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Removal of antibiotics from piggery wastewater by biological aerated filter system: Treatment efficiency and biodegradation kinetics

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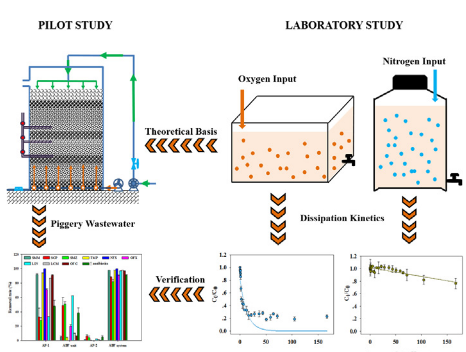
HIGHLIGHTS

- A pilot BAF system built in a swine farm to treat the piggery wastewater.
- Antibiotics could be efficiently removed by the BAF system.
- Antibiotic dissipation in the simulated systems followed the first order kinetics.
- The biodegradation kinetics predicted the fate of antibiotics in the BAF system.
- BAF is a promising technology for treating antibiotics-containing wastewaters.

GRAPHICAL ABSTRACT



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ABSTRACT

This study aimed to investigate the removal efficiency and mechanism for antibiotics in swine wastewater by a biological aerated filter system (BAF system) in combination with laboratory aerobic and anaerobic incubation experiments. Nine antibiotics including sulfamonomethoxine, sulfachloropyridazine, sulfamethazine, trimethoprim, norfloxacin, ofloxacin, lincomycin, leucomycin and oxytetracycline were detected in the wastewater with concentrations up to 192,000 ng/L. The results from this pilot study showed efficient removals (>82%) of the conventional wastewater pollutants (BOD_5, COD, TN and NH_3-N) and the detected nine antibiotics by the BAF system. Laboratory simulation experiment showed first-order dissipation kinetics for the nine antibiotics in the wastewater under aerobic and anaerobic conditions. The biodegradation kinetic parameters successfully predicted the fate of the nine antibiotics in the BAF system. This suggests that biodegradation was the dominant process for antibiotic removal in the BAF system.

1. Introduction

Veterinary antibiotics have been widely used in the animal husbandry for the prophylactic and therapeutic purposes (Sarmah et al., 2006). China is now the biggest producer and user of antibiotics in the world, with its annual usage of 84,240 tons in animals

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(Zhang et al., 2015b). After use, antibiotics and their metabolites are excreted with 30–90% of the parent compounds in animal feces and urine, and finally end up in the receiving environment (Watanabe et al., 2010; Zhou et al., 2013a,b). In recent years, antibiotics in the environment have received a great attention because of their potential adverse effects to non-target organisms and public health due to development and dissemination of bacterial resistance (He et al., 2014, 2016). Therefore, it is essential to eliminate antibiotics in animal wastes before discharge into receiving environments.

In China, most animal farms are often equipped only with simple treatment facilities such as anaerobic lagoons and digesters to treat animal wastes, while some even have no waste treatment facility with wastewater being directly discharged into the environment (Tong et al., 2009; Zhou et al., 2013a,b). In a previous study, seventeen antibiotics were found in the aqueous phase of the wastewater from the three swine farms, with the concentrations ranging from 13.7 ng/L (sulfamethazine) to 166,000 ng/L (lincomycin); while eighteen antibiotics were found in the digester sludge samples from the swine farms, with the concentrations ranging from 4.43 μg/kg (trimethoprim) to 47,100 μg/kg (chlortetracycline) (Zhou et al., 2013a). The treatment facilities used in the farms are ineffective in the elimination of antibiotics in animal waste (Zhou et al., 2013b). Hence, there is a need for cost-effective wastewater treatment techniques to remove not only conventional nutrients but also emerging contaminants like antibiotics in animal waste (Wen et al., 2016; Zhang et al., 2016).

Biological aerated filter system or anaerobic–aerobic biological filter system (BAF system) has been used for treating various wastewater because of its high efficiency and low cost, including industrial wastewater (Zhang et al., 2015a), municipal wastewater (Yang et al., 2015), domestic wastewater (Tao et al., 2016), pharmaceutical wastewater (Priya and Philip, 2015), drinking water pre-treatment (Han et al., 2013), floatation wastewater (Cheng et al., 2012), swine flush water (Westerman et al., 2000), 2,4,6-trinitrotoluene red water (Zhang et al., 2015a), and even some refractory wastewater such as textile and oil field wastewater (Chang et al., 2002; Zhao et al., 2006; He et al., 2013). The BAF unit is a wastewater treatment reactor that consists of three phases: a solid phase that acts as the support media for microbial growth, a liquid phase in which the solid material is submerged, and a gas phase created by the input of air into the reactor (Mendoza-Espinosa and Stephenson, 1999). Currently, it has been proven that BAF system can serve as a promising alternative technology to remove or reduce a wide variety contaminants such as ammonia nitrogen and total nitrogen, phosphorus, total suspended solids (TSS), metals, and volatile organic compounds (VOCs) (Albuquerque et al., 2012; Chen et al., 2015b; Han et al., 2013; Priya and Philip, 2015; Yang et al., 2015). However, it remains unknown whether a BAF system is able to remove antibiotics in animal wastewater.

