Effects of pharmaceutical load on the effluent quality using sequencing batch reactor

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Abstract. The effect of pharmaceutical load on the performance of laboratory scale sequencing batch reactor (SBR) in terms of effluent quality and sludge settling ability was studied. The SBR was operated using low-strength domestic wastewater and spiked with a unique combination of drugs namely atenolol, gliclazide and prazosin after 21 days of reactor operation. Atenolol and gliclazide were spiked at concentration of 1000 µg/l while 100 µg/l for prazosin. Several parameters were observed which includes concentration of COD, ammonia, MLSS and SVI_{30}. The effluent quality and sludge settling ability before and after drugs introduction were compared. The average COD removal efficiency before and after the introduction of pharmaceutical was 85% and 74%, respectively. The average ammonia removals were 84% and 62% before and after addition of pharmaceutical respectively. MLSS and SVI_{30} values was successfully maintained within desired value at the end of experiment indicating good sludge accumulation and sludge settling ability.

1. Introduction

Pharmaceutically active compound (PhAC) in environment is classified as one of micropollutants. Pharmaceuticals are widely used to treat diseases in human as well as in veterinary practices. However, these chemicals will be excreted, as human and animal will not able to retain or absorb them 100 percent [1]. Occurrence of PhAC around the world is widely reported by researchers. In Malaysia, many pharmaceutical compounds such as carbamazepine, enalapril, nifedipine, prazosin, simvastatin, levonorgestrel, gliclazide and atenolol detected in effluent of wastewater treatment plant and surface water [2], [3]. In biological wastewater treatment using conventional aerobic system, organic pollutants undergo biodegradation to become a simpler compound. However, some PhAC are not biodegradable and are toxic or resistant to microorganisms [4]. Previous research reported that the success rate PhAC removal in aerobic conditions is parallel to the removal of COD and nitrogen [5]. Many studies have reported the successful rate of removal of PhAC in lab scale experiment utilizing aerobic sludge in sequencing batch reactor. However, assessing the effect of loading additional PhAC in raw wastewater for experimental purpose towards the quality of effluent and sludge is also one of the important elements. In this study, the quality of effluent in terms of COD and ammonia as well as the physical characteristics of activated sludge, before and after introduction of selected pharmaceutical (atenolol, gliclazide and prazosin) was evaluated and compared.

2. Materials and methods

2.1. Experimental setup

Experiments were performed in an acrylic sequencing batch reactor (SBR) with a total volume of 5 L and 3 L working volume as shown in Figure 1. The height to diameter (H/D) ratio of the reactor is 5 whereby the height and diameter was 55 cm 11 cm, respectively. Reactor was equipped with air bubble diffuser and air compressor to supply air. Dissolved oxygen inside the reactor was monitored at 4.00 ± 0.5 mg/L. Overhead stirrer was installed to ensure homogeneous sludge mixing. The reactor operated in a cycle of which one cycle take 4 h (240 min) to complete. Each cycle consist of filling phase (15 min), anoxic phase (58 min), aeration phase (140 min), settling phase (20 min), withdrawal phase (5 min) and idle phase (2 min). 1500 ml of influent and effluent was pumped in and out of the reactor using peristaltic pumps. The volumetric exchange ratio (VER %) of the reactor was 50%.

