Integration of Membrane Bioreactor and Nanofiltration for the Treatment Process of Real Hospital Wastewater in Ho Chi Minh City, Vietnam

Authors:

Thanh Tran, Thanh Binh Nguyen, Huu Loc Ho, Duc Anh Le, Tri Duc Lam, Duy Chinh Nguyen, Anh Tuan Hoang, Trung Sy Do, Luong Hoang, Trinh Duy Nguyen, Long Giang Bach

Date Submitted: 2019-07-05

Keywords: pathogen contents, nitrogen compounds, total phosphorus, chemical oxygen demand, hospital wastewater, membrane bioreactor, nanofiltration

Abstract:

Hospital wastewater contains pharmaceutical residues, chemicals, and pathogens that cause coloration and nourish pathogenic microorganisms. The objective of the study was to evaluate the effectiveness of a medical wastewater treatment system at Military Hospital 175 (Ho Chi Minh City, Vietnam) that combined a membrane bioreactor (MBR) system with nanofiltration (NF). The influent of the system was the wastewater discharged from the operating rooms of the hospital. The system has a capacity of 50 L/day and operates at three organic load rates (OLR) of 0.5, 1.5 and 2.5 kgCOD/m3day (COD: Chemical oxygen demand), in which each load rate operates for 40 days. The results showed that most nutritional criteria generally achieved positive results. Specifically, the average COD removal was shown to be consistently high throughout the three phases at 94%, 93.3%, and 92.7%, respectively. For removal of nitrogen, the system demonstrated efficiencies of 75%, 79%, and 83%, respectively, to three phases. The log removal value (LRV) for Escherichia coli and coliform bacteria were higher than four throughout the study period. The average removal efficiency for color and total iron was approximately 98% and 99%, respectively. The water quality after treatment, especially after NF, meets the Vietnamese standard of grade A. The arrangement in which the MBR preceded NF was also found to limit the amount of soil and solids entering subsequent treatment, which therefore improved the efficiency of NF, as demonstrated by the stability of post-NF transmembrane pressures throughout three cycles renewed by two backwashes.

Record Type: Published Article

Submitted To: LAPSE (Living Archive for Process Systems Engineering)

Citation (overall record, always the latest version):	LAPSE:2019.0616
Citation (this specific file, latest version):	LAPSE:2019.0616-1
Citation (this specific file, this version):	LAPSE:2019.0616-1v1

DOI of Published Version: https://doi.org/10.3390/pr7030123

License: Creative Commons Attribution 4.0 International (CC BY 4.0)



Article

Integration of Membrane Bioreactor and Nanofiltration for the Treatment Process of Real Hospital Wastewater in Ho Chi Minh City, Vietnam

Thanh Tran¹, Thanh Binh Nguyen¹, Huu Loc Ho^{1,2}, Duc Anh Le¹, Tri Duc Lam¹, Duy Chinh Nguyen¹, Anh Tuan Hoang³, Trung Sy Do⁴, Luong Hoang⁵, Trinh Duy Nguyen¹, and Long Giang Bach^{1,*}

- ¹ NTT Hi-Tech Institute, Nguyen Tat Thanh University, Ho Chi Minh 700000, Vietnam; thanhtran2710@gmail.com (T.T.); ntbinh28081996@gmail.com (T.B.N); huuloc20686@gmail.com (H.L.H.); leducanh1707@gmail.com (D.A.L.); ltduc3096816@gmail.com (T.D.L.); ndchinh@ntt.edu.vn (D.C.N.); ndtrinh@ntt.edu.vn (T.D.N.)
- ² Nanyang Environment and Water Research Institute, Nanyang Technological University, Singapore 639798, Singapore
- ³ Military Hospital 175, Ho Chi Minh 700000, Vietnam; hoanganhtuan175@gmail.com
- ⁴ Institute of Chemistry, Vietnam Academy of Science and Technology, Hanoi 100000, Vietnam; dosyvhh@gmail.com
- ⁵ Institute of Environmental Technology, Vietnam Academy of Science and Technology, Hanoi 100000, Vietnam; indenpendenthanoivn@yahoo.com
- * Correspondence: blgiang@ntt.edu.vn; Tel.: +84-969294297

Received: 29 January 2019; Accepted: 20 February 2019; Published: 27 February 2019



Abstract: Hospital wastewater contains pharmaceutical residues, chemicals, and pathogens that cause coloration and nourish pathogenic microorganisms. The objective of the study was to evaluate the effectiveness of a medical wastewater treatment system at Military Hospital 175 (Ho Chi Minh City, Vietnam) that combined a membrane bioreactor (MBR) system with nanofiltration (NF). The influent of the system was the wastewater discharged from the operating rooms of the hospital. The system has a capacity of 50 L/day and operates at three organic load rates (OLR) of 0.5, 1.5 and 2.5 kgCOD/m³day (COD: Chemical oxygen demand), in which each load rate operates for 40 days. The results showed that most nutritional criteria generally achieved positive results. Specifically, the average COD removal was shown to be consistently high throughout the three phases at 94%, 93.3%, and 92.7%, respectively. For removal of nitrogen, the system demonstrated efficiencies of 75%, 79%, and 83%, respectively, to three phases. The log removal value (LRV) for Escherichia coli and coliform bacteria were higher than four throughout the study period. The average removal efficiency for color and total iron was approximately 98% and 99%, respectively. The water quality after treatment, especially after NF, meets the Vietnamese standard of grade A. The arrangement in which the MBR preceded NF was also found to limit the amount of soil and solids entering subsequent treatment, which therefore improved the efficiency of NF, as demonstrated by the stability of post-NF transmembrane pressures throughout three cycles renewed by two backwashes.

