Distribution and Removal of Nonylphenol Ethoxylates and Nonylphenol from Textile Wastewater—A Comparison of a Cotton and a Synthetic Fiber Factory in Vietnam

Hien Ho [1] and Tsunemi Watanabe [1,2,*]

[1] Graduate School of Engineering, Kochi University of Technology, Tosayamada, Kami City, Kochi 782-8502, Japan; thanhhien119.ho@gmail.com

[2] School of Economics and Management, Kochi University of Technology, 2-22 Eikokuji, Kochi City, Kochi 780-8515, Japan

* Correspondence: watanabe.tsunemi@kochi-tech.ac.jp; Tel.: +81-805-6662-2871

Abstract: The textile industry is a significant source of nonyphenol and their ethoxylates, which are suggested to be responsible for endocrine disruption in wildlife and humans. This study is a comparison of two conventional wastewater treatment processes in a cotton and a synthetic fiber factory in Vietnam, with regard to the distribution and removal of nonyphenol ethoxylates and nonyphenol throughout each process. Diverse trends in the distribution of nonyphenol ethoxylates in wastewater from factories, distinguished by their raw materials, could be revealed. Primary coagulation might not perfectly facilitate nitrification in the secondary activated sludge process regarding pH. Nevertheless, satisfactory removals were achieved during coagulation and activated sludge processes in both systems. The roles of long hydraulic retention times (21 and 16 h, respectively), low organic loadings (0.1 and 0.2 gCOD/gMLVSS.day, respectively), extended solids retention times (61 and 66 days, respectively), and mixed liquor suspended solids of greater than 2000 mg/L have been demonstrated. The findings provide evidence and a better understanding of nonyphenol ethoxylate and nonyphenol removal efficacy as well as influencing factors in Vietnamese textile wastewater treatment. The results are beneficial for the textile industry in Vietnam regarding investment decisions for wastewater treatment.

Keywords: coagulation; activated sludge process; temperature; nitrifying conditions; hydraulic retention time; solids retention time; cotton; synthetic fibers; textile wastewater

1. Introduction

Vietnam is one of the top ten textile exporters in the world. From 2004 to 2014, the textile industry achieved a compound annual growth rate (CAGR) of about 19% per year, increasing its contribution to Vietnam's GDP by 5% to 15% [1]. About 62% of factories are established in southern Vietnam [2]. Raw materials in the textile industry include natural fibers (cotton, wool, silk), synthetic fibers (nylon, polyester, viscose), and a blend of natural and synthetic materials [3,4]. The textile industry is one of the most polluting industries, releasing highly toxic and persistent chemicals into the environment, especially the watercourse [3,5]. It is suggested that nonylphenol ethoxylates (NPEOs) are still widely used in the textile industry [6] as detergents and auxiliaries in wool scouring, hydrogen peroxide bleaching [7], washing, dyeing, and printing [8,9]. Studies by Brigden et al. [10–12] have demonstrated the worldwide presence of NPEOs in the majority of textile products, regarding all materials, and across most of the countries of manufacture and consumption. The authors also suggest that lower

traces of NPEOs in textile products may imply a higher discharge of NPEOs into the watercourse during production.

NPEOs are indirectly responsible for endocrine disruption among wildlife and human beings via their metabolites, especially nonylphenol (NP) [13,14]. NP, on the one hand, is shown to be persistent at different levels in the environment [15–17]. On the other hand, NP has a high affinity for solids such as sediment, sewage sludge, and soil amended with sewage sludge [18,19], as well as for lipids [20]; hence, it has been shown to accumulate in organisms [21,22]. Past research has demonstrated the presence of NP in all trophic levels, such as plankton, benthic invertebrates, fish, birds, and mammals [23–29], via a complex food web [30]. NP has been detected in foodstuffs [26,31–34], drinking water [35], human adipose tissue [36], urine [37], maternal blood plasma and amniotic fluid [38], blood serum [33], and breast milk [20,31,39]. The presence of NP in pregnant women's decidua and early embryos along with maternal transfers has also been observed [40].

Adverse effects of NP on reproductive, immune, and central nervous systems have been discovered in fish, rats, birds, and humans with possible abnormalities in embryos and offspring [41–53]. Recent studies on carcinogenesis have reflected the relation of exposure to NP to the possibilities of breast cancer in women [54] and prostate cancer in men [55,56]. A study of Lepretti et al. [57] revealed negative impacts on human intestinal homeostasis and functionality. The mechanisms of action of NP are related to xenoestrogens [58–60], antiestrogens [61], and disruption of thyroid function [62]; which occur on nuclear (genomic) [54], extranuclear (non-genomic) [63], and cross-talk between genomic and non-genomic pathways [64,65].

Concerning the negative impacts on the ecosystem and human beings, NP/NPEOs have been added to the list of chemicals for priority action since 1998 by the OSPAR Commission [66]. From 2000, NP has been classified as a priority hazardous substance under the Directive 2000/60/EC of the European Parliament and the Council. Marketing and use restrictions of NP/NPEOs have also been put into place under Directive 2003/53/EC, and the concentration of NP in surface waters has been regulated in Directive 2008/105/EC. Accordingly, the average annual level of NP should not exceed 0.3 µg/L. Nevertheless, NP and NPEOs are allowable for use today in many Asian countries including Vietnam.

In Vietnam, NP has been detected at extremely high levels in urban watercourses such as in the cities of Ha Noi and Hochiminh, in the range of 0.02–9.7 µg/L (mean 3.0 µg/L) and 2.0–20.0 µg/L (mean 9.7 µg/L), respectively [67]. Regarding environmental risk, it is suggested that NP may cause ecological effects due to its high risk quotient (expressed as the ratio of Measured Environmental Concentration (MEC) to Predicted No-Effect Concentration (PNEC). An MEC/PNEC ratio of greater than 1 indicates a potential hazard or risk) of 128 [68]. Recent investigations in textile manufacture in developing countries such as Thailand, China, Mexico and Indonesia by Greenpeace have revealed that NP/NPEOs are among the most commonly detected hazardous chemicals in the effluent of wastewater treatment plants (WWTPs) [8]. NP and NPEOs with one and two ethoxylate group(s) have been identified as the most dominated alkylphenols in the effluent of PT Gistex, one of the biggest textile factories in Indonesia [69]. Brigden et al. [70] reported a level of up to 14 µg/L of NP at the effluent of Youngor Textile Complex, Yangtze River Delta, China. These concentrations of NP and NPEOs are not representative of Vietnam, but they illustrate the current problematic condition of textile production in developing countries in Asia [8].

Every textile manufacturer in Vietnam utilizes WWTPs. However, their designs are only for addressing macro-pollutant issues with a major application of conventional processes [71]. It is suggested that conventional processes are insufficient to effectively remove NPEOs and NP in wastewater [69,72]. Therefore, the effective elimination of endocrine disrupting compounds is a great challenge for the Vietnamese textile industry. More investigations on the existing textile WWTPs in Vietnam regarding the removal of nonylphenol are needed. On this basis, the objectives of this study are:

- To investigate the distribution and removal of NPEOs and NP across two typical textile wastewater treatment processes.

- To assess the influence of hydraulic retention time (HRT), nitrifying conditions, solids retention time (SRT), mixed liquor suspended solids (MLSS), and water temperature on the NPEO and NP removal.

