The Biodegradation of Antibiotics from Cow Wastewater using Sequencing-Batch Membrane Bioreactor

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Abstract

The present study investigated the ability of a lab-scale sequencing-batch membrane bioreactor (SMBR) to remove antibiotics from cow wastewater. We evaluated three veterinary antibiotics from different families:sulfamethoxazole (SM), oxytetracycline (OT), and ciprofloxacin (CF). We found that the SMBR had a high capacity for removing SM (>90%) and OT(>88%), while its ability to remove CF was weaker (>60%). Mass balance analysis revealed that biodegradation is the primary pathway for antibiotic removal in the SMBR system, with sludge adsorption and membrane retention playing a minor role. It is worth noting that OT and CFexhibited higher accumulation in biosolids thanSM. Furthermore, the SMBR system effectively removed nutrients and organic matter from cow wastewater.

Keywords: Antibiotics removal, Adsorption, Biodegradation, Cow wastewater, Sequencing-batch membrane bioreactor.

Introduction

Large-scale beef farms are being progressively established to meet the growing meat demand in Egypt. However, this development has substantially increased the production and release of high-strength cow wastewater [1]. Additionally, veterinary antibiotics are extensively employed in developing countries as growth enhancers and therapeutic agents in the livestock industry to enhance productivity [2].

In Egypt, improper and excessive use of veterinary antibiotics has resulted in the emergence and proliferation of antibiotic-resistant genes (ARG) and antibiotic-resistant bacteria (ARB),particularly among enteric bacteria such as *Salmonella* species and *Escherichia coli*. This phenomenon has a detrimental impact on the effectiveness of antibiotics for future infectious disease treatments [3].

Due to poor absorption and excessive dosing, many antibiotics (70–90%) are released into the environment, including surface water and soil, through cow waste such as manure and urine. This poses a considerable risk to public health and other organisms, as it contributes to the dissemination of ARG and ARB[2,4]. Consequently, recent endeavors have focused on treatingcow farm wastewater to mitigate health and environmental hazards[5].

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Anaerobic digestion is a widely adopted method worldwide for treating livestock waste [5,6]. During the anaerobic digestion process, organic matter in cow waste produces biogas and digestate. Biogas serves as a renewable energy source, while digestate is highly valued as an organic fertilizer. In practical applications, digestate is typically divided into solid and liquid fractions to facilitate efficient management and storage. The liquid fraction of digestate undergoes a series of treatment processes, such as the anaerobic-anoxic-oxic bioreactor, the anoxic-oxic bioreactor, the sequencing-batch reactor (SBR), and the sequencing-batch biofilm reactor. These treatments are necessary because the liquid fraction is generated continuously during the anaerobic digestion process but used seasonally for agricultural applications [4,7].

Nevertheless, applying these treatment procedures diminishes the advantages of anaerobic digestion for livestock manure. The SBR, an alternative to the conventional activated sludge (AS) treatment method, is a biological wastewater treatment approach. Previous studies have employed SBR for treating cow wastewater and/or the liquid fraction of digestate. The cycling between anoxic and aerobic conditions constitutes the primary mechanism for removing organic nutrients and matter in the SBR system [8].

SBR has proven its effectiveness in removing over 94% of TN and total organic carbon from pig wastewater [5]. Similarly, Lee et al.[9]obtained comparable results, demonstrating that SBRachieved a 95% removal of organic matter from pig manure with the appropriate aeration intensity. Wu et al.[8]proposed that the high TN removal capacity of SBR can be attributed to the alternating aerobic and anaerobic conditions that facilitate nitrification and denitrification processes. Furthermore, Amorim et al. [7]discovered that the aerobic and anaerobic conditions in SBR contributed significantly to the efficient removal of total phosphorus (TP), particularly by promoting phosphorus release and uptake by phosphate-accumulating organisms.

However, regrettably, SBRs have limited effectiveness in removing antibiotics from wastewater. Studies have indicated that SBRs achieved less than 50% removal of sulfamethoxazole (SM). The inefficiency of SBR in effectively removing antibiotics from wastewater could be attributed to its low sludge concentration (<3 g/l) and short sludge retention time (SRT; 5.9 days) [4,9].

The sequencing-batch membrane bioreactor (SMBR) is an advanced variation of the conventional SBR that offers improved effluent quality and enhanced simultaneous removal of organic matter and nutrients from wastewater [10]. SMBR incorporates membrane filtration technologies such as ultrafiltration (UF) and microfiltration (MF) to extract water from bioreactors during intermittent aeration [5]. In comparison to conventional SBR, SMBR provides several advantages, including a longer SRT and higher biomass concentration, as reported by Cheng et al. [4].

