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Membrane Bioreactor (MBR) for the Removal of **Emerging Contaminants** from **Municipal Wastewater** and its Viability of Integrating **Advanced Oxidation Processes**

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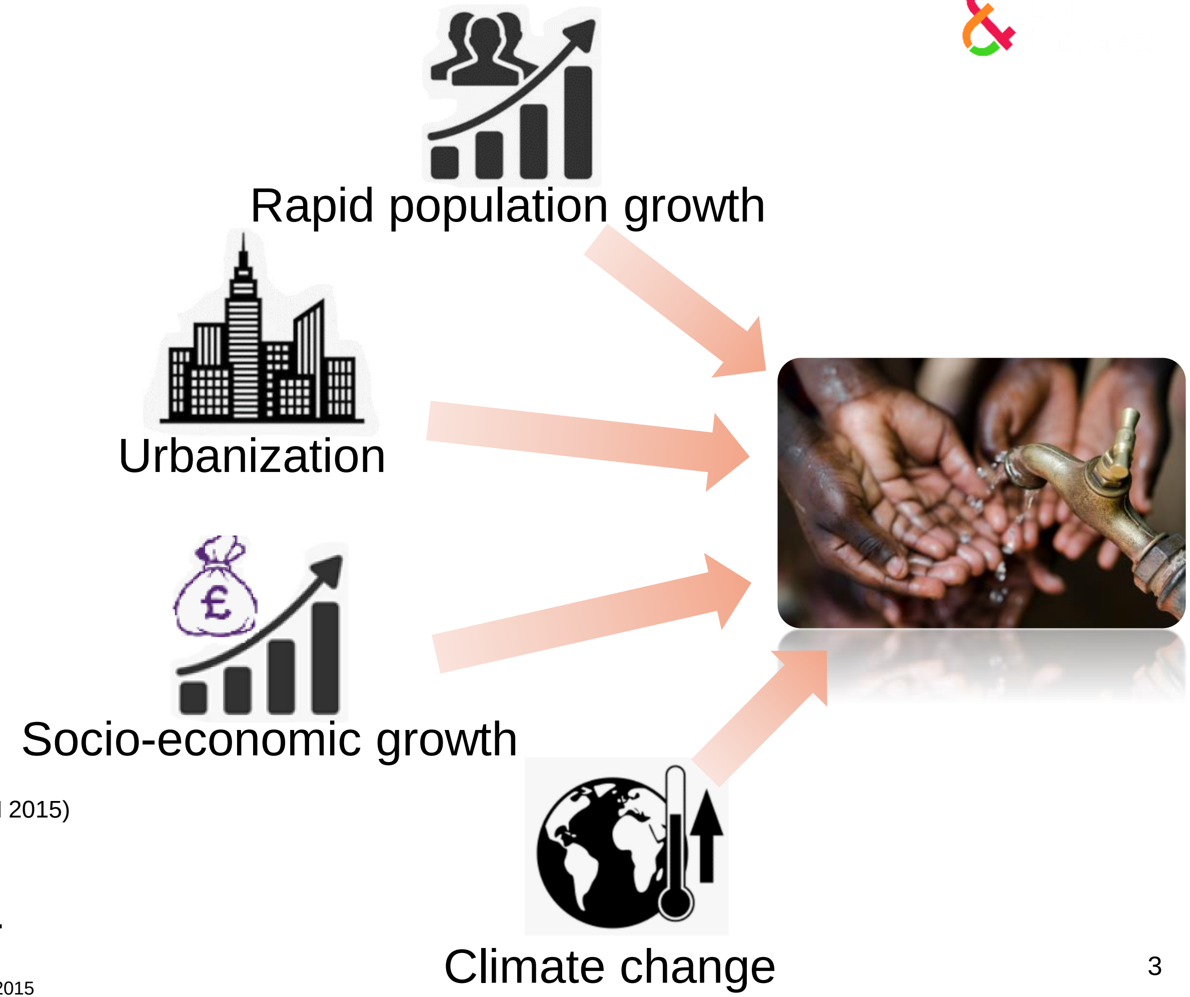
Water scarcity

- Global issue!!
- About 2 billion people worldwide experience high water stress
- About 3 billion people experience severe water crisis at least one month of a year
- More than 5 billion people will face water shortage by 2050[1].



