Plant Archives Vol. 24, No. 2, 2024 pp. 1717-1724 e-ISSN:2581-6063 (online), ISSN:0972-5210

Plant Archives

Journal homepage: http://www.plantarchives.org
DOI Url : https://doi.org/10.51470/PLANTARCHIVES.2024.v24.no.2.245

ENHANCING PHOSPHORUS REMOVAL EFFICIENCY: MODELING INSIGHTS FOR PULP MILL WASTEWATER TREATMENT PLANTS

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(Date of Receiving-24-01-2024; Date of Acceptance-04-04-2024)

ABSTRACT

Using the program Bio Win, the efficacy of a pulp mill wastewater treatment plant (WWTP) was evaluated to offer possibilities for further lowering the phosphorus (P) concentration in the treated effluent. The pulp and paper effluent typically contains insufficient amounts of nutrients, the WWTP was not built with the ability to remove nutrients. But compared to other woods, the hardwood (Eucalyptus) processed in this plant has a higher P concentration, and some of that P ended up in the raw wastewater that needed to be treated. The Dutch STOWA methodology was followed in characterizing the wastewater. Historical data from various periods was utilized to validate the model after it had been calibrated. Both the plant's past performance and current operations may be described by the model. Additionally, the model was applied to assess various possible upgrading scenarios for the treatment plant to improve plant performance about P removal. The model indicated that introducing an anaerobic phase before the aerobic process was a realistic option that would help reduce the amount of total phosphorus (TP) in the treated effluent by about 58%. Chemical precipitation is another method that can lower the TP concentration below 0.1 mg/L. To validate the earlier recommendations of implementing enhanced biological and chemical P removal at such pulp mill wastewater treatment plants, additional research activities, such as pilot testing, might be required.

***Key words*:** Wastewater; Pulp and Paper industry; Phosphate; Treatment

Introduction

The majority of developing and underdeveloped nations have always struggled with air and water pollution (Toczyłowska-Mamiñska, 2017). One of the main causes of the smoke that clogs roads obstructs commercial aircraft operations from rural airports, and causes disorder on the roads is the burning of agricultural byproducts. Upon harvesting rice, farmers are left with rice straw that cannot be utilized for animal feed, thus they must burn it. This causes a sharp increase in CO_2 emissions, destructive fires, the formation of smog, and the repercussions of global warming. Wood, Paper, polypropylene, cardboard, and other single-use disposable plastic materials are used to make packing units all over the world (Jin *et al.*, 2013; Umar *et al.*, 2021).

According to Wagemann and tipkötter (2018), agricultural residue and rice strawcan be converted into a practical and affordable organic source. Sustainable paper packaging products are made in the pulp and paper biorefinery by simplifying complicated cellulose (Naushad, 2014; Nandiyanto *et al.*, 2017). The packing materials decompose naturally and are friendly to the environment. The products accommodate nearly all food types and are safe for both the environment and people. Nevertheless, pulp and paper biorefinery effluent (PPBE) is produced by this industry. Due of its toxicity, thermal impacts, low biodegradability, and aesthetic concerns, the PPBE has the potential to negatively impact both the vulnerable people and the surrounding aquatic ecosystem (Lawal *et al.*, 2021).

Wastewater discharge is a continuous threat to the world's supply of clean water since nutrients are wastewater contaminants that lower water quality by causing eutrophication in rivers, lakes, and seas (Jagaba *et al.*, 2022d). Recently, a number of treatment facilities have reduced the amount of nutrients in water and wastewater by increasing the biological nutrient removal processes. Increased biological phosphorus removal, biological nitrification, and biological denitrification are all included in biological nutrient removal (Nath and Bhakhar, 2011).

Reduction of the reflux ratio, elevation of wastewater temperature, and stream flow velocity can all improve the efficiency of ammonia removal (Faizal, 2014). Increasing the aeration period also results in a significantly higher elimination of ammonia. This may be explained by a decrease in total nitrogen increasing the rate of nitrification (Al-Rekabi *et al*., 2017). One crucial component of wastewater treatment systems is the removal of phosphorus (Ayaz *et al*., 2012; Bawiec, 2019). The hydraulic load and fractionation of daily bioreactors are the primary determinants of the efficacy of phosphorus removal (Dan *et al*., 2020). The need for ecologically sustainable wastewater treatment and reuse has grown in tandem with the ongoing depletion of water supplies. Consequently, effluent from the pulping process can be managed in a variety of methods (Assadi *et al*., 2018; Koupaie *et al*., 2013; Naushad *et al*., 2019).