Previous studies have documented that SMBR achieved removal efficiencies of TN(TN) and chemical oxygen demand (COD) of less than 94% in swine wastewater, considering various nitrogen concentrations [10]. Nevertheless, the extent to which SMBR can effectively remove antibiotics from cattle and cattle manure remains inadequately explored[5].

This study aims to assess the removal efficiency of veterinary antibiotics commonly found in cattle effluents (urine and dung) using SMBR. Additionally,the performance of SMBR will be evaluated by monitoring the quality of the treated water and the characteristics of the biomass. Furthermore, the study aims to elucidate the mechanisms involved in removing antibiotics by investigating their transport and fate within the SMBR system.

Material and methods

Natural cow wastewater

1. Urine sample collection and storage

The fresh cow urine samples were collected from a dairy farm situated in Giza, Egypt. The sampling point chosen was the collection tank of the cow milking station due to its lower solid concentration. The samples were collected using sterile 2-L polyethylene bottles and immediately stored in an icebox at 4℃. Subsequently, they were transferred to the laboratory for further experimentation.

2. Urine characterization

The urine samples were initially thawed to room temperature before the characterization process. Several physicochemical parameters were measured, including pH, COD, TN, mixed liquor-suspended solids (MLSS), and TP, following the guidelines outlined in APHA (2017). Furthermore, fecal pollution bacterial indicators such as total coliforms (TC), *Escherichia coli*, and *Enterococci* (EC) were also examined using the membrane filtration technique described in APHA[11]. All experiments were conducted in triplicate.

Sequencing-batch membrane bioreactor(SMBR)

Figure 1 illustrates the bench-scale submerged SMBR (Submerged Membrane Bioreactor) system. The system comprises three glass tanks arranged in sequence: the feed tank, the bioreactor (housing a submerged hollow fiber membrane filtration unit), and the effluent collection tank. Additionally, the system includes a power timing controller, air diffuser, mixed liquor stirrer, a sensor to monitor the transmembrane pressure for evaluating MF membrane fouling behavior, and two peristaltic pumps. The effective volume of the bioreactor tank is 10 liters, and the MF membrane has an effective surface area of 740 cm^2 with a pore size of 0.4 μm. A power timing controller regulates the aeration process, the mixed liquid stirrer, and the two peristaltic pumps. The first peristaltic pump is responsible for introducing cow wastewater into the bioreactor, while the second peristaltic pump is used to extract treated water from the bioreactor unit.

Experimental protocol

A modified experimental protocol based on Xu et al. [5] was employed in this study. Initially, the collected cow wastewater underwent filtration to eliminate solid wastes, and the resulting filtrate was introduced into the feed tank. The air diffuser was adjusted to alternate the SMBR system between aerobic and anoxic modes. Every 4 h, cow wastewater was fed into the bioreactor tank, with a duration of 1 h for anoxic conditions followed by 1 h for aerobic conditions. A consistent aeration intensity of 20 l/min was maintained throughout the process. During the final hour of each operation cycle, the treated effluent was drawn through the MF membrane. The SMBR system operated continuously for 60 days under controlled temperature conditions of 25 ± 2°C. To maintain a SRT of 30 days, 400 ml of mixed liquor was extracted from the bioreactor tank daily. The MF membrane was operated at a constant hydraulic pressure to achieve an initial flux of 15 ml/min. Every 10 days, the MF membrane was removed, subjected to a 20-minbackwash using a 1% NaOCl solution, and then rinsed with deionized

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water. In the second aerobic phase of the SMBR, mixed liquor samples were collected every 5 days from the three tanks for water quality analysis and biomass characterization.

Antibiotic analysis

Sulfamethoxazole (SM),oxytetracycline (OT), and ciprofloxacin (CF) antibiotics were administered to the cow farm through injection and feed addition.Following the methodology outlined by Liu et al. [12], the three antibiotics were analyzed every 10 days using samples obtained from the feed tank, bioreactor, and treated effluent tank (400 ml each). The analysis involved solid phase extraction, derivatization, and quantification using ultrahigh-performance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS). The removal of antibiotics through biological treatment (R_{Bio}), membrane rejection (R_{MF}), and the SMBR system (R_{SMBR}) was calculated using the following equations:

$$R_{Bio} = (1 - C_{supernatant} / C_{feed}) \times 100\% \tag{1}$$
$$R_{SMBR} = (1 - C_{permeate} / C_{feed}) \times 100\% \tag{2}$$
$$R_{MF} = R_{SMBR} - R_{Bio} \tag{3}$$

where $C_{feed}$is themeasured antibiotic concentrations in feed, $C_{Supernatant}$is themixed liquor supernatant, $C_{Permeate}$ is the permeate.

