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Can strict water reuse standards be the drive for the wider implementation of MBR technology?

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ABSTRACT

Membrane bioreactors (MBRs) are currently considered a mature technology for municipal wastewater treatment with many full scale applications worldwide. The drive for the wider implementation of MBR technology can be the increasingly stringent legislation concerning the reuse or discharge of the treated effluent. In this work it is shown that the strict limits recently adopted by Greece concerning reclaimed water reuse can be consistently met when MBR technology and suitable disinfection are applied. MBR permeate met the Greek limit of 5 FC/100 mL for 80% of samples and 50 FC/100 mL for 95% of samples required for unrestricted irrigation when a chlorination dosage of 10 mg min/L was applied and an ultraviolet (UV) radiation dosage of 10 mW s/cm^2 . On the contrary, secondary effluent from the activated sludge process could not satisfy the given limits even at a chlorination dosage of 600 mg min/L and a UV radiation dosage of 120 mW s/cm^2 to satisfy the same limits. Therefore, the dosage of UV radiation and chlorine required to meet the microbiological limits for unrestricted irrigation were much lower for MBR permeate than for conventional tertiary effluents.

Keywords: Membrane bioreactors; Water reuse; Chlorination; UV radiation; Disinfection

1. Introduction

During the early stages of its development the adoption of membrane bioreactor (MBR) technology at a worldwide level took place at a fast rate. As pointed out by Lesjean et al. [1], this led to certain misconceptions and/or overstatements concerning this technology related to the treated effluent quality, the energy requirements and economics, and its potential dominance over the conventional activated sludge (CAS) process. MBRs are now considered a mature technology for municipal wastewater treatment since many full scale applications are operating at a worldwide level. By the end of 2008, in Europe more than 800 full scale applications of MBRs had been documented, with 37 MBR plants having a design capacity higher than 5,000 m³/d. The increasing application of full scale MBRs in recent years for municipal wastewater treatment may lead to the impression that MBRs will

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extensively replace the CAS processes. However, this is unlikely to happen in the medium to long term range. In 2009, only 0.5% of the population in Europe was serviced by MBRs for municipal wastewater treatment [2]. Despite the increase in competitiveness of MBR technology resulting from the decrease in membrane cost, the increase in the life span of the membranes, the increase in the permeate flux, the decrease of energy requirements, and the proven process reliability, MBRs still remain an energy intensive process when compared to the CAS processes [3-7]. The bottlenecks are related to the increased energy requirements (usually around 0.2 kW h/m^3 higher than CAS) the requirements for membrane cleaning and for module replacement after some years [1]. Given the aforementioned bottlenecks the MBRs are expected to continue to grow, but will not outcompete the CAS processes. Furthermore, in the future MBR technology will have to compete with low carbon footprint technologies that are currently growing such as the upflow anaerobic sludge blanket and the completely autotrophic nitrogen removal process.

Nevertheless, the drive for the wider penetration of MBR technology can be the gradual enforcement of strict legislation concerning the reuse and/or discharge of the treated effluent. In Venice, the strict limits imposed on nitrogen and phosphorus forms of the discharged effluents and on targeted micropollutants into the Lagoon of Venice, favored the implementation of MBR technology for domestic wastewater treatment, with 43 decentralized MBRs operating in the historical centre and with the largest MBR plant in the world for petrochemical effluents operating in Porto-Marghera [8,9]. In Greece, the penetration of MBR technology has been sluggish with few applications of small capacity for industrial and municipal wastewatreatment. However, the recently adopted ter reclaimed water reuse standards can favor the implementation of MBR technology in Greece. The limits are summarized in Table 1.

Table 1

Parameter	Restricted irrigation, industrial use (cooling water for single use) and groundwater recharge by filtration through suitable soil layer for aquifers used for non-potable applications	Unrestricted irrigation and industrial use ^a	Urban use, peri-urban green and groundwater recharge with drilling
Escherichia coli (EC/100 mL)	≤200 median value	≤5 for 80% of samples ≤50 for 95% of samples	_
Total coliforms (TC/100 mL)		_	≤2 for 80% of samples ≤20 for 95% of samples
$BOD_5 (mg/L)$	Specified by the common ministerial decision KYA 5673/400/1997	≤ 10 for 80% of samples	≤ 10 for 80% of samples
Turbidity (NTU)		≤2 median value	≤2 median value
TSS (mg/L)	Specified by the common ministerial decision KYA 5673/400/1997	≤ 10 for 80% of samples	\leq 2 for 80% of samples
Minimum treatment requirements	Secondary biological treatment ^b and disinfection ^c	Secondary biological treatment ^d , tertiary treatment ^e and disinfection ^b	Secondary biological treatment ^f , advanced treatment ^g and disinfection ^b

^aExcept cooling water for single use.