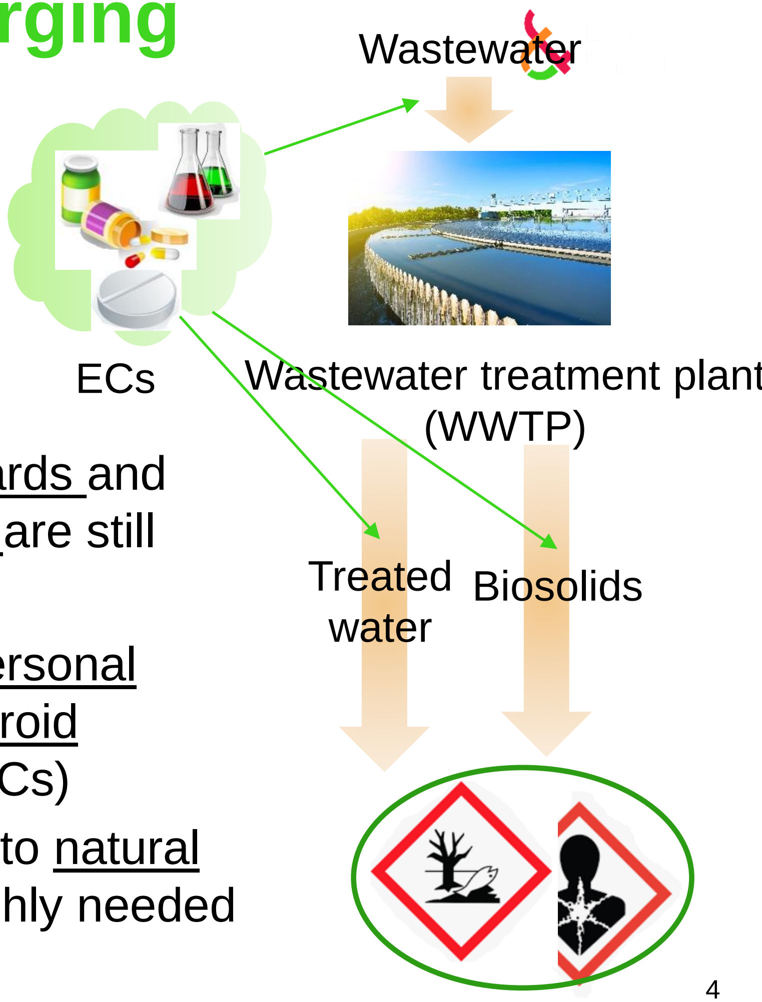
(WRI 2015)

[1] World water development report , United Nations 2019

[2] Aqueduct Projected Water Stress Country Rankings, World Resources Institute 2015

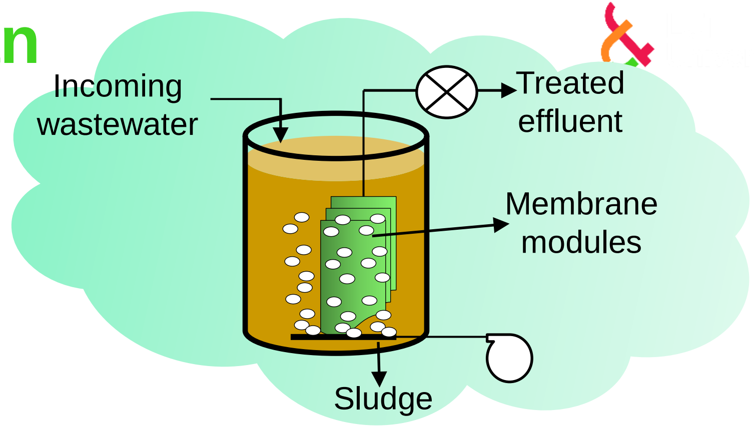
Municipal wastewater and Emerging contaminants (ECs)

- Comprised of wastewaters generated from various standpoint domains
- About 70-130% of municipal freshwater consumption becomes wastewater [1]
- ECs are new chemicals with no regulatory standards and whose effects on environment and human health are still largely unknown
- Pharmaceutically active compounds (PhACs), personal care products (PCPs), stimulants, pesticides, steroid hormones, endocrine disrupting compounds (EDCs)
- To avoid releasing of contaminated wastewaters to natural environment- effective treatment systems are highly needed

