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Biodegradation of persistent organics can overcome adsorption–desorption hysteresis in biological activated carbon systems

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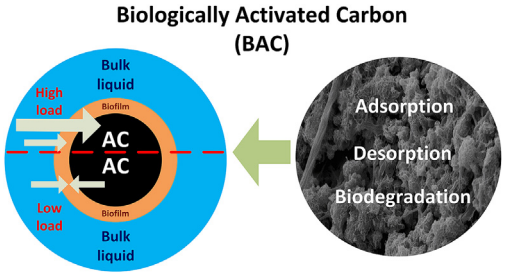
HIGHLIGHTS

- It was studied how persistent organics are removed by Biological Activated Carbon.
- Adsorption–desorption hysteresis limits activated carbon bioregeneration.
- Acetate enhanced the biodestruction of the persistent pharmaceutical metoprolol.
- Metoprolol can be biodegraded to below 0.08 μg/L, which can overcome hysteresis.

GRAPHICAL ABSTRACT



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ABSTRACT

In Biological Activated Carbon (BAC) systems, persistent organic pollutants can be removed through a combination of adsorption, desorption and biodegradation. These processes might be affected by the presence of other organics, especially by the more abundant easily-biodegradable organics, like acetate. In this research these relations are quantified for the removal of the persistent pharmaceutical metoprolol.

Acetate did not affect the adsorption and desorption of metoprolol, but it did greatly enhance the metoprolol biodegradation. At least part of the BAC biomass growing on acetate was also able to metabolise metoprolol, although metoprolol was only converted after the acetate was depleted. The presence of easily-degradable organics like acetate in the feeding water is therefore beneficial for the removal of metoprolol in BAC systems.

The isotherms obtained from metoprolol adsorption and desorption experiments showed that BAC systems are subject to hysteresis; for AC bioregeneration to take place the microbial biomass has to reduce the concentration at the AC-biomass interface 2.7 times compared to the concentration at which the carbon was being loaded. However, given the threshold concentration of the MET degrading microorganisms (<0.08 μg/L) versus the average influent concentration (1.3 μg/L), bioregeneration is feasible.

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1. Introduction

Biological Activated Carbon (BAC) systems are applied as post-treatment during wastewater treatment and as pre-treatment during the production of potable or ultrapure water (Aktas and Cecen, 2007). Typically BAC systems are filters loaded with granular activated carbon (GAC) that also contain biomass (Seredynska-Sobecka et al., 2006). The GAC serves as porous carrier material to which bacteria can attach; the AC granules can be completely covered with a biofilm, although part of the biomass can also be present as flocks suspended in the bulk liquid. Pollutants are removed by both adsorption to the GAC and biodegradation by the biomass (Xiaojin et al., 1991). BAC systems provide the same effluent quality as abiotic AC systems, but due to the contribution of the biomass to the removal the lifetime of the adsorbent is longer. The biomass is thought to even regenerate the AC, due to the simultaneous desorption and biodegradation (Aktas and Cecen, 2007).

An advantage of BAC over activated sludge systems is the ability to cope with peak loads, since the AC can relatively quickly take-up large amounts of pollutants (Cha et al., 1998). After a peak load the AC could be regenerated over time due to simultaneous desorption and biodegradation (Ha and Vinitnantharat, 2000). Furthermore, BAC systems are relative resistant to toxic compounds, since the AC reduces the concentration of those compounds to non-toxic levels (Chevallier et al., 1984; Ng and Stenstrom, 1987; Kohler et al., 2006). A third advantage of BAC systems is that the relative long solid retention times allows microorganisms degrading persistent organics to maintain themselves even at low growth rates (Aktas and Cecen, 2007).

A limiting factor for BAC is the low bioavailability of persistent organics due to the adsorption to AC (Guerin and Boyd, 1997). In addition, the slow-growing persistent-organic degrading microorganisms might be overgrown by faster-growing organisms that only utilize the easily-degradable organics. Usually the concentrations of the persistent organic metoprolol in secondary effluent of domestic wastewater treatment plants varies from 0.019 to 1.7 μg/L, while the BOD (biochemical oxygen demand) is usually between 1.7 and 7.7 mg/L (Beheerm, 2010; Deblonde et al., 2011). Both adsorption and biodegradation of persistent organics might be affected by the presence of other organics. In the case of adsorption, this may be due to the competition for adsorptive sites on the AC (Pelekani and Snoeyink, 1999; Ha and Vinitnantharat, 2000).

