Comparison of PPCPs removal on a parallel-operated MBR and AS system and evaluation of effluent post-treatment on vertical flow reed beds

R. Reif, A. Besancon, K. Le Corre, B. Jefferson, J. M. Lema and F. Omil

ABSTRACT

The presence in the aquatic environment of xenobiotics such as Pharmaceutical and Personal Care Products (PPCPs) has emerged as an issue of concern. Upgrading sewage treatment quality with modern technologies such as Membrane Bioreactors (MBRs) and/or implementing a further post-treatment might mitigate the release of xenobiotics into surface waters. The performance of two processes treating municipal sewage, a MBR and an Activated Sludge (AS) unit, have been compared in terms of PPCPs removal. Moreover, their effluents were treated using vertical flow reed beds. Both systems were operated under similar conditions, more specifically Hydraulic Retention Time (HRT), maintained at 8 hours, and Sludge Retention Time (SRT) set at 6 and 20 days. Pharmaceuticals belong to therapeutic groups such as antiepileptics (carbamazepine) and analgesics (ibuprofen, naproxen, diclofenac), whereas the personal care products are musk fragrances (galaxolide and tonalide). Xenobiotics removals achieved in the MBR showed better results, particularly for the acidic drugs ibuprofen (87% vs. 50%) and naproxen (56% vs. 6%) operating at low SRT. After filtration through vertical flow reed-beds, PPCPs content in effluents was decreased, below 1 ppb in most cases, improving the effluent quality and confirming reed-beds as an interesting low cost alternative in order to attenuate xenobiotics contamination.

Key words | activated sludge, MBR, micropollutants, post-treatment, wetlands

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INTRODUCTION

Trace amounts of micropollutants such as Pharmaceutical and Personal Care Products (PPCPs) are found ubiquitously in the aquatic environment (Ternes 1998). Although these substances are present at low concentrations, their occurrence has raised potential toxicological concerns, due to their unique characteristics and the paucity of information available regarding their potential toxicological effects, more particularly when they are present as components of complex mixtures (Schwarzenbach *et al*. 2006). To date, the number of scientific papers published assessing the potential long-term effect on aquatic environment is growing. Well-known examples are the development of pathogenic microorganisms resistant to antibiotics, suspected to be linked with their widespread use (Dantas *et al*. 2006) or the dramatic decrease of vulture species population in India, caused by traces of the anti-inflammatory drug diclofenac which is present on carrion (Oaks *et al*. 2004).

PPCPs are released into the environment through different pathways. The most relevant is initiated after excretion via urine or faeces of unmetabolized fractions of pharmaceuticals or the rinsing off of cosmetics during shower, eventually reaching Sewage Treatment Plants (STPs). However, STPs have not been specifically designed to remove micropollutants. Along the different treatment steps, PPCPs concentration might be reduced following different mechanisms: volatilisation, sorption to either suspended solids or biological sludge and chemical or biological transformation (Suárez *et al*. 2008). The dominant mechanism (generally sorption or biodegradation) and the extent of the micropollutants removal will depend on their specific physico-chemical properties and biodegradability, and on

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the design and operational parameters of the STP considered. However, STPs are not specifically designed to achieve a complete removal of xenobiotic substances, and a significant depletion of the micropollutants content from the final effluent is not achieved in many cases.

Traditionally, activated sludge (AS) processes have been the most representative technology at full-scale STPs, but such systems require a final settling step in order to separate the biological sludge from the effluent. In contrast, Membrane Biological Reactors (MBR) combine the biological process with a membrane filtration step within one process unit, overcoming clarification and producing a high quality effluent. Key operational parameters in STPs considered crucial for improving micropollutants removal are the Hydraulic Retention Time (HRT) and Sludge Retention Time (SRT). Some studies dealing with the influence of SRT conclude that values longer than 15–20 days, easily achievable in well-designed AS systems, are high enough to develop a sustainable population of nitrifiers, considered to play a relevant role on the biodegradation of PPCPs (Clara *et al.* 2005). Up to now, some authors have compared the fate of micropollutants in AS and MBR systems, reporting better results for MBRs in many cases (Weiss & Reemtsma 2008). Nevertheless, both systems of reactors were operated with different parameters making direct comparison difficult to interpret. Potential differences include: the influence of the membrane separation process directly (membrane composition, configuration and pore size), the formation of a cake layer over the membrane surface or the different structural conformation of biomass observed in MBRs (Massé *et al.* 2006).

A further post-treatment of the effluent might attenuate the release of xenobiotics into the aquatic environment. Modern STPs are equipped with technologies for a polishing step such as ozonation or activated carbon sorption. These technologies are also being tested in terms of micropollutants removal, proving to be effective in many cases (Huber *et al.* 2005). However, their capital and operational costs are high. Consequently, available low-cost alternatives like phytoremediation are worth consideration. Wetlands have shown a potential for removing persistent substances. For example, plants like *phragmites australis* contain the appropriate enzymes able to detoxify organic pollutants (Schwitzguébel 2004). However, the potential of constructed wetlands has only been partially studied (Matamoros *et al.* 2008). In this work, the effluent/permeate produced in the pilot-scale AS/MBR has been treated in two separated Vertical Flow Reed Beds (VFRB). Information obtained in this study would be useful to evaluate the potential of green emerging technologies based on phytoremediation from an aspect that has not been sufficiently studied.

METHODS

Pilot-scale system: MBR, AS and VFRB

Three vertical flow reed beds were designed and developed at Cranfield University (UK) based on the model developed by Grant & Griggs 2001. The reed beds are part of a demonstration treatment train which includes a membrane bioreactor (MBR), and a conventional activated sludge process (AS) (Figure 1). The sewage treatment work in Cranfield provides the primary effluent for pilot plant. Both MBR and AS systems are operated at 6 days sludge retention time and 8 hours hydraulic retention time. The

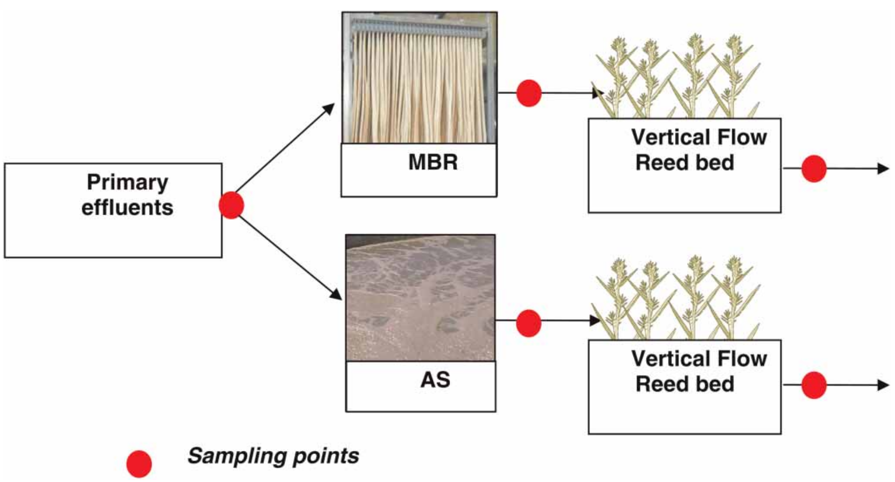

Figure 1 | Schematic of the pilot unit and main characteristics of the primary effluent.

vertical flow reed beds are fed 8 times per day for 10 minutes at 125 mL min^{-1} for the ones following the AS (VFRB AS) and the MBR (VFRB MBR) systems.

The MBR (Figure 2) is composed of an aerated cylindrical tank of 35 L capacity and is set up in submerged membrane configuration. The membrane module has a working length of 50 cm and is made of 21 tubes at pore size of 0.08 μm and lumen diameter of 6 mm. As a result the membrane surface area is of 0.2 m^2 as required to ensure the working flux of 21.9 L·(h $m^2)^{-1}$ LMH.

The AS is composed of an aerated rectangular tank of 30 L capacity, a pre-anoxic zone of 2 L capacity and a clarifier of 7 L capacity (Figure 2). The AS plant was operated with a sludge recirculation rate of 4 times the feed flow rate (Table 1).

Analytical methods

Routine analyses were carried out to assess the performance of the system with respect to aerobic carbonaceous and nitrogen removal. Chemical Oxygen Demand (COD), ammonia (NH_4^+) and nitrate (NO_3^-) were determined using a Spectroquant Cell Test and measured on a Nova 60 model spectrophotometer. Total suspended solids (TSS), Mixed Liquor Suspended Solids (MLSS), Mixed Liquor Volatile Suspended Solids (MLVSS) and pH were determined according to APHA/AWWA/WEF (1999). Particle size density distribution was analysed using a Malvern Instruments Mastersizer 2000 laser diffractometer with a shear stress of 30 rpm, refractive index of 1.33 and dilution of 1:5 for MBR and 1:10 for AS.

Table 1 | Main operational parameters and biomass characteristics of the studied pilot-plant

			Flow rate (L/d)	
Sampling	**HRT (h)**	**SRT (d)**	**AS**	**MBR**
1st	8	6	90	105
2nd	8	20	90	105
3rd	8	20	90	105

Selected PPCPs are the anti-inflammatory drugs ibuprofen (IBP), naproxen (NPX) and diclofenac (DCF), the musk fragrances galaxolide (GLX) tonalide (TON) and celestolide (CLT), the tranquilizer diazepam (DZP) and the antiepileptic carbamazepine (CBZ). Five discrete samples, taken between 8:00 and 20:00 on each day of the three sampling campaigns (12th December, 27th March and 26th July). Sample points were the sewage, MBR permeate, effluent AS and the effluents from both vertical flow reed beds. After collection, liquid samples were filtered through glass-fibre and nitrate cellulose membrane filters (0.45 μm). PPCPs content was determined after solid-phase extraction of 100 mL of sewage or 250 mL of effluent, according to a previously explained method (Reif et al. 2008). During the

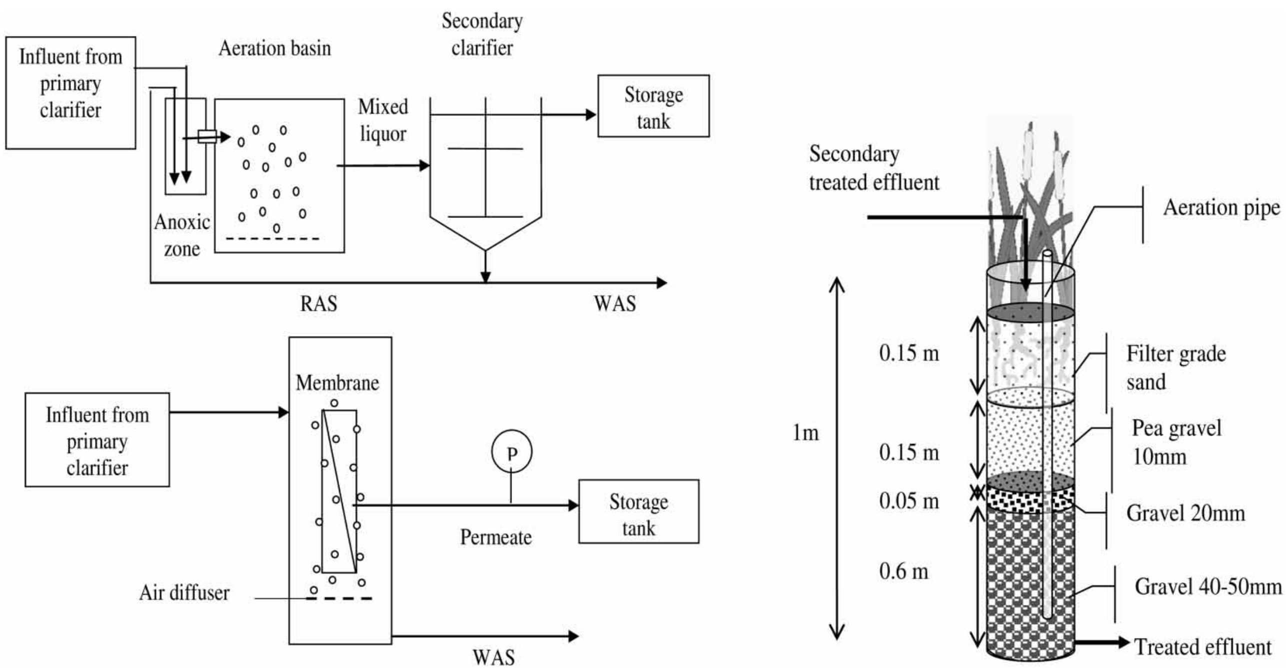

Figure 2 | Diagram of MBR and AS pilot plants, and vertical flow reed bed design.

second sampling campaign, grab samples of primary and biological sludge were taken from both bioreactors in order to determine the amount of PPCPs sorbed onto the solid fraction perform using an ultrasonic solvent extraction method (Ternes *et al.* 2005).

RESULTS AND DISCUSSION

In the present study, the performance of a pilot-scale AS and MBR system treating municipal sewage is studied, but keeping equal operational parameters. Two different SRT have been considered (6 and 20 days) maintaining a constant HRT of 8 hours, typical of full-scale STPs.

Conventional parameters analyses at the different sampling points are summarized in Table 2. AS and MBR systems showed good performance in terms of COD removal. Improved results were always observed in the MBR most probably due to the effect of membrane filtration. Ammonia removal was higher, above 95% even at low SRT (1st sampling). Membrane process separated efficiently the suspended solids. With the exception of the first sampling campaign, the clarifier from the AS process achieved good suspended solids removal.

Table 2 | Conventional parameters measures (mg L^{-1}) in the sampling points of the MBR and AS pilot plants

	Conventional parameters (mg L^{-1})					
	pH	TSS	COD	NH4+	NO3−	DO
1st sampling						
Sewage	7.79	89	306	26.30	0.00	n.a.
AS effluent	7.89	45	97	0.17	1.60	6.03
MBR Permeate	7.29	0	32	1.05	16.10	8.34
VFRB AS	7.66	3	15	0.32	23.60	n.a.
VFRB MBR	7.80	15	44	1.29	14.90	n.a.
2nd sampling						
Sewage	7.92	102	260	22.60	0.00	n.a.
AS effluent	7.82	23	63	0.35	19.30	5.22
MBR Permeate	7.06	0	34	0.06	22.90	9.11
VFRB AS	7.31	0	20	0.36	25.00	n.a.
VFRB MBR	7.38	2	60	2.62	17.80	n.a.
3rd sampling						
Sewage	7.89	386	240	28.50	0.60	n.a.
AS effluent	7.82	22	55	0.94	33.60	4.64
MBR Permeate	7.27	0	27	0.03	14.20	7.80
VFRB AS	7.31	4	24	0.08	24.00	n.a.
VFRB MBR	7.38	10	29	0.05	23.30	n.a.

Note: n.a: not available.

Comparison between membrane bioreactor and conventional activated sludge systems

DZP, CLT and CBZ were below the detection limit. The anti-inflammatory DCF was also not detected during the first sampling campaign, and consequently it was spiked during the next months. Measured concentrations for musk fragrances were low, and TON was not detected during the second sampling campaign. Since the study of these substances is interesting due to their lipophillic character, GLX was also spiked in order to slightly increase its concentration. For the third sampling campaign, the antiepileptic CBZ was also spiked, but only in both VFRBs since it is well-known that this substance is not removed during secondary treatment.

Figure 3 shows calculated removal efficiencies measured along the three sampling campaigns in both bioreactors. First sampling campaign is characterised by a considerably low SRT (6 days). Removal rates increased gradually when this parameter was set at 20 days and acclimation phenomena was achieved.

Anti-inflammatory drugs: IBP, NPX and DCF

These hydrophillic substances are characterized by a high polarity and solubility in water. Analysis performed in the solid phase confirmed their low tendency to be sorpted onto suspended solids, since their concentration remained below detection limit. Therefore, their main removal mechanism will mainly depend on their biodegradability. In general, MBR biomass was more efficient in removing polar micropollutants under the set conditions. When SRT was increased, the differences among AS and MBR were almost negligible. IBP showed the highest removal rates, up to 95%, and NPX removal was particularly influenced by SRT in the AS system. There are no data available for DCF during the first sampling campaign. The removal rates for this substance were slightly better in AS (70–80%), but decreased during the third sampling, observing the opposite tendency in the MBR.

Musk fragrances: GLX and TON

Musk fragrances are lipophillic substances with high solid-water distribution coefficients reported (Suárez *et al.* 2008). Therefore, their removal is mainly based on a sorption

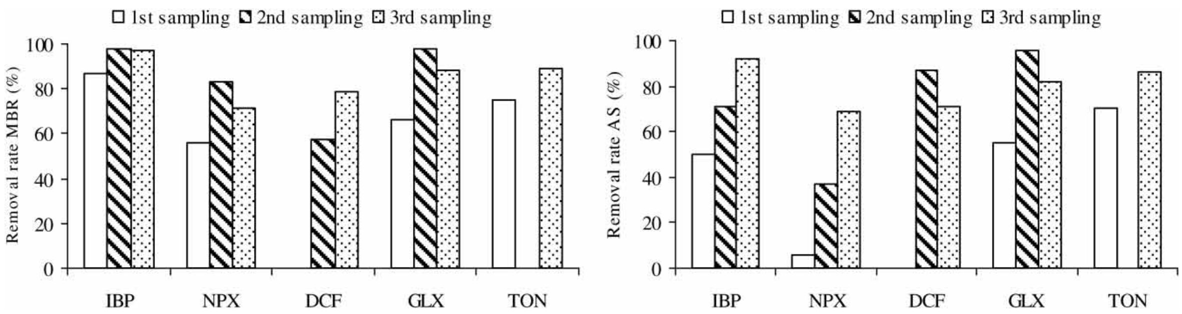

Figure 3 | Removal rates for PPCPs in MBR and AS along the sampling campaigns.

mechanism. During the second sampling campaign, musk fragrances content associated with the solid phase was measured and significant amounts of GLX were detected on the sewage suspended solids and sorpted onto the biological sludge of both reactors (Table 3). Calculated galaxolide solid-water distribution coefficients (K_d) for primary sludge and AS biological sludge were in good agreement with previous researching works (Suarez *et al.* 2008). On the contrary, and according to K_d data, MBR biological sludge showed higher sorption potential compared with the AS sludge. In despite of this tendency, calculated removal rates considering both liquid and solid phase were only slightly higher (4%) in the case of the MBR, mostly due to the low amount of suspended solids present in the AS effluent and the similar performance of both bioreactors.

A minimum amount of the musk fragrance tonalide was detected on biological sludge samples but calculation of K_d was not possible in this case, since its concentrations on the liquid phase were below detection limit during this sampling campaign.

In general, the removal rates from the liquid phase ranged from intermediate to high (60–90%) following the same trend in both systems, and higher eliminations were achieved after increasing the SRT of both systems. Extended SRT implies higher retention times inside the reactor for lipophilic substances. Under these conditions, biological transformation might occur to a certain extent. A more feasible explanation might be based on the increase of suspended solids (Table 4) inside both systems which should enhance the fraction removed by sorption. Removal rates are in good agreement with MLSS content with the exception of galaxolide in the AS system during the third sampling, when the highest removal rate should be observed.

As shown in Table 4, significant differences were found regarding the biomass particle size measured in both bioreactors. The particle sizes of the AS biomass are comparable to values reported in the literature (Massé *et al.* 2006), while MBR biomass particles sizes are substantially smaller and seemed to decrease when increasing the SRT from 6 to 20 days.

This factor might explain the differences between galaxolide solid-water distribution coefficients calcultated for the MBR and AS biomass, enhancing the availability of contaminants and oxygen for the biodegradation mechanisms. However, sorption is a complex phenomenon which involves different processes such as absorption and adsorption. Absorption is referred to the hydrophobic interactions of the micropollutants with the lipophilic cell membrane, the lipid fractions of the slude or the colloidal organic matter present in the mixed liquor. Adsorption is referred to the electrostatic interactions between micropollutants and the solids present (Meakins *et al.* 1994). As a consequence of this variety of mechanisms, links between biomass properties and the micropollutants removal capacity have not been completely established yet. In

Table 3 | Musk fragrances concentrations detected on the solid phase in the sampling points of the MBR and AS pilot plants

	μg GLX g TSS^{-1}	Log *Kd*	μg TON g TSS^{-1}	Log *Kd*
Primary sludge	51.97	3.63	n.d.	–
Secondary sludge (CAS)	2.12	3.57	0.33	–
Secondary sludge (MBR)	6.07	4.44	0.24	–

Table 4 | MLSS/MLVSS content inside aeration tanks and biomass particle size in AS and MBR systems. Particle size unit correspond with the median value of the particle size distribution

	Particle size *d*0.5 (μm)		MLSS (g L^{-1})		MLVSS (g L^{-1})	
Sampling	AS	MBR	AS	MBR	AS	MBR
1st	252	85	0.20	0.41	0.18	0.33
2nd	381	39	0.73	1.90	0.67	1.48
3rd			3.33	1.50	3.08	1.39

order to identify the most effective technology to remove lipophilic substances from sewage, further work should include batch sorption test comparing different types of biomass and longer CAS-MBR paralell operation under constant spiking conditions including solid-phase analysis on each sampling campaign. Regarding the overall removal rates, the MBR achieved slightly better results, but the differences were not high enough to confirm this trend.

