Research Article

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Effective Removal Of Nitrogen And Phosphorus From Saline Refinery Wastewater Utilizing A USBF Reactor.

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ABSTRACT

This study investigates the efficacy of a modified Upflow Sludge Blanket Filtration (USBF) system in the biological removal of nitrogen and phosphorus from saline wastewater generated by refineries. In this research, the experimental design was conducted utilizing two retention times of 12 and 24 hours, two nitrogen concentrations of 200 and 400 mg/l, a single phosphorus concentration of 20 mg/l, three levels of Chemical Oxygen Demand (COD) at 800, 1000, and 1200 mg/l, and three salt concentrations of 10, 12, and 20 g/l.
The findings reveal that the Chemical Oxygen Demand (COD) removal rate is exceptionally high, ranging from 89.4% to 98.9%. Furthermore, an increase in the nitrification rate correlates with a rise in the quantity of nitrate entering the anaerobic reactor, where the nitrate removal rate fluctuates between 47.9% and 82.8%. The percentage of phosphorus removal is observed to be between 81.7% and 96.1%. These results underscore the effectiveness of the modified Upflow Sludge Blanket Filtration (USBF) system in the treatment of industrial saline wastewater. The system's capability to concurrently eliminate organic pollutants, nitrogen, and phosphorus positions it as a viable solution for the treatment of complex refinery effluents.

Keywords : *USBF System, Saline Wastewater, Refinery, Biological Treatment of Nitrogen, Biological Treatment of Phosphorus.*

INTRODUCTION

This study investigates the deployment of an advanced Upflow Sludge Blanket Filtration (USBF) system for the biological remediation of nitrogen and phosphorus in high-salinity effluents generated by refineries. The water-intensive operations of the petroleum industry produce substantial quantities of contaminated wastewater, which pose significant threats to global water resources, human health, and ecosystems (Zhao et al. 2024; Ting et al., 2024). Conventional methods for salt removal, such as reverse osmosis and ion exchange, when utilized as biological pre-treatment strategies, often incur high costs and exhibit inadequate removal efficiencies (Delashoob et al., 2020). The persistent issues of eutrophication and unpleasant odors continue to endanger aquatic environments, exacerbated by increasing nutrient loads, particularly nitrogen and phosphorus (Lu et al., 2024). The research protocol encompassed a wide range of operational parameters, including hydraulic retention times of 12 and 24 hours, nitrogen concentrations of 200 and 400 mg/l, a constant phosphorus level of 20 mg/l, Chemical Oxygen Demand (COD) levels of 800, 1000, and 1200 mg/l, and salinity levels of 10, 12, and 20 g/l. Staphylococcus warneri ATCC 27863 (T) was utilized as the primary microbial agent in the treatment process, reflecting the growing focus on reducing phosphorus levels in aquatic systems (Cheng et al., 2023). The USBF system exhibited exceptional effectiveness, achieving COD reduction rates of up to 89%.
The Upflow Sludge Blanket Filtration (USBF) technology has attracted considerable interest as a novel solution to the challenges associated with saline wastewater treatment. This process signifies a significant advancement in the domain of environmental engineering, providing a distinctive blend of efficiency and cost-effectiveness in tackling the intricate issues related to high-salinity effluents. A pioneering study conducted by Jorge Ricardo Cunh in 2019 highlighted the potential for improving phosphorus removal in anaerobic treatment systems. The research concentrated on the anaerobic digestion of vacuum-collected black water (BW) utilizing an up-flow anaerobic sludge bed (UASB) reactor, a system closely associated with the USBF process. Cunh's findings indicated a substantial enhancement in total phosphorus (TP) retention upon the introduction of calcium into the treatment process (Ricardo Cunha et al., 2019). Additionally, Lea Chua Tan's 2018 research examined the removal of selenate (SeO42−), sulfate (SO42−), and nitrate (NO3−) at varying influent pH levels

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ranging from 7.0 to 5.0 and at a temperature of 20 °C in an up-flow anaerobic sludge blanket (UASB) reactor using lactate as an electron donor. The study achieved removal efficiencies of 43% for total selenate and 61% for dissolved selenate at a pH of 5.0. The USBF system is characterized by its elegant and straightforward design, with cost-effectiveness being one of its primary advantages.

Recent studies have proposed a kinetic model incorporating intrinsic reaction kinetics, as well as a simplified model with apparent reaction kinetics for denitrification in up-flow sludge bed (USB) reactors (Chua Tan et al., 2018). The simulated residual nitrate concentrations derived from the simplified model were found to be in good agreement with experimental data, and the results from the simplified model closely aligned with those obtained from the kinetic model (Chatterjee et al, 2016). Furthermore, Gholamreza Moussavi's 2015 research investigated the effects of inlet concentrations of total petroleum hydrocarbons (TPH), hydraulic retention time (HRT), salinity, and the presence or absence of nitrate on TPH biodegradation in a continuous up-flow sludge-blanket/fixed-film hybrid bioreactor (UAnSFB) operated under nitrate-reducing conditions. The study concluded that TPH biodegradation was unaffected by sodium chloride concentrations up to 30 g/l (Moussavi et al., 2015). The USBF process employs the phenomenon of sludge filtration (dewatering) within the sedimentation site. This sedimentation system is designed as a trapezoidal tank, through which the mixed fluid enters the sedimentation area from the lower section via a specialized baffle. This design facilitates flow entry hydraulics, coagulation, and flocculation at the sediment entry point. The trapezoidal configuration of the sedimentation tank enhances the cross-sectional area of the ascending fluid from the bottom to the top of the tank, thereby creating a gradual downward gradient within the vertical sedimentation site. Additionally, the sedimentation tank features a unique baffle at the inlet that promotes sludge sedimentation at the tank's base. The settled sludge can subsequently be utilized as return sludge to the aerobic or anoxic zones. In standard design practices, the return flow rate is established at four times the daily average wastewater inflow into the system. This elevated return sludge flow, originating from the lower section of the sedimentation tank, generates a downward velocity gradient within the tank. This gradient significantly influences the hydraulic flow dynamics and enhances both sedimentation and flocculation processes, thereby improving the overall efficiency of the tank in comparison to conventional sedimentation tanks. The return flow between the aerobic and anoxic zones supplies the essential carbon source required for nitrate reduction in the anoxic zone. Additionally, this return flow facilitates the migration of phosphorus-accumulating microorganisms from the aerobic zone back to the anoxic zone. The return flow rate is calibrated according to the ratios of biochemical oxygen demand (BOD), total phosphorus, and ammonia nitrogen present in the influent. Key design parameters for the system include the strength and biodegradability of the wastewater, temperature, and concentrations of BOD, nitrogen, and phosphorus at both the inlet and outlet. The typical hydraulic retention time (HRT) for the anoxic zone varies based on specific applications. The Design Solids Retention Time (SRT) is regulated in accordance with the temperature-dependent kinetics of BOD removal, nitrification, and ammonia nitrogen requirements in the effluent. Operational SRTs are generally maintained at the prevailing operating temperature to ensure adequate safety margins and to facilitate adjustments in system performance in response to variations in influent wastewater characteristics.

