Journal of Environmental Management 193 (2017) 491–502

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Pilot-scale removal of pharmaceuticals in municipal wastewater: Comparison of granular and powdered activated carbon treatment at three wastewater treatment plants

Victor Kårelid*, Gen Larsson, Berndt Björlenius

Division of Industrial Biotechnology, KTH Royal Institute of Technology, AlbaNova University Center, SE-106 91 Stockholm, Sweden

ARTICLE INFO

Article history:
Received 17 October 2016
Received in revised form
15 February 2017
Accepted 16 February 2017
Available online 28 February 2017

Keywords:
Advanced wastewater treatment
Adsorption
GAC
PAC
Pharmaceutical removal

ABSTRACT

Adsorption with activated carbon is widely suggested as an option for the removal of organic micropollutants including pharmaceutically active compounds (PhACs) in wastewater. In this study adsorption with granular activated carbon (GAC) and powdered activated carbon (PAC) was analyzed and compared in parallel operation at three Swedish wastewater treatment plants with the goal to achieve a 95% PhAC removal. Initially, mapping of the prevalence of over 100 substances was performed at each plant and due to low concentrations a final 22 were selected for further evaluation. These include carbamazepine, clarithromycin and diclofenac, which currently are discussed for regulation internationally. A number of commercially available activated carbon products were initially screened using effluent wastewater. Of these, a reduced set was selected based on adsorption characteristics and cost. Experiments designed with the selected carbons in pilot-scale showed that most products could indeed remove PhACs to the target level, both on total and individual basis. In a setup using internal recirculation the PAC system achieved a 95% removal applying a fresh dose of 15–20 mg/L, while carbon usage rates for the GAC application were much broader and ranged from <28 to 230 mg/L depending on the carbon product. The performance of the PAC products generally gave better results for individual PhACs in regards to carbon availability. All carbon products showed a specific adsorption for a specific PhAC meaning that knowledge of the target pollutants must be acquired before successful design of a treatment system. In spite of different configurations and operating conditions of the different wastewater treatment plants no considerable differences regarding pharmaceutical removal were observed.

1. Introduction

The removal of organic micropollutants within wastewater treatment has become a matter of urgent concern during recent years. This is a result of comprehensive research on the effects that environmentally relevant concentrations may pose on living species occupying the receiving waters. Residues of pharmaceutically active compounds (PhACs) are arguably of specific concern since they primarily end up in the municipal wastewater treatment plants (WWTPs) where a large share passes relatively undisturbed through the treatment processes (Joss et al., 2006). Adverse effects in fish have previously been reported e.g. on organ tissue (Hoeger et al., 2005) and gene expression (Cuklev et al., 2011) caused by diclofenac and on natural behavior (Brodin et al., 2013) caused by oxazepam. Moreover, the emergence of antibiotic resistance presents a matter of global concern (Larsson, 2014).

During the quest to achieve a reliable and efficient removal of organic micropollutants in general, adsorption onto activated carbon and chemical transformation with ozone have emerged as the main alternatives, and a few pioneering studies in pilot and full scale have been performed on effluent wastewater proving the efficacy (Boehler et al., 2012; Mailler et al., 2015; Margot et al., 2013; Meinel et al., 2015). These convincing results suggest that adsorption is a first choice for high-level removal and is favorable compared to oxidation by e.g. ozone. The presentation of activated carbon in purification systems is however subjected to specific constraints. Conventionally, adsorption is performed using either the granular (GAC) or the powdered (PAC) form. Promising results regarding adsorption of organic compounds, e.g. dyes and

* Corresponding author.
E-mail addresses: karelid@kth.se (V. Kårelid), gen@biotech.kth.se (G. Larsson), berndtb@kth.se (B. Björlenius).

http://dx.doi.org/10.1016/j.jenvman.2017.02.042
0301-4797/

surfactants have also been observed using activated carbon cloths (Ayranci and Duman, 2009; Duman and Ayranci, 2010). GAC is contained in a column, which facilitates regeneration and reactivation while specific measures are required for PAC. On the other hand, PAC has a larger available specific surface area resulting in faster adsorption kinetics (Meinel et al., 2015). The stationary application of GAC ensures that saturation of the carbon can be reached, while for PAC applications this can generally only be achieved through internal recirculation due to the long contact times required to reach adsorption equilibrium (Meinel et al., 2015; Nowotny et al., 2007). Previous research has shown that a comparable removal of organic micropollutants has been achieved using separation and recirculation with significant reductions of the PAC dose in both bench-scale (Meinel et al., 2016) and in pilot and full scale (Boehler et al., 2012). In Sweden, however, the desire to recycle sludge for agricultural purposes restricts the direct dosing of PAC into the biological treatment step and forces the design of a final, separate process. The choice between adsorption with GAC and PAC currently remains uncertain due to the advantages of the respective methods, thus invoking a demand for an increased knowledge base before widespread full-scale implementation and for site-specific guidance.