The mass balance was used to calculate the biodegradation of antibiotics as follows:

$$C_{Feed} \times V_{Feed} = (C_{MLSS} \times X_{MLSS} \times V_{MLSS}) + (C_{Supernatant} \times V_{Supernatant}) + (C_{Permeate} \times V_{Permeate})$$
$$+ \text{Biodegradation} \tag{4}$$

where $C_{MLSS}$is the concentration of antibiotics measured in sludge; $X_{MLSS}$is the concentration of MLSS;andV_{Feed}, V_{MLSS}, $V_{Supernatant}$, and $V_{Permeate}$are the volumes of wastewater fed into the bioreactor, mixed liquor, and its supernatant, and permeate, respectively.
Antibiotic hydrolysis and volatilization were negligible when treated with the AS process and were therefore not considered in the mass balance calculations [4,5].

Statistical analysis

The samples from all experiments were analyzed independently, and replicates were performed. Average values and corresponding standard deviations were calculated for the analysis. One-way analysis of variance (ANOVA) was employed to conduct the statistical analysis.
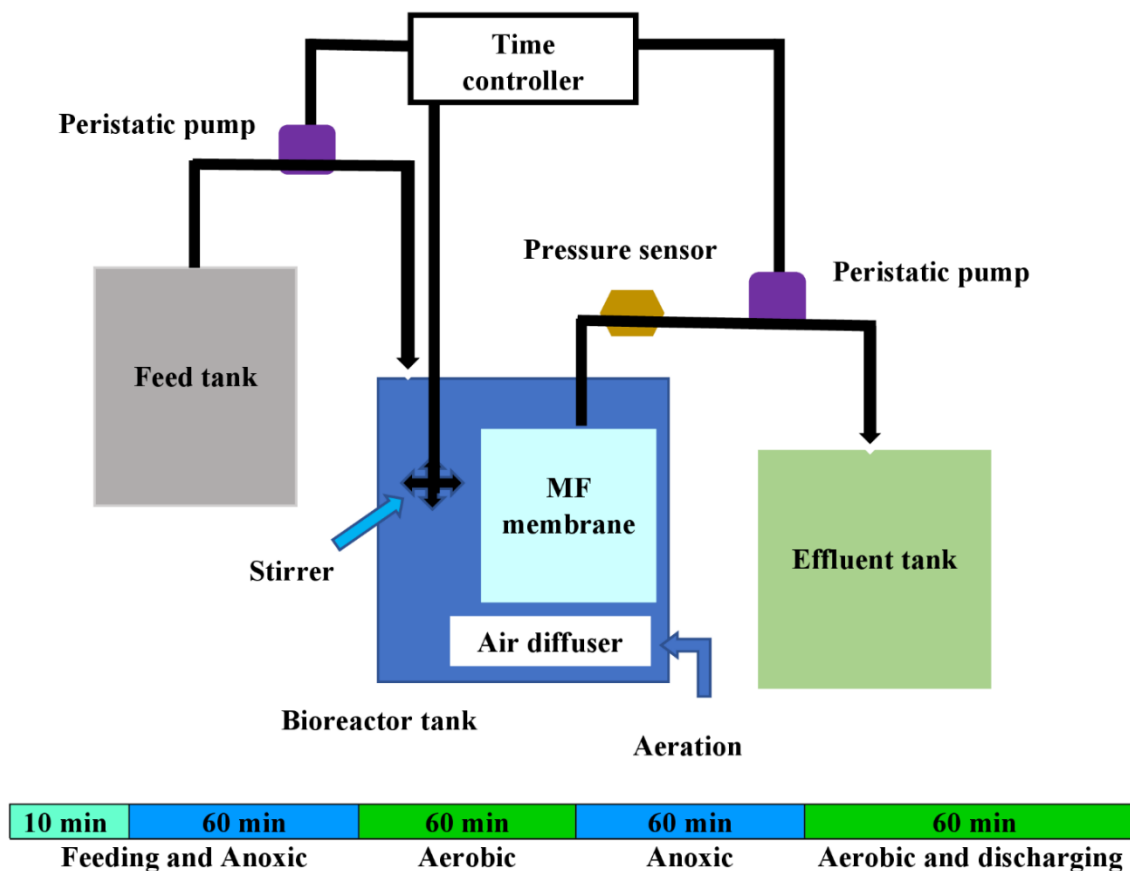
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10 min	60 min	60 min	60 min	60 min
Feeding and Anoxic		Aerobic	Anoxic	Aerobic and discharging

Figure 1. Schematic diagram and operational mode of sequencing-batch membrane bioreactorsystem.