It appears that effluent from pulp and paper mills has been treated biologically by aerobic, anaerobic, and aerobic/anaerobic procedures (Ahmed *et al*., 2020; Noor *et al*., 2021). Historically, industrial wastewater was treated using the traditional activated sludge technique. Its short hydraulic retention time (HRT), cheaper capital cost, generally more environmental friendliness, and less operational requirements than other physical processes are some of the reasons for this. The aeration tank, settling tank, and sludge recirculation comprise the AS system (Jagaba *et al*., 2022). According to Ng *et al*. (2021) it is the most used biological treatment technique. Depending on its design and intended usage, a wastewater treatment facility can remove both organic and biological inorganic materials (Mareai *et al*., 2020).

Paper and pulp wastewater have been the subject of some modelling attempts, but none of these models have included the properties of pulp mill effluent with a higher than typical P content. In conventional pulp and paper WWTPs, P is often dosed to meet the nutrient requirements for biological wastewater treatment processes that are healthy. Phosphate not dosage, needs to be eliminated in this investigation. This led to a comprehensive overhaul of the existing mathematical models structure and validation and calibration procedures. It is impossible to forecast future circumstances for assessing additional P removal options, such as the use of chemical and/or biological P removal, without a validated model. These studies directly addressed those demands.

The purpose of this study was to describe the performance and operation of a pulp mill WWTP with varying P-removal and P-assimilation capabilities. In order to demonstrate and ensure that the treated wastewater produced at the local WWTP complies satisfactorily with the effluent standards established by the local authorities, the ultimate goal of this research was to create a model of such a WWTP using the software BioWin.

Materials and Methods

The investigation centered on the scrutiny of the operational dynamics and efficacy of individual components within a wastewater treatment plant (WWTP). This scrutiny was based on historical data spanning from June to August 2020, from which an initial model of the plant was formulated. To evaluate the WWTP's performance, diverse mass balances were executed, focusing on parameters such as water flow, suspended solids, chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP).

From August 11 to 18, 2021, a comprehensive sampling campaign was carried out to assess wastewater characteristics, COD fractionation, and flow rates. Over a five-day period, representative composite samples were systematically collected at various junctures in the treatment process. These points included locations before screening and per-neutralization, post-cooling towers and pre-aeration basins, along aeration basins, after aeration basins, and at the final effluent discharge point. The primary objective was to characterize both raw and treated wastewater throughout the specified timeframe.

In this research, the aerobic degradation of organic matter and nutrient (phosphorus and nitrogen) requirements were calculated utilizing a steady-state model developed by Ekama and Wentzel in 2008. This model was initially applied before implementing the BioWin software. Simultaneously, a laboratory-based aerobic batch activity test was conducted using activated sludge biomass obtained from a specific pulp mill wastewater treatment plant (WWTP). The experimental setup involved aeration of mixed liquor activated sludge, followed by a six-hour activity test. Maintaining a pH of 7.5, a nitrification inhibitor (Allyl-N-thiourea) was introduced, along with urea and sodium tri-phosphate as nitrogen and phosphorus sources, respectively. In the experiment, influent wastewater was introduced to the batch reactor, and samples were gathered at various intervals, including the first 120 minutes, as well as after 24 and 48 hours.

The subsequent model construction and organization were facilitated using BioWin software. Following the structuring phase, a stepwise calibration process ensued, involving adjustments to wastewater fractions, as well as kinetic and stoichiometric model parameters. Calibration efforts persisted until the model accurately mirrored the WWTP parameters, ensuring a deviation of less than 10% from the periodically measured average values.

Result and Discussion

WWTP configuration

The wastewater treatment plant (WWTP) designed to handle an effluent flow of 35 m^3/air dry ton (ADT) from a pulpmill, primarily originating from the bleaching operation. The plant removed organic materials by mechanically pre-treating it and then using the CAS method. The treated effluent consisted of two streams: one with high solids concentration from various industrial processes, and another with low solids concentration from bleaching processes. The high solids effluent underwent coarse screening, primary clarification, and then was directed to safety and equalization basins. The low solids effluent, comprising two-thirds of the total wastewater, originated mainly from acid and alkaline bleaching processes, with smaller contributions from other plant sections.

