX system. The diluted sludge volume index (DSVI) were determined following the method reported by Ekama et al. (1997). The settled sludge volume after a 30 min sedimentation period should be between 150 and 250 mL/L. Permeate was used to carry out the dilutions. The DSVI is usually utilized to determine the settleability of concentrated sludge (e.g., above 15 g/L of TSS). The DSVI determinations were conducted between 2 and 3 times a week.

2.3.4. Extraction and analytical determination of soluble microbial products and extracellular polymeric substances

Analytical determinations of SMP and EPS were carried out following the method described by Le-Clech et al. (2006). Mixed liquor samples containing a volume of 60 mL were centrifuged for 5 min at 5, 000 g using a Rotina 35 centrifuge (Hettich, Germany). The supernatant was then filtered through a 1.2 μm Minisart® syringe filter (Sartorius, Germany). The filtrate represented the SMP solution. The SMP was determined as reported by Jarusutthirak and Amy (2006) both by measuring the total organic carbon (TOC) content using a TOC analyzer (TOC-5000 A, Shimadzu, Japan) as non-purgeable organic carbon, and by measuring the ultraviolet absorbance at 254 nm (UV_{254}) using a spectrophotometer UNICAM Helios Beta (Thermo Fisher Scientific, USA). The samples were filtered through a 0.45 μm polypropylene filter (Whatman, USA) to determine both the TOC and UV_{254}. Therefore, the measured TOC values represented the dissolved organic carbon (DOC) values. The specific ultraviolet absorbance ($SUVA_{254}$) was calculated dividing the UV_{254} by the DOC. Since SMPs are characterized by major components of dissolved organic matter in wastewater, the $SUVA_{254}$ was reported as the SMPs (Jarusutthirak and Amy, 2006).

After removing the supernatant from the sample for the SMP determination, the remaining pellet retained at the bottom of the centrifuge tube was re-suspended with demineralized water. The mixture was then heated for 10 min at a temperature of 80 °C in a water bath (Memmert, Germany), which was then centrifuged for 10 min at 7,000 g. The supernatant was then filtered through a 1.2 μm Minisart® syringe filter (Sartorius, Germany). Such filtrate contained the EPS compounds. The EPS can be further classified into carbohydrate EPS (EPS_c) and protein EPS (EPS_p). The EPSc was determined following a photometric method performing a H_2SO_4/phenol oxidation followed by a colorimetric determination method using a DR3900 spectrophotometer (Hach, USA) (Lowry et al., 1951); the EPSp was determined following the Folin–Ciocalteu method (Dubois et al., 1956). Both SMP and EPS were determined between 2 and 3 times a week.

2.3.5. Determination of the characteristics and elemental composition of the compounds deposited on the membrane surface

After determining the permeability of the fouled membrane in demineralized water, the membranes were dried at atmospheric conditions. Then, membrane fragments of approximately 1 cm^2 were prepared. Such membrane fragments were later observed under the microscope (JEOL JSM-7500 F Field Emission SEM, Japan) to determine the characteristics of the compounds that were deposited on the surface of the membrane. In addition, the elemental composition of such compounds was determined. Each sample was coated with gold (Au) by using a JFC-1300 high-vacuum auto fine sputter coater (JEOL, Japan) connected to a DUOLINE vacuum pump (Pfeiffer vacuum GmbH, Germany). The coated samples were placed on a sample holder, and the membrane surfaces were scanned using a JSM-7500 F Field Emission SEM (JEOL, Japan). Each sample was photographed using secondary electrons (SE) at 5,000× magnification at 5 kV. For characterizing the chemical elemental composition of the compounds deposited on the membrane surface, an EDX spectroscopy (Noran system 6, Thermo Fisher Scientific, USA) was employed with the SEM images taken at the low magnifications of 5,000×. The determination of the characteristics and elemental composition of the compounds deposited on the membrane surface was performed on the following membranes: an unused membrane, two membranes used during P1, and three membranes used during P2.

3. Results and discussion

3.1. Effect of the supersaturated dissolved oxygen system on the transmembrane pressure and membrane permeability

The MBR operation was started immediately after inoculation (day 1) with a new (unused) membrane. During the entire evaluated period, the membranes were replaced every time the TMP exceeded the value of 0.45 bar. That is, 13 new membranes were used to carry out the entire evaluation. Fig. 2 presents the evolution of the TMP as a function of the operational time. In addition, Table 2 describes the TMP increment rates for each membrane (slow, fast, and total), the permeability for each unused and fouled membrane, and the loss of permeability. Regarding the TMP increments, two fouling rates were determined: the slow membrane fouling rate (exhibiting TMP values below 0.06 bar), and the fast membrane fouling rate (exhibiting TMP values above 0.06 bar).

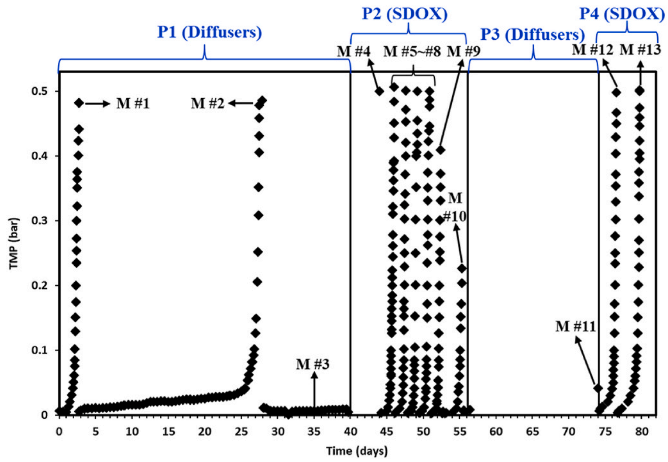

Fig. 2. Transmembrane pressure profile as a function of the exposure time. M#1 to M#13 denote the 13 different new membranes used in this research. The four phases of this research are indicated in the figure.

Each membrane was identified with a unique number and code as indicated in Table 2. The first and second column in Table 2 display the membrane number (in a chronological order as used in this research) and the code assigned to each membrane, respectively.

The MBR was continuously operated at the same effluent (permeate) flowrate of 40.6 L/d setting a membrane flux of approximately 15 $L/m^2/h$. Therefore, the TMP values reported in Fig. 2 and Table 2 were obtained at the very same flux for all the evaluated membranes. Usually, MBR systems provided with flat sheet (Kubota) membranes (as used in this research) are operated in cycles introducing relaxation phases in between the filtration phases to enhance the membrane filtration performance. In this evaluation, to push the fouling limits of the membrane filtration process, the relaxation phase was not included in between the filtration phases.

