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Improved elimination of organic micropollutants by a process combination of membrane bioreactor (MBR) and powdered activated carbon (PAC)

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ABSTRACT

Membrane bioreactors (MBR) are increasingly considered for de-centralized wastewater treatment. As MBR can be operated with higher sludge retention time and total suspended solids than conventional wastewater treatment plants (WWTP) the elimination of micropollutants is slightly better for MBR than for WWTP. However to be able to use the MBR filtrate for artificial ground water recharge it is necessary to further improve elimination of micropollutants. In this study, the MBR process has therefore been combined with the adsorption on powdered activated carbon (PAC). PAC was dosed to the MBR filtrate of a pilot plant and removed again after a contact time of 30 min. PAC was recycled to the MBR process tank to use the residual adsorption capacity. The process was operated continuously for a period of 110 d. With 5–10 mg l^{-1} PAC dosage a 50–80% improved elimination of carbamazepine and diclofenac could be achieved compared to the process without PAC.

Keywords: Organic micropollutants; Membrane bioreactor MBR; Powdered activated carbon (PAC); Waste water treatment; Fouling; Elimination of persistent organic pollutants (POPs)

1. Introduction

As part of the SMART Jordan Valley project [1], new integrated approaches for water management, aquifer recharge, and wastewater reuse are developed. Decentralized membrane bioreactor technologies combined with consecutive subsoil conditioning are studied with respect to the removal of persistent organic pollutants (POPs) and pathogenic organisms. Pharmaceutical active substances enter the surface water with effluents of wastewater treatment plants (WWTP) due to incomplete elimination by conventional biological wastewater treatment [2]. Considering the fate of organic micropollutants in the aquatic environment both partitioning and persistence are important. Partitioning includes sorption by sediments, evaporation and uptake by biota. The persistence of pharmaceuticals is governed by biodegradation, sunlight photolysis and other abiotic transformation such as hydrolysis [3].

Biodegradability varies significantly for the different pharmaceutical residues. For example, the antiepileptic drug carbamazepine was not removed in various WWTPs [4,5]. For diclofenac, in some wastewater treatment processes elimination rates of 50%–90% appeared, whereas only slight removal was observed in other WWTPs [4–6]. Other compounds such as the analgesics ibuprofen and naproxen or the lipid regulators gemfibrozil and bezafibrate were removed up to >90% [5–7]. X-ray contrast agents belong to the persistent substances and are more or less not degraded in WWTPs.

Improvement of sewage treatment, for instance through membrane technologies, by higher solid

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retention times or combinations of aerobic and anoxic conditions, is considered to enhance biological elimination of pharmaceuticals and other degradable micropollutants to reduce their emission into aquatic environments [4,8–10]. In this research a membrane bioreactor (MBR) has been operated with the objective of an optimized elimination of POPs [10].

Studies on the reuse and recycling of wastewaters show that the possibilities for application very much depend on factors such as wastewater composition and availability with regard to amount and fluctuations as well as requirements and obligations connected with the type of reuse. These factors have to be considered when choosing a treatment strategy. Especially the presence of organic micropollutants limits the reuse of treated wastewaters. Therefore treatment technologies have to be checked for their elimination potential.

In order to be able to reuse treated wastewater for ground water recharge the aim is to completely eliminate organic micropollutants.

Due to the complete retention of biomass MBR can be operated at much higher sludge concentrations resulting in a higher biological activity per unit volume. Because of this feature the MBR process was said to have a higher elimination capacity for such substances than conventional WWTP [11–14]. However others found out that only a few substances showed better removal in MBRs than in WWTPs [5,15].

To further improve the elimination of organic micropollutants MBR may be combined with adsorption to powdered activated carbon (PAC). Tests were carried out to determine the required amount of PAC, the optimal contact time, the type of PAC, its capability to eliminate pollutants and the evaluation of an optimized process technology.

2. Materials and methods

2.1. MBR operation

The MBR pilot plant (HUBER SE, Berching, Germany) consists of a process tank (volume of 800 l) in which two plate and frame membrane modules of 2 m² membrane area each are submerged. Two different membrane types were tested in parallel.

A schematic of the MBR process is shown in Fig. 1.

Properties of the membranes are given in Table 1; the operation conditions in Table 2.