In order to control pH and get ready for biological treatment, the first effluent stream was treated in the per-neutralization tank. The high solids effluent and neutralized wastewater were then mixed together in the equalization and safety basins to create a consistent mixture. After that, the effluent was sent to cooling towers to reduce its temperature from 55°C to about 30°C in order to avoid having any negative effects on the CAS system's biological performance. Before entering the CAS system, the effluent underwent post-neutralization for pH adjustment, which involved the addition of urea, after the temperature was adjusted. Two parallel aerobic reactors with a combined maximum capacity of 75,000 m^3 were used in the CAS process.

The dimensions of the aeration tanks were 9.5 metres in height, 42 metres in width, and 190 metres in length. Every one of the four air blowers in each of the two aeration basins had a 20,000 Nm^3/h capacity.

The variations in dissolved oxygen (DO) concentrations were primarily due to the degradation of organic matter within the reactor. However, the submerged air bubble diffuser effectively maintained adequate DO levels and promoted proper mixing. The CAS system operated with a solids retention time (SRT) of 32 days, functioning as an extended aeration process

Table 1: Comparing the wastewater characteristics of the studied pulp mill WWTP with other similar facilities, using historical data from the Yamuna-Nagar plant spanning June to August 2020.

Parameter	Bleached Kraft Pulp mill (Yamuna-nagar) June to August 2020 (avg)	Bleached Kraft Pulp mill (Uruguay)
BOD_5	1115	911
COD	1839	1588
Phosphate	15	3.7
Nitrate	30	6.5
Ammonia	3	0.2
TSS	390	220

to reduce sludge production. The treated wastewater from the aerobic reactors underwent clarification in two parallel secondary clarifies, each with a 65-meter diameter. Subsequently, the treated and clarified effluent was discharged.

A portion of the sedimented sludge in the secondary settling tank underwent re-circulation to the aeration basins, labeled as Re-circulation of Activated Sludge (RAS), while the remaining fraction was directed to sludge handling facilities as Waste Activated Sludge (WAS). Two extraction lines facilitated the transfer of WAS to a side-stream sludge treatment line, amounting to around 5% of the influent flowrate. Throughout the sampling period, the observed RAS flowrate averaged 115% relative to the influent flowrate, fluctuating between 110% and 150%. The aeration basins maintained a biomass concentration of about 5.3 grams of mixed liquor suspended solids (MLSS) per liter.

With a combined capacity of 33.8 total dry solids (t DS) per day, the two belt filter presses were used to dewater the sludge that had settled in the primary sedimentation tank. The solids were moved to a specified disposal location in forest plantings after being dewatered. A gravity table and a decanter centrifuge with a daily capacity of 17 t DS were also used to treat the WAS. The wastewater treatment plant's headworks received filtrates from both the main and WAS dewatering procedures (WWTP).

Collection of Data , prelim characterization, and WWTP assessment

The flow-rate of Influent to the wastewater treatment plant (WWTP) was assessed by analyzing historic data from June to Aug 2020. During this period, the flow-rates remained relatively constant, with an average Influent flow-rate of 59,538 ± 8,731 m^3/d. The minand maxvalues were 52, 491 ± 2,335 m3/d and 73,971 ± 671

m^3/d. The composition of wastewater in the pulp mill Yamuna-Nagar, obtained from historical data during the same period, is presented in Table 1. This data is compared to the wastewater compositions of other pulp mill.

In the examination of pulp mill wastewater, it was found that the organic matter concentrations, including the biodegradable fraction were higher. The COD/BOD ratio in the pulp mill wastewater from Uruguay indicated a significant presence of biodegradable components. Municipal wastewater exhibited higher TN and TP concentrations, while the wastewater of pulp mill in Yamuna-Nagar had lower levels. The characteristics of this wastewater in Yamuna-Nagar were similar to those reported in other PPM wastewaters regarding NH_4^+, TSS, TN, COD. However, BOD_5 and TP were notably higher, potentially due to the higher phosphorus content in the wood used as raw material.