At the startup of the MBR system during the phase P1, as shown in Fig. 2, the TMP of the first membrane (M#1) sharply increased after only two days of operation. In addition, Table 2 exhibits slow, fast, and total fouling rates experienced for M#1. During phase P1, the system was equipped with the fine bubble diffusers. The sludge taken from the municipal WWTP for inoculating the MBR was concentrated from 4 g/L (sludge concentration at the WWTP in Zagreb) to 15 g/L (sludge concentration at the startup of the MBR). In addition, at the startup of the MBR, the MBR was fed synthetic influent wastewater whose characteristics differed from the real municipal wastewater at which the sludge was already exposed and acclimated. Thus, the sludge needed to be acclimatized again to a new set of operational conditions. Therefore, during the first few days of operation the biomass could have been stressed eventually causing a detrimental effect on the membrane filtration processes (Shah et al., 2021; Wu et al., 2011). In addition, while concentrating the sludge, some sludge flocs-breakage (sludge disintegration) could occur contributing to the presence of large amounts of small-size loose particles that could also affect the membrane filtration processes (De Temmerman et al., 2015; Wisniewski and Grasmick, 1998). Both the stress exerted on the biomass, as well as the potential presence of biomass fragments could eventually explain such a pronounced increase on the observed TMP values during the initial operational days. After reaching the TMP value of 0.45 bar, M#1 was replaced by M#2. The MBR system was operated with M#2 for approximately 25 days before experiencing a severe increase on the TMP; that is, a much better membrane filtration performance was observed compared to M#1, as also indicated in Table 2 by a lower fouling rate for M#2 (slow, fast, and total) compared to M#1. Nevertheless, a pronounced and fast fouling was also observed on M#2 after 25 days of operation; at this point, M#2 was replaced by a new membrane (M#3). The MBR system was then operated for additional 12 days during which membrane fouling was not observed.

On day 41, the aeration system was changed from the bubble diffusers to the SDOX system indicating the beginning of the phase P2; a new membrane (M#4) was placed to better observe the effects of the SDOX system on the membrane filtration performance. After approximately 4 days the maximum allowed TMP value was reached. Eventually, the sludge was adapting to the new conditions introduced by the SDOX system. As the MBR continued to run with the SDOX system, a rise in the TMP values for all the subsequent evaluated membranes was also noticed. Six additional unused membranes were introduced (M#5 to #10), and all of them lasted for approximately 1.5 days. As observed in Fig. 2 and Table 2, the TMP increase over time during P2 was much more pronounced than during P1. On day 53, the membrane scouring air flowrate was increased from 2.0 to 2.5 $m^3/m^2/h$ to evaluate the possibilities of achieving a more prolonged membrane operational period. Indeed, such operational change seemed to play a certain positive effect on delaying the replacement of the membranes; e.g., membrane #10 lasted for almost three days compared to the 1.5 days of the previous membranes (M#5 to #9). However, such improvements on the membrane filtration were still below the performance obtained in P1 where the membrane was operating for 25 days before reaching the maximum TMP value. The fouling rates observed in P2 were higher than in P1 for all the evaluated membranes confirming a worse membrane filtration performance in P2 than in P1. Thus, it can be inferred that the SDOX system introduced changes on the sludge characteristics in the MBR with a negative short-term impact on the membrane filtration performance.

Right before M#10 reached the maximum allowed TMP value, the phase P3 was initiated; that is, the aeration system was switched back from the SDOX system to the bubble diffusers. The phase P3 was evaluated starting with a new membrane in the MBR (M#11). A marginal

Table 2
Transmembrane pressure increment rates and membrane permeability for the evaluated membranes.

Membrane number	Membrane code	Operational days	Days of exposure	dTMP/dt (bar/day)			Permeability (K) (L/m^2-h-bar)		Loss of permeability (%)
				Slow	Fast	Total	Clean	Fouled	
1	DIFF1	0 to 2.75	2.75	0.0262	0.6230	0.1728	7529.2	96.1	98.7
2	DIFF2	2.95 to 28.06	25.11	0.0024	0.2375	0.0192	6946.7	58.7	99.2
3	DIFF3	28.26 to 39.74	11.48	–	–	–	9505.5	1840.2	80.6
4	SDOX1	39.94 to 44.23	4.29	–	–	0.1155	7816.3	51.3	99.3
5	SDOX2	44.43 to 46.24	1.81	0.0422	0.9247	0.2773	7816.3	77.6	99.0
6	SDOX3	46.44 to 47.84	1.40	0.0511	1.3232	0.3554	8016.4	65.5	99.2
7	SDOX4	48.04 to 49.45	1.41	0.0555	0.9616	0.3511	7103.5	70.0	99.0
8	SDOX5	49.65 to 51.06	1.41	0.0543	0.9617	0.3511	8462.0	64.9	99.2
9	SDOX6	51.26 to 52.58	1.32	0.0432	0.9228	0.3079	8739.9	113.0	98.7
10	SDOX7	52.78 to 55.52	2.74	0.0087	0.3416	0.0820	9696.1	423.9	95.6
11	DIFF4	55.72 to 74.16	18.44	–	–	0.0020	5523.4	527.0	90.5
12	SDOX8	74.36 to 76.7	2.34	0.0229	0.9913	0.2097	5891.8	130.7	97.8
13	SDOX9	76.90 to 79.94	3.04	0.0228	0.7646	0.1633	5829.6	92.1	98.4

increase on the TMP was observed during phase P3, and the system was operated all the time with the same membrane (M#11) until reaching operation day 74. The negative membrane filtration effects introduced by the SDOX system observed in P2 were not noticeable in P3. This was also observed on the total fouling rate as shown in Table 2 exhibiting almost the lowest fouling-rate values comparable to those observed during P1. The bubble diffusers operated almost at atmospheric pressure and with the absence of the high-shear forces introduced by the SDOX. That is, the sludge characteristics were much less affected and thereby yielding a better membrane performance compared to when the system was equipped with the SDOX system.

Right after the initiation of the phase P4 on day 75, a new membrane was introduced into the MBR (membrane M#12) to evaluate the effects of re-introducing the SDOX system on the membrane filtration performance. Increments on the TMP values were observed in P4, although the TMP values were lower than the values observed in P2 when the MBR was also equipped with the SDOX unit. The M#12 only lasted for little longer than two days, when a new membrane was introduced (membrane M#13). Both P2 and P4 exhibited almost the same fouling pattern. Therefore, it can be concluded that the SDOX aeration system had a detrimental effect on the membrane filtration performance observed as a fast increase on the TMP values.