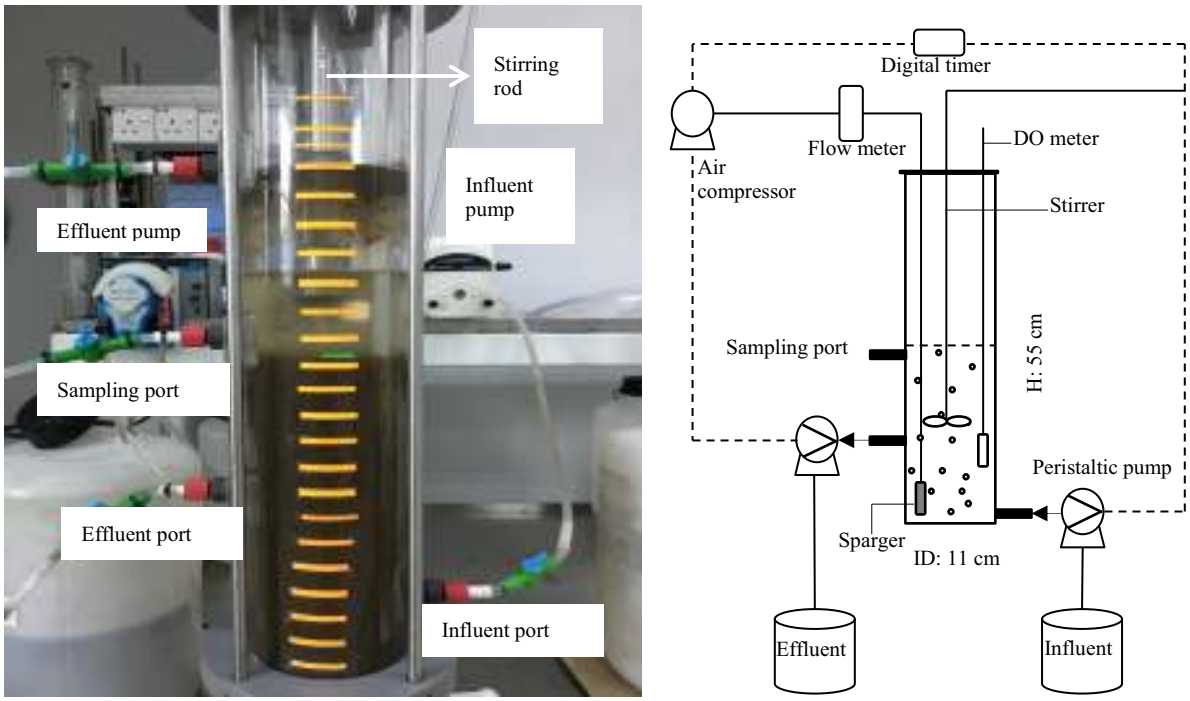

Figure 1. Lab scale SBR column fabricated using Perspex and the schematic diagram

2.2. Wastewater and sludge

Raw domestic wastewater and aerobic sludge was collected from local Sewage Treatment Plant (STP) in Kuala Lumpur. After collection, the wastewater and sludge was kept at 4°C to minimize nutrient degradation. Prior used, the sludge was filtered and washed to remove large particles. The characteristics of sampled raw wastewater collected are as in Table 1.

Table 1. Characteristics of raw domestic wastewater.

	1	2	3	4	Average
pH	7.26	7.12	7.15	7.05	7.15
COD (mg/l)	208	164	175	183	183
Ammonia (mg/l)	19.0	19.2	22.5	22.9	20.9

2.3. Experimental procedures

The reactor was first inoculated with 1500 mL of concentrated activated sludge taken from another SBR in the lab facilities where it was operated for more than 120 days, resulting in MLSS concentration of 2800 to 3300 mg/L at the start of experiment. Raw domestic wastewater fed into the reactor for 20 days. During the start up period without presence of pharmaceuticals, the reactor was

operated at HRT of 8h and organic loading rate of 0.45 to 0.65 g COD/L/d. From day 21 onwards, atenolol and gliclazide were spiked in the influent at concentration of 1000 µg/L while 100 µg/L for prazosin and feed into the reactor for another 30 days. The concentration of pharmaceuticals was chosen in such way to allow reliable measurement of 90% decrease in concentration during experiment [6]. Figure 2 shows timeline of experiment. The effluent of reactor and aerobic sludge samples were taken from the reactor at predetermined days for analysis. Samples were then characterized in terms of COD, ammonia, MLSS and SVI_{30}.

Figure 2. Timeline of experiment

3. Results and Discussion

3.1. Impact of pharmaceutical load on COD and ammonia removal

Figure 3 shows the removal percentage and concentration of COD in influent and effluent samples throughout the experiment. The average COD removal efficiency before and after addition of pharmaceutical is 85% and 74% respectively. Before addition of pharmaceuticals, the reactor demonstrated good removal of COD as presented in Figure 3 (a). The highest COD removal efficiency achieved on day 6 with 91% at minimum final concentration of 19 mg COD L-1 in the effluent. These results show that the reactor has achieves stable condition in terms of ability to remove organic matters. The results also was comparable with previous study [7].

