Municipal Wastewater Reclamation and Water Reuse for Irrigation by Membrane Processes

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Municipal wastewater was treated by membrane bioreactor (MBR), and the obtained MBR effluent was then treated by reverse osmosis (RO), and nanofiltration (NF). The MBR effluent was additionally treated by reverse osmosis (XLE) and nanofiltration (NF90 and NF270) membranes. RO and NF permeate output streams were assessed for their utilization in agricultural irrigation. The MBR used a hollow fiber ZeeWeed 1 ultrafiltration membrane. Conductivity, turbidity, total suspended solids, chemical oxygen demand, and dissolved organic carbon were rejected by MBR with average values of 10 %, 100 %, 99.8 %, 96 %, and 88 %, respectively. Further treatment with RO/NF membranes showed additional reduction in all measured parameters. According to results, MBR effluent belongs to the 'slight to moderate' degree of restriction on use due to conductivity, chloride, and sodium concentrations. RO/NF permeate, based on all parameters, belongs to the 'none' degree of restriction on use, except on sodium adsorption ratio (SAR), where it belongs to the 'severe' degree of restriction on use. Based on conductivity and SAR parameters, assessment of produced water quality obtained by blending of two effluents (50 % of MBR and 50 % of NF270 permeate) resulted in an output stream appropriate for irrigation, proving that the blending of output streams in this ratio is a good strategy for agricultural irrigation.

Keywords:

water reuse, municipal wastewater, membrane processes, irrigation

Introduction

Climate change, along with industrialization and population growth, is becoming a global problem in both the supply of drinking water and water for industrial and agricultural purposes. Climate change is primarily seen through the rise in temperature and rapid and intense droughts and floods. The Intergovernmental Panel on Climate Change published its 'Special report on global warming of 1.5 °C', demonstrating how important it is to keep temperature increases below 2 °C, which would reduce the risks to human well-being, ecosystems, and sustainable development.¹ The negative impact of climate change will continue at least throughout the next decades; thus, the development and application of new technologies aimed at the mitigation of negative effects of climate change are needed.²

In the last decades, water is no longer considered an inexhaustible resource.³ Water scarcity and droughts (caused by climate change), which are increasingly frequent and widespread across Europe, have become a major challenge. This is evident from the fact that water scarcity is affecting at least 11 % of the European population and 17 % of its territory.⁴ Therefore, the European Innovation Partnership gave high importance to water reuse and recycling by included them in its top five priorities.⁴ However, on the European level, there are no regulations concerning wastewater reuse, apart from the Urban Wastewater Treatment Directive (91/271/EEC), which under Article 12 states that "Treated wastewater shall be reused whenever appropriate".⁵ Thus, wastewater reuse across Europe is carried out under national regulation or guidance. So far, wastewater reuse in Croatia is nonexistent (because of nonexistent regulations and prohibition on use of unregulated water).

In times of prolonged drought, agriculture has an increased need for water; thus, it is important that reclaimed wastewater meets international guidelines for irrigation, which assures the safety of crops and minimizes the risks on human health and the environment. These objectives can be achieved by promising treatment technologies, such as membrane technologies of microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis

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Fig. 1 – Schematic representation of MBR-RO/NF treatment

(RO), and membrane bioreactor (MBR). In the last decade, membrane technologies have been used for wastewater reuse.^{6–11} However, most recent papers ^{6,7,11,12} reported processing of MBR effluents by NF and RO membranes in order to improve its quality and meet irrigation regulations. General characteristics of reclaimed wastewaters intended for irrigation should be compared, and must comply with appropriate standards.⁹

The aim of this study was to characterize and evaluate MBR, NF, and RO treated municipal wastewater for its reuse in agricultural irrigation in accordance with various international guidelines.^{13,14}

Materials and methods

Experimental set-up

A small lab-scale submerged MBR used for municipal wastewater treatment was equipped with adequate sensors (measurement of pressure, flow, temperature, and level regulation), an Alpha Programming (SW0D5-ALVLS-EU) (Mitsubishi Electric Corporation, Japan) data acquisition system, in order to monitor the system (Fig. 1). The hydraulic volume of the MBR was 5 L. The MBR was seeded with biological sludge from a municipal WWTP (Čakovec, Croatia).

