Investigation of Wood Chips as Internal Carbon Source Supporting Denitrification Process in Domestic Wastewater Treatment

Ruth Lorivi, Jianzheng Li, John J. Ambuchi, Kaiwen Deng

Abstract—Nitrogen removal from wastewater is accomplished by nitrification and denitrification processes. Successful denitrification requires carbon, therefore, if placed after biochemical oxygen demand (BOD) and nitrification process, a carbon source has to be reintroduced into the water. To avoid adding a carbon source, denitrification is usually placed before BOD and nitrification processes. This process however involves recycling the nitrified effluent. In this study wood chips were used as internal carbon source which enabled placement of denitrification after BOD and nitrification process without effluent recycling. To investigate the efficiency of a wood packed aerobic-anaerobic baffled reactor on carbon and nutrients removal from domestic wastewater, a three compartment baffled reactor was presented. Each of the three compartments was packed with 329 g wood chips 1x1cm acting as an internal carbon source for denitrification. The proposed mode of operation was aerobic-anoxicanaerobic (OAA) with no effluent recycling. The operating temperature, hydraulic retention time (HRT), dissolved oxygen (DO) and pH were 24 ± 2 °C, 24 h, less than 4 mg/L and 7 ± 1 respectively. The removal efficiencies of chemical oxygen demand (COD), ammonia nitrogen (NH4+-N) and total nitrogen (TN) attained was 99, 87 and 83% respectively. TN removal rate was limited by nitrification as 97% of ammonia converted into nitrate and nitrite was denitrified. These results show that application of wood chips in wastewater treatment processes is an efficient internal carbon source.

Keywords—Aerobic-anaerobic baffled reactor, denitrification, nitrification, wood chip.

I. INTRODUCTION

WASTEWATER treatment is among the most severe environmental challenges due to cumulative population and mounting standards for pollutant discharge. Anaerobic wastewater treatment had been proposed to be the best on site domestic wastewater treatment technology [1]. The study done by Barber and Stucky, however, discovered that effluents from anaerobic treatment could not meet the discharge standards. Both of these studies suggested aerobic polishing stage for the effluents coming from anaerobic treatment for better organic pollutant removal and more importantly nutrients (nitrogen (N) and phosphorus (P)) removal [2].

Nitrogen removal entails alternating anaerobic-aerobic conditions because it is accomplished in two processes: the first one is nitrification which is the oxidation of ammonia into nitrite and then nitrates. And the second process is denitrification which is the sequential reduction of nitrate and nitrites into nitrogen gas [3]. Nitrifying bacteria (*Nitrosomonas*)

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are obligate aerobes whereas a study executed by Reza and Cuenca [4] compared denitrification with biological oxidation of organic matter in which they found no difference except for the fact that denitrification occurs in the absence of DO. Albeit on its presence, denitrifying bacteria (*Nitrobacter*) prefer DO rather than NO₃⁻ to oxidize organic matter [5], [6]. Xu et al clarified a significant aspect in the accomplishment of the biological nutrient removal process is the availability of suitable carbon source for denitrification process [7].

Since 1970s AO treatment technology have been employed [8]. Currently, AO is used to treat diverse kinds of wastewater especially the one characterized by high nutrients concentrations. A study done by Justo et al for instance integrating an expanded granular sludge bed and a sequential batch reactor treating beet sugar industrial wastewater realized removal rates of more than 97.3 and 99.3% of TN and soluble COD, respectively [9] and another study by Xiang et al utilizing an A²O membrane bioreactor (MBR) treating municipal wastewater with low C/N ratio obtained good removal efficiencies of 84.5% and 98.1% for COD and NH₄⁺ respectively [10]. The tremendous results from these two studies resembles many other AO studies reported.