This study seeks to assess the efficacy of the USBF reactor in the removal of nitrogen and phosphorus from saline refinery wastewater, with the objective of establishing a robust and efficient methodology for managing these complex effluents. The results of this research may have considerable implications for the treatment of high-salinity industrial wastewater, particularly in areas facing water scarcity and stringent environmental regulations. Operational solids retention times (SRTs) are typically maintained to ensure optimal operating temperatures, thereby providing the necessary safety margins and the capacity to adapt system performance in response to variations in the characteristics of the incoming wastewater. Field research concerning the conditions of refinery effluent storage ponds in southern Iran commenced in 2022. In this investigation, a pilot study was conducted at two retention times of 12 and 24 hours, with nitrogen concentrations of 200 and 400 mg/l, a phosphorus concentration of 20 mg/l, and three levels of chemical oxygen demand (COD) at 800, 1000, and 1200 mg/l, alongside three salt concentrations of 10, 12, and 20 g/l. Additionally, the microorganism utilized in this study was *Staphylococcus warneri* ATCC 27863 (T) type.

MATERIALS AND METHODS

System Design

The modified Upflow Sludge Blanket Filtration (USBF) process represents a sophisticated advancement of traditional activated sludge treatment methodologies, incorporating significant innovations aimed at improving nutrient removal and overall operational efficiency. This system features an anoxic selective zone, an upstream flow sedimentation unit, and an aerobic zone, collectively fostering a synergistic environment conducive to comprehensive wastewater treatment. In this configuration, the anoxic zone fulfills a dual role. It serves as a selective environment that enhances sludge deposition while regulating the growth of filamentous

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microorganisms, which is essential for effective carbon removal. Furthermore, this zone creates optimal conditions for nitrate reduction and phosphorus removal, thereby facilitating both denitrification and biological phosphorus elimination processes. The treatment mechanism is characterized by a series of biochemical reactions occurring across the various zones. Within the aerobic zone, ammonia nitrogen is oxidized by Nitrosomonas and Nitrobacter bacteria, resulting in the formation of nitrate, which is subsequently recirculated to the anoxic zone via return flow for reduction under low anaerobic conditions. This denitrification process utilizes incoming biochemical oxygen demand (BOD) as an electron donor, effectively converting nitrate into nitrogen gas. Phosphorus removal within the USBF system is accomplished through a complex biological mechanism. In the anoxic zone, soluble BOD is converted into fermentation products that are selectively utilized by phosphorus-accumulating organisms (PAOs). These specialized microorganisms store phosphorus internally, and during the aerobic phase, a significant population of these phosphorus-storing bacteria, cultivated in the anoxic zone, assimilates soluble phosphorus. The excess phosphorus is ultimately removed from the system as surplus biological sludge. The efficiency of this phosphorus removal process is significantly influenced by the BOD/P ratio of the influent wastewater. The operational dynamics of the modified USBF system involve a meticulously coordinated flow pattern. Incoming wastewater is introduced into the anoxic zone of the bioreactor, where it is mixed with return activated sludge (RAS) from the bottom of the sedimentation tank. A low-speed stirrer operating at 25 rpm ensures thorough mixing and the maintenance of appropriate dissolved oxygen levels. The mixed liquor then progresses into the aeration section, adhering to a plug-flow reactor (PFR) model. Following aeration, a portion of the mixed liquor is directed into the sedimentation tank, where solids separation occurs through an upflow process, facilitated by a specifically designed velocity gradient. Clarified effluent overflows from the top, while settled sludge is recirculated to the anoxic zone at a 4:1 return ratio, thereby completing the internal cycle.This study provides a comprehensive analysis of various nitrogen and phosphorus removal systems, including lagoons, activated sludge, rotating biological contactors (RBC), extended aeration, oxidation ditches, and moving bed biofilm reactors (MBBR). The selection of the USBF system was based on its superior treatment efficiency and favorable cost-benefit ratio. The USBF system offers several advantages over fixed-bed alternatives, particularly in the treatment of saline wastewater, as it mitigates common operational challenges such as pressure drop, clogging, and channelization. Additionally, the USBF demonstrates enhanced removal rates due to a higher specific surface area available for microbial growth. The dynamic nature of the system ensures uniform distribution of the microbial biofilm, thereby facilitating improved mass transfer of substrates and oxygen, and optimizing reactor space utilization.

