Science of the Total Environment 565 (2016) 672–681

Contents lists available at ScienceDirect

Science of the Total Environment

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

# Effect of operating conditions in soil aquifer treatment on the removals of pharmaceuticals and personal care products

Kai He *, Shinya Echigo, Sadahiko Itoh

*Department of Environmental Engineering, Graduate School of Engineering, Kyoto University, Nishikyo, Kyoto 615-8540, Japan*

## HIGHLIGHTS

- Soil organic matter and cation exchange capacity enhanced the removals of antibiotics in SAT.
- A hydraulic retention time (HRT) of 7 days was sufficient for the removals of most PPCPs.
- The removals of most selected PPCPs were similar under vadose and saturated conditions.
- Vadose condition contributed to the removals of sulfamethoxazole and crotamiton.

## GRAPHICAL ABSTRACT

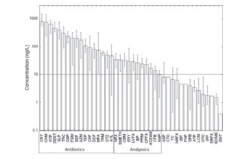

Figure-PPCP concentrations in the A, O water (n=11).

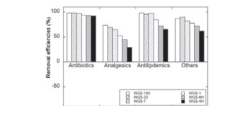


Figure-Removal efficiencies of PPCPs with respected to classification in WGS columns with HRTs. WGS referred to the columns using weathered granite soil, and 180, 30, 7, 1, 8H, and 4H referred to the HRT of 180 days, 30 days , 7 days, 1 day, 8 hours, and 4 hours, respectively.

## ARTICLE INFO

*Article history:*
Received 1 March 2016
Received in revised form 21 April 2016
Accepted 21 April 2016
Available online xxxx

Editor: D. Barcelo

*Keywords:*
PPCPs
SAT
Packing materials
Hydraulic retention time
Saturated condition

## ABSTRACT

Soil aquifer treatment (SAT) is an alternative advanced treatment for wastewater reclamation, and it has the potential to control micropollutants including pharmaceuticals and personal care products (PPCPs). However, the relationship of operating conditions in SAT and removals of micropollutants was not clear. In this study, the effects of operating conditions on the removals of PPCPs were evaluated by using lab-scale columns and plant pilot-scale reactors under different operating conditions. Firstly, weathered granite soil (WGS), standard sand (SAND) and Toyoura standard sand (TS) have different soil characteristics such as total organic carbon (TOC) and cation exchange capacity (CEC). In the columns with these packing materials, the removals of carboxylic analgesics and antilipidemics were effective regardless packing materials. The removals of antibiotics were more effective in WGS than in TS and SAND, indicating high TOC and CEC enhance the sorption in SAT. Secondly, with the extension of hydraulic retention time (HRT), the removals of sulfamethoxazole, acetaminophen, crotamiton, and antipyrine were improved in WGS columns, and adaptable biodegradation for moderately removable PPCPs was formed. Thirdly, the removal efficiencies of sulfamethoxazole and crotamiton were higher in the WGS column under vadose condition than in the WGS column under saturated condition, because of aerobic condition in WGS column under vadose condition. Though long HRT and vadose condition had positive influence on the removals of several PPCPs such as sulfamethoxazole, WGS column with an HRT of 7 days under saturated condition removed most PPCPs.

© 2016 Elsevier B.V. All rights reserved.

## 1. Introduction

Micropollutants including pharmaceuticals and personal care products (PPCPs) are ubiquitous (from ng/L to low μg/L) in the effluents

* Corresponding author.
*E-mail address:* hekai@urban.env.kyoto-u.ac.jp (K. He).

http://dx.doi.org/10.1016/j.scitotenv.2016.04.148
0048-9697/© 2016 Elsevier B.V. All rights reserved.