The most widely used AO orientation is anaerobic-anoxicaerobic (AAO) referred to as pre-denitrification. This orientation however entails effluent recycling as denitrification process is placed before nitrification [8]. Recycling of the effluent can be avoided by placing denitrification process after nitrification (post-denitrification). A significant factor for a successful post-denitrification is the availability of adequate carbon source. This is achieved by regulating C/N ratio which Fu et al suggested a ratio of 6 to be sufficient [11]. Regulating C/N ratio can be attained through adding an extra carbon which can either be an external carbon source like methanol, acetate, glucose [12], food waste [13] and ethanol [14] as many literatures suggest or internal carbon sources like readily biodegradable organics like volatile fatty acids (VFAs) obtained through sludge pretreatment [15]. The drawback of utilizing external carbon sources lies with not only raising operational cost [16] but also multiplies sludge production considerably [7]. Moreover, Xu et al reported that most applied internal carbon sources contain organic nitrogen such as protein which increased effluent nitrogen loading [7]. Therefore, it is

crucial to look for more efficient and yet economical carbon sources.

This study presented a reverse anaerobic-anoxic-aerobic (RAAO) process named as OAA process without effluent recycling. It utilized wood chips as internal carbon source. The logic behind the novelty is that wood is made up of carbon and once kept inside water for some-time it starts decaying making its carbon available for bacteria in water. Since the decaying wood supplies carbon to the bacteria, the carbon requirement for denitrifying bacteria is cut off and therefore neither effluent recycling nor addition of an external carbon source is essential.

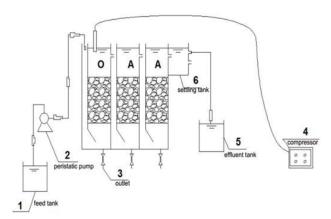


Fig. 1 Schematic lab-scale OAA baffled reactor

II. MATERIALS AND METHODS

A. Reactor Setup and Operation

The presented OAA baffled reactor had a rectangular shape and is divided into three compartments by vertical high/low baffles of 2.5 cm length (Fig. 1). 329 g of wood chips 1x1 cm was packed into each of the three compartments consuming 2.1 L of the reactor volume. The lower portion of the hanging baffles was bent at 45° to route the flow to the center of the up flow chamber, to achieve better contact of feed and bio-solids. The reactor has a total volume of 18 L however after addition of wood chips the volume dropped to 15.9 L. During operation, the reactor had a total functioning volume of 14.8 L. The reactor was made of a Plexiglas, and each of the three compartments had an equal volume of 6 L and dimensions ((length x width x depth) = $(10 \times 10 \times 54)$ cm). The last compartment was attached to a small settling tank of 1.5L of dimensions ((length x width x depth) = (12 x 10 x 24) cm). The overall configuration dimensions were ((length x width x depth) = $(37.5 \times 10 \times 54)$ cm). The air diffuser was used to supply DO to the first compartment maintaining aerobic conditions whereas the remaining two compartments were kept free of DO. In order to control temperature, the reactor was wound up with an electric wire connected to a temperature controller. The peristatic pump (Lange YZ515X) was used to pump the influent into the reactor.

3 g/L MLVSS inoculum obtained from a Harbin local sewage treatment plant with MLSS and MLVSS of 10.37 and 5.783 g/L was filled into the reactor and then topped up with raw sewage. After 48 h detention, the raw wastewater was

pumped into the first compartment stimulating the flow of water to the effluent tank. Throughout the experiment, temperature, HRT, DO and pH were maintained at $24 \pm 2\,^{\circ}C$, $24\,\text{h}$, less than 4 mg/L and 7 ± 1 respectively. The samples from the influent, each of the three compartments and the effluents were collected periodically for analysis.

B. Chemical Analysis

Parameters COD, NH₄⁺- N, nitrite nitrogen (NO₂⁻-N), nitrate nitrogen (NO₃⁻-N), and total phosphorus (TP) were detected using potassium dichromate titration, N- (1- naphthyl) ethylene diamine spectrophotometry, Nessler reagent spectrophotometric method, thymol and Potassium persulfate digestion ammonium molybdate spectrophotometric method respectively. Detection of pH was performed using pH meter (Shanghai Lei magnetic, PHS-3c), while DO using DO line detector (Taiwan Heng Xin, AZ 8403). All the parameters were analyzed based on China standard methods for water and wastewater monitoring and analytical. The samples were withdrawn for analysis at regular intervals (2 days) from inflow and from each compartment.

C. Wood Chips Preparation

The wood was obtained from multiple tree species around Harbin Institute of Technology (HIT) tree garden. The tree barks were removed to avoid blocking water movement inside the reactor. The wood was then cut into small pieces of approximately 1x1 cm and then packed into each of the three compartments where it was left to stand in clean water for one week before the experiment.