^bSuggested methods of secondary biological treatment include the activated sludge process, biological filters and rotating biological discs or other systems that produce equivalent treated effluent quality. Nitrogen in the treated effluent should have a concentration lower than 45 mg N/L expect in cases where the treated effluents are stored for long periods in reservoirs, irrigation in nitrite vulnerable zones is practiced and groundwater recharge takes place. In these cases nitrogen should be lower than 15 mg N/L.

^cDisinfection options include chlorination, UV radiation and ozonation or other disinfection system that can guarantee the required limits. ^dAs specified in b. In the case of irrigation in nitrate vulnerable zones nitrogen removal through nitrification/denitrification is required as the treated effluent should have a total nitrogen concentration lower than 15 mg N/L and ammonium concentration lower than 2 mg N/L. ^eTypical tertiary treatment includes coagulant addition and filtration through conventional sand filters.

 $^{^{\}rm f}$ As specified in b with the extra requirement of biological nitrogen removal through nitrification/denitrification as the treated effluent should have a total nitrogen concentration lower than 15 mg N/L and ammonium concentration lower than 2 mg N/L.

^gAdvanced treatment means ultrafiltration membrane system or other equivalent system that can guarantee the required limits. In the case MBRs are used, it is possible to integrate biological and advanced treatment in one stage.

This work evaluated whether the Greek water reuse limits can be the drive for the greater penetration of MBR technology in the Greek market.

2. Materials and methods

2.1. MBR system and sample collection

The MBR pilot system was installed at the premises of the Sanitary Engineering Research and Development Centre owned by the Athens Water Supply and Sewerage Company (EYDAP S.A.), situated at Metamorphosi Attiki. Primary treated municipal effluent was fed to a 2 m³ tank and then to a pilot scale MBR (working volume of 210 L) where the membrane ultrafiltration module was immersed. The hollow fiber membrane module (ZeeWeed 10) was supplied by GE Water and Process Technologies. It was made of polyvinylidene fluoride and had a nominal pore size of 0.04 µm and an absolute pore size of 0.1 µm. Permeate was filtered directly from the aeration reactor. Coarse bubble aeration was supplied at a constant rate of 4.0 m³/h to minimize membrane fouling, while fine bubble aeration was supplied through air diffusers to maintain the dissolved oxygen level higher than 2 mg/L. The filtration pattern took place at 10 min cycles during which 9.5 min of filtration were followed by 0.5 min of backwash. Grab samples from the primary effluent and the MBR permeate were collected during a 4 month period and were analyzed for their physicochemical and microbiological characteristics. During the sampling period, the solids retention time was 10 d and the hydraulic retention time was 11.2 h.

2.2. UV radiation and chlorination

Ultraviolet (UV) radiation was accomplished using a low pressure lamp. The lamp emitted radiation through a tube which was able to move vertically, thus determining, based on distance, the intensity of radiation that was emitted to the sample. A diaphragm located at the base of the tube prevented the radiation. Once this diaphragm was removed, radiation was emitted to the sample located below, inside, a 50 mL dish. Before the initiation of the experiment the lamp was heated for approximately 15 min. Its intensity was measured by a radiometer (IL 1700) using a properly adjusted UV sensor (SED 240). The intensity was determined using formula [11]:

$$I_{\text{average}} = I_{\text{applied}} (1 - e^{-aL}) / aL \tag{1}$$

where I_{average} is the average radiation intensity (mW/cm²); I_{applied} is the applied radiation intensity at the surface of the sample (mW/cm²); *a* is the absorption unit per cm at 254 nm (cm⁻¹); *L* is the height of the sample (cm).

To measure correctly the applied intensity, the UV filter of the radiometer's sensor should be at the same level as the surface of the sample. First the absorption was measured at 254 nm based on the following equation:

$$T = 100 \times 10^{-a} \Rightarrow a = \log \left(\frac{100}{T} \right) \tag{2}$$

where T is the UV transmittance of each sample at 254 nm.