Wastewater Specifications

This study conducted an analysis of the influent to the wastewater retention basin of the Parsian refinery. Based on this analysis, an artificial feed was formulated in the laboratory of Sharif University of Technology. Urea was utilized as the nitrogen source, while KH_2PO_4 served as the phosphorus source. Additionally, a mixture of benzene, toluene, and xylene compounds was employed as carbon sources, simulating refinery sludge, in a ratio of 100:1:5. Given the insolubility of the carbon source materials, acetone was incorporated as a co-solvent to ensure the carbon remained in solution. The composition of the artificial feed, derived from the analysis of the Parsian Refinery wastewater, is detailed in **Table 1**.

Table 1. Artificial Feed Composition.

No.	Material	Amount (per 100l)
1	NaCl	1000 g
2	$(CNH_2)_2$	27 g
3	kH_2PO_4	6.2 g
4	Acetone	200 ml
5	Benzene	50 ml
6	Toluene	50 ml
7	Xylene	30 ml

Testing Process

In the initial phase of the study, microorganisms were subjected to artificial feed. Following the observed growth of microorganisms on the carrier surface at a concentration of 190 mg/(l.10 Packing),, the system was transitioned into a continuous state with a retention time of 24 hours and a flow rate of 18 ml/min. To mitigate the risk of backflow shock, the system was calibrated to maintain the same flow rate before being adjusted to 4 after a designated retention period. **Figure3** illustrates the phase during which the microorganisms adapted to phosphorus, nitrogen, and salt. In the second phase of the

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research, experiments were conducted utilizing two retention times of 12 and 24 hours, three concentrations of Chemical Oxygen Demand (COD) at 800, 1000, and 1200 mg/l, a single phosphorus concentration of 20 mg/l, three salt concentrations of 10, 12, and 20 g/l, and two ammonia concentrations of 200 and 400 mg/l, as depicted in **Figure 2**.

Figure 1. The schematic view of the USBF system; 1) Feed tank, 2) Feed pump, 3) Anoxic reactor, 4) Agitator, 5) Sludge blanket (USBF) separation, 6) Aerobic reactor, 7) Air pump, 8) Backflow pump, 9) Balancer tank, 10) USBF Clarifier.

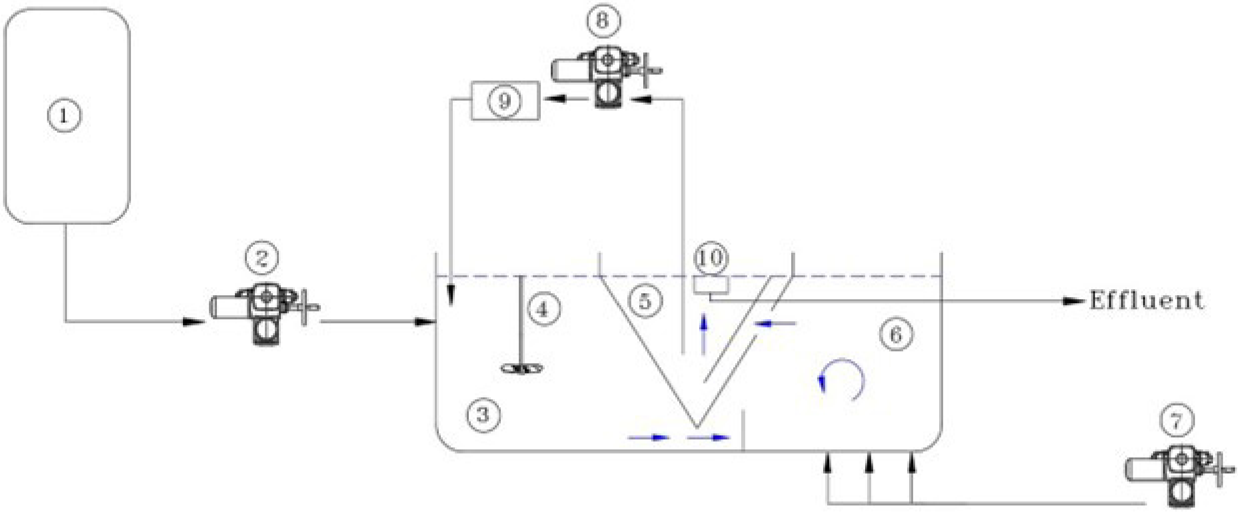
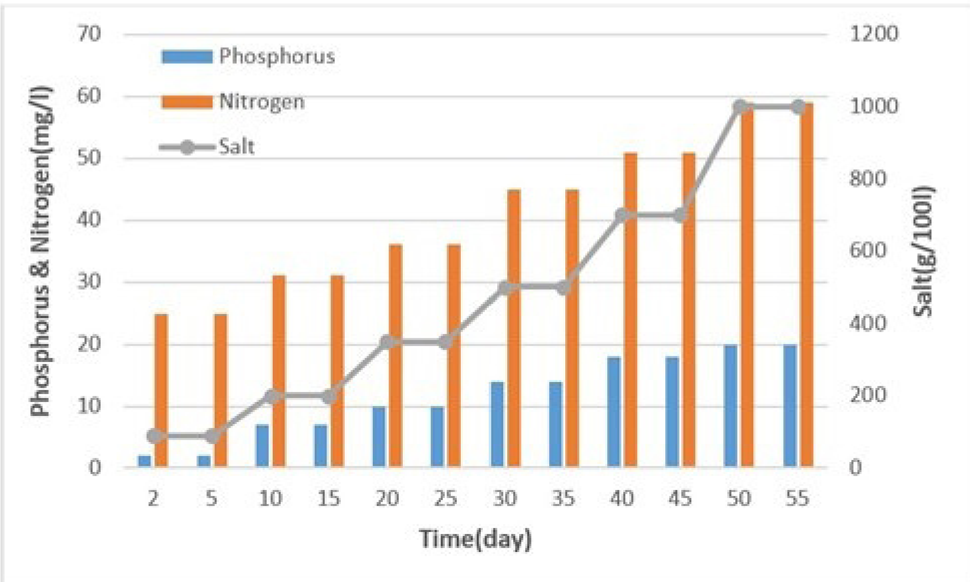


Figure 2. Adaptation of Microorganisms to phosphorus, nitrogen, and salt.