The objectives of this study were: (1) to evaluate treatment efficiency for antibiotics in swine wastewater by a BAF system, (2) to study the degradation kinetics of the detected antibiotics in the laboratory simulated systems under aerobic and anaerobic conditions, and (3) to investigate the antibiotic removal mechanism by the BAF system. This is the first report on the removal of antibiotics in swine wastewater by a BAF system.

2. Materials and methods

2.1. Biological aerated filter system

For the purpose of investigating the removal of antibiotics in piggery wastewater by BAF system, a pilot BAF system including 2 anaerobic pools (AP) and 1 biological aerated filter unit (BAF unit) (Fig. 1) was set up to treat the raw flush water from a swine farm in Gaoming county, Guangdong province, South China. The raw flush water from the piggery was pumped into the BAF system from a settling tank by a centrifugal pump (0.25 kW·h, 10 min one time and 8 times within-day), with a daily treatment capacity of 1.5 m^3/d. The effective volumes of the two anaerobic pools AP-1 and AP-2 were approximately 3.45 m^3 and 2.60 m^3 and filled with elastic solid materials for microbial attachment, while the BAF unit size was 1.5 m in length, 1.5 m in width and 3.0 m in height. The BAF unit was filled with elastic solid materials and gravel (2 meters high), with their particle size of 3–9 mm and 15–20 mm, respectively (Fig. 1). The BAF unit was designed to be a contact reactor with downward flow, and its average void fraction evaluated by immersion test was 39.9%. The AP-1 and AP-2 were connected with a communicating pipe for keeping the same water level and providing enough carbon source (from AP-1 to AP-2 via the communicating pipe), which is conducive to the denitrification of nitrate nitrogen (from BAF unit) in AP-2. The hydraulic retention times (HRT) of AP-1 and AP-2 were 48 h and 40 h, respectively. BAF unit was operated once every 3 h with a hydraulic loading rate (HLR) of 16 cm, that is: 0.5 h HRT, and 2.5 h empty bed contact time (EBCT). To supply sufficient oxygen for microbial metabolism, each cycle included three 15 min aerations after 1.0 h, 2.0 h and 2.5 h delay from the start of pumping by an air blower (0.10 kW·h). The wastewater supply, aeration and effluent discharging were all controlled by a microcomputer. The effluent from the BAF system was discharged into a big oxidation lagoon (approximately 1000 m^2) for further treatment and storage. The daily operational costs were mainly from energy consumption of different equipment, especially the air blower and centrifugal pump (0.62 kW·h/(d·m^3).

2.2. Laboratory simulation experiment

Aerobic and anaerobic biodegradation of antibiotics was simulated in the laboratory under the room temperature conditions for 166 days without light with the same swine wastewater used for the BAF system. The aerobic experiment was conducted in three 25 L Teflon containers with continuous aeration, while the anoxic experiment was conducted in three 25 L Teflon sealed barrel filled with nitrogen gas. The concentrations of antibiotics in the wastewater were monitored at different time intervals, while physicochemical parameters (temperature, pH, dissolved oxygen (DO), conductivity, redox potential) and conventional wastewater quality parameters (biochemical oxygen demand after 5 days (BOD_5), chemical oxygen demand (COD), total nitrogen (TN) and ammonia nitrogen (NH_3-N)) were simultaneously monitored.

2.3. Sampling campaign

As to the BAF system, water samples (W0, W1, W2, W3, and W4) were collected from different points of the BAF system for analysis of conventional wastewater quality parameters and antibiotics (Fig. 1). The wastewater samples for analysis of antibiotics were collected in 1-L pre-cleaned brown glass bottles (1 L wastewater for one sample) with three replicates, then approximately 50 mL of methanol was added to each bottle and the pH value adjusted to 3 by using 4 M H_2SO_4 to prevent microbial growth of the water samples.