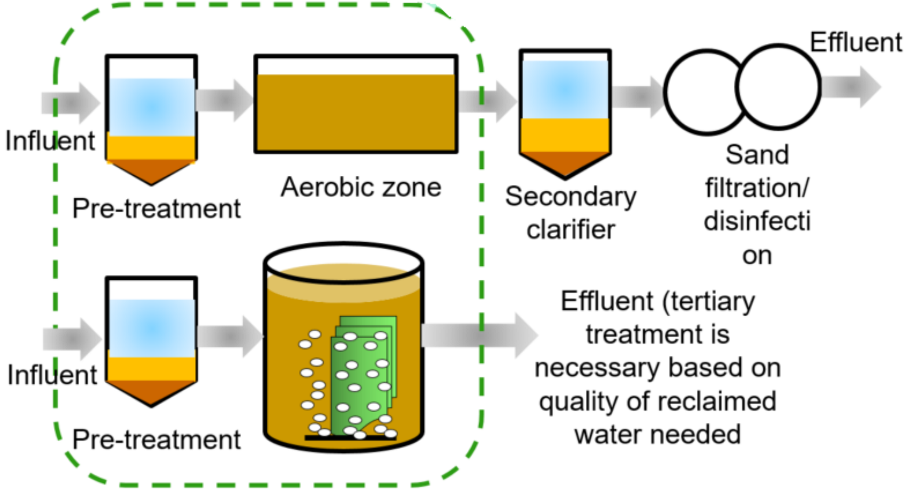


1 Wastewater Treatment Plants: Planning, Design, and Operation, Qasim, 2017

Membrane bioreactor (MBR) as an Advanced wastewater treatment:

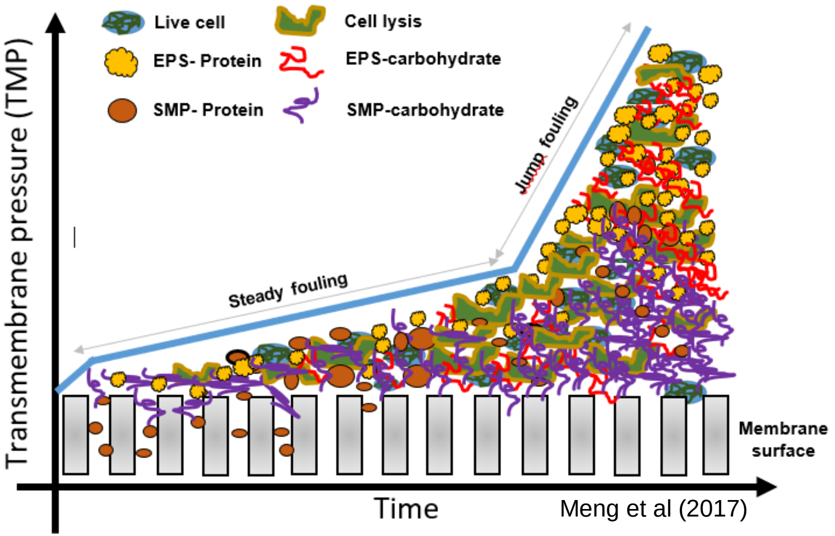


- Combination of biological treatment and membrane filtration system
- High quality effluent, less sludge production, low-demand of tertiary treatment and less space requirement
- The global MBR market was about 2 billion in 2018 and expected to reach 3.8 billion by 2023.
- Dramatic reductions in the membrane cost (1/10) over the last two decades



Membrane fouling: a big challenge in MBR operation

- Inevitable phenomena- major bottleneck!
- Very complex process- possible deposition of organic, inorganic and biological compounds on/in the membrane surface - deteriorates membrane permeability
- Fouling mitigation requires intensive energy and chemicals
- About 50-70% of OPEX is attributed to physical fouling controlling in MBRs



Meng et al (2017)

Meng, F., Zhang, S., Oh, Y., Zhou, Z., Shin, H.-S., Chae, S.-R., 2017. Fouling in membrane bioreactors: An updated review. Water Res. 114, 151–180. https://doi.org/10.1016/j.watres.2017.02.006

Integrated MBR-Advanced oxidation processes (AOPs) concept in wastewater treatment

- For high grade effluent quality
- AOPs involve aqueous phase oxidation of ECs by *in-situ* generated powerful reactive species, e.g., hydroxyl radicals
- MBRs offer solids and turbidity free water- gives high technical flexibility to be integrated with AOPs
- Electrochemical, photochemical, ozonation, Fenton or sonochemical processes.