RESULTS

Investigating ammonia removal in the system by varying the inlet COD and different salt concentrations

The research examined the efficiency of ammonia removal under varying influent Chemical Oxygen Demand (COD) and salt concentrations. Over a hydraulic retention time (HRT) of 24 hours, ammonia removal efficiency varied between 82.8% and 47.9%, while for a 12-hour HRT, it ranged from 80.9% to 51.4%. The highest removal efficiencies were recorded at the lowest salt concentration, whereas the lowest efficiencies were noted at the highest salt concentration.

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A significant observation was the decline in ammonia removal efficiency with increasing influent COD across all ammonia concentrations. This trend can be attributed to the competition for oxygen between heterotrophic and nitrifying bacteria. As the concentration of organic matter (indicated by COD) increases, heterotrophic bacteria consume a greater amount of oxygen, thereby reducing the availability of oxygen for nitrifying bacteria to oxidize ammonia. Furthermore, the study indicated that elevated inlet ammonia concentrations exerted an inhibitory effect on nitrification, resulting in diminished ammonia removal rates. This finding aligns with previous research that suggests high ammonia levels can be detrimental to nitrifying bacteria. The effects of salinity were particularly significant. At a salt concentration of 10 g/l, the system's performance was comparable to that observed under non-saline conditions. However, at 12 g/l, minor changes were noted, and at 20 g/l, there were substantial reductions in performance. This indicates the presence of a salinity threshold between 10-12 g/l, beyond which considerable inhibition occurs, as illustrated in **Figure 3-8**.

Figure 3. Ammonia removal efficiency in relation to varying inlet COD concentrations, maintaining a 24-hour retention time and 10 g/l salt concentration.

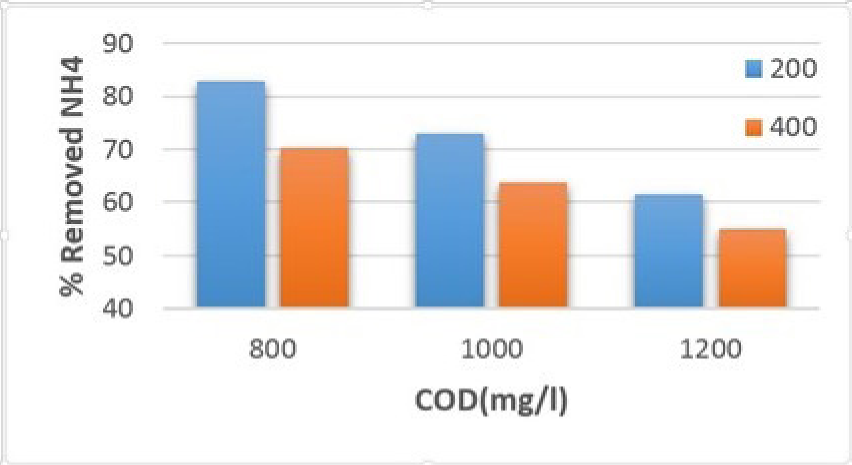

Figure 4. Ammonia removal based on inlet COD in different concentrations of inlet COD for 12-hour retention time and 10g/l salt concentration.

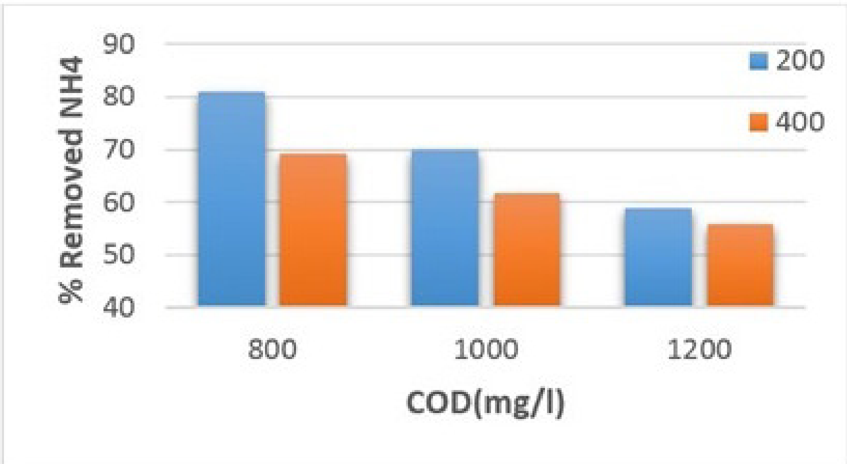

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Figure 5. Ammonia removal efficiency in relation to varying inlet COD concentrations, utilizing a 24-hour retention time and 12 g/l salt concentration.

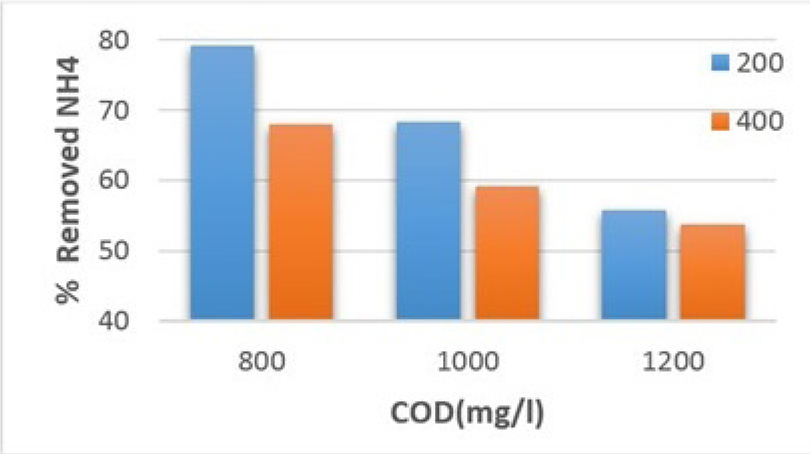

Figure 6. Ammonia removal effectiveness across various inlet COD concentrations, employing a 12-hour retention time and 12 g/l salt concentration.

