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L'utilisation de murs végétalisés pour éliminer les contaminants émergents des eaux urbaines

The use of green walls to remove the emerging contaminants from urban waters

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RÉSUMÉ

Les murs végétalisés constituent une nouvelle méthode sur site pour traiter et réutiliser les eaux urbaines dans des milieux construits denses. Mais ils doivent être conçus pour éliminer efficacement les contaminants émergents tels que les composés organiques xénobiotiques (COX). Dans cette étude, nous avons utilisé un modèle de colonne de laboratoire et des expériences par lots pour étudier la capacité de différents types de murs végétalisés et de leurs mélanges à éliminer un grand nombre de COX hydrophiles, hydrophobes et chargés, potentiellement présents dans les eaux urbaines. Les résultats indiquent que l'élimination de tous les COX est excellente (>95 %) avec des configurations utilisant fibres de coco et mélange de matériaux pendant les dosages initiaux à 2 volumes de pores. L'élimination des COX hautement hydrophobes et chargés positivement est la plus élevée (>95 %) dans toutes les conditions opérationnelles, tandis que l'élimination d'autres COX a diminué à 25 PV et 50 PV, probablement en raison des polluants de fond dans les eaux urbaines qui peuvent entrer en compétition pour les sites d'adsorption à la surface du matériau. La plupart des COX présentaient une élimination similaire avec des matériaux autoclavés et biologiquement actifs, ce qui indique que l'adsorption est un mécanisme d'élimination dominant.

ABSTRACT

Green walls can offer novel on-site approach for urban water treatment and reuse in densely build environments. However, they need to be engineered for effective removal of emerging contaminants such as xenobiotic organic compounds (XOCs). In this study, we used laboratory column design and batch experiments to investigate the performance of different green wall media and their mixtures for removing wide range of hydrophilic, hydrophobic, and charged XOCs potentially found in urban waters. Results indicate that all XOCs showed excellent removal (>95%) using coco coir and media mix configurations during initial 2 pore volumes (PV) of dosing. The removal of highly hydrophobic and positively charged XOCs remained highest (>95%) under all operational conditions, while other XOCs experienced decline in removal at 25 PV and 50 PV, possibly due to background pollutants in urban water that may compete for the adsorption sites on media surface. Most XOCs showed similar removal using autoclaved and biologically active media, indicating adsorption as a dominant removal mechanism.

MOTS CLÉS/KEYWORDS

Mur végétal, micropolluants, milieu naturel, solution basée sur la nature, jardins verticaux

Living wall, micropollutants, natural media, nature-based solution, vertical gardens

INTRODUCTION

With increasing demand of fresh water in urban cities, there is a dire need to find decentralized solutions to manage existing water resources, such as onsite treatment and reuse of urban waters (i.e., stormwater, greywater) that has significantly lower levels of pathogens, nutrients, and organic matter than blackwater (i.e., toilet effluent). Green wall systems are novel on-site technologies for urban water treatment that can offer multiple benefits to the environment (cooling, noise isolation, amenity, etc.), while providing reliable alternative water resource. However, the challenge is to critically engineer the green wall systems to remove wide variety of pollutants, such as pathogens, nutrients, heavy metals, but also xenobiotic organic compounds (XOCs; organic micropollutants from anthropogenic source) that are of serious concern due to their aquatic toxicity, antimicrobial resistance, and endocrine disrupting properties (Glover et al., 2021). Therefore, XOCs removal from urban waters is necessary to make it a safe water source for potential reuse (e.g., garden irrigation, toilet flushing, food production, etc.).

In green walls, XOCs can be removed through synergetic contribution of plants (i.e., phytoremediation) and growing media (i.e., adsorption, biodegradation). However, the major contribution in removing XOCs is offered by media, implying the need to find such media types that can facilitate healthy growth of plants and can remove wider range of XOCs (i.e., hydrophilic, hydrophobic, charged). Therefore, in this study, we used laboratory column design and batch experiments to investigate the performance of green wall media types and their mixtures for removing wide range of hydrophilic, hydrophobic, and charged XOCs found in urban water. Specific objectives of this study are to (i) quantify the removal of XOCs using different green wall media types and their mixtures, (ii) investigate the effect of operational parameters on the removal of XOCs, and (iii) understand the underlying mechanisms of XOCs adsorption and biodegradation during removal processes.

1 MATERIALS AND METHODS

1.1 Selection of XOCs

To understand the effect of XOCs' properties on the removal performance of different green wall media, twelve commonly found urban water XOCs were selected as representative pollutants, covering both neutral (hydrophilic to hydrophobic) and charged (negative and positive) groups. These XOCs included caffeine (CAF; stimulant), acetaminophen (ACT; analgesic), atrazine (ATZ; herbicide), and tris(2-chloroethyl) phosphate (TCEP; flame retardant) as hydrophilic pollutants (logK_{ow}<2.5); N, N-diethyl-meta-toluamide (DEET; insect repellent), diuron (DUR; herbicide), bisphenol A (BPA; plasticizer), and triclocarban (TRC; antimicrobial agent) as hydrophobic pollutants (logK_{ow}<2.5); sulfamethoxazole (SMX; antibiotic) and ibuprofen (IBF; anti-inflammatory) as negatively charged pollutants, while atenolol (ATN; beta blocker) and trimethoprim (TRP; antibiotic) as positively charged pollutants.

1.2 Column study

1.2.1 Media selection

This work tested three lightweight green wall media types (i.e., coco coir, zeolite, and perlite) and three different combinations of their mixes (i.e., coco coir + perlite, coco coir + zeolite, and coco coir + perlite + zeolite). The selected media types have low bulk density value $(0.09-0.63 \text{ g/cm}^3)$ which is desirable to reduce the weight of green wall system.

1.1.1 Experimental column design

The columns were constructed using 32 mm diameter PVC pipe with 350 mm total height, containing 300 mm media and 50 mm free gap for water detention (Figure 1(a)). All columns were fitted with stainless-steel mesh (150 micron) at the bottom to avoid media leaching and a layer of washed gravel (~10 mm) was introduced at the top to avoid media floating. The column experiment lasted for 10 weeks, during which different operational conditions were simulated (Figure 1(b)). Before starting experiments, all columns were flushed with tap water for one week to remove any residual fines. After that, each column was irrigated with ~2 pore volume (PV) of synthetic greywater (SGW), five days a week for 6 weeks. The infiltration rate of SGW during week 1–5 was adjusted to 170 mm/h (standard condition) using peristaltic pump and three sampling events (S1–S3) were performed to analyze XOCs removal by different media types over time. Fourth (S–4) and fifth (S-5) sampling events were conducted to analyze XOCs removal under different infiltration rate and drying period, respectively. This was followed by breakthrough curve analysis under continuous SWG dosing and samples were collected after 0.5, 1, 1.5, 2, 4, 6, 8, 10, 12, 15, 25, 50, and 100 PV.

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Inflow Flushing Week 1 Week Week 3 Week Week 5 Week 6 (S-2) **(S-1)** 2 4 (S-3)(S-4)Ponding zone (50 mm) Fresh Aged Aged media (~ 3 media (~ 3 media 4x lower Gravel layer (10 mm) month month Tap water infiltration equivalent) equivalent) rate Standard operational conditions Media depth (300 mm) Week Week Week 9 Week 10 **Batch study** 7 8 (S-5) Stainless steel mesh Adsorption Column Standard Breakthrough (150 micron) destruction vs Drying operational curve Biodegradation Outflow conditions

(b) Experimental timeline

Figure 1. (a) Column design (b) Experimental timeline for column experiments. S1–S5 represent sampling events. Standard operating condition were 2 pore volume of dosing at 170 mm/h infiltration rate.

1.3 Batch study

(a) Column design

At the completion of column experiment, pure media columns (i.e., coco coir, zeolite, and perlite) were emptied in a large container and gently homogenised. One half of the homogenised media was autoclaved (121°C for 30 minutes) to deactivate biological activities, while another half was kept in glasshouse under same experimental environment to maintain its biological activity.

The batch experiments consisted of two groups of aged materials, i.e., (1) biologically active media to represent combined effect of adsorption and biodegradation on the removal of XOCs (2) autoclaved media to represent adsorption as a sole contributor in removing XOCs. For each group, batch experiments were performed using 50 ml centrifuge tubes in which media (5 ml) and SGW spiked with 500 μ g/l of XOCs (20 ml) was added to attain solute to solvent volumetric ratio of 1:4. The detailed method for batch experiments is provided in our previous study (Abd-ur-Rehman et al., 2022).

1.4 Water quality analysis

The concentration of XOCs in all samples was analysed using 1200 series Agilent HPLC coupled with mass spectrometer (Sciex API 4000 triple-quadrupole). The detailed method is provided elsewhere (Vanderford and Snyder, 2006).