2. Treatment Processes and Factors Influencing Nonylphenol and Nonylphenol Ethoxylate Removal

In aqueous phase, NPEOs undergo biotransformation as a predominant process under aerobic conditions [16,73–76], and partly under anaerobic conditions [18,77]. Consequently, metabolites such as nonylphenol mono- and di-ethoxylate (NP$_{1-2}$EO), nonylphenol mono- and di-ethoxy carboxylate (NP$_{1-2}$EC), and the final refractory product NP are formed [18,19,78–80]. They have great hydrophobicity except for NP$_{1-2}$EC and less biodegradability. Due to their lipophilicity, NP and NP$_{1-2}$EO can be eliminated via biotransformation and sorption [18,81,82]. Although a variety of wastewater treatment processes have been studied for their ability to remove nonylphenolic compounds from wastewater, such as ozonation [83], in particular for the treatment of textile wastewaters [84], it seems that conventional processes still dominate in practice. Primary coagulation/flocculation followed by a secondary anaerobic/anoxic/aerobic activated sludge has been shown to be popularly applied in the textile industry [85], particularly in Vietnam [3,71]. The removal of nonylphenolic compounds depends on the nature of the processes [72], and factors such as the population served by the sewage system [86], wastewater compositions [81,87], hydraulic retention time (HRT) [88], biomass concentration (expressed as MLSS), solids retention time (SRT), pH and temperature [81]. Table 1 demonstrates the removal efficacies of NPEOs and NP by some commonly used wastewater treatment processes.

2.1. Removal by the Coagulation Process

The main implication of coagulation is to remove total suspended solids and colloids in wastewater [17]. In textile wastewater treatment, taking advantage of particle removal, coagulation also functions for the elimination of dye agents that are microbial inhibitory, and contribute to the color of wastewater [3]. Regarding nonylphenolic compounds, the coagulation process has shown its capacity for removing NP, NP$_1$EO and NP$_2$EO [18], which are considered hydrophobic with high partition coefficients (log K_{ow}) of 4.48, 4.17, and 4.21, respectively [89]. High removals of over 90% of NP [90], and over 90% of NP$_{1-2}$EO [87] were achieved during this process. Coagulation and flocculation efficiencies are dependent on wastewater compositions, coagulants, and process conditions such as pH and mixing [91].

2.2. Removal by Anaerobic Processes

Under anaerobic conditions, a complete de-ethoxylation of NPEOs with the formation of NP could be obtained [18]. The results of Zhou, et al. [92] demonstrated that up to 200% of the NP was formed from the conversion of NPEOs in anaerobic units, suggesting the elimination of NP by this process was negligible. In contrast, the anaerobic biological process was shown to contribute a great deal to NP removal, at 82.5%, with a total removal efficiency of the whole biological system of 89% [93]. Similarly, a majority of NP being removed in the anaerobic bioreactor, a rate of 42%, with a total efficacy of 67%, through a combined anaerobic-activated sludge process was reported [72]. Diverse potencies have been documented probably because the biotransformation of NP in anaerobic conditions may strictly rely on the availability of nitrate in wastewater as an electron acceptor [94]. Indeed, the study of Wang et al. [95] revealed that NP was mainly removed via sorption on sludge, and no biodegradation was observed under anaerobic-without-nitrate conditions, suggesting the dominant role of nitrate in NP removal via anaerobic processes.

2.3. Removal by Anoxic Processes

Literature on the elimination of NPEOs and NP in anoxic conditions is scarce. However, Ferguson and Brownawell [96] suggested that only anoxic conditions were able to complete the de-ethoxylation of NPEOs. However, anoxic degradation of NPEOs might not last long since it requires a combination of an electron acceptor and a specialized microbial consortium [94] or denitrifying conditions [97]. Lu et al. [97] suggested a more efficient removal of NPEOs by denitrifying activated sludge, rather than by anaerobic activated sludge. Regarding NP, H. Zhou et al. [98] observed an approximately 82% reduction in NP within 8 h. Whereas, Wang et al. [95] reported a removal of 89% after 120 h. The authors demonstrated a rise in removal efficacy at an increased nitrate concentration, where the most suitable COD/nitrate ratio was of 15:1. It may imply that the degradation of NP is also facilitated by denitrification.

2.4. Removal by Aerobic Activated Processes

Removal efficiency of NPEOs and NP in wastewater by aerobic activated sludge processes (ASPs) has been demonstrated by numerous scholars (Table 1). Tan et al. [93] revealed that conventional activated sludge systems could be particularly effective and had an NP removal potential of 85% to 99% in the influents comprising municipal, industrial and hospital wastewater in mixture. Zhou Z. et al. [92] affirmed the major role of ASPs, which contributed 93–94% over total NP removals of three denim WWTPs. Though biotransformation dominates in anaerobic and anoxic processes, it appears that in aerobic ASPs the removal of nonylphenolic compounds takes place via biodegradation and wastage of excessive sludge [98,99]. Accordingly, a major portion of the removal of long-chain NPEOs (with log K_{ow} less than 2.5) occurs via biotransformation [100], whereas removal of NP and short-chain NPEOs (with log K_{ow} greater than 4) is more likely via adsorption onto biomass [101,102]. Factors such as HRT, nitrification, organic loading, SRT, biomass concentration (expressed as MLSS), temperature, and pH have been suggested to be significant for governing those mechanisms [81,103–105].

Table 1. Removal efficiency of nonylphenol and nonylphenol ethoxylates.

Substance	Anaerobic Process [a]/ Coagulation Process [b]	Activated Sludge Process	Conditions	Source
NP	82.5% [a]	6.5%	Anaerobic followed by aerobic activated sludge	[93]
NP	42% [a]	25%	Anaerobic activated sludge followed by oxidation ditch SRT~16–17 days	[72]
NP	Negligible	93–94%	Anaerobic followed by aerobic activated sludge * HRT *~7.8–9.8 h SRT *~10.5–11.9 days MLSS *~3500–4000 mg/L	[92]
NP	-	~99%	Activated sludge Long SRT~25 days	[106]
NP	-	97–99%	Activated sludge with nitrifying Ammonia removal ~62% to >90% HRT~10 h SRT~3–20 days MLSS~500–3000 mg/L	[107]
NP	-	80%	Activated sludge Temperature~20–30 °C MLSS~1676 mg/L HTR~64 h Relatively low loadings	[87]
NP$_{1-2}$EO	-	>90%		
NP		70%	Activated sludge Long SRT~27 days HRT~16 h	[108]
NP$_{1-3}$EO	-	93%		
NP$_{4-12}$EO		93%		

NP		37%		
NP$_1$EO	-	−3% (produced)	Activated sludge	[109]
NP$_2$EO		−5% (produced)	High loading/ non-nitrifying	
NP$_6$EO		78%		
NP		77%		
NP$_1$EO	-	31%	Activated sludge	[109]
NP$_2$EO		91%	Low loading/ nitrifying	
NP$_6$EO		98%		
NP and NPEOs	-	>80%	Activated sludge Low loading/ nitrifying STR > 10 days	[110]
NP and NP$_{1\text{-}2}$EO	-	>90%	Activated sludge Low loading/ nitrifying-denitrifying	[111]
NP	>90% [b]	-	Coagulation	[90]
NP	53% [b]			
NP$_1$EO	91% [b]	-	Coagulation	[87]
NP$_2$EO	94% [b]			

Notes: * Operational parameters of aerobic activated sludge process; [a] Anaerobic process; [b] Coagulation process; - no available data.

3. Materials and Methods

3.1. Selection of Wastewater Treatment Processes

Surveys of 120 textile manufactures in Vietnam in 2010 revealed two types of WWTPs [71]. For the first type, a primary coagulation followed by a sand- or powder-activated carbon filtration was connected to the central WWTP of an industrial zone. The second type included WWTPs, in which effluent wastewater was discharged directly into the watercourse. These WWTPs employed a biological process in addition to physico-chemical processes. An investigation of forty textile WWTPs in southern Vietnam revealed that wastewater treatment processes for cotton or blended fabrics mainly involved the physico-chemical process (coagulation) and the activated sludge process (ASP), whereas those for polyester required both primary and tertiary physico-chemical processes in combination with a secondary biological process. In some cases, ozone oxidation, filtration, or electrochemical processes were used in place of tertiary treatment [112]. In general, ASP is the most widely used wastewater treatment process in the Vietnamese textile industry [3].