The filtrate flux of both membranes was adjusted to 15 l m^{-2} h^{-1} and kept constant during long term operation. Filtration time was set to 2 min. For mechanical cleaning of the membrane surface the filtration process is interrupted regularly for 1 min of relaxation time (pause). Membranes are aerated continuously

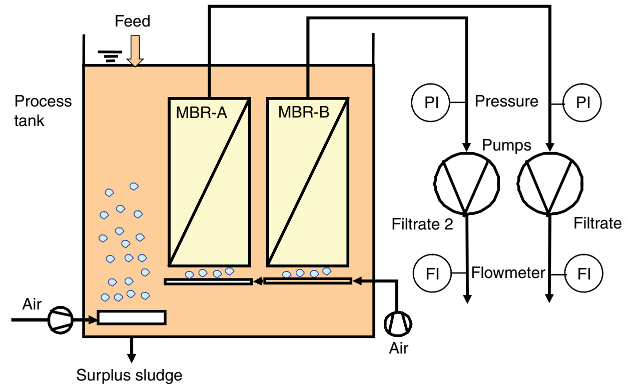

Fig. 1. Schematic illustration of the membrane test unit.

(1.3 Nm³ m^{-2} h^{-1}) in order to support removal of deposits on the membrane surface. For biological degradation processes the process tank is aerated intermittently (10 s on, 10 s off) with 3 Nm³ h^{-1}. Flux should stay below critical flux because high flux may accelerate adsorption and therefore fouling processes [16]. Lowest fouling was observed when sludge retention time (SRT) is adjusted between 20 and 50 d [16].

The MBR was operated continuously from November 2007 to November 2010 at the wastewater treatment plant in Eggenstein–Leopoldshafen, Germany. It received the effluent of the primary treatment of the full-scale WWTP which consisted of mechanical pre-treatment (screening/sieving (8 mm) and sand and grease removal). The mean flow rate of the WWTP was in the range of 2800 and 3500 m³ d^{-1} at dry weather conditions. The plant mainly received domestic wastewater with an industrial input of less than 5%. The main treatment was an activated sludge reactor with intermitting aeration and a dosage of sodium aluminates for phosphorous removal. Hydraulic retention time is 5 h, sludge age is 20 d and TSS is 5 g l^{-1}.

2.2. Sampling

Time proportional 24 h composite samples of influent and MBR effluent were regularly taken. Effluent sampling was adjusted according to the HRT of 24 h under the present conditions. Influent samples were filtered over 0.2 µm cellulose nitrate filters. During the process combination with PAC grab samples were taken from the MBR filtrate before and after PAC treatment.

2.3. Analytical methods

Analysis of pharmaceutical residues and X-ray contrast agents was done by high-performance liquid chromatography with tandem mass spectrometric detection (HPLC-MS-MS) after solid-phase extraction at pH 3

Table 1
Membrane specification for MBR

Properties	MBR-A	MBR-B
Membrane type	Ultrafiltration	Microfiltration
Material (active layer)	Polyethersulfone	Polyvinylidenefluoride
Pore size, nm	38	200
Permeability (20°C) (clear water), $l\ m^{-2}\ h^{-1}\ bar^{-1}$	350–620	180–200
Membrane area, m^2	2	2

Table 2
MBR operation and sludge conditions

Parameters		MBR
Filtrate flux	$l\ m^{-2}\ h^{-1}$	15
Transmembrane pressure	mbar	–20––800
Filtration time interval	min	2
Time for relaxation (pause)	min	1
Air for cleaning	$Nm^3\ m^{-2}\ h^{-1}$	1.3
Sludge age	d	25
TSS within process tank of MBR	$g\ l^{-1}$	8–11
Hydraulic retention time HRT	h	24
Temperature	°C	10–16

using styrene divinylbenzene material. The detection limit of X-ray contrast agents was 10 ng l^{-1}. However depending on the water type due to the need of dilution during sample preparation the detection limit may go up to 100 ng l^{-1}. Details of the analytical procedures as well as of the instrumentation are described elsewhere [17,18]. A list of the pharmaceuticals analyzed and some of their properties are given in Table 3.

The octanol–water partitioning coefficient (log K_{OW}) is a measure for the hydrophobicity of neutral organic substances. Values larger than 2 refer to hydrophobic, values smaller than 2 to hydrophilic compounds. If substances dissociate with various pH the octanol–water distribution coefficient log D_{OW} is used. The p_{Ka}-values give the pH when there is no dissociation. There were no values available for X-ray contrast agents.