For the laboratory biodegradation experiments, 24 wastewater samples were collected at different sampling time intervals within 166 days (SI Table S1). The samples for analysis of antibiotics were collected in 1-L pre-cleaned brown glass bottles (500 mL each sample) with three replicates, then approximately 25 mL of methanol was added to each bottle and the pH value adjusted to 3 by using 4 M H_2SO_4 to inhibit microbial growth. All the samples from the

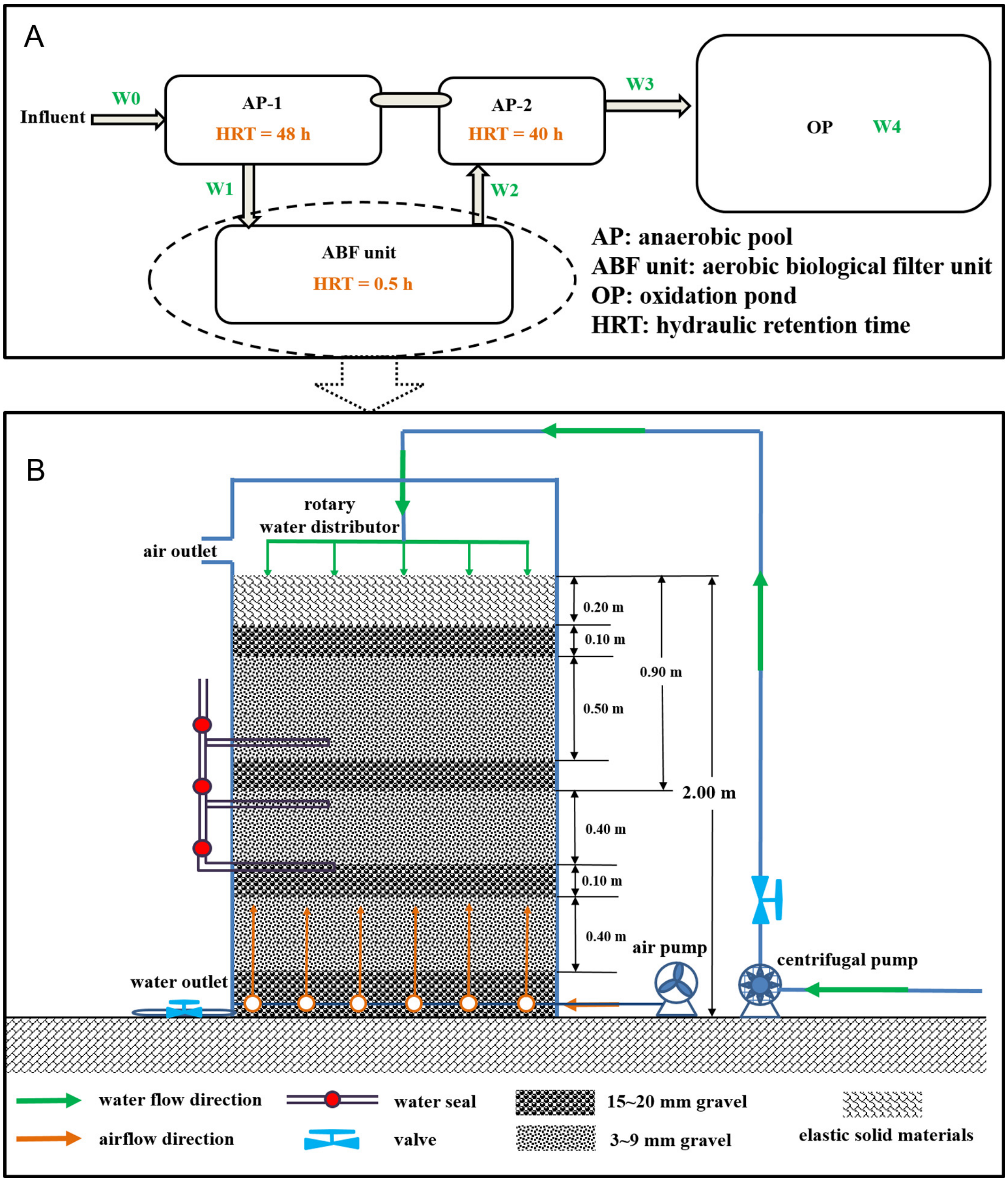

Fig. 1. Process flow diagram of the BAF system and schematic design of the BAF unit.

pilot treatment system and laboratory experiments were stored at 4 °C before analysis and then processed within 24 h.

2.4. Measurement of conventional wastewater quality parameters

Physicochemical parameters including pH, DO, temperature, conductivity and redox potential of the BAF system and laboratory simulation system were measured by the YSI meter (YSI-Pro2030; YSI Incorporated, Yellow Springs, OH, USA). Conventional wastewater quality parameters (BOD_5, COD, TN and NH_3-N) were determined according to the Chinese standard methods (e.g. **HJ 399-2007**, and **HJ 505-2009**). BOD_5 was determined by the dilution and seeding method (**HJ 505-2009**) while COD was measured by the fast digestion-spectrophotometric method (**HJ/T 399-2007**). TN and NH_3-N were determined with a UV–vis spectrophotometer (Shimadzu Instrument Co. Ltd., UV-2450, Japan) according to alkaline potassium persulfate digestion UV spectrophotometric method (**HJ 636-2012**) and Nessler's reagent spectrophotometry method (**HJ 535-2009**).