Removal of micropollutants is governed by factors such as the physico-chemical properties of substances, wastewater compositions and characteristics, and wastewater treatment processes linked with treatment conditions [113]. With given properties of NPEOs and their metabolites, differences in the nature of wastewater and treatment processes for cotton/ blended fabrics and synthetic materials were of interest.

Our criteria for selecting WWTPs for the investigation were inherited from the outcome of the surveys by the Vietnam Environment Administration—VEA [71] and the Center for Environmental Technology and Management—CENTEMA [112], taking into account the influent concentrations of NPEOs and NP as well as treatment processes, as previously discussed. Since there is no regulated limit for the discharge of NP in Vietnam, we supposed that WWTPs that directly discharge into waterbodies might pose a threat to the aquatic ecosystem. Therefore, one textile WWTP connecting to the central WWTP of an industrial zone and another connecting to a river were selected. Second, the textile factories should involve a dying—finishing process. This process comprises three main steps—preparing, dyeing and/or printing, and finishing—in all of which surfactants and auxiliaries are used for impurity removal, enhancement of color agent dispersion, abrasion resistance and improvement of the tear strength of fabric [3]. Lastly, materials of each factory should be typically different.

Consequently, among some factories that met the three aforementioned criteria, permission from two factories to carry out the survey was granted. Hoa Sen factory was established in 2002 and is specialized in the production of cotton fabric. Chyang Sheng factory has been in operation since 1996 and produces synthetic garment products. Both are Taiwan-owned factories. The WWTPs of

both factories employ a physico-chemical process followed by a biological process. The WWTP of Hoa Sen factory, namely as F1, adopts a primary coagulation prior to a consecutive ASP and disinfection. For the WWTP of Chyang Sheng factory, namely as F2, a primary coagulation is followed by a secondary ASP and a tertiary coagulation before disinfection.

3.2. Field Survey and Sampling

The survey campaign took place from 5 September to 7 September 2016, at Hoa Sen and Chyang Sheng. Since this is the first investigation with the primary purpose of demonstrating the presence (with distribution) or absence of nonylphenolic compounds in textile wastewater in Vietnam, grab sampling could be sufficient [114]. Sampling was performed as suggested by Pothitou and Voutsa [87]. On each day, grab wastewater samples were collected from the equalization tanks and at the outlets of treatment facilities such as primary clarifiers (following coagulation process), secondary clarifiers (after ASP), and a disinfection tank (for F1) or tertiary clarifier (after advanced treatment by coagulation, for F2). The wastewater containing activated sludge was collected directly from the aeration tanks, and the returning sludge sample was collected from the sludge outlets of the secondary clarifiers. Wastewater samples were stored in 1 L glass bottles, which had been well rinsed with acetone solution, followed by Milli-Q solution, and finally evaporated by a drying facility. Sludge samples were stored in 1 L plastic bottles. All samples were kept in ice containers when they were conveyed to laboratories on the day. For those parameters such as temperature, pH, and DO, we conducted the measurements on site. Sampling positions are shown in Figure 1. Treatment processes and technical specifications such as design capacity and operating capacity, facility volume, hydraulic retention time, sludge returning schedule, and chemical use of the two wastewater treatment facilities were also investigated.

The estimations of solids retention time (SRT), returning activated sludge ratio, organic loading, and removal rate were detailed as follows.

Traditional mass-balance approach for the estimation of SRT (day) [92]:

$$\text{SRT} = \frac{V_A X_A}{\left(Q - Q_W\right) X_E + Q_W X_R} \tag{1}$$

where V_A denotes the volume of the aeration tank (m³). X_A, X_E, and X_R indicate the concentration of biomass in the aeration tank (g/m³), the concentration of biomass in the effluent (g/m³), and the concentration of returning activated sludge (g/m³), respectively. In our study, the effluent total suspended solids (TSS) was assumed to be X_E, and the mixed liquor suspended solids (MLSS) to be X_A. Q and Q_W represent the wastewater flowrate (m³/day) and the waste activated sludge flowrate (m³/day), respectively.

Estimation of returning activated sludge ratio (R) [91]:

$$R = \frac{1 - \left(\text{HRT}/\text{SRT}\right)}{\left(X_R/X_A\right) - 1} \tag{2}$$

where HRT denotes the hydraulic retention time in the aeration tank (day).

Estimation of organic loading (F/M, g/g.day) [91]:

$$F/M = \frac{QS_o}{VX} \tag{3}$$

where S_o, V, and X indicate influent concentration (g/m³), volume (m³), and biomass concentration (g/m³), respectively. In this case, S_o was the filtered COD in the influent, V was equal to V_A, and X was the mixed liquor volatile suspended solids (MLVSS) in the aeration tank.
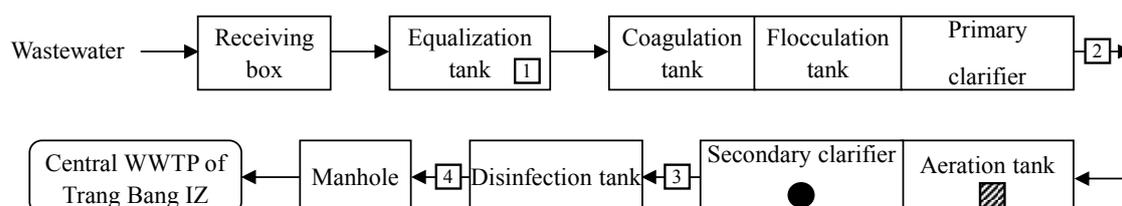
Estimation of removal rate (%):

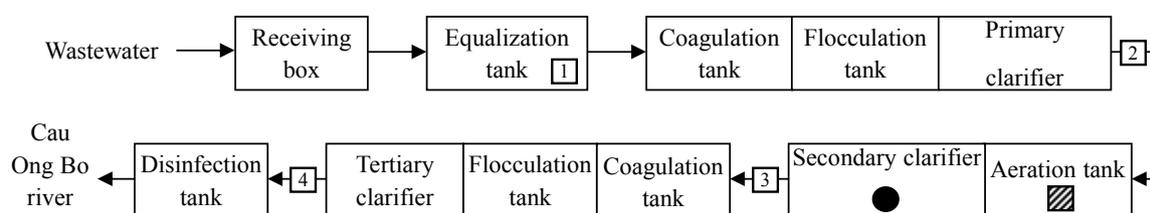$$\text{Removal rate} = \frac{(S_o - S_e) \times 100}{S_o} \tag{4}$$

where S_o and S_e represent the influent and the effluent concentrations (g/m³), respectively.

　　Nonylphenol ethoxylates are denoted as NPEOs or NP$_n$EO, where n indicates the numbers of ethoxylate unit.

(a) Wastewater treatment process of F1:

(b) Wastewater treatment process of F2:

1 2 3 4　Wastewater sampling points

▨　Wastewater containing activated sludge sampling point

●　Returning sludge sampling point

Figure 1. Wastewater treatment processes and sampling points at (**a**) Hoa Sen factory—F1 and (**b**) Chyang Sheng factory—F2.

3.3. Method of Analysis

　　Macro-compositions such as color, total suspended solids (TSS), mixed liquor suspended solids (MLSS), alkalinity, organic matter in terms of chemical oxygen demand (COD) and biological oxygen demand (BOD$_5$), ammonia (N–NH$_4^+$), nitrite (N–NO$_2^-$), and nitrate (N–NO$_3^-$) were analyzed at the laboratory of CENTEMA, Vietnam, following the standard methods for the examination of water and wastewater [115].