COD was measured according to DIN 38409-H41 using cuvette tests LCK414 and 514 (Hach Lange). Colour was measured by a spectrophotometer (CADAS 200, Hach Lange, Düsseldorf, Germany) according to DIN EN ISO 7887-C1 at a wavelength of 436 nm.

2.4. Powdered activated carbon

Two types of PAC from DONAU CARBON were tested. Specific properties are given in Table 4. According to standardization (DIN EN 12903) PAC usually has a mean particle size in the range of 10–50 µm with 95% of all particles having a size <150 µm. The PAC types used here have a larger amount (60–70%) of particles smaller than 40 µm.

Table 3
List of pharmaceuticals analyzed and properties of interest

Analytical group	Substances	Properties of interest [3,25,26]		
		p_{Ka}	log D_{OW} (at pH 7)	log K_{OW}
Pharmaceuticals	Bezafibrate	3.3 [3]	2.25	3.44 [26]
	Carbamazepine	13.9 [3]	2.67 [3] 2.58 [26]	2.45 [26]
	Clofibric acid	3.64 [25]	0.57	2.57 [25]
	Diclofenac	3.2 [3]; 4.15 [25]	1.7 [27]	4.51 [25]
	Fenofibric acid	–	1.35	–
	Gemfibrocil	4.7 [3]	2.77	4.77 [26]
	Ibuprofen	4.9 [3,25]	1.16 [3] 0.77 [26]	3.97 [25]
	Ketoprofen	4.5 [3]	1.12	–
	Naproxen	4.2 [3,25]	0.34 [26]	3.18 [25]
Not detected	Diazepam, etofibrate, fenoprofen, fenofibrate, indomethacine, pentoxifyllin, phenacetine			
X-ray contrast agents	Amidotrizoic acid, iodipamide, iohexol, iomeprol, iopamidol, iopromid, iotalamic acid, ioxaglic acid, ioxithalamic acid			

Table 4
Specific properties of PAC used

	PAC1 Carbopal AP[a]	PAC2 Hydraffin P800[a]
Water content, wt.%	<5	<8
Ash content, wt.%	–	<10
pH-value	7–9	9–10
Iodine number, mg g^{-1}	1250	>900
BET, m^2 g^{-1}	1300	ca. 900
Particles <40 µm, wt.%	70	ca. 60
Elimination of polyaromatics, %	>90	–

[a]Donaucarbon GmbH & Co KG.

Table 5
Composition of MBR influent and effluent

	Influent ranges (no.)	Effluent ranges (no.)
COD, mg l^{-1}	132–1290	13–23
Temperature, °C	9–29	9–29
Bezafibrate, ng l^{-1}	540–3200	<50–110
Carbamazepine, ng l^{-1}	390–1800	600–1600
Clofibric acid, ng l^{-1}	<100	<50
Diclofenac, ng l^{-1}	550–5200	1200–2800
Fenofibric acid, ng l^{-1}	360–1400	<50
Gemfibrozil, ng l^{-1}	810–1500	<50–180
Ibuprofen, ng l^{-1}	3800–11,000	72–300
Naproxen, ng l^{-1}	530–3700	<50–250

In order to test the capacity of the PACs for colour removal, samples of the MBR filtrate from the pilot plant were spiked with PAC (various dosing rates). After a given contact time (10–60 min) PAC was removed by filtering and samples were analyzed for colour (UV absorption at 436 nm). For the determination of the removal capacity with regard to POPs such experiments were carried out with a dosage of 10 mg l^{-1} PAC and analysis of filtered samples after a contact time of 30 min.

3. Results and discussion

3.1. Influent quality

The MBR was operated with pre-treated wastewater. Seasonal fluctuations resulted in a range of values for COD content and temperature as given in Table 5. Mean value for COD was 612 mg l^{-1} with maximum values of 1290 mg l^{-1} COD depending on wastewater throughput and rainfall events.

Temperature changed between 9°C and 29°C from winter to summer time.

Temperature plays an important role for the performance of the MBR. As viscosity decreases with increasing temperature TMP decreases accordingly. Although permeability is normalized to a temperature of 20°C there is still an influence of temperature observable. This may be explained by the change in the composition of the sludge due to the seasonal variations [19]. It was shown that EPS is more likely released at low temperature than at higher temperature. This may be the cause for a stronger increase in TMP during cold weather conditions which was observed in the presented experiments as well.