The immersed laboratory MBR used a submersible UF hollow fiber module ZeeWeed 1 (ZW-1) from GE Water & Process Technologies (Hungary). The mode of filtration in hollow fiber membrane, depending on the direction of permeate flow, was outside-in. Permeate flow was measured with Cole Parmer (C3290843) flowmeter and connected to a



Fig. 2 – Schematic representation of RO/NF laboratory set-up

computer. The nominal membrane surface area was 0.046 m² with nominal membrane pore diameter (size) of 0.02 μ m. Membrane characteristics given by the manufacturer are listed in Table 1.

NF and RO were performed with NF270 (loose NF), NF90 (tight NF), and XLE (dense RO) membranes (Dow-Filmtec, USA) at 12 bar in a laboratory set-up (Fig. 2) and as described in a previous publication.¹⁸ The feed (MBR effluent) from a 10-L tank was circulated through the membrane cell at a flow rate of 3 L min⁻¹ (fluid velocity 0.75 m s⁻¹). The membrane characteristics are listed in Table 1. Prior to the experiment, the pristine membranes were washed with demineralized water (7 L) to remove the conserving agent, precompressed for 1 h at 15 bar, and stabilized for 30 min at working pressure. NF/RO experiments were carried out in batch

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	ZW-1	NF270	XLE	NF90
Typical transmembrane pressure (bar)	0.10-0.50	41	41	41
Maximum operating temperature (°C)	40	45	45	45
Maximum cleaning temperature (°C)	40	35	35	35
Operating pH	5–9	2–11	2–11	2–11
Pore diameter (µm)	0.02	-	-	_
Cleaning pH range	2.0-10.5	1–12	1–12	1–12
Maximum OCl ⁻ exposure (mg L ⁻¹)	1000	-	-	_
Allowed flux (m ³ day ⁻¹)	55-110 ^a	200.59 (4.56) ^b	133.41 (2.04) ^b	139.95 (0.90) ^b
Material ^{16,17}	Polyvinildenfluorid (PVDF)	Semi-aromatic poly (piperazin-amide)	Polyamide	Polyamide
MWCO ^c (Da)	200 000	150-30018	10018	100-20018

Table 1 – Characteristics of used membranes^{15–17}

^a – determined by manufacturer (m³ day⁻¹)

^b – demineralized water flux and standard deviation measured in this study (N = 54), L m⁻² h⁻¹

^c – MWCO – molecular weight cut-off

circulation mode for 3 h, meaning that permeate and retentate were circulated back into the feed tank, and one permeate sample of each membrane was taken after 3 h and analyzed.

Municipal wastewater

The municipal wastewater was sampled in a WWTP, with a capacity of 75 000 population equivalent, located in Čakovec, Croatia, after large screening (removal of large floating objects) and grit chamber (removal of sand and grease). The wastewater is 1/3 of industrial origin and 2/3 of domestic origin. Its composition is given in Table 2. Municipal wastewater samples were brought in 2 batches. Each batch was characterized and continuously fed to the MBR. The quality of water samples collected during investigation was evaluated using both Food and Agricultural Organization (FAO) and World Health Organization guidelines.13,14 Summarized criteria for agricultural irrigation are given in Table 3. The 'severe' degree of restriction means that this type of water may cause serious drop in infiltration (downward movement of water through soil) and it is unusable for irrigation; 'slight to moderate' degree of restriction means that it may have some negative effects on infiltration, but it can be used with caution; and 'none' degree of restriction on use refers to water that will have no harmful effects on infiltration.⁶

Operating conditions

The MBR experiments were carried out for 3 days at the following operating conditions: 26.8 ± 0.8 °C, transmembrane pressure (TMP) around -0.035 bar, permeate flux of 18 L m⁻² h⁻¹, hydraulic retention time (HRT) of 6 h, air supply rate 20 L min⁻¹, and

