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Research article

Effects of recirculation in a three-tank pilot-scale system for pharmaceutical removal with powdered activated carbon

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ABSTRACT

The removal of pharmaceutically active compounds by powdered activated carbon (PAC) in municipal wastewater is a promising solution to the problem of polluted recipient waters. Today, an efficient design strategy is however lacking with regard to high-level overall, and specific, substance removal in the large scale. The performance of PAC-based removal of pharmaceuticals was studied in pilot-scale with respect to the critical parameters; contact time and PAC dose using one PAC product selected by screening in bench-scale. The goal was a minimum of 95% removal of the pharmaceuticals present in the evaluated municipal wastewater. A set of 21 pharmaceuticals was selected from an initial 100 due to their high occurrence in the effluent water of two selected wastewater treatment plants (WWTPs) in Sweden, whereof candidates discussed for future EU regulation directives were included. By using recirculation of PAC over a treatment system using three sequential contact tanks, a combination of the benefits of powdered and granular carbon performance was achieved. The treatment system was designed so that recirculation could be introduced to any of the three tanks to investigate the effect of recirculation on the adsorption performance. This was compared to use of the setup, but without recirculation. A higher degree of pharmaceutical removal was achieved in all recirculation setups, both overall and with respect to specific substances, as compared to without recirculation. Recirculation was tested with nominal contact times of 30, 60 and 120 min and the goal of 95% removal could be achieved already at the shortest contact times at a PAC dose of 10–15 mg/L. In particular, the overall removal could be increased even to 97% and 99%, at 60 and 120 min, respectively, when the recirculation point was the first tank. Recirculation of PAC to either the first or the second contact tank proved to be comparable, while a slightly lower performance was observed with recirculation to the third tank. With regards to individual substances, clarithromycin and diclofenac were ubiquitously removed according to the set goal and in contrast, a few substances (fluconazole, irbesartan, memantine and venlafaxine) required specific settings to reach an acceptable removal.

1. Introduction

The removal of pharmaceutically active compounds (PhACs) from wastewater has been of increasing concern and a focus for research and development during the last decade. For treatment in municipal wastewater treatment plants (WWTPs), this is mainly because conventional unit operations remove these substances inefficiently (Joss et al., 2006) and furthermore, environmentally relevant concentrations of certain PhACs, e.g. diclofenac and oxazepam, might negatively affect a range of aquatic species (Brodin et al., 2013; Hoeger et al., 2005). While discharge concentrations will largely depend on several factors such as the consumption by the attached population, the degree of degradation within the WWTP and the dilution factor of the final effluent, measures have been sought to reduce this inevitable pollution for future sustainability.

Several technologies have been evaluated to counter this issue. Promising results have e.g. been achieved with nanofiltration and reverse osmosis (Snyder et al., 2007; Vergili, 2013), however not with the same cost efficiency as the current main alternatives; oxidation with ozone and activated carbon adsorption (Joss et al., 2008), which have been deemed ready for full scale implementation (Hollender et al., 2009; Mailler et al., 2015; Margot et al., 2013).

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Neither of these two methods are however without drawbacks. The total energy demand for application with activated carbon has been suggested to be considerably higher (Mousel et al., 2017) while ozonation, on the other hand, can lead to oxidation products with uncertain effects (Stalter et al., 2010).

The efficacy of activated carbon is generally depending both on the contact time, which needs to be designed to satisfy the kinetic limitations for adsorption of a particular pollutant, and furthermore on the available adsorption sites, i.e. the amount of carbon in the system. Both can easily be manipulated. Adsorption using different carbon varieties has frequently been discussed in the literature, where the use of powdered activated carbon (PAC) has been suggested to give better results with regard to carbon consumption as compared to the granular form (GAC) mainly due to the larger surface area of the finer grains (Meinel et al., 2015). Furthermore, the stationary setup of GAC filtration naturally saturates the adsorption sites over time and if PAC is the preferred adsorbent, similar conditions can be achieved by the intentional use of an extended carbon retention time. This can be actualized e.g. in a fluidized bed (Mailler et al., 2015) or by use of PAC recirculation over the contact zone (Meinel et al., 2016a, 2016b). Pre-embedding of PAC on existing tertiary filters has also been suggested to minimize the need for constructional change (Hu et al., 2016). Recirculation can principally be applied in two different configurations i.e. either over one single contact tank or with a set of sequential contact tanks (cf. tanks-in-series modeling). Examples of the latter can be found in the literature but the application was here mainly for facilitation of PAC separation by flocculation (Abegglen and Siegrist, 2012; Metzger and Kapp, 2008). The design and efficient operation of treatment with PAC is thus presently under considerable development. However, few conclusive studies on pilot and full-scale performance are yet available to guide the design for very high-level removal of PhACs.