　　Solid phase extraction of NP and NPEOs was conducted at the Laboratory for Advanced Waste Treatment Technology of the National University of Hochiminh city, Vietnam. We followed the solid phase extraction procedures as described by Tuc Dinh, et al. [116]. First, 200 mL of each wastewater sample was filtered through 0.7 μm glass fiber filters (GF/F, Whatman) to remove suspended solids. Second, the cartridges were conditioned with 3 mL of high performance liquid chromatography grade methanol (MeOH) of Merck, and then equilibrated with 3 mL of Milli-Q water. SampliQ solid phase extraction cartridges C18 ODS (Agilent, Santa Clara, CA, USA) were used. Third, wastewater samples were loaded through the cartridges at a flow rate of 3–5 mL per minute. Depending on the characteristic of the wastewater samples, loading volumes were adjusted from 100 to 134 mL so that the cartridges would not get blocked. When the loading was almost finished, the cartridges were rinsed with 3 mL of MeOH:Milli-Q (5:95) solution, and then vacuum dried for 10 min. Next, elution was performed with 5 mL of MeOH solution each. After that, evaporation of the extracts was

conducted under a gentle 99.999% pure nitrogen stream at 40 °C to dryness. Residues were dissolved in 1 mL of gradient grade for liquid chromatography acetonitrile (CH_3CN) solution of Merck each and well shacked. Finally, the resulting solid phase extracts were injected through 0.45 μm syringe filters into 1.5 mL vials and preserved at 4 °C. The analysis of extracted samples was performed in Japan to take advantage of modern and more sensitive equipment needed for data accuracy. During the transportation to Japan, the samples were preserved in an ice container [67].

Analysis of NP and NPEOs was conducted at the laboratory of the Foundation for Promotion of Material Science and Technology of Japan. NP and NPEOs (from NP_1EO to $NP_{15}EO$) were quantified using a liquid chromatograph (LC-Prominence, Shimadzu, Kyoto, Japan) followed by a tandem mass spectrometer (MS-4500 Qtrap, AB SCIEX, Framingham, MA, USA). In liquid chromatography, an Inertsil PH column (150 mm × 2.1 mm, 5 μm, GL Science, Japan) was employed and maintained at 40 °C, where the mobile phase A was a 5 mmol/L ammonium acetate (CH_3COONH_4) solution and the mobile phase B was a MeOH solution. The injection volume was 10 μL and the flow rate was at 0.2 mL per minute. In mass spectrometry, an electrospray ionization (ESI) method and a multiple reaction monitoring (MRM) mode were employed. The details of MRM transitions are given as supplementary data (Table S1).

Recovery experiments were conducted with 100 mL filtered water samples spiked with 400 ng/L of each aforementioned alkylphenolic substances. The same solid phase extraction method and quantification method were applied.

4. Results and Discussion

4.1. Design Figures and Operating Conditions of the Factories during the Campaign

The design capacities of the WWTPs of F1 and F2 were 1000 m³/day and 3500 m³/day, respectively. Each factory had just resumed operations after a three-day holiday when we started the campaign. During the campaign, the flowrate of F1 was at about 44% of its full capacity, and F2 was operating at approximately 54% of its full capacity, where the full flowrates were assumed to be equal to 80% of the design capacity (a safety factor of 1.25). Operating figures of the biological process such as HRT in the aeration tank and SRT were achieved from the data of flowrates, aeration volumes, sludge returning scheme, and the analyzed results of MLSS, effluent solids concentration, and returning solids concentration. Technical figures of the two WWTPs that were necessary for the assessment of NPEO and NP removal were summarized in Table 2.

Table 2. Design and operating figures of the WWTPs of Hoa Sen and Chyang Sheng factories.

Factory	Wastewater Flowrate Q (m³/Day)		Aeration Volume V_A (m³)	MLSS/MLVSS X_A [a] (mg/L)	Waste Solids Flow Rate Q_W (m³/d)	Returning Solids Concentration X_R [a] (mg/L)	Effluent Solids Concentration X_E [a] (mg/L)	HRT [b] (h)	SRT [b] (Day)	R [b]
F1	During the campaign	700	607.2	2267/1651	0.8	9913	21	20.8	60.8	0.29
	At full capacity [b]	800	607.2	-	-	-	-	18.2	-	-
F2	During the campaign	1500	1013.8	2657/2141	1.6	4753	22	16.2	66.3	1.25
	At full capacity [b]	2800	1013.8	-	-	-	-	8.7	-	-

Notes: MLSS/ MLVSS: mixed liquor suspended solids/ mixed liquor volatile suspended solids in aeration tank; HRT: Hydraulic retention time in the aeration tank; SRT: Solids retention time in the activated sludge process; R: returning activated sludge ratio. Correspondingly, returning sludge flowrates for F1 and F2 were 205 m³/day and 1,881 m³/day, respectively. [a] Analysis result; [b] Estimation by the authors; "-" means data not available.

4.2. Textile Wastewater Compositions

Wastewater released from manufacturing processes differs in both quantity and quality, depending on dyeing technologies as well as types and doses of chemicals [112]. Combined streams from equalization tanks of various textile factories across Vietnam are characterized by high values

of pH (over the favorable range for microbial growth of 8.5), temperature (36–52 °C), color (350–3710 Pt-Co), organic matter expressed in COD (360–2448 mgO$_2$/L) and BOD$_5$ (200–1450 mgO$_2$/L) [112].

Influent wastewater of both factories was collected from equalization tanks with aeration where wastewater flowrates and compositions were homogenized. In comparison with the reported compositions and characteristics of textile wastewater in Vietnam, influent wastewater compositions of F1 and F2 (Table 3), were rather low, with average TSS values of 139 and 159 mg/L, and COD of 317 and 541 mgO$_2$/L, respectively. Exceptionally, the wastewater had high color values of 848 Pt-Co (F1) and 945 Pt-Co (F2). The influent pH of F1 ranged from 8.6 up to 10.0, while that of F2 was moderately lower, between 7.9 and 8.6. As a characteristic of the textile industry, the influent wastewater was high in temperature, at an average of 40 °C and 41 °C for F1 and F2, respectively. Comparing compositions of the disinfection effluent (F1) and the tertiary effluent (F2: during the campaign, the disinfection of F2 was not in operation) with the national technical regulation on the effluent of textile industry, QCVN 13:2008/BTNMT, it was suggested that only F1 might fully comply with the standard in terms of pH, color, TSS, and organic matter.

Table 3. Compositions of influent and effluent textile wastewater at the two factories.

Composition	Unit	Hoa Sen Factory—F1 [a]		Chyang Sheng Factory—F2 [a]		QCVN 13:2008/BTNMT	
		Influent	Disinfection Effluent	Influent	Tertiary Effluent	Class A	Class B
pH	-	8.6–10.1	7.8–8.0	7.9–8.6	6.8–7.4	6–9	5.5–9.0
Temperature	°C	40	-	41	-	40	40
Color (pH = 7)	Pt-Co	848	31	945	87	20 [b]/50 [c]	150
TSS	mg/L	139	19	159	19	50	100
COD$_{total}$	mgO$_2$/L	317	-	541	-	50	150
COD$_f$	mgO$_2$/L	247	36	416	101	-	-
BOD$_{5-f}$	mgO$_2$/L	163	7	180	57	30 [d]	50 [d]

Notes: [a] Average compositions in the three-day campaign in this study; [b] For newly established factories; [c] For the existing factories; [d] Maximum allowable level for total BOD$_5$. COD$_f$ and BOD$_{5-f}$: COD and BOD$_5$ of wastewater samples that had passed through 0.7 µm glass fiber filters, respectively. Class A: wastewater to be discharged into the waters for domestic supply; Class B: wastewater to be discharged into the waters for other purposes except for domestic supply; "-" means data not available.