 Table 2 – Physicochemical characteristics of municipal wastewater

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Parameter	Unit	Feed
κ	$\mu S \ cm^{-1}$	1174±2
рН	_	7.22±0.11
turbidity	NTU	248±11
DOC	mg L^{-1}	126.6±7.3
COD	mg L^{-1}	478±132
F-	mg L^{-1}	0.096±0.003
Cl⁻	mg L ⁻¹ (meq L ⁻¹)	156.0±2.4 (4.40±0.07)
NO_2^{-}	mg L ⁻¹	64.35
NO ₃ ⁻	mg L ⁻¹	44.53±42.17
PO ₄ ³⁻	mg L ⁻¹	9.631±1.428
SO ₄ ²⁻	mg L ⁻¹	36.33±0.84
Li+	mg L ⁻¹	n.d.
Na ⁺	mg L ⁻¹ (meq L ⁻¹)	71.14±0.48 (3.09±0.02)
NH_4^+	mg L ⁻¹	n.d.
K^+	mg L ⁻¹	11.85±0.14
Mg^{2+}	mg L ⁻¹ (meq L ⁻¹)	22.05±0.04 (1.815±0.005)
Ca^{2+}	mg L ⁻¹ (meq L ⁻¹)	110.7±0.2 (5.525±0.005)
SAR	meq L ⁻¹	1.61±0.01
TSS	mg L ⁻¹	488±48

mixed liquor suspended solids (MLSS) of 10.9 - 11.5 g L⁻¹. The operation mode of the system was suction 10 min, and 1 min of backwash. Sludge retention time (SRT) is not mentioned here since no sludge was removed from the reactor throughout

Table 3 – Water criter	ria for agricultural irri	gation				
Parameter		Unit	Degree of restriction on use			
			None	Slight to moderate	Severe	
Salinity	$\kappa (EC_w)$	μS cm ⁻¹	<700	700 - 3000	>3000	
	TDS	mg L ⁻¹	<450	450 - 2000	>2000	
Infiltration (affects infilt	tration rate of water int	to soil. Evaluate using EC	w and SAR toge	ther)		
SAR	=0-3	EC _w =	>700	700 - 200	<200	
	=3 - 6	$EC_{w} =$	>1200	1200 - 300	<300	
	=6 - 12	$EC_w =$	>1900	1900 - 500	<500	
	=12 - 20	$EC_{w} =$	>2900	2900 - 1300	<1300	
	=20 - 40	$EC_{w} =$	>5000	5000 - 2900	<2900	
Na ⁺	Surface irrigation	mg L ⁻¹	<69	69 - 207	>207	
	Sprinkle irrigation	mg L ⁻¹	<69	>69		
Cl-	Surface irrigation	mg L ⁻¹	<142	142 - 354	>354	
	Sprinkle irrigation	mg L ⁻¹	<106.5	>106.5		
Total nitrogen (TN)		mg L ⁻¹	<5	5 - 30	>30	
Nitrogen (NO ₃ ⁻ N)		mg L ⁻¹	<5	5 - 30	>30	
TSS		mg L ⁻¹	<50	50 - 100	>100	
pН		_	6.5 - 8 (8.5)			
turbidity		NTU	<2			
DOC		mg L ⁻¹	-			
COD		mg L ⁻¹	_			
Anions and cations ¹⁸						
NO ₂ ⁻		mg L^{-1} (meq L^{-1})	_			
SO ₄ ²⁻		mg L^{-1} (meq L^{-1})	960 (20)			
Mg_2^+		mg L^{-1} (meq L^{-1})	61 (5)			
Ca ₂ ⁺		mg L ⁻¹ (meq L ⁻¹)	400 (20)			
Nutrients ¹⁸						
NO ₃ ⁻		mg L^{-1} (meq L^{-1})	140 (10)			
NH_4^+		mg L^{-1} (meq L^{-1})	90 (5)			
PO ₄ ³⁻		mg L^{-1} (meq L^{-1})	194 (2)			
<u>K</u> ⁺		mg L ⁻¹ (meq L ⁻¹)	78 (2)			

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the experimentation period (except small samples for analytical purposes). MBR permeate was collected for further RO/NF treatment.

Analytical methods and water analysis

The water analysis was conducted for the main wastewater parameters according to Standard methods¹⁹, which included conductivity (κ), pH, turbidity, organic content (chemical oxygen demand (COD), dissolved organic carbon (DOC)), anions (F⁻, Cl⁻, NO₂⁻, NO₃⁻, PO₄³⁻, SO₄²⁻) and cations (Ca²⁺,

 Mg^{2+} , Na⁺, NH₄⁺, K⁺), sodium adsorption ratio (SAR), and total suspended solids (TSS).