D.Experimental Water

The raw sewage was collected from Harbin community septic tank between 9:00 am and 10:30 am every day. Throughout the experiment the soluble COD ($_{\rm s}$ COD), NH_4^+-N , TP and TN concentrations fluctuated between 110 and 390 mg/L, 50 and 95 mg/L, 3 and 9 mg/L, 50 and 110 mg/L respectively and pH range was 7 ± 1 .

E. Estimating Parameters

1) Volumetric Hydraulic Load [17]

Volumetric Hydraulic Load explains the amount of wastewater applied regularly to the reactor, per unit of volume $(L, m^3/(m^3 \cdot d))$

$$L = \frac{Q}{V} \tag{1}$$

Q- Flow rate (m³/d), and V-total volume of the reactor (m³).

2) The HRT

The HRT expressed in days is presented as

$$HRT = \frac{v}{0} \tag{2}$$

3) Volumetric Organic Load (VOL)

Volumetric organic loading explains the amount of organic matter fed into the reactor per unit volume

$$L_0 = \frac{QS}{V} \tag{3}$$

 L_0 - COD volumetric organic load (kg/ (m³·d)), and S - influent substrate concentration (kg/m³).

4) Efficiency of Reactor

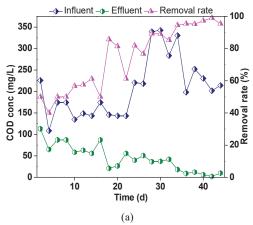
The removal efficiency of the reactor refers to the difference between the influent concentration and effluent concentration. The removal rate is presented by:

$$R = \frac{c_{inf} - c_{eff}}{c_{inf}} x \ 100\% \tag{4}$$

III. RESULTS AND DISCUSSION

Wood chips were presented in this study as an internal carbon source for denitrification process. The use of wood chips saved two main significances, first, it eliminated the need for effluent recycling and secondly, eliminated re-introduction of an external carbon requirement. These two significances simplify the process of nitrogen removal from wastewater.

A. Analysis of Overall COD Removal Efficiency



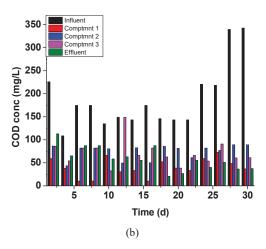


Fig. 2 Soluble sCOD concentrations (a) influent, effluent and removal rate of AO wood packed baffled reactor (b) COD removal rate in each compartment (day 16-44)

The COD was analyzed using potassium dichromate method which is preferred due to its higher oxidation rate and good reproducibility in determining the total organic matter in wastewater samples [18]. Fig. 2 (a) shows the influent, effluent and removal efficiency of COD in the system. Throughout the experiment, the influent COD was in range of 143-343 mg/L with an average of 229.19 mg/L. The effluent soluble COD was in range 2-87.2 mg/L with an average of 36.27 mg/L which is below stage one domestic wastewater discharge standard for China. The maximum COD removal efficiency attained was 99% with an overall average of 82.7%.

Fig. 2 (b) shows the soluble COD removal efficiency in each compartment. It can be seen that the influent COD concentration is the highest, removal efficiencies of 84-100% of the influent COD were realized in the first compartment. These high removal rates in the first compartment shows that an aerobic treatment is efficient in COD removal on its own in resemblance with studies [19], [20]. The COD concentration rise of about 20-93 mg/L was observed in the second compartment, this COD rise requires further investigation as it could be due to dead aerobic biomass or could be contributed by the wood chips. The COD in the third compartment and the effluent dropped slowly, this could be the result of the utilization done by denitrifying bacteria.

B. Analysis of NH₄⁺- N Removal Efficiency

The results for ammonia removal are presented in Fig. 3. The influent NH_4^+ - N concentration fluctuated between 30 and 80 mg/L with an average of 59.62 mg/L. The effluent a NH_4^+ - N concentration was in range of 7-30mg/L with the highest values recorded from the first week and decreased with time increase. The maximum NH_4^+ - N removal efficiency attained was 87% with a total average of 62%. The efficiency with which ammonia was removed increased slightly with time revealing the slow growth of ammonia oxidizing bacteria (AOB) as explained by [4], [21].

