

Comparing pharmaceutical removal in wastewater treatment plant and lab-scale membrane bioreactor system

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ABSTRACT

This study aims to assess the removal efficiency of nine pharmaceutical compounds along with COD, BOD₅, nitrate (NO₃⁻), and phosphate (PO₄³⁻). Results showed that the removal of antibiotics – sulfamethoxazole, erythromycin, and ofloxacin – ranged from 30–34% in the HWWTP, and lab-scale MBR achieved significantly higher rates, between 60–78%. For non-steroidal anti-inflammatory drugs such as diclofenac, paracetamol, and ibuprofen, the HWWTP removed 35–54%, compared to 53–81% in the lab-scale system. Compounds like carbamazepine, diazepam, and furosemide showed modest removal in HWWTP (11%, 20%, and 34%, respectively), but improved performance in MBR (31%, 39%, and 45%). The tubesettler alone contributed to pollutant removal in the range of 15–31% in HWWTP and 24–51% in the lab-scale setup, suggesting its potential as a polishing step when used alongside an MBR. Future research should also investigate the impact of sudden chemical loads – such as surfactants and disinfectants on HWWTP performance.

Keywords: hospital wastewater, pharmaceuticals, MBR, tubesettler, HQ.

INTRODUCTION

Pharmaceutical compounds are increasingly being detected in water resources, especially in surface waters, as reported by several studies (Praseratkulsak et al., 2019). This growing presence is closely linked to the rising consumption of medications in daily life – driven largely by population growth, which in turn increases pharmaceutical production and use (Lopez-Herguedas et al., 2024). Modern analytical tools, especially high-resolution techniques, now make it easier to detect these substances even at trace levels (Tang et al., 2020).

Among the main contributors to pharmaceutical pollution in the environment are wastewater treatment plants (WWTPs), which were not originally designed to remove such compounds (Leiviskä and Risteelä, 2022; Moya-Llamas et al., 2023). As a result, conventional WWTPs often discharge effluent containing residual pharmaceuticals, due to their limited removal efficiency. Although this issue has been known for more than two decades, WWTPs still face major challenges

in adapting to current demands—ranging from high operational costs to inadequate removal of emerging contaminants like pharmaceuticals (Gharibian and Hazrati, 2022).

In response, membrane-based treatment technologies have gained attention for their efficiency and selectivity (Farah et al., 2023). One such approach is the membrane bioreactor (MBR), which integrates the biological degradation of the activated sludge process with membrane filtration (ultrafiltration or nanofiltration). The bioreactor promotes microbial breakdown of pollutants, while the membrane acts as a physical barrier, ensuring clean separation. MBRs combine low operational costs with high treatment performance and have demonstrated superior removal of pharmaceuticals, organic matter, and nutrients. Their effectiveness has been confirmed in both pilot studies and full-scale treatment plants (Moya-Llamas et al., 2023; Sutthiwanit et al., 2023).

Several studies have investigated the effectiveness of different wastewater treatment approaches for removing pharmaceutical compounds. Tang

et al. (2020) examined a municipal wastewater treatment plant using a hybrid system that combined attached biofilm with activated sludge. They found that 14 out of the 21 pharmaceutical compounds analyzed were removed at efficiencies above 50%. In a separate study, Kim et al. (2014) evaluated a MBR system and reported removal rates exceeding 90% for all 23 pharmaceutical compounds they tested. Al-Khafaji et al. (2023) focused on the performance of a hospital wastewater treatment plant in Basrah, Iraq. While nitrate levels in the effluent were within acceptable limits, the concentrations of COD and BOD exceeded permissible standards, suggesting insufficient removal of organic matter. Meanwhile, Vo et al. (2019) explored the use of a sponge-based MBR system paired with ozonation for treating hospital wastewater. Their results showed that the addition of ozone significantly improved removal efficiencies: 66% for sulfamethoxazole, 83% for ciprofloxacin, 88% for ofloxacin, 90% for erythromycin, 92% for norfloxacin, and 97% for trimethoprim. Alrhoun et al. (2014) assessed the performance of a pilot-scale MBR treating hospital wastewater by analyzing the behavior of extracellular polymeric substances (EPS) in the sludge. They found that a 20-day exposure to pharmaceutical compounds had no negative impact on COD removal or nitrification. In another comparative study, Hamon et al. (2018) evaluated MBRs fed with biomass from both municipal treatment systems and hospital oncological units. Their findings supported MBRs as a reliable solution, reporting over 75% removal for sulfamethoxazole and more than 90% for codeine. These studies show a clear interest in enhancing pharmaceutical compound removal from both municipal and hospital wastewater. However, the literature still lacks comparative evaluations between full-scale hospital wastewater treatment plants and controlled lab-scale setups using membrane bioreactor technology. This gap highlights the need for further research to better understand how each system performs under different conditions.

While many studies have identified wastewater treatment plants (WWTPs) as a major source of pharmaceutical contaminants in the environment, hospital and healthcare facilities—despite being widespread in urban areas—have often been overlooked as significant contributors (Abad et al., 2023). The literature also highlights that MBRs, whether used in hybrid forms or in combination with other technologies, have been

increasingly studied for their improved treatment efficiency. What sets this study apart is its focus on a direct comparison between a full-scale hospital wastewater treatment plant using MBR and a controlled lab-scale MBR setup. Specifically, the study aims to evaluate and compare the treatment performance of both systems, with particular attention to the role of the associated tubesettler and its potential use as a polishing unit in pharmaceutical removal. The literature does not directly compare full-scale hospital wastewater treatment plants (HWWTPs) and controlled lab-scale MBR systems treating hospital wastewater under the same influent conditions, despite significant advancements in pharmaceutical removal studies. Furthermore, even while tubesettlers have shown encouraging polishing performance, there is still a lack of documentation about their combined use with MBR for pharmaceutical removal from hospital effluents. With an emphasis on the removal of nine representative pharmaceutical compounds and related conventional parameters (COD, BOD₂, NO₃⁻, PO₄³⁻), this study intends to systematically compare the treatment efficiency of a full-scale HWWTP and a lab-scale MBR, both coupled with a tubesettler, in order to close these gaps.

Because of the controlled operating conditions and lack of hydraulic and toxic shock loads, we therefore predict that: (i) the lab-scale MBR will achieve higher and more stable removal; and (ii) the tubesettler, when used as a post-treatment step, will further improve overall pharmaceutical removal, thereby reducing ecotoxicological risk in the final effluent.