Vertical flow reed beds performance

After post-treatment, a significant depletion of PPCPs concentration was achieved (Table 5).

In the case of anti-inflammatories, this finding confirms that biodegradation mechanisms are viable in reed beds, probably due to the prevalent aerobic conditions. Ibuprofen and naproxen concentrations at the outlet of both reed beds were comparable, and slightly affected by the influent variability. However, concentrations were below 1.0 ppb in most cases, and particularly lower for VFRB MBR, probably due to the low content of suspended solids and organic matter in the MBR permeate. No significant differences were found along the sampling campaigns for these substances. Diclofenac content was substantially higher at the outlet during the second sampling campaign, probably due to the similarly higher concentrations in sewage (up to 25 ppb). The increase on diclofenac concentration after the spike probably helped to improve the acclimation of microorganisms to this substance, enhancing its availability for transformation despite its recalcitrant character (Omil *et al*. 2010). Consequently, diclofenac concentration in effluents was the lowest during the third sampling. The antiepileptic carbamazepine was also spiked for the third sampling campaigns, in order to gather information of recalcitrant substances in the reed bed system, but its removal was almost negligible.

Similar conclusions are drawn for musk fragrances. No differences were found among VFRB MBR and VFRB AS, and their concentrations at the outlet ranged from 0.1 to 0.3 ppb. Only slightly high levels were found for galaxolide during the third sampling campaign (1 ppb). Matamoros *et al*. (2007) reported substantially higher removal efficiencies (up to 90%) for musk fragrances on a vertical flow constructed wetland, using a larger pilot plant (5 m^2). However, these experiments were performed with urban wastewater, and higher levels of musk fragrances were available at the inlet of the pilot-plant.

Table 5 | PPCPs concentration at the inlet and outlet of the vertical flow reed beds

	Liquid phase PPCPs analysis (μg L^{-1})					
	IBP	NPX	DCF	GLX	TON	CBZ
1st sampling						
VFRB AS						
Inlet	3.11	1.81	n.d.	0.65	0.19	n.d.
Outlet	0.53	0.27	n.d.	0.27	0.12	n.d.
VFRB MBR						
Inlet	0.80	0.84	n.d.	0.50	0.16	n.d.
Outlet	0.09	0.15	n.d.	0.31	0.12	n.d.
2nd sampling						
VFRB AS						
Inlet	2.04	0.89	3.18	0.54	n.d.	n.d.
Outlet	1.73	0.69	1.32	0.19	n.d.	n.d.
VFRB MBR						
Inlet	0.15	0.24	10.79	0.21	n.d.	n.d.
Outlet	0.02	0.16	2.83	0.12	n.d.	n.d.
3rd sampling						
VFRB AS						
Inlet	0.50	0.89	0.69	1.73	0.32	4.19
Outlet	<LC	0.80	<LC	1.02	0.26	3.10
VFRB MBR						
Inlet	0.29	0.83	0.51	1.14	0.25	6.75
Outlet	0.18	0.66	0.09	1.01	0.25	6.81

CONCLUSIONS

Conventional and innovative wastewater treatment technologies (AS and MBR) which were operated under strictly similar conditions have been tested in terms of micropollutants removal. MBR showed higher removal efficiencies particularly at low SRT (6 days), conditions that are commonly considered less favorable for micropollutants removal. After establishing SRT of 20 days, both systems reduced the discharge of micropollutants at comparable rates, ranging from 0.3 to 0.9 ppb for acidic drugs and from 0.2 to 1.7 ppb in the case of musk fragrances in the final effluents. Consequently, upgrading an existing STP based in the AS process by coupling membrane filtration technology would not be justified only in terms of micropollutants removal. Nevertheless, MBRs main advantages are still attractive for optimising the quality of wastewater treatment.

A green emerging technology used for post-treatment, filtration through vertical flow reed-beds, led to a general improvement of effluent quality, confirming them as an interesting alternative due to their low operational and maintenance costs, especially in sensitive or remote locations where other more expensive post-treatment systems might not be viable.

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