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Integrating granular activated carbon in the post-treatment of membrane and settler effluents to improve organic micropollutants removal

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HIGHLIGHTS

- The application of GAC filters was evaluated to treat effluents of different quality.
- The GAC breakthrough was quicker for diclofenac than for carbamazepine and diazepam.
- The high TSS content in settler effluent strongly limited the lifespan of GAC filters.
- The 2 types of GAC tested exhibited very similar technical performance in OMP removal.
- Economical and environmental criteria were included to select the most suitable GAC.

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ABSTRACT

Granular activated carbon (GAC) is applied as post-treatment technology in wastewater treatment plants (WWTPs) in order to increase the elimination of organic micropollutants (OMPs). However, the efficiency and life-time of GAC depend on several parameters, such as the quality of the effluent to be treated or the type of GAC. In the present paper, two types of GAC, based on bituminous carbon (BC-GAC) and coconut shell (CS-GAC), were assessed from a technical, economic and environmental point of view to further remove OMPs present in two secondary effluents, coming from integrated biological systems with a membrane or a settler, respectively. Although all GAC filters were efficient in removing selected OMPs, the quality of the secondary effluent had a strong influence on the lifespan of adsorbent material and the technical operability of the filtration systems. While GAC filters treating membrane effluent were highly effective to remove recalcitrant compounds, such as carbamazepine and diazepam (> 80%), even after 430 d of operation (> 30,800 BV), the efficiency of GAC filters treating settler effluent quickly lowered to 50% after 100 d of operation (< 7200 BV). Both types of GAC showed similar adsorption capacities and only slight differences were found in terms of costs (2.4 €/kg vs 2.7 €/kg). However, CS-GAC has a lower carbon footprint than BC-GAC, mainly due to the more environmentally friendly production process of CS-GAC.

1. Introduction

The appearance of new challenges in wastewater policy concerning organic micropollutants (OMPs), such as the inclusion of four pharmaceuticals and three hormones in the watch list defined by European Commission (Decision 2015/495/EU), have promoted that some countries, such as Switzerland, have already established OMPs discharge thresholds in WWTP effluents [1]. Others, such as Austria, Germany and The Netherlands, are considering some initiatives to increase the removal of OMPs during wastewater treatment (overall elimination of 80%) [2]. A potential solution to achieve these challenges is the application of post-treatment technologies in order to increase the removal of OMPs in WWTPs and, consequently, to enhance the quality of treated effluents before their discharge into the environment. Adsorption on activated carbon and ozonation are the most applied tertiary treatment technologies in WWTPs since both were proven as effective to reduce the concentration of different OMPs [2–4] and are economically viable [5]. Despite its high capacity for OMPs abatement, the possible appearance of toxic by-products during the ozonation stage compromises its sole application as final treatment unit in WWTPs [6]. Activated carbon can be used as granular activated carbon (GAC) or powdered activated carbon (PAC) in wastewater treatment. PAC is normally applied in combination with biological treatment [7] whereas GAC is used as post-treatment system with the

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additional advantage that can be regenerated once its adsorption capacity has been exhausted [2,8].

The application of GAC filters to remove OMPs from WWTP effluent was widely studied in literature [2,9,10]. The properties of activated carbon, the empty bed contact time (EBCT) and the composition of the effluent to be treated were identified as key parameters affecting the process efficiency [8,9,11].

Different types of activated carbon exist in the market, being their properties (specific surface area, pore size and composition) dependent on the raw material used as precursor and the production process [12]. The selection of the raw material together with the method employed for its activation (physical or chemical) also affects the market price and the carbon footprint [13]. Previous studies evaluated the application of GAC and PAC with different origin (hard coal, coconut shell, wood and peat), observing a strong link between the efficiency of activated carbon to remove OMPs and its specific surface area [14,15]. The pore size is also a key factor controlling the effectiveness of the adsorbent material for a given OMP [16].

The influence of EBCT on the efficiency of GAC filters to remove OMPs was extensively studied [9,11,17], being the typical values employed in WWTPs between 10 and 30 min [2]. EBCT mainly affects the adsorption kinetics of OMPs on the adsorbent material and the size of the filtration system.

Most studies found in literature are based on the post-treatment of secondary effluents coming from conventional activated sludge (CAS) systems and few information is available about the lifespan of these technologies treating effluents from membrane bioreactors (MBR) [2,18]. While the effect of dissolved organic matter on the saturation of GAC filters is unquestionable [9,10], the influence of total suspended solids (TSS) (either colloids and/or sedimentable solids) on the adsorption capacity of filters is not clear. Even though a negative effect is expected on the adsorption process, because TSS can block the pores of the adsorbent material hampering the arrival of OMPs to the surface of GAC, other factors, such as the frequency of backwash cycles and the development of a biofilm on the surface of GAC should be also considered [19,20]. This unclear effect was evidenced in literature since Bornemann et al. [21] observed higher removals for carbamazepine and sulfamethoxazole in GAC filters preceded by a sand filter where TSS were separated. In contrast, Benstoem et al. [22] could not demonstrate that the sand prefiltration extended the lifespan of GAC.