In the case of biodegradation, both competitive inhibition and co-metabolism may play a role. There will be competitive inhibition when one enzymatic pathway is involved in the conversion of multiple compounds. Due to a difference in reaction rate and concentration, persistent compounds may only be metabolised after the depletion of other substrates. However, in literature the bioconversion of persistent organics is often linked to co-metabolism (Tran et al., 2013). During co-metabolism, the degradation of a secondary (more persistent) substrate depends on the simultaneous degradation of a primary substrate. The metabolism of a secondary substrate may be a detoxify mechanism or an artefact of the enzymatic system, and not allow for energy conservation by the microorganisms (Wey et al., 2009; Liu et al., 2013). In this way, easy biodegradable organic compounds (eg. amino acids, glucose) stimulate the biodegradation of persistent organics (Hess et al., 1993; Zhong et al., 2007). For bioregeneration of AC in BAC systems to occur, the persistent organics have to be available for microorganisms, which is not the case for the fraction that is adsorbed to the AC (Guerin and Boyd, 1997; Ha and Vinitnantharat, 2000; Aktas and Cecen, 2006). Thus, the bioavailability depends on the relation between the dissolved and adsorbed persistent organics; e.g. hydrophobic, polar, less soluble, long chain organics adsorb stronger than hydrophilic, non-polar organic compounds (Ridder et al., 2009; Kovalova et al., 2013). This relation, described by an isotherm, is different for sorption and desorption, which causes so-called adsorption–desorption hysteresis (Toro and Horzempa, 1982; Ma et al., 1993; Huang and Weber, 1997). Therefore, the concentration at which desorption takes place will be lower than the concentration at which the AC is loaded. Thus, for AC regeneration to occur, the persistent organic concentration has to be reduced below the minimal concentration achieved by adsorption. This might be possible through biodegradation by the microbial biomass growing in BAC systems. However, to the best of our knowledge it was never been investigated if the BAC biomass can indeed overcome the adsorption–desorption hysteresis. This study will therefore investigate whether, despite the adsorption–desorption hysteresis, activated carbon regeneration can take place in BAC systems.

To do that, the adsorption–desorption hysteresis and biodegradation for the persistent pharmaceutical metoprolol, in presence of different acetate concentrations, were quantified. Metoprolol (MET) was used as model compound for persistent organics as it was the most dominant pharmaceutical found in the influent of the full-scale BAC filter from which the biomass was obtained. Acetate (ACET) was used as model compound for easily degradable organic matter, as it is a major intermediate in the biodegradation process of complex organic matter.

2. Material and methods

2.1. Buffer and medium solution

The adsorption experiments, desorption experiments and biomass washing were done in Phosphate Buffered Saline (PBS) containing 800 mg/L NaCl, 20 mg/L KCl, 144 mg/L Na_2HPO_4 and 24 mg/L KH_2PO_4. The pH was checked at the start, during and at the end of each experiment, in all experiments where PBS was used the pH remained between 6.7 and 7.4.

Biodegradation experiments were done with PBS supplemented with nutrients: 170 mg/L NH_4Cl, 8 mg/L $CaCl_2 \cdot 2H_2O$, 9 mg/L $MgSO_4 \cdot 7H_2O$ and 1 mL/L of trace element's solution. The trace element solution contained 2000 mg/L $FeCl_3 \cdot 4H_2O$, 2000 mg/L $CoCl_2 \cdot 6H_2O$, 500 mg/L $MnCl_2 \cdot 4H_2O$, 30 mg/L $CuCl_2 \cdot 2H_2O$, 50 mg/L $ZnCl_2$, 50 mg/L H_3BO_3, 90 mg/L $(NH_4)_6Mo_7O_{24} \cdot 4H_2O$, 100 mg/L $Na_2SeO_3 \cdot 5H_2O$ and 50 mg/L $NiCl_2 \cdot 6H_2O$ (Mrafkova et al., 2003).

2.2. Full-scale BAC filter

Biomass was obtained from the high-loaded BAC filters at the Puurwaterfabriek (Nieuw Amsterdam, the Netherlands). The Puurwaterfabriek produces ultrapure water for oil extraction from municipal wastewater treatment plant effluent. The Puurwaterfabriek comprises the following subsequent treatment steps: sieving, ultrafiltration, high-loaded BAC filter, low-loaded BAC filter, Reserve Osmosis (RO) and Electro Deionisation. The main function of the BAC filters was to remove fouling precursors for RO.

2.3. Biomass

The full-scale BAC filter was sampled during the periodic backwashing. Subsequently, the bulk of the biomass was detached from the activated carbon (AC) particles by severe agitation and filtration with a 215 μm sieve. The biomass was washed twice and