Table 2 displays the results of assessing the performance of a pulp mill (WWTP) in Yamuna-Nagar based on historical data gathered between June and December 2020. The evaluation indicates that the WWTP employed state-of-the-art technologies during that period and demonstrated commendable performance across the evaluated parameters.

Wastewater fractionation and characterization

Table 3 displays the constituent COD (chemical oxygen demand) fractions in the influent wastewater of a pulp mill in Yamuna-Nagar, as observed during a sampling campaign conducted from October 11 to 18, 2021. The COD distribution is compared with that other pulp and paper mill wastewater (Diez *et al.*, 2002; Keskitalo and Leiviskä, 2010; Meijer and Brdjanovic, 2012). Additionally, the characteristics of both influent and effluent wastewater from the pulp mill's wastewater treatment plant (WWTP) in Yamuna-Nagar are detailed in Table 4, based on samples collected during the same October 2021 campaign.

Steady-state model and aerobic batch activity test

In the initial assessment of the pulp mill wastewater treatment plant (WWTP) performance, a steady-state model was employed. This model utilized data from the previous section, which included the characterization and fractionation of wastewater, to evaluate its effectiveness. The assessment was conducted at a temperature of 30°C and a solid retention time (SRT) of 32 days, with stoichiometric and kinetic parameters aligned with Henze *et al.*, (2008). The model considered an average annual influent wastewater temperature of 30°C, and adjustments were made for the temperature-dependent kinetic endogenous respiration rate (bH) coefficient, as per Henze *et al.*, (2008). It's noted that high temperatures, such as 30°C, may adversely impact the performance of enhanced biological phosphorus removal (EBPR) due to the preferential growth of glycogen accumulating

Table 2: June to December 2021, the removal efficiencies were monitored at a flow rate of 71,418 m^3/d on average.

Sampling	COD	BOD5	Phosphate	Nitrate	TSS
Influent	1533mg/L	1029mg/L	35mg/L	15mg/L	417mg/L
Effluent	356mg/L	15.6mg/L	9.3mg/L	3.6mg/L	24mg/L
Removal efficiency	76.8%	98.5%	73.5%	76%	94.3%

Table 3: Thepulp mill WWTP COD influent fractions based on a sample program conducted from October 11–18, 2021, and a comparison of those fractions with industrial wastewater from pulp and paper mill (Morse *et al.*, 1998).

Influent fractions	Pulp and paper mill (Yamuna-Nagar)	Pulp mill (Uruguary)	Stora Enso Fine Paper Oulu pulp mill (Finland)
Particulate slowly and biodegradable COD (XS)	0.28	0.19	0.35
Soluble inert COD (SI)	0.24	0.14	0.26
Particulate inert COD (XI)	0.09	0.03	0.08
Total soluble readily biodegradable COD (SS)	0.83	0.64	0.31

Table 4: Wastewater characterizationduring sampling campaign from October 11 to 18th of 2021. (Unit g/ m^3).

Parameters	Influent	Effluent
Total Influent COD	1839	384.3
Influent COD soluble	1503	341.5
Influent BOD_{10}	1115	9.3
Influent BOD_{10} soluble	941	-
Total Nitrogen	30	1.93
Total Kjehldahl Nitrogen	30	1.18
Ammonium	3	0.08
Total Phosphorus	15	0.5
Total Posphorus soluble	14	0.4
Orto-phosphate	9.8	0.4
Total Suspended Solid	390	10.3
Inorganic Suspended Solid	56.1	-
Volatile Suspended Solid	130	-

organisms (GAOs) over phosphorus accumulating organisms (PAOs), as reported by various studies (Whang and Park, 2002, 2006; Wang *et al*., 2020; López-Vázquez *et al*., 2008).

In the study using the steady-state model, it was found that the nutrient concentrations in the influent pulp mill wastewater were not enough to meet the growth needs of the microorganisms responsible for organic matter removal. This suggests a potential nutrient limitation in the plant. Nevertheless, the effluent characteristics and removal efficiencies of the pulp mill wastewater treatment plant (WWTP) show that it was operating efficiently despite this.