After pharmaceutical loading, the average removal efficiency of COD slightly decreased. The COD removal efficiency fluctuates for the first 15 days after introduction of pharmaceuticals. Sudden drop on day 24 in Figure 3 was due to drastic rise of COD concentration in influent samples at 1434 mg COD/L (Figure 4). This caused by presence of methanol in sample used as solvent during pharmaceutical preparation. To address this issue, methanol was not further utilized and pharmaceutical tablets were purely dissolved and diluted in pure water. For the remaining experimental days, the removal efficiency of COD was stable at removal percentage of 81% to 85%. From the result it can be concluded that introduction of pharmaceutical in feed will not inhibit and does not shows a significant difference on COD removal. This finding was similar as reported by previous study whereby presence of PhAC does not affect the degradation of COD [8].

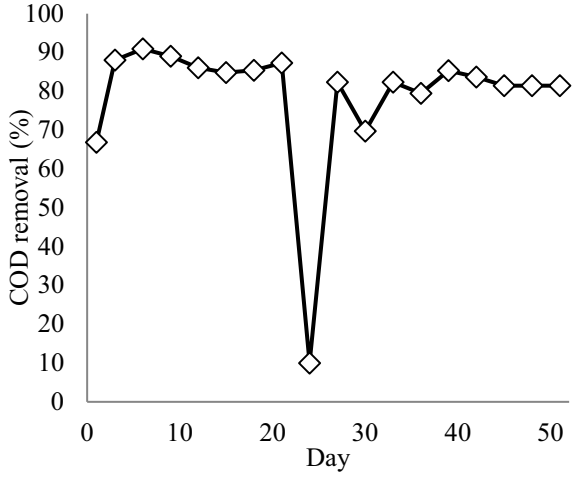

Figure 3. Removal efficiency of COD over time

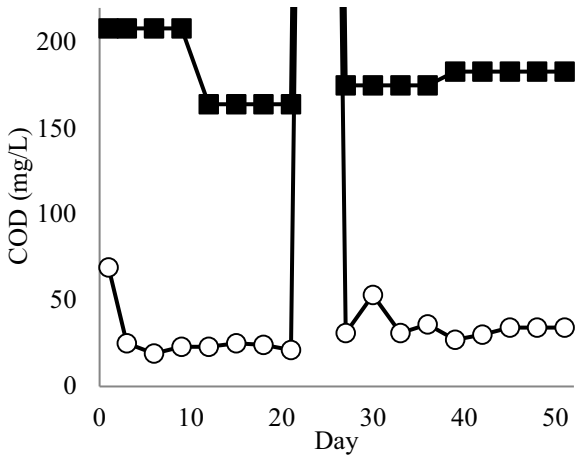

Figure 4. Concentration of COD in influent (■) and effluent (○) of the SBR

In terms of ammonia removal, the average removal efficiency before and after addition of pharmaceutical were 84% and 62%, respectively. Comparatively, the removal of ammonia before pharmaceutical load was more stable than after pharmaceutical load, as shown in Figure 5. The concentration of ammonia in effluent before introduction of pharmaceutical was less than 3 mg/L (Figure 6), which follows the effluent discharge requirement by Department of Environment Malaysia (DOE). Low remaining concentration of ammonia in effluent also indicates that nitrification process occurred, converting ammonia into another compound, nitrate and subsequently nitrite. The highest removal efficiency of ammonia was on day 3 at 98% and ends up with only 0.4 mg/L ammonia in effluent. In contrast, ammonia removal was unstable after day 21, whereby the lowest removal recorded on day 24 with no significant difference between concentration in influent (22.5 mg/L) and effluent (22.4 mg/L). This may due to sudden high COD concentration in influent. Another possible reason for decline in ammonia removal could be attributed by the inhibition of drugs on the activity of ammonia oxidizing bacteria (AOB). Previous study also reported small decline in the removal of ammonia when antibiotics were introduced to the influent [9]. From day 30, the removal efficiency steadily increases and stable towards the end of experiment.