2.5. Extraction and instrumental analysis of antibiotics

Fifty antibiotics of different classes (sulfonamides, diaminopyrimidines, tetracyclines, fluoroquinolones, macrolides, polyether ionophores, aminocoumarins, polypeptides, lincosamides, chloramphenicol derivatives, and β-lactams) were investigated according to the previous method (Zhou et al., 2012). Detailed information about the analytical method is given in the Supporting Information (SI Text1). In brief, the wastewater samples were filtered through 0.7 mm glass fiber filters (Whatman GF/F), and then the filtered water samples were extracted by solid phase extraction method with Oasis HLB cartridges (6 mL, 500 mg). The samples of particle phase were extracted by the ultrasonic-assisted extraction method with acetonitrile and citric acid buffer, followed by an enrichment and clean-up step with SAX-HLB cartridges in tandem. The target antibiotics were determined with an Agilent 1200 series ultrahigh-performance liquid chromatograph (Agilent, USA) coupled to an Agilent 6460 triple quadrupole mass spectrometer with electrospray ionization in both positive and negative ionization modes (UHPLC-ESI-MS-MS). The quantitative analysis of the target compounds was carried out in dynamic multiple reaction monitoring (DMRM) mode. Laboratory blanks and laboratory controls were also analyzed along with the samples as quality controls.

2.6. Data analysis

First-order reaction kinetic model was applied to fit the laboratory degradation data of antibiotics from the laboratory simulation experiments. The first-order kinetic model can be expressed by the following equation:

$$\frac{dC}{dt} = -k \cdot C \leftrightarrow C_t = C_0 \cdot e^{-kt}$$

where C_0 is the initial concentration of target compound; C_t is the concentration of target compound at time t; and k is the first order rate constant. Using this equation, the half-life ($t_{1/2}$) for the target compound was calculated by the following equation:

$$t_{1/2} = (\ln 2)/k$$

where k is the first order rate constant for the degradation of the target compound. The physiochemical properties and biodegradability of the detected antibiotics were also predicted using the U. S. EPA modeling software EPI Suite v4.1 version.

3. Results

3.1. Operational performance of the BAF system

The general wastewater physiochemical parameters (temperature, pH, DO, conductivity, and redox potential) and conventional wastewater quality parameters (BOD_5, COD, TN and NH_3-N) in the BAF system and biodegradation experiments are summarized in Table 1 and Supporting Information (SI Tables S2–S4). The DO values in the effluents of AP-1 and AP-2 were 0.47 and 0.20 mg/L, respectively, indicating anoxic even anaerobic conditions in AP-1 and AP-2 (anaerobic zone: DO < 0.2 mg/L; anoxic zone: DO 0.2–0.5 mg/L). In contrast, the DO value in the effluent of BAF unit was 2.28 mg/L, indicating aerobic condition in BAF unit (oxic zone: DO > 2 mg/L). As shown in Table 1, the removal rates for the conventional wastewater pollutants (BOD_5, COD, TN and NH_3-N) by the BAF system were found to be 85.0–97.2%, indicating good treatment efficiency.

3.2. Occurrence and removal of antibiotics in the BAF system

Among the 50 target antibiotics, 9 antibiotics of different categories including 4 sulfonamides and diaminopyrimidines (sulfamonomethoxine, SMM; sulfachloropyridazine, SCP; sulfamethazine, SMZ; trimethoprim, TMP), 2 fluoroquinolones (norfloxacin, NFX; ofloxacin, OFX), 1 lincosamides (lincomycin, LIN), 1 macrolides (leucomycin, LCM) and 1 tetracyclines (oxytetracycline, OTC) were detected in the piggery wastewater (Fig. 2 and SI Table S5). Thus, only these nine antibiotics were presented and discussed in the following sections. In influent (W0) of the BAF system, the total concentration of the nine detected antibiotics was 279,000 ± 19,600 ng/L. SCP had the highest concentration of 192,000 ± 7270 ng/L, followed by SMM, LIN and OTC with their concentrations of 45,400 ± 1380, 19,600 ± 407 and 18,700 ± 3090 ng/L, respectively; while LCM, TMP, SMZ, OFX and NFX were detected at the relatively lower concentrations of 2670 ± 243, 220 ± 1.19, 131 ± 1.49, 78.5 ± 4.00 and 40.1 ± 0.67, respectively. After treatment of the BAF system, the detected antibiotics were decreased to various extents, with their total concentration being reduced to 25,900 ± 2210 ng/L in the effluent of BAF unit, and further down to 572 ± 14 ng/L in the oxidation lagoon. It should be noted that the nine antibiotics were also detected in the substrates of the BAF unit, with their concentrations ranging from a few ng/g to several hundred ng/g in the substrates (Table 2).