An aerobic batch activity test was used to ascertain the precise nutritional requirements. Based on the elimination of organic matter (COD), phosphorus (P), and nitrogen (N), the nutrient requirements were calculated. The amount of phosphorus removed for biomass growth (fP) was around half of what was found in municipal wastewaters, at 0.015 mg P/mg VSS. In a similar vein, biomass nitrogen requirements (fN) were calculated to be less than the average value for municipal wastewater treatment plants, at about 0.039 mg N/ mgVSS.

To design a plant-wide model for the pilot plant, Kazadi *et al*., 2019 modified the Benchmark Simulation Model No. 2 (BSM2) and enhanced the physico-chemical framework (PCF). The model and measured phosphorus concentrations agreed well, with differences as minor as 5-10% between the two sets of data. It was discovered that operational parameters like FeP molar ratios and environmental factors like pH had an impact on the amount of dissolved phosphate in the effluent. Because of a step decrease in the dosage of $FeSO_4$, the calibrated model's time constant for simultaneous precipitation was roughly 5 days. This suggests a multi-step process comprising dissolution, oxidation, precipitation, ageing, adsorption, and co-precipitation, which is responsible for the sluggish dynamic response. Because Fe_2oxidises quickly to Fe3, the model predicts that aerobic tanks are the best places to dose $FeSO_4$.

Model calibration and validation

In BioWin, the layout of the wastewater treatment plant (WWTP) for the pulp mill was designed. An initial simulation using influent data from wastewater characterization was conducted, employing default values for kinetic and stoichiometric parameters in BioWin. Subsequently, a step-wise calibration, following the methods of Hulsbeek *et al*., (2002), and Meijer and Brdjanovic (2012) was carried out. In order to correlate the COD biodegradable and non-biodegradable particle fractions (XS and XI) with the plant's MLSS concentration, a number of parameters were changed during calibration. The nutrients content in biomass, other particulate compound and endogenous residues reduced, leading to adjustments in the fP (phosphorus fraction) to 0.005 and fN (nitrogen fraction) to 0.034. These adjustments aimed to account for the lower nutrients absorptionas certained in the sludge of the pulp mill wastewater treatment plant (WWTP), as well as the lower concentrations of effluent NH_4^+ and PO_4^{3-}. The calibration phase involved modifying certain values, with only three kinetic parameters adjusted for adequate description of effluent TSS, TN, TP, NH_4^+, COD, and NO_3^- concentration. Notably, the calibrated stoichiometric parameter fP and fN differed significantly from the default values in BioWin for conventional municipal wastewaters, being much lower at 0.004 mg P/mg COD and 0.025 mg N/mg COD, respectively, compared to BioWin's defaults of 0.022 mg P/mg COD and 0.07 mg N/mg COD.

To validate the limited nutrient assimilation observed, a comprehensive mass balance, involving N and P analysis in both primary and secondary sludges, was carried out. The mass balance data confirmed the modest requirements for nutrient absorption. This suggests that microbial populations with low nutritional requirements may have developed as a result of the remarkably low N/COD and P/COD ratios in the pulp mill wastewater. Past studies on activated sludge treatment for pulp and paper mill wastewater have also noted low nutrient concentrations in such effluents. For instance, Cabrera (2017) found a BOD:N:P ratio of 100: (1-2):(0.15-0.3) in wood processing industry effluents, suggesting the common need for supplementary nutrient addition. Other researchers, such as Davis (2005) and Bashaar (2004), have highlighted the deficiency of N and P in pulp and paper industry effluents, necessitating the addition of these nutrients for optimal microbial growth. Notably, the nutrient requirements observed in this study for pulp mill wastewater in Yamuna-Nagar were even lower than those reported for other pulp mill wastewaters.

The calibrated model's predictions were verified using data from two distinct operational periods (June-August 2020 and September-December 2020) of the pulp mill WWTP. The effluent model's predictions were compared to the average values from these periods, demonstrating a satisfactory alignment between the model predictions and the actual plant performance. While certain parameters, like TSS and NO_3^-, exhibited notable differences, it is important to note that these concentrations are exceptionally low. Even a minimal increase of 0.1 mg/L could result in a seemingly significant deviation between predicted and measured values. Despite these small differences, they were considered acceptable, and it was assumed that the model had been adequately validated.