Results and discussion

Characteristics of cow wastewater

Table 1 summarizes the physicochemical and bacterial characteristics of the raw cow wastewater samples. Throughout the entire experiment, 12 rounds of cow wastewater samples were collected. The cow wastewater exhibited high concentrations of organic matter, which were comparable to the findings reported by Othman et al. [13] for raw cattle wastewater, specifically for COD (3600 mg/l), TN (650 mg/l), and TP (380 mg/l). The bacterial indicators of fecal pollution counts were observed to range between $10^7–10^8$cfu/100 ml, $10^6–10^7$cfu/100 ml, and $10^4–10^5$cfu/100 ml for fecal coliforms, *Escherichia coli*, and *Enterococci*, respectively. These results aligned with the outcomes reported by Wang et al. [14] in both fresh and dry manure of cows.

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Table 1. Characteristics of raw cow wastewater.

Sample round	Physicochemical parameters (mg/l)[*]				Bacterial indicators(cfu/100 ml)		
	pH	COD	TN	TP	FC	*E. coli*	EC
1	7.4	3840	760	420	1.1×10^7	3.1×10^6	6.1×10^4
2	7.6	3500	680	352	2.0×10^7	2.7×10^6	4.8×10^5
3	8.0	4800	620	380	9.8×10^6	2.7×10^6	3.4×10^4
4	7.9	4120	742	426	3.4×10^8	1.5×10^7	2.8×10^5
5	8.1	4470	640	395	7.1×10^8	3.3×10^7	6.7×10^4
6	8.2	3500	620	416	4.9×10^7	5.8×10^6	2.9×10^4
7	8.0	3940	764	420	6.0×10^7	8.2×10^6	6.8×10^5
8	7.6	3960	606	400	3.8×10^8	6.8×10^7	2.9×10^5
9	7.8	3780	720	422	5.8×10^7	8.7×10^6	1.7×10^4
10	8.0	4560	686	372	6.0×10^7	1.8×10^6	6.6×10^4
11	7.9	4048	692	318	4.2×10^7	9.4×10^6	8.7×10^4
12	8.2	3820	655	368	1.9×10^8	9.9×10^6	5.8×10^5
Average	7.89	4028	682	390	1.6×10^8	1.4×10^7	2.2×10^5

* All parameters are in mg/l except pH.COD: chemical oxygen demand, TN: total nitrogen, TP: total phosphorus, FC: fecal coliforms, EC: *Enterococci*.

Removal of nutrients and organic matters

Figures 2–4 provide evidence of the SMBR system's capability to remove both organic nutrients and matter. In this study, SMBR exhibited an average removal efficiency of 94–98% for COD (Figure 2) and 92.8–94.2% for TN (Figure 3). These results surpass those reported by Othman et al. (2013), where aerobic granular sludge achieved removal efficiencies of 70–74% for COD and 60–73% for TN in cattle wastewater treatment. The improved performance in our study can be attributed to the utilization of a longer SRT (30 days). However, these outcomes were slightly lower than the results reported by Han et al. [10], who achieved removal efficiencies of 97–99% for COD and >89% for TN using a submerged membrane sequencing-batch reactor (SMSBR) for swine wastewater treatment. Concerning TP removal, the SMBR system demonstrated removal efficiencies ranging between 53.9–61.2% (Figure 4). Notably, TP removal decreased from 62% to 43% within the initial 25 days, followed by a gradual recovery in removal efficiency after day 40 of SMBR operation. This removal pattern aligns with previous studies [5]. The decline in TP removal can be attributed to the inhibitory effect of antibiotics on polyphosphate-accumulating organisms (PAOs), subsequently affecting phosphorus release and uptake under anoxic and aerobic conditions, respectively [15,16]. The conventional membrane bioreactors' ability to remove TP has been established, relying on microbial assimilation due to the retention of phosphorus cations by microporous membranes [17].