Fig. 3 showed aqueous-phase removal rates of different antibiotics by the BAF system and each processing unit. For most

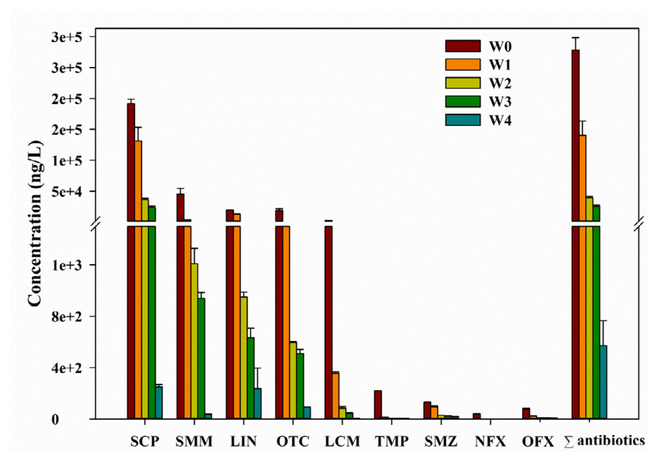

Fig. 2. Concentrations (ng/L) of detected antibiotics in the BAF system. Influent (W0), effluent of the AP-1 (W1), effluent of the BAF unit (W2), effluent of the AP-2 (W3) and the water in the oxidation pond (W4).

Table 1
Concentrations (mg/L) and removal rates (%) of the conventional wastewater pollutants in the BAF system.

Parameter	Concentration (mg/L)					Removal (%)			
	W0	W1	W2	W3	W4	AP-1[e]	BAF unit[f]	AP-2[g]	BAF system
BOD_5[a]	781	53.2	25.8	22.0	5.19	93.2	3.51	0.47	97.2
COD[b]	2410	806	272	268	54.8	66.5	22.2	0.22	88.9
TN[c]	954	607	226	154	15.0	36.3	40.0	7.54	83.8
NH_3-N[d]	564	436	112	84.9	8.41	22.7	57.4	4.85	85.0

[a] Biochemical oxygen demand.
[b] Chemical oxygen demand.
[c] Total nitrogen.
[d] Ammonia nitrogen.
[e] First anaerobic pool.
[f] Biological aerated filter unit.
[g] Second anaerobic pool.

Table 2
Concentrations (ng/g) of the detected antibiotics in the substrates of the BAF unit.

Substrate	Sulfonamides and diaminopyrimidines				Fluoroquinolones		Lincosamides	Macrolides	Tetracyclines
	SMM[a]	SCP[b]	SMZ[c]	TMP[d]	NFX[e]	OFX[f]	LIN[g]	LCM[h]	OTC[i]
Plastic solid material	1.39 ± 0.43[j]	27.6 ± 7.90	0.91 ± 0.35	2.63 ± 0.13	93.9 ± 11.3	43.5 ± 3.68	1.94 ± 0.21	77.2 ± 16.3	507 ± 62.1
Gravel	0.57 ± 0.03	14.5 ± 1.90	0.45 ± 0.15	1.44 ± 0.41	3.07 ± 0.39	1.37 ± 0.00	2.02 ± 0.29	6.97 ± 1.32	85.8 ± 1.82

[a] Sulfamonomethoxine.
[b] Sulfachloropyridazine.
[c] Sulfamethazine.
[d] Trimethoprim.
[e] Norfloxacin.
[f] Ofloxacin.
[g] Lincomycin.
[h] Leucomycin.
[i] Oxytetracycline.
[j] Mean ± standard deviation.

detected antibiotics, the removal rates by AP-1 basically achieved as high as 70.8 ± 1.34% to 100 ± 0.00%, except for SCP (32.9 ± 12.2%), SMZ (27.6 ± 3.93%) and LIN (33.0 ± 0.42%) (SI Table S6). The BAF unit showed good removal for residual antibiotics after AP-1 treatment, especially for SCP (48.5 ± 10.9%), SZM (50.5 ± 3.13%) and LIN (62.2 ± 0.34%). The removal rates for the detected antibiotics by AP-2 were very low (0.05 ± 0.00% to 6.49 ± 2.33%). Totally, the BAF system produced high removal rates for the nine individual antibiotics (82.1%–100%). Moreover, the total removal efficiency for all nine antibiotics by the BAF system also reached to 91.1 ± 0.71%.