Keywords: nanofiltration; membrane bioreactor; hospital wastewater; chemical oxygen demand; total phosphorus; nitrogen compounds; pathogen contents

1. Introduction

In hospitals of various countries including Vietnam, China, Japan, and Greece [1–3], wastewater is biologically treated on-site or pre-treated. The primary benefit of this method is two-fold.



First, it prevents hospital wastewater from diluting with wastewater in urban sewage. Second, it also mitigates the risk of leakage of hazardous wastewater into the environment. However, on-site water treatment has to deal with substantial and denser pollutants [4,5]. Since large hospitals use an abundant amount of disinfectants for daily operations [6,7], their generated wastewater is characterized by the distinct yellow color and a large concentration of toxic or persistent substances, carrying dangerous hazards including infected pathogens, toxic chemicals, radioactive elements, organics and nutrients [8]. The release of these mixtures to the environment will adversely impact the ecosystem and human health, causing biological imbalances and environmental damage [9]. On the other hand, an excess amount of disinfectants also hinders the growth of sludge biomass in biological processes, in turn reducing the efficiency of the treatment system.

In general, the treatment of wastewater and of hospital effluent, in particular, have to deal with a number of significant contaminants including phosphorous (P), nitrogen (N), iron (Fe), and non-essential heavy metals. The two first notable contaminants, phosphorus and nitrogen, are widely released to waterways in high quantities from agricultural activities and manufacturing industries and could impair the water ecosystems in two ways. First, these elements are mainly responsible for the occurrence of eutrophication, the phenomenon in which plants and algae excessively grow on a water surface, blocking sunlight and in turn causing oxygen depletion [10–12]. Second, substances that are made up of nitrogen, such as ammonia, nitrate, and nitrite, are capable of forming highly complex carcinogens when released into the water source. The third pollutant, iron, is characteristic of hospital wastewater due to the presence of pharmaceutical compounds. High total iron in water could cause numerous consequences. First, iron and iron precipitates, in conjunction with filthy collecting and disorganized classification process, give water undesirable color and taste. Second, iron accumulated in pipes could reduce the efficiency of water treatment and the distribution system. Third, iron causes indirect harms to the ecosystem by nourishing bacteria in water [13]. Other concerning contaminants include non-essential heavy metals, especially the five elements: As (arsenic), Pb (lead), Hg (mercury), Cr (chromium), and Cd (cadmium), which have been reported to influence cellular organelles and cell components, causing carcinogenesis or apoptosis even in low concentrations [14]. Treatment of these heavy metals is also strongly accentuated in hospitals, where X-rays and laboratory-related operations generate high amounts of Pb, Hg, and Cd.

Membrane bioreactor (MBR) is a technology of interest in wastewater treatment for the purpose of re-using treated water and improving the sustainability of the water environment. Compared with conventional aerobic treatment, MBR could limit the amount of generated sludge, which is toxic and dangerous for the environment. The technology is compact and has many advantages over traditional biological systems, including high output water quality, excellent bacteriological separation, improved sludge retention time, high biomass content, and operational flexibility [15]. The effectiveness of MBR, especially in complement with other treatment methods such as nanofiltration (NF) and reverse osmosis (RO), has been demonstrated in various studies. For example, in a treatment model that incorporated MBR and RO processes to treat landfill leachate, the final treatment efficiencies for the chemical oxygen demand (COD), biological oxygen demand (BOD), suspension solids, NH_4^+ -N, and $NO_3^{--}N$ were reported to reach 97%, 97%, 99%, 96%, and 93%, respectively [16]. Also for treatment of landfill leachate, evaluation of another pilot-scale MBR system integrated with granular active carbon (GAC) found that the integrated system achieved a significantly higher efficiency at above 80% for both the COD and NH₃⁺-N. In addition, the study also suggested the addition of NF or RO for further advanced treatment of post-MBR effluents. To be specific, the NF showed advantages over RO when it came to removing Pb (95% versus 2%), Cr (95% versus 2%) and coloration (93% versus 41%) from the MBR effluents [17]. For the removal of organic trace pollutants, an MBR-NF/RO combination also showed excellent efficiency in another laboratory-scale study [18]. Of 40 selected organic contaminants, 37 of which were removed by NF with an efficiency higher than 90%. Furthermore, both NF and RO in the study also demonstrated a high removal rate (>80%) of compounds that persist through MBR including DEET, Atrazine, Meprobamate, Primidone, and Dilantin. This result is strongly supported by another