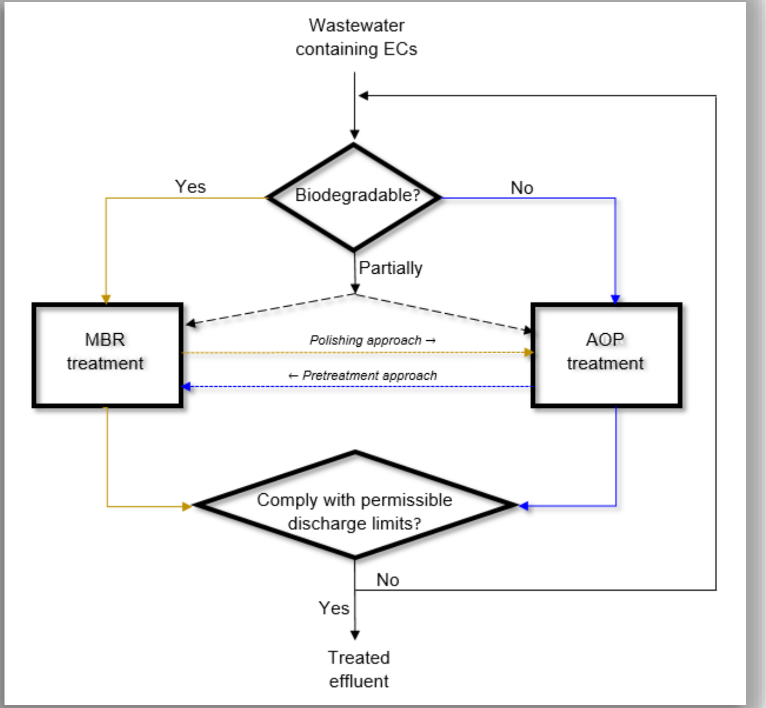


Fig. Decision tree for MBR-AOP integration concept

Results

» **Assessment of MBR performance at different operating conditions**

- A pilot-scale aerobic submerged MBR was operated for more than 200 days, including Nordic cold periods (< 10°C) and varying solid retention times

