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# Transformation of erythromycin during secondary effluent soil aquifer recharging: Removal contribution and degradation path

Liangliang Wei<sup>1,2</sup>, Kena Qin<sup>1</sup>, Ningbo Zhao<sup>1</sup>, Daniel R. Noguera<sup>2</sup>, Wei Qiu<sup>1</sup>, Qingliang Zhao<sup>1,\*</sup>, Xiangjuan Kong<sup>3</sup>, Weixian Zhang<sup>1</sup>, Felix Tetteh Kabutey<sup>1</sup>

1. School of Municipal & Environmental Engineering, State Key Laboratory of Urban Water Resources and Environment (SKLUWRE), Harbin Institute of Technology, Harbin 150090, China. E-mail: [weill333@163.com](mailto:weill333@163.com)

2. Department of Civil and Environmental Engineering, University of Wisconsin-Madison, Madison, WI 53706, USA

3. Center of Science & Technology of Construction of the Ministry of Housing and Urban Rural Development of P. R. China, Beijing 100835, China

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## ABSTRACT

Erythromycin (ERY), a widely used antibiotic, has recently been detected in municipal secondary effluents and poses serious threats to human health during wastewater reusing. In this study, the removal, fate, and degradation pathway of ERY in secondary effluent during soil aquifer treatment was evaluated via laboratory-scale SAT tests. Up to a 92.9% reduction of ERY in synthetic secondary effluent was observed in 1.0 m depth column system, which decreased to 64.7% when recharged with wastewater treatment plant secondary effluent. XRD-fractionation results demonstrated that the transphilic acid and hydrophobic acid fractions in secondary effluent compete for the adsorption sites of the packed soil and lead to a declined ERY removal. Moreover, aerobic biodegradation was the predominant role for ERY removal, contributing more than 60% reduction of ERY when recharged with synthetic secondary effluent. Destruction of 14-member macrocyclic lactone ring and breakdown of two cyclic sugars (L-cladinose and D-desosamine) were main removal pathways for ERY degradation, and produced six new intermediates.

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## Introduction

Antibiotics are the most widely used pharmaceutical compounds for preventing or treating human/animal diseases, however, excretion of those incompletely metabolized antibiotics into ecosystem has become a worldwide environmental issue (Chang et al., 2010; Niu et al., 2013). For example, it is estimated that about 138 g antibiotics/year were consumed per person in China (ten times higher than in America), and 60–85% of those antibiotics were directly discharged into sewage drains and the environment via human excrement (Tian, 2010; Niu et al., 2013). Recent studies reported that

abundant antibiotics were detected in wastewater treatment plant (WWTP) effluent (Costanzo et al., 2005; Batt et al., 2006; Arye et al., 2011), with macrolides, quinolone and sulfonamide being most prevalent. For their genotoxicity to bacterial biomass even under low concentration (Hernando et al., 2006; Watkinson et al., 2009), these antibiotics are recognized as priority pollutants during wastewater treatment and secondary/tertiary effluent reuse (Onesios and Bouwer, 2012).

Of the pharmaceutical antibiotics in use, erythromycin (ERY) is one of the most frequently detected compounds in surface water, secondary effluent, and other water bodies, which is used worldwide to treat infectious diseases (Kummerer,

\* Corresponding author. E-mail: [zhql1962@163.com](mailto:zhql1962@163.com) (Qingliang Zhao).

2009; Serisier and Martin, 2011; Gao et al., 2012). Specifically, high ERY concentrations (0.08–2.5 µg/L) in Spain and Switzerland WWTP effluents have been previously reported (Alder et al., 2001; Suárez et al., 2010). For its continual input and persistence in natural environment (Hernando et al., 2006), ERY has been selected as a “pseudopersistent” contaminant by USEPA and “high priority level” contaminant by the Global Water Research Coalition (GWRC, 2008; USEPA, 2010). Alexy et al. (2004) reported that ERY could not be readily biodegraded in the Closed Bottle Test at initial concentration of 2.46 mg/L, and Gartiser et al. (2007) found a concentration of 167 mg/L ERY inhibited carbon removal.

Considering the environmental toxicity and potential health concerns, advanced oxidation processes (AOP) of ultraviolet (UV) irradiation, ozonation, and hydrogen peroxide have been widely applied for ERY removal. Nakada et al. (2007) revealed that the combination of sand filtration/ozonation with activated sludge treatment gave an efficient removal (>90%) of ERY. Similarly, Kim et al. (2009) demonstrated that the combining UV with H<sub>2</sub>O<sub>2</sub> removed more than 90% of ERY within the secondary effluent. Moreover, Fan and He (2011) found that the biodegradation of ERY improved significantly via carbon source (e.g., glucose) and nutrients (N, P) addition. Despite AOP technologies showed promise for the removal of ERY, costs associated with nutrient augmentation and high-energy consumption severely limited its adoption.

In addition to AOP, the artificial recharge technique of soil aquifer treatment (SAT) was recently widely employed to treat those ERY polluted waterbodies for its relatively easy operation, low operational cost, and high efficiency (Pavelic et al., 2011). Several studies have reported the interactions between the removal trend of antibiotics/personal care products and the operational parameters of SAT systems. Arye et al. (2011) revealed that the average removal rate of carbamazepine was 30.56%–79.20% in 1.2 m depth SAT. Hua et al. (2003) found that greater than 90% removal of ibuprofen, iopromide, iohexol, and naproxen was achieved in biologically active sand columns. These studies principally focused on removal characteristics and transformation of typical antibiotics, while few studies focused on the aerobic or anaerobic degradation contribution of ERY following SAT treatment.