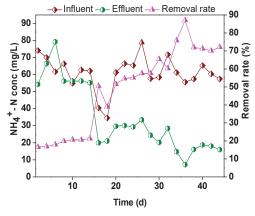


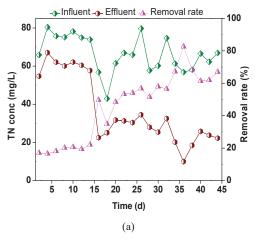
Fig. 3 NH₄⁺- N removal efficiency in an OAA baffled reactor

Ammonia removal from wastewater depends on the concentration of ammonia oxidizing bacteria (AOB). These bacteria are highly affected by DO concentration in the system (DO saturation rate for AOB is 0.2) [22]. Featuring the

observation by [23], small DO variation in the system was observed to affect nitrification rates, lower DO concentrations hindered nitrification whereas higher DO concentrations facilitated higher nitrification rates. Therefore, DO concentration in range of 3-4 mg DO/L proved to be sufficient in supporting nitrification process as also proposed by [23]. Therefore, these results show that the presented system is ideal for removal of NH₄⁺- N from domestic wastewater.

C. Analysis of TN Removal Efficiency

The removal efficiency of TN is presented in Fig. 4 (a). The influent TN concentration fluctuated from 35-79 mg TN/L with an average of 60.94 mg TN/L. The effluent TN concentration was 9-34 mg TN/L with an average of 25.14 mg TN/L which complied with stage 2 china TN discharge standards for domestic wastewater. The maximum TN removal rate attained was 83% with a total average of 58.1%.



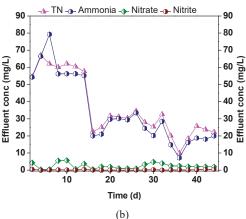


Fig. 4 Total ammonia removal efficiency (a) the influent, effluent and removal rate of TN (b) comparison of the influence of ammonia, nitrate and nitrite in the TN effluent concentration

Factors contributing to TN concentration in the effluent are presented in Fig. 4 (b). TN concentrations in the effluent is a function of organic nitrogen, ammonia, nitrite and nitrates. However, in this study, 96% of TN concentrations observed in

the effluent is largely contributed by ammonia concentrations. Nitrates and nitrites were also observed; however, its values were insignificant. These results suggest that huge amounts of ammonia oxidized into nitrate were converted into nitrogen gas (denitrified). Therefore, presented OAA configuration denitrified 97% of ammonia oxidized and therefore it is efficient in supporting denitrification.

D.Advantages of Wood and Comparison to Other Carbon Sources

1) Wood Chips in Aerobic Compartment

The wood packed in the aerobic zone acted as carriers to aerobic microorganisms. In the first compartment which was aerobic 84 to 100% of COD removal was attained. Wang et al compared the performance of a baffled reactor with and without burnt coke carrier, his results showed that without burnt coke carrier the reactor performance could only reach 96% COD removal rates whereas addition of burnt coke rises COD removal rates to 98.7% [23]. Ma et al. [24] also using Latex to enhance aerobic treatment observed 98% COD removal rates. Therefore, the removal efficiencies of 84 to 100% attained in the first compartment could be contributed by wood chips. Therefore, wood turns out to be the more efficient carrier in aerobic zones.

2) Wood Chips as an Extra Carbon Source

A comparative study of glucose, sodium acetate and food wastes efficiencies in denitrification process executed by Zhang et al. showed incomplete denitrification due to higher nitrate and nitrite concentrations in the effluent 22.2 mg/L and 6.6 mg/L respectively when glucose was used. When sodium acetate and food wastes was used no nitrite observed in both effluents and nitrate was very low less than 1mg/L. both sodium acetate and food wastes were able to achieve 98% of nitrogen removal [25]. Comparing this study to Zhang et al. external carbon sources study, effluent nitrites were insignificant whereas nitrates were less than 4 mg/L.