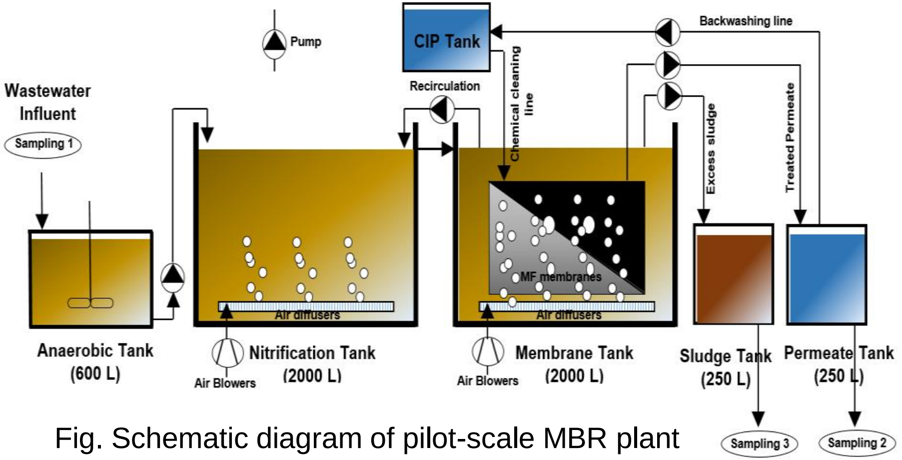


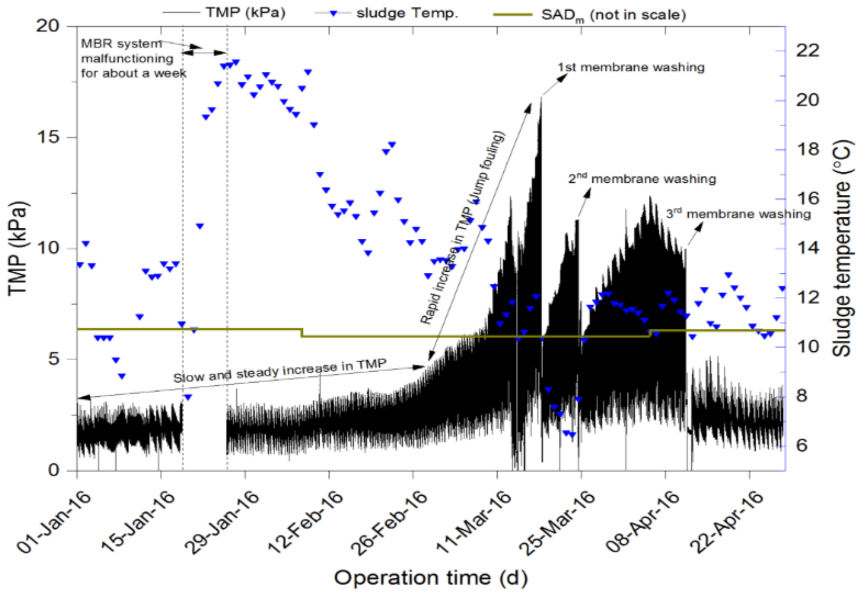
Fig. Schematic diagram of pilot-scale MBR plant

Parameter	Units	Values			
		Paper I	Paper II	Paper III	Paper IV
Hydraulic retention time (HRT)	Hours	35	40-45	40-45	40-45
Sludge retention time (SRT)	Days	25 - 30	60 and 21	55-60	55-60
Avg. Flux (Continuous)	$L\ m^{-2}\ h^{-1}$	7.80	4-6	4-6	4-6
MLSS concentration	$mg\ L^{-1}$	5300 - 9800	8550 and 3748	5000-8000	5000-8000
F/M ratio	$kg\ COD\ (kg\ MLSS.\ d)^{-1}$	0.02 – 0.05	0.027 and 0.09	0.02 – 0.09	0.02 – 0.09
Aeration intensity	$m^{-3}\ m^{-2}\ h^{-1}$	0.4 – 0.6	0.2	0.2 -0.23	0.2 -0.23
Sludge temperature	°C	6.5 - 21	19 ± 2	15-22	15-22
pH	Unitless	6.6 – 7.3	6-7.4	6-8	6-8
Suction cycle	Minutes	9-ON/1-OFF	9-ON/1-OFF	9-ON/1-OFF	9-ON/1-OFF

Results

» Assessment of MBR performance <u>Nordic cold conditions</u>

- Significant membrane <u>permeability reduction</u> (~75%) - at low temperatures <u>(7-10°C)</u>. However;
- Consistently <u>high removal</u> of <u>organics</u>, <u>nutrients</u> and <u>solids</u>.
- <u>High reductions</u> of <u>pathogens</u>, e.g., <u>human enteric viruses</u> (NoV GII > NoV GI> AdV) and <u>faecal indicators</u> (*E*-coli and enterococcus)
- Relatively <u>high heavy metals removal</u>, which meets <u>EU</u> and <u>WHO</u> guidelines

