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Removal of acidic pharmaceuticals within a nitrifying recirculating biofilter

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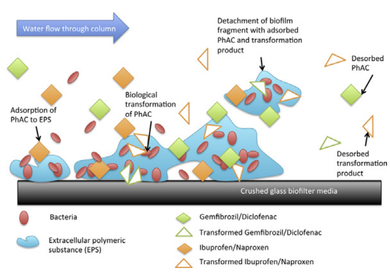
HIGHLIGHTS

- Ibuprofen and naproxen were mainly removed through biological transformation.
- Main removal mechanism for gemfibrozil and diclofenac was adsorption to the biofilm.
- Recirculating biofilters were robust, repeatable and able to recover from major system upsets.

GRAPHICAL ABSTRACT



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ABSTRACT

The fate of pharmaceutically active compounds (PhACs) in wastewater treatment systems is an area of increasing concern. Little research has been done to understand this issue in rural or decentralized communities. The objective of this research was to examine the ability of a bench scale nitrifying recirculating biofilter (RBF) to remove four acidic PhACs: gemfibrozil, naproxen, ibuprofen and diclofenac from secondary treated municipal wastewater at concentrations of 20 and 200 μg/L. The average removals in this study were between 92 and 99% for ibuprofen, 89 and 99% for naproxen, 62 and 92% for gemfibrozil and 40 and 76% for diclofenac, which is consistent with literature. Ibuprofen and naproxen were largely removed through biological transformation; whereas gemfibrozil and diclofenac showed more variable removal, likely due to both biological transformation and sorption processes. PhAC removal in the RBFs was repeatable between trials, robust and responsive to system upsets, and the presence of PhACs as a single compound versus mixtures had no impact on PhAC removal efficiency. In summary, this study indicates that RBFs as a nitrifying stage of a multi-stage filtration process could be a viable technology for removal of some acidic pharmaceuticals in small onsite wastewater treatment facilities.

1. Introduction

Approximately one-quarter of households in the United States utilize onsite wastewater treatment systems [1], the majority of which have both a septic tank and soil adsorption field [2,3]. Large footprint requirements for soil adsorption fields make traditional onsite systems a less desirable wastewater treatment option, and therefore alternative small wastewater treatment technologies have become an attractive option.

Recirculating biofilters (RBFs), a modification of intermittent sand filters, offer an alternative treatment approach with a smaller footprint requirement. According to the United States Environmental Protection Agency [4], RBFs are reliable, robust technologies resistant to flow variations that can be used for single households and small communities. Compared to single pass filters, recirculation of wastewater can provide improved wastewater effluent quality (biochemical oxygen demand (BOD), total suspended solids (TSS), ammonia) [5]. During RBF treatment, settled wastewater

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flows from a septic tank into a recirculation tank, where it is pumped to a distribution network and onto the surface of the filter bed. Traditionally sand has been used as the filter media; however, crushed glass, geotextile and peat have also been used successfully [2,6]. Wastewater percolates through the filter bed and is gathered as effluent through a drain collection system. A portion of this filter effluent is pumped back to the recirculation tank and the remainder is discharged from the treatment system. Feeding frequency onto the filter bed is usually 1–3 times per hour and typical recirculating ratios are between 3:1 and 5:1 [4]. RBFs can either be used as a single stage filter to provide bulk organics removal, or could be used as the latter part of a multi-stage filtration system to provide increased ammonia removal through nitrification.

Microorganisms form a biofilm on the surface of the filter media in a RBF, where they absorb soluble and colloidal waste material (organic material and nutrients) in the wastewater. Generally, RBFs can reduce BOD and TSS to less than 10 mg/L and they also promote nitrification through long solids retention times (SRT) that enable growth of ammonia oxidizing bacteria (AOB) [2,5,7], especially when operated as part of a multi-stage filter process.

Many studies report on the general occurrence and removal of pharmaceutically active compounds (PhACs) within centralized wastewater treatment plants [8–13]; however, the removal of PhACs through RBF treatment is not widely reported [1,14,15]. PhAC removal is highly dependent on many factors: the physical and chemical properties of the PhAC, the microbial ecology in the treatment system, the type of treatment, water quality (e.g. pH, temperature), and process conditions (e.g. flow rate, SRT, biomass concentration, hydraulic retention time, oxygen availability) [16–20].

