Ecological Engineering 90 (2016) 234–243

Contents lists available at ScienceDirect

Ecological Engineering

journal homepage: www.elsevier.com/locate/ecoleng

Escherichia coli removal in copper-zeolite-integrated stormwater biofilters: Effect of vegetation, operational time, intermittent drying weather

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ARTICLE INFO

Article history:
Received 2 March 2015
Received in revised form
17 December 2015
Accepted 26 January 2016
Available online 16 February 2016

Keywords:
Stormwater treatment
Biofiltration
Bacteria treatment
Antimicrobial media
Copper ion-exchanged zeolite

ABSTRACT

Existing biofiltration systems have shown variable and often inadequate bacterial removal efficacy. Previous work has shown antimicrobial media copper-zeolite as a promising alternative to reduce the variability and excessive discharge of faecal indicator bacteria such as *Escherichia coli*. A large-scale biofilter column study was conducted over eight months to investigate the benefits of incorporating copper-zeolite into biofilters on *E. coli* removal. The incorporation of copper-zeolite into biofilters improved *E. coli* log removal rate by 53% reducing *E. coli* concentration from 21,800 MPN/100 mL (median inflow) to 126 MPN/100 mL (median outflow) comparable to international primary contact recreational water quality. In addition, the *E. coli* removal performance of copper-zeolite amended biofilters increased after intermittent dry weather periods; this is notable, especially considering biofilter performance usually decreases after drying. Furthermore, these designs reduced inflow copper concentration by 91% (comparable to the metal removal performance of traditional biofilters) and provided a median effluent copper concentration of 8 μg/L. The vegetation in copper-zeolite filters survived. These results validate the use of copper-zeolite as bioretention media, particularly for sites requiring microbial reduction. Future research will include systematic investigation of the processes involved in reduction of bacteria in copper-zeolite filters and optimise filter design to augment the system performance to meet more stringent stormwater reuse requirements.

1. Introduction

Stormwater biofilters are gravity-fed filter beds, often located within an urban environment while creating an amenity feature (Dietz and Clausen, 2006; FAWB, 2009; Zinger et al., 2013). They remove faecal microbes mainly through sedimentation, straining, adsorption and natural die-off; however leaching of faecal microbes occurs through, for example, survival or regrowth in the media, resuspension, and desorption (Stevik et al., 2004; Zhang et al., 2011; Chandrasena et al., 2014a). Indeed, monitored systems range from providing net leaching to reasonable faecal microbe removal capability (over 90%, with most being trapped in the top media layer); regardless of such, the effluent water quality rarely meets stormwater harvesting requirements (Rusciano and Obropta, 2007; Hathaway et al., 2009; NRMMC et al., 2009; Zhang et al., 2011; Chandrasena et al., 2012; Li et al., 2012). These findings suggest that both inadequate microbial removal capacity of traditional filter media and low die-off rate of bacteria trapped in the media contribute to the low and variable performance of existing biofilters. Therefore, recent research has been to investigate alternative filter media with more favourable surface properties for microbial removal (Mohanty et al., 2013; Mohanty and Boehm, 2014).

Antimicrobial materials exert microbiocidal effects through contact with microbial solution or slow release of antimicrobial agents (Milan et al., 2001; Hrenovic et al., 2012). The utilisation of antimicrobial filter media to replace inert filter media has the potential to enhance microbial removal through improved adsorption and inactivation during filtration, and accelerated inactivation of trapped bacteria during dry periods. A recent study, which

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http://dx.doi.org/10.1016/j.ecoleng.2016.01.066
0925-8574/

developed and evaluated a wide range of antimicrobial filter media, demonstrated that, Cu^{2+}- and $Cu(OH)_2$-treated media showed consistently effective 2 log removal of *Escherichia coli* from natural stormwater for over five months under typical stormwater operational conditions (Li et al., 2014a). The Cu^{2+}-exchanged zeolite was further calcined at 400 °C or *in situ* $Cu(OH)_2$ coated to prepare stable copper-zeolite (ZCu400 and ZCuCuO180, respectively) showing over 95% reduction in copper leaching (Li et al., 2014b). ZCu400 showed 2 log bacterial inactivation during a 24 h drying period, and ZCuCuO180 showed consistent 2 log *E. coli* removal during filtration. In the same study, laboratory trials using sand filter columns integrated with different layered configurations of copper-zeolite, showed that ZCu400 at the top and ZCuCuO180 in the middle provided the most effective *E. coli* removal (1.7 log).

The research by Li et al. (2014a,b) was conducted in small, non-vegetated columns (diameter of 30 mm), under highly controlled experimental conditions, making it hardly possible to transfer these findings to field-scale biofilters. For example, water turbidity levels were not representative of natural stormwater, which could overestimate the true performance of the system; indeed, these experiments were unable to account for the effect that sediments have on shielding of the microbes, fouling of the filter media or the unsaturated conditions caused by surface clogging. Furthermore, the testing duration was just 8 days using a daily dosing regime, limiting our understanding of the long-term system stability and performance and whether the systems were resilient against strenuous drying conditions or seasonality effects.

