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Advanced biological activated carbon filter for removing pharmaceutically active compounds from treated wastewater

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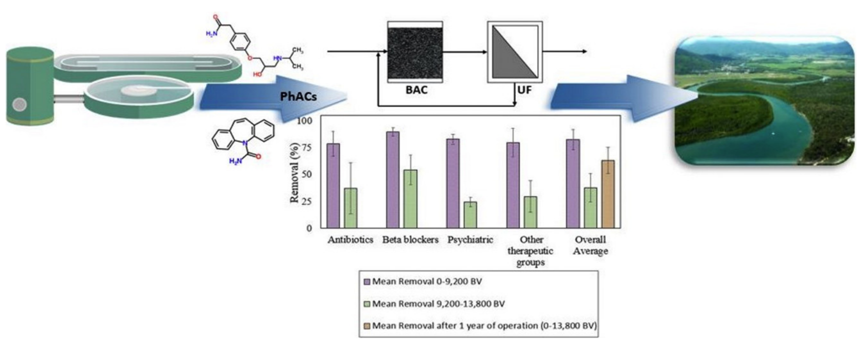
HIGHLIGHTS

- BAC-UF was evaluated for PhACs removal from wastewater.
- The one year monitoring allowed for both biofilm development and GAC saturation
- PhACs were removed to high extent during the first 9200 bed volumes.
- Adsorption was identified as main removal mechanisms for most of the PhACs.
- Biofilm contributed to improve the overall removal of nine PhACs.

GRAPHICAL ABSTRACT



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ABSTRACT

Through their release of effluents, conventional wastewater treatment plants (WWTPs) represent a major pollution point sources for pharmaceutically active compounds (PhACs) in water bodies. The combination of a biological activated carbon (BAC) filter coupled with an ultrafiltration (UF) unit was evaluated as an advanced treatment for PhACs removal at pilot scale. The BAC-UF pilot plant was monitored for one year. The biological activity of the biofilm that developed on the granular activated carbon (GAC) particles and the contribution of this biofilm to the overall removal of PhACs were evaluated. Two different phases were observed during the long-term monitoring of PhACs removal. During the first 9200 bed volumes (BV; i.e., before GAC saturation), 89, 78, 83 and 79% of beta-blockers, psychiatric drugs, antibiotics and a mix of other therapeutic groups were removed, respectively. The second phase was characterized by deterioration of the overall performances during the period between 9200 and 13,800 BV. To quantify the respective contribution of adsorption and biodegradation, a lab-scale setup was operated for four months and highlighted the essential role played by GAC in biofiltration units. Physical adsorption was indeed the main removal mechanism. Nevertheless, a significant contribution due to biological activity was detected for some PhACs. The biofilm contributed to the removal of 22, 25, 30, 32 and 35% of ciprofloxacin, bezafibrate, ofloxacin, azithromycin and sulfamethoxazole, respectively.

1. Introduction

During the last 20 years, the presence of several pharmaceutically active compounds (PhACs) in water bodies has garnered increasing attention and worldwide concern. The effluents from wastewater