MBR-NF/RO evaluation, where 27 common antibiotics and pharmaceuticals were tested, showing an excellent removal rate of the system for all compounds (95% on average) and a high removal rate (>50%) of NF for Metoprolol, Norfloxacin, Enrofloxacin, and Carbamazepine compounds [19]. With regard to the removal of pathogens, NF has been shown to be able to remove *Bacillus subtilis* bacteria and MS2 bacteriophage at very high efficiencies, ranging from 3.3 to 6.3 log removal value (LRV) for *Bacillus subtilis* bacteria and from 4.1 to 4.5 LRV for MS2 bacteriophage [20].

For treatment of hospital wastewater, previous studies have specifically suggested that a microscale MBR was insufficient to eliminate most medicines (especially iodinated contrast media), antibiotics and anti-epilepsy drugs, and that additional filtration (NF or RO) is required to effectively remove these compounds [21,22]. Among the two advanced filtration methods, NF is preferred to RO in the treatment of hospital wastewater for various reasons. First, since NF could retain high-valence ions while allowing monovalent ions to pass, it is particularly suitable for water softening. Second, in comparison with RO, NF requires lower operation pressures (<40 atm). Third, NF allows backwashing, which could significantly reduce the frequency of membrane fouling [21]. Fourth, NF is particularly effective when it comes to removing pharmaceuticals and personal care products [23]. Lastly, the costs of implementation and operation of an NF system are also lower than those of an RO system. To the best of our knowledge, most MBR-NF systems that have been evaluated only involve treatment of landfill [17], municipal wastewater [24,25], or laboratory-simulated sewage [18]. The examination of integrated systems that exclusively treat wastewater from healthcare facilities is therefore lacking. One exception is a particular study, where a laboratory-scale MBR equipped with nanomembranes was evaluated, which showed high-performance in terms of the COD, NH_3^+-N , $NO_2^{-}-N$, $NO_3^{-}-N$, and $PO_4^{3-}-P$ removal [8]. However, other indicators such as total iron, coloration and bacteria were missing in that study.

Given these notions, the present study aims to investigate the effectiveness of a pilot-scale wastewater treatment system that links a membrane biofilm reactor with a separate NF system. We consider various indicators including the COD, total phosphate (TP), concentrations of nitrogen compounds, total iron, coloration, and amounts of *Escherichia coli* and coliform bacteria. In addition, two backwashing operations were also conducted and post-backflow pressure was measured. The results act as a precursor for future investigation regarding advanced treatments of pharmaceutical compounds and feasibility assessment of the MBR-NF system in larger scale.

2. Materials and Methods

2.1. Experimental Model

The integrated system employed in this study was a pilot-scale MBR-NF model specifically used to treat wastewater discharged from operating rooms in the Military Hospital 175 (Nguyen Thai Son St., Go Vap District, Ho Chi Minh City, Vietnam). The MBR-NF model consisted of six main components, including a 180 L inlet tank, an anoxic tank, a membrane biofilter, a submersible membrane module with a surface area of 0.9 m², an intermediate tank and the NF system. The system is depicted in Figure 1, where the injection pump, anoxic tank mixer, circulation pump, MF pump, NF feed pump, and backwash pump are represented as P-01/02, Mixer, P-05, P-03/04, P-06, and P-07, respectively.



Figure 1. Experimental model process of membrane bioreactor nanofiltration system.

The inlet pump was a Pulsafeeder KX100 (AC 23 V) dosing pump: Qmax = 15.75 L/hour, a power of 161 W, and a pressure of 4.2 bar. The outlet pump was a blower Resun-ACO 006 (AC230 V): Qmax = 90 L/min, a power of 80W, and a pressure of 0.03 Mpa.

The membrane filter used in this study was a membrane microfilter (MF) module of hollow fiber (Motimo, China) with a throughput of 12–18 $1/m^2$.h, a filter size of 0.1 µm, and an operation pressure of ≤ 40 kPa. The inner and outer diameters of the MF pore were 0.6 mm and 1.1 mm, respectively. The ratio of activation to the rest period of the membranes (in minutes) was 10:1. The NF was the FILMTECTM with a pore size of 1 nm. The NF membrane was a thin polymer with a flux of 6–8 1/h.

The seed sludge was gathered from another MBR system that treated hazardous wastewater in Ho Chi Minh City. The mixed liquor suspended solids (MLSS) with a concentration of approximately 5100 mg/L was deposited into the MBR tank. The average ratio of the mixed liquor volatile suspended solids (MLVSS) to MLSS of the seed sludge was 0.79.

To maintain dissolved oxygen (DO) of $\geq 4.0 \text{ mg/L}$ during operation, this study used an air supply equipment with a flow of 1.7 m³/h. The filter efficiency was equivalent to 15–20 l/ (m².h). Air was supplied to microorganisms to break down organic matter, promoting nitrification and reducing membrane congestion. The initial MLSS concentration in the reactor was maintained at 10,000 mg/L minimum.