In addition, Table 2 describes the permeability of each clean and fouled membrane and the loss of permeability expressed as a percentage. The loss of permeability was almost the same for all the evaluated membranes, since the permeability was measured when each membrane reached its limit of functionality. Certainly, the SDOX system exhibited a negative effect on the membrane filtration performance. When the MBR was equipped with the SDOX unit, the sludge was recirculated through the SDOX system by a high-pressure peristaltic pump through a small-diameter piping (6 mm). The sludge was introduced to the SDOX pressurized-chamber through a small orifice in the SDOX system (6 mm), and exposed to high-pressure conditions (6.9 bar) inside the pressurized-chamber of the SDOX unit. Therefore, the sludge was exposed to high-shear forces and friction that could have an influence on the structure and nature of the sludge flocs. Consequently, changes on the size distribution of the sludge were expected, as reported later in this study, eventually negatively affecting the membrane filtration performance. Nevertheless, the biomass activity was not affected with respect to organic matter removal and nitrification (not reported in this study but thoroughly discussed in Kim et al. (2021); however, such high stress conditions at which the microorganisms were subjected, could have contributed to changes on the sludge characteristics with a negative impact on the membrane filtration performance.

3.2. Effects of the supersaturated dissolved oxygen system on the sludge physical-chemical properties

3.2.1. Total suspended solids and volatile suspended solids concentration

Fig. 3 describes the changes on the TSS and VSS concentrations as well as the VSS/TSS ratio as a function of the operational time. During P1, the TSS and VSS concentrations increased right after inoculating the reactor until reaching stable TSS and VSS concentrations by the end of the phase P1 at approximately 16.9 and 14.9 g/L, respectively. The VSS/TSS ratio also increased until reaching a 0.89 value towards the end of P1. At the beginning of P2 an operational problem occurred with the level sensor of the MBR allowing an excess of influent wastewater to reach the MBR and therefore to slightly dilute the mixed liquor. The TSS and VSS concentrations dropped to approximately 13.5 and 12.6 g/L, respectively. Such a decrease on the TSS and VSS concentrations was not due to the action of the SDOX system (i.e., shear forces and high-pressure conditions), but rather to the dilution effect. The TSS and VSS concentration increased towards the end of P2 reaching TSS and VSS values of 14.1 and 13.0, respectively. During P3, the TSS and VSS concentrations kept increasing reaching concentrations on average of 18.5 and 17.2 g/L, respectively. Finally, during P4 similar TSS and VSS concentrations as in P3 were reported confirming that the SDOX system did not contribute to reduce the TSS and/or the VSS concentration or to change the VSS/TSS ratio. During the entire evaluated period, the TSS and VSS concentration ranged from 13.5 to 18.8 and from 11.7 to 17.4 g/L, respectively. Even though some variation on the TSS and VSS were reported, those changes in the TSS and VSS concentration could not explain the different membrane filtration performance observed on the different phases as discussed in previous section 3.1. Likewise, the VSS/TSS ratio adopted values from 0.76 to 0.93 for the entire evaluated period. Accordingly, the VSS/TSS ratio did not reflect major changes on the organic fraction content of the sludge.

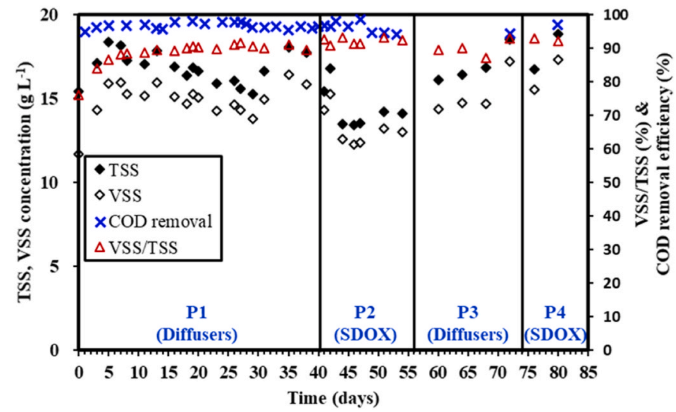

Fig. 3. Concentration of total suspended solids and volatile suspended solids, ratio Volatile suspended solids/Total suspended solids and efficiency of chemical oxygen demand removal as a function of operating time.

Although not reported in this paper, the biological performance of the MBR was also evaluated and reported in Kim et al. (2021). The biological performance regarding COD removal and nitrification did not vary during the evaluated period, confirming that the activity of the sludge was not affected depending on the utilized aeration system. That is, the deterioration on the membrane performance observed primarily in P2 cannot be explained due to changes on the TSS and/or VSS concentrations in the mixed liquor, or due to the changes on the biological activity of the sludge.

3.2.2. Particle size distribution, median particle size, and diluted sludge volume index

The PSD of the sludge has been considered one of the most critical sludge properties affecting the membrane filtration performance (Diaz et al., 2016; Drews, 2010; Shen et al., 2015). Fig. 4a shows the MPS as a function of the operational time. Right after inoculating the MBR, the sludge exhibited an MPS of approximately 114 μm. During P1 the MPS continuously decreased until reaching a value of approximately 39.8 μm towards the end of P1. Such a reduction on the MPS during P1 was only due to the action of the shear forces introduced by the bubble diffusers and the filtration process. The sludge was taken from a municipal WWTP, so during the acclimation period to the MBR such a reduction on the MPS was expected due to the action of the shear forces (Stricot et al., 2010). Noticeably, as described in Section 3.1 such shift on the PSD of sludge towards smaller particle sizes did not have a detrimental effect on the membrane filtration performance during P1. On the other hand, the membrane filtration exhibited a better performance at the end of P1compared to at the beginning of P1. On the day 41, the SDOX system was introduced initiating the phase P2. The PSD kept shifting towards the low size particle range. As shown in Fig. 4a, the MPS further decreased from 39.8 μm at the beginning of P2 to 28.9 μm by the end of the P2. The reduction on the MPS was not as evident during P2 as compared to during P1. However, P2 started at a much lower MPS than P1; the sludge in P2 was already acclimatized to the operational conditions in the MBR. In this regard, the changes on the MPS on P2 were