The municipal wastewater used for the operation of the MBR contained many of the substances of interest in sufficient concentration so that no spiking of POPs was required. Table 5 (left column) gives a list of the substances and the ranges measured for the analgesics and anti-inflammatory drugs (ibuprofen, naproxen, diclofenac), the lipid regulators (clofibric acid, gemfibrozil, bezafibrate, fenofibric acid) and the antiepileptic drug (carbamazepine). Similar concentrations of these substances have been also measured by others in municipal wastewaters for most of the substances [5,11,12,20]. Clofibric acid, which is a very refractory compound and was detected in river water [21] and wastewater [12], has not been detected in the wastewater used in this study. Ketoprofen and iopromide were also not detected.

3.2. Removal of pollutants by MBR

From the values in Table 5 (right column) it can be seen, that some substances (ibuprofen, bezafibrate, fenofibric acid, gemfibrozil), are easily biodegradable, whereas others (carbamazepine, diclofenac) passed the MBR almost without changes in their concentrations. In Fig. 2 concentrations of these substances in the influent and the effluent of the MBR are shown in comparison to the values for the WWTP.

The elimination of organic micropollutants is caused by two mechanisms: sorption to biomass and biodegradation. It is dependent on the specific properties of the substances [3]. The main elimination mechanism of organic micropollutants was found to be biodegradation [3,11]. However in the case of diclofenac the decrease in concentration by sorption to the sludge was demonstrated to be an important factor [11]. Kimura et al. found that MBR sludge has a higher specific sorption capacity than sludge in WWTP [11]. Therefore higher elimination may be observed in MBR compared to WWTP.

Carbamazepine and clofibric acid were found to be more persistent than ibuprofen [3]. The results shown in Fig. 2 confirm this at least for carbamazepine and diclofenac. There is no difference in the removal rates between MBR and WWTP for these substances.