The carbon content was measured with Carbon Analyzer Shimadzu TOC- V_{WS} (Japan). Turbidity was measured with WTW Turb 430 (Germany) turbidimeter. Conductivity and pH were measured with SI Analytics HandyLab680 (Germany). The COD was determined with COD cell tests on spectrophotometer Hach Lange DR3900 (Germany), and ion content with Ionic chromatograph DIONEX ICS-3000 Thermo Fischer Scientific (SAD).

Results and discussion

In addition to the mitigation of possible health effects associated with the use of treated municipal wastewater in agriculture, good irrigation practices will need to be followed to ensure a good crop yield and to minimize risks to the environment.13 Therefore, the parameters of municipal wastewater and those of MBR and NF/RO effluents were compared to the criteria for agricultural irrigation (Table 3). The characteristics of sampled municipal wastewater used in this study are given in Table 2. The municipal wastewater was characterized by high turbidity and TSS, and relatively high COD. According to the parameters defined by FAO and WHO guidelines, this wastewater was not suitable for agricultural irrigation and the most problematic parameters were turbidity of very high value (260 and 237 NTU) and TSS (536 and 440 mg L⁻¹). According to other parameters, the wastewater could be used for irrigation with 'slight to moderate' restriction on use, only nitrate in the first sample (86.70 mg L^{-1}) belongs to 'severe' restriction on use.

MBR treatment

Municipal wastewater was first processed by MBR under conditions specified in the section *Operating conditions*. MBR effluent was characterized 3 times since it was treated with 3 different membranes in the second step (Table 4). MBR-1, MBR-2, and MBR-3 were effluents used for XLE, NF90, and NF270 membranes, respectively. Nevertheless, an explanation will be given for all three MBR effluents together, since they were fairly similar in composition.

Conductivity is a very important water quality factor for crop production because water with high conductivity causes physiological drought (this causes the inability of plants to compete with ions in the soil solution and water, which affects the crop).⁶ As expected, conductivity decreased slightly (9.3–10.2 %) since MBR is inappropriate for the retention of electrolytes (ions), which is in agreement with previous studies.^{12,20} According to the conductivity, MBR effluents satisfy 'slight to moderate' restriction on use. In addition to conductivity, sodium imbalance in irrigation water can have a substantial impact on crop production. When irrigation water has high sodium content relative to the calcium and magnesium contents, water infiltration decreases.⁶ Excessive levels of exchangeable sodium adversely affect the soil physicochemical properties7 and needs to be considered. The most used index is SAR. The decrease in SAR was very low since removal of cations (Na⁺, Ca²⁺, and Mg²⁺) with MBR and UF membranes was not expected.¹² Concerning SAR and conductivity, the MBR effluent fell into the category of 'none' degree of restriction on use. The pH of permeate increased above 8, which was in compliance with FAO guidelines, but a little high according to WHO guidelines. It was probably because the air flow in the MBR tank resulted in the desorption of CO₂ from the MBR mixed liquor.²¹ Turbidity and TSS are visual indicators of water quality, and MBR showed to be very effective since both decreased >99.8 %. This confirms the effectiveness of MBR for turbidity and TSS removal. Concentrations of chloride and sodium were also regulated, meaning that the MBR effluent fell into the category of 'slight to moderate' degree of restriction on use. Organic matter, expressed as COD and DOC, were decreased below 14.1 mg L⁻¹ and 13.98 mg L⁻¹, respectively, suggesting that their removal was higher than 96 % and 88 %, respectively. High removal of organics confirmed the high efficiency of MBR, and since sludge was taken from WWTP Čakovec, it was obvious that the sludge had acclimated to the used wastewater from the start of the experiment. The nutrients in the wastewater were below the level required by regulations, and after MBR, no significant changes were present. For nitrate and potassium, the concentrations were similar, while phosphate had decreased by 35 % - 54 % during the MBR treatment.