from wastewater treatment plants (WWTPs) (Boyd et al., 2003; Ellis, 2006; Focazio et al., 2008; Kim et al., 2009; Lin et al., 2009; Okuda et al., 2009; Ort et al., 2010; Stuer-Lauridsen et al., 2000; Yonetani et al., 2012), and they may pose risks to both human and ecosystems (Nakada et al., 2010). Thus, an additional process after the treatments in WWTPs is required on the improvement of water quality (Suárez et al., 2008), and soil aquifer treatment (SAT) is an option for this purpose. Though SAT has benefits such as low operating cost and stable performance (Gerba et al., 1991; Idelovitch and Michail, 1984), not all micropollutants are degraded completely in SAT (Hernández et al., 2012). Degradation of micropollutants including EDCs and PPCPs in soil were affected by environmental conditions such as temperature, pH, moisture content, organic carbon, the presence of specific microorganisms, and dissolved oxygen (DO) conditions (Aga, 2007; Colucci et al., 2001; Monteiro and Boxall, 2009). Schaffer et al. (2015) demonstrated that the hydrophobicity of compounds and the solid organic matter in SAT column enhanced the sorption of neutral and anionic compounds. However, our understanding on the relationship of these operating conditions and the properties of micropollutants is still poor, and the roles of SAT as an additional process to wastewater treatment remain unclear.

This study addresses these unanswered questions by monitoring the removals of various PPCPs in SAT. PPCPs are an ideal group of compounds for this purpose because of their variety of chemical properties (Suárez et al., 2008), giving insight into the removal mechanism in SAT. The removals of PPCPs in SAT were assessed in both field- and laboratory-scale studies: Drewes et al. (2001) mentioned that six to ten years of saturated anoxic flow led to 60% removal of X-ray contrast media in a field-scale groundwater recharge. In another experiment, the removal efficiency of sulfamethoxazole in a bank filtration system was 95%, but only 53% in an artificial recharge system (Grünheid et al., 2005). Xu et al. (2009) observed that the removals of six PPCPs varied in four agricultural soils. These studies indicated that the operating conditions influenced the removals of PPCPs in SAT, but the number of target compounds is limited.

Though previous studies revealed the removal characters of several PPCPs in SAT, no study systematically evaluated the relationship between the removals of PPCPs and operating conditions with respected to the properties of PPCPs. In this study, the effects of operating conditions (packing materials, hydraulic retention time (HRT), and saturated conditions) on the removals of PPCPs were evaluated in the SAT columns under setting operating conditions. Furthermore, the cooperation of SAT and conventional activated sludge (CAS) treatment was discussed with respected to the removals of micropollutants.

## 2. Materials and methods

In this study, the experiments were divided into three parts: the effects of 1) packing materials, 2) HRT and 3) vadose condition on the removals of PPCPs in SAT. Firstly, columns packed with weathered granite soil (WGS), standard sand (SAND) and Toyoura standard sand (TS) were operated to evaluate the effects of packing materials on the removals of PPCPs. Secondly, two series of experiments were conducted to evaluate the effect of HRT. In the first set, WGS with HRTs of typical SAT operation (i.e., 7, 30 and 180 days); in the second set, shorter HRTs (i.e., 4 h, 8 h, 1 day and 7 days) were used to understand the removals of PPCPs in SAT with short-term HRT. Thirdly, WGS columns under vadose and saturated conditions were operated to evaluate the effect of vadose condition on the removals of PPCPs in SAT.

### 2.1. Reagents

In this study, 42 PPCPs were selected according to the frequency of their detection in the effluents of WWTPs (Okuda et al., 2009; Yonetani et al., 2012) (Table 1). DEET is the abbreviation of *N*,*N*-Diethyl-meta-toluamide. CAM and SDIME were dissolved into acetone (Okuda et al., 2009). CPFX, LVFX, NRFX, and ERFX were dissolved into a water/acetonitrile solution, and all the other PPCPs were dissolved into acetonitrile to prepare the stock solutions. For the analysis of PPCPs with a LC–MS/MS system, formic acid, methanol, and acetonitrile (LC–MS grade) were used for the preparation of mobile phase.

CAF and CTC were purchased from Cosmo Bio Co., Ltd; IPP and LVFX were purchased from Tokyo Chemical Industry Co., Ltd; ATP, LCM, ATL, AZM, CPFX, DTZ, ERFX, FNP, FSM, GFB, MFA, OTC, SMT, TC, IBP, SMETH, and TRM were purchased from Funakoshi Co., Ltd, and all other compounds were purchased from Wako Pure Chemical Co., Ltd. All the aqueous solutions were prepared with ultra-pure water produced with Millipore Academic-A10 system (hereinafter call MQW).