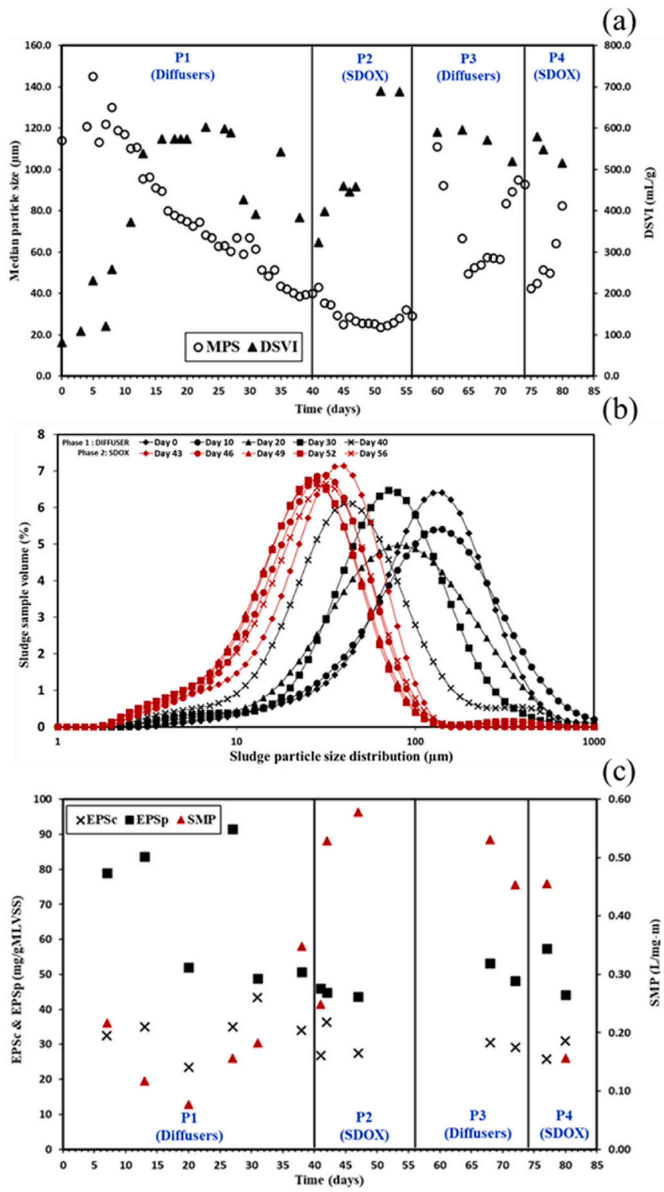

Fig. 4. (A) Median particle size and diluted sludge volume index of the sludge as a function of the operational time, (b) Particle size distribution of the activated sludge during the experiment, (c) Extracellular polymeric substances and soluble microbial products presence in the activated sludge during the evaluated period.

mostly due to the action of the SDOX unit. Even though the starting MPS was smaller (40 μm) than the MPS at the start of P1 (114 μm), the MPS kept decreasing during P2 until reaching an MPS of 28.9 μm. Significantly, during P2 the membrane filtration performance was negatively affected as delineated in Section 3.1. Thus, the changes on the PSD on P2 could eventually contribute to such reduced membrane filtration performance.

When the SDOX system was replaced again by the diffuser (P3), the MPS of the sludge increased in the MBR; some re-flocculation of the sludge could eventually take place. During P3 a better membrane filtration performance was observed than during P2. Therefore, there is a potential indication that the PSD of the sludge may exhibit a key role on the performance of the membrane filtration. The SDOX unit could introduce an adverse effect on lowering the size of the sludge particles, and ultimately exhibiting a negative effect on the membrane filtration performance. However, when the high-shear effects introduced by the SDOX system were no longer present, the MPS of the sludge increased improving the membrane filtration performance. When changing the aeration system back to the SDOX system in P4, the filtration performance of the new membranes was again strongly affected. The MPS sharply decreased immediately after introducing again the SDOX unit, and the membrane filtration performance was negatively affected. However, towards the end of P4 the MPS gradually increased. This was also noticed at the end of P2. The fact that the MPS was increasing towards the end of the evaluation in P2 and P4 could imply that the long-term exposure of the sludge to the SDOX unit (not evaluated in this study) may not be that much negative in terms of the reduction of the MPS and membrane filtration performance. However, more research would be needed to confirm that hypothesis.

Fig. 4b shows the PSD of the sludge evaluated during phases P1 (black dots) and P2 (red dots). The PSD continuously shifted towards the low particle size range (left side of Fig. 4b). That is, the fraction of small-sized particles in the sludge gradually increased. Particularly, during P1 an increase on the proportion of particles with a particle size range from approximately 10 to 100 μm was observed. However, during P2 the shift of the PSD occurred at lower particle sizes compared to P1, increasing the ratio of particles in the sludge with particle sizes lower than 10 μm. Such an increase on the fraction of particles with sizes lower than 10 μm was quite notorious when looking at the left side of Fig. 4b during P2. The presence of a left shoulder in Fig. 4b can be observed indicating an increase on the high amount of particles with particle sizes lower than 10 μm.

Based on Fig. 4b, a much larger fraction of small particles was present during P2 (SDOX unit) as compared to during P1 (bubble diffusers). Guo et al. (2012) reported that particulate and colloidal solids contributed significantly to pore blocking (i.e., internal fouling) leading to a reduced membrane filtration performance. Small particles can easily adhere to the surface of the membrane due to their inherent low diffusive back-transport forces (Belfort et al., 1994). That is, lower reverse transport velocities from the cake layer to the bulk mixed liquor solution occurred for small particles than for large particles. This phenomenon could be explained considering that the scouring effect of the air when passing through the membrane surface is not as effective for removing small particles as for removing large particles. As a result, the cake layer on the surface of the membrane would consist mostly of small particles forming a thin and compact cake layer that cannot be effectively removed by air scouring Park et al. (2015), and thereby contributing to deteriorate the membrane filtration performance.

The MPS, also known as the D50, represents the value of the particle size at 50% in the cumulative distribution. Lin et al. (2010) could not establish a clear relation between the MPS and the membrane filtration performance. However, the authors observed a link between the D10, the value of the particle size at 10% in the cumulative distribution, and the membrane filtration performance. Particularly, the authors reported that the membrane filtration performance was affected when increasing the fraction of particles with a particle size smaller than 10 μm. In addition, Bai and Leow (2002) and Qi et al. (2016) had shown that membrane filtration was strongly affected by sludge flocs sizes less than 50 μm.

Similar findings were observed in this research. Table 3 indicates the fraction of particles organized in two categories: (i) 1–50 and 50–100 μm, and (ii) 1–10 and 10–100 μm. As observed in Table 3, when focusing on the 1–50 μm range fraction, the operation during P1 increased such range fraction from approximately 13.8%, when the MBR system was just inoculated, up to 56.1% at the end of P1. However, as soon as the SDOX system was introduced, the 1–50 μm fraction range sharply increased reaching a maximum value of 85.4% during P2. That is, the SDOX system indeed exerted a strong influence increasing the fraction of particles within the 1–50 μm range, and that was detrimental for the membrane filtration performance. Similar effects were observed when focusing on the 1–10 μm range. The incorporation of the SDOX unit increased the fraction of small particles from 6.1% at the end of P1, up to

Table 3
Fractions of sludge particle sizes organized in two different ranges for all the evaluated phases.