The model was evaluated for total operating efficiency in 120 days, exclusive of the adaptation time of two weeks. Each organic load rate was held for 40 days. Details of operating parameters are shown in Table 1.

Table 1. Operating parameters of MBR-NF model during testing period.

Parameter	Unit –	Organic Load Rate		
		OLR ₁	OLR ₂	OLR ₃
Q	litre/day	2.4	6.3	10.5
F/M	day ⁻¹	0.003 ± 0.0001	0.006 ± 0.0009	0.010 ± 0.00011
OLR	kgCOD/m ³ day	0.5	1.5	2.5
HRT	Hour	24	8.0	4.8
MLSS	mg/L	5041.6 ± 137.9	5101.9 ± 209.5	5110.8 ± 241
pН	-	7.2 ± 0.4	7.5 ± 0.5	7.5 ± 0.4
DO	mg/L	5.2 ± 0.5	4.7 ± 0.2	4.9 ± 0.3
SRT	Days	40	40	40

2.2. Sampling and Analysis

The wastewater samples were taken once a day in the morning at three locations: The inlet, intermediate (after aerobic treatment with MF) and the outlet tank (after NF), and were put in cold

storage. The process of sampling is in accordance with the Tieu Chuan Viet Nam (TCVN) Vietnamese standards 4556-88 [26]. Characteristic to the operating room, the color of the effluent was dark yellow. The temperature of the wastewater ranged from 25 to 32 °C. The pH of the wastewater ranged from 6.82 to 8.21 and the concentration of pollutants was measured in mg/L (physical–chemical parameters). To be specific, pollutants and corresponding concentration ranges were as follows: the COD (475–868 mg/L), TSS (26.8–90.5 mg/L), total Kjeldahl Nitrogen, or TKN (19.7–57.3 mg/L), NH₄-N (1.6 to 16.1 mg/L), TP (1.3–5.5 mg/L), NO₂ (0–0.7 mg/L), NO₃ (0.1–2.6 mg/L), Fe (1.4–7.3 mg/L), coloration (503–1358 Pt-Co), coliform (1,50E+6–4,30E+6) and *E. coli* (1,40E+4–4,50E+4). These parameters were consistent with Verlicchi et al., who analyzed hospital wastewater and reported a significant difference in the COD, suspended solids, BOD, and chlorides between the hospital and local wastewaters [5].

The examined indicators and methods of analysis are as follows: The COD (SMEWW 5220 C:2012), TP (SMEWW 4500-P B&E:2012), coloration (SMEWW 2120B:2012), NH₄ (TCVN 5988:1995), NO₂ (SMEWW 4500-NO₂-.B:2012), NO₃ (SMEWW 4500-NO3-.E:2012), total iron (TCVN 6177:1996), and *E. coli* and coliform (SMEWW 9222). QCVN28:2010/MONRE of Vietnam was the reference standard in this study.

The MBR membrane was replaced after 130 days of stable operation. The stability of toxins in color and total iron removal was achieved after 25 days of solid retention time (SRT) of MBR-NF. Moreover, the membrane module (MF) was backwashed, followed by physical and chemical cleaning after the transmembrane pressure (TMP) reached 40 kPa, to recover the membrane permeability. The TMP after the NF membrane was also recorded. The sludge concentration was calculated through the MLSS concentration of the biomass.

3. Results and Discussion

3.1. Characterization of the Active Sludge

Figure 2 presents the MLSS and MLVSS in the MBR tank during three operating load rates. The average concentration of the sludge in the MBR tank was nearly 5000 mg/L. Overall, in each period of load, the trend of the MLSS was found to increase. As the load rate changed to heavier loads on the day 41 and 81, the biomass of the sludge was observed to decline slightly due to the shock load, as demonstrated by the drops in the MLSS. The range of the MLVSS/MLSS ratio was in agreement with that in the conventional active sludge tank (0.7–0.8).



Figure 2. The concentration of mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) of the sludge throughout three load rates.

Active sludge in the MBR-NF was recovered from a treatment plant for hazardous wastewater. After a short period of operation, the sludge was found to be well-adapted to the wastewater, as indicated by the color change from dark brown to light brown and the declining sludge volume index (SVI) over time as displayed in Figure 3. The SVI in this study fluctuated from 76–113 mL/g, which was in line with the ideal SVI range of an aerotank of 50–90 mL/g.



Figure 3. The sludge volume index (SVI) throughout three load rates.