Antibiotics removing

1. General removal of antibiotics

As per Xu et al. [5], during the biodegradation of antibiotics, certain metabolites and intermediate compounds can be generated in the bioreactor and treated effluent. Therefore, in the current study, the examination of antibiotic removal focused solely on the disappearance of the parent compound rather than the complete mineralization of the antibiotic.
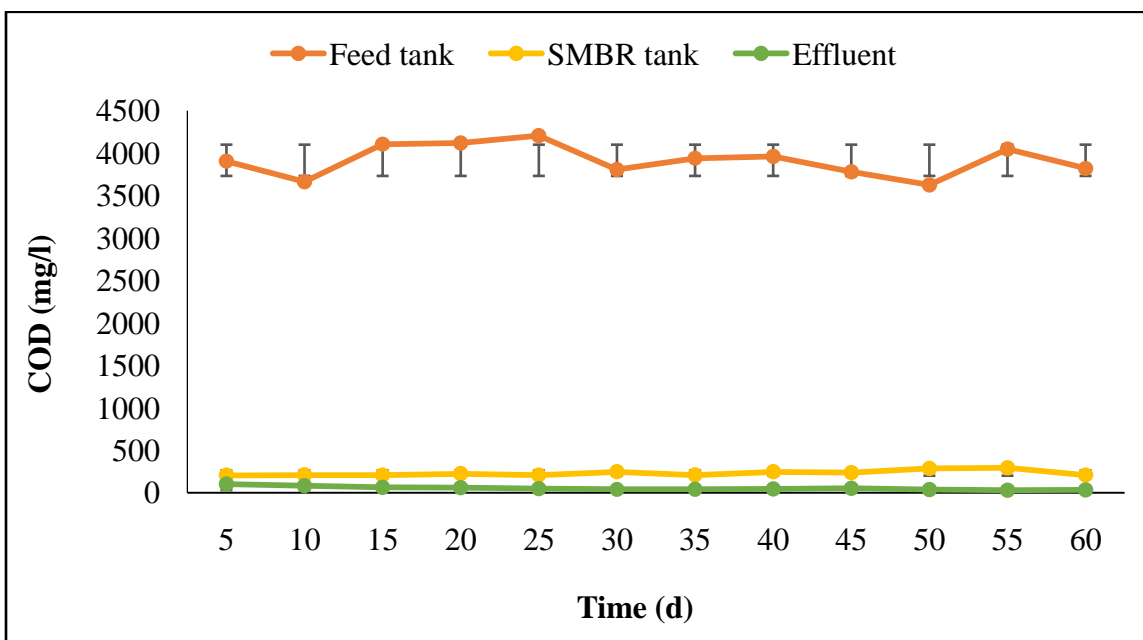
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Figure 2. The chemical oxygen demandremoval efficiency of the SMBR system. Results are expressed as the average of triplicates, and error bars show the standard deviation.
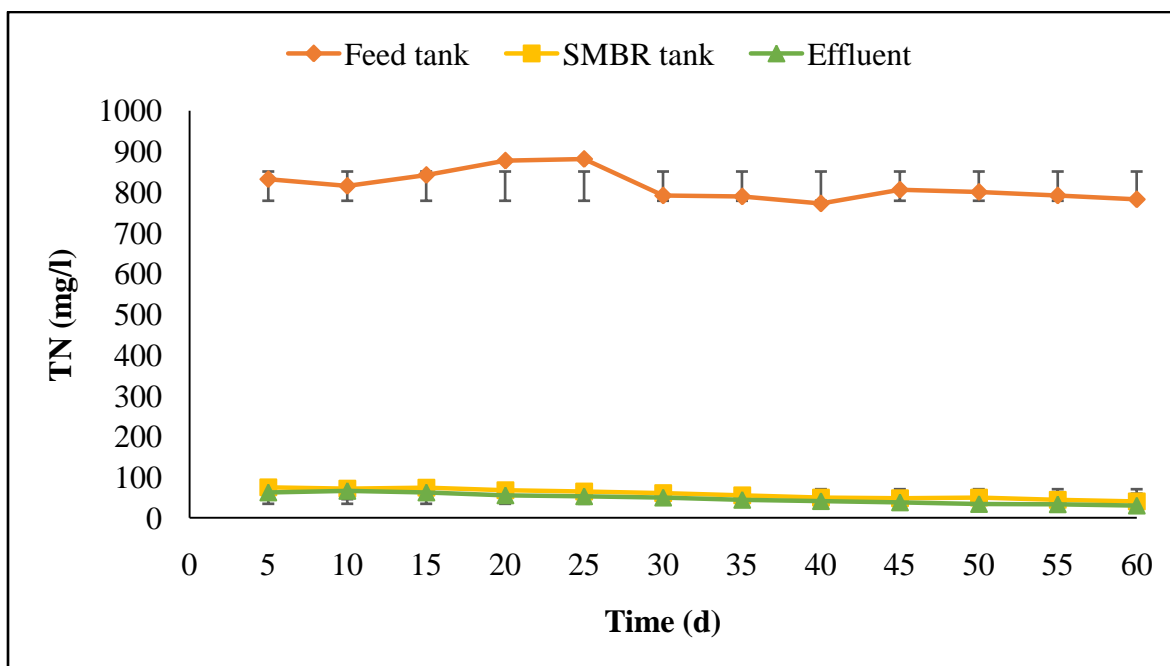