Sampling points (days)		Proportion (%)			
		Particle size (1–50 μm)	Particle size (50–1000 μm)	Particle size (1–10 μm)	Particle size (10–1000 μm)
Inoculated sludge	0	13.8	85.8	1.56	98.3
DIFFUSER (Phase 1)	10	15.9	83.5	2.1	97.7
	20	29.1	71.0	2.9	97.5
	30	29.1	71.1	3.5	96.8
	40	56.1	43.9	6.1	94.8
SDOX (Phase 2)	43	72.9	27.1	10.9	90.6
	46	81.8	18.2	12.5	89.7
	49	85.3	14.7	15.2	87.4
	52	85.4	14.6	15.5	87.0
	56	80.0	20.0	14.3	87.7
DIFFUSER (Phase 3)	60	24.1	70.9	4.8	90.6
	64	36.8	59.6	6.8	90.1
	68	36.7	58.9	7.6	88.5
	71	29.8	64.0	5.4	88.7
	74	27.8	61.5	4.3	85.4
SDOX (Phase 4)	76	48.3	50.3	9.9	89.5
	77	42.7	57.4	9.4	91.4
	78	44.4	53.9	9.2	89.9

a maximum of 15.5% at P2. As previously reported by Lin et al. (2010), large fractions of particles with diameters lower than 10 μm could be adverse impact on the membrane filtration performance.

Table 3 also indicates that as soon as the SDOX system was taken out of the MBR and replaced by the bubble diffuser, both the 1–50 μm fraction as well as the 1–10 μm fraction decreased back to similar values as observed during P1. That is, the fraction of small particles in the mixed liquor decreased during P3. This is reflected in an improved membrane filtration performance during P3. Finally, when the SDOX system is introduced back in P4, again the fraction of both the 1–50 μm and 1–10 μm ranges increased with a consequent deterioration of the membrane filtration performance. Therefore, the amount of small particles presented in the mixed liquor sludge had a detrimental effect on the membrane filtration, and the high-shear forces introduced by the SDOX system contributed to create that shift on the PSD increasing the proportion of small particles.

Kim et al. (2001) also reported on the influence of the shear effect on the membrane filtration performance of MBRs. The authors evaluated the impacts of the shear effects created by two different types of pumps, a centrifugal pump and a rotary pump, on the membrane filtration performance of a crossflow MBR. The authors concluded that the rotary pump imposed greater shear to the microbial flocs than the centrifugal pump. The percentage of flocs below 10 μm increased from 23% to 61% in the rotary pump compared to the centrifuge pump. Thus, lower fluxes were obtained when operating with the rotary pump (20 L/m^2/h) compared to when operating with the centrifuge pump (36 L/m^2/h) after only six days of operation. Likewise, as observed in our research, the shear effects exerted an impact on increasing the fraction of small particles with a detrimental effect on the membrane filtration processes. Similarly, Wisniewski and Grasmick (1998) also operated an MBR exposing the sludge at different shear forces by increasing the cross flow velocity. As the cross flow velocity increased, the fraction of small particles with average sizes below 10 μm increased, showing detrimental effects on the membrane filtration performance. In addition, higher resistance to filtration was reported when operating at cross flow velocities of 5 m/s than at 0.5 m/s. Therefore, indeed the introduction of the SDOX system increased the shear on the sludge shifting the PSD of the sludge to the low-size range of the distribution, increasing the percentage of small particles existed in the sludge. Consequently, the presence of a large fraction of small particles contributed to the formation of a more compact cake layer (less permeable) worsening the membrane filtration performance as observed during P2 and P4 in this research.

Fig. 4a also indicates the DSVI of the sludge as a function of the operational time. The DSVI is utilized to determine the settleability of concentrated sludge. Although the solid-liquid separation in MBRs is achieved by a membrane filtration process and not by gravity settling (Van den Broeck et al., 2012), an increased on the DSVI values could indicate some deterioration of the membrane filtration performance. At the beginning of P1, good settleabilities of the sludge were observed with DSVI values lower than 100 mL/g; afterward, the DSVI increased indicating a poor settleability of the sludge. The deterioration of the sludge settleability was observed when the MPS decreased below 100 μm. However, at that point, still during P1, a deterioration on the membrane performance was not observed. After the introduction of the SDOX unit (beginning of P2), the PSD kept shifting to the lower side of the range, and the sludge settling properties became progressively worse in such period. The increase on the DSVI implies deflocculation of the sludge shifting the PSD towards low particles sizes (i.e., increasing the fraction of small particles in the sludge). The presence of a non-settleable sludge could also possibly indicate a reduced membrane filtration performance (Ognier et al., 2002; Rosenberger et al., 2006). The DSVI during P3 also exhibited a better sludge settleability compared to P2, and it kept improving towards the end of the phase. Thus, the results may suggest that the sludge originally exposed to high extra shear conditions could recover in terms of membrane filtration performance once the conditions causing the high-shear are eliminated. During P4 an increase on the fraction of small particles were observed; however, no major changes on the DSVI were noticed.

3.2.3. Presence of soluble microbial products and extracellular polymeric substances

Organic components such as EPS and SMP are regarded as the main contributors to membrane fouling (Drews, 2010; Shah et al., 2021); the membrane pore blocking process is also intimately associated with the presence of EPS and SMP compounds (Guo et al., 2012; Zhang et al., 2015). EPS can be described as a mixture of high molecular polymers. Proteins, polysaccharides, humic acids, lipids, and nucleic acids, are the major constituents of EPSs (Frolund et al., 1996). EPS can make up to 50–80% of the total sludge floc weight (Meng et al., 2016). Because of their strong adhesive and cohesive characteristics, EPS acts as a bridge to allow bacteria and their surrounding particles to clump together (Wingender et al., 1999). SMP consists of soluble organic compounds generated because of the bacterial metabolism and autolysis; they can also be present in the influent wastewater (Barker and Stuckey, 1999).

The concentrations of EPS and SMP were determined during the evaluated period, and they are presented in Fig. 4c. EPS_p and EPS_c concentrations of 78.9 and 32.5 mg/L, respectively were observed right after inoculating the MBR at the beginning of P1. The EPS_p decreased towards the end of P1 reaching a concentration of approximately 50.7 mg/L, while the EPS_c did not fluctuate much during the P1 observing an EPS_c concentration of 33.9 mg/L at the end of P1. Right after introducing the SDOX unit during P2, no major changes on the EPS concentrations were noticed. Similar EPS concentration as in P2 were reported for P3 and P4. It can be concluded that after the sludge was acclimated to the new operational conditions in the MBR, similar EPS concentrations were observed for the entire evaluated period.

Judd and Judd (2011) reported severe fouling at EPS concentrations ranging from 20 to 80 mg g/MLVSS. On the contrary, Geng and Hall (2007) reported the absence of a correlation between membrane fouling and the presence of EPS. The authors claimed that the presence of SMP in the mixed liquor floc was more influential to membrane fouling than the presence of EPS. In our study, the EPS values were within the range reported by Judd and Judd (2011). The membrane filtration performance was not much affected during P1 and P3, in spite of the reported EPS concentrations, whereas the membrane filtration was severely affected during P2 and P4. Therefore, in our study the presence of EPS could not be linked to the membrane filtration performance. The sludge flocs during P2 and P4 were exposed to high-shear conditions; thus, EPS could have been released into the sludge mixture. However, marginal changes on the EPS concentrations were observed during P2 and P4 disregarding such a hypothesis. The SDOX unit shifted the PSD of the sludge to smaller-size particles. However, such changes on the PSD could not be reflected on the increase on the EPS concentrations that could have affected the membrane filtration performance.