3.2. Treatment Efficiencies of Organic Matter and Nutrients

Figure 4 presents three organic loads of 0.5, 1.5 and 2.5 kgCOD/m³.day and their corresponding COD indicators. Overall, the MBR-NF system maintained a COD removal efficiency of above 90% during the study period, with average COD removal efficiencies in the three load rates of 94%, 93.3%, and 92.7%, respectively. The average COD output throughout the three operating phases reached 39 ± 9 mg/L, which was relatively consistent and was below the QCVN 28 standard of 50 mg/L, in spite of significant variations of the COD in input wastewater, from 457 to 868 mg/L. This was similar to the results from Nguyen et al. where the COD removal rate ranged from 96 to 97% in different sponge MBRs [27]. The difference in the COD efficiencies of the three organic load rates was statistically significant using the Bonferroni test at a 5% confidence interval.

The described MBR–NF system had two main advantages. First, the system was irresponsive to the shock load, as demonstrated by the stability in the COD output, regardless of the fluctuation in the COD input. Second, despite the impermissible and highly fluctuated COD efficiency of the outflow of the aerobic tank with MF membranes, the minimum COD removal efficiency of the NF was still higher than 80% and the average COD in the effluent was less than 25 mg/L. These results also match with those of Choi et al., where the total organic carbon (TOC) concentration after MBR-NF ranged between 0.5 and 2.0 mg/L and the average COD was 5 mg/L [28]. Accordingly, the low TOC and COD could be explained by the smaller size of NF membrane pores in comparison with those of the MF membrane, which suggests an improved COD removal of the system that combines the NF with the MBR.

Figure 5 details the patterns of TP removal of the integrated system with respect to the time of treatment. Overall, the P concentration of the final effluent met both QCVN standards and was well maintained in the range from 0.008 to 0.69 mg/L, with an average concentration of 0.25 ± 0.1 mg/L. This is in line with Wang et al., who reported that the mean final TP of the MBR-NF reached 0.34 mg/L [29]. TP removal efficiencies of MBR-NF were 95.27%, 94.57% and 96.39%, corresponding to a TP input ranging from 1.4 to 5.4 mg/L (average of 3.5 ± 0.8 mg/L) in three operating phases. This high TP removal efficiency was similar to Cartagena et al., who found that MBR with subsequent NF can achieve a TP reduction of 99% [25].



Figure 4. The chemical oxygen demand (COD) removal efficiency at three organic load rates of the MBR-NF system.



Figure 5. The total phosphate (TP) removal efficiency at three organic load rates in duration 120 days of MBR-NF system.

Table 2 summarizes concentrations of nitrogen compounds corresponding to different load rates. In wastewater, although decomposed products such as ammonium, nitrite, and nitrate play a pivotal role in the water ecosystem, and nitrogen content is essential to oxidation capacity of the activated sludge, excessive amounts of such substances may form carcinogens. In the study of Al Qarni et al., and Liu, it is reported that the maximum removal of NH₄⁺ and NH₃ reached 99% and 95% for aerobic and MBR processes, respectively [6,30]. This is comparable with the results of this study, where nitrification and nitrate reduction achieved more than 80%, corresponding to 4.9 g/L of average total nitrogen in the effluent. Regarding the TN output, the results showed that the relative stability of the TN output was maintained with increasing load rates (8.23 \pm 1.61 mg/L on average), fluctuating from 4.78 to 11.59 mg/L. The removal efficiencies of nitrogen at three loads were 75%, 79%, and 83%, where the highest efficiency was achieved at an organic load rate (OLR) of 2.5 kgCOD/m³day.

Parameter		Concentrations of Nitrogen Compounds			QCVN 28: 2010/MONRE
		OLR 0.5	OLR 1.5	OLR 2.5	(Grade A)
NH4 ⁺ -N	in	22.8 ± 4.4	23.7 ± 3.2	25.0 ± 3.2	5
(mg/L)	out	0.2 ± 0.3	0.3 ± 0.3	0.2 ± 0.2	5
$NO_2^{-}-N$	in	0.67 ± 0.26	0.70 ± 0.26	0.74 ± 0.33	-
(mg/L)	out	0.36 ± 0.27	0.14 ± 0.19	0.02 ± 0.03	-
$NO_3^{-}-N$	in	0.20 ± 0.20	0.22 ± 0.25	0.16 ± 0.29	30
(mg/L)	out	4.91 ± 0.61	4.50 ± 0.50	3.90 ± 0.62	30
TKN (mg/L)	in	37.7 ± 5.6	37.5 ± 4.8	40.1 ± 6.8	-
	out	5.0 ± 0.8	4.5 ± 0.5	2.9 ± 0.7	-
TN (mg/L) i o	in	35.84 ± 5.51	38.43 ± 4.84	41.04 ± 6.88	-
	out	9.46 ± 1.45	8.14 ± 1.11	6.81 ± 1.13	-

Table 2. Concentrations of nitrogen compounds in hospital wastewater after treatment in various organic load rates.

Regarding the NH₄ and NO₃ removal efficiencies, the results in various operating phases indicated that these compounds were removed with average efficiencies varying from 94.52% to 99.64%, and NH₄ and NO₃ content in the outputs met the Vietnamese standard of grade A. To be specific, the input of NH₄⁺ in three operating phases ranged from 1.6 to 16.1 mg/L (average of 9.36 ± 12.89 mg/L) and corresponding outputs were all less than 5 mg/L. For the NO₃ treatment, average NO₃ concentrations in three load rates were also minimal, at 2.18, 1.05 and 1.99 mg/L, respectively.