In order to assess the removal of NPEOs and NP, wastewater samples at the effluents of each treatment stage were analyzed for both macro-pollutants and micro-pollutants, such as NP$_{1-15}$EO and NP. The analysis results are presented in Table 4. The facilities demonstrated rather high efficacy in overall removal of COD$_f$ (75% and 82%), BOD$_{5-f}$ (95% and 69%), and TSS (effluent TSS of 18 and 19 mg/L), for F1 and F2, respectively. During the campaign, F2 might have performed a partial nitrification. The ammonia removal efficiency was 23%, equivalent to 20 mg/L. The loss in ammonia might be attributed to the ammonia oxidation and the conversion of slowly biodegradable organic matter (a COD$_f$ reduction of 73%) into new cells. Nitrite concentration was shown to accumulate with an additional amount of 8 mg/L. A small decrease in nitrate (0.6 mg/L) was probably due to the assimilation by heterotrophic bacteria or/and denitrification occurring in one section (one third) of the aeration tank where moving bed media were introduced [117,118]. However, ammonia was not detected from the primary and secondary effluents of F1. It was suggested that ammonia in the equalization tank of F1 probably evaporated at high pH (pH = 10.1), so a conclusion on the nitrification of F1 could not be made. Further discussion on nitrification conditions linked with NPEO and NP removal will be provided in the next section.

Regarding micro-pollutants, both NPEOs and NP were detected in the influent wastewater of the two factories. The concentration of total NP$_{1-15}$EO of F1 was 117,594 ng/L, which was about 40 times higher than that of F2, as 3099 ng/L. The presence of NP at 474 ng/L and 551 ng/L in the influents of F1 and F2, respectively, was due to the degradation of the parent ethoxylates in the sewage systems. Our study demonstrated rather good overall removals of NPEOs (at the rates of >99% and >79%), and NP (at the rates of 77% and 56%) for F1 and F2, respectively. NP concentrations of

the secondary effluents were below the European Union environmental quality standard of 0.3 μg/L [119]. The reported results of NP and NPEOs had been adjusted using the recovery rates of individual compounds (Figure 2).

Table 4. Wastewater compositions after each treatment process.

Composition	Unit	Hoa Sen Factory [a]				Chyang Sheng Factory [b]			
		Equalization	Primary Effluent	Secondary Effluent	Disinfection Effluent	Equalization	Primary Effluent	Secondary Effluent	Tertiary Effluent
pH	-	10.1	6.7	7.7	7.9	8.1	7.1	7.8	7.4
Temperature	°C	39	-	34 [c]	-	41	-	37 [c]	-
DO	mgO_2/L	-	-	4.8 [c]	-	-	-	4.3 [c]	-
Alkalinity	$mgCaCO_3/L$	700	280	320	-	450	300	190	-
TSS	mg/L	46	25	25	18	212	3	15	19
COD_f	mgO_2/L	265	157	58	66	570	504	138	104
BOD_{5-f}	mgO_2/L	165	28	25	8	297	246	53	92
$N–NH_4^+$	mg/L	55	<MDL	<MDL	-	85	87	67	-
$N–NO_2^-$	mg/L	0.4	0.2	0.0	-	0.8	0.4	8.4	-
$N–NO_3^-$	mg/L	1.0	2.2	2.1	-	3.4	2.3	1.7	-
NP	ng/L	474	487	109	-	551	1141	244	-
$\Sigma NP_{1-15}EO$	ng/L	117,594	6469	<SQL	-	3099	4453	<SQL	-

Notes: [a] Wastewater compositions on 6 September 2016; [b] Wastewater compositions on 7 September 2016; [c] Parameters of wastewater in aeration tanks. MDL: Method detection limit; MDL_{NH4+} = 5 mg/L; SQL: Sample Quantification Limit; $SQL_{NPEOs-F1}$ = 540 ng/L; $SQL_{NPEOs-F2}$ = 660 ng/L; "-" means data not available.
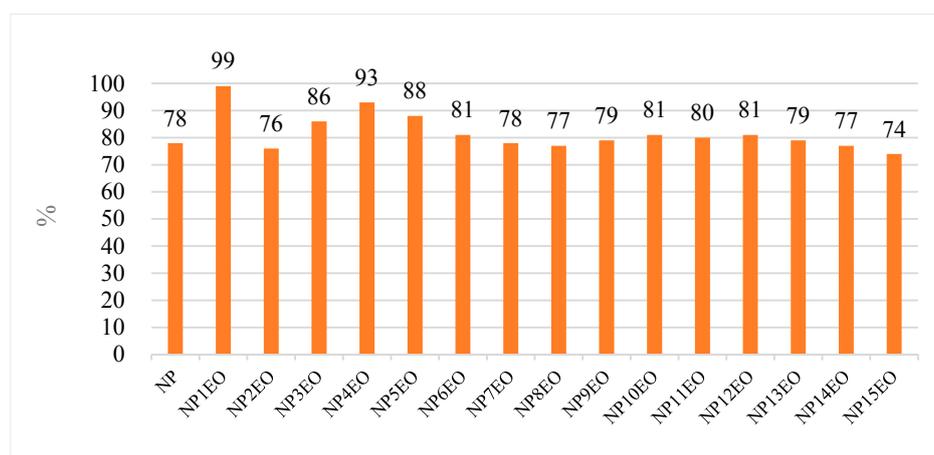

Figure 2. Recovery rate for nonylphenol and individual nonylphenol ethoxylate(s).

An inspection of oligomer distributions (Figures 3 and 4) revealed similar patterns for the influent wastewater and the primary effluents for both WWTPs. The influent wastewater from F1 (specializing in cotton materials) contained $NP_{4-17}EO$ in majority, accounting for 98.5% of the detected homologues, which seemed close to the distribution of NPEO oligomers found by Loos et al. [7]. Accordingly, the proportion of individual oligomers of this group varied between 3.0% and 13.7%. In contrast, influent wastewater from F2, which is based on synthetic fabrics, was dominated by short-chain oligomers (NP_1EO: 61.4% and NP_2EO: 12.4%), reflecting the similarity with the findings of Pothitou and Voutsa [87]. Because the total NP_{1-15}–EO levels of the secondary effluents were below the sample quantification limits for both factories, they were not shown in the following figures.

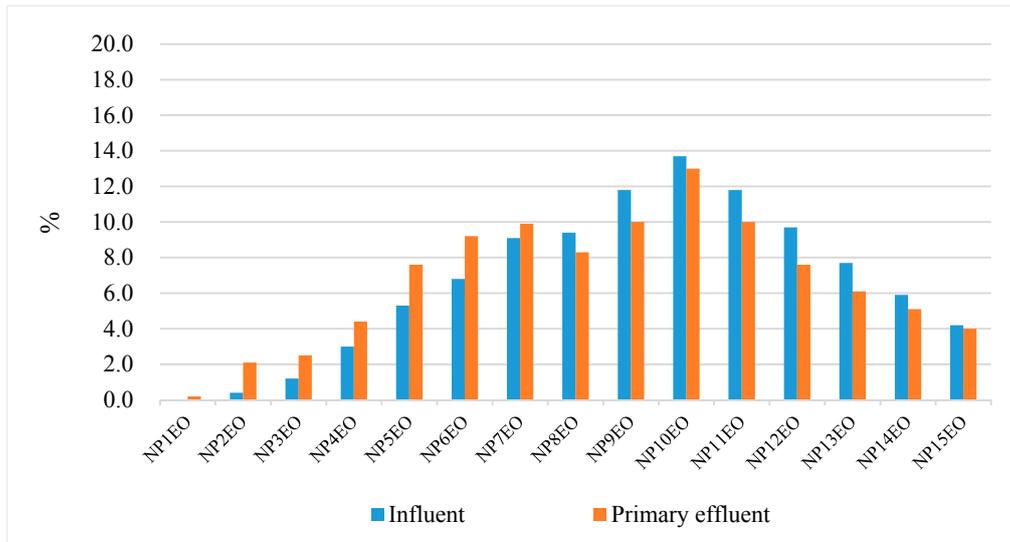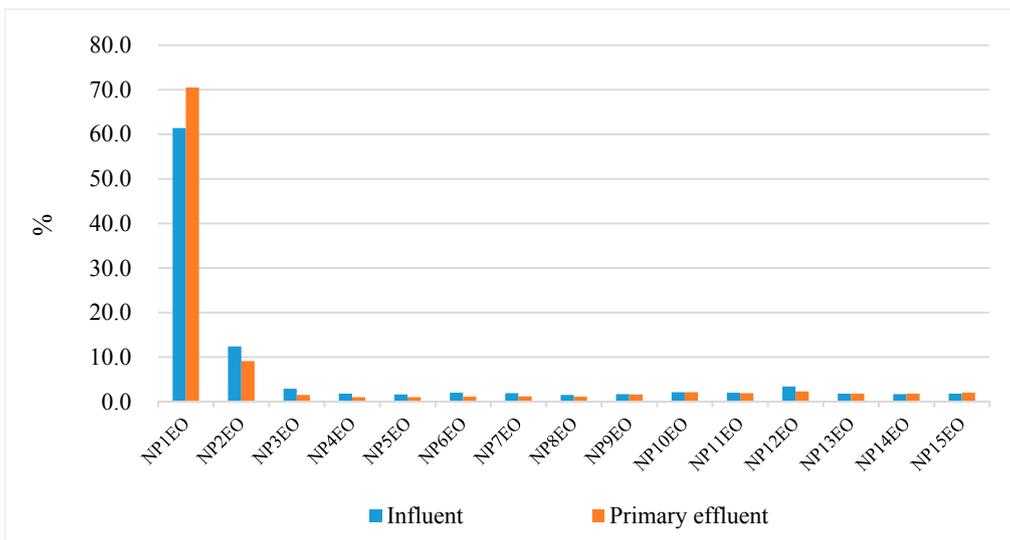**Figure 3.** Distribution of nonylphenol ethoxylates for F1.