RO/NF treatment

The membranes for the second step were selected according to their characteristics to cover the range of loose NF to dense RO membranes. Characteristics of XLE, NF90, and NF270 effluents are given in Table 5. All monitored parameters were decreased additionally with tested NF/RO membranes. For conductivity, the highest removal was obtained with XLE membrane (96.9 %), and the smallest for NF270 (62.6 %). For NF90, the retention was 96.0 %, which was similar to XLE membrane. Turbidity was already low after MBR, but after treatment with RO/NF membranes, an additional small decrease was obtained. Organic compounds, expressed with DOC and COD, were below 0.35 mg L⁻¹ and <5.0 mg L⁻¹, respectively. Anions, cations, and nutrients (Table 4) were, in this case, below FAO and WHO regulations (Table 3), but after RO and NF treatment, they had additionally decreased. Depending on membrane type, their concentrations in the permeates differ. The lowest concentration of these parameters in permeate were obtained with XLE membrane, followed by NF90 and NF270, except for NO $_3^-$, PO $_4^{3-}$, and SO $_4^{2-}$. For these anions, the concentrations were very low; thus, their change was within measurement error, but the concentrations significantly decreased.

In general, the results after treatment with RO/ NF membranes are in accordance with the mem-

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Parameter	Unit	MBR-1	MBR-2	MBR-3
ĸ	μS cm ⁻¹	1057	1061	1063
рН	_	8.18	8.17	8.15
turbidity	NTU	0.51	0.39	0.27
DOC	mg L ⁻¹	8.98	13.63	13.98
COD	mg L ⁻¹	13.1	14.1	13.3
F-	mg L ⁻¹	0.0937	0.0929	0.1003
Cl-	mg L^{-1} (meq L^{-1})	151.0 (4.26)	157.1 (4.43)	161.7 (4.56)
NO ₂ ⁻	mg L ⁻¹	n.a.	n.a.	0.8532
NO ₃ ⁻	mg L ⁻¹	82.3	81.4	83.4
PO ₄ ³⁻	mg L ⁻¹	5.32	5.30	5.11
SO ₄ ²⁻	mg L ⁻¹	34.30	33.86	34.10
Li ⁺	mg L ⁻¹	n.a.	n.a.	n.a.
Na ⁺	mg L^{-1} (meq L^{-1})	69.57 (3.03)	69.09 (3.00)	69.66 (3.03)
NH_4^+	mg L ⁻¹	n.d.	n.d.	n.d.
K^+	mg L ⁻¹	10.69	16.83	19.53
Mg^{2+}	mg L^{-1} (meq L^{-1})	22.39 (1.84)	22.32 (1.84)	22.36 (1.84)
Ca ²⁺	mg L^{-1} (meq L^{-1})	116.6 (5.82)	112.8 (5.63)	111.5 (5.57)
SAR	meq L ⁻¹	1.55	1.55	1.57
TSS	mg L ⁻¹	0	0	0

Table 4 - Characteristics of MBR effluent

Table 5 – Characteristics of XLE, NF90, and NF270 permeate

Parameter	Unit	XLE	NF90	NF270
κ	μS cm ⁻¹	33.2	42.6	397
pН	_	6.94	7.15	8.06
turbidity	NTU	0.22	0.12	0.23
DOC	mg L ⁻¹	0.33	0.29	0.35
COD	mg L ⁻¹	<5.0	<5.0	<5.0
F-	mg L ⁻¹	n.a.	n.a.	n.a.
Cl-	mg L^{-1} (meq L^{-1})	11.18	22.13	63.77
NO ₂ ⁻	mg L ⁻¹	0.1557	n.a.	0.3728
NO ₃ ⁻	mg L ⁻¹	13.4	9.58	63.1
PO ₄ ³⁻	mg L ⁻¹	0.770	n.a.	n.a.
SO ₄ ²⁻	mg L ⁻¹	2.054	0.381	0.464
Li ⁺	mg L ⁻¹	n.a.	n.a.	n.a.
Na ⁺	mg L^{-1} (meq L^{-1})	4.68 (0.204)	6.24 (0.271)	38.01 (1.65)
NH_4^+	mg L ⁻¹	0.22	0.13	n.a.
K^+	mg L ⁻¹	1.77	17.65	5.90
Mg^{2+}	mg L^{-1} (meq L^{-1})	0.32 (0.026)	0.30 (0.025)	3.04 (0.250)
Ca ²⁺	mg L^{-1} (meq L^{-1})	1.68 (0.084)	1.58 (0.079)	29.9 (1.49)
SAR	meq L ⁻¹	0.87	1.19	1.77
TSS	mg L ⁻¹	0	0	0

brane characteristics, since XLE is a dense RO membrane with the smallest pore sizes (<1 nm), and NF270 is a loose NF membrane with pores up to 2 nm²², while NF90 membrane is a tight NF membrane showing characteristics very similar to RO membranes.