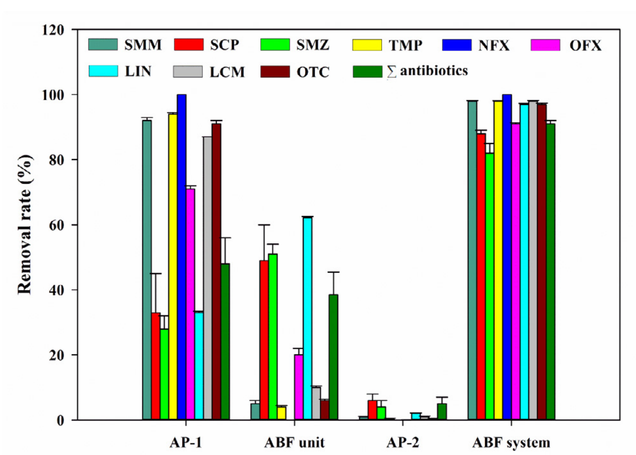

Fig. 3. Removal rates (%) of detected antibiotics by each unit and the BAF system.

3.3. Laboratory biodegradation kinetics of antibiotics

Aerobic and anaerobic treatments of swine wastewater in the laboratory showed different dissipation behaviors for the nine antibiotics with incubation time as shown Fig. S1. Within 166 days of incubation, their concentrations of the detected antibiotics displayed decreasing trends (SI Tables S7 and S8). At the end of the incubation, 90% (aerobic) and 80% (anaerobic) of the antibiotics in the swine wastewater were dissipated, except for SMZ and NFX with 77.3% and 81.7% being lost under aerobic conditions, and 23.7% and 43.4% being lost under anaerobic conditions, respectively (Table 3). The dissipation of these antibiotics followed the first order kinetic model (Fig. 4). It is worth noting that the dissipation rates for SMZ, LIN, LCM and OTC in aerobic treatment were higher than in anaerobic treatment, while the opposite trend was observed for TMP and NFX. The rest three antibiotics OFX, SMM and SCP showed similar dissipation rates under both aerobic and anaerobic conditions.

Table 3
Dissipation kinetics for the target antibiotics in simulated biodegradation systems.

Compound[a]	Aerobic condition				Anaerobic condition			
	Equation	R^2[j]	Dissipation (%)[k]	$t_{1/2}$ (d)[l]	Equation	R^2	Dissipation (%)	$t_{1/2}$ (d)
SMM[a]	$y = 38950e^{-0.023x}$	0.77	96.3	30.1	$y = 107296e^{-0.024x}$	0.75	99.0	28.9
SCP[b]	$y = 94223e^{-0.015x}$	0.81	93.6	46.2	$y = 230052e^{-0.016x}$	0.92	91.8	43.3
SMZ[c]	$y = 156.5e^{-0.01x}$	0.46	77.3	69.3	$y = 263.71e^{-0.002x}$	0.75	23.7	347
TMP[d]	$y = 314.44e^{-0.028x}$	0.92	98.3	24.8	$y = 205.95e^{-0.267x}$	0.83	100	2.6
NFX[e]	$y = 2285.1e^{-0.011x}$	0.63	81.7	63.0	$y = 485.66e^{-0.018x}$	0.68	88.9	38.5
OFX[f]	$y = 3614.3e^{-0.033x}$	0.75	98.6	21.0	$y = 1385.5e^{-0.03x}$	0.79	98.1	23.1
LIN[g]	$y = 22108e^{-0.029x}$	0.92	99.5	23.9	$y = 28392e^{-0.003x}$	0.46	43.4	231
LCM[h]	$y = 1316.4e^{-0.038x}$	0.51	99.8	18.2	$y = 12867e^{-0.011x}$	0.88	84.1	63.0
OTC[i]	$y = 109592e^{-0.036x}$	0.64	94.2	19.3	$y = 311731e^{-0.015x}$	0.97	94.5	46.2

[a] Sulfamonomethoxine.
[b] Sulfachloropyridazine.
[c] Sulfamethazine.
[d] Trimethoprim.
[e] Norfloxacin.
[f] Ofloxacin.
[g] Lincomycin.
[h] Leucomycin.
[i] Oxytetracycline.
[j] Fitness of the kinetic equation.
[k] Loss at the end of incubation (166 d).
[l] Half-life.