In the present study, we chose to use three consecutive aerated contact tanks, partly to avoid the escape of untreated water, which poses a risk during short retention times in a single tank, but partly to provide a larger design space, with the possibility of using different entry points for the PAC recirculation. Apart from the recirculation point, the contact time and the PAC dose were chosen as critical parameters in this study, where the aim was a minimum of 95% PhAC removal from municipal wastewater.

The pilot experiments were preceded by bench-scale experiments, screening different PAC products, to guide the operational design for pilot-scale. Over one hundred pharmaceuticals were initially monitored before a set of twenty-one was finally selected for further studies due to their high occurrence in the effluent of the selected treatment plants.

2. Materials and methods

2.1. Site description

Pilot and bench-scale experiments were performed during 2015 at Käppalaverket (Käppala), the second largest WWTP in the Stockholm region. The plant treats 167 000 m^3 wastewater each day, corresponding to 440 000 population equivalents and the treatment is located in temperature controlled caverns. 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.

Bench-scale experiments were also performed at Kungsängens WWTP (Västerås) which treats 50 000 m^3 wastewater per day, corresponding to 105 000 population equivalents. Most of the treatment is located outdoors and consists of pre-treatment (screening and grit removal), primary sedimentation and biological treatment. The wastewater is treated in a conventional activated sludge pre-denitrification setup, where methanol and ethylene glycol are used as carbon sources to improve the nitrogen removal process. Pre-precipitation is used to achieve phosphorous removal through addition of ferrous sulfate. Polymeric coagulants are added to the secondary sedimentation tanks to improve particle separation before the final effluent.

2.2. Pharmaceuticals

The selection of PhACs started with a set of over 100 substances that has shown high potency and potential for bioaccumulation in fish according to (Grabic et al., 2012). A subset of these was then selected based on 50% or higher occurrence in the effluent wastewater at Käppala. The properties of the 21 substances that were selected are given in the supplementary material – Table S1. Data is presented as either overall removal, i.e. for the sum of substances or as individual substance removal. In case of detection below the limit of quantification (LOQ), the concentration was set to LOQ/2. For the overall removal, PhACs that were observed below 5xLOQ in the effluent sample were excluded, due to the high analytical uncertainty in this range (Martin Ruel et al., 2011). This coincided with detection below the LOQ in the treated samples. Removal rates were denoted as sufficient or adequate in the case they reached either 95% or the detected concentration in the treated sample was below the LOQ.

2.3. Bench-scale experiment setup

Wastewater for the bench scale experiments was collected intermittently and pooled for 30 min from the final effluent. Experiments were performed in Erlenmeyer flasks with a liquid volume of 1 L into which dry PAC, carefully weighed on an analytical balance (Sartorius) to achieve the correct dose, was dispensed. Mixing was maintained through aeration. After the desired contact time was reached, samples were immediately filtered to remove residual PAC with 0.45 μm cellulose nitrate filters (Whatman). Temperature, pH and conductivity of the treated wastewater were recorded before and after each experiment (average values are presented in Table S2). The samples were frozen before PhAC analysis.

Dose response experiments were performed in Käppala with Aquasorb 5000P and Aquasorb MP20 (both Jacobi carbons, Table S3) with PAC doses of 2.5, 5, 10, 15 and 20 mg/L at 30 min contact time. Experiments where the contact time was varied were performed in Västerås and Käppala with Aquasorb 5000P at 30, 60 and 120 min with a PAC dose of 10 mg/L.

2.4. Pilot plant setup and operation

Pilot-scale operation was performed in three parallel lines, each consisting of an initial mixing tank, three sequential contact tanks, a sedimentation tank and a concluding sand filter (Fig. 1). The setup with three consecutive tanks was adapted from previous technical reports, which however used PAC separation by flocculation and sedimentation (Abegglen and Siegrist, 2012; Metzger and Kapp, 2008). Recirculation of PAC was accomplished by pumping with an airlift pump from the bottom of the sedimentation tank back to the first, second or third contact tank depending on the experiment. These recirculation configurations are denoted R1, R2 and R3,