3.3. Treatment Effectiveness of Other Pollutants

Among two forms of iron existing in wastewater, ferrous ion II (Fe²⁺) only stays in small amounts, and ferric iron III (Fe³⁺) in dissolved forms are usually dominant. Iron is often removed from the wastewater by physical means through membrane pores in MBR tanks. The average total iron removal efficiencies of the MBR-NF system of three organic load rates were 99.37%, 98.14% and 98.82%, respectively. The output total iron content was stable, fluctuating from 0 mg/L to 0.37 mg/L. From Figure 6, it is shown that the MF filter can reduce 60–80% the iron in the input and the NF can block most of the remaining iron ions. This suggests that the MBR-NF combination system is suitable for treatment of solute residue such as drug residues accrued in medical operations in hospitals.



Figure 6. Total iron removal efficiency at three organic load rates in a duration of 120 days of the MBR-nano system.

Figure 7 describes a coloration analysis of different flows in different load rates. In general, the average output color following Pt-Co measurements were 21.37, 11.55 and 8.89, corresponding to efficiencies of 97%, 98%, and 99% in the three load rates. In general, the removal efficiency remained above 90% during the study period, averaging at 14 ± 9 Pt-Co, irrespective of the variation of the input wastewater from 503 to 1358 Pt-Co. Considering the dark yellow and opaque color of the initial wastewater, which was contrasted by relative transparency of the effluent, this suggested that the coloration was well-handled by the MBR-NF system. In this case, wastewater discoloration resulted from the system also indicated the absence of pus, blood and iron precipitates.



Figure 7. Color removal efficiency at three organic load rates in a duration of 120 days of the MBR-Nano system.

To reinforce the results of pathogen removal, we performed quantification tests for coliform and *E. coli* bacteria once every four days. The result was expressed as the log removal value (LRV), as in Figure 8. The results showed that the coliform efficiency of the system approximates 100%. Despite the relatively high and fluctuated bacteria amount found in the input flows at three phases, fluctuating between 1.50×10^4 and 4.30×10^6 , bacteria found in the intermediate tank were approximately zero, which satisfied Vietnamese discharge standards.



Figure 8. Removal efficiencies of coliform and E. coli.

3.4. Evaluation of Membrane Resistance during Operation

Two backwashes on the MBR tank were performed on day 58 and 102. The membrane resistance in the two membranes was measured by the TMP, as illustrated in Figure 9. In the first 41 days, coinciding with the first operating phase, the TMP of the MBR gradually increased from 3.3 to 6.6 kPa. From day 42 to 57, the rise of the TMP became more rapid, surging from 8.3 to 32 kPa. After the first membrane wash on day 58, the TMP dropped from 32 to 5 kPa on the following day, followed by a similar TMP trend to the first cycle. However, the second cycle occurred at a faster pace, whereby only 35 days elapsed before a second wash. After the second wash on day 102, the TMP lowered to approximately 10 kPa and began to rise rapidly thereafter. In contrast with the instability of the TMP in the aerobic tank, the TMP after NF kept at consistently low levels and was stable. These results suggest that the preceding MBR could significantly relieve the filtration burden for the subsequent treatment.



Figure 9. Transmembrane pressure (TMP) over time of MF membrane filtration in aerobic tanks.

4. Conclusions

We evaluated a pilot-scale wastewater filtration system that combined an MBR with NF through various criteria, including the COD, the TP, nitrogen compounds and pathogen contents. Two main limitations were recognized in this study. First, the evaluation of pharmaceuticals widely present in hospital wastewater is lacking. Second, the considered OLRs were small. Overall, the system showed excellent capacity for removal of common organic pollutants, nutrients, and pathogenic microorganisms existing in hospital water discharge. The result was consistent and comparable with that of similar treatment systems in previous studies. After NF, the water quality of the outflow met the Vietnamese standard of grade A. The system arrangement, in which an MF membrane precedes an NF membrane, was shown to limit the amount of soil going through the NF system, thereby improving processing efficiency and possibly reducing fouling of the NF membranes. Hence, an MBR-NF combination could be a promising technology for the treatment of hospital wastewater.

Author Contributions: Investigation, T.T., T.B.N., H.L.H., D.A.L., T.D.L., A.T.H. and T.S.D.; Supervision, T.D.N. and L.G.B.; Writing-original draft, T.T. and D.C.N.; Writing-review and editing, D.C.N. and L.H.

Funding: This research received no external funding.

Acknowledgments: The authors acknowledge Nguyen Tat Thanh University for providing facilities, chemicals, and permission during the research period.

Conflicts of Interest: The authors declare no conflicts of interest.