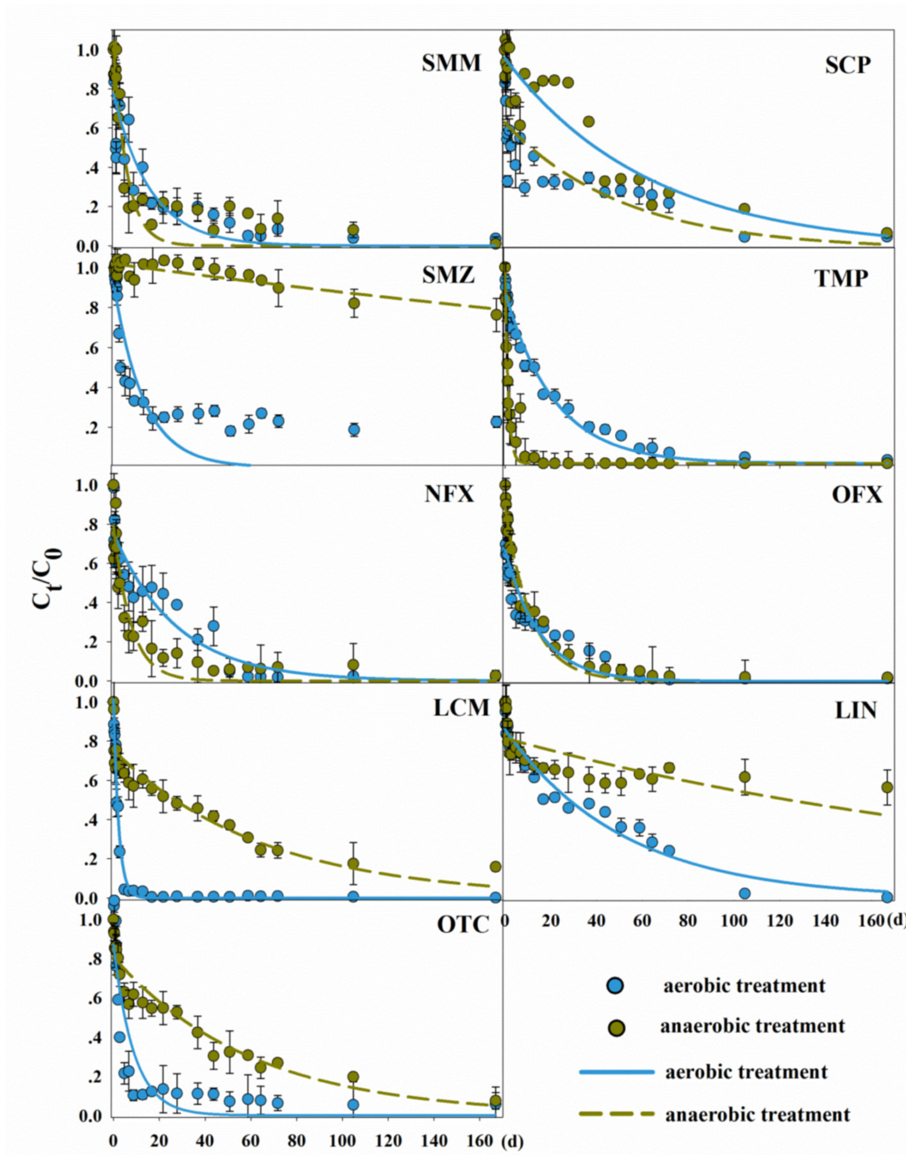

Fig. 4. Dissipation curves for the detected antibiotics under aerobic and anaerobic conditions in the laboratory biodegradation experiments. C_t/C_0 is the concentrations at sampling time (t) divide the initial concentrations.

4. Discussion

The results from the present study showed that the BAF system produced good elimination of the conventional wastewater pollutants (BOD_5, COD, TN and NH_3-N) and the emerging contaminants antibiotics. In terms of conventional wastewater pollutants, the removal efficiencies by this down-flow BAF system were consistent to those obtained from the up-flow BAF system for the treatment of flushed swine manure in a previous study (Westerman et al., 2000). The wastewater quality in the final effluent of the lagoon could meet the Chinese discharge standard of pollutants for the livestock and poultry breeding (GB 18596-2001, $BOD_5 < 150$ mg/L, COD < 400 mg/L and NH_3-N < 80 mg/L). This suggests that the BAF unit in combination with anaerobic and aerobic lagoons can be applied as an effective treatment facility in swine farms to remove conventional wastewater pollutants with high treatment efficiency.

In terms of antibiotic residues in swine wastewater, the present study also showed high treatment efficiencies with more than 90% of total antibiotics removed by the BAF system. This is the first report of removal of antibiotics in swine wastewater by a BAF system. The removal of antibiotics in the BAF system can be attributed to adsorption and biodegradation as demonstrated by the present study and previous studies in various wastewater treatment systems (Li and Zhang, 2010; Chen et al., 2015a; Yang et al., 2011; Zhou et al., 2013a). In the BAF unit, antibiotics could be adsorbed onto supporting media (gravel and elastic materials) and sludge based on their physiochemical properties (Table 2). Tetracyclines and fluoroquinolones are more prone to be adsorbed onto the supporting media and sludge than sulfonamides and macrolides (Zhou et al., 2013a). Biodegradation has been known to be an important removal mechanism for most antibiotics in various wastewater treatment processes such as activated sludge (Li and Zhang, 2010; Yang et al., 2011), enzymatic membrane reactor (Becker et al., 2016), composting system (Yu et al., 2013), anoxic/aerobic membrane bioreactors (Xia et al., 2012) and constructed wetlands (Chen et al., 2015a; Fernandes et al., 2015; Chen et al., 2016a,b). The U.S. EPA EPI Suite model predicted aerobic biodegradation with half-lives of weeks, and slow anaerobic degradation (Table S9). The present study also showed that the nine antibiotics

Table 4
Prediction of antibiotic concentrations by the dissipation kinetic equations, and comparison with the measured concentrations in the BAF system.