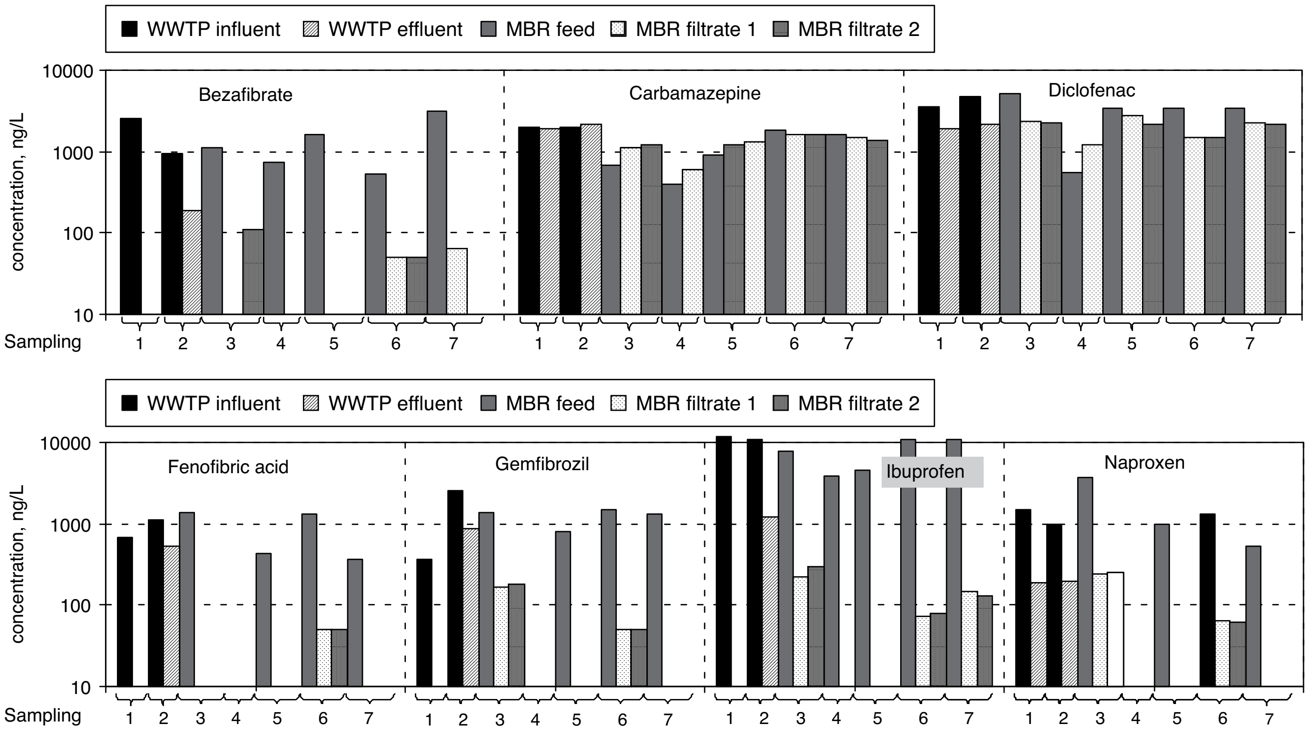

Fig. 2. Concentration of organic micropollutants in influents and effluents of the conventional WWTP (sampling times 1 and 2) and MBR (sampling times 3–7) (dark colours = influent; light colours = effluent).

As elimination of some POPs is not at all sufficient in regular operation of MBRs a tertiary treatment of wastewater is recommended by [20]. At some WWTPs tertiary treatment steps are already investigated. As a process for tertiary treatment the adsorption to activated carbon is considered. It is a feasible step to eliminate substances that are well adsorbed to activated carbon.

3.3. Removal of pollutants by a process combination of MBR and PAC

In order to be able to reuse treated wastewater for artificial ground water recharge the aim is to fully eliminate organic micropollutants. In this research PAC has been used as a tertiary treatment step for this purpose. In order to find a PAC which was most suitable for the elimination of POPs the experience with adsorption processes for drinking water was considered [22]. Two PACs were recommended and have been tested for their removal capacity of colour and POPs. From the results shown in Table 6 it can be concluded, that PAC2 is not as efficient for colour removal as PAC1.

For PAC1 colour removal increased with increasing PAC dosage as was expected. At 10 mg l^{-1} the elimination of colour was almost 10%. However from this experiment no conclusions can be drawn to the elimination of POPs. For a dosage of 10 mg l^{-1} PAC and a contact time of 30 min removal of POPs has been measured in another experiment. Results are given in Table 7. For most of the substances PAC1 showed higher values of elimination than PAC2. Differences in removal efficiency of the two PACs tested may be related to their different physical–chemical properties and the interactions between PAC and the organic compounds which are dependent on surface potential gradients between adsorption sites of the PAC and functional groups of the organic compounds.

Table 6
Colour removal efficiency of PAC for varying dosage rates at a contact time of 30 min

Dosage of PAC, mg l^{-1}	PAC1 (%)	PAC2 (%)
10	9 ± 1	<8 ± 1
20	25 ± 3	<4 ± 1
30	39 ± 2	<8 ± 1
40	48 ± 1	<9 ± 1

Therefore PAC1 was used for the long-term experiment where PAC was dosed to the MBR filtrate for a period of 12 weeks and recycled to the MBR after a second filtration step. The scheme of the set up is shown in Fig. 3.

PAC was continuously dosed to the MBR filtrate. The resulting "MBR filtrate/PAC-suspension" was stirred

Table 7
Removal efficiency of PAC for POPs at a dosage rate of 10 mg l^{-1} and a contact time of 30 min

Substance	PAC1 (%)	PAC2 (%)
Diclofenac	99.2	67.5
Carbamazepine	99.4	92.0
Naproxen	87.3	95.1
Amidotrizoic acid	66.1	73.8
Iopamidol	98.6	33.3

continuously in a separate tank (reactor). The dosage was adjusted to 10 mg l^{-1} PAC. After a mean contact time of 30 min the suspension was pumped through a second UF membrane to remove the PAC in order to be able to recycle it to the MBR process tank. If the used PAC is recycled to the MBR process tank it has an additional effect on the elimination process in the MBR as it still has a residual adsorption capacity.

With this MBR/PAC process combination a further elimination of POPs was achieved. Fig. 4 shows the elimination measured for some POPs during the long-term run.