The anticipated scientific contribution of this work is to clarify the added value of tubesettler integration as a compact polishing stage for hospital wastewater management and to provide previously unavailable comparative evidence on how full-scale and lab-scale systems differ in pharmaceutical removal performance.

DATA AND METHOD USED

Hospital wastewater treatment plant

HWWTP setup is given in Figure 1. Instead of whole treatment plant setup the reduced setup is presented to ensure that the influent of MBR is passing through two treatment phases i.e., grit chamber/screening and equalization tank before undergoing MBR process. This was done to bring

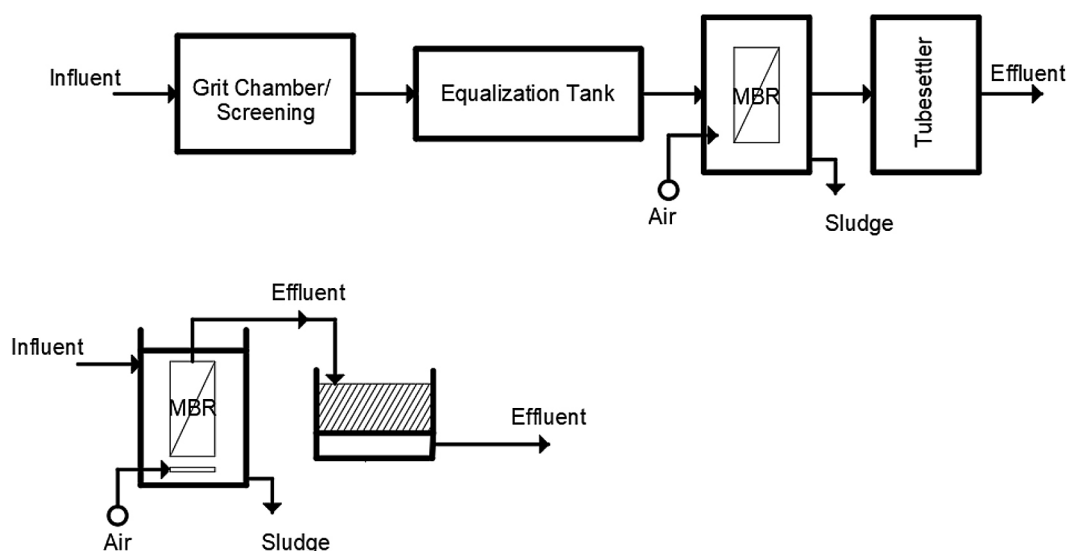


Figure 1. Hospital wastewater treatment plant schematic diagram (top) and lab-scale setup of MBR with tubesettler (below)

some relevance between HWWTP and LSMBR setup as LSMBR influent is not influenced by continuous discharge of surfactant, disinfectants and other major pollutants which may not only alter the hospital wastewater composition but also the efficiency of the treatment. If the wastewater samples were collected prior to its entry to treatment plant it is prone to sudden spikes in concentration of the pollutants. The comparison nature of this study calls for no interference from sudden changes which are not present in lab scale setup. This will compromise the comparison of the results as one is affected by spikes while another is free from spikes. Hence using wastewater samples after equalization tanks will eliminate such discrepancies. Tubesettler is currently installed at hospital wastewater treatment plant. Also, tubesettler pose higher advantage to serve as polishing unit for effluent from secondary or tertiary treatment plant. This is attributed to the fact that it requires far less area as compared to secondary or tertiary clarifiers and for the same reason have been investigated in several studies. As health care facilities and hospitals are often restricted with space availability especially for wastewater treatment plants tubesettler is a preferred choice (Abad et al., 2023; Al-subih et al., 2022a; Khan et al., 2020, 2019, 2022).

Lab setup

In this study flat sheet of membrane was used which was placed inside the tank as presented in Figure 2. The seed sludge was obtained from

hospital wastewater treatment plant which was acclimatized with hospital wastewater for lab scale setup. The lab scale reactor comprised of 4.9 L volume glass cylinder. Peristaltic pump was employed to control wastewater feed into the reactor. Permeate suction was achieved by deploying vacuum pump for the membrane ($0.12\text{--}0.15\text{ m day}^{-1}$). The dissolved oxygen ($7\text{--}8\text{ mgL}^{-1}$), mixing in reactor and formation of cake later on membrane surface was controlled by using air diffuser.

Because of the controlled operating conditions and lack of hydraulic and toxic shock loads, we therefore predict that: (i) the lab-scale MBR will achieve higher and more stable removal; and (ii) the tubesettler, when used as a post-treatment step, will further improve overall pharmaceutical removal, thereby reducing ecotoxicological risk in the final effluent. The anticipated scientific contribution of this work is to clarify the added value of tubesettler integration as a compact polishing stage for hospital wastewater management and to provide previously unavailable comparative evidence on how full-scale and lab-scale systems differ in pharmaceutical removal performance.

Wastewater sampling

Composite samples were obtained from reactor and hospital wastewater treatment plant (effluent from the equalization tank) over a period of 24 hours. Grab samples were collected every 4-hour interval. Composite sample comprised of 6 grab samples over time period of 24 hours.