Figure 3. Total nitrogenremoval efficiency of SMBR system. Results are expressed as the average of triplicates, and error bars show the standard deviation.
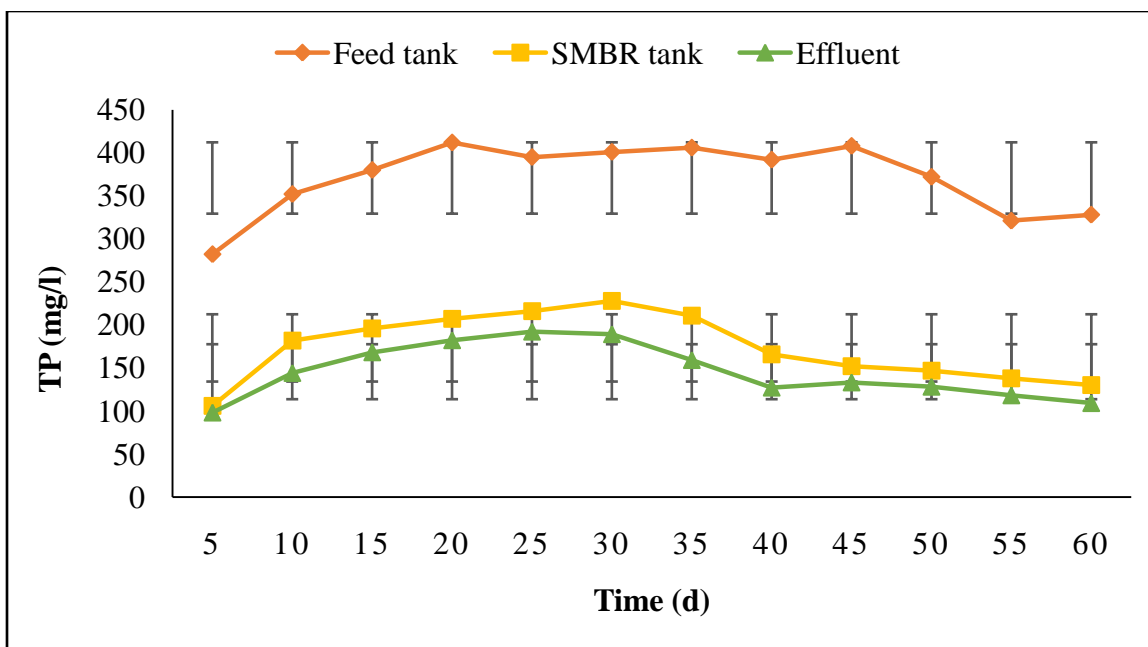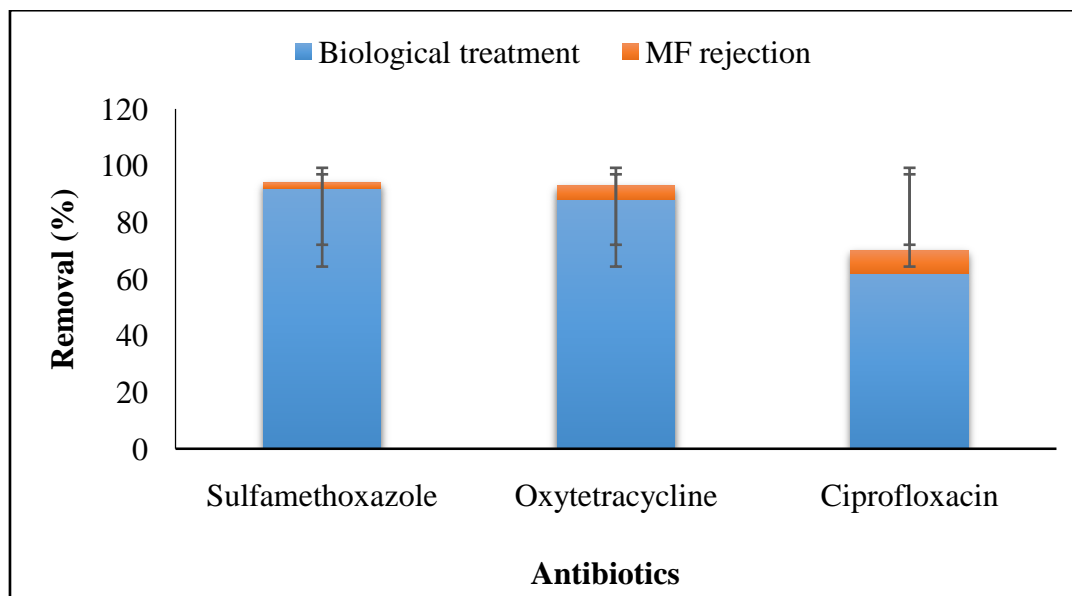
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Figure4. Total phosphorusremoval efficiency of SMBR system. Results are expressed as the average of triplicates, and error bars show the standard deviation.

Figure 5. Removal of antibiotics during SMBR using the biological treatment and MF rejection.Results are expressed as the average of triplicates, and error bars show the standard deviation.