The cost spent for external and internal carbon sources reported by Xu et al. shows that the amount spent was 57.13 CNY/Kg N for C-acetate (external carbon source) and 54.48 CNY/Kg N for C-MHP (internal carbon source produced from sludge pretreatment by microwave-H₂O₂ process). 57.08%, and 69.66% nitrogen removal rates were attained when C-MHP and C-acetate was added respectively [7]. The author concludes that C-MHP has a good technical and economic feasibility to be used however it was observed that C-MHP addition limited the nitride removal. In comparison to the above studies, the use of wood in this study did not have any costs and although the maximum of 83% of TN was removed, 97% of oxidized ammonia was able to be denitrified. Therefore, wood can be an alternative, cost free internal carbon source.

IV. CONCLUSION

An integrative aerobic-anaerobic baffled reactor packed with wood chips was proposed to treat domestic wastewater at temperature 24±2 °C and HRT of 24 h. Domestic wastewater is characterized by C/N ratio of 3 which is low in supporting post-

denitrification without an extra carbon addition. Therefore, the objective of this study was to pack the reactor with wood chips as internal carbon source and then operate the reactor in OAA mode which is a post-denitrification mode without recycling the effluent or adding an external carbon source and observe its efficiency. The overall COD, NH₄⁺-N and TN removal efficiency fluctuated between 50-99%, 40-87% and 34-83% respectively and 97% of the oxidized ammonia was able to be denitrified. Therefore, wood proved to be an efficient and economical internal carbon source.

ACKNOWLEDGMENT

The authors appreciatively acknowledge the National Natural Science Foundation of China (Grant No. 51478141), the Major Science and Technology Program for Water Pollution Control and Management (No. 2013ZX07201007), and the State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology (Grant No. 2016DX06) for valuable financial support.

REFERENCES

- M. H. G. Nadais, M. I. A. Capela, L. M. G. Arroja, and Y.-T. Hung, "Anaerobic treatment of milk processing wastewater," in *Environmental Bioengineering*, ed: Springer, 2010, pp. 555-627.
- [2] W. P. Barber and D. C. Stuckey, "The use of the anaerobic baffled reactor (ABR) for wastewater treatment: A review," Water Research, vol. 33, 1999, pp. 1559-1578.
- [3] C. Mendes, K. Esquerre, and L. M. Queiroz, "Modeling simultaneous carbon and nitrogen removal (SCNR) in anaerobic/anoxic reactor treating domestic wastewater," *Journal of environmental management*, vol. 177, 2016, pp. 119-128.
- [4] M. Reza and M. A. Cuenca, "Nitrification and denitrifying phosphorus removal in an upright continuous flow reactor," *Water Science and Technology*, vol. 73, 2016, pp. 2093-2100.
- [5] T. Kuba, M. Van Loosdrecht, and J. Heijnen, "Phosphorus and nitrogen removal with minimal COD requirement by integration of denitrifying dephosphatation and nitrification in a two-sludge system," *Water research*, vol. 30, 1996, pp. 1702-1710.
- [6] M. R. Templeton and D. Butler, Introduction to Wastewater Treatment: Bookboon, 2011.
- [7] R. Xu, Y. Fan, Y. Wei, Y. Wang, N. Luo, M. Yang, et al., "Influence of carbon sources on nutrient removal in A 2/O-MBRs: Availability assessment of internal carbon source," *Journal of Environmental* Sciences, 2016.
- [8] Z. Zhou, X. Shen, L.-M. Jiang, Z. Wu, Z. Wang, W. Ren, et al., "Modeling of multimode anaerobic/anoxic/aerobic wastewater treatment process at low temperature for process optimization," *Chemical Engineering Journal*, vol. 281, 2015, pp. 644-650.
- [9] A. J. Justo, L. Junfeng, S. Lili, W. Haiman, M. R. Lorivi, M. O. Mohammed, et al., "Integrated expanded granular sludge bed and sequential batch reactor treating beet sugar industrial wastewater and recovering bioenergy," Environmental Science and Pollution Research, 2016, pp. 1-9.
- [10] H. Xiang, X. Li, S. Hojae, S. ZHANG, and Y. Dianhai, "Biological nutrient removal in a full scale anoxic/anaerobic/aerobic/pre-anoxic-MBR plant for low C/N ratio municipal wastewater treatment," *Chinese Journal of Chemical Engineering*, vol. 22, 2014, pp. 447-454.
- [11] Z. Fu, F. Yang, Y. An, and Y. Xue, "Simultaneous nitrification and denitrification coupled with phosphorus removal in a modified anoxic/oxic-membrane bioreactor (A/O-MBR)," *Biochemical Engineering Journal*, vol. 43, 2009, pp. 191-196.
- [12] D. Wang, X. Li, Q. Yang, W. Zheng, Y. Wu, T. Zeng, et al., "Improved biological phosphorus removal performance driven by the aerobic/extended-idle regime with propionate as the sole carbon source," water research, vol. 46, 2012, pp. 3868-3878.