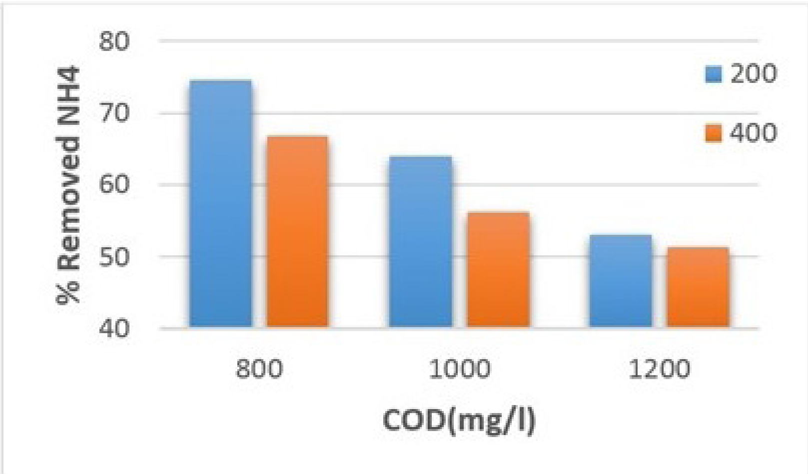

Figure 7. Ammonia removal efficiency in relation to varying inlet COD levels, using a 24-hour retention time and 20 g/l salt concentration.

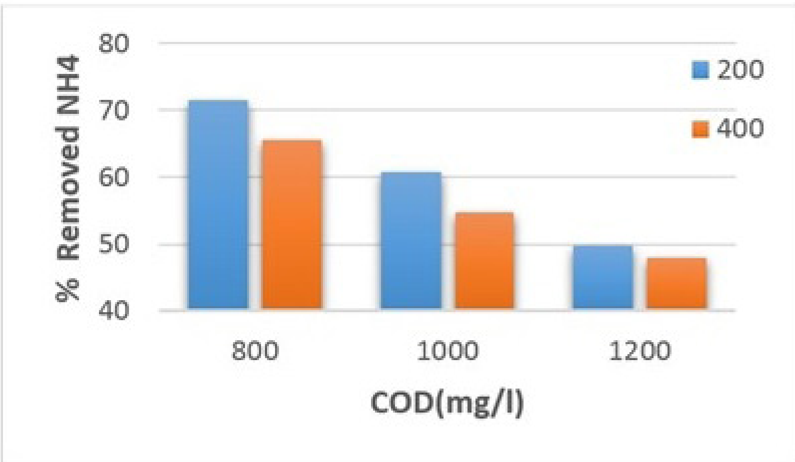

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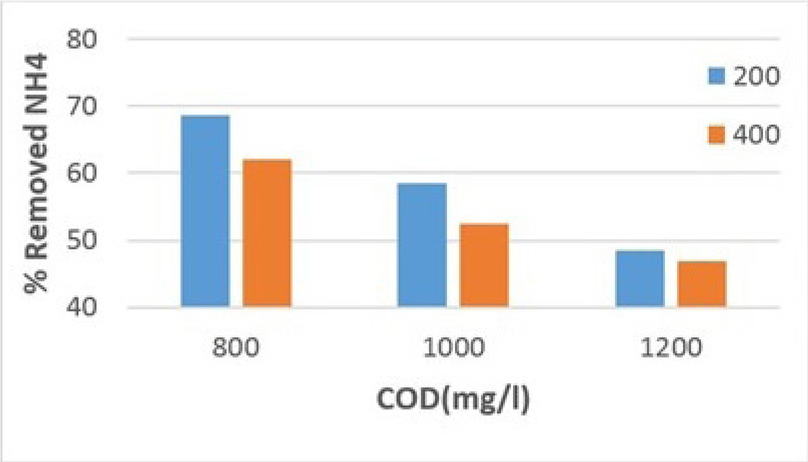
Figure 8. Ammonia removal performance across diverse inlet COD concentrations, utilizing a 12-hour retention time and 20 g/l salt concentration.



Investigating the nitrification rate by varying the inlet COD and different salt concentrations

The research indicated that an increase in inlet Chemical Oxygen Demand (COD) concentrations resulted in a decline in nitrification rates. This phenomenon can be attributed to the competitive advantage gained by heterotrophic organisms, which outcompete nitrifying bacteria for both oxygen and spatial resources. The nitrification factor, which is defined by the quantity of ammonia removed, exhibited a positive correlation with the concentrations of ammonia at the inlet.

Notably, at lower inlet ammonia concentrations, the reduction in nitrification rates in response to rising COD levels was less pronounced, as demonstrated by the more gradual slopes observed in the graphical representations. This observation implies that the influence of COD on nitrification is more pronounced at elevated ammonia loads, potentially due to the increased stress experienced by nitrifying bacteria. Furthermore, the study demonstrated that variations in Hydraulic Retention Time (HRT) had a significant impact on nitrification rates. Shorter HRTs, which are typically associated with higher inlet flow rates, appeared to hinder nitrification, particularly at increased ammonia concentrations. This underscores the necessity of achieving a balance between HRT and inlet nutrient loads to optimize nitrification efficiency, as illustrated in **Figures 9-14**.

Figure 9. Nitrification rates in relation to varying inlet COD and ammonia concentrations, employing a 24-hour retention time and 10 g/l salt concentration.