Carbamazepine and diclofenac showed elimination efficiencies larger than 50%–80% and even the X-ray contrast agent amidotrizoic acid which belongs to the group of persistent substances with a very low bio-degradation was partly removed at least in five out of eight cases. Fluctuations are caused by differing influent concentrations. This also causes negative values for the elimination when effluent concentrations were higher than influent concentration. In the case of iopamidol concurring adsorption takes place as it is less absorptive than other X-ray contrast agents.

It is not clear which properties of the compounds are responsible for the elimination. From the p_{Ka}-values in Table 3 it can be derived that besides carbamazepine all substances are dissociated at the pH-value of 7 in the wastewater. Because of the higher log K_{OW}-value diclofenac showed a better elimination in the PAC treatment compared to carbamazepine which has a lower log K_{OW}-value. To elucidate the influencing factors more research is required.

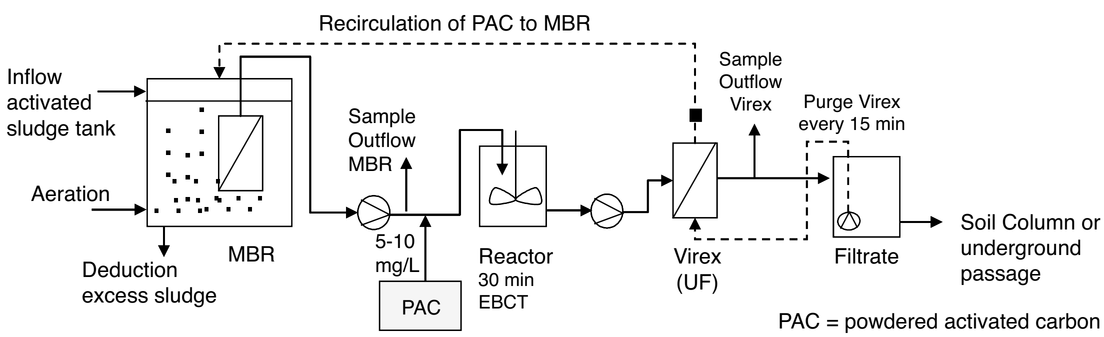

Fig. 3. Schematic illustration of the experimental set-up for the MBR filtrate post treatment by PAC.

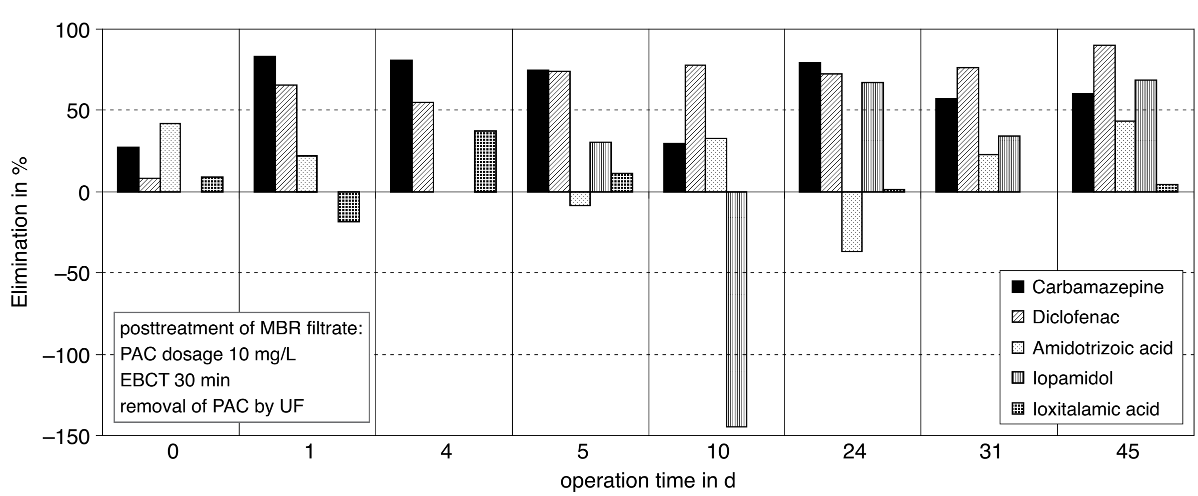

Fig. 4. Elimination of POPs by the combination of MBR and PAC post treatment.