The time t_{exposed} that the samples were exposed to specific UV radiation dosage was found:

$$t_{\text{exposed}} = \frac{D}{I_{\text{average}}} \Rightarrow t = \frac{DaL}{I_{\text{applied}}(1 - e^{-aL})}$$
 (3)

where *D* is UV radiation dosage (mW s/cm²).

In all the UV radiation experiments, 50 mL of wastewater sample was added to the dish. The dish was then placed on the magnetic stirring plate in order to be under continuous agitation throughout the experiment. The height that this sample occupied was L = 2.2 cm. Each time, the applied intensity was measured and the average intensity was determined using Eq. (1). The applied UV radiation dosages were 3, 5, 10, 20, 30, and 40 mW s/cm². Eq. (3) was used to determine the time of sample exposure for the given dosage. During the experiments, the time of exposure was recorded using a stop watch.

In the chlorination experiments a commercial solution of NaOCl was used having a residual chlorine concentration of 4.8% w/w. During each experiment the initial chlorine concentration was always measured for confirmation purposes. The dosage of residual chlorine was determined from the following equation:

 $CD = C_{residual} \times t_{contact}$

where CD is chlorine dosage (mg/L min); C_{residual} is residual chlorine concentration (mg/L); t_{contact} is contact time (min).

The addition of chlorine was conducted in 300 mL of water samples (i.e. MBR permeate) and the dosages tested were 5, 10, 25, 50, 75, and 100 mg/L min. The contact time was kept constant at 30 min during which the water sample was kept under agitation. Once the experiment was completed the residual chlorine was measured in each sample. Then, the samples were

dechlorinated using sufficient amount of sodium metabisulfite so that the microbiological analysis could follow.

The samples obtained after the UV radiation and the chlorination processes, were analyzed to determine their content in fecal coliforms (FC) and total coliforms (TC), and was compared to the FC and TC content in MBR permeate and in the primary effluent.

2.3. Analytical methods

Total suspended solids (TSS), volatile suspended solids (VSS), biochemical oxygen demand (BOD₅), turbidity, TC, and FC were determined according to standard methods [12]. As Escherichia coli (E. coli) are part of the group of FC, the latter were measured instead of E. coli. Chemical oxygen demand (COD), total nitrogen (TN) and ammonium nitrogen (NH₄-N) were determined using the Spectroquant kits and the NOVA60 photometer of Merck. Turbidity was determined using the turbidity meter Turb 550 IR model. Residual chlorine was determined using the spectrophotometer Hach DR/2000 and suitable reagents.

Table 2Characteristics of the influent and effluent streams

Parameter	Influent to MBR	MBR permeate
TSS (mg/L)	276 ± 125	< 0.5
VSS (mg/L)	218 ± 78	-
Turbidity (NTU)	196 ± 27	0.17 ± 0.03
COD (mg/L)	634 ± 40	35 ± 7
$BOD_5 (mg/L)$	287 ± 19	2.8 ± 2.2
TN (mg/L)	61.2 ± 12.5	41.5 ± 8.0
$NH_4-N(mg/L)$	45.7 ± 7.7	0.9 ± 1.8
$NO_3-N (mg/L)$	<0.5	33.2 ± 6.6
TC (CFU/100 mL)	60×10^{6}	700
FC (CFU/100 mL)	19×10^5	10

3. Results and discussion

Table 2 shows the characteristics of primary effluent which was fed to the MBR and the permeate. Strictly speaking the UF membrane of MBR should reject completely the bacteria and MBR permeate should be free from FC and TC. According to the manufacturer's specifications FC should be less than 2.2 CFU/100 mL after disinfection [13]. In our case, infection in the outlet tube of the MBR permeate resulted in increased levels of TC and FC. Fig. 1 shows the cumulative distribution of TC and FC in the treated effluent. As seen in Fig. 1, the MBR permeate was characterized by significant levels of TC and FC. These values are higher than the required Greek limits for unrestricted irrigation, urban use, peri-urban green and groundwater recharge. The MBR permeate was then subjected to UV radiation and chlorination to determine the dosage required to reach the microbiological reuse limits. These disinfection methods were tested since they are used in Greek wastewater treatment plants.

