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How to dose powdered activated carbon in deep bed filtration for efficient micropollutant removal

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ABSTRACT

Direct addition of powdered activated carbon (PAC) to the inlet of a deep bed filter represents an energy- and space-saving option to remove organic micropollutants (OMPs) during advanced wastewater treatment or drinking water purification. In this lab-scale study, continuous dosing, preconditioning a filter with PAC and combinations thereof were investigated as possible dosing modes with respect to OMP adsorption efficiency. Continuous dosing resulted in decreasing effluent concentrations with increasing filter runtime due to adsorption onto accumulating PAC in the filter bed. Approximately constant removal levels were achieved at longer filter runtimes, which were mainly determined by the dose of fresh PAC, rather than the total PAC amount embedded. The highest effluent concentrations were observed during the initial filtration stage. Meanwhile, preconditioning led to complete OMP adsorption at the beginning of filtration and subsequent gradual OMP breakthrough. PAC distribution in the pumice filter was determined by the loss on ignition of PAC and pumice and was shown to be relevant for adsorption efficiency. Preconditioning with turbulent upflow led to a homogenous PAC distribution and improved OMP adsorption significantly. Combining partial preconditioning and continuous dosing led to low initial effluent concentrations, but ultimately achieved concentrations similar to filter runs without preconditioning. Furthermore, a dosing stop prior to the end of filtration was suitable to increase PAC efficiency without affecting overall OMP removals.

1. Introduction

Increasing numbers of organic micropollutants (OMPs), such as pharmaceuticals or industrial chemicals, have been detected in the aquatic environment in recent years (Putschew et al., 2000; Reemtsma et al., 2010). While the overall effects of life-long exposures to a mixture of OMPs are still being assessed (Leusch et al., 2014; Magdeburg et al., 2014), negative effects on aquatic life have been reported for individual compounds at environmentally relevant concentrations (Melnyk-Lamont et al., 2014; Pomati et al., 2006). Conventional wastewater treatment plants (WWTPs) do not serve as an effective barrier for many common OMPs and thus represent a major source of OMP entry into receiving water bodies (Loos et al., 2013; Michael et al., 2013). Therefore, advanced treatment options, such as adsorption of OMPs onto powdered activated carbon (PAC), are currently being investigated as preventive measures (Kovalova et al., 2013; Margot et al., 2013).

Application of PAC typically consists of addition to an adsorption stage with subsequent separation by deep bed or

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membrane filtration. A less energy and space consuming alternative is the direct dosing of PAC to the influent of a filtration unit (Böhler et al., 2012). In a study investigating dosing strategies in a PAC/membrane filtration system, Hoffmann and Hobby (2013) observed improved OMP removal if PAC was added in shorter dosing intervals at the beginning of filtration. Elimination was highest when PAC was dosed as a single-pulse within the first minute of filtration. Similarly, Campos et al. (2000a, b) developed models for step and pulse addition of PAC to a membrane filtration process. Validation experiments confirmed that dosing the entire PAC at the beginning results in lower cycle-averaged effluent concentrations compared to continuous PAC addition. However, those findings may not be applicable to PAC dosing to a deep bed filter, as comparably short membrane filtration cycles (30–120 min in the studies mentioned) greatly favor single-pulse addition at the beginning of filtration, because the PAC capacity is usually not exhausted during the filtration time.

In combination with deep bed filtration, PAC is continuously added to the filter influent, leading to increased PAC accumulation within the filter bed with increasing filter runtime until backwashing. As contact times are typically not sufficient to exhaust the adsorption capacity of freshly dosed PAC before separation, embedded PAC will still remove OMPs. Accordingly, improving OMP removals with increasing filter runtime have been observed in pilot-scale PAC filtration (Altmann et al., 2014). Continuous PAC addition leads to an average PAC retention time in the filter bed of half the total filtration cycle. Alternatively, dosing a higher amount or the entire PAC mass at the beginning of filtration increases its retention time and thus may enhance PAC efficiency. Haist-Gulde and Baldauf (2007) tested a dosing sequence consisting of high PAC doses at the beginning of the filtration cycle and no PAC addition towards the end instead of constant volume proportional dosing. Results showed a considerable improvement of OMP removal, particularly during the initial filtration stage. Similarly, Haberer and Normann-Schmidt (1991) embedded PAC into filter material made of foamed polystyrene beads in a pretreatment step with subsequent filtration without additional PAC dosing. Besides the total PAC amount embedded, the PAC distribution within the filter bed was determined to be relevant for efficient OMP removal.