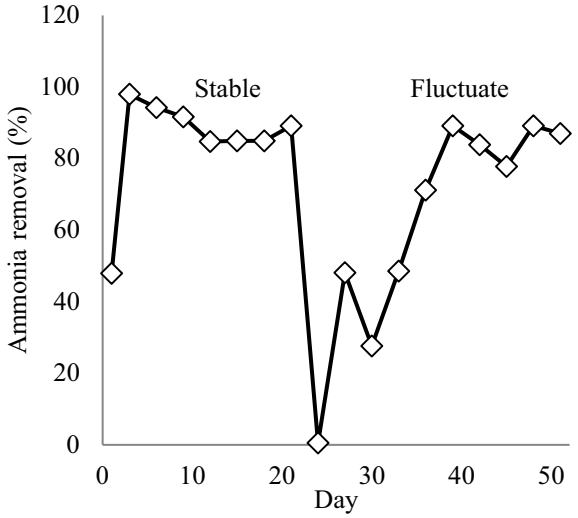

Figure 5. Removal efficiency of ammonia (NH_3-N) over time

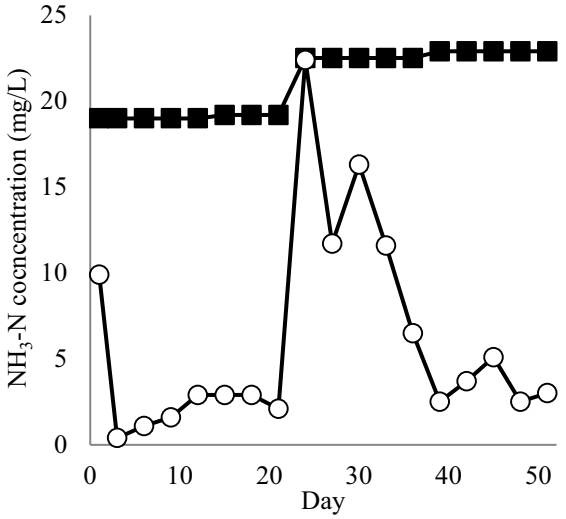

Figure 6. Concentration of ammonia (NH_3-N) in influent (■) and effluent (○) of the SBR

3.2. Impact of pharmaceutical load on sludge concentration and settling ability

The biomass concentration was measured in suspended phase once a week. The MLSS value after inoculation was 3400 mg/L. From day 0 to 20, the value maintained within the range of 3000 to 3500 mg/L. Moreover, SVI_{30} value of sludge decrease constantly, which indicates good sludge settling ability. However, after introduction of PhAC in influent wastewater the value drops from 3000 mg/L to 2100 mg/L on day 27 and further drop to 1500 mg/L on day 33. During this phase, a lot of biomass washout from reactor as the sludge settling ability worsens at SVI_{30} value of 127. The common range of SVI_{30} for activated sludge in wastewater treatment should be between 70 and 100 [10]. This phenomenon may happen due to drastic increase in organic loading rate as mentioned in Section 3.1. Based on Figure 7, MLSS and SVI_{30} values begin to stabilize starting from day 39 until end of experiment.

Flocculation of sludge was observed in the reactor throughout the experiments. Figures 8 shows sludge floc on day 21 and day 51, respectively. On day 21, there were small sludge flocs sized less than 1 cm and brown in colour. At the end of experiment, bigger flocs or bio-solid were found in the reactor. The surface of the bio-solid was slimy and dark brown in color. The bacteria produced the slime around the bio-solid in order to flocculate with each other. However, the formation of flocs or bio-solid was not significant due to presence of mechanical stirrer, thus, hindered floc formation due to high shear force that makes the flocs easily ruptured or burst.

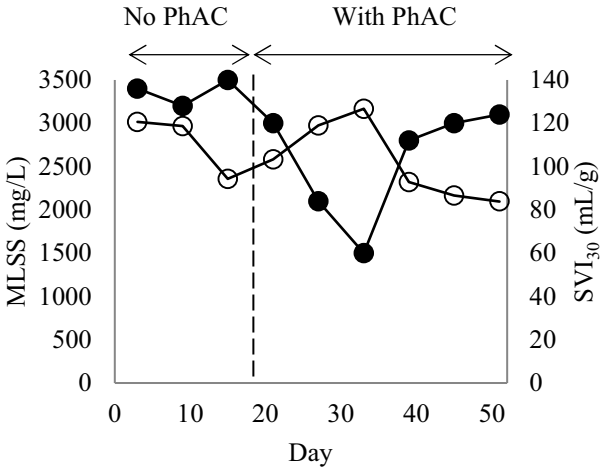

Figure 7. Profile of MLSS (●) and SVI_{30} (○).