Tables 3(a) and 3(b) show the TC and FC for different UV radiation dosages. The dosage of 30 mW s/cm^2 resulted in attaining the Greek limits of 2 TC/100 mL for 80% of the samples and 20 TC/100 mL for 95% of the samples required for urban use, peri-urban green and groundwater recharge. A lower dosage of 10 mW s/cm² was sufficient to obtain ≤ 5 FC/100 mL for 80% of samples and ≤50 FC/100 mL for 95% of samples required for unrestricted irrigation. Previous work has shown that the required dosage of tertiary effluent in order to meet the aforementioned FC limits for unrestricted irrigation is much higher (45 mW s/cm^2) [14]. For secondary effluents from a CAS process having TSS = 35 mg/L no dosage in the range of 20–150 mW s/cm² could satisfy the required limits of FC. Specifically, the application of 150 mW s/cm² resulted in approximately ≤300 CFU/100 mL of FC for 80% of the samples and ≤900 CFU/100 mL of FC for 95% of the samples [10,14].

Tables 4(a) and 4(b) show the TC and FC for different chlorine dosage. The dosage of 50 mg min/L



Fig. 1. Cumulative distribution of TC and FC in MBR permeate.

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Percentile	UV dosage (mW s/cm ²)	0	3	5	10	20	30	40
50	TC (CFU/100 mL)	475	76	48	17	3	0	0
80	TC (CFU/100 mL)	820	140	92	44	6	0	0
95	TC (CFU/100 mL)	2,050	480	360	200	84	16	4

Table 3(a) TC in MBR permeate by applying different UV radiation dosages

Table 3(b)

FC in MBR permeate by applying different UV radiation dosages

Percentile	UV dosage (mW s/cm ²)	0	3	5	10	20	30	40
50	FC (CFU/100 mL)	8	4	1	0	0	0	0
80	FC (CFU/100 mL)	22	17	9	2	0	0	0
95	FC (CFU/100 mL)	165	118	68	33	11	3	0

Table 4(a) TC in MBR permeate by applying different chlorination dosages

Percentile	Cl ₂ dosage (mg min/L)	0	5	10	25	50	75	100
50	TC (CFU/100 mL)	530	190	12	0	0	0	0
80	TC (CFU/100 mL)	1,410	605	180	23	1	0	0
95	TC (CFU/100 mL)	2,900	800	420	45	6	0	0

Table 4(b) FC in MBR permeate by applying different chlorination dosages

Percentile	Cl_2 dosage (mg min/L)	0	5	10	25	50	75	100
50	FC (CFU/100 mL)	32	4	0	0	0	0	0
80	FC (CFU/100 mL)	70	16	2	0	0	0	0
95	FC (CFU/100 mL)	390	85	48	7	0	0	0

resulted in attaining the required limits of 2 TC/100 mL for 80% of the samples and 20 TC/100 mL for 95% of the samples for urban use, peri-urban green and groundwater recharge. A lower dosage of 10 mg min/L was sufficient to meet the values of \leq 5 FC/100 mL for 80% of samples and \leq 50 FC/100 mL for 95% of samples required for unrestricted irrigation. For secondary effluents from the CAS process a chlorine dosage of up to 600 mg min/L could not fulfill the required limits. For tertiary effluents the required dosage was 100 mg min/L [14].

To conclude, MBR permeate required much lower dosages of UV radiation and chlorination to achieve the required limits compared to tertiary effluents (treated by filters). The secondary effluents could not meet the required limits even when very high UV radiation and chlorination dosages were applied. The recent implementation of the Greek water reuse standards is thus expected to favor the future implementation of MBR plants. In the Greek reclaimed water reuse standards, it is explicitly specified that in the cases where advanced treatment is required (i.e. for urban use, peri-urban green and groundwater recharge), if MBR is implemented no further treatment apart from disinfection is required. On the contrary, if conventional biological treatment is adopted it has to be coupled with further treatment such as membrane filtration. This restriction is expected to favor the development of MBRs in Greece.

4. Conclusion

The drive for the greater penetration of MBR technology can be the increasingly stricter standards that are specified concerning reclaimed water reuse. Experimental results show that the dosage of UV radiation and chlorine that is required for the MBR permeate to meet the Greek microbiological limits for unrestricted irrigation is much lower than the dosage required for tertiary effluents (i.e. treated by filters). For secondary effluents from the CAS process, even the highest dosage could not attain the required reuse limits.

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