Nomenclature

BOD	Biological oxygen demand
COD	Chemical oxygen demand
DO	Dissolved oxygen
F/M	Food to microorganism ratio
GAC	Granular active carbon
HRT	Hydraulic retention time
LRV	Log removal value
MBR	Membrane bioreactor
MF	Microfiltration
MLSS	Mixed liquor suspended solids
MLVSS	Mixed liquor volatile suspended solids
NF	Nanofiltration
OLR	Organic load rate
Pt-Co	Platinum-cobalt
RO	Reverse osmosis
SRT	Solid retention time
SVI	Sludge volume index
TCVN	Tieu Chuan Viet Nam (Vietnamese standards)
TKN	Total Kjeldahl nitrogen
TMP	Transmembrane Pressure
TP	Total phosphate
TSS	Total suspended solids

References

- 1. Kosma, C.I.; Lambropoulou, D.A.; Albanis, T.A. Occurrence and removal of PPCPs in municipal and hospital wastewaters in Greece. *J. Hazard. Mater.* **2010**, *179*, 804–817. [CrossRef] [PubMed]
- 2. Zheng, X.; Zhou, Y.; Chen, S.; Zheng, H.; Zhou, C. Survey of MBR market: Trends and perspectives in China. *Desalination* **2010**, *250*, 609–612. [CrossRef]
- 3. Pauwels, B.; Verstraete, W. The treatment of hospital wastewater: An appraisal. *J. Water Health* **2006**, *4*, 405–416. [CrossRef] [PubMed]
- 4. Joss, A.; Siegrist, H.; Ternes, T. Are we about to upgrade wastewater treatment for removing organic micropollutants? *Water Sci. Technol.* **2008**, *57*. [CrossRef] [PubMed]
- 5. Verlicchi, P.; Galletti, A.; Petrovic, M.; Barceló, D. Hospital effluents as a source of emerging pollutants: An overview of micropollutants and sustainable treatment options. *J. Hydrol.* **2010**, *389*, 416–428. [CrossRef]
- 6. Liu, Q.; Zhou, Y.; Chen, L.; Zheng, X. Application of MBR for hospital wastewater treatment in China. *Desalination* **2010**, *250*, 605–608. [CrossRef]
- Rodriguez-Mozaz, S.; Chamorro, S.; Marti, E.; Huerta, B.; Gros, M.; Sànchez-Melsió, A.; Borrego, C.M.; Barceló, D.; Balcázar, J.L. Occurrence of antibiotics and antibiotic resistance genes in hospital and urban wastewaters and their impact on the receiving river. *Water Res.* 2015, *69*, 234–242. [CrossRef] [PubMed]
- 8. Nasr, M.M.; Yazdanbakhsh, A. Study on wastewater treatment systems in hospitals of Iran. *J. Environ. Health Sci. Eng.* **2008**, *5*, 211–215.
- 9. Suarez, S.; Lema, J.M.; Omil, F. Pre-treatment of hospital wastewater by coagulation–flocculation and flotation. *Bioresour. Technol.* 2009, *100*, 2138–2146. [CrossRef] [PubMed]
- Zhu, M.; Zhu, G.; Zhao, L.; Yao, X.; Zhang, Y.; Gao, G.; Qin, B. Influence of algal bloom degradation on nutrient release at the sediment–water interface in Lake Taihu, China. *Environ. Sci. Pollut. Res.* 2013, 20, 1803–1811. [CrossRef] [PubMed]