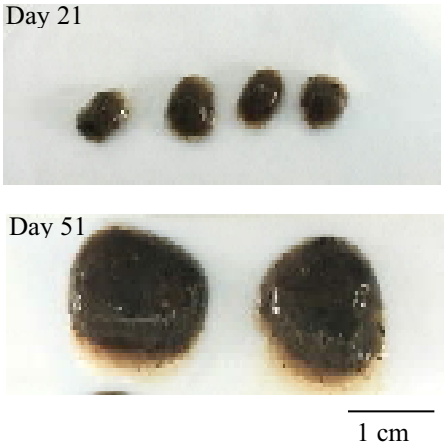

Figure 8. Formation of flocs or bio-solid at Day 21 and Day 51 in the bioreactor.

4. Conclusion

Pharmaceutical loads were not significantly affecting the performance of SBR in terms of COD concentration removal. The average COD removal efficiency before and after addition of pharmaceutical is 85% and 74% respectively. However, ammonia removal is slightly affected with an average of ammonia removal of 84% and 62% before and after addition of pharmaceutical, respectively. MLSS and SVI_{30} value were achieved and able to be maintained within desired values indicating good sludge accumulation and sludge settling ability. Based on this study, experiment should be continued to longer period and pharmaceutical concentration in effluent should be quantified for more comprehensive results.

5. References

[1] E. N. Evgenidou, I. K. Konstantinou, and D. A. Lambropoulou, "Occurrence and removal of transformation products of PPCPs and illicit drugs in wastewaters: A review," *Sci. Total Environ.*, vol. 505, pp. 905–926, 2015.

[2] F. F. Al-qaim, M. P. Abdullah, and M. R. Othman, "Multi-residue analytical methodology-based liquid chromatography-time-of-flight-mass spectrometry for the analysis of pharmaceutical residues in surface water and effluents from sewage treatment plants and hospitals," *J. Chromatogr. A*, vol. 1345, pp. 139–153, 2014.

[3] F. F. Al-qaim, Z. H. Mussa, A. Yuzir, N. A. Tahrim, N. Hashim, and S. Azman, "Transportation of Different Therapeutic Classes of Pharmaceuticals to the Surface Water , Sewage," *Water*, vol. 10, no. 916, pp. 1–16, 2018.

[4] N. H. Tran, T. Urase, H. H. Ngo, J. Hu, and S. L. Ong, "Insight into metabolic and cometabolic activities of autotrophic and heterotrophic microorganisms in the biodegradation of emerging trace organic contaminants," *Bioresour. Technol.*, vol. 146, pp. 721–731, 2013.

[5] M. E. Casas *et al.*, "Biodegradation of pharmaceuticals in hospital wastewater by staged Moving Bed Bio fi lm Reactors (MBBR) nica Escol a," *Water Res.*, vol. 83, pp. 293–302, 2015.

[6] M. Markiewicz, C. Jungnickel, S. Stolte, A. Białk-Bielińska, J. Kumirska, and W. Mrozik, "Primary degradation of antidiabetic drugs," *J. Hazard. Mater.*, vol. 324, pp. 428–435, 2017.

[7] X. Hu, L. Xie, H. Shim, S. Zhang, and D. Yang, "Biological nutrient removal in a full scale anoxic/anaerobic/aerobic/pre- anoxic-MBR plant for low C/N ratio municipal wastewater treatment," *Chinese J. Chem. Eng.*, vol. 22, no. 4, pp. 447–454, 2014.

[8] C. Grandclément *et al.*, "From the conventional biological wastewater treatment to hybrid

processes, the evaluation of organic micropollutant removal: A review," *Water Res.*, vol. 111, pp. 297–317, 2017.

[9] Z. Xu, X. Song, Y. Li, G. Li, and W. Luo, "Removal of antibiotics by sequencing-batch membrane bioreactor for swine wastewater treatment," *Sci. Total Environ.*, vol. 684, pp. 23–30, 2019.

[10] M. Zhang, C. Wang, Y. Peng, S. Wang, F. Jia, and W. Zeng, "Organic substrate transformation and sludge characteristics in the integrated anaerobic anoxic oxic-biological contact oxidation (A^2/O-BCO) system treating wastewater with low carbon/nitrogen ratio," *Chem. Eng. J.*, vol. 283, pp. 47–57, 2016.

Acknowledgement

The authors would like to thank the Ministry of Education Malaysia (MOE) for the Fundamental Research Grant (FRGS) No. 5F003. Many thanks to Malaysia-Japan International Institute of Technology (MJIIT) and Universiti Teknologi Malaysia (UTM) for financially supporting this research under the Tier 1 Grant No. 17H11.