In general, the mechanisms and removal pathways are not completely understood for each PhAC; however, it is generally accepted that biodegradation and sorption to wastewater biosolids are important removal processes [8,9,13]. It has been observed that wastewater treatment plants (WWTPs) operating under nitrifying conditions can lead to higher removal of PhACs, compared to WWTPs with no nitrification [12,16,21–23]. It has also been reported that aerobic conditions often result in higher PhAC removal efficiencies compared to anaerobic systems [1,15,16,24]. Biofilters have been shown to have increased removal efficiencies per unit biomass for certain PhACs compared to activated sludge systems [13,22,25].

The objective of this research was to examine the ability of bench scale RBF systems to remove gemfibrozil (a lipid-regulating drug), ibuprofen, naproxen and diclofenac (all non-steroidal anti-inflammatory drugs) under aerobic nitrifying conditions. There are numerous different PhACs representing different classes of compounds and the four acidic PhACs were chosen for this study based on their widespread use, the existing literature on the behavior of these pharmaceuticals in wastewater treatment systems [8,19,26], and the analytical capacity within the laboratory. Results for other classes of compounds may vary. The impacts of PhAC concentration, PhAC mixtures and system upset on PhAC removal in RBFs was investigated through two experimental trials using secondary treated municipal wastewater.

2. Materials and methods

2.1. RBF bench scale setup

RBF system setup consisted of a septic tank, a recirculation tank and a biofiltration column as shown in Fig. 1 and previously described by Hu and Gagnon [6]. The treatability trial consisted of six parallel recirculating biofilter units. The concentration trial consisted of four parallel recirculating biofilter units. The experimental conditions for each of these trials are described under experimental design. The septic tank, effluent tank and recirculation tank were all 150 mL Erlenmeyer flasks.

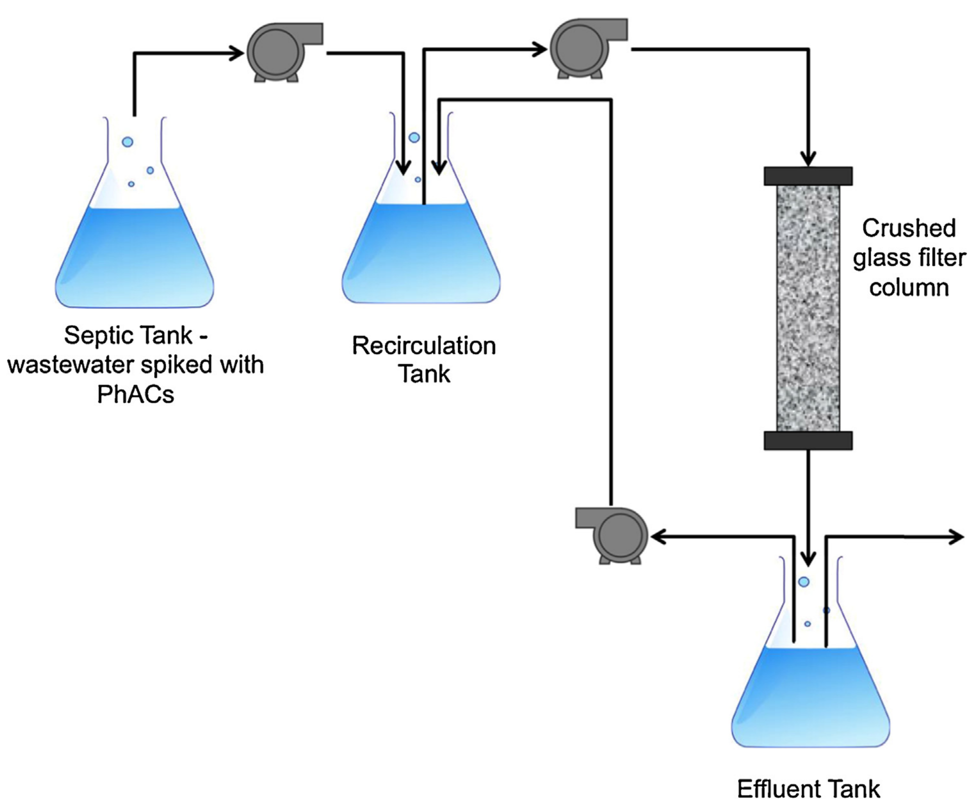

Fig. 1. Schematic of RBF setup, adapted from Hu and Gagnon [6].