Comparing the experimental results (Table 5) with the parameters given by regulation (Table 3), it can be concluded that the permeates of XLE, NF90, and NF270 membranes satisfy the requirements for the category of 'none degree of restriction on use for all parameters if conductivity is taken into account. According to SAR, membrane permeates belong to the category of 'severe' degree of restriction on use. Infiltration problem, expressed by SAR and EC_w , was caused by unbalanced removal of sodium,

calcium, and magnesium ions affecting SAR values.⁷

Salinity affects crop water availability, while SAR affects infiltration of water into the soil. It can be concluded that MBR effluent, RO, and tight NF permeates are unsuitable for irrigation. MBR effluent is very often unsuitable because of high salinity (conductivity) and specific ions (in this case chloride, sodium, nitrate).^{6,12} The RO and tight NF permeates are also unsuitable due to high removal of sodium, calcium, and magnesium, which directly impact the SAR values.¹⁰ Bearing this in mind, mixing RO/NF permeates with MBR effluent in appropriate ratios may have suitable composition values for irrigation. Therefore, calculated values are presented in Table 6.

 Table 6 – Water quality obtained with XLE, NF90, and NF270 membranes using various blending ratios of MBR effluent and RO/NF permeate

Parameters	MBR effluent	0	0.25	0.5	0.75	1	
	RO/NF permeate	1	0.75	0.5	0.25	0	
XLE membrane							
κ , μ S cm ⁻¹		33.2	289	545	801	1057	
Cl ⁻ , mg L ⁻¹		11.18	46.13	81.09	116.0	151.0	
Na ⁺ , mg L ⁻¹		4.680	20.90	37.12	53.35	69.57	
K^+ , mg L^{-1}		1.770	4.000	6.230	8.460	10.69	
Mg^{2+} , mg L^{-1}		0.320	5.837	11.35	16.87	22.39	
Ca^{2+} , mg L^{-1}		1.6800	30.41	59.14	87.87	116.6	
SAR, meq L ⁻¹		0.87	1.04	1.21	1.38	1.55	
NF90 membrane							
κ , μ S cm ⁻¹		42.60	297.2	551.8	806.4	1061	
Cl ⁻ , mg L ⁻¹		22.13	55.87	89.61	123.4	157.1	
Na ⁺ , mg L ⁻¹		6.240	21.95	37.66	53.38	69.09	
K^+ , mg L^{-1}		17.65	17.44	17.24	17.03	16.83	
Mg^{2+} , mg L^{-1}		0.300	5.805	11.31	16.82	22.32	
Ca ²⁺ , mg L ⁻¹		1.580	29.38	57.19	84.99	112.8	
SAR, meq L ⁻¹		1.19	1.28	1.37	1.46	1.55	
NF270 membrane							
κ , μ S cm ⁻¹		397.0	563.5	730.0	896.5	1063	
Cl ⁻ , mg L ⁻¹		63.77	88.25	112.7	137.2	161.7	
Na ⁺ , mg L ⁻¹		38.01	45.92	53.83	61.75	69.66	
K^+ , mg L^{-1}		5.900	9.307	12.71	16.12	19.53	
Mg^{2+} , mg L^{-1}		3.040	7.870	12.70	17.53	22.36	
Ca^{2+} , mg L^{-1}		29.90	50.30	70.70	91.10	111.5	
SAR, meq L ⁻¹		1.77	1.72	1.67	1.62	1.57	

The blending of 50 % of MBR effluent with 50 % of NF270 permeate gives a SAR value of 1.67 with a conductivity of 730 μ S cm⁻¹. In this case, the infiltration hazard is resolved, since the WHO and FAO guidelines for water of the 'none' degree of restriction on use is $\kappa > 700 \ \mu\text{S cm}^{-1}$ (10 % deviations are acceptable) for a SAR range of 0-3. XLE and NF90 membranes had a most significant conductivity decrease (Table 6), so it was difficult to solve the infiltration hazard, since SAR value varied from 0.87 to 1.55 and from 1.19 to 1.55, respectively, and for the 'none' degree restriction on use, water conductivity must be $>700 \ \mu\text{S cm}^{-1}$. For these two membranes, none of the ratios was appropriate for irrigation. For this kind of situation, water can be treated to reduce SAR and increase conductivity. It can be accomplished by continually adding soluble calcium (for example, gypsum, calcium chloride) to the irrigation water.²³ Addition of calcium reduces sodium hazard by reducing water SAR and increasing water conductivity, and the treated water can be used for irrigation with a 'none' degree restriction on use.