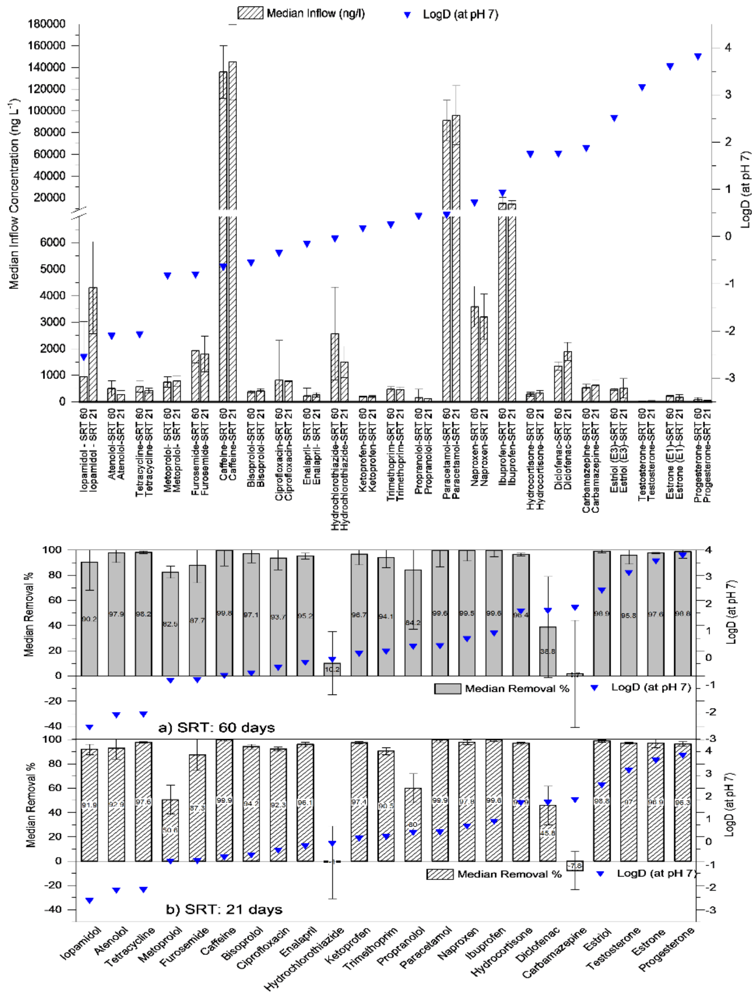


https://doi.org/10.1016/j.scitotenv.2016.11.122

Results

» Assessment of the performance of MBR at different solid retention times

- Removal and fate of 23 diverse ECs were studied at different operational solid retention times (SRTs): 21 days and 60 days.
- Large Variations in removal efficiencies of ECs were observed (non-removal to > 99.9%)- MBR is not the optimal solution!
- Physico-chemical (pKa, logD, log K_d) and molecular properties of ECs and plant operating conditions - greatly influence ECs removal in MBR
- Major mechanism of ECs removal: biotransformation and biosorption



https://doi.org/10.1016/j.scitotenv.2019.02.308

Results

- **Integration assessment- <u>Electrochemical oxidation</u> (ECO) for treating/polishing MBR effluent:**

– Model EC compound: <u>Carbamazepine</u> (CBZ)

– Novel MMO electrode: <u>Ti/Ta_2O_5-SnO_2</u>

– <u>As prepared</u> electrodes were <u>characterized</u> using SEM, AFM, CV etc.

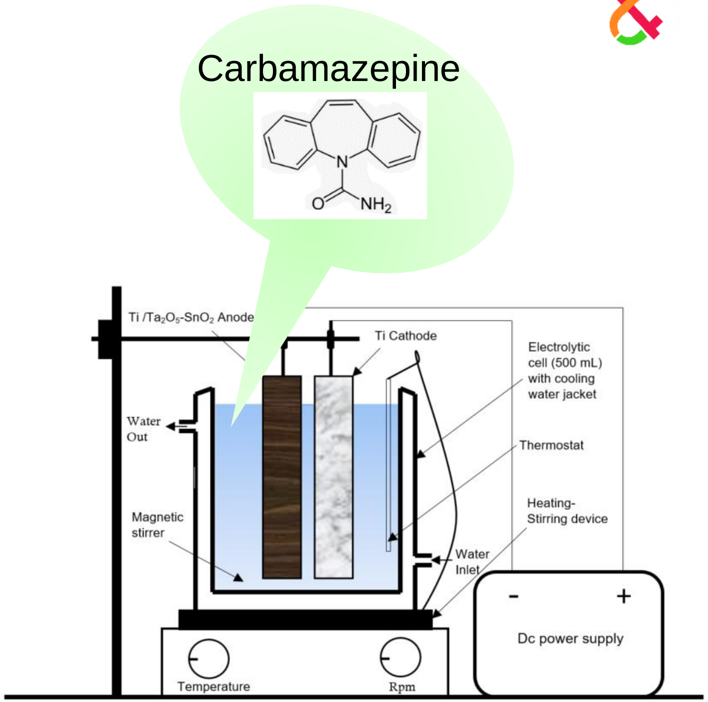


Fig. Experimental set-up for electrochemical oxidation.

Results

» **Integration assessment- Electrochemical oxidation (ECO) for treating/polishing MBR effluent:**

- Operating parameters ,such as current density, initial ECs concentration, pH, temperature - effecting CBZ degradation efficiency, were studied in aqueous solution.
- Optimized condition of current density= 9 mA cm^{-2}; pH= 6; T= 11 ± 1 °C was applied to real MBR effluent

Table : Removal of CBZ in real MBR effluent by using Ti/Ta2O5-SnO2 electrode.

