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Research article

Impact of exogenous organic carbon on the removal of chemicals of concern in the high rate nitrifying trickling filters

Lei Mai [a], Ben van den Akker [a, b], Jun Du [c], Rai S. Kookana [c, d], Howard Fallowfield [a, *]

[a] Department of Environmental Health, School of Environment, Flinders University, Adelaide, South Australia, 5042, Australia
[b] Australian Water Quality Centre, Adelaide, South Australia, 5000, Australia
[c] Commonwealth Scientific and Industrial Research Organisation (CSIRO) Land and Water, Waite Campus, Glen Osmond, South Australia, 5064, Australia
[d] University of Adelaide, Waite Campus, Glen Osmond, South Australia, 5064, Australia

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ABSTRACT

The application of fixed bed high rate nitrifying trickling filters (NTFs) for the removal of track organic chemicals of concern (CoC) is less well known than their application to nutrient removal in water treatment. Particularly, the effect of exogenous organic carbon substrate (sucrose) loading on the performance of NTFs is not well understood. A laboratory-scale NTF system was operated in recirculation mode, with the objective of removing ammonia and CoC simultaneously. The efficiency of a high rate NTF for removal both of low concentration of ammonia (5 mg NH_4–N L^{-1}) and different concentrations of CoC in the presence of an exogenous organic carbon substrate (30 mg total organic carbon (TOC) L^{-1}) was investigated. In the presence of exogenous organic carbon, the results demonstrated that the high rate NTF was able to successfully remove most of the CoCs investigated, with the removal ranging from 20.2% to 87.54%. High removal efficiencies were observed for acetaminophen (87.54%), bisphenol A (86.60%), trimethoprim (86.24%) and 17α-ethynylestradiol (80.60%). It was followed by the medium removal efficiency for N, N-diethyl-m-toluamide (61.31%) and atrazine (56.90%). In contrast, the removal of caffeine (28.43%) and benzotriazole (20.20%) was poorer in the presence of exogenous organic carbon. The removal efficiency for CoC was also compared with the results obtained in our previous study in the absence of exogenous organic carbon. The results showed that the addition of exogenous organic carbon was able to improve the removal of some of the CoC. Significant TOC percentage removals (45.68%–84.43%) and ammonia removal rate (mean value of 0.44 mg NH_4–N L^{-1} h^{-1}) were also achieved in this study. The findings from this study provide valuable information for optimising the efficiency of high rate NTF for the removal of ammonia, CoC and TOC.

1. Introduction

The presence of track organic chemicals of concern (CoCs), for example surfactants, antibiotics, endocrine disrupting chemicals, in inadequately treated wastewaters discharged into the environment can be harmful to the aquatic organisms in surface waters (Benotti et al., 2009). This is because most of the conventional wastewater treatment plants (WWTPs) are not designed to eliminate these CoCs. The presence of these chemicals in the surface waters and environment may adversely affect human and ecosystem health. The adverse environmental effects include hermaphroditism in frogs caused by atrazine (Hayes et al., 2002), toxicity to fish and invertebrates by N, N-diethyl-m-toluamide (DEET) (Costanzo et al., 2007) and inhibition of egg fertilization and development of regular secondary sexual characteristics in male fish caused by 17α-ethynylestradiol (EE2) (Kidd et al., 2007). Therefore, it is imperative to effectively remove these CoCs from wastewater in a cost-effective manner.

Biological treatment processes have been considered to be an efficient way to remove these CoCs, e.g. activated sludge (Johnson and Sumpter, 2001) and membrane bioreactors (Trinh et al., 2012). Considering the energy consumption for aeration and the maintenance, a passively aerated and easily maintained nitrifying trickling filter (NTF) may be a more cost effective option for the water industry. The microorganisms in the NTF are capable of removing some organic contaminants (Roh et al., 2009; van den

* Corresponding author.
E-mail address: howard.fallowfield@flinders.edu.au (H. Fallowfield).

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Akker et al., 2011). Thus, the high rate NTFs were considered to be an attractive technology for potential removal of CoCs.

The NTFs have been increasingly introduced to remove nutrients from waters. Operational parameters important to optimise include packing media (Lekang and Kleppe, 2000), influent ammonia concentration (Almstrand et al., 2011), the volumetric flow rate (Tekerlekopoulou and Vayenas, 2003) and organic carbon loading (Zhu and Chen, 2001). One of the most important parameters influencing NTF performance is the organic carbon loading because it may affect the biofilm structure and microorganisms activity in the NTFs (Pearce and Edwards, 2011; van den Akker et al., 2011). High concentrations of organic carbon favour the growth of heterotrophic microorganisms. This growth, potentially changes the structure of the biofilm, adversely affecting nutrient and gas diffusion, and competes directly with the slow growing nitrifying bacteria for dissolved oxygen and biofilm space (van den Akker et al., 2010a; van den Akker et al., 2011). Nevertheless, both nitrifying bacteria and heterotrophic microorganisms in activated sludge have the ability to degrade the CoCs, such as bisphenol A (BPA) (Roh et al., 2009) and EE2 (Larcher and Yargeau, 2013). Little is known about the effect of a readily available exogenous organic carbon source on the efficiency of the high-rate NTFs for CoC removal.