The aim of this study was to achieve a 95% removal, in relation to the effluent, of a selected set of 22 PhACs frequently occurring in municipal wastewater of representative Swedish WWTPs through adsorption with activated carbon. This degree of removal is chosen to surpass future effluent quality standard requirements by a safe margin. To appropriately target the removal of these substances, sets of activated carbons were selected through screening where a relevant load of PhACs was determined after mapping the effluent wastewater with respect to over 100 substances. The initial selection covered a broad range of adsorption specific properties, e.g. specific surface area, iodine number and particle size. To account for site-specific conditions three different WWTPs were chosen for evaluation. These plants differed regarding wastewater characteristics and plant configuration to give a diverse representation of Swedish wastewater effluents. To accommodate operation at different locations a mobile pilot plant, specifically designed for comparison of PAC and GAC purification performance, was constructed.

2. Materials and methods

2.1. Mobile pilot plant

A mobile pilot plant was constructed in-house for application at the three WWTPs. Treatment tanks, and equipment for sampling and process control were installed into a 20-foot shipping container allowing for operation at outdoor temperatures down to freezing. The temperature of the wastewater treated in the pilot plant varied with the seasons according to the WWTP effluent temperature. The pilot plant consisted of eleven treatment lines; three designed for GAC application, another three for PAC, two ozonation lines, two lines using biofilm (MBBR) and finally one line with sand filtration after ozonation. This work, however, only covers operation with the GAC and PAC treatment lines. During operation, effluent wastewater was pumped by a submerged impeller pump via coarse particle filters (2 mm perforation) to a leveling tank located outside the container. The PAC and GAC lines were fed from the leveling tank by separate screw pumps.

GAC filtration was performed in two identical treatment lines. The design was based on both literature (Tchobanoglous et al., 2003) and previous in-house tests (Wahlberg et al., 2010). Each line consisted of two stainless steel tubes of 2 m height in series and an inner diameter of 0.15 m, corresponding to a horizontal filter area of 0.018 m^2 and an operational volume for each line of ~60 L. The filters were filled with 1 m of GAC supported by filter bottoms with four nozzles in each filter. The operation was performed in down-flow configuration. A fixed, but adjustable, water level was kept in the GAC filter by a control valve connected beneath the filter bottom in each column. The GAC filters were supplied with manual valves to enable backwashing with tap water.

PAC treatment was likewise performed using two identical treatment lines. The design of the PAC treatment system was adapted from previous technical studies, primarily Metzger and Kapp (2008) and Abegglen and Siegrist (2012). Each line consisted of an initial 1.7 L tank for mixing of effluent wastewater and dosed PAC, followed by three sequential aerated contact tanks, a sedimentation tank and a final sand filter. After exiting the third contact tank, wastewater enters the sedimentation tank 0.7 m below the surface level. The five main tanks were all made of stainless steel, while the mixing tank was made from plastic filter housing. Each contact tank was 0.9 m in height (the operating water level was 0.70–0.75 m) with an inner diameter of 0.25 m, amounting to a total operating volume of ~100 L. The sedimentation tank measured 1.46 m in height (of which the bottom cone height is 0.46 m) and had an inner diameter of 0.5 m (operating volume of ~180 L). The sand filter tank was 1.75 m in height with an inner diameter of 0.25 m (empty bed volume of 66 L) supported by a filter bottom with four nozzles. The sand filter was composed of two different media: a bottom layer constituting one third of the volume of filter sand (1.2–2 mm particle size, Rådasand AB) and a top layer constituting two thirds of the volume of Filtralite MC 2.5–4 (2–4 mm particle size, Saint Gobain Byggevarer AS). The total sand filter height was maintained at 0.8–1.0 m during the experiments. Recirculation was accomplished by pumping with an airlift pump from the bottom of the sedimentation tank back to the first contact tank. Schematic setups of the treatment systems are presented in Fig. 1.

2.2. Selected wastewater treatment plants

Three plants were chosen due to considerable differences in attached populations, disparities between their tertiary treatment processes, as well as differences regarding treatment load such as the total hydraulic retention time (HRT) and sludge loading rate.

Käppalaverket (Käppala) is the second largest WWTP in the Stockholm region and treats 149 000 m^3 wastewater per day, corresponding to 425 000 population equivalents. The treatment consists of pre-treatment (screening and grit removal), primary sedimentation, biological treatment and sand filtration. Two thirds of the wastewater is treated in a conventional activated sludge pre-denitrification setup using simultaneous chemical precipitation of phosphorous with ferrous sulfate. One third of the wastewater is treated in the UCT setup (University of Cape Town; Ekama et al., 1983), which allows for enhanced biological phosphorous removal.

Kungsängsverket (Uppsala) treats 50 000 m^3 wastewater per day, corresponding to 148 000 population equivalents. The treatment consists of pre-treatment (screening and grit removal), primary sedimentation, biological treatment, concluding with flocculation and lamella separation. Approximately 40% of the wastewater is treated in a conventional activated sludge pre-denitrification setup, while the remaining 60% is treated using activated sludge in a step-feed pre-denitrification setup. Both pre- and post-precipitation are used to achieve phosphorous removal through addition of ferric chloride. In Uppsala, the mobile pilot plant was extended with a pretreatment step in the form of two shallow sand filter lines, after early observations of a fast clogging process in the upper surfaces of the GAC filters. The filters were installed before the leveling tank in the pilot plant.