Parameter	Initial concentration in MBR effluent	Final concentration after 8 h of electrolysis	Electrolyte	Removal (%)	EC (kWh m^{-3})
Carbamazepine (µg L^{-1})	10.75 ± 0.35	< 0.07 (LOD)*	No electrolyte	>99.99	109.4
		< 0.07 (LOD)*	0.1 M Na_2SO_4	>99.99	57.2

http://dx.doi.org/10.1016/j.apcatb.2017.09.017

Results

» **Integration assessment- Photochemical oxidation (PCO) for treating/polishing MBR effluent:**

- Model compounds: CBZ and diclofenac (DCF)- not efficiently removed in MBR
- Heterojunction photocatalyst: Ag_2O/TiO_2(P-25) composite is used

- Operating parameters were optimized both in aqueous (DW) and real MBR effluent (RME) matrices

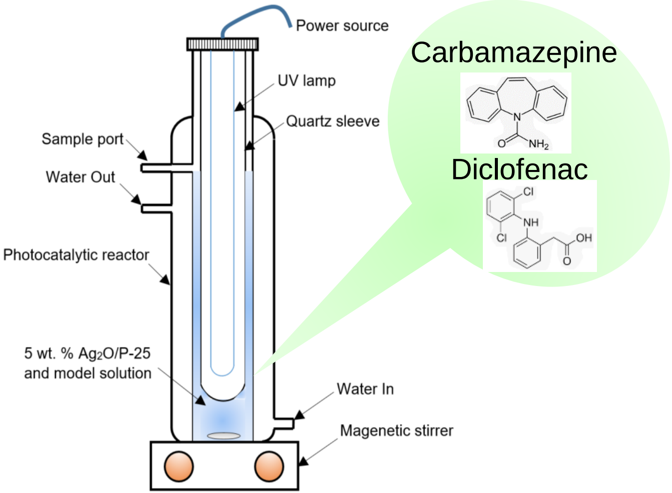


Fig. Experimental set-up for photocatalytic oxidation

Results

›› Integration assessment- Photochemical oxidation (PCO) for treating/polishing MBR effluent:

- About 90% ECs degradation under optimized catalyst dosage in both the matrices
- About 2-fold catalyst dosage was required in RME matrix than in DW matrix to achieve same level of ECs removal
- Mineralization rate of about 55 to 65% for both the compounds in different matrices.

Table : Removal rates (% ±SD) of ECs under the varying catalyst dosages and the extent of mineralization (%) in two different water matrices.

Solution Matrices	Target Pollutants	Catalyst concentration (g L^{-1})	Removal efficiency (%)	Mineralization (%)
ECs in deionized water (DW)	CBZ	0.2	80.40 ± 0.5	
		0.4	89.10 ± 1.5	67.90
		0.6	88.60 ± 1.4	
		0.8	91.70 ± 1.5	
		1.0	89.02 ± 4.7	
	DCF	0.2	87.60 ± 2.2	
		0.4	93.50 ± 0.1	64.80
		0.6	93.60 ± 0.1	
		0.8	93.04 ± 0.1	
		1.0	93.40 ± 0.3	
ECs in real MBR effluent (RME)	CBZ	0.4	76.60 ± 5.2	
		0.6	85.40 ± 6.5	
		0.8	89.74 ± 0.4	60.30
		1.0	90.50 ± 0.4	
		1.2	90.95 ± 2.3	
	DCF	0.4	86.60 ± 0.3	
		0.6	90.70 ± 4.5	55.20
		0.8	88.30 ± 3.4	
		1.0	90.40 ± 0.40	
		1.2	92.10 ± 1.10	

https://doi.org/10.1016/j.seppur.2018.12.069

Conclusion

- Severe membrane fouling (permeability drop) was observed in MBRs operation in Nordic real cold-water conditions- needs urgent solution!
- Diverse removal efficiencies of ECs during MBR treatment due to several influencing factors
- MBR is not the optimal solution for complete remediation of many recalcitrant ECs
- AOPs, such as ECO and PCO showed promising integration alternatives to achieve complete removal of highly recalcitrant ECs- i.e., enhanced treatment efficiency

Future research prospects

- Developing novel integrated processes and materials, such as microalgae cultivation, microbial electrolysis cells, engineered nanoparticle coated membranes etc. – for reduced energy consumption and recovery of value-added products (Nutrients, biofuels, electricity etc.)

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