2 RESULTS AND DISCUSSION

All XOCs showed excellent removal (>90%) at the start of the experiment (i.e., fresh media and initial 2 PV) in pure coco coir (Figure 2(a)) and all media mix columns (Figure 2(b)), indicating extensive availability of active adsorption sites on fresh media surface. However, most XOCs experienced steep decline in performance (>50%) at 25 or 50 PV, possibly due to the presence of background pollutants (i.e., organics, inorganics, nutrients, suspended solids) that could compete for the adsorption sites on media surface. The decline in XOCs removal was lowest in coco coir column (<25%), suggesting ample availability of adsorption sites to accommodate XOCs in the presence of background pollutants. Few exceptions include significantly lower removal of TCEP and DEET (p>0.05) at 25 PV that can be attributed to their least number of hydrogen bond doner/acceptor among all studied XOCs, limiting their ability to form H-bonding with available functional groups (i.e., -COOH, -OH) of coco coir surface.

The effect of lower infiltration rate on the removal of most XOCs was not significant (p>0.05) (Figure 2), however, the removal was significantly increased after 2-weeks drying (p>0.05) in coco coir (Figure 2(a)) and media mix columns (Figure 2(b)). As 2-weeks drying showed no significant effect in zeolite and perlite columns (p>0.05) (Figure 2(a)), the improvement in media mix columns (Figure 2(b)) was majorly controlled by coco coir. This indicate that daily greywater dosing may have created a resistive water layer around media surface, limiting the access to active adsorption sites. These adsorption sites became available after drying period, resulting in significantly higher removal (>50%) of XOCs. Another possibility could be the change in moisture content of media during dry period that may have resulted in favourable environment for microbial degradation of XOCs. Moreover, the dry period may have forced the heterotrophic microbes to consume XOCs as source of energy in the absence of SGW dosing that otherwise provide a primary source of organic carbon for microbial growth.

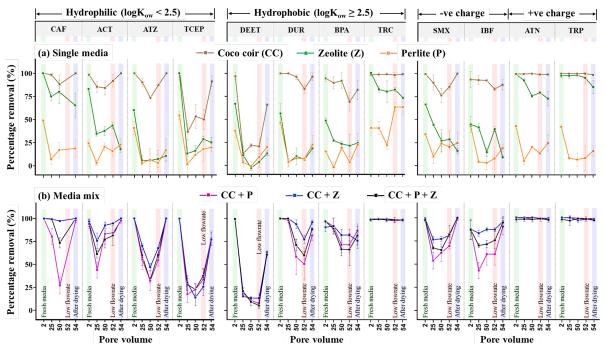


Figure 2. Removal of XOCs under different operational conditions in (a) single media columns (b) media mix columns. The initial concentration of all XOCs was ~10 µg/l.

All XOCs showed similar removal performance using autoclaved media and biologically active media for 24 hours (p > 0.05), indicating adsorption as a dominant mechanism of removal. However, hydrophilic XOCs, ACT and ATZ, showed significantly higher removal after longer contact time (i.e., 5 days) using biologically active media in comparison to autoclaved media (p < 0.05), suggesting the contribution of both adsorption and biodegradation in removal process. Hydrophobic (i.e., DUR, BPA, TRC) and positively charged (i.e., ATN and TRP) XOCs experienced no biodegradation, possibly due to higher adsorption affinity for media, resulting in reduce mobility (and bioavailability) of XOCs. Negatively charged XOCs (SMX and IBF) also showed resistance to biodegradation that may be linked to their functional groups (i.e., aromatic ring structure, sulphur, and amines) that showed recalcitrant behaviour towards biodegradability.

3 CONCLUSIONS

This study explored the performance of three lightweight media types (coco coir, zeolite, and perlite) and their mixtures for the removal of twelve XOCs that cover both neutral (hydrophilic to hydrophobic) and charged (positive and negative) pollutants in urban waters. Results indicate that all XOCs showed excellent removal (>90%) at the start of the experiment in pure coco coir columns and all media mix columns. However, the removal of most XOCs declined at 25 PV and 50 PV. The removal of XOCs was not affected under varying infiltration rate, however, significantly higher removal of XOCs was achieved after 2-weeks drying (p<0.05). Most XOCs showed similar removal performance using autoclaved and biologically active media, indicating adsorption as a dominant mechanism of removal. These findings will be further tested during long-term experiments on real green wall system focusing on XOCs removal from urban waters.

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