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RESEARCH ARTICLE

Nonylphenol ethoxylate degradation in detergents during shelf time, a new exposure pathway, and a perspective on their substitution

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Abstract

Detergents are highly produced pollutants with environmental problems like foam generation and toxic effects in biota. Nonylphenol ethoxylates (NPEs) are efficient, economical, and versatile surfactants, used in detergents for more than 40 years due to their detergency capacity. In the environment, NPE biodegrades into the metabolite nonylphenol (NP), classified as an endocrine disruptor. The identification and quantification of 4-NP in a designed detergent and 30 commercially available detergents were performed to prove the degradation of NPE into 4-NP during storage time. This investigation introduces the first evidence of NPE degradation during storage in commercially available detergents, demonstrating a novel exposure pathway in humans that has not been explored before, representing potential human health risks. Therefore, simple, easy, low-cost, and available approaches to remove and substitute NP is paramount. Alkyl polyglucoside (APG) was assessed as a substitute, and the feasibility of this substitution was proven according to physical and chemical properties, cleaning performance, and antimicrobial properties. NPE substitution in detergents is demonstrated as a viable strategy to minimize exposure risks in humans and the environment.

Keywords Nonylphenol ethoxylate · 4-nonylphenol · Endocrine disruptor · Dermal exposure · Alkyl polyglucoside · Sustainable detergent

Introduction

Alkylphenol ethoxylates (APEs) are non-ionic surfactants widely used in detergents, plastic additives, emulsifiers, and pesticide manufacturing (Najim and Radeef 2023). Therefore, most of APEs are removed from wastewater with conventional treatment plants (Grześkowiak et al. 2021); however, APEs are one of the most reported surfactants in wastewaters, effluents, rivers, drinking water, sediments, and soil (Belmont et al. 2006; Chen et al. 2013; Dong et al. 2015; Jie et al. 2017; Van Zijl et al. 2017). Nonylphenol ethoxylate (NPE) is an efficient, low-cost, and versatile surfactant. It has been used in detergents for more than 40 years due to its ability to form micelles in solution and detergency capacity (Araujo et al. 2018; Yu et al. 2008). In some cases, domestic and industrial wastes are discharged without previous treatment, affecting significantly water quality (Bilal et al. 2019). Therefore, NP is one of the most frequently identified organic pollutants in wastewater, rivers, drinking water, sediments, and soil (Belmont et al. 2006; Dong et al. 2015; Van Zijl et al. 2017). After its disposal, NPE

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is degraded by microorganisms or ultraviolet light in environmental conditions, transforming into nonylphenol (NP) (Cheng et al. 2017). NP and intermediate biodegradation products are known for their estrogenic properties (Bhandari et al. 2021). NP mimics the female hormone 17β-estradiol, causing adverse health effects on organisms and humans; therefore, it is classified as an endocrine disruptor (Hong et al. 2020). Previous studies have shown adverse effects on humans, like decreased sperm count, reproductive malformations, immune deficiency, and an increment in prostate, breast, ovarian, and testicular cancer. Moreover, effects on children, fetuses, and newborns include neurological effects and poor intellect development (Kim et al. 2020; Lussier et al. 2000; Prasad et al. 2023; Scaia et al. 2019; Shirdel et al. 2020; Tabassum et al. 2017), and concentrations have been found in rivers (Lalonde and Garron 2021, Pakzad Toochaei and Kazemi 2022, Zhao et al. 2024), lakes (Liu et al. 2020), sediments (Crane 2021, Komaki and Riyahi Bakhtiari 2020), and sewage (Ryu et al. 2024; Sá et al. 2022). Consequently, regulations to restrict the use of NPE have been implemented. The European Union (EU), through Directive 2003/53/EC, establishes that NPE "…may not be placed on the market or used as a substance or constituent of preparations in concentrations equal or higher than 0,1% by mass…" (Directive 2003), and Directive 775/2004(02/2076) prohibits the use of NPE in pesticides formulations (Commission Regulation 2004). Also, the Environmental Protection Agency (EPA) from the United States added NPE to the Toxics Release Inventory (TRI) list. Also, the NPE meets the toxicity listing criteria of the EPA's Emergency Planning and Community Right-to-know Act (EPCRA) Section. 313(d)(2)(C), indicating that the NPE is highly toxic to aquatic organisms (EPA 2005). Likewise, Canada enlists NP as a toxic substance in the Canadian Environmental Protection Act (Act and Pentafluoroethane 1999), and the Rotterdam Convention classifies it as a severely restricted compound. However, there are still countries without regulations, like Latin American countries (Vargas-Berrones et al. 2020a). Due to the occurrence and potential exposure risks, the substitution of NPE in detergents as a development of sustainable technology is proposed. Usually, NPE is substituted with alcohol ethoxylates, less effective surfactants with side effects like eye irritation, skin irritation and sensitization, genetic toxicity, oral and dermal acute toxicity, and oral repeat-dose toxicity (Li et al. 2018a, Li et al. 2021, Maurer and Kung 2020, Yu et al. 2008). Therefore, natural surfactants from renewable sources like fatty acid esters, amino acid amides, lecithin, and various sugar types have been introduced. Sugar-based surfactants are of primary interest because of their cleaning performance, consumers' health, and environmental compatibility (Deleu and Paquot 2004, Uzwatania et al. 2017, Holmberg 2001). Alkyl polyglucosides (APG) are sugar-based surfactants with synergic effects, low irritation potential, full biodegradation, and nontoxic (Wasilewski et al. 2016; Yu et al. 2008). More evidence of sustainable alternatives is needed, mainly in developing countries, where higher concentrations in water bodies and health effects of exposure have been found (Vargas-Berrones et al. 2020a). There is a major concern about the future and transport of NP through the environment and humans because of its continuous detection and identification in water sources. In addition, due to the lack of regulations in developing countries, most commercially available detergents have NPE in their formula. This study suggests NPE degradation during the shelf time of detergents, introducing a new exposure pathway with associated potential health risks. Thus, the aim of this study is to investigate the NