**Submerged membrane filtration process coupled with powdered activated carbon for nonylphenol ethoxylates removal**

Phuoc-Dan Nguyen\textsuperscript{a,b}, Thi-Minh-Tam Le\textsuperscript{c}, Thi-Kim-Quyen Vo\textsuperscript{d,e}, Phuong-Thao Nguyen\textsuperscript{b,d}, Thi-Dieu-Hien Vo\textsuperscript{a,e}, Bao-Trong Dang\textsuperscript{f}, Nguyen-Thanh Son\textsuperscript{b,g}, Dinh Duc Nguyen\textsuperscript{b,h} and Xuan-Thanh Bui\textsuperscript{b,d,*}

\textsuperscript{a}Asian Center for Water Research (CARE-RESCIF), Ho Chi Minh City University of Technology (HCMUT), Ho Chi Minh City 700000, Vietnam  
\textsuperscript{b}Key Laboratory of Advanced Waste Treatment Technology, Ho Chi Minh City University of Technology (HCMUT), Vietnam National University Ho Chi Minh (VNU-HCM), Ho Chi Minh City 700000, Vietnam  
\textsuperscript{c}Faculty of Environment – Natural Resources and Climate Change, Ho Chi Minh City University of Food Industry (HUFI), 140 Le Trong Tan street, Tay Thanh ward, Tan Phu district, Ho Chi Minh city 700000, Vietnam  
\textsuperscript{d}Faculty of Environment and Natural Resources, Ho Chi Minh City University of Technology (HCMUT), Ho Chi Minh City 700000, Vietnam  
\textsuperscript{e}Faculty of Environmental and Food Engineering, Nguyen Tat Thanh University, 300A Nguyen Tat Thanh, District 4, Ho Chi Minh City 700000, Vietnam  
\textsuperscript{f}Ho Chi Minh City University of Technology (HUTECH) 475A, Dien Bien Phu, Ward 25, Binh Thanh District, Ho Chi Minh City 700000, Vietnam  
\textsuperscript{g}Center for Space and Remote Sensing Research, National Central University, Zhongli District, Taoyuan City, Taiwan  
\textsuperscript{h}Department of Environmental Energy Engineering, Kyonggi University, Suwon 442-760, Republic of Korea  
\textsuperscript{*}Corresponding author. E-mail: bxthanh@hcmut.edu.vn

**ABSTRACT**

A combination of a submerged membrane filtration system and powdered activated carbon (PAC) was investigated for nonylphenol ethoxylates removal. Both filtration flux and initial powdered activated carbon dosage had significant effects on the micropollutants removal efficiency. The best performance was achieved under the filtration flux of 20 L/m\textsuperscript{2}.h and the initial powdered activated carbon of 50 mg/L. The removal efficiencies of nonylphenol ethoxylates was obtained at 75\textpm{}5\% in the first 60 hours, and then decreased at 55\textpm{}7\% and 23\textpm{}11\% in the following hours, respectively. As observed, over 65\% of dissolved organic carbon mass adsorbed into powdered activated carbon that was suspended in the bulk phase, and the remainder was adsorbed into powdered activated carbon that deposited on the membrane surface. It reveals that the combination between submerged membrane filtration and PAC could be an effective solution for enhancing removal of micropollutants from water.

**Key words:** adsorption, hybrid microfiltration membrane, nonylphenol ethoxylates, organic micropollutants, powdered activated carbon, wastewater

**HIGHLIGHTS**

- Continuous microfiltration coupled with PAC effectively removed nonylphenol ethoxylate.  
- 75\textpm{}5\% of nonylphenol ethoxylate was rejected after 60 hours of operation.  
- Removal efficiencies of dissolved organic carbon could reach 53\textpm{}4\%.  
- Over 65\% of dissolved organic carbon mass adsorbed into powder activated carbon.

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INTRODUCTION

Nonylphenol ethoxylates (NPEs) and derivatives are widely used in industrial applications as detergents, dispersing agents, paints, plastics, pesticides, emulsifiers, and demulsifiers, and so on. NPEs are non-ionic surfactants, unstable, and biotransformed to metabolites such as nonylphenol monoethoxylate (NPE1), nonylphenol diethoxylate (NPE2), and nonylphenol triethoxylate (NPE3) nonylphenols (NPs) in the aquatic environment (Ho & Watanabe 2017). NPEs have estrogenic properties and a hydrophobic nature so they are highly toxic to human health and organisms (Soares et al. 2008). NPEs are the most cytotoxic compound to mice, Biomphalaria tenagophila, and Drosophila melanogaster when pesticide formulation has NPEs as an additive (Grisolia et al. 2004). NPs adversely affect aquatic organisms, such as by feminization, male fertility reduction, and low survival rate of juveniles (Soares et al. 2008). On the other hand, Harréus et al. (2002) demonstrated by comet assay that at a concentration of 10–15 μg/mL, NPEs made levels of cytotoxicity increased to damage DNA in human cells. Regular exposure with NPs leads to female breast cancer, male prostate cancer, and negative effects on human intestinal functionality (Kim et al. 2016). Due to these adverse impacts, many countries have started to limit the production and usage of NPEs compounds. The allowable concentration of NPs was recommended as below 6.6 μg/L in freshwater, 1.7 μg/L in saltwater (Brooke & Thursby 2005), and 0.3 μg/L in surface waters (EC 2008). The presence of NPEs in the aquatic environment has been recorded in several countries. NPE1 and NPE2 were detected in Greek sewage treatment plants (0.15–5.76 μg/L), in Polish agricultural drains (0.02–0.70 μg/L) (Stasinakis et al. 2008; Zgola-Grzesikowiak et al. 2009). NPs concentration was observed as 14 μg/L in the effluent of a textile factory in China (Brigden et al. 2011), and 0.5–1.1 μg/L in sewage treatment plants in Japan (Nakada et al. 2006). In Vietnam, NP was detected in urban surface water sources with the concentration of 0.025–0.160 μg/L in Hanoi and 0.026–0.149 μg/L in Ho Chi Minh city (Ho & Watanabe 2017). Consequently, to have safe drinking water supplies, the removal of NPEs and derivatives should be carefully considered.
Many methods have been studied to eliminate NPEs and NPs from water such as adsorption, ozonation, photocatalysis, coagulation, advanced oxidation processes, activated sludge, membrane bioreactor, nanofiltration, reverse osmosis, and so on. Among them, adsorption, oxidation, and membrane filtration are the three main proposed processes. Moreover, NPEs and nonylphenols are hydrophobic compounds and poorly soluble in water, thus adsorption to the solid phase is an effective process (Gao et al. 2014). Some adsorbents have been used to remove NPEs and nonylphenols including silica gel, clays, alginate, cyclodextrins, hyper crosslinked resins, biomass, algae, and magnetic particles. Due to some properties such as being effective, less expensive, available, and suitable for large-scale applications, activated carbon is commonly utilized to remove various organic micropollutants. Choi et al. (2005) investigated different activated carbon types (made from coal/coconut/wood) to remove 0.5 mg/L of nonylphenol by packed-bed reactors filled with 30 cm³ of activated carbon and under a flow rate of 2 mL/min. The results showed that coal-based activated carbon was the most effective adsorbent to remove 72 ± 3% of nonylphenol with an adsorption capacity of 0.06 mg/mg. In a study of Altmann et al. (2014), powdered activated carbon (PAC) was used as an advanced treatment for wastewater treatment plants in Germany, organic micropol-lutants (e.g., carbamazepine, diclofenac) were removed more than 90% with the PAC concentration of 20 mg/L.

Membrane technology has been widely used for water and wastewater treatment in recent decades to remove micropollu-tants that cannot be removed by conventional methods such as sedimentation, adsorption, biodegradation. Nanofiltration membrane (NF) and reverse osmosis (RO) can remove low molecular weight matter such as microparticles, colloidal materials, and most microorganisms while the addition of adsorbents in a microfiltration membrane (MF) or ultrafiltration membrane (UF) assists in micropollutant removal for MF/UF systems (Guo et al. 2005; Cao et al. 2016; Bui et al. 2019; Al-Obaidi et al. 2020). Compared to the above membrane technologies, MF consumes less energy than NF and RO, exhibits similar micropollutant removal to UF, and is easier to operate than RO (Lee et al. 2009). Based on these advantages, a hybrid MF membrane process combined with powdered activated carbon has been employed to enhance micropollutant removal in this study. The addition of PAC in the PAC-MF system would enhance the physical removal of micropollutants that can pass through membranes, decrease the dissolved organic compounds loading on the membrane surface, and reduce membrane fouling (Guo et al. 2005). Gai & Kim (2008) demonstrated that a PAC-MF system operated with an initial PAC dosage of 20 g/L was able to remove 70% of total organic carbon (TOC). Besides, Khan et al. (2009) also indicated that a PAC-MF system with an initial PAC concentration of 40 g/L removed 65–95% of TOC and trihalomethanes (THMs) from river water; however, the removal efficiency only achieved 5–18% in the MF system without adding PAC. In another study, a combination of granular activated carbon and MF was able to remove 45–80% of hydrophobic organics, 50–80% of hydrophilic organics, and 33–92% of pharmaceuticals and personal care products (Shanmuganathan et al. 2015).

Overall, the combination of PAC and MF is very effective due to its advantages such as preventing the PAC washing out, high removal of micropollutants, small footprint, and ease of operation. Although the effectiveness of the PAC-MF process in micropollutant treatment has been demonstrated by various studies, the application of a PAC-MF system for NPEs removal from water and wastewater is still lacking. Therefore, the aim of this study was to investigate the feasibility of the combination of microfiltration membrane and powdered activated carbon for nonylphenol ethoxylates removal, and determine appropriately operational parameters such as PAC dosage and permeate flux.

**MATERIALS AND METHODS**

**Experimental setup**

In this study, three experimental stages were sequentially conducted including batch experiments for evaluating effects of PAC concentration on NPEs removal, determining an appropriate flux for the PAC-MF system, and continuously operating the system. Firstly, batch experiments were conducted to evaluate the effects of PAC concentration on NPEs removal. Six sealed bottles were filled with 200 mL of deionized water and PAC was added to each bottle with various dosages of 25, 50, 100, 150, 200, and 250 mg/L. The initial NPEs concentration was controlled at 4 mg/L. These bottles were shaken at a rate of 25 rpm, and a temperature of 32 °C, within 30 minutes. Then, the NPEs concentration in each bottle was measured to determine the removal efficiency. The batch experiment was replicated two times.

In the next stage, a lab-scale PAC-MF system was set up as shown in Figure 1. A flat sheet membrane module (250-B8, Korea), made from polyethylene with a surface area of 0.15 m² and a pore size of 2.5 μm, was submerged in the reactor (L×W×H of 0.25×0.10×0.45 m) with a working volume of 10 L. The trans-membrane pressure (TMP) was daily recorded by a digital pressure gauge. Air was supplied with a flow rate of 65 L/min to avoid PAC depositing in the reactor. PAC
was added to the system with the dosage chosen by batch experiments. The PAC-MF system was sequentially operated at different fluxes of 10, 15, 20, 25, and 30 L/m²/h (LMH). Each flux was operated for 8 h, the reactor and membrane module were cleaned to start checking the following flux. The suspended and deposited PAC concentrations corresponding to each flux were measured after 8 h of operation. The flux that creates a high suspended PAC concentration would be chosen for the continuous operation.

After determining the appropriate PAC dosage and operational flux, the PAC-MF system was operated within 288 h (12 days) to evaluate the organic micropollutant removal performance. The raw water was composed of tap water and NPEs compound to achieve an influent NPEs concentration of 4 mg/L. Then, PAC was added on the first day and no PAC addition was conducted during the continuous operation period. Additionally, due to continuous sampling (every 3–4 h), a large number of samples were collected. Therefore, the adsorption capacity of PAC for the trace organic compounds in the PAC-MF system was determined via dissolved organic carbon (DOC) concentration. To ensure the accuracy of this, the correlation between NPEs and DOC was tested. The result indicated that NPEs had a high correlation with DOC ($r = 0.990$ and $p = 0.000$). The influent DOC concentration was measured at $2.64 \pm 0.04$ mg/L.

### Characteristics of PAC and NPEs

Powdered activated carbon (Norit SA UF, Netherland) used in this study has the characteristics presented in Table 1. The mean size of PAC was determined by a particle size analyzer (HORIBA, model LA950V2, United Kingdom). NPEs compounds ($C_{15}H_{24}O-(C_2H_4O)nH$ with $n = 1–15$) were purchased from Sigma – Aldrich (USA) with the purity of 98% and have a trading name of Nonidet™ P 40 Substitute.

### Table 1 | Characteristics of powdered activated carbon used in this study

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Iodine number</td>
<td>1100</td>
</tr>
<tr>
<td>Moisture (w/w %)</td>
<td>2</td>
</tr>
<tr>
<td>Average size ($\mu$m)</td>
<td>22</td>
</tr>
<tr>
<td>Density (kg/m$^3$)</td>
<td>225</td>
</tr>
<tr>
<td>pH</td>
<td>Alkaline</td>
</tr>
<tr>
<td>Specific surface area (m²/g)</td>
<td>1200</td>
</tr>
<tr>
<td>Methylene blue adsorption (g/100 g)</td>
<td>24</td>
</tr>
<tr>
<td>Phenol adsorption (g/100 g)</td>
<td>5</td>
</tr>
</tbody>
</table>
Analytical methods

The samples were filtered through 0.45 μm filter paper (GF/F, Whatman, United Kingdom) before measuring the DOC concentration by Total Organic Carbon Analyzer (Model: TOC-VVPH/CPN, Shimadzu, Japan). The solid-phase extraction procedure for NPEs analysis was carried out according to the research results of Dinh et al. (2011). First, 250 mL of water sample had suspended solids removed by being filtered through 0.7 μm filter paper (GF/F, Whatman, United Kingdom). Second, an Oasis hydrophilic-lipophilic-balance (HLB) cartridge (60 mg, 3 mL, Waters Corporation, USA) was conditioned with 5 mL of MeOH, and equilibrated with 5 mL of Up-water. Third, the water sample was loaded through the cartridge at a flow rate of 3–5 mL/min. After loading, the cartridge was rinsed with 3 mL of MeOH:Up-water (5:95, v/v). Then, the cartridge was vacuum dried for 10 minutes and eluted with 5 mL of MeOH. The extract was evaporated by a pure nitrogen stream (99.9999%) at a temperature of 40 °C. The ultra-pure water:CH₂CN (90:10, v/v) with 0.01% formic acid was added into the extract to achieve a volume of 0.5 mL. Finally, the extract was preserved at 4 °C before analyzing by high-performance liquid chromatography (HPLC/UV, Shimadzu, Japan).

For the continuous operation period, the collected sample suspended PAC concentration was immediately measured by a turbidity meter (HACH DR/2010, USA) and then phases separated by centrifugation. The liquid phase DOC and NPEs concentrations were analyzed while PAC was returned to the system to avoid PAC loss. The mass of DOC adsorbed in PAC in the bulk and PAC deposited on the membrane surface was calculated by mass balance equations (Lee et al. 2009).

\[
\frac{dm_{DOC,PAC_t}}{dt} = C_{DOC,f} \cdot F \cdot A - \frac{dc_{DOC,b}}{dt} \cdot V - C_{DOC,p} \cdot F \cdot A \\
\frac{dm_{DOC,PAC_b}}{dt} = C_{DOC,f} \cdot F \cdot A - \frac{dc_{DOC,b}}{dt} \cdot V - C_{DOC,p} \cdot F \cdot A - \frac{dm_{PAC_m}}{dt} \cdot \left( \frac{m_{DOC,PAC_m}}{m_{PAC_m}} \right) \\
\frac{dm_{DOC,PAC_m}}{dt} = C_{DOC,b} \cdot F \cdot A - C_{DOC,p} \cdot F \cdot A + \frac{dm_{PAC_m}}{dt} \cdot \left( \frac{m_{DOC,PAC_m}}{m_{PAC_m}} \right)
\]

where:
- \(m_{DOC,PAC_t}\): mass of DOC adsorbed on the total PAC mass (mg);
- \(m_{DOC,PAC_b}\): mass of DOC adsorbed on PAC suspended in the bulk (mg);
- \(m_{DOC,PAC_m}\): mass of DOC adsorbed on PAC deposited on the membrane surface (mg);
- \(C_{DOC,f}\): DOC concentration in the influent (mg/L);
- \(C_{DOC,b}\): DOC concentration in the bulk (mg/L);
- \(C_{DOC,p}\): DOC concentration in the permeate (mg/L);
- \(m_{PAC_b}\): mass of PAC suspended in the bulk (mg);
- \(m_{PAC_m}\): mass of PAC deposited on the membrane surface (mg);
- \(F\): flux (LMH);
- \(A\): membrane surface area (m²);
- \(V\): working volume of the reactor (L);
- \(t\): sampling time (h).

RESULTS AND DISCUSSION

Determining the PAC dosage and operational flux

The initial PAC dosage and filtration flux are the major operating parameters of PAC-MF systems (Shanmuganathan et al. 2015). Thus, to elucidate the effect of PAC dosage, different concentrations of 25–250 mg/L were tested. Data in Table 2 indicates that more NPEs compounds were removed with increasing PAC dosage. The NPEs removal efficiency was obtained at 58 ± 2% under PAC dosage of 25 mg/L and improved by 26–29% as PAC dosage increased to 250 mg/L. In addition, there was no significant difference in NPEs removal when PAC dosage was varied from 50 to 250 mg/L, but the difference in dosage used was remarkable. This may affect the operation of membrane-based systems and increase the treatment cost. Consequently, the appropriate PAC dosage chosen for the PAC-MF system was 50 mg/L.
Due to the suction, PAC in the system tends to move to the membrane module and deposit on the membrane surface. As a result, PAC in the system is divided into PAC deposited on the membrane surface and PAC suspended in the bulk phase. Lee et al. (2009) demonstrated that the micropollutant adsorption capacity of PAC in the bulk phase was higher compared to deposited PAC. Moreover, the permeate flux is one of the factors influencing the PAC distribution in reactors. Increasing operational flux strengthens the suction force on the membrane surface, which develops PAC deposition. The suspended PAC concentration at different fluxes is shown in Table 2. After 8 h of operation, the PAC suspended in the bulk phase at fluxes of 10–25 LMH was insignificantly different but under the flux of 30 LMH, the suspended PAC concentration was markedly reduced. A similar trend was observed in the study of Lee et al. (2009); the suspended PAC concentration was obtained around 0.9–1.0 mg/L at fluxes of 40–80 LMH and decreased to 0.2 mg/L at the flux of 120 LMH after 8 h. In addition, the suspended PAC concentration at 20 LMH was slightly higher than that of other fluxes; consequently, 20 LMH was an appropriate flux for the continuous operational period. This flux was also chosen to operate the PAC-MF system in some studies (Gai & Kim 2008; Khan et al. 2009).

### NPEs and DOC removal by PAC-MF system

Based on the results from batch experiments, the PAC-MF system was operated continuously for 288 h (12 days) under a flux of 20 LMH and a PAC dosage of 50 mg/L. PAC was added on the first day and no PAC addition was conducted during the operational period. As mentioned before, PAC in the system tends to move to the membrane module and deposit on the membrane surface. Thus, the PAC concentration in the bulk phase decreased with time, whereas the mass of deposited PAC increased. Data from Figures 2 and 3 show the correlation between micropollutant removal and suspended PAC concentration. In the first 40 hours, although suspended PAC in the bulk phase was strongly decreased from 50.00 ± 0.30 mg/L to 16.4 ± 3.3 mg/L, the concentration of suspended PAC existing in the bulk phase was still high of 36.4 ± 4.4 mg/L.

![Sample PAC](image_url)

**Figure 2** | Variation of powdered activated carbon (PAC) concentration in the bulk phase during the operation period.

### Table 2 | Batch experiments to determining PAC dosage and operational flux for the PAC-MF system

<table>
<thead>
<tr>
<th>PAC dosage (mg/L)</th>
<th>25</th>
<th>50</th>
<th>100</th>
<th>150</th>
<th>200</th>
<th>250</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effluent NPEs concentration (mg/L)</td>
<td>1.70 ± 0.01</td>
<td>0.63 ± 0.01</td>
<td>0.55 ± 0.00</td>
<td>0.58 ± 0.00</td>
<td>0.51 ± 0.01</td>
<td>0.51 ± 0.01</td>
</tr>
<tr>
<td>NPEs removal efficiency (%)</td>
<td>58 ± 2</td>
<td>84 ± 2</td>
<td>86 ± 1</td>
<td>86 ± 1</td>
<td>87 ± 1</td>
<td>87 ± 1</td>
</tr>
<tr>
<td>Suspended PAC concentration (mg/L)</td>
<td>30 ± 2.6</td>
<td>29.7 ± 2.1</td>
<td>33.2 ± 1.1</td>
<td>30.4 ± 1.5</td>
<td>23.3 ± 2.1</td>
<td></td>
</tr>
</tbody>
</table>

Operational flux (LMH)

- 10
- 15
- 20
- 25
- 30
DOC concentration in the permeate was $1.25 \pm 0.12$ mg/L corresponded to a removal efficiency of $58 \pm 4\%$. However, from 43rd to 168th hours, the suspended PAC concentration was decreased to $12.20 \pm 2.31$ mg/L and the DOC removal efficiency achieved was $45 \pm 5\%$. By the remaining hours, the PAC concentration in the bulk phase was only $9.4 \pm 1.1$ mg/L, and the lowest DOC removal efficiency was observed at $23 \pm 11\%$. The effluent DOC concentration ($2.05 \pm 0.32$ mg/L) obtained in this period was approximate to that of the influent ($2.64 \pm 0.04$ mg/L). It is clear that the depletion of PAC in the bulk phase resulted in a reduction in micropollutant removal. The DOC concentration in the permeate from the 200th hour onwards was two-fold higher than that of the first hours. From these results, PAC should be supplemented to the system after 40 h to maintain the removal performance. The organic micropollutant removal capacity of PAC-MF system was demonstrated in previous studies, the TOC removal efficiency achieved was 70% (Gai & Kim 2008), or 65–95% of Trihalomethanes and TOC from river water was removed by adding PAC in an MF reactor (Khan et al. 2009).

NPEs concentration was measured during the operational period. The influent NPEs was maintained at 4 mg/L, and the NPEs concentration in the permeate fluctuated at $1.34 \pm 0.41$ mg/L (Figure 3). Like DOC removal performance, the PAC concentration in the reactor affected the NPEs treatment. Due to the high suspended PAC concentration in the first 50 hours, the

**Figure 3** | Variation of DOC and NPEs concentrations in the permeate during the operation period.

**Figure 4** | Mass of DOC adsorbed on total PAC mass during the operation period.
NPEs removal efficiency was achieved at 81 ± 3%, then reduced to 66 ± 5% in the following hours. Choi et al. (2005) demonstrated that the coal-based activated carbon was effective for NPs removal. A column filled with 30 cm³ of coal-based activated carbon was operated at a flow rate of 2 mL/min, an empty bed contact time of 15 min, and the initial NPs concentration of 0.5 mg/L. The results indicated that NPs was completely adsorbed by activated carbon, and NPs was detected in the effluent after 29 days of operation. Compared with biological treatment processes, NPEs removal by the PAC-MF system in this study showed to be more effective. For example, Patureau et al. (2008) evaluated the NPEs removal efficiency of several biological processes operated in continuously stirred tank reactors with a hydraulic retention time (HRT) of 20 days. The anaerobic process removed 25–30% of NPEs, and the aerobic process achieved 39%. While the combined process increased the NPEs removal efficiency by comparison to the single process, such as a combination of aerobic and anaerobic processes (45%) or anaerobic and ozonation processes (48%).

Table 3 | Removal efficiencies of NP, NPE1, NPE2 by various methods

<table>
<thead>
<tr>
<th>Systems</th>
<th>Mechanisms</th>
<th>Operational conditions</th>
<th>Removal efficiencies</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conventional activated sludge process (Aerotank)</td>
<td>- Biodegradation - Adsorption onto sludge</td>
<td>- Municipal wastewater - Initial NPs, NPE1, NPE2 concentrations: 0.55–3.02, 0.47–4.03, 0.49–2.67 μg/L - HRT: 64 h</td>
<td>- NPs: 35–77% - NPE1: 86–97% - NPE2: 92–95%</td>
<td>Pothitou &amp; Voutsia (2008)</td>
</tr>
<tr>
<td>Anaerobic/Oxic system</td>
<td>- Biodegradation - Adsorption onto sludge</td>
<td>- Municipal wastewater - Initial NPs, NPE1, NPE2 concentrations: 1.80–2.00 μg/L - Capacity: 1.6 × 10⁵ m³/d</td>
<td>- NPs: 78% - NPE1: 84% - NPE2: 89%</td>
<td>Gao et al. (2014)</td>
</tr>
<tr>
<td>Biological Aerated Filter system</td>
<td>- Biodegradation - Adsorption onto sludge</td>
<td>- Municipal wastewater - Initial NPs, NPE1, NPE2 concentration: 1.80–2.00 μg/L - Capacity: 4.0 × 10⁴ m³/d</td>
<td>- NPs: 55% - NPE1: 76% - NPE2: 79%</td>
<td>Gao et al. (2014)</td>
</tr>
<tr>
<td>Conventional activated sludge process (Aerotank)</td>
<td>- Biodegradation - Adsorption onto sludge</td>
<td>- Textile wastewater - Initial NPs concentration: 0.47–0.53 μg/L - HRT: 16.2–20.8 h - SRT: 60.8–66.3 d</td>
<td>- NPs: 56–77%</td>
<td>Ho &amp; Watanabe (2017)</td>
</tr>
<tr>
<td>Packed-bed reactor</td>
<td>- Adsorption on granular activated carbon</td>
<td>- Synthetic wastewater - Flow rate: 2 mL/min - EBCT: 15 min - Initial NPs concentration: 500 μg/L - GAC volume: 30 cm³</td>
<td>- NPs: 72 ± 3%</td>
<td>Choi et al. (2005)</td>
</tr>
<tr>
<td>Coagulation</td>
<td>- Coagulation</td>
<td>- Textile wastewater - Initial NPs, NPE1, NPE2 concentrations: 1.16, 7.90, 10.50 μg/L</td>
<td>- NPs: 53% - NPE1: 91% - NPE2: 94%</td>
<td>Pothitou &amp; Voutsia (2008)</td>
</tr>
<tr>
<td>Membrane filtration coupled with powdered activated carbon</td>
<td>- Adsorption onto PAC; - Filtration by membrane</td>
<td>- Synthetic wastewater - Initial NPEs concentration: 4.00 × 10⁵ μg/L - HRT: 3 h - PAC dosage: 50 mg/L</td>
<td>- NPEs: 75 ± 5%</td>
<td>This study</td>
</tr>
</tbody>
</table>