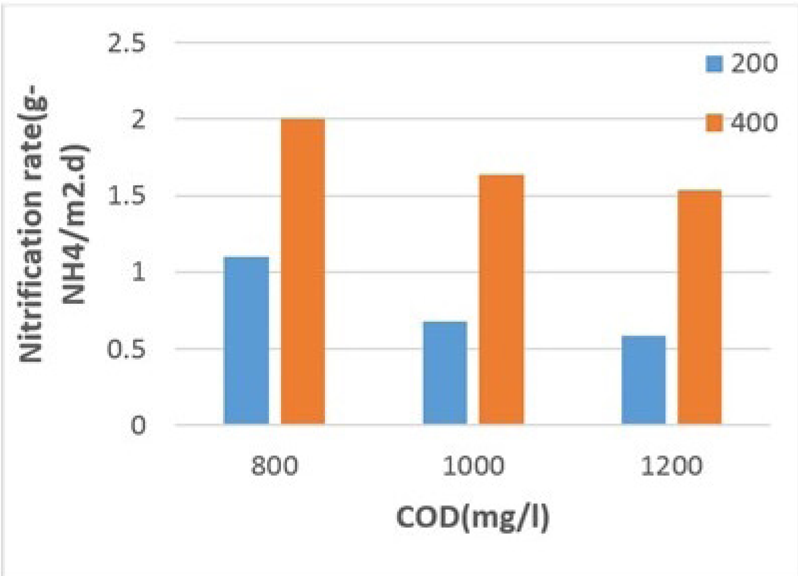


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Figure 10. Nitrification kinetics in response to varying inlet COD and ammonia levels, using a 12-hour retention time and 10 g/l salt concentration

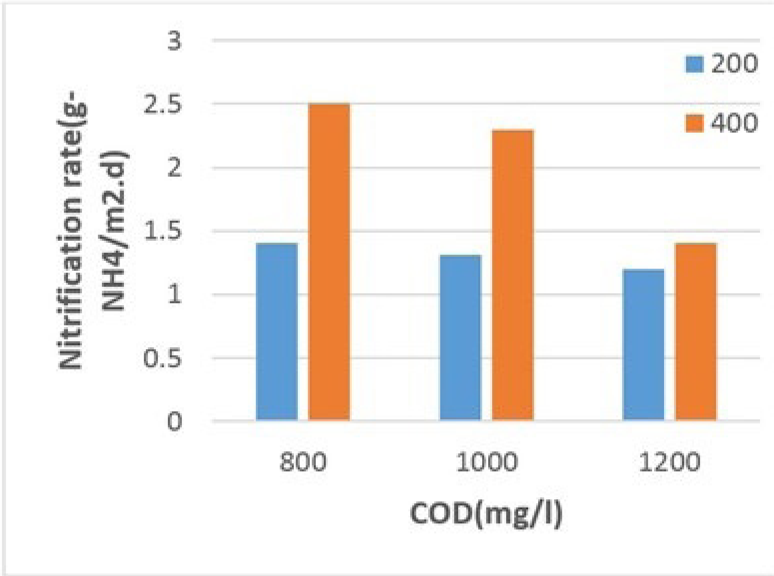

Figure 11. Nitrification rates as a function of inlet COD and ammonia concentrations, utilizing a 24-hour retention time and 12 g/l salt concentration

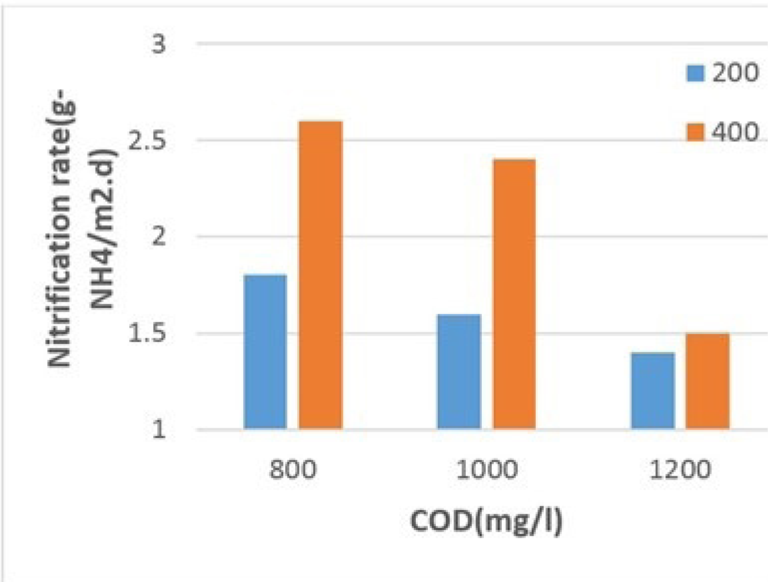

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Figure 12. Nitrification velocity in relation to varying inlet COD and ammonia levels, employing a 12-hour retention time and 12 g/l salt concentration

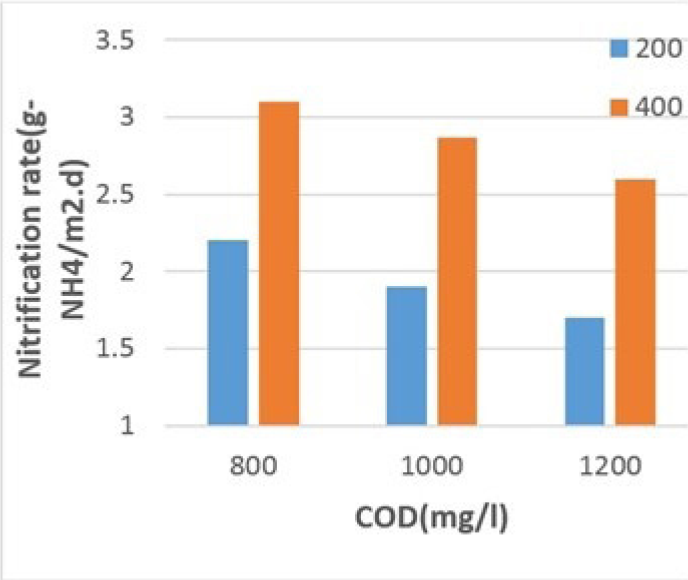


Figure 13. Nitrification rates in response to different inlet COD and ammonia concentrations, using a 24-hour retention time and 20 g/l salt concentration.