The main objective of this work was to assess the application of GAC filters to remove OMPs from two secondary wastewater effluents of different quality (from a membrane and a settler, respectively). Moreover, the influence of the type of GAC was assessed not only in terms of technical performance but also under an economic and environmental perspective. This would allow us to directly relate the results since the comparison among different studies is hampered by the application of filters of varying sizes and the use of various types of GAC and wastewaters with different composition.

2. Materials and methods

2.1. Target organic micropollutants

Three antiphlogistics (ibuprofen (IBP), naproxen (NPX) and diclofenac (DCF)), four antibiotics (erythromycin (ERY), roxithromycin (ROX), sulfamethoxazole (SMX) and trimethoprim (TMP)), an antidepressant (fluoxetine (FLX)), an antiepileptic (carbamazepine (CBZ)), a tranquillizer (diazepam (DZP)), three musk fragrances (galaxolide (HHCB), tonalide (AHTN) and celestolide (ADBI)) and five endocrine disruptor compounds (estrone (E1), 17β-estradiol (E2), 17α-ethinylestradiol (EE2), bisphenol A (BPA) and triclosan (TCS)), were selected in this work.

Table 1
Characterization of secondary effluents from membrane (n = 50) and settler (n = 28). n represents the number of samples considered to calculate the average value and the standard deviation for each parameter.

	Membrane effluent	Settler effluent
pH	8.1 ± 0.3	8.0 ± 0.3
DOC (mg/L)	3.5 ± 2.7	8.6 ± 4.9
Transmittance at 254 nm (%)	93.1 ± 2.6	89.0 ± 2.4
$N\text{-}NH_4^+$ (mg/L)	n.d.	12.0 ± 7.2
$N\text{-}NO_2^-$ (mg/L)	n.d.	0.4 ± 0.2
$N\text{-}NO_3^-$ (mg/L)	41.9 ± 12.0	19.0 ± 4.7
TSS (mg/L)	n.d.	32.0 ± 15.3

*n.d.: not detected

2.2. Selected secondary effluents

Two different secondary effluents were selected to assess the influence of effluent quality on the efficiency of GAC filters to remove OMPs. The effluents were produced in two biological systems treating the same urban wastewater composed of anaerobic treatment followed by anoxic/aerobic chambers (Fig. S1). The difference between both systems is the technology employed to separate the biomass from the treated effluent: one system employed a settler (Fig. S1A), whereas an ultrafiltration membrane was used in the second system (Fig. S1B) [23]. The physicochemical characterization and the concentrations of target OMPs detected in membrane and settler effluents are shown in Tables 1 and 2, respectively. It can be observed that the method employed to retain the biomass not only affects the concentration of suspended solids detected in the effluents (Table 1) but also the biological activity developed in the integrated system due to the effect of the recirculation between anoxic and aerobic chambers (nitrification was complete in membrane system, but only partial in settler system, Table 1). Consequently, the concentrations of OMPs detected in membrane and settler effluents were slightly different (Table 2).

2.3. Characterization of granular activated carbons

Two different types of granular activated carbon were tested: one based on bituminous carbon (BC-GAC) (MG 1050, ChiemiVall) and the second produced from coconut shell (CS-GAC) (CG 1000, ChiemiVall). These adsorbent materials were selected in order to evaluate and compare from a technical, environmental and economic point of view

Table 2
Concentrations of OMPs (μg/L) detected in membrane (n = 10) and settler (n = 12) effluents during whole operation of GAC filters. n represents the number of samples considered to calculate the average value and the standard deviation for each parameter.

	Membrane effluent	Settler effluent
CBZ	12.46 ± 4.78	14.00 ± 2.88
DZP	8.22 ± 2.91	9.29 ± 2.56
DCF	3.17 ± 1.38	5.73 ± 3.74
ADBI	3.99 ± 1.27	2.86 ± 0.66
HHCB	3.67 ± 0.94	2.33 ± 0.65
AHTN	2.90 ± 0.57	1.96 ± 0.42
FLX	1.24 ± 0.40	2.03 ± 0.28
NPX	0.55 ± 0.21	0.49 ± 0.20
ROX	0.25 ± 0.25	1.45 ± 0.99
BPA	0.38 ± 0.14	0.73 ± 0.38
IBP	0.28 ± 0.17	0.69 ± 0.30
TCS	0.29 ± 0.22	0.59 ± 0.22
ERY	0.04 ± 0.04	0.62 ± 0.54
SMX	0.005 ± 0.003	0.53 ± 0.53
TMP	0.01 ± 0.00	0.02 ± 0.00
EE2	0.07 ± 0.02	0.18 ± 0.02
E2	0.02 ± 0.01	0.02 ± 0.01
E1	0.02 ± 0.00	0.03 ± 0.02