Using a sequencing batch reactor (SBR) at 20°C, Roots *et al.*, 2020 investigated the effective and dependable removal of total nitrogen (N), phosphorus (P), and chemical oxygen demand (COD) from wastewater. To achieve consistent removal rates and high effluent quality, the process combines occasional oxicanoxic phases during aeration with anaerobic-aerobic cycling. The study shows that, as opposed to simultaneous nitrification-denitrification, energy and carbon efficient nitrification-denitritation achieves the combined removal of N and P.P elimination is combined with a powerful biological process.

According to the research, even in the presence of changing influent concentrations and rainy weather flows, consistent and high removal rates can be achieved without the use of exogenous chemicals. The suppression of nitrite oxidizing organism (NOO) activity is confirmed by process modelling, which also explains how nitrite draw-down through denitrification works.

Future Recommendations

- **Pilot Testing and Validation:** It is recommended to carry out extensive pilot testing at the pulp mill wastewater treatment plant (WWTP) in order to verify the suggested improvements in the removal of phosphorus both chemically and biologically. This real-world validation will offer vital information about the planned changes' efficiency, practicability, and possible drawbacks.
- **Optimization and Fine-Tuning:** The suggested anaerobic phase introduction and chemical precipitation techniques should be optimized with more study. The most economical and efficient parameters will be found by fine-tuning these procedures through monitoring and testing, guaranteeing excellent performance in removing phosphorus.
- **Long-Term Performance Review:** To gauge the improved treatment plant's ongoing efficacy, consider a long-term performance review. Long-term, ongoing observation will shed light on the durability and dependability of the phosphorus removal improvements that have been put in place.
- **Life Cycle Analysis:** To evaluate the suggested upgrades' overall environmental impact and sustainability, conduct a life cycle analysis. To provide a comprehensive picture of the environmental footprint, this analysis should take into account not only the efficiency of phosphorus removal but other variables like energy usage, greenhouse gas emissions, and resource utilization.
- **Technological Developments:** Keep up with developments in wastewater treatment technology. Examine new developments and methods that may improve the effectiveness of phosphorus removal in pulp mill WWTPs. Work together with academics and business leaders to investigate innovative approaches that support sustainable objectives.
- **Information Transfer and Industry use:** Promote the use of effective phosphorus removal techniques in the pulp and paper sector by facilitating information transfer. Work together with politicians, regulatory agencies, and industry stakeholders to encourage the adoption of environmentally friendly practices and distribute research findings.
- **Economic Viability evaluation**: To determine whether the suggested upgrades are cost-effective, perform an economic viability evaluation. Think about things like the initial outlay, ongoing expenses, and possible savings from better phosphorus removal. Decision-makers will need to know this information when deciding whether to apply the recommended improvements.

Conclusion

In conclusion, this research used the BioWin program to evaluate the efficiency of a pulp mill wastewater treatment plant (WWTP) with a specific focus on reducing phosphorus (P) concentrations in the treated effluent. Despite the WWTP being initially designed without nutrient removal capabilities, the study identified the higher phosphorus concentration in hard wood (*Eucalyptus*) processed at the plant as a significant contributor to the raw wastewater. The Dutch STOWA methodology was applied to characterize the wastewater, and historical data validated the calibrated model, allowing for a comprehensive assessment of both past and present plant performance.

The findings from the model suggested potential strategies for upgrading the treatment plant, emphasizing the introduction of an anaerobic phase before the aerobic process as a realistic option to achieve a substantial reduction of approximately 58% in total phosphorus (TP) concentrations in the treated effluent. Furthermore, chemical precipitation emerged as a viable method capable of lowering TP concentrations below 0.1 mg/L. However, the implementation of these strategies, particularly enhanced biological and chemical P removal, may necessitate additional research activities such as pilot testing to validate their practicality and effectiveness in real-world scenarios.

This research contributes valuable insights into improving the phosphorus removal efficiency of pulp mill WWTPs, offering concrete recommendations for potential upgrades. The identified strategies present promising opportunities to enhance the overall environmental performance of such facilities. Nevertheless, the transition from theoretical models to practical implementation requires careful consideration, and further research activities are recommended to validate and optimize these proposed upgrades in real-world conditions. Ultimately, this study lays the foundation for advancing sustainable practices in pulp mill wastewater treatment, emphasizing the importance of ongoing research and innovation in addressing environmental challenges.

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