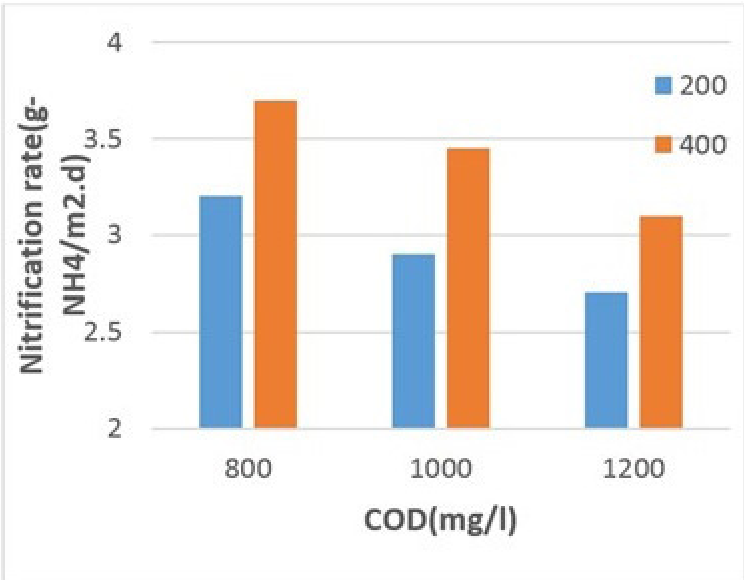
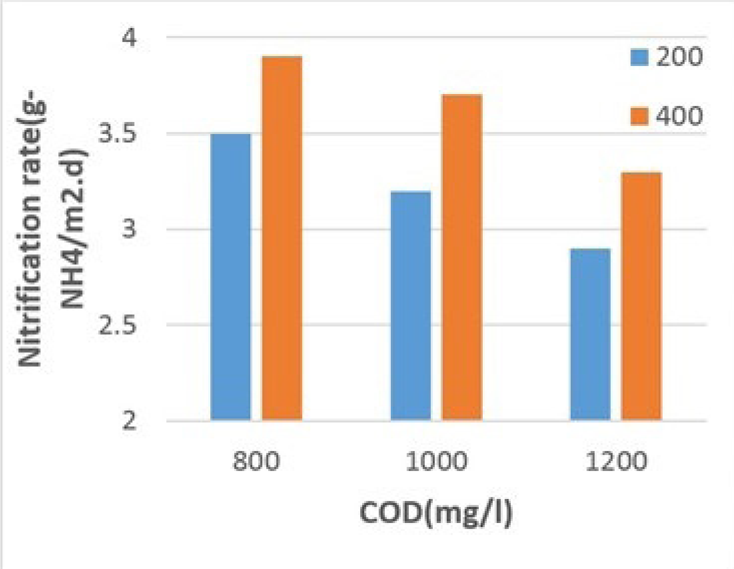


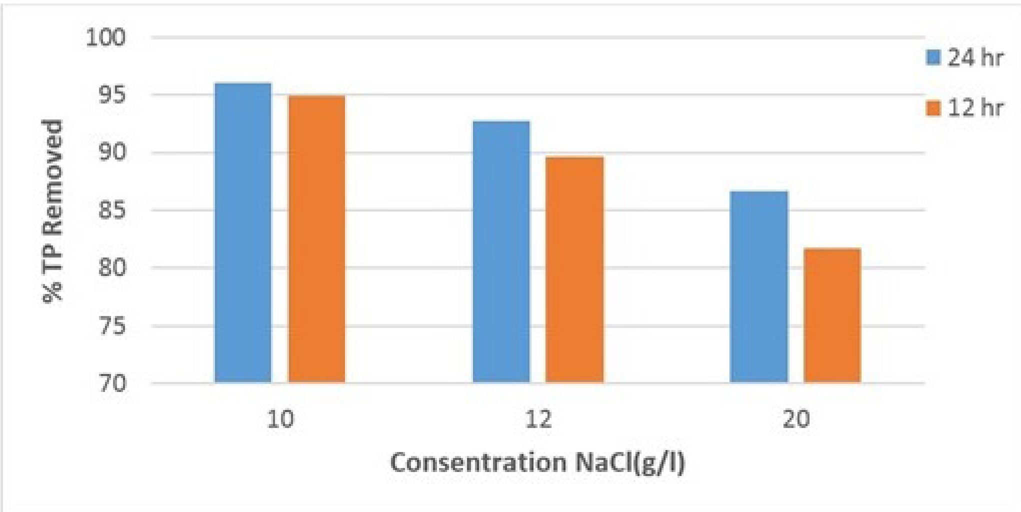
Figure 14. Nitrification kinetics across varying inlet COD and ammonia levels, employing a 12-hour retention time and 20 g/l salt concentration



Investigating changes in phosphorus concentration at retention time and different salt concentrations

Phosphorus removal was evaluated across various salt concentrations, revealing removal efficiencies ranging from 96.1% to 86.7% for a residence time of 24 hours, and from 94.9% to 81.7% for a residence time of 12 hours. These findings indicate that the highest removal percentages were achieved at the lowest salt concentrations, while the lowest removal percentages were observed at the highest salt concentrations. The efficacy of the biological phosphorus removal process is influenced by the growth dynamics of microorganisms, which exhibit varying behaviors across different phases to ensure survival and productivity. During the anaerobic phase, microorganisms consume carbon while simultaneously releasing orthophosphate. Conversely, in the aerobic phase, the dynamics shift, resulting in a greater uptake of phosphorus by microorganisms from the effluent than the amount released during the anaerobic phase. This study concluded that a residence time of 24 hours is optimal, as it provides sufficient opportunity for phosphate release. Furthermore, it was noted that an increase in salt concentration correlates with a decrease in the percentage of phosphate removal, as illustrated in **Figure 15**.