This paper presents, for the first time, a large-scale laboratory study on the development of stormwater biofilters that integrate copper-zeolite for effective *E. coli* treatment. Since this is the first attempt to develop such biofilters, the study focuses on understanding how key design components (i.e. vegetation, filter media type, saturated zone) and stormwater operational conditions (i.e. intermittent wetting/drying, seasonal variation, operational time) impact on *E. coli* removal. *E. coli* was selected as it is a commonly applied indicator for assessing overall water quality and is a conservative indicator for evaluating soil filters (VGG, 2003; NRMMC et al., 2009; Li et al., 2012).

2. Materials and methods

2.1. Preparation of copper zeolites

Natural zeolite (0.1–0.6 mm; Escott Zeolite, Zeolite Australia) was used as the base medium. It was washed 10 times with 10 volumes of tap water and air dried before use. Preparation of copper-zeolite was conducted following procedures specified by Li et al. (2014b). In brief, natural zeolite was treated with NaCl to produce Na-zeolite, followed by treatment with $CuSO_4$ solution to produce Cu^{2+}-exchanged zeolite. The Cu^{2+}-exchanged zeolite was then calcined at 400 °C to prepare ZCu400, or *in situ* coated with $Cu(OH)_2$ followed by heat treatment at 180 °C to prepare ZCuCuO180. The modified media ZCu400 and ZCuCuO180 were analysed using inductively coupled plasma mass spectrometry (ICP-MS) to contain 10 mg Cu/g media, and 13 mg Cu/g media, respectively.

2.2. Experimental set-up

Biofilters were constructed from 240 mm diameter PVC pipes 860 mm in depth, with a transparent Perspex top section 280 mm in depth allowing for plant growth and ponding of water (Table 1). The inner walls of the filters were sand blasted to minimise preferential flow effects. The filters were placed in a greenhouse constructed with a clear, impermeable roof.

The biofilter construction followed industry standards (FAWB, 2009) and previous biofilter studies (Blecken et al., 2009a; Bratieres et al., 2009; Li et al., 2012). In brief, the filters had 70 mm of coarse gravel at the base, including a slotted drainage pipe connected to a vertical riser pipe (Table 1b). The riser created a saturated zone (SZ) in the system 440 mm high with an outlet at the end. 70 mm of coarse sand lay on top of the gravel (as a transition layer), topped by 300 mm of triple-washed sand (0.075–0.6 mm, Daisy Garden Supplies, Melbourne) mixed with 270 g of carbon source (1/4 sugarcane mulch and 3/4 pine wood chips), and 400 mm of filter media layer which included the copper-zeolite layers (ZCu400 and ZCuCuO180, prepared as explained above). Two configurations of the 400 mm top filter media layer were tested (Table 1b):

- '*Layered*' – 50 mm ZCu400 at the top (found to be effective for inactivation while systems were resting during dry weather periods, and was therefore placed at the top where the majority of bacteria are concentrated; 100 mm washed sand for supporting vegetation (in which the top 50 mm was ameliorated with appropriate organic matter, fertiliser and trace elements as per Australian biofiltration design guidelines (Bratieres et al., 2009)); 50 mm of ZCuCuO180 (found to be efficient at bacteria removal during rain events); 50 mm of natural zeolite to adsorb heavy metals leached from the copper-zeolite media, and 150 mm of washed sand at the base to protect the underlying SZ.
- '*Mixed*' – 100 mm of ZCu400/ZCuCuO180 mixture in 1:1 ratio at the top; 50 mm of natural zeolite; and 250 mm of washed sand just above the SZ.

Different vegetation designs were adopted in the cases of "*layered*" and "*mixed*" configurations, because the former was preferred configuration due to its superior *E. coli* removal performance in a feasibility study (Li et al., 2014b) while the latter was to assess the potential of retrofitting existing biofilters with antimicrobial media (by simply adding a new top layer). The '*layered*' configuration was investigated (1) without vegetation (termed SCu) to assess the 'reference characteristics' of copper-zeolite media, (2) with *Palmetto soft leaf buffalo* (termed PBCu) and (3) with *Leptospermum continentale* (termed LCCu, one plant each column) to assess performance of antimicrobial media in the presence of vegetation. The same designs, replacing copper-zeolite with untreated natural zeolite, served as controls (termed S, PB, and LC). The '*mixed*' configuration was planted with *Leptospermum continentale* (termed LCCu-T). The two selected plant types have shown effective removal of both nutrients (Payne et al., 2014) and *E. coli* (Chandrasena et al., 2014b) in traditional biofilters. In total, 7 types of biofilter designs (5 replicates each) were constructed.

During construction, the filter media were added in segments of 75 mm, and each layer was smoothed and compacted by dropping a 3 kg weight once from 100 mm. The filters were then vegetated with plants established over 4 months within 220 mm diameter sand planter bags. Once constructed, all filters were subjected to 6 weeks of twice-weekly watering using dechlorinated tap water to establish the plants and achieve hydraulic compaction.

2.3. Experimental procedure

2.3.1. Dosing

Synthetic stormwater was prepared in a continuously mixed, 1500 L tank using dechlorinated tap water and raw sewage collected from Pakenham Treatment Plant, sediment from a stormwater wetland (sieved through a 1000 μm sieve), and the chemicals listed in Table 2. The target concentrations described in Table 2 were matched to 'typical' worldwide and Australian urban stormwater quality characteristics (Duncan, 1999; NRMMC et al., 2009; McCarthy et al., 2012).