3.4. Performance of MBR

During operation of MBRs a decrease of permeability occurs with operation time caused by fouling processes. Main mechanisms of fouling are adsorption of solutes or colloids within or on the membrane, deposition of sludge flocs onto the membrane surface, detachment of foulants due to shear forces and spatial and temporal changes of the foulant composition during long-term operation (e.g., change of bacteria community and biopolymer components in the cake layer).

The long-term operation with regard to COD removal is comparable to results published by Meuler et al. who operated many small-scale MBRs [23].

During PAC dosage and removal to the MBR process tank no TMP increase was observed with the MBR-A. However, MBR-B showed a clear TMP increase with time. This effect is shown in Fig. 5 where the change in permeability (normalized to 20°C) is plotted versus operation time for 150 d for both membranes.

It seems that the larger pores of the microfiltration membrane MBR-B are increasingly blocked by the PAC whereas the smaller pores of the ultrafiltration membrane MBR-A were not. Additionally the membrane material is different. Polyvinylidenefluoride is less hydrophilic than polyethersulfone and may therefore be more prone to organic fouling [21].

According to Meng et al. the function of the PAC in membrane processes may be twofold [16]. On the one hand PAC adsorbs the micropollutants. On the other hand it reduces fouling because it also adsorbs foulants. This positive effect was observed also by Remy et al. who demonstrated a PAC dosage of 0.5 g l^{-1} to be suitable to reduce fouling of MBRs [24]. From several possible mechanisms the formation of stronger sludge flocs in the presence of PAC and therefore a higher shear resistance and lower release of foulants was demonstrated to be the most likely explanation. However, the PAC dosage of the investigations presented here was 50 times smaller.

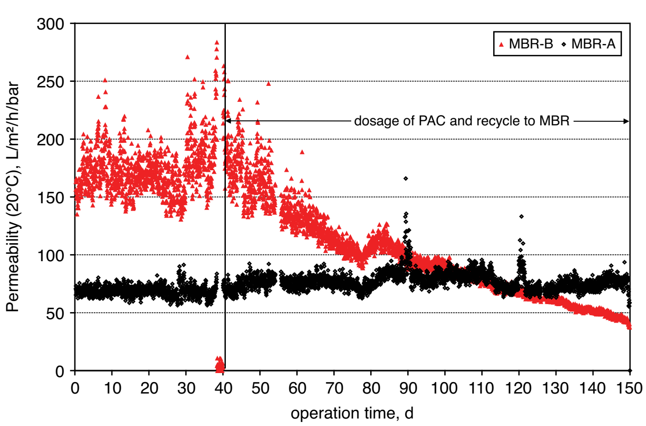

Fig. 5. MBR performance during operation with PAC.

4. Conclusions

Removal rates for organic micropollutants in the MBR were in the same range as for WWTP and considered not to be sufficient for the requirements of an artificial ground water recharge. Despite the fact that soil passage also has a potential for biodegradation of organic micropollutants under certain conditions [27], the intension is to improve removal rates especially for the persistent POPs like carbamazepine and diclofenac or X-ray contrast agents by using other processes prior to soil passage. In the case study presented here the efficiency of an adsorption to activated carbon was investigated as a tertiary treatment step in pilot scale.

The results have shown that the process combination of MBR and PAC has positive as well as negative effects. With a PAC dosage of 10 mg l^{-1} a further removal of POPs of 60–80% can be achieved especially for carbamazepine and diclofenac. Some of the X-ray contrast agents can be also absorbed to a certain percentage. However a drawback is that for the removal of PAC from the suspension a second filtration step is required. Moreover the presence of activated carbon in the MBR process tank caused fouling of the MF membrane, whereas the UF membrane was not affected. The PAC removed by the second filtration step can be recycled back to the MBR process tank where residual adsorption capacity can be used.

The rather good filtrate quality of the MBR is suitable for artificial ground water recharge, however, the operation of the MBR process results in a rather high energy demand mainly because of the required aeration of the membrane surface to avoid fouling.

One possibility to minimize energy demand and avoid a second filtration step, PAC may be directly dosed to the MBR process tank. In a further research project this will be evaluated in detail.

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Symbols

EPS	—	extra cellular polymeric substance
HRT	—	hydraulic retention time
MBR	—	membrane bioreactor
PAC	—	powdered activated carbon
POPs	—	persistent organic pollutants
SRT	—	sludge retention time
TMP	—	transmembrane pressure

TSS	—	total suspended solids
WWTP	—	conventional wastewater treatment plant

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