Figure 15. the total phosphorus removal efficiency under various salt concentrations and retention times.



Investigating changes in COD of the whole system

The removal efficiencies of Chemical Oxygen Demand (COD) varied between 98.9% and 91.8% for a 24-hour Hydraulic Retention Time (HRT), and between 97.6% and 89.4% for a 12-hour HRT. The findings of the study indicate that alterations in HRT within the examined range did not have a significant impact on COD removal rates. This observation implies that heterotrophic

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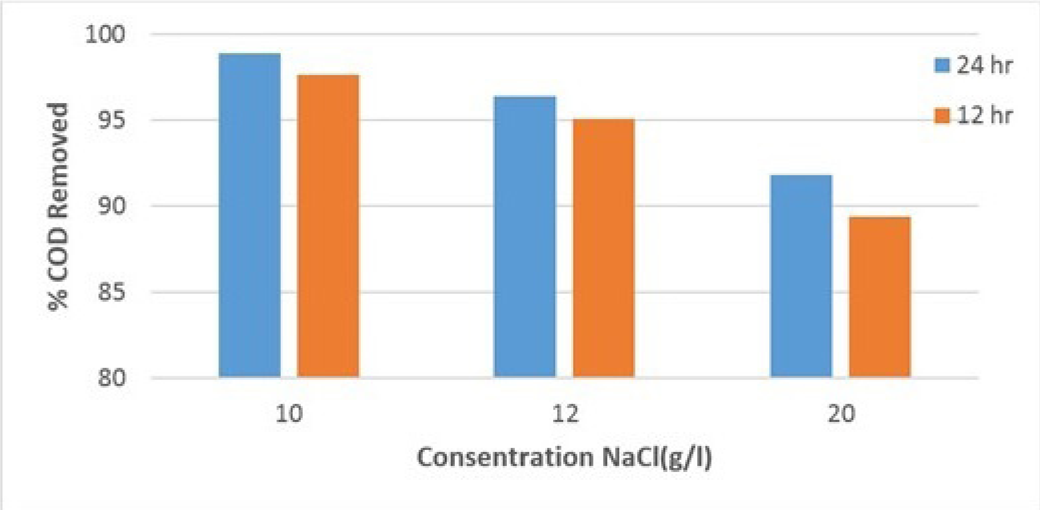
bacteria, which are primarily responsible for COD removal, exhibit a lower sensitivity to variations in HRT within this range when compared to nitrifying bacteria or Phosphate Accumulating Organisms (PAOs). Nevertheless, an increase in salinity was found to adversely affect COD removal efficiency, suggesting that while heterotrophic bacteria may demonstrate greater resilience to changes in HRT, they remain vulnerable to the effects of salt stress. (Refer to **Figure 16** for further details.)

Figure 16. COD removal rate of the whole system at different salt concentrations and retention times.



CONCLUSION

This study utilized a continuous Upflow Sludge Blanket Filtration (USBF) system to biologically treat phosphorus and nitrogen in saline wastewater originating from refineries. The results indicated remarkable Chemical Oxygen Demand (COD) removal efficiencies, ranging from 89.4% to 98.9%. However, the researchers noted a decline in efficiency under specific conditions, including increased influent COD concentration, reduced retention time, and elevated salt concentration. Notably, the system exhibited resilience in ammonia removal, with fluctuations in inlet ammonia concentration having a negligible effect on the removal process. The investigation also revealed a complex interplay between nitrification and denitrification processes; as nitrification rates increased, higher nitrate concentrations were observed in the anoxic reactor. Consequently, nitrate removal rates varied significantly, ranging from 47.9% to 82.8%. Similarly, phosphorus removal efficiency varied from 81.7% to 96.1%, with performance diminishing as retention time decreased and salt concentration increased. The researchers conducted a comparative analysis of the USBF system against existing wastewater treatment technologies, as detailed in **Table 2**. To substantiate their findings, they employed Pearson analysis, which indicated minimal data scattering, as presented in **Table3**. This statistical methodology enhances the credibility of the study's conclusions and highlights the system's potential for practical applications.

Table 2. Compares the USBF process with other wastewater treatment systems, analyzing design principles, operational parameters, and nutrient removal efficiencies to evaluate its effectiveness in managing industrial effluents.

TP removal (%)	TN removal (%)	SRT(d)	HRT(h)	Settlers	Reactor	Process	References
96.1	82.8	20	24	1	2	USBF	This study
99	90	20	24	1	3	AOA	Delashoob et al., 2020
98	75	15-20	9.31	1	4	Modified AOA	Zeng et al., 2011
94.1	86.8	36.3	10.2	1	6	BCFS	Puig et al., 2008
83	81	10-12	15	3	4	Modified DEPHANOX	Kapagiannidis et al., 2011
95	76	12	6	2	5	Modified DEPHANOX	Kim et al., 2009
95.3	76.7	15	9	1	3	Modified A2N	Hongbo et al., 2008

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Table 3. Statistical analysis.

	% TN removal	% TP removal
% TN removal Pearson Correlation	1	-0.200
Sig. (2-tailed)		0.635
N	7	7
% TP removal Pearson Correlation	-0.200	1
Sig.(2-tailed)	0.635	
N	7	7

In summary, this research demonstrates that the continuous USBF system is highly effective in achieving substantial removal rates of phosphorus and nitrogen from saline wastewater generated by refineries. The system's performance exceeds that of current technologies, as evidenced by the comparative analysis and statistical validation. These findings position the USBF system as a promising solution for the treatment of complex industrial wastewaters, particularly in contexts where high salinity and nutrient loads present significant challenges to conventional treatment methods.

Acknowledgement

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Data Availability Statement

Due to the nature of the research, due to ethical supporting data is not available.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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