Figure 4. Distribution of nonylphenol ethoxylates for F2.

4.3. Removal of NPEO and NP in the Two Textile Factories of Investigation

In this section, critical factors influencing the removal of NPEOs and NP by the wastewater treatment processes, which have been applied in the two textile factories, are addressed and discussed. With the achieved data of NPEOs and NP, we focus on the primary coagulation and the secondary ASP. Consequently, the effects of temperature and TSS in the primary coagulation, as well as the roles of HTR, nitrification, SRT, and MLSS in the ASP on NPEO and NP removal will be discussed in depth.

4.3.1. The Role of Primary Coagulation

Coagulation has been widely applied as primary treatment (e.g., at Hoa Sen factory—F1), as tertiary treatment, or both (e.g., at Chyang Sheng factory—F2) in textile wastewater treatment in Vietnam [71]. It is capable of eliminating 73% to 85% of the influent TSS [112], and about 50% of the color [3]. Regarding nonylphenolic compounds, Stackelberg et al. [120] and Nam et al. [121] reported an NP removal of about 15–16% during the coagulation process. Nevertheless, greater removals of

60% of nonylphenol equivalent (NP$_{equ}$) [18] or up to 75% of NP [121] could be obtained with an increased turbidity in terms of solids concentration. On the contrary, a shortening of medium-long NPEOs, which are more hydrophilic, to produce more persistent metabolites has been observed in primary treatment by Ahel et al. [18] and Loyo-Rosales et al. [122]. This process is elevated with an increased wastewater temperature [104].

Results from F1 revealed that NP$_{2-15}$EO concentration was reduced by 94% from 117,594 ng/L to 6469 ng/L during the primary coagulation process (Figure 5). At the same time, a small elevation of NP and NP$_1$EO by about 5%, was observed. The net increase in NP and NP$_1$EO suggests that their formation from the degradation of the parent oligomers (NP$_{2-15}$EO) slightly exceeds their removal via adsorption onto solids. This result , on the one hand, confirms the finding of Vogelsang et al. [90] that the coagulation process primarily removes the most long-chain ethoxylates, where a temperature of 40 °C of the influent wastewater greatly contributes to the elimination of these compounds. On the other hand, it has reflected a high affinity of NP and NP$_1$EO on particulate matter due to their great hydrophobicity.

We found from F2 that there was a 19% reduction in NP$_{3-7}$EO, from 337 ng/L to 281 ng/L, in relation to a 66% increase of the metabolites mainly as NP and NP$_1$EO, from 2745 ng/L to 4552 ng/L, during the primary coagulation process (Figure 6). At the same time, a small amount of NP$_{8-15}$EO was produced, raising their concentration from 569 ng/L to 762 ng/L. As discussed earlier, the observed behaviors of NP and their ethoxylates followed two main mechanisms as sorption and biotransformation, as suggested by Fauser et al. [82]. Particularly, our results have shown dissimilar patterns of transformation between intermediate oligomers (NP$_{3-7}$EO) and higher oligomers, which have been described by Ahel et al. [18]. The increase in NP$_{8-15}$EO level was probably due to the degradation of longer-chain oligomers. Indeed, higher oligomers with more than 15 ethoxylate groups such as NP$_{4-17}$EO [7] and NP$_{35-40}$EO [90] have been reported for textile wastewater.
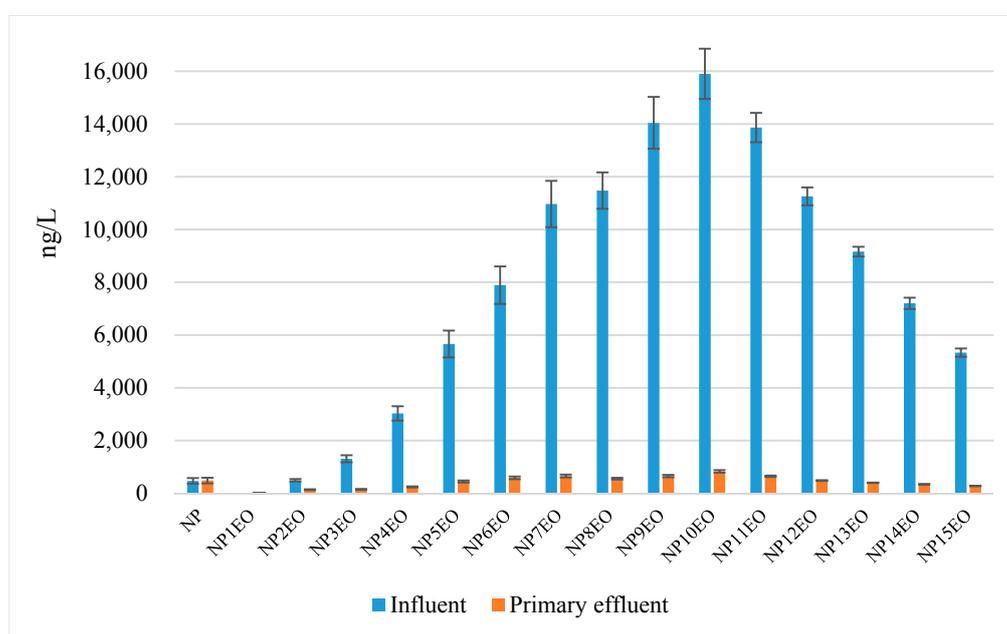

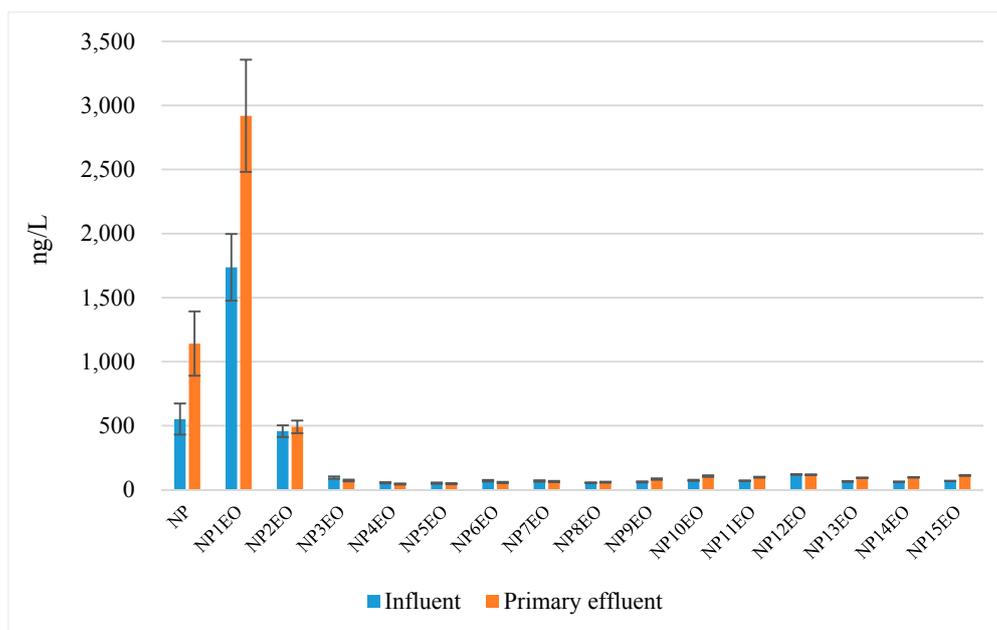**Figure 5.** Concentration of NP and individual NP$_n$EO for F1.