Compounds	Anaerobic treatment						Aerobic treatment		
	AP-1			AP-2			BAF		
	Measured[a]	Predicted[b]	Error[c]	Measured	Predicted	Error	Measured	Predicted	Error
SMM	3970 ± 371[d]	14,900 ± 4500	0.57 ± 0.09	939 ± 45.6	1200 ± 120	0.11 ± 0.02	1210 ± 120	3950 ± 370	0.51 ± 0.00
SCP	131,000 ± 22,200	149,000 ± 28,900	0.06 ± 0.01	23,700 ± 2300	36,300 ± 2090	0.19 ± 0.07	36,400 ± 2100	10,400 ± 5280	0.45 ± 0.00
SMZ	95.2 ± 5.72	109 ± 12.7	0.06 ± 0.02	23.6 ± 3.43	28.7 ± 1.20	0.09 ± 0.05	28.8 ± 1.21	95.1 ± 5.72	0.52 ± 0.01
TMP	14.2 ± 0.96	9.91 ± 0.38	-0.15 ± 0.01	3.53 ± 0.19	4.40 ± 0.01	0.10 ± 0.02	4.54 ± 0.01	14.1 ± 0.91	0.49 ± 0.03
NFX	–[e]	–	–	–			–	–	–
OFX	23.5 ± 0.57	34.5 ± 0.00	0.17 ± 0.03	7.54 ± 0.49	7.6 ± 0.00	0.00 ± 0.00	7.58 ± 0.48	23.4 ± 0.00	0.49 ± 0.04
LIN	3180 ± 168	6070 ± 712	0.28 ± 0.03	634 ± 74.6	948 ± 40	0.18 ± 0.03	949 ± 40.9	3170 ± 167	0.52 ± 0.00
LCM	355 ± 12.0	633 ± 81.7	0.25 ± 0.04	44.5 ± 6.81	82 ± 14.1	0.26 ± 0.01	82.4 ± 44.5	352 ± 11.9	0.64 ± 0.06
OTC	1830 ± 28	1990 ± 77.5	0.04 ± 0.02	510 ± 32.1	596 ± 4.2	0.07 ± 0.03	–	–	–

[a] The measured concentrations of antibiotics in the BAF system.
[b] The concentrations were predicted by the dissipation kinetics.
[c] Error equals to the log 10 (simulated value) minus log 10 (measured value), used to check if these two values are in the same order of magnitude.
[d] Mean ± SD (standard deviation).
[e] Not detected in the lagoons.

in swine wastewater could undergo biodegradation under both aerobic and anaerobic conditions (Fig. 4). The present study demonstrated the removal efficiency by the BAF system, which is comparable to or even higher than some other treatment technologies, such as Fenton's reagent with sequencing batch reactor (Ben et al., 2009), constructed wetlands (Huang et al., 2016), and composting (Ho et al., 2013). Thus BAF unit in combination with anaerobic and aerobic lagoons would be a promising technology for treatment of antibiotics-containing swine wastewater.

In order to verify the role of biodegradation in the BAF system, the laboratory dissipation kinetic parameters for the nine antibiotics (Table 3) were used to predict the antibiotic concentrations in the effluents of different units of the BAF system based on the hydraulic retention times of AP-1, AP-2 and BAF unit (Table 4). It was found that the predicted concentrations were mostly within the same order of magnitude when compared to the measured concentrations (Tables 3 and 4). Anaerobic dissipation kinetic parameters predicted better for the two anaerobic pools than the aerobic kinetic parameters used for the BAF unit. This is mostly probably due to the partial contribution of adsorption onto supporting media in the BAF unit. In general, biodegradation played the dominant role in the removal of antibiotics in the BAF system. Hence, the established first-order kinetic models for the nine antibiotics can be applied in predicting the fate of antibiotics in aerobic and anaerobic treatment units of the BAF system.

5. Conclusion

The results from this study demonstrated the efficient treatment of the conventional wastewater pollutants and emerging contaminants antibiotics in the swine wastewater by the designed BAF system. The laboratory biodegradation study showed that the nine antibiotics could be biodegraded under aerobic and anaerobic conditions. In terms of removal mechanisms, biodegradation played the dominant role in the elimination of antibiotics in the BAF system, with anaerobic degradation being complementary to aerobic degradation. Therefore, BAF system could be a promising technology for treatment of piggery wastewater in removing both conventional pollutants such as nutrients and emerging pollutants such as antibiotics.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.biortech.2017.04.023.

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