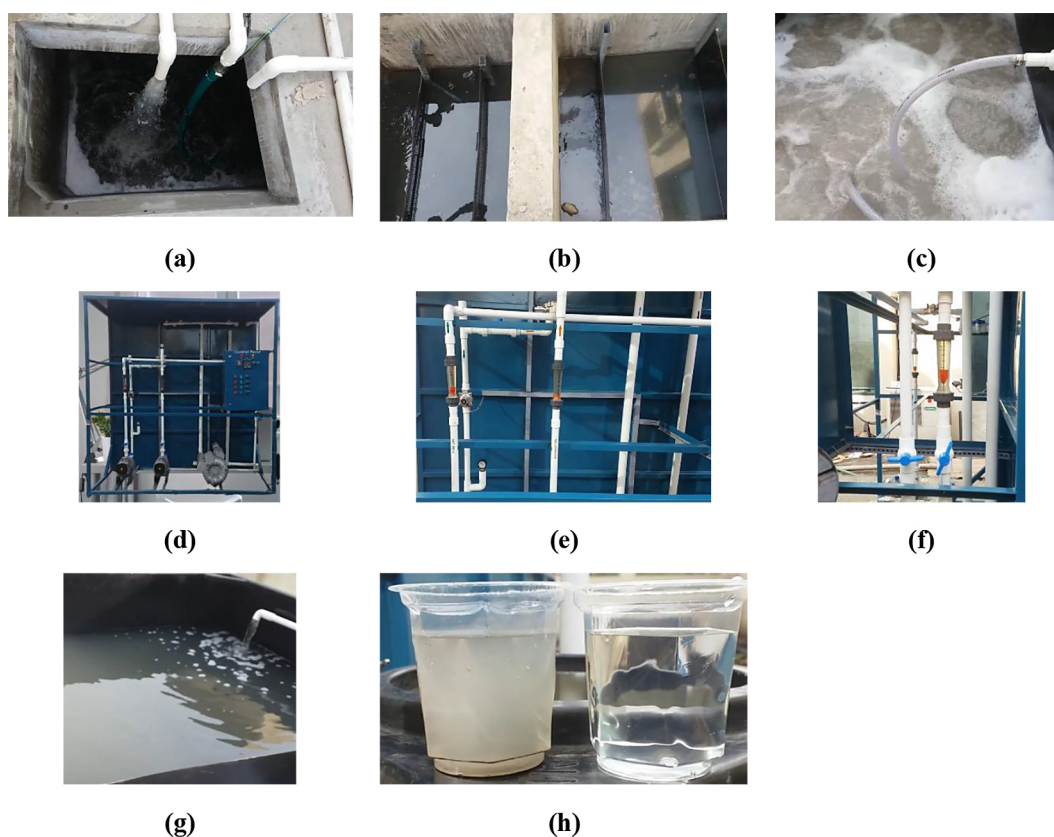


Figure 2. MBR setup at wastewater treatment plant influent & processing (a-c), setup (d-f) and effluent (g-h)

The grab samples were stored in amber glass bottles in a refrigerator at $-4\text{ }^{\circ}\text{C}$. The laboratory analysis for characterization of wastewater samples has been provided in (Alsubih et al., 2022b, 2022a). The conventional parameters analysed in this study consisted of chemical oxygen demand (COD), biological oxygen demand (BOD₅), nitrate (NO_3^-) and phosphate (PO_4^{3-}) analysis. The pharmaceutical compounds analysed in this study are presented in Table 1. Three antibiotics, three NSAIDs (non-steroidal anti-inflammatory drugs), 1 anticonvulsant, 1 diuretic and 1 benzodiazepine.

Pharmaceutical compound analysis

To extract pharmaceutical residues from wastewater samples before HPLC analysis, we used a solid phase extraction (SPE) method based on reverse-phase interactions. First, we adjusted the pH of the filtered samples to around 3–4 using a few drops of formic or phosphoric acid. This step helps enhance the retention of acidic and weakly basic compounds like ketoprofen, sulfamethoxazole, and diazepam. We then conditioned the SPE cartridges – either C18 or Oasis HLB – with 5 mL of methanol followed by 5 mL

of ultrapure water to activate the sorbent surface. After conditioning, we loaded between 250 and 1000 mL of the wastewater sample onto the cartridge at a slow, steady rate to allow the analytes to bind properly. Next, we rinsed the cartridge with water to remove polar interferences, and in some cases, used a mild wash of water with 5% methanol to eliminate weakly bound matrix components. Once washed, we dried the cartridges under vacuum for about 10 to 15 minutes to remove residual moisture. To elute the retained pharmaceutical compounds, we passed through 5 to 10 mL of methanol – or a mix of methanol and acetonitrile – directly into glass vials. The eluted extracts were then evaporated gently under a nitrogen stream at $35\text{--}40\text{ }^{\circ}\text{C}$ to avoid thermal degradation. Finally, we reconstituted the dried residue in 1 mL of a water–methanol solution (or directly in the mobile phase used for HPLC), making the samples ready for injection and analysis.

All water samples underwent solid phase extraction (SPE) for pre-concentration prior to HPLC analysis. Calibration standards were prepared in both ascending and descending concentrations for various pharmaceutical compounds, including aspirin, amoxicillin, levofloxacin,

Table 1. Pharmaceutical compounds investigated in this study

Pharmaceutical compound	Acronym	Class	Chemical formula	Molecular weight	Log Kow
Diclofenac	DIC	NSAID	C ₁₄ H ₁₁ Cl ₂ NO ₂	296.15 g/mol	4.51
Ketoprofen	KTF	NSAID	C ₁₆ H ₁₄ O ₃	254.28 g/mol	3.12
Paracetamol	PAR	NSAID	C ₈ H ₉ NO	155.19 g/mol	0.46
Carbamazepine	CAB	Anticonvulsant	C ₁₅ H ₁₂ N ₂ O	236.27 g/mol	2.45
Diazepam	DZM	Benzodiazepine	C ₁₆ H ₁₃ ClN ₂ O	284.74 g/mol	2.82
Sulfamethoxazole	SMZ	Antibiotic	C ₁₀ H ₁₁ N ₃ O ₃ S	253.28 g/mol	0.89
Erythromycin	ERY	Antibiotic	C ₃₇ H ₆₇ NO ₁₃	733.9 g/mol	3.06
Ofloxacin	OFL	Antibiotic	C ₁₈ H ₂₀ FN ₃ O ₄	361.4 g/mol	-0.39
Frusemide	FSM	Diuretic	C ₁₂ H ₁₁ ClN ₂ O ₅ S	330.74 g/mol	2.03

fluconazole, and ketorolac. Initial water samples were analyzed in duplicate; since no significant differences were observed, the remaining samples were analyzed in single runs. Both wastewater samples and standards followed the preparation protocol outlined by Sabri et al. (2020).

Analysis of all pharmaceutical compounds was carried out using HPLC. The methodology, as previously detailed by Sabri et al. (2020), used an isocratic HPLC system (Shimadzu SPD-M20A 230 V) equipped with a Luna C18 column (5 µm, 250 × 4.6 mm). The mobile phase consisted of buffer solution (NaH₂PO₄·H₂O and Na₂HPO₄·7H₂O) and methanol in a 55:45 volume ratio. The pH was adjusted to 3.5 using phosphoric acid, with a constant flow rate of 1 mL/min. The method was validated for linearity, stability, accuracy, precision, and detection limit.