Remarks: NPs, nonylphenols; NPEs, nonylphenol ethoxylates; NPE1, nonylphenol monoethoxylate; NPE2, nonylphenol diethoxylate; HRT, hydraulic retention time; SRT, sludge retention time; EBCT, empty bed contact time; PAC, powdered activated carbon; GAC, granular activated carbon.
Additionally, the adsorption capacity of PAC in the system is shown in Figure 4. The suspended PAC always exhibits higher adsorption capacity compared to deposited PAC. Average 65 ± 10% of DOC mass adsorbed on PAC suspended in the bulk, and this result is consistent with the study of Lee et al. (2009).

Membrane fouling

PAC in the system tends to deposit on the membrane surface, resulting in the TMP development. The results showed that TMP rapidly increased from 1.65 to 2.25 kPa in the first 40 hours and stabilized at around 2.25–2.70 kPa in 41–288 h. In the first 40 hours, the PAC concentration in the bulk phase decreased dramatically from 50 to 16.4 mg/L, which means more PAC deposited on the membrane surface. Consequently, TMP value rapidly increased with the fouling rate of 0.015 kPa/h. In the following hours, there was no significant increase of PAC deposited on the membrane surface, thus the fouling rate was lower than 0.002 kPa/h. This result was similar to the study of Uyak et al. (2014), TMP also increased rapidly over a period of 70 hours of operation.

CONCLUSIONS

The PAC-MF system shows a higher nonylphenol ethoxylates removal rate (24 g NPEs/m³/d) than that of other technologies (Table 3). Consequently, the submerged membrane filtration system coupled with powdered activated carbon is a prospective method for NPEs removal from natural water bodies, especially nonylphenol ethoxylates. Besides, PAC-MF system can be employed as an advanced process for wastewater treatment, especially for treated wastewater containing micropollutants like NPEs. PAC also serves as an effective adsorbent for micropollutants that cannot be rejected by membrane filtration alone. PAC existing in the system includes PAC suspended in the bulk phase and PAC deposited on the membrane surface. The suspended PAC showed a higher adsorption capacity compared to deposited PAC due to over 65% of dissolved organic carbons being adsorbed in it. However, the reduction of suspended PAC in the bulk phase leads to the deterioration of NPEs removal efficiency which is the main drawback of the PAC-MF system. Thus, to maintain the desired removal, the suspended PAC mass should be supplemented. Determining how much (same with initial dosage or less) and when PAC is added (every 40 h or longer) should be carried out in further studies.

COMPETING INTERESTS

The authors declare that they have no competing interests.

AUTHORS’ CONTRIBUTIONS

Phuoc-Dan Nguyen, Thi-Minh-Tam Le: Investigation; Formal analysis; Visualization; Writing- Original draft preparation. Thi-Kim-Quyen Vo: Methodology; Writing- Reviewing and Editing.

Xuan Thanh Bui & Phuoc-Dan Nguyen: Conceptualization; Project administration; Funding acquisition, Supervision; Phuong-Thao Nguyen, Bao-Trong Dang: Software, Resources. Thi-Dieu-Hien Vo: Validation, Editing; Dinh Duc Nguyen, Nguyen-Thanh Son.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES


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