The SMP value gradually decreased during the acclimatization period in P1 right after inoculating the MBR. However, after day 20, the SMP concentration continuously increased until the end of P1. In P2, the SDOX unit was introduced and the SMP concentration rapidly increased approximately doubling the SMP values from 0.25 to 0.53 L/mg/m. During P3, the SMP values slightly decreased, although showing similar values compared to P2. At the start of phase P4, similar SMP values as observed in P3 were reported. However, the SMP concentration largely dropped at the end of P4.

The increase in the SMP values during P2 was consistent with the increase in the TMP as reported in Fig. 2. That is, the presence of such SMPs values during P2 could contribute to a reduced membrane filtration performance. The SMPs produced by the microbial metabolism, although not fully characterized during this research, consists of dissolved macromolecules and colloids surrounding the sludge flocs. As the SMPs penetrate through the membrane surface, such substances could be readily adsorbed onto the membrane pores. Thereby, they may accumulate on the membrane surface, eventually forming a gel layer. The gel layer can bring more foulants such as sludge flocs. Particularly, small-sized sludge particles can be easily adhered to the membrane surface, forming a cake, which may ultimately lead to a higher degree of irreversible membrane fouling (Zhang et al., 2015). It is generally known that a cake layer plays a larger role than the internal membrane pore clogging in terms of membrane fouling (Park et al., 2015). The membrane filtration performance was deteriorated during P2, and that could be eventually related to the presence of SMP. However, same SMP values were reported during phase P3, and the membrane filtration performance was not worsened. In this context, the membrane filtration performance cannot be directly linked to the presence of SMP.

3.3. Effects of the supersaturated dissolved oxygen system on deposits on membrane surface

Fig. 5a shows the images of the fouled membranes when the MBR was equipped with the diffuser aeration system (M#1 to M#3) or with the SDOX unit (M#4 to M#9). Membrane fouling can be expressed by the overall resistance to filtration composed of three different resistances: the membrane intrinsic resistance, the cake layer resistance, and the pore blocking resistance. Membrane fouling in a submerged MBR stems from a combination of cake layer formation and pore blocking (Chen et al., 2017), both of which always coincide because of the interaction between the biomass metabolism and the membrane (Meng et al., 2007a). Among the two phenomena, the cake layer formation on the surface of the membrane has been considered the primary membrane fouling-causing factor (Hu et al., 2016; Turken et al., 2019); the cake layer accounts for approximately 90% of the total resistance to filtration (Guo et al., 2012). Meng et al. (2007b) in a study aimed at evaluating the characterization of the cake layer in submerged MBRs reported that the cake layer resistance contributed the most to the overall resistance. The authors reported resistances of approximately 8.7, 84, and 7.3% for the membrane intrinsic, cake layer, and pore blocking resistances, respectively. Thus, the cake layer resistance contributed by far the most to the membrane fouling in submerged MBRs. The cake layer resistance can be caused by the presence of: (i) microorganisms (biofouling); (ii) biopolymers or organic matter (organic fouling); and/or (iii) inorganic matter (inorganic fouling). The authors reported that the cake layer fouling in submerged MBRs consisted of sludge – biomass/VSS (63%), inorganic compounds (23%),

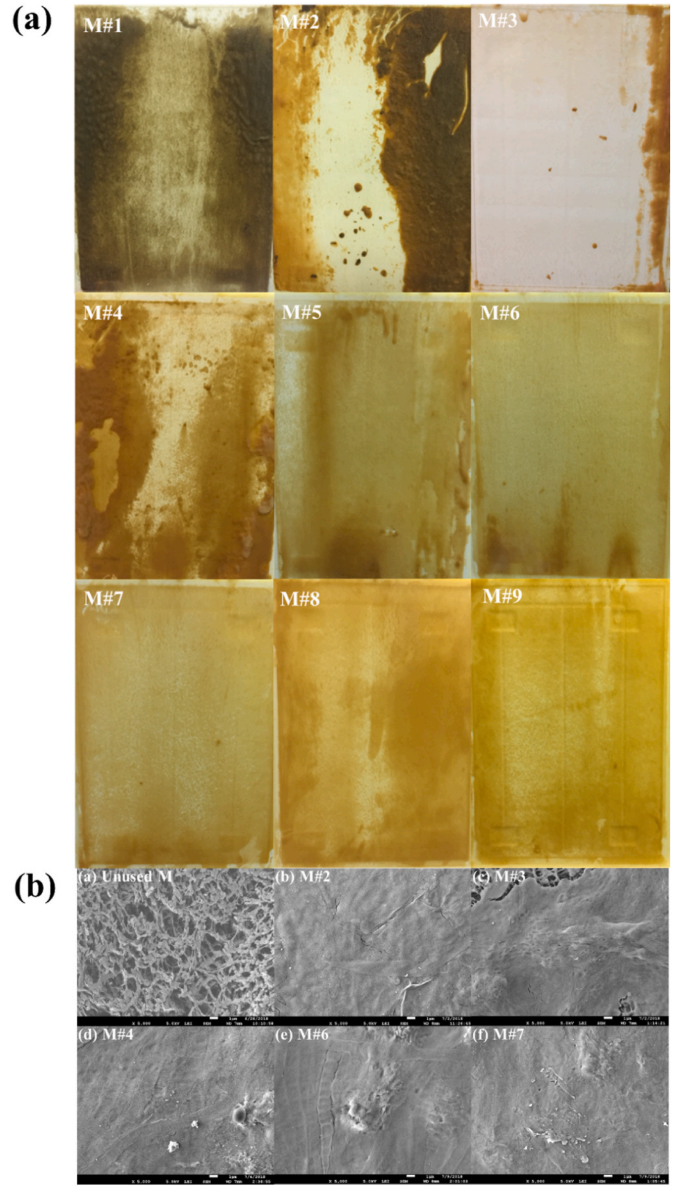

Fig. 5. (A) Images of the fouled membranes for the membrane bioreactor equipped with bubble diffusers (M#1 to M#3) and with the supersaturated dissolved oxygen unit (M#4 to M#9); and (b) Scanning electron microscope images of the fouled membranes: Unused membrane (a), M#2 (b), M#3 (c), M#4 (d), M#6 (e), and M#7 (f).