### 2.2. Feed water

An effluent from an actual WWTP using anaerobic–anoxic/oxic ($A_2O$) process (hereinafter called $A_2O$ water) in Kyoto City (Japan) was used as the feed water for the experiments. Water quality of feed water is shown in Table 3.

**Table 1**
List of the target PPCPs.

| Compounds | $K_{oc}$ | p$K_a$ | COOH | Use/category | Abbr. |
|---|---|---|---|---|---|
| Ampicillin | 6.423 | 2.5, 7.3 | + | Antibiotic | AMP |
| Azithromycin | 3100 | 8.74 | | Antibiotic | AZM |
| Chlortetracycline | 6.575 | 1.4 | | Antibiotic | CTC |
| Ciprofloxacin | 61,000 | 6.09 | + | Antibiotic | CPFX |
| Clarithromycin | 150 | 8.99 | | Antibiotic | CAM |
| Enrofloxacin | 15,800 | 5, 8–9 | + | Antibiotic | NRFX |
| Erythromycin | 570 | 8.88 | | Antibiotic | ERY |
| Levofloxacin | 44,143 | 6.24, 8.74 | + | Antibiotic | LVFX |
| Lincomycin | 69 | 7.6 | | Antibiotic | LCM |
| Norfloxacin | | 6.34, 8.75 | + | Antibiotic | NRFX |
| Oxytetracycline | 4.319 | 9.5 | | Antibiotic | OTC |
| Sulfadimethoxine | 285.2 | 6.91 | | Antibiotic | SDIME |
| Sulfamethazine | 208 | 7.59 | | Antibiotic | SMT |
| Sulfamethoxazole | 72 | 1.6, 5.7 | | Antibiotic | SMETH |
| Tetracycline | 0.69 | 3.3 | | Antibiotic | TC |
| Triclosan | 19,952 | 7.9 | | Antibiotic | TRC |
| Trimethoprim | 75 | 7.12 | | Antibiotic | TRM |
| Acetaminophen | 41 | 9.6 | | Analgesic | ACEAM |
| Antipyrine | 11.65 | 4.15 | | Analgesic | ATP |
| Aspirin | 100 | | + | Analgesic | ASP |
| Crotamiton | 244 | 10.45 | | Analgesic | CRT |
| Diclofenac | 245 | 4.5 | + | Analgesic | DCF |
| Fenoprofen | 233.8 | 4.15 | + | Analgesic | FNP |
| Ibuprofen | 3400 | 9.38 | + | Analgesic | IBP |
| Isopropylantipyrine | 84.96 | 4.91 | | Analgesic | IPP |
| Ketoprofen | 229 | 4.20 | + | Analgesic | KTP |
| Mefenamic acid | 880.1 | 4.20 | + | Analgesic | MFA |
| Naproxen | 330 | 4.45 | + | Analgesic | NPX |
| Atenolol | 4.081 | 3.83 | | Antiarrhythmic | ATL |
| Disopyramide | 288 | 3.18 | | Antiarrhythmic | DSP |
| Bezafibrate | 204 | 3.61 | + | Antilipidemic | BZF |
| Clofibric acid | 43.7 | 3.18 | + | Antilipidemic | CFB |
| Gemfibrozil | 430 | 4.5 | + | Antilipidemic | GFB |
| DEET | 300 | | | Rejectant | DEET |
| Benzophenone | 517 | | | Ultraviolet absorber | BP |
| Caffeine | 741 | | | Cardiac | CAF |
| Carbamazepine | 510 | 13.9 | | Antipilepsy | CBM |
| Primidone | 23.84 | 11.5 | | Antipilepsy | PRM |
| Sulpiride | 13.85 | 9.12 | | Peptic ulcer | SLP |
| Theophylline | 517 | | | Cardiac | TEP |
| Diltiazem | 197.8 | 8.06 | | Blood-vessel dilator | DTZ |
| Furosemide | 302 | 3.8.7.5 | + | Blood-vessel dilator | FSM |

Download English Version:

https://daneshyari.com/en/article/6322093

Download Persian Version:

https://daneshyari.com/article/6322093

Daneshyari.com