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treatment plants (WWTPs) are the main sources of PhACs in aquatic environments (Daughton and Ruhoy, 2009). In fact, most WWTPs are designed to comply with conventional pollutant thresholds such as those for nutrients, organic matter and solids, but they are not efficient at completely eliminating organic compounds at low concentrations ranging from ng/L to μg/L (Verlicchi et al., 2012). Advanced tertiary treatments are required to improve the quality of WWTP effluents discharged into sensitive receiving water bodies and those utilized for potable reuse, industrial reuse and irrigation purposes (Siegrist and Joss, 2012). Among the advanced tertiary treatments, adsorption onto granular activated carbon (GAC) has been proven to be a viable technique to remove PhACs (Boehler et al., 2012; Hu et al., 2016a; Mailler et al., 2016a). The main drawbacks of GAC applications are (i) the need to properly treat and dispose of both the spent carbon and the sludge generated during the filtration process, and (ii) the need to remove the effluent organic matter before filtration. The effluent organic matter significantly diminishes the adsorption capacity of GAC and thus the operating lifetime. An alternative may be to take advantage of the organic matter retained in the GAC filter to support the growth of autochthonous biomass attached to the GAC particles. This colonization implies the development of biological activity inside the filter bed, and the process can thus be defined as biological activated carbon (BAC) filtration. BAC filters, compared to other biofiltration systems, have the advantage of presenting a high adsorption capacity, which contributes to the removal of PhACs as well as to biofilm development. It has been demonstrated that the GAC structure with its roughness and high porosity, possesses characteristics that promote microbial growth, thus providing better support compared to other materials such as sand, clay and anthracite (Luo et al., 2014). With regard to the abatement of PhACs, BAC filters combine biodegradation with physical adsorption (Zhang et al., 2010). By combining these two mechanisms, a wider range of PhACs can be addressed, thus increasing the potential of such technology for application as a tertiary treatment (Kalkan et al., 2011; Velten et al., 2011). Justo et al. (2015) showed that during BAC filtration treatment of reverse osmosis brine, approximately 60% of the pharmaceutical content was depleted by the biological treatment. When applied after coagulation and clarification of surface water for drinking purposes, this technique was confirmed to be a sustainable treatment process for PhACs, with removals higher than 75% (Zhang et al., 2017). In this context, the biofiltration process is often evaluated as a part of larger treatment trains in which BAC filters represent only one step of the entire treatment line, according to the multiple barrier criteria. Few studies have documented situations in which BAC filters, applied as part of wastewater reuse schemes to treat wastewater for reclamation purposes, were proven to effectively remove many PhACs (Rattier et al., 2012; Reungoat et al., 2012). Rattier et al. (2012) suggested that biodegradation can enhance the removal of the compounds that are less adsorbed onto GAC. Similarly, BAC filters have also been applied as the last step of an experimental indirect potable reuse treatment train comprising UF and ozone (Gerrity et al., 2011). When preceded by UF and ozone, BAC filtration effectively decreased the concentration of PhACs in the treated WWTP effluent to below the method detection limit (MDL). Nevertheless, to the best of our knowledge, little is known regarding the application of BAC filters as a stand-alone tertiary treatment for the removal of PhACs. Paredes et al. (2016) conducted a comparative study of BAC and sand biofiltration, which were both applied as stand-alone tertiary treatments. However, this study was conducted at lab-scale with synthetic water and spiked concentrations of PhACs. Rather than investigating the real operating conditions, the authors mainly focused on the specific removal mechanisms. Understanding the mechanisms shed light on the contributions of the biological processes taking place in these biofiltration systems. To assess whether biodegradation allows for greater removal of PhACs by BAC filters rather than by conventional GAC treatment, gaining insights into the mechanisms involved is key in biofiltration studies. As far as we know, only Rattier et al. (2012) and Paredes et al. (2016) have distinguished between biodegradation and adsorption during the BAC filtration of wastewater. Studying the BAC filtration of real treated wastewater at pilot scale, Gerrity et al. (2011) found higher concentrations of both total coliform and fecal coliform after BAC passage. Since bacteria grow on GAC particles, they can be detached and washed out from the biofilter. Aiming to prevent this escape, an ultrafiltration (UF) step can be performed after the BAC filter. When this BAC-UF combination was applied as a tertiary treatment, total coliforms, *E. coli*, Enterococci and Somatic coliphages were almost completely removed due to the UF step (Weemaes et al., 2011). On the other hand, single UF is ineffective for the removal of most of the PhACs found in secondary effluents of WWTPs. Several studies have shown limited removal of PhACs by UF membrane alone (Sheng et al., 2016; Snyder et al., 2007). Ultrafiltration alone is ineffective for PhACs removal because of its high molecular weight cut off and the unavoidable disadvantage of fouling when it is applied to wastewater treatment (Secondes et al., 2014). Weemaes et al. (2011) tested the use of this system for removal of PhACs from secondary effluents in short-term experiments. This concept, applied at the laboratory scale, was able to almost completely remove antibiotics, iodinated contrast media and analgesic anti-inflammatory compounds. Hence, to gain useful information for a real application of this BAC-UF technology, further investigations need to verify this hypothesis at a larger scale. Moreover, the long-term performance of the system after reaching PhACs breakthrough must also be investigated.

In summary, only a limited number of studies have used BAC filters as advanced technology to remove PhACs from secondary effluents. Furthermore, in these few studies, BAC filters were usually preceded by other treatment units (i.e., the technology was applied to refine tertiary effluents). When this was not the case, the studies were conducted at the laboratory scale and, therefore, within a controlled environment. Moreover, a long-term investigation of the BAC-UF performance, including the start-up phase, a few months of steady operation, and the GAC breakthrough, was not performed in any of the previous studies. To the best of our knowledge, this is the first time that a BAC filter has been evaluated as a stand-alone advanced treatment for the removal of PhACs at pilot scale during one year of operation; the results of this study will provide additional information about the extent to which biological processes can enhance the overall abatement of PhACs. Our hypothesis is that BAC, applied as a stand-alone tertiary treatment, can achieve better performance than other biofiltration systems and guarantee higher short-term levels of PhACs abatement than the conventional GAC treatment while providing the same long-term achievements.

The objective of the current work was to evaluate the long-term performance of the BAC-UF process in treating secondary effluents with environmentally relevant concentrations of PhACs at pilot scale. A lab-scale study with more controlled conditions was also performed to gain insights into the respective roles of biodegradation and adsorption.

2. Materials and methods

2.1. BAC-UF pilot plant

The BAC-UF pilot plant setup coupled two separate units, as shown in Fig. 1. The pilot plant was fed by secondary effluent (the wastewater characteristics are reported in Table S1) from a municipal WWTP designed for biological nutrient removal. The BAC filter was filled with ORGANOSORB ® 10-CO coconut shell-based GAC (Table S2) to an operating volume of 2 m^3. This resulted in an empty bed contact time (EBCT) of 50 min. The BAC bed, operated in downstream mode, was maintained submerged by a control valve placed at the outlet of the filter. The backwash of the BAC bed filter was controlled on-line when reaching a head loss threshold in the filter. The secondary effluent was pumped into the fixed bed BAC filter in a downstream configuration with a flow rate of 48 m^3/day. The BAC filter operating conditions are reported in Table S3. Taking into account this influent flow, one day of operation