solutes (8%), and colloidal material (6%). The last two exhibited a higher tendency to membrane fouling by pore blocking (Du et al., 2020; Meng et al., 2007b). The larger the particles, the easier to detached from the membrane surface and not contributing much to the cake layer resistance, or forming a loose cake layer. On the other hand, the smaller the particles, the stronger the tendency to deposit onto the membrane surface and create a compact cake later; i.e., the accumulation of small particles on the surface of the membrane contributes to form a more compact cake layer decreasing the cake layer porosity and thereby contributing to membrane fouling (Chen et al., 2022). In addition, solutes (macromolecules) and colloidal material can be adsorbed onto the cake layer and/or are directly deposited on the surface of the membrane contributing also to the formation of a less permeable cake layer. In addition, macromolecules and colloidal material can easily penetrate into the membranes pores and lead to severe membrane pore blocking. Meng et al. (2007b) reported that the causes of biofouling, organic fouling, and inorganic fouling could coexist in the cake layer resistance; the authors observed bacteria clusters covered with biopolymers/organic matter in the cake layer. Mostly, the biopolymers consisted of macromolecules such as polysaccharides. In addition, the presence of inorganic compounds could contribute to precipitate out certain biopolymers having ionic constituents such as SO_4^{-2}, CO_3^{-2}, PO_4^{-3} and OH^-. Cations such as Ca^{+2}, Mg^{+2}, Al^{+3}, and Fe^{+2} could origin the precipitation of such substances contributing to inorganic fouling. In addition, the presence of carbonate species (due to the constant production of CO_2 due to the aerobic respiration) can contribute as well as to inorganic fouling. Thus, there is a synergistic interaction in the cake layer of bacterial, colloids, macromolecules, and inorganic elements all contributing to the cake layer resistance and membrane fouling. Biopolymers (colloids and macromolecules – such as polysaccharides) have the strongest effect on membrane fouling (Kim et al., 2001; Wang and Li, 2008). The provision of aeration (membrane scouring) partially contributes to remove the cake layer, although cannot be completely removed; particularly, the more loose the compact layer, the better the performance of the membrane scouring on preventing membrane fouling.

At the beginning of the evaluation, during P1 when the MBR was provided with the bubble diffuser aeration, the material attached to the surface of the fouled membrane exhibited a dark brown color as observed in images M#1, M#2, and M#3 in Fig. 5a. The MPS and PSD described in Fig. 4a and b, respectively indicate the presence of relatively large-size particles in the sludge. Thus, the cake layer observed in Fig. 5a probably consisted mostly of biomass (VSS) as previously described by Bell et al. (2016) and Meng et al. (2007b); thus, the cake layer was not much compacted yielding a relatively porous cake layer. That was confirmed considering the relatively good membrane filtration performance described in Fig. 2 and Table 2, also considering that a relaxation phase was not included in the membrane filtration cycles. In addition, Fig. 5a (images M#1, M2, and M#3) show on the left and right sides of the membrane more biomass deposited and accumulated on the surface of the membrane than on the center of the membrane. This could also indicate the presence of a relatively loose cake layer since the provision of the membrane air scouring was able to remove partially the cake layer; particularly, the cake layer located in the center of the membrane where the air scoring seemed to be more intense. Thus, biofouling seemed to account as the major fouling mechanisms in P1 contributing to the cake layer resistance.

The appearance of the cake layer on the membranes during P2, i.e. M#4 to M#9 in Fig. 5a, is different from the membranes during P1 - M#1 to M#3. A most homogeneous cake layer could be observed on the membranes in P2. In addition, the cake layer in P2 seemed thinner than the cake layer observed in P1. In essence, the cake layer in P2 looks more compact than in P1. When looking at the MPS and PSD described in Fig. 4a and b, respectively, an important reduction on the particle size of the sludge was observed during P2. Such a reduction on the particle size was due to the strong shear forces introduced by the SDOX system. Therefore, there were higher fractions of small-sized particles present in the sludge mixture eventually contributing to the formation of a more compact cake layer. This was also noticed by a decrease on the membrane filtration performance as indicated in Fig. 2 and Table 2, and the presence of a more frequent fouling. In addition, the exposure of the sludge mixture to such high-shear forces introduced by the SDOX unit could promote the release and/or generation of colloidal material and macromolecules that could be adsorbed onto the cake layer/membrane surface promoting a less permeable cake layer (Judd and Judd, 2011; Stricot et al., 2010). Moreover, such substances could easily penetrate through the membrane pores causing severe membrane pore blocking (Guo et al., 2012). Thus, the presence of higher concentrations of small particles, colloidal material and macromolecules compared to P1 seemed to contribute to membrane fouling deteriorating in such a way the membrane filtration performance during P2. The main contributor to membrane fouling in the cake layer during P2 seemed to be the organic fouling (macromolecules and small particles); the provision of air membrane scouring seemed not to play a major effect to prevent fouling as was the case in P1.

Fig. 5b shows the SEM images of the selected membranes photographed at × 5000 resolution to investigate the morphological features of the cake layers. The unused membrane, Fig. 5b (a), exhibited a clean surface with the membrane pores clearly visible on the membrane surface. The rest of the SEM photographs show the membrane surface completely covered by the cake layer. M#3, Fig. 5b (c) exhibits some of the membranes pores visible together with the cake layer. Similar observations are shown in Fig. 5a for the same membrane. Not much difference was observed on the SEM photographs for the evaluated membranes. Certainly, the fouling was associated with the presence of a cake layer in all the evaluated membranes.

The elemental composition of the surface of the fouled membranes was also evaluated by conducting EDX analyses, and the results are presented in Table 4. The filtration membrane was made of chlorinated polyethylene, and the elemental composition presented in Table 4 for the unused membrane was in accordance showing the presence of C, O, and Cl. The membranes analyzed during P1 exhibited a changed on the elemental composition with respect to the unused membrane. M#2 and M#3 still exhibited the presence of C, and O; however, higher percentages of O were reported than in the unused membrane (13.56% in M#3 compared to 1.26% in the unused membrane), and the presence of N was observed (10.61% in M#3) but not present in the unused membrane. A low proportion of metallic elements along with non-metallic elements such as Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc, and Fe, were present on the membrane surface. The presence of inorganic elements except Cl and Sc was in line with the results from Meng et al. (2007b) when operating a submerged MBR treating synthetic wastewater. Thus, the presence of C, O, N, and trace of inorganic elements in the EDX analysis of the surface of the cake layer is in agreement with the findings reported in Fig. 5. Such findings could support the presence of biomass – VSS in the cake lake formation in the membranes during P1, and thereby also contributing to support that biofouling could have been the main cause of fouling during P1.