Risk assessment

Hazard quotient approach was employed to assess potential impact of pharmaceutical compounds on the receiving environment. Equation 1 was used to estimate HQ values for each pharmaceutical compound. Hazard index (HI) value was estimated using equation 2 to determine the cumulative risk posed by all the pharmaceutical compounds (Lancheros et al., 2019). The PNEC (predicted no effect concentration) value adopted in this study was 16300, 164000, 134000, 810, 26.6, 900, 36.6, 900 and 322000 for diclofenac, ketoprofen, paracetamol, carbamazepine, diazepam, sulfamethoxazole, erythromycin, and ofloxacin respectively (Besse et al., 2008; Database, 2023; Frédéric and Yves, 2014; Huschek et al., 2004; Minguez et al., 2016; Park et al., 2019;

Razak et al., 2021; Straub, 2016). The MEC values were obtained from the laboratory analysis:

- Hazard quotient (HQ) = 1,
- Hazard index (HI) = ΣHQ 2.

RESULTS AND DISCUSSION

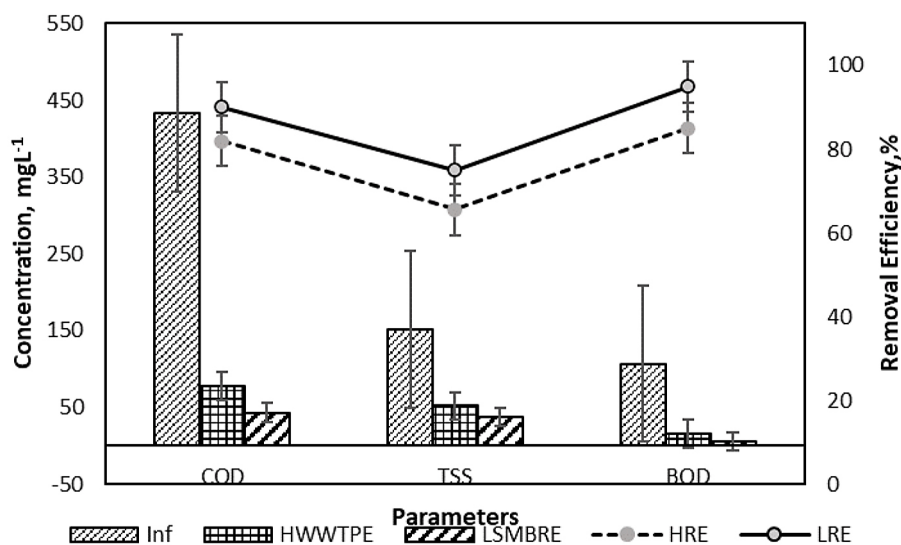
Removal of organic matter and nutrients

Organic matter and nutrients removal in hospital wastewater treatment plant and lab scale MBR setup is presented in Figure 3a and Figure 3b.

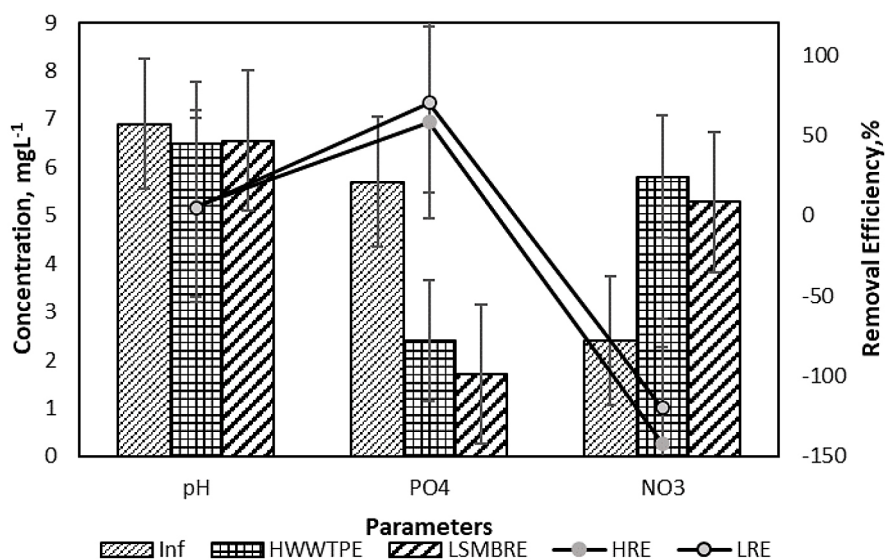
The pH of hospital wastewater was observed to be acidic with average pH of 6.9. In HWWTP the pH reduced to 6.5 while in Lab scale MBR setup it was 6.5. Total suspended solids influent concentration was 151 mgL⁻¹ which reduced to 52 mgL⁻¹ in HWWTP effluent. In LSMBR, TSS reduced to 37 mgL⁻¹. BOD₅ reduction in HWWTP was achieved up to 85% and in LSMBR setup it was 95% with effluent BOD₅ being 16 mgL⁻¹ and 5 mgL⁻¹ respectively. COD influent 433 mgL⁻¹ and its reduction was 78% and 90% in HWWTP and LSMBR setup. Phosphate was reduced by 57% and 70% in HWWTP and LSMBR respectively. There was increase of nitrate concentration by 141% and 120% in HWWTP and LSMBR respectively.

Removal of pharmaceutical compounds

Figure 4a, 4b and 4c presents the removal efficiency, influent and effluent concentration of pharmaceutical compound from HWWTP and LSMBR. The antibiotics, erythromycin (ERY), sulfamethoxazole (SMZ) and ofloxacin (OFL) reduction were low in HWWTP. sulfamethoxazole



(a)



(b)

Figure 3. Influent, effluent and removal efficiency of Hospital wastewater treatment plant (HWWTPE) and Lab scale MBR setup (LSMBRE) for a) BOD, COD and TSS and (b) pH, PO₄ and NO₃ (Inf = influent, E = effluent, HRE = HWWTPE removal efficiency and LRE = LSMBRE removal efficiency)

removal was 34%, Erythromycin removal was observed to be 32% and Ofloxacin removal was found to be 30% in HWWTPE. In LSMBRE, antibiotic removal efficiency achieved was 68%, 78% and 60% respectively for SMZ, ERY and OFL. Sulfamethoxazole and erythromycin higher removal can also be attributed to their lower concentration of 2258 ngL⁻¹ and 3276 ngL⁻¹ as compared to OFL concentration of 9857 ngL⁻¹. Vo et al., (2019) have observed removal of ofloxacin from

hospital wastewater up to 88% using sponge MBR coupled with ozonation with influent concentration of 6200 ngL⁻¹. The same study also reported 90% removal of erythromycin using same setup for influent concentration of 540 ngL⁻¹. (Mamo et al., 2016) observed 16–60% removal from urban wastewater for reclamation under varying aeration and nitrification condition (nitrifying and denitrifying). In the first study MBR was hybrid setup in itself accompanied by ozonation which enhanced

its removal efficiency associated with lower influent concentration of antibiotics as compared to concentrations observed in this study. In the other study it has to be noted that it was urban wastewater not hospital wastewater and even then, the removal efficiency was lower as compared to the lab scale study result of this study and concluded diclofenac to be lowest among the compounds to be removed through biodegradation.

NSAIDs removal was higher as compared to antibiotic in HWWTP. Diclofenac removal efficiency reached 34% while ketoprofen removal efficiency was 31% and the highest removal efficiency was achieved for paracetamol reaching 54%. The LSMBR removal efficiency for NSAIDs was 53%, 81% and 78% for diclofenac, ketoprofen and paracetamol respectively. similar to antibiotics it was observed that removal efficiency was much higher in LSMBR as compared to HWWTP. Farah et al., (2023) have reported 72% removal of diclofenac from wastewater by using hollow fiber liquid membrane. dos Santos et al., (2022) have termed diclofenac as most

recalcitrant in comparing it for biodegradation against other pharmaceutical compounds analysed in the study. Park et al., (2018) used WWTP wastewater as influent to investigate efficiency of coagulant-MBR for removal of diclofenac and ketoprofen and observed it to be 42% and 77% respectively. However, they also stated that upon addition of coagulant to MBR setup the removal efficiency increased by 21% for ketoprofen and 23% for ketoprofen.

The other three classes of pharmaceutical compounds viz. carbamazepine, diazepam, furosemide showed low removal efficiency both in HWWTP and LSMBR. Carbamazepine was removed with efficiency of 11%, diazepam removal was achieved up to 20% and furosemide removal reached up to 34%. However, the LSMBR performance was better with carbamazepine removal of 40%, diazepam was removed up to 25% and furosemide up to 45%. Monteoliva-García et al., (2020) investigated MBR combined with advanced oxidation process for its efficiency in removing pharmaceutical compounds from urban

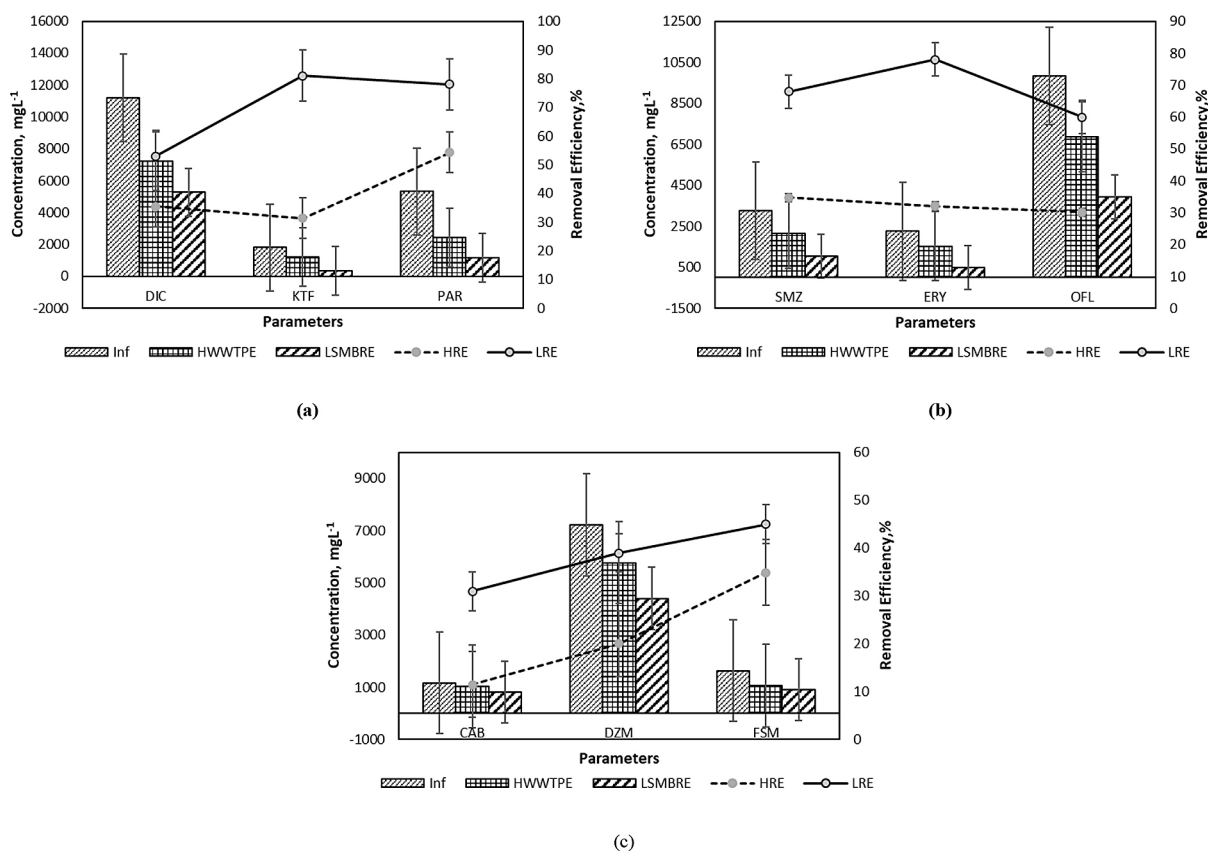


Figure 4. Influent, effluent and removal efficiency of hospital wastewater treatment plant and lab scale MBR setup for a) NSAIDs, b) Antibiotics and c) anticonvulsant, diuretic and Benzodiazepine (Inf = influent, HWWTPE = hospital wastewater treatment plant, LSMBRE = lab scale MBR, HRE = hospital wastewater treatment plant removal efficiency and LRE = lab scale MBR removal efficiency)

wastewater. It was observed that diazepam was removed with an efficiency of 69% with MBR alone and in hybrid combination with AOP it was observed to be 75%. They also reported in their study that biodegradation of diazepam is difficult attributed to its strong electron withdrawing ability in its molecular structure. Wijekoon et al., (2013) have observed carbamazepine removal of 58% by employing MBR treatment alone. They also reported that low removal of carbamazepine is due to its strong EWGs (Electron withdrawing group) in their molecular structure along with its low hydrophobicity. Reif et al., (2013) has reported another study which upon addition of powdered activated carbon (PAC) in MBR setup achieved diazepam removal efficiency in range of 93–99%. Tiwari et al., (2017) also reported the same study for high rate removal of diazepam upon introduction of PAC in MBR setup. Cheng et al., (2016) has cited an study employing aerobic + anerobic hollowfiber coupled MBR setup achieving furosemide removal efficiency of 68% on investigating full scale wastewater treatment. Kim et al., (2014) has reported furosemide removal to be greater than 97% while investigating MBR wastewater treatment plant. One of the factors that the study mentioned was the temperature of 21 °C which may have optimized the removal efficiency of the treatment plant.

Removal of pharmaceutical compounds from tub settler

As it was evident that from literature that combination of wastewater treatment process enhances the removal efficiency of MBR, this study investigated the associated tub settler with the HWWTP and also with LSMBR setup. The associated tub settler results both for HWWTP and LSMBR setup showed reduction in all pharmaceutical compounds analysed in this study. In HWWTP tub settler DIC effluent concentration was 5784 ngL⁻¹, KTF concentration was 918 ngL⁻¹ and PAR concentration reduced to 1920 ngL⁻¹. Among the antibiotic SMZ concentration in effluent was observed to be 1689 ngL⁻¹, ERY concentration reduced to 1303 ngL⁻¹ and OFL concentration were observed to be 5637 ngL⁻¹. CAB in effluent from HWWTP tub settler was found to be 755 ngL⁻¹, Diazepam was observed to be 3918 ngL⁻¹ and FSM was present in concentration of 714 ngL⁻¹. In tub settler associated with LSMBR setup, DIC, KTF and PAR concentration in the

effluent were 3898 ngL⁻¹, 220 ngL⁻¹ and 854 ngL⁻¹ respectively. Similarly, antibiotics SMZ, ERY and OFL concentration was 691 ngL⁻¹, 377ngL⁻¹ and 1851ngL⁻¹ respectively. CAB, DZM and FSM concentrations reduced up to 467 ngL⁻¹, 2160 ngL⁻¹, and 656 ngL⁻¹ respectively. The lower concentration in tub settler coupled with LSMBR are attributed to lower concentration in effluent of LSMBR. This again is related to LSMBR higher removal efficiency as compared to HWWTP. Second, it was free from any fluctuations and changes in wastewater characteristics which occurs in HWWTP but not in LSMBR setup.

When overall removal efficiency was considered, HWWTP removal efficiency reached for NSAIDs was in range of 48–84%, antibiotics 42–48%, CAB, DZM and FSM range was 35–50%. Nonetheless LSMBR setup coupled with tub settler achieved removal efficiency of 65%, 87%, 84% for DIC, KTF and PAR respectively. for antibiotics SMZ, ERY and OFL the removal efficiency of 78%, 85% and 70% was achieved respectively. CAB was removed with efficiency of 60%, DZM 70% and FSM 60%. Based on overall efficiency and the tub settler individual performance it can be inferred the combination of MBR with tub settler can achieve satisfactory removal efficiency for pharmaceutical compounds. The lower results of MBR as individual treatment process upon coupling with tub settler can now relate to the results already published in the literature.

ECOTOXICITY ASSESSMENT

The HQ was estimated to determine the potential environmental risk that effluent from hospital wastewater treatment plant and lab scale reactor. The HQ was categorized into four categories; HQ > 10 high risk, HQ in range 1–10 moderate risk, HQ in range 0.1–1 low risk and HQ < 0.1 no risk (Ávila et al., 2021). The summation of all the HQ values gave the hazard index value to sum the overall environmental risk from all the targeted compound analysed as combined. The category of risk is same as that for HQ values. The risk assessment in this study revealed that there was no risk to low risk posed to fish and crustacea from the influent to begin with, both in case of Fish and crustacea/Daphne. Which further reduced in the effluent from HWWTP. Same was observed in LSMBR, with removal efficiency of pollutants higher, the risk posed to environment

reduced even more from the effluent. However, with hazard quotient value of 4 in influent from SMZ revealed moderate hazard for algae, the HQ values of 10.9 and 61 from OFL and ERY indicated highly hazardous state of the influent. Upon treatment in HWWTP, HQ value reduced to 2.6 which is significant although the risk category remains the same i.e., moderately hazard from SMZ. Nonetheless in case of ERY and OFL, HQ values did not reduce significantly as compared to SMZ. In case of OFL risk reduced from high risk to moderate risk, but in case of ERY the risk remained in the same category i.e., high risk with HQ value of 59. In LSMBR effluent the risk was significantly reduced to HQ value of 1.29 for SMZ, 4 for OFL and 13 for ERY. The high risk to algae arises from the fact that the PNEC value for algae is much lower as compared to the PNEC values for fish and crustacea/daphne. In tubesettler effluent the HQ values decreased further. The effluent from hospital tubesettler depicted HQ value of 0.5 for fish, 1.39 for daphnia and crustacea and 14 for algae. For lab setup tubesettler HQ values were 0.37 for fish, 0.94 for daphnia and crustacea and 7 for algae. The risk for fish and daphnia/crustacea reduced to low risk for both setups as compared to moderate risk is HWWTP and LSMBR setup. The HQ value of 14 and 7 for algae in HTS (HWWTP tubesettler) and LTS (LSMBR tubesettler) respectively further reduced the risk. From LSMBR tubesettler the risk to fish reduced to low risk and for hospital tubesettler moderate risk. It can be inferred that tubesettler can serve as polishing treatment unit for MBR to reduce risk posed for the receiving environment. Lozano Avilés et al., (2022) has also observed moderate to high risks posed to aquatic organisms from SMZ, OFL and ERY, while investigating effect of WWTP upgradation on pharmaceutical compounds risks in receiving water bodies on scale of 0–1. Which if inferred to the scale of this study pose low risk for all the investigated pharmaceutical compounds. So how do we interpretate the significantly varying scale of HQ? WWTP in developed countries are optimized, controlled and monitored as per the guiding standards where they achieve the design efficiency for wastewater treatment. However, in developing countries, where government agencies lack funds to provide basic living amenities to its population. Running, operating and maintaining WWTPs are still far from reality, even though it is not zero. So, government agencies came up with

solution, where for concentrated pollution source of specific pollution source is expected to enter public sewers, the facility of institute requires to install its own WWTPs before the effluent can be discharged into the public sewers. This addresses two issues first the cost of treatment is solely borne by the running facility and second even if efficiency is low, it can be further reduced when it reaches urban WWTPs. This again reduces the load of pollutant in this case pharmaceutical compounds to be removed at urban WWTPs.

CONCLUSIONS

This study compared the hospital wastewater treatment performance between hospital wastewater treatment plant and lab scale treatment setup. The treatment employed was MBR associated with tubesettler. The lab scale setup exhibited higher removal efficiency as compared to hospital wastewater treatment plant.

Among the NSAIDs removal, paracetamol removal efficiency was highest with 54% in hospital wastewater using MBR. While in lab scale MBR setup ketoprofen was removed up to 72%. The antibiotic removal in hospital wastewater removal was similar with range of 30–34%. While in lab scale MBR setup erythromycin removal was 67%, and sulfamethoxazole removal was 50% followed by 42% removal of ofloxacin. Frusemide was removal was 34% followed by diazepam 20% and carbamazepine 11% in hospital MBR. At lab scale MBR carbamazepine and diazepam was removed by 22% and 23% respectively, while frusemide removal was 15%. The variation in performance arises from the fact that lab-scale systems operate under controlled and idealized conditions while hospital wastewater treatment plant is affected by load variation, shock loading of pollutants and inconsistent characteristics of the influent. Compounds like carbamazepine are identified as recalcitrant and have low removal efficiency while compounds like ketoprofen are hydrophobic and are readily absorbed by sludge or membrane in lab scale setup. The greater control of hydraulic retention time and solids retention time at lab scale setup enables it achieve greater removal efficiency. On the contrary hospital wastewater treatment plant shorter retention time which hinders biodegradation of the compounds leading to reduced efficiency. Also, the microorganisms in hospital wastewater vary significantly

as compared to lab scale setup. In lab scale setup the microbial community is restricted to the sample obtained and will not be subjected to any shock loading. Thereby enabling the microbial community to adapt and achieve greater efficiency at lab scale setup. However, in hospital the influent characteristic varies giving shock loads to the microbial community. Additionally, the contaminants vary from time to time depending on the activity undertaken in the hospital. Which again may not provide enough time for the microbial community to adapt and achieve the optimized performance of the setup.

Tubesettler can be employed as polishing unit for effluent from MBR both in HWWTP and LSMBR scale treatment. As an individual unit MBR performance for removal of pharmaceutical compound was not satisfactory. However, in combination with tubesettler the performance achieves satisfactory results to be adopted in real time scenario. It has to be noted that pharmaceutical compounds with higher concentration have lower removal in same category of pharmaceutical compound. The hazard to receiving environment is reduced from high to moderate and low risk pertaining to NSAIDs and antibiotics in MBR and coupled tubesettler. Future studies are required to explore the biodegradation pathway in MBR and tubesettler. Additional studies are required to analyze impact of surfactants and disinfectants on the microbial activity in MBR and tubesettler.

Acknowledgement

The authors extend their appreciation to the Deanship of Research and Graduate Studies at King Khalid University for funding this work through Large Research Project under grant number RGP2/09/46.

REFERENCE

1. Abad, R., Khan, N.A., El, R., Alsubih, M., Rahman, A., Khan, S., Mubashir, M., Balakrishnan, D., Shiong, K., (2023). Comparison of constructed wetland performance coupled with aeration and tubesettler for pharmaceutical compound removal from hospital wastewater. *Environ. Res.* 216, 114437. <https://doi.org/10.1016/j.envres.2022.114437>
2. Alsubih, M., El Morabet, R., Khan, R.A., Khan, N.A., Khan, A.R., Khan, S., Mubarak, N.M., Dehghani, M.H., Singh, L., (2022a). Field performance investigation for constructed wetland clubbed with tubesettler for hospital wastewater treatment. *J. Water Process Eng.* 49, 103147. <https://doi.org/10.1016/j.jwpe.2022.103147>
3. Alsubih, M., El, R., Abad, R., Khan, N.A., (2022b). Performance evaluation of constructed wetland for removal of pharmaceutical compounds from hospital wastewater : Seasonal perspective. *Arab. J. Chem.* 15, 104344. <https://doi.org/10.1016/j.arabjc.2022.104344>
4. Ávila, C., García-Galán, M.J., Uggetti, E., Montemurro, N., García-Vara, M., Pérez, S., García, J., Postigo, C., (2021). Boosting pharmaceutical removal through aeration in constructed wetlands. *J. Hazard. Mater.* 412, 1–10. <https://doi.org/10.1016/j.jhazmat.2021.125231>
5. Besse, J.P., Kausch-Barreto, C., Garric, J., (2008). Exposure assessment of pharmaceuticals and their metabolites in the aquatic environment: Application to the French situation and preliminary prioritization. *Hum. Ecol. Risk Assess.* 14, 665–695. <https://doi.org/10.1080/10807030802235078>
6. Cheng, Y.-L., Lee, C.-Y., Huang, Y.-L., Buckner, C.A., Lafrenie, R.M., Dénomme, J.A., Caswell, J.M., Want, D.A., Gan, G.G., Leong, Y.C., Bee, P.C., Chin, E., Teh, A.K.H., Picco, S., Villegas, L., Tonelli, F., Merlo, M., Rigau, J., Diaz, D., Masuelli, M., Korrapati, S., Kurra, P., Puttugunta, S., Picco, S., Villegas, L., Tonelli, F., Merlo, M., Rigau, J., Diaz, D., Masuelli, M., Tascilar, M., de Jong, F.A., Verweij, J., Mathijssen, R.H.J., (2016). We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists TOP 1%. *Intech* 11, 13.
7. Database, E.C., (2023). PARACETAMOL PNEC VALUE [WWW Document]. Eur. Chem. Agency. URL <https://echa.europa.eu/registration-dossier/-/registered-dossier/12532/6/1> (accessed 4.26.23).
8. dos Santos, C.R., Lebron, Y.A.R., Moreira, V.R., Koch, K., Amaral, M.C.S., (2022). Biodegradability, environmental risk assessment and ecological footprint in wastewater technologies for pharmaceutically active compounds removal. *Bioresour. Technol.* 343, 126150. <https://doi.org/10.1016/j.biortech.2021.126150>
9. Farah, M., Giral, J., Stüber, F., Font, J., Fabregat, A., Fortuny, A., (2023). Hollow fiber liquid membrane: A promising approach for elimination of pharmaceutical compounds from wastewater. *J. Environ. Chem. Eng.* 11, 111544. <https://doi.org/10.1016/j.jece.2023.111544>
10. Frédéric, O., Yves, P., (2014). Pharmaceuticals in hospital wastewater: Their ecotoxicity and contribution to the environmental hazard of the effluent. *Chemosphere* 115, 31–39. <https://doi.org/10.1016/j.chemosphere.2014.01.016>
11. Huschek, G., Hansen, P.D., Maurer, H.H., Kregel,

- D., Kayser, A., (2004). Environmental risk assessment of medicinal products for human use according to European Commission recommendations. *Environ. Toxicol.* 19, 226–240. <https://doi.org/10.1002/tox.20015>
12. Khan, N.A., El Morabet, R., Khan, R.A., Ahmed, S., Dhingra, A., Alsubih, M., Khan, A.R., (2020). Horizontal sub surface flow Constructed Wetlands coupled with tubesettler for hospital wastewater treatment. *J. Environ. Manage.* 267, 110627. <https://doi.org/10.1016/j.jenvman.2020.110627>
 13. Khan, N.A., Morabet, R. El, Khan, R.A., Ahmed, S., Dhingra, A., (2019). Extended Aeration Tubesettler Performance Evaluation for Hospital Wastewater Treatment.
 14. Khan, R.A., Morabet, R. El, Khan, N.A., Ahmed, S., Alsubih, M., Mubarak, N.M., Dehghani, M.H., Karri, R.R., Zomorodiyani, N., (2022). Removal of organic matter and nutrients from hospital wastewater by electro bioreactor coupled with tubesettler. *Sci. Rep.* 12, 1–12. <https://doi.org/10.1038/s41598-022-12166-9>
 15. Kim, M., Guerra, P., Shah, A., Parsa, M., Alae, M., Smyth, S.A., (2014). Removal of pharmaceuticals and personal care products in a membrane bioreactor wastewater treatment plant. *Water Sci. Technol.* 69, 2221–2229. <https://doi.org/10.2166/wst.2014.145>
 16. Lancheros, J.C., Madera-Parra, C.A., Caselles-Osorio, A., Torres-López, W.A., Vargas-Ramírez, X.M., (2019). Ibuprofen and naproxen removal from domestic wastewater using a horizontal subsurface flow constructed wetland coupled to ozonation. *Ecol. Eng.* 135, 89–97. <https://doi.org/10.1016/j.ecoleng.2019.05.007>
 17. Lozano Avilés, A.B., Del Cerro Velázquez, F., Lozano Rivas, F., (2022). Ultrafiltration Membranes System: A Proposal to Remove Emerging Pollutants in Urban Wastewater. *Membranes (Basel)*. 12. <https://doi.org/10.3390/membranes12121234>
 18. Mamo, J., Insa, S., Monclús, H., Rodríguez-Roda, I., Comas, J., Barceló, D., Farré, M.J., (2016). Fate of NDMA precursors through an MBR-NF pilot plant for urban wastewater reclamation and the effect of changing aeration conditions. *Water Res.* 102, 383–393. <https://doi.org/10.1016/j.watres.2016.06.057>
 19. Minguez, L., Pedelucq, J., Farcy, E., Ballandonne, C., Budzinski, H., Halm-Lemeille, M.P., (2016). Toxicities of 48 pharmaceuticals and their freshwater and marine environmental assessment in north-western France. *Environ. Sci. Pollut. Res.* 23, 4992–5001. <https://doi.org/10.1007/s11356-014-3662-5>
 20. Monteoliva-García, A., Martín-Pascual, J., Muñoz, M.M., Poyatos, J.M., (2020). Effects of carrier addition on water quality and pharmaceutical removal capacity of a membrane bioreactor – Advanced oxidation process combined treatment. *Sci. Total Environ.* 708. <https://doi.org/10.1016/j.scitotenv.2019.135104>
 21. Park, J., Lee, S., Lee, E., Noh, H., Seo, Y., Lim, H.H., Shin, H.S., Lee, I., Jung, H., Na, T., Kim, S.D., (2019). Probabilistic ecological risk assessment of heavy metals using the sensitivity of resident organisms in four Korean rivers. *Ecotoxicol. Environ. Saf.* 183, 109483. <https://doi.org/10.1016/j.ecoenv.2019.109483>
 22. Park, J., Yamashita, N., Tanaka, H., (2018). Membrane fouling control and enhanced removal of pharmaceuticals and personal care products by coagulation-MBR. *Chemosphere* 197, 467–476. <https://doi.org/10.1016/j.chemosphere.2018.01.063>
 23. Razak, M.R., Aris, A.Z., Zakaria, N.A.C., Wee, S.Y., Ismail, N.A.H., (2021). Accumulation and risk assessment of heavy metals employing species sensitivity distributions in Linggi River, Negeri Sembilan, Malaysia. *Ecotoxicol. Environ. Saf.* 211, 111905. <https://doi.org/10.1016/j.ecoenv.2021.111905>
 24. Reif, R., Omil, F., Lema, J.M., (2013). *Removal of pharmaceuticals by membrane bioreactor (MBR) technology, 2nd ed, Comprehensive Analytical Chemistry*. Elsevier B.V. <https://doi.org/10.1016/B978-0-444-62657-8.00009-4>
 25. Straub, J.O., (2016). Aquatic environmental risk assessment for human use of the old antibiotic sulfamethoxazole in Europe. *Environ. Toxicol. Chem.* 35, 767–779. <https://doi.org/10.1002/etc.2945>
 26. Tiwari, B., Sellamuthu, B., Ouarda, Y., Drogui, P., Tyagi, R.D., Buelna, G., (2017). Review on fate and mechanism of removal of pharmaceutical pollutants from wastewater using biological approach. *Biore Sour. Technol.* 224, 1–12. <https://doi.org/10.1016/j.biortech.2016.11.042>
 27. Vo, T.K.Q., Bui, X.T., Chen, S.S., Nguyen, P.D., Cao, N.D.T., Vo, T.D.H., Nguyen, T.T., Nguyen, T.B., (2019). Hospital wastewater treatment by sponge membrane bioreactor coupled with ozonation process. *Chemosphere* 230, 377–383. <https://doi.org/10.1016/j.chemosphere.2019.05.009>
 28. Wijekoon, K.C., Hai, F.I., Kang, J., Price, W.E., Guo, W., Ngo, H.H., Nghiem, L.D., (2013). The fate of pharmaceuticals, steroid hormones, phytoestrogens, UV-filters and pesticides during MBR treatment. *Biore Sour. Technol.* 144, 247–254. <https://doi.org/10.1016/j.biortech.2013.06.097>