Conclusion

This study shows that smart management of biologically treated municipal wastewater and RO/NF permeate could be helpful in producing a reliable source of water for agricultural irrigation.

The results demonstrated a stable and suitable quality of the permeate in MBR with regard to the removal of turbidity (99.8 %), TSS (100 %), COD (96 %), and DOC (88 %). Nevertheless, MBR effluent fell into the category of 'slight to moderate' degree of restriction on use, due to low rejection of conductivity (10 %), sodium (2.4 %), and no changes for chloride.

RO (XLE) and NF (NF90 and NF270) membranes additionally decreased all parameters in significant amounts, due to their tight porous structure. The permeate with the highest quality was achieved with XLE membrane, while NF270 membrane showed the lowest. The greatest differences were observed in the conductivity and content of chloride, nitrate, sodium, magnesium, and calcium. RO/ NF permeate alone showed 'severe' degree of restriction on use due to very high removal of conductivity.

The management of both effluents (MBR and RO/NF) consisted in finding the ideal blending ratio to meet specific water reuse guidelines. The optimum blending ratio was found to be 50 % of MBR effluent with 50 % of NF270 membrane permeate. By blending the MBR effluent and NF permeate, a more sustainable treatment could be achieved, i.e., less energy consumption and less NF/RO concentrate generation.

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References

- Masson-Delmotte, V., Portner, H.-O., Skea, J., Zhai, P., Roberts, D., Shukla, P. R., Pirani, A., Moufouma-Oika, W., Pean, C., Pidcock, R., Connors, S., Matthews, J. B. R., Chen, Y., Zhou, X., Gomis, M. I., Lonnoy, E., Maycock, T., Tignor, M., Waterfield, T., Global warming of 1.5 °C – An IPCC Special Report on the impact of global warming of 1.5 °C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global response to the threat of climate change, sustainable development, and efforts to eradicate poverty, International Panel on Climate Change, 2019.
- http://public.wmo.int/en/media/press-release/2015-hottestyear-record (25.04.2016.)
- Bixio, D., Thoeye, C., De Koning, J., Joksimovic, D., Savic, D., Wintgens, T., Melin, T., Wastewater reuse in Europe, Desalination 187 (2006) 89. doi: https://doi.org/10.1016/j.desal.2005.04.070
- Sanz, L. A., Gawlik, B. M., Water Reuse in Europe Relevant guidelines, need for and barriers to innovation, Joint Research Centre (European Commission), Luxembourg, 2014.
- https://eur-lex.europa.eu/legal-content/EN/TXT/ PDF/?uri=CELEX:31991L0271&from=EN (30.05.2019.)
- 6. Falizi, N. J., Hacıfazlıoğlu, M. C., Parlar, İ., Kabay, N., Pek, T. Ö., Yüksel, M., Evaluation of MBR treated industrial wastewater quality before and after desalination by NF and RO processes for agricultural reuse, J. Water Process Eng. **22** (2018) 103.

doi: https://doi.org/10.1016/j.jwpe.2018.01.015

 Gündoğdu, M., Jarma, Y. A., Kabay, N., Pek, T. Ö., Yüksel, M., Integration of MBR with NF/RO processes for industrial wastewater reclamation and water reuse-effect of membrane type on product water quality, J. Water Process Eng. 29 (2019) 100574.

doi: https://doi.org/10.1016/j.jwpe.2018.02.009

 Zanetti, F, De Luca, G., Sacchetti, R., Performance of a full-scale membrane bioreactor system in treating municipal wastewater for reuse purposes, Bioresour. Technol. 101 (2010) 3768.