Figure 6. Concentration of NP and individual NP$_n$EO for F2.

4.3.2. The Role of the Secondary Activated Sludge Process

- Removal Efficacy of NPEOs and NP

Although anaerobic processes and advanced aerobic processes (e.g., membrane bioreactor—MBR, moving bed biofilm reactor—MBBR) have been introduced, conventional ASPs still dominate in textile wastewater treatment in Vietnam. Hence, discussing the removal efficacy of NPEOs and NP as well as influencing factors of this process is of great importance that contributes to the practice of wastewater treatment design and operation in Vietnamese textile industry.

Our study at Hoa Sen factory (F1) and Chyang Sheng factory (F2) revealed good removals of NP by the ASPs, at 78% and 79%, respectively. The estimation of removal efficacy did not take into account the formation of NP during the ASPs. The effluent concentrations of NP, as low as 109 ng/L (F1) and 244 ng/L (F2) are comparable to those from a similar treatment process reported by Vogelsang et al. [90]. Both facilities also showed satisfactory reductions of NP$_{1-15}$EO to <540 ng/L (F1) and <660 ng/L (F2), equivalent to removal rates of >92% and >85%, respectively. Our results of NPEO removal are slightly lower than those achieved from ASPs (92–96%) reported by Petrie et al. [108]. This is possibly because the study of Petrie et al. [108] is based on well-controlled pilot-scale models. The following will discuss the possible operational conditions that may affect the NPEO and NP removal efficacies of F1 and F2.

- Persistence of NP and NPEO Metabolites and the Implication of HRT

In an activated sludge system designed for endocrine disruption compound (EDC) removal, HRT is a parameter of great importance since a complete degradation can be achieved with an adequate HRT [103,105]. The biotransformation of NPEOs occurs in two steps. Firstly, the oxidation of ethoxylate chain as primary degradation may take place as quickly as within 10 h of aeration [123,124]. The next phase as mineralization of decomposed products may consume an additional 10 h for the induction of specialized enzymes or shifts in bacterial inoculum that may degrade those substances via different mechanisms [109,123,125]. Consequently, an ultimate degradation time of about 20 h at the F/M (substrate/biomass) ratio of 0.24–0.86 is suggested by Carvalho et al. [123]. Nevertheless, Petrie et al. [108] argues that the shortening of medium-to-long nonylphenolics (NP$_{4-12}$EO) may not be dependent on the HRT, whereas the degradation of short-chain compounds (NP$_{1-4}$EO) and NP is probably mediated by increasing HTR from 8 to 24 h.

During our campaign, the activated sludge processes of F1 and F2 were operated at HRT values of 21 h and 16 h, corresponding to operational capacities of 700 m³/day and 1500 m³/day, respectively. High removals of NP and NP$_{1-15}$EO obtained for both F1 and F2 were consistent with the findings of Petrie et al. [108], as mentioned above. However, at full operation (F1: 800 m³/day, F2: 2800 m³/day), HRT values would be shortened to 18 h and 9 h for F1 and F2, respectively. In that case, F1 may be still on the safe side regarding nonylphenolic removal, but F2 may be at risk in terms of complete mineralization of NPEO metabolites, as suggested by Birkett and Lester [109], Carvalho et al. [123], Maki et al. [125], and Petrie et al. [108].

From experience of design and operation of WWTPs in Vietnam, it is suggested that HRTs of between 6 and 8 h would be sufficient for textile wastewater treatment [112]. Taking into account of micro-pollutant removal, our study, amongst others, has provided evidence that HRTs of no less than 16 h might be favorable for removing NP and its parent compounds from textile wastewater. Therefore, we suggest that the existing ASPs with extended HRTs for textile wastewater treatment in Vietnam should be maintained. Nevertheless, long HRT systems may result in large footprints. Therefore, for those enterprises of space constraint who are likely to improve their WWTPs' performance, a modified approach would be to introduce carrier materials, specifically fixed-film, into activated sludge systems, where EDC removal could be enhanced as hydrophobic organic substances are trapped onto the surface of floc [126].

- The Role of Nitrification

It has been indicated that nitrifying conditions may play an important role in NP and short-chain NPEO removal with ASPs [81,104,105,109]. At the same time, NPEOs with longer chain (more ethoxylate groups) are more eager to decompose. In nitrifying conditions, co-metabolic oxidation with ammonium monooxygenase enzyme possibly allows the assimilation of unreadily biodegradable EDCs [110,127,128]. Körner et al. [129] reported a marginal estrogenic response, with a Relative Proliferative Effect (RPE) of 10% (in the study of Körner et al. [129], RPE indicated the proliferative effect of effluent wastewater on the MCF-7 breast cancer cells relative to the positive control 17β-estradiol (E2)) of the effluent from a textile WWTP adopting an ASP with nitrification-denitrification. In a strong agreement, Vader et al. [130] suggested that high removal of estrogens (in the range of 79–95%) was mainly attributed to the nitrifying stage. Findings of McAdam et al. [131] also showed the same patterns where nitrifying appeared in higher de-ethoxylation of long-chain nonylphenolic compounds (NP$_{5-12}$EO), at approximately 92%, and partial removal of NP, whereas non-nitrifying process yielded poorer de-ethoxylation and an accumulation of NP.

Conditions for nitrifying in wastewater treatment have been widely studied. It is suggested that nitrification could be affected by various factors, such as pH coupled with alkalinity, temperature, DO, organic loading, toxic compounds available in wastewater, HRT and SRT, which may vary between ammonium oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB) [132–138]. Accordingly, alkaline pH of 8.4–8.5, temperature of 28–36 °C, and organic loading in the range of 0.15–1.1 gCOD/gMLVSS.day [135] were reported with uninhibited nitrification. A study of Park et al. [139] provided insights of the optimal pH values of 8.2 ± 0.3 for AOB and 7.9 ± 0.4 for NOB. Increasing HRTs from 12 to 20 and to 22 h exhibited a poor nitrification, a partial nitrification with peak nitrite accumulation, and a complete nitrification, respectively [138].

Compared with the reported nitrifying conditions, the pH values in the ASPs of the two factories of 7.7–7.8 did not lie in the optimal range but might sustain the development of both AOB and NOB [139]. The pH in the ASP is partly influenced by the primary process where pH adjusting chemicals are introduced to facilitate the coagulation reactions. The DO levels of 4.8 and 4.3 mgO$_2$/L indicated an aerobic environment, and the temperatures of 34 and 37 °C might provide proper conditions for nitrifiers in F1 and F2, respectively. Our estimation of organic loadings resulted in acceptable values for nitrification, at 0.1 gCOD/gMLVSS.day (F1) and 0.2 gCOD/gMLVSS.day (F2). An HRT of 20.8 h might favor both nitritation and nitratation in F1, whereas an HRT of 16.2 h in F2

was beneficial for the growth of AOB superior to NOB, leading to an accumulation of nitrite as mentioned above [138,140].