Adsorption capacity may be significantly depleted during typical filter runtimes of deep bed filters (usually >12 h), necessitating addition of fresh PAC. Ruhl et al. (2014) investigated preconditioning of a deep bed filter with PAC for treatment of WWTP effluent. While PAC efficiency was higher than in comparable batch adsorption experiments, complete breakthrough of weakly adsorbing OMPs due to PAC exhaustion was observed after 24 h of filtration.

The objective of this study was to better understand the adsorption of OMPs in a PAC/filtration system. The influence of PAC accumulation during continuous dosing and its significance for overall OMP removal in comparison to adsorption onto fresh PAC was assessed. OMP adsorption in a preconditioned PAC filter was characterized and the influence of contact time in the filter bed was investigated. Based on the results, optimization of PAC utilization by partial preconditioning with subsequent continuous dosing and a dosing stop before the end of the filter run were tested.

2. Materials and methods

2.1. Experiments

A glass tube of 150 cm length and an inner diameter of 2.2 cm filled with 280 g pumice (grain size 1.5–2.5 mm, 102–105 cm bed height) was used as a bench-scale filter. Before each experiment, virgin pumice was moistened with deionized water and placed in a vacuum unit for 5 min to remove residual gas from the inner pores, filled into the tube and subsequently backwashed with deionized water for at least two days to wash out the fine pumice fraction.

The experiments were conducted using Berlin tap water (≈5 mg/L dissolved organic carbon (DOC), pH 7.5–7.7, conductivity 600–640 μS/cm), which was spiked with 1 μg/L each of acesulfame, benzotriazole, bezafibrate, carbamazepine, diclofenac, 4-formyl-aminoantipyrine (FAA), methyl-benzotriazole, primidone and sulfamethoxazole (additional information on area of application, molecular mass and log D value of the OMPs are given in Table S1 in the Supporting Information). The OMPs were chosen based on their permanent occurrence in elevated concentrations in Berlin WWTP effluents, due to their consideration as priority substances in the European water framework directive and for comparison purposes because they are frequently considered in similar studies. Stock suspensions of PAC Carbopal AP (Donau Carbon, Germany, d_{70} < 40 μm, B.E.T. surface >1300 m^2/g according to manufacturer) were prepared in deionized water and treated in an ultrasonic bath for 15 min for full wetting of the PAC. The PAC was chosen based on high removals of a wide range of OMPs in preliminary tests (Zietzschmann et al., 2014).

The filter was fed in downflow direction with a filtration rate of 7 m/h (44 mL/min) for 24 h. Two basic modes of PAC dosing were applied: a) continuous addition of PAC and b) downflow and upflow single-pulse preconditioning with subsequent filtration. For continuous dosing, 25 L stock suspension (6, 12, 26 and 29 mg/L PAC) was continuously stirred and pumped to the filter inlet at a constant flow rate of 11 mL/min for PAC doses of 1.5, 3.0, 6.5 and 7.2 mg/L. The OMP concentrations were adjusted accordingly to compensate for the dilution. In downflow preconditioning experiments, 400 mg PAC was suspended in 200 mL deionized water and added directly to the filter inlet at a preconditioning filtration velocity of 7 or 15 m/h. For turbulent upflow conditioning, a stirred stock suspension (2 g/L) was pumped with a high velocity to expand the filter bed until PAC was visible at the filter outlet. Washed out PAC was reapplied to the top of the filter in downflow mode. The remaining PAC in the stock suspension and in the collected effluent was quantified to calculate the embedded PAC mass. Following preconditioning, 4 × 500 mL of deionized water was filtrated and collected to quantify PAC releases.

In additional experiments, 50 and 100 mg PAC were added as a single-pulse in a preconditioning step with subsequent continuous PAC dosing during the filter run. The experimental procedure was similar to the approach for each individual dosing method. In further experiments, continuous PAC dosing was turned off before the end of filtration. The PAC