NTFs have shown to be able to remove low concentrations of ammonia and organic carbon (van den Akker et al., 2010a; van den Akker et al., 2010b). Therefore, the aims of this study were (i) to observe the nitrification performance in the NTFs in the presence of exogenous organic carbon and; (ii) CoCs to investigate the effect of exogenous organic carbon on the CoC removal efficiency in the high-rate NTFs; and (iii) to determine the organic carbon removal efficiency in the NTFs in the presence of exogenous organic carbon and CoCs. To cover a variety of the CoCs, eight commonly occurring compounds from a wide range of chemical classes were selected in this study, include caffeine (stimulant), benzotriazole (detergent additive), DEET (insect repellent), EE2 (hormone), acetaminophen (analgesic agent), atrazine (herbicide), trimethoprim (antibiotic) and bisphenol A (plasticizer).

2. Materials and methods

2.1. Chemicals and reagents

High performance liquid chromatography (HPLC)-grade acetonitrile and methanol were purchased from Sigma–Aldrich (Sigma–Aldrich, Australia). Ammonia chloride (≥99.99% purity) was provided by Fisher Scientific (Thermo Fisher Scientific Australia). All the selected CoCs including caffeine (CAF), acetaminophen (ACE), N, N-diethyl-m-toluamide (DEET), benzotriazole (BEZ), 17α-ethynylestradiol (EE2), trimethoprim (TRI), atrazine (ATZ) and bisphenol A (BPA) were purchased from Sigma–Aldrich (Sigma–Aldrich, Australia) with purity >98%. A stock solution (1 g L^{-1}) of each authentic standard was prepared in HPLC-grade methanol.

2.2. Recirculating water preparation

Ammonia stock solution (1 g NH_4–N L^{-1}) was prepared by dissolving 3.819 g ammonium chloride in 1 L Milli-Q water. The exogenous organic carbon source was 15 g sucrose (Thermo Fisher Scientific, Australia) dissolved in 1 L Milli-Q water. 4 bottles (1 L for each) of recirculating water were prepared in tap water, which had a very low total organic carbon (TOC) concentration of 0.21 mg L^{-1}. Each bottle of prepared water was made by adding 5 mL ammonia stock solution and 5 mL sucrose stock solution. The initial concentration of ammonium and sucrose in the recirculating water was 5 mg NH_4–N L^{-1} and 30 mg TOC L^{-1}, respectively. Each CoC was added individually into three of the bottles, the fourth bottle was a control in the absence of CoC. In order to determine the effect of exogenous organic carbon on the efficiency of NTFs for CoC removal, high initial concentrations of these compounds were introduced in this study. CAF, ACE, TRI, BEZ and BPA were all individually added at an initial concentration of 1 mg L^{-1}. The initial concentrations for other CoC in water were 2 mg ATZ L^{-1}, 5 mg EE2 L^{-1} and 10 mg DEET L^{-1}.

2.3. Nitrifying trickling filters (NTFs)

The NTFs consisted of four parallel filter columns (0.8 m in height and 0.09 m in diameter), with each column packed with two layers (0.3 m high for each layer) of polypropylene media (2H Plastics, Victoria Australia). The total specific packing media surface area in each column was 1.09 m^2. The packing media were immersed in the diluted activated sludge for a week to encourage the rapid development of nitrifying microorganisms, before it was inserted into the NTF columns. The NTFs were supplied with the prepared water, recirculating at constant flow rate (0.335 L min^{-1}) by peristaltic pump (Watson Marlow, Germany), resulting in a specific hydraulic loading rate of 18.44 L m^{-2} h^{-1}.

2.4. Experimental design

The activated sludge conditioned packing media were installed in the NTF columns, and the NTFs were then fed with ammonia (5 mg NH_4–N L^{-1}) for one month. In order to maintain this feeding concentration in the recirculating NTFs, 1 mL of ammonia stock solution (1 g NH_4–N L^{-1}) was added every 5 h during the day and after 12 h overnight. Firstly, the representative nitrification rate of the NTFs operated in this mode was determined over 5 h. Secondly, to investigate the effect of exogenous organic carbon on nitrification performance, the NTFs were again fed ammonia (5 mg NH_4–N L^{-1}) but with the addition of sucrose (30 mg TOC L^{-1}) as an exogenous organic carbon source, which is a readily biodegradable carbon source, for one month. The representative nitrification rate of the NTFs operated in the presence of sucrose was also determined over 5 h. Finally, in the presence of ammonia (5 mg NH_4–N L^{-1}) and sucrose (30 mg TOC L^{-1}), three of the NTFs were challenged with individual CoC. The fourth NTF was operated as a control column with initial ammonia concentration of 5 mg NH_4–N L^{-1} and sucrose concentration of 30 mg TOC L^{-1} in the absence of CoCs. The NTFs were operated in recirculation mode for 27 h. In order to avoid the depletion of nitrogen source during the 27 h operation, each recirculation reservoir was supplemented with ammonia after the collection of each sample via adding 1 mL of ammonia stock solution (1 g NH_4–N L^{-1}). Because sucrose is a readily degradable organic carbon source, 5 mL sucrose stock solution was also added into the recirculation reservoirs of the NTFs every 12 h to compensate for the organic carbon removed over time. In the 27 h experiments, the concentration changes of CoCs were determined to evaluate the CoC removal efficiency. The TOC concentration changes after 12 h recirculation were also determined to investigate the correlation between TOC loadings and removals. The concentration of ammonium removed by the NTFs was calculated. All experiments were conducted in duplicate.

2.5. Sampling

The water samples were taken directly from the tubing returning the effluent to the reservoir after passage through each NTF. A 10 mL sample was collected hourly for 5 h to determine nitrification rate. The samples were stored at 3 °C ± 2 °C and analysed within 2 days for ammonia-N, nitrite-N and nitrate-N. To