Importantly, nitrification can take place in the presence of autotrophic bacterial species that are believed to be slowly growing [113,135]. Thus, nitrifying usually involves an extended SRT that sustains the development of a diverse microbial population including the nitrifiers [81,104]. The role of STR in NP and NPEO removal will be discussed in the next section.

- Extended SRT as an Essence of Microbial Development and Biomass Quality

A complete biotransformation of short-chain NPEOs, as aforementioned, may require a specialized microbial inoculum that could be enriched in an elevated SRT system [81,105,110,141]. According to Cirja et al. [81], microbial communities including nitrifying bacteria are able to acclimate and develop at the SRTs of longer than 8 days. Langford et al. [103] reported an SRT of 14 days which was sufficient to obtain a diverse population of microorganisms. The authors also indicated a greater production of shorter-chain NPEOs along with rapid losses of long-chain oligomers. Indeed, an investigation of three WWTPs in Beijing China by Zhou H. et al. [72] demonstrated the role of SRT. The results showed that the system with longer SRT (16–17 days) obtained a higher NP removal (65%) compared with the other two. Satisfying elimination of NP was achieved at the SRT of 25 days [106] up to 30 days [88]. It should be noticeable that the degradation of medium-long NPEOs may not be necessarily acquired by long SRTs, which may be only the case for short-chain oligomers (NP$_{1-4}$EO) and NP [108].

In addition, sorption onto activated sludge flocs of hydrophobic nonylphenolic compounds could be enhanced at high SRTs [103]. An extended sludge age may be concurrent with an increase in MLSS [102], where the partitioning of NP between dissolved and particulate phases is attributed to the SS or MLSS concentration in wastewater [142]. Moreover, it is suggested that bacteria surface may become more hydrophobic and less negatively charged at high SRTs, resulting in more partition [143].

From the operating figures at F1 and F2 (Table 2) such as aeration volume, biomass concentration, waste solids generation and concentration, and effluent solids concentration, SRT values were estimated using the mass balance approach [91]. As a result, SRT values for F1 and F2 of approximately 61 days and 66 days, respectively, exceed the expected SRT for short-chain NPEO decomposition. Therefore, it is suggested that the success in NPEO and NP removal at the two factories is attributed to satisfactory SRTs although the pH might be unsupportive for an optimal nitrifying.

In the majority of textile WWTPs in Vietnam, the parameter of SRT is not sufficiently taken into account. Information on SRTs was only minimally derivable from a survey of 120 textile WWTPs by VEA [71]. It was also indicated that small and medium enterprises paid insufficient attention on operating WWTPs as well as training responsible staffs. According to Aboobakar et al. [144], most of the existing ASPs have been designed at mid-ranged conditions such as 10-day SRT and 8 h HRT, which may not support a complete NPEO and NP removal.

- The Implication of MLSS in Removing Hydrophobic Compounds

ASPs have an advantage in the removal of nonylphenolic compounds due to the abundance of bio-solids as suggested by Brunner et al. [142]. Partition of nonylphenolic compounds in secondary sludge has been reported by various researchers. Petrie et al. [108] documented 58–99% of NP, 68–91% of NP$_{1-3}$EO, and 77–85% of NP$_{4-12}$EO in attachment to bio-solids at the SRT of 27 days and HRTs of 8–24 h. Similarly, NP was found in association with activated sludge from 62% [145] up to 93% [146], or in the range of 80–90% at 1000–1700 mgMLSS/L [142]. Consequently, sorption in addition to biodegradation made up high removals, up to 99% of NP and NP$_{1-2}$EO at a 1676 mgMLSS/L [87], and over 99% of NP at 3000 mgMLSS/L [107]. Indeed, adsorption of NP and NPEOs on solids may keep increasing till the Critical Micelle Concentration (CMC: the concentration at which the system's free energy can be reduced by the aggregation of the surfactant molecules into clusters with the hydrophobic groups located at the center of the cluster and hydrophilic head groups towards the solution) is

reached [147]. Hence, a certain MLSS level should be sustained for nonylphenolic removal, which probably depends on their concentration in wastewater.

Regarding the two factories under investigation, the average MLSS values of F1 and F2 during the campaign were 2267 mg/L and 2657 mg/L, respectively, which fell between the MLSS range suggested for complete-mixing ASPs as 1500–4000 mg/L [91]. In combination with field observation at the secondary clarifiers and data of the effluent TSS concentration (as 19 mg/L—Table 3), it could be suggested that good removals of bio-solids were achieved. This may lead to a considerable reduction in NP and short-chain NPEOs by sorption at the detected MLSS levels of F1 and F2.

4.4. Limitations of This Study

This study is not without shortcomings. First, grab sampling insufficiently reflects trends and "real" concentrations of wastewater from the two factories, which would lead to bias in the conclusion on consistently good nonylphenolic removals. Results could only reflect the concentrations and removal efficacies at the time of survey. Second, although the equalization tanks function in regulating flows as well as variations in wastewater compositions on a day, short-term variations in terms of hourly events could occur, leading to variations in NPEO and NP removal efficacies which were still undiscovered by this study. Third, this study detected NPEOs and NP in Vietnamese textile wastewater only in the aqueous phase. This should be addressed by future investigations over the solid phase.

5. Conclusions

This study, among others, has demonstrated the presence and distribution of NPEOs and their final metabolite as NP in textile wastewater. To the best of our knowledge, this is the first study on textile wastewater regarding a wide range of NPEOs (from NP_0EO to $NP_{15}EO$) and their elimination across two typical textile wastewater treatment processes in Vietnam. The insights of NPEO and NP removal capacity, as well as influencing factors in the Vietnamese textile industry, are crucial to improving the current wastewater treatment situation.

Our findings revealed diverse trends in the distribution of $NP_{1-15}EO$ in wastewater of the two selected factories. The highest concentrations centered at $NP_{9-11}EO$ for F1, which was specialized for cotton products. On the contrary, metabolites including $NP_{1-2}EO$ and NP dominated in wastewater from the synthetic fibers process, as F2. The trends showed insignificant changes from the influents to the primary effluents.

The primary coagulation of F1 functioned well in eliminating NP and $NP_{1-2}EO$, while a significant number of medium-long oligomers—as much as 94%—was transformed into metabolites. Whereas, a lower removal efficacy was obtained in F2, probably due to a continuing decomposition of more complex oligomers containing more than 15 ethoxylate groups.

ASPs shown to be crucial phases in reducing NPEOs and NP in textile wastewater, taking advantage of long HRTs (21 and 16 h), low organic loadings (0.1 and 0.2 gCOD/gMLVSS.day), and extended SRTs (61 and 66 days), which sustained the MLSS concentration of greater than 2000 mg/L for F1 and F2, respectively. Treatment conditions such as DO, temperature, organic loading, and SRT were likely to be suitable for nitrification. However, the pH of 7.8 and the HRT of 16 h in F2 seemed not to be optimal for a complete nitrifying process.

Concerning the disadvantage of a prolonged HRT linked with a footprint shortage, future studies need to focus on optimizing nonylphenolic removal from the conditions of Vietnam, taking into account the relationship between HRTs and alternatives, such as MBR and attached-growth ASPs. Although satisfactory elimination of NPEOs and NP has been achieved, further research may extend the investigation for nonylphenol ethoxy carboxylates (NPECs), especially $NP_{1-2}EC$, since they can contribute to the NP concentration in the aquatic environment. Last but not least, additional investigations on both aqueous and solid phases are suggested, to gain more insight into the fate of NPEOs and NP during textile wastewater treatment processes.

Author Contributions: Hien Ho conceived the idea of the study, carried out data collection and participated in the analysis team. Tsunemi Watanabe supervised data collection and data analysis. Both authors drafted the manuscript and approved the final manuscript.

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

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