The objectives of this study were to (1) investigate the removal efficiency of ERY during SAT operation; (2) examine the contribution of aerobic biodegradation on bulk ERY reduction; and (3) identify putative degradation intermediates and the potential effects of dissolved organic matter (DOM) on ERY removal.

## 1. Materials and methods

### 1.1. Chemicals

ERY (CAS no. 114-07-8) used in this study was of analytical grade and purchased from Sigma-Aldrich (St. Louis, MO, USA). High performance liquid chromatography (HPLC) grade methanol (99.9%; CAS no. 67-56-1), acetonitrile (99.9%; CAS no. 75-05-8) and triethylamine (CAS no. 121-44-8), analytical grade NaHCO<sub>3</sub> (CAS no. 144-55-8), CaCl<sub>2</sub> (CAS no. 10043-52-4), MgSO<sub>4</sub>·7H<sub>2</sub>O (CAS no. 10034-99-8), CO(NH<sub>2</sub>)<sub>2</sub> (CAS no.

57-13-6), KH<sub>2</sub>PO<sub>4</sub> (CAS no. 7778-77-0), K<sub>2</sub>HPO<sub>4</sub> (99%; CAS no. 7758-11-4), et al. were purchased from Tianjin benchmark chemical reagent Co., LTD (Tianjin, China). Milli-Q water was used for all dilutions, samples and chemicals preparation, and final glassware cleaning. Stock solutions of 100 mg/L ERY were prepared in methanol and stored at 4°C (Gao et al., 2015).

### 1.2. Chemical characteristics of experimental soil sample and wastewater

Experimental soil samples were collected from the campus of Harbin Institute of Technology and were classified as sandy loam soil, its physicochemical characteristics are presented in Appendix A Table S1. The main compositions of the soil were found to be 56.83% SiO<sub>2</sub>, 18.19% Al<sub>2</sub>O<sub>3</sub>, 8.92% CaO, 2.84% MgO, 7.73% Fe<sub>2</sub>O<sub>3</sub>, 0.89% TiO<sub>2</sub>, 3.12% K<sub>2</sub>O, and 0.86% Na<sub>2</sub>O (W/W). The loss on ignition (LOI) was observed to be 5.4%. This soil samples had a pH of 8.1, organic carbon (OC) content of 3.63%, cation exchange capacity (CEC) of 16.3 cmol/kg, and Brunauer–Emmitt–Teller (BET) specific surface area of 45.0 m<sup>2</sup>/g. The average pore size distributions of the soil were 49.4 Å.

During the acclimation period (3 months) and first-stage operation (4 months) of SAT, synthetic wastewater was chosen as influent, which was prepared according to OECD guidelines (1996) (in which, glucose 28.2 mg/L, amyllum 28.2 mg/L, NaHCO<sub>3</sub> 13.5 mg/L, CaCl<sub>2</sub> 3.3 mg/L, KH<sub>2</sub>PO<sub>4</sub> 2.2 mg/L, NH<sub>4</sub>Cl 30.6 mg/L, MgSO<sub>4</sub>·7H<sub>2</sub>O 10.2 mg/L, CO(NH<sub>2</sub>)<sub>2</sub> 26.5 mg/L, and K<sub>2</sub>HPO<sub>4</sub> 2.8 mg/L). To clarify the effect of DOM existence in WWTP effluent on ERY removal, secondary effluent obtained from Taiping WWTP of Harbin was recharged simultaneously for a comparison; the recharged secondary effluent had a pH of 7.6 ± 0.3, chemical oxygen demand (COD) of 46.7 ± 4.8 mg/L, total organic carbon (TOC) of 14.7 ± 2.7 mg/L, dissolved organic carbon (DOC) of 12.5 ± 1.8 mg/L, UV-254 of 16.3 ± 1.7/m, nitrate of 5.4 ± 2.7 mg N/L, ammonia of 9.3 ± 2.7 mg N/L, and total phosphorus of 2.3 ± 0.8 mg/L.

### 1.3. Operation of the lab-scale SAT columns

The SAT reactor consisted of two acrylic soil columns (50 cm in length, 10 cm in diameter), with top and bottom caps sealed with rubber gaskets, those columns were operated in-series at a flow rate of 15 mL/hr under gravity flow conditions (16 hr wetting cycle and 8 hr drying cycle). Air-dried soil samples described above were sieved through a 2 mm mesh screen and packed into the SAT column, then compacted to field density (with a density of 1.45 g/cm<sup>3</sup>). The columns were operated in down flow mode and were therefore assumed to be predominantly unsaturated. Each SAT column was fitted with a water sampling port at the middle and bottom of the column. The average porewater velocities of the operated SAT columns were ranged from 2.31 to 2.44 cm/hr, demonstrating a residence time of about 10.2–10.8, 20.4–21.6, 30.6–32.4 and 40.8–43.2 hr for 25, 50, 75 and 100 cm depth sampling port, respectively. To prevent the growth of algae within the SAT column, all columns were wrapped with aluminum foil. Soil pore water extraction from different SAT soil layers was performed by inserting the porous plastic tubing of the Rhizon (capped with 0.45 µm nylon membrane) into the sampling ports firstly. At the other end of Rhizon soil moisture sampler,

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