doi: https://doi.org/10.1016/j.biortech.2009.12.091

 Quist-Jensen, C. A., Macedonio, F., Drioli, E., Membrane technology for water production in agriculture: Desalination and wastewater reuse, Desalination 364 (2015) 17. doi: https://doi.org/10.1016/j.desal.2015.03.001

- Bunani, S., Yörükoğlu, E., Yüksel, Ü., Kabay, N., Yüksel, M., Sert, G., Application of reverse osmosis for reuse of secondary treated urban wastewater in agricultural irrigation, Desalination 364 (2015) 68. doi: https://doi.org/10.1016/j.desal.2014.07.030
- Sert, G., Bunani, S., Kabay, N., Egemen, Ö., Arda, M., Pek, T. Ö., Yüksel, M., Investigation of mini pilot scale MBR-NF and MBR-RO integrated systems performance—Preliminary field tests, J. Water Process Eng. 12 (2016) 72. doi: https://doi.org/10.1016/j.jwpe.2016.06.008
- Sert, G., Bunani, S., Yörükoğlu, E., Kabay, N., Egemen, Ö., Arda, M., Yüksel, M., Performances of some NF and RO membranes for desalination of MBR treated wastewater, J. Water Process Eng. 16 (2017) 193. doi: https://doi.org/10.1016/j.jwpe.2016.11.009
- WHO, WHO guidelines for the safe use of wastewater, excreta and greywater: Volume II Wastewater use in agriculture, World Health Organization, Geneva, 2006.
- Pescod, M. B., Wastewater treatment and use in agriculture – FAO irrigation and drainage paper 47, Food and Agricul-ture Organization of the United Nations, Rome, 1992.
- Tang, C. Y., Kwon, Y.-N., Leckie, J. O., Effect of membrane chemistry and coating layer on physiochemical properties of thin film composite polyamide RO and NF membranes: I. FTIR and XPS characterization of polyamide and coating layer chemistry, Desalination 242 (2009) 149. doi: https://doi.org/10.1016/j.desal.2008.04.003
- Simon, A., McDonald, J. A., Khan, S. J., Price, W. E., Nghiem, L. D., Effects of caustic cleaning on pore size of nanofiltration membranes and their rejection of trace organic chemicals, J. Membr. Sci. 447 (2013) 153. doi: https://doi.org/10.1016/j.memsci.2013.07.013

- 17. Dolar, D., Vuković, A., Ašperger, D., Košutić, K., Effect of water matrices on removal of veterinary pharmaceuticals by nanofiltration and reverse osmosis membranes, J. Environ. Sci. 23 (2011) 1299.
 - doi: https://doi.org/10.1016/S1001-0742(10)60545-1
- Drazevic, E., Bason, S., Kosutic, K., Freger, V., Enhanced partitioning and transport of phenolic micropollutants within polyamide composite membranes, Environ. Sci. Technol. 46 (2012) 3377. doi: https://doi.org/10.1021/es204188j
- Standard Methods for the Examination of Water and Wastewater, American Public Health Association/American Water Works Association/Water Environment Federation, Washington DC, 1995.
- Cartagena, P., El Kaddouri, M., Cases, V., Trapote, A., Prats, D., Reduction of emerging micropollutants, organic matter, nutrients and salinity from real wastewater by combined MBR–NF/RO treatment, Sep. Purif. Technol. 110 (2013) 132.

doi: https://doi.org/10.1016/j.seppur.2013.03.024

- Munz, G., Gualtiero, M., Salvadori, L., Claudia, B., Claudio, L., Process efficiency and microbial monitoring in MBR (membrane bioreactor) and CASP (conventional activated sludge process) treatment of tannery wastewater, Bioresour. Technol. 99 (2008) 8559. doi: https://doi.org/10.1016/j.biortech.2008.04.006
- 22. Košutić, K., Dolar, D., Kunst, B., On experimental parameters characterizing the reverse osmosis and nanofiltration membranes' active layer, J. Membr. Sci. 282 (2006) 109. doi: https://doi.org/10.1016/j.memsci.2006.05.010
- Hopkins, B. G., Horneck, D. A., Stevens, R. G., Ellsworth, J. W., Sullivan, D. M., Managing irrigation water quality – for crop production in the Pacific Northwest, Oregon State University, Oregon, 2007.