Figure 5 illustrates the average removal percentages of SM, OT, and CF. It is evident that the SMBR system effectively eliminated nearly all antibiotics due to the combined effects of membrane retention and biodegradation. The disparity in removal efficiency between biological treatment and SMBR was demonstrated by the MF rejection (Figure 5). Since microporous

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membranes are unable to retain antibiotics, the primary pathway for their removal was throughbiological treatment processes involving biodegradation, adsorption, and biotransformation (Figure 6). Xu et al. [5] reported that the enhanced retention of antibiotics by membrane clogging and the fouling layer on the membrane surface facilitated their partial removal using an MF membrane. Among the antibiotics examined, SM exhibited the highest average removal rate (92%), followed by OT (88%), while CF displayed the lowest removal rate (62%) (Figure 5). The higher biodegradability of SM can account for its greater removal efficiency in the SMBR system [18]. Previous studies have also indicated the ability of SMBR to remove tetracycline by adsorption onto biosolids, thereby enabling subsequent biodegradation [5,19]. The lower removal rates observed for CF were attributed to the resistance of fluoroquinolones to biodegradation in AS methods [20,21]. Additionally, it is evident that CF and tetracycline, in comparison to SM, were more effectively rejected by the MF membrane, which can be attributed to their lower biodegradability and higher adsorption onto the biofilm layer formed on the membrane surface [19].
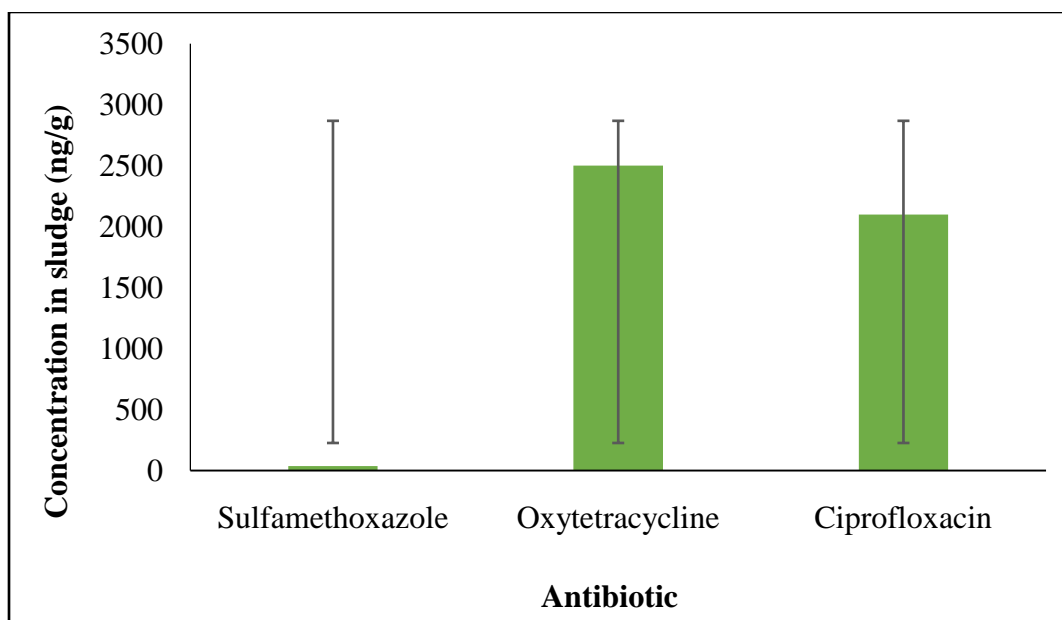

Figure 6. Residuals of antibiotics in biosolids during operation of SMBR. Results are expressed as the average of triplicates, and error bars show the standard deviation.

2. Antibiotics residuals in biosolids

Figure 6 presented the residual concentrations of SM, OT, and CF in biosolids. It was evident that the residuals of OT and CF (2000-2500 ng/g) were more prominent compared to SM (< 100 ng/g). This disparity can be attributed to the higher adsorption and lower biodegradability of OT and CF in sludge as compared to SM[21]. Despite the hydrophilic nature of OT and CF, their substantial adsorption onto sludge may be attributed to electrostatic attractions such as ion bridging, cation exchange, surface complexation, and bonding [20]. Moreover, the positively charged functional groups of OT and CF could undergo electrostatic attraction with the negatively charged AS[4,22]. The minimal adsorption of SM (< 100 ng/g) onto sludge aligns with the findings reported by Li and Zhang [22], highlighting its high biodegradability and low adsorption to biosolids.

Furthermore, the experimental conditions, particularly the neutral pH of the mixed liquor, increased the negative charges of SM, resulting in strong electrostatic repulsion and limited adsorption onto AS. Additionally, the hydrophilic nature of SM further contributed to its lower adsorption onto AS[5]. Considering these findings (Figure 6), additional attention should be given to the discharge and reuse of sludge from the SMBR system as agricultural fertilizer. The generated sludge should undergo further treatment, such as composting, to mitigate any potential risks[23].