- Le Moal, M.; Gascuel-Odoux, C.; Ménesguen, A.; Souchon, Y.; Étrillard, C.; Levain, A.; Moatar, F.; Pannard, A.; Souchu, P.; Lefebvre, A.; et al. Eutrophication: A new wine in an old bottle? *Sci. Total Environ.* 2019, 651, 1–11. [CrossRef] [PubMed]
- Conley, D.J.; Paerl, H.W.; Howarth, R.W.; Boesch, D.F.; Seitzinger, S.P.; Havens, K.E.; Lancelot, C.; Likens, G.E. ECOLOGY: Controlling Eutrophication: Nitrogen and Phosphorus. *Science* 2009, 323, 1014–1015. [CrossRef] [PubMed]
- 13. Naeem, M.; Idrees, M.; Khan, M.M.A.; Moinuddin; Ansari, A.A. *Task of Mineral Nutrients in Eutrophication*; Springer: Dordrecht, The Netherlands, 2014; Volume 2, pp. 223–237.
- Tchounwou, P.B.; Yedjou, C.G.; Patlolla, A.K.; Sutton, D.J. Heavy Metal Toxicity and the Environment. In *Molecular, Clinical and Environmental Toxicology*; Experientia Supplementum; Luch, A., Ed.; Springer: Basel, Switzerland, 2012; Volume 3, pp. 133–164.
- 15. Krzeminski, P.; Leverette, L.; Malamis, S.; Katsou, E. Membrane bioreactors-A review on recent developments in energy reduction, fouling control, novel configurations, LCA and market prospects. *J. Membrane Sci.* **2017**, 527, 207–227. [CrossRef]
- 16. Ahn, W.-Y.; Kang, M.-S.; Yim, S.-K.; Choi, K.H. Advanced landfill leachate treatment using an integrated membrane process. *Desalination* **2002**, *149*, 109–114. [CrossRef]
- 17. Wang, G.; Fan, Z.; Wu, D.; Qin, L.; Zhang, G.; Gao, C.; Meng, Q. Anoxic/aerobic granular active carbon assisted MBR integrated with nanofiltration and reverse osmosis for advanced treatment of municipal landfill leachate. *Desalination* **2014**, *349*, 136–144. [CrossRef]
- Alturki, A.A.; Tadkaew, N.; McDonald, J.A.; Khan, S.J.; Price, W.E.; Nghiem, L.D. Combining MBR and NF/RO membrane filtration for the removal of trace organics in indirect potable water reuse applications. *J. Membr. Sci.* 2010, 365, 206–215. [CrossRef]
- Wang, Y.; Wang, X.; Li, M.; Dong, J.; Sun, C.; Chen, G. Removal of Pharmaceutical and Personal Care Products (PPCPs) from Municipal Waste Water with Integrated Membrane Systems, MBR-RO/NF. Int. J. Environ. Res. Public Health 2018, 15. [CrossRef] [PubMed]
- 20. Patterson, C.; Anderson, A.; Sinha, R.; Muhammad, N.; Pearson, D. Nanofiltration Membranes for Removal of Color and Pathogens in Small Public Drinking Water Sources. J. Environ. Sci. 2012, 138, 48–57. [CrossRef]
- 21. Lan, Y.; Groenen-Serrano, K.; Coetsier, C.; Causserand, C. Nanofiltration performances after membrane bioreactor for hospital wastewater treatment: Fouling mechanisms and the quantitative link between stable fluxes and the water matrix. *Water Res.* **2018**, *146*, 77–87. [CrossRef] [PubMed]
- 22. Kovalova, L.; Siegrist, H.; Singer, H.; Wittmer, A.; McArdell, C.S. Hospital wastewater treatment by membrane bioreactor: Performance and efficiency for organic micropollutant elimination. *Environ. Sci. Technol.* **2012**, *46*, 1536–1545. [CrossRef] [PubMed]
- 23. Snyder, S.A.; Adham, S.; Redding, A.M.; Cannon, F.S.; DeCarolis, J.; Oppenheimer, J.; Wert, E.C.; Yoon, Y. Role of membranes and activated carbon in the removal of endocrine disruptors and pharmaceuticals. *Desalination* **2007**, 202, 156–181. [CrossRef]
- 24. Chon, K.; Sarp, S.; Lee, S.; Lee, J.-H.; Lopez-Ramirez, J.A.; Cho, J. Evaluation of a membrane bioreactor and nanofiltration for municipal wastewater reclamation: Trace contaminant control and fouling mitigation. *Desalination* **2011**, *272*, 128–134. [CrossRef]
- 25. Cartagena, P.; El Kaddouri, M.; Cases, V.; Trapote, A.; Prats, D. Reduction of emerging micropollutants, organic matter, nutrients and salinity from real wastewater by combined MBR–NF/RO treatment. *Sep. Purif. Technol.* **2013**, *110*, 132–143. [CrossRef]
- 26. Ngoc, N.N.; Van Cach, N.; Ha, T.L.; Giang, P.T.T. Microbiologycal characterization and potential application of indigenous B. Methylotrophycus Ba1 in handling of Canna Edulis. Ker processing craft village wastewater. *J. Forestry Sci. Technol.* **2016**, *5*, 3–9.
- 27. Nguyen, T.-T.; Bui, X.-T.; Luu, V.-P.; Nguyen, P.-D.; Guo, W.; Ngo, H.-H. Removal of antibiotics in sponge membrane bioreactors treating hospital wastewater: Comparison between hollow fiber and flat sheet membrane systems. *Bioresource Technol.* **2017**, *240*, 42–49. [CrossRef] [PubMed]
- Choi, J.-H.; Fukushi, K.; Ng, H.; Yamamoto, K. Evaluation of a long-term operation of a submerged nanofiltration membrane bioreactor (NF MBR) for advanced wastewater treatment. *Water Sci. Technol.* 2006, 53, 131–136. [CrossRef] [PubMed]

- Wang, J.; Li, K.; Wei, Y.; Cheng, Y.; Wei, D.; Li, M. Performance and fate of organics in a pilot MBR–NF for treating antibiotic production wastewater with recycling NF concentrate. *Chemosphere* 2015, 121, 92–100. [CrossRef] [PubMed]
- 30. Al Qarni, H.; Collier, P.; O'Keeffe, J.; Akunna, J. Investigating the removal of some pharmaceutical compounds in hospital wastewater treatment plants operating in Saudi Arabia. *Environ. Sci. Pollut. Res. Int.* **2016**, *23*, 13003–13014. [CrossRef] [PubMed]



© 2019 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).