- [13] A. Oehmen, P. C. Lemos, G. Carvalho, Z. Yuan, J. Keller, L. L. Blackall, et al., "Advances in enhanced biological phosphorus removal: from micro to macro scale," Water research, vol. 41, 2007, pp. 2271-2300.
- [14] U. Bracklow, A. Drews, R. Gnirss, S. Klamm, B. Lesjean, J. Stüber, et al., "Influence of sludge loadings and types of substrates on nutrients removal in MBRs," *Desalination*, vol. 250, 2010, pp. 734-739.
- [15] R. Xu, Q. Zhang, J. Tong, Y. Wei, and Y. Fan, "Internal carbon source from sludge pretreated by microwave-H2O2 for nutrient removal in A2/O-membrane bioreactors," *Environmental technology*, vol. 36, 2015, pp. 827-836.
- [16] P. Elefsiniotis and D. Li, "The effect of temperature and carbon source on denitrification using volatile fatty acids," *Biochemical Engineering Journal*, vol. 28, 2006, pp. 148-155.
- [17] J. L. C. Ladu, X. W. Lu, and A. M. Osman, "Integrated Processes of Anoxic/Oxic Bioreactor and Artificial Wetland for Rural Domestic Wastewater Treatment," in *Advanced Materials Research*, 2014, pp. 2526-2529
- [18] H. Wu, J. Zhang, H. H. Ngo, W. Guo, Z. Hu, S. Liang, et al., "A review on the sustainability of constructed wetlands for wastewater treatment: design and operation," *Bioresource technology*, vol. 175, 2015, pp. 594-601
- [19] A. L. Eusebi, N. Martin-Garcia, E. J. McAdam, B. Jefferson, J. N. Lester, and E. Cartmell, "Nitrogen removal from temperate anaerobic–aerobic two-stage biological systems: impact of reactor type and wastewater strength," *Journal of Chemical Technology and Biotechnology*, vol. 88, 2013, pp. 2107-2114.
- [20] Y. J. Chan, M. F. Chong, C. L. Law, and D. Hassell, "A review on anaerobic-aerobic treatment of industrial and municipal wastewater," *Chemical Engineering Journal*, vol. 155, 2009, pp. 1-18.
- [21] A. Katsogiannis, M. Kornaros, and G. Lyberatos, "Enhanced nitrogen removal in SBRs bypassing nitrate generation accomplished by multiple aerobic/anoxic phase pairs," *Water science and technology*, vol. 47, 2003, pp. 53-59.
- [22] C. W. Knapp and D. W. Graham, "Nitrite-oxidizing bacteria guild ecology associated with nitrification failure in a continuous-flow reactor," FEMS microbiology ecology, vol. 62, 2007, pp. 195-201.
- [23] R.-M. Wang, Y. Wang, G.-P. Ma, Y.-F. He, and Y.-Q. Zhao, "Efficiency of porous burnt-coke carrier on treatment of potato starch wastewater with an anaerobic-aerobic bioreactor," *Chemical Engineering Journal*, vol. 148, 2009, pp. 35-40.
- [24] G.-P. Ma, Y.-F. He, B.-Y. Yang, F.-R. Li, and R.-M. Wang, "Effect of latex carrier on treating potato starch wastewater in Anaerobic-aerobic Integrative Bioreactor," *Technology of Water Treatment*, vol. 33, 2007, pp. 75-77.
- [25] Y. Zhang, X. C. Wang, Z. Cheng, Y. Li, and J. Tang, "Effect of fermentation liquid from food waste as a carbon source for enhancing denitrification in wastewater treatment," *Chemosphere*, vol. 144, Feb 2016, pp. 689-96.