3. Fate and transport of antibiotics

The mass balance was employed to calculate the fate and transport of SM, OT, and CF throughout the SMBR operation. The results of the massbalance analysis (Figure 7) revealed that biodegradation served as the primary mechanism for eliminating the three antibiotics in the SMBR system. Notably, SM demonstrated a removal percentage exceeding 90% in the mixed liquor and permeate, highlighting the efficacy of biodegradation. These findings align with the results ofZheng et al. [24], who investigated the fate and transport of sulfonamides in AS.
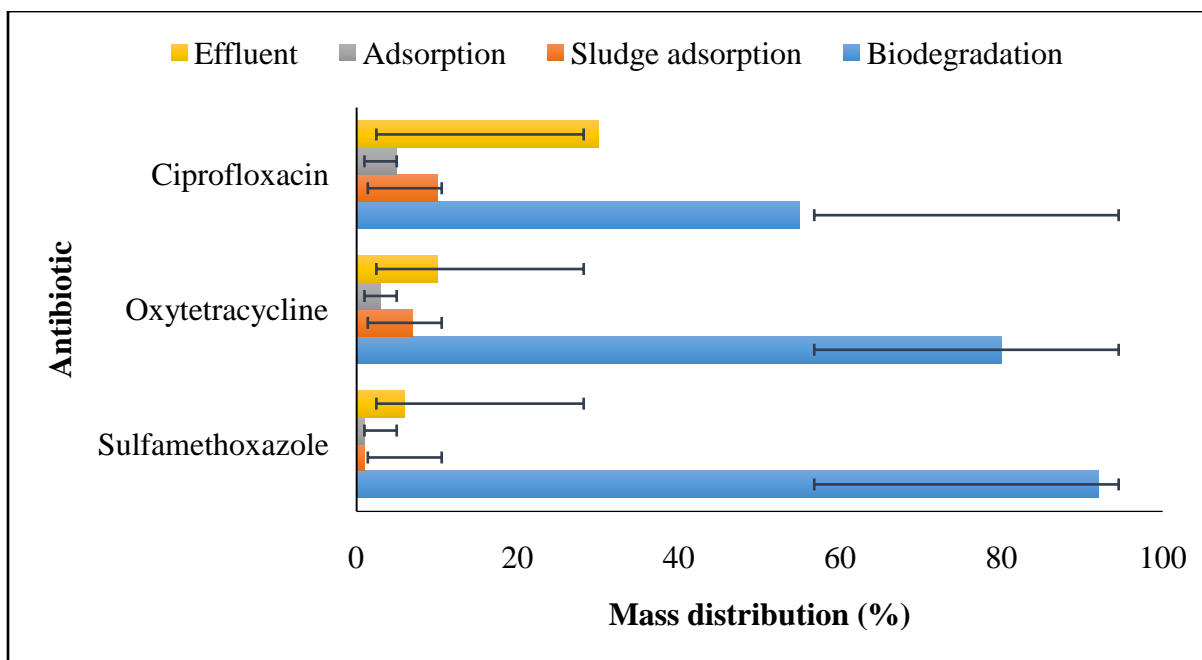

Figure 7. Mass distribution of antibiotics during SMBR operation. Results are expressed as the average of triplicates, and error bars show the standard deviation.

In terms of OT, biodegradation emerged as the primary removal pathway in SMBR operation, accounting for 79% of the removal (Figure 7). Sludge adsorption contributed to 7% of the removal, aligning with the findings reported by Gao et al. [25] and Xu et al. [5]. However, these results diverged from studies that identified sludge adsorption as the main route, followed by biodegradation, for tetracycline removal in conventional SBR[26,27]. In this study, the significantly higher biodegradation of OT could be attributed to the longer SRT and higher sludge concentration characteristic of the SMBR system compared to conventional SBRs.

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Furthermore, biodegradation was identified as the primary pathway for the removal of CF, despite its significant accumulation in the sludge (Figure 7). However, certain previous studies have indicated that sludge adsorption serves as the main removal mechanism for fluoroquinolones in conventional MBRs[20]. These conflicting outcomes suggest that the alternating anoxic and aerobic conditions in SMBR facilitate the biodegradation of CF[12].

Conclusion

The present study demonstrates the effectiveness and efficiency of SMBR as a biological method for removing antibiotics from cow wastewater, thereby reducing the environmental impacts of these antibiotics. SM exhibited high removal efficiency (> 90%), followed by OT (> 88%) and CF (> 60%). Biodegradation served as the primary pathway for antibiotic removal, supplemented by sludge adsorption, with a minor contribution from membrane retention. Notably, residuals of antibiotics, particularly OT and CF, remained prominent in the sludge, emphasizing the need for additional treatment before discharging it into the environment. Overall, SMBR proved to be effective in efficiently removing organic nutrients and matter from cow wastewater.

Conflicts of Interest
The authors declare that there are no conflicts of interest regarding the publication of this paper.

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