The EDX analysis of the evaluated membranes during P2 exhibited some changes compared to the EDX analysis of the membranes during P1. A higher proportion of C and O than in P1 was reported as described in Table 4. Moreover, N was no longer detected, and the proportion of inorganic elements increased. The absence of N could indicate that the biomass and VSS were no longer the major contributors to the cake layer; in addition, the larger proportion of C and O could also suggest the presence of biopolymers such as polysaccharides as reported by Meng et al. (2007b). Particularly, the high-shear effects introduced by the SDOX unit were eventually favoring the deposit of such macromolecules in the cake layer rather than biomass. In addition, the increase on the

Table 4
Elemental composition of the surface of the selected evaluated membranes.

Element	Weight (%)					
	Unused membrane	M#2	M#3	M#4	M#6	M#7
C	74.57	62.95	65.57	74.79	70.07	74.38
N		13.19	10.61			
O	1.26	11.88	13.56	20.47	24.54	20.22
Na	0.07	0.37	0.05	0.38	0.33	0.66
Mg		0.1	0.16	0.52	0.93	0.38
Al		0.01				
Si		0.1			0.09	
P			0.29	1.51	1.24	1.53
S				0.07	0.04	
Cl	24.1	11.15	8.81	0.4	0.22	1.29
K		0.09	0.63	0.86	0.7	0.8
Ca		0.13	0.18	0.67	1.59	0.48
Sc		0.03				
Fe			0.13	0.33	0.24	0.25
Total	100	100	100	100	100	100

proportion of inorganic elements occupying the fouled membranes could also influence the cake layer by promoting the precipitation of macromolecules and by introducing a bridging effect (Hong and Elimelech, 1997; Seidel and Elimelech, 2002). Thus, the EDX analysis of the elemental composition of the surface of the membranes also suggested some changes on the cake layer structure supporting the membrane filtration performance findings previously reported.

The high-shear forces introduced by the SDOX unit introduced changes on the sludge mixture reducing the particle size of the mixture and promoting a different composition and structure of the cake layer. The cake layer changed from P1 to P2 from a more loose cake layer to eventually a more compact layer. The presence of biomass as the main component of the cake layer during P1 was most likely replaced by biopolymers. Thus, the major fouling contributor due to the cake layer seemed to change from biofouling in P1 to organic fouling in P2. However, due to the strong reduction on the particle size, and the presence of macromolecules and colloidal mater, membrane pore blocking cannot be rejected as an important contributor to membrane fouling during P2.

3.4. Limitations on the supersaturated dissolved oxygen-MBR concept

This study aimed at evaluating the membrane filtration performance of an MBR when equipped with an innovative oxygen transfer technology supplying concentrated oxygen, the SDOX system. Concentrated oxygen supply systems have been widely used in water remediation applications and in aquaculture systems for supplying DO; however, they were barely used in wastewater treatment applications. Kim et al. (2019) indicated that the operation of MBRs is limited by inefficiencies introduced by conventional bubble-diffuser aeration systems. Conventional bubble diffusers exhibit an inefficient oxygen transfer performance at the MLSS concentrations higher than 10 g/L, commonly reported for operating MBR systems. The oxygen transfer inefficiencies are exacerbated when working at even higher MLSS concentrations limiting either expanding the treatment capacity, or reducing the footprint requirements of MBR systems. Concentrated oxygen supply technologies, such as the SDOX system, can uncap such limitations imposed by conventional bubble diffusers. Higher OTRs, higher alpha factors, and higher OTEs were reported for the concentrated oxygen supply system compared to bubble diffusers (Kim et al., 2020). Particularly, the higher the MLSS concentration in the MBR, the better the oxygen transfer performance of the concentrated oxygen supply system compared to the bubble diffusers (Kim et al., 2020). In addition, the biological performance of the MBR was not negatively affected by the action of the concentrated oxygen supply system (Kim et al., 2021); similar performance in terms of carbon removal and nitrification was reported by Kim et al. (2021) in an MBR operated either with bubble diffusers or with the SDOX system. Moreover, no major changes in the microbial population dynamics were reported by the authors. Therefore, concentrated oxygen supply systems, such as the SDOX system, introduce major advantages for operating biological wastewater treatment system, particularly MBR systems at much higher MLSS concentrations, uncapping their operational limitations and increasing the overall energy efficiency due to the improved oxygen transfer (higher OTRs, higher alpha factors, and higher OTEs). Compared to conventional diffused aeration systems, the SDOX system requires much less energy consumption especially at high MLSS above 20 g/L, which could be applicable to the MBR-SDOX concept. However, this study demonstrated, at the evaluated experimental conditions in this research, that the membrane filtration performance could be negatively affected by the shear forces and high-pressure conditions introduced by the concentrated oxygen supply systems. The changes in the PSD increasing the concentration of particles with diameters less than 50 and 10 μm exhorted a detrimental effect on the membrane filtration performance. Nevertheless, the present study was carried out using a bench scale SDOX system provided with small size diameter tubing and a small orifice at the inlet of the SDOX; these conditions imposed additional shear forces that would not have been observed when operating a full-scale SDOX system, thereby calling for large-scale experiments to support the absence of lower intensity of shear forces.

3.5. Practical applications and future research prospects on the supersaturated dissolved oxygen-MBR concept

In this study, only one type of membrane was used and backwash was impossible to perform. Moreover, to push the limits of the membrane filtration a relaxation phase in between filtration phases was not added. Besides, this study only evaluated the short-term exposure effect of the membrane to the SDOX systems and the sludge in the bioreactor was cultivated on synthetic wastewater. It was assumed that real sewage sludge may have different properties and may be less prone to cause fouling when exposed to SDOX. During P2 and P4, the MPS increased (or at least stopped decreasing) towards the end of these phases, so the membrane filtration performance could recover during a long-term exposure to the SDOX unit; this, however, was not evaluated in this research. Therefore, further research is required to explore membrane fouling propensity of MBR operated with SDOX under following conditions (i): long-term periods (at least 3 times SRTs); (ii) different membrane types (hollow fiber and/or ceramic membranes); (iii) under relaxation and backwash modes; and (iv) real municipal and/or industrial wastewater. Finally, the SDOX system could be applied as an oxygen supply source for biological treatment of industrial/municipal WWTPs that take advantage of SDOX's low footprint.

4. Conclusions

The membrane filtration performance under the experimental conditions investigated in this study was negatively affected by the short-term exposure of the membranes to the concentrated oxygen supply system (i.e., the SDOX). It was impossible to establish a clear relationship between the presence of EPS and SMP and the membrane filtration performance. Eventually, the presence of high fractions of small particles (size 1–10 and 1–50 μm) had a stronger negative effect on membrane filtration performance than the presence of EPS and SMP. The cake layer formation was observed on the fouled membranes exposed to the SDOX system, which contributed to the deterioration of membrane filtration performance. Biofouling appeared to be the major contributor to the persistence of the cake layer when the membranes were not exposed to the SDOX system, while organic fouling appeared to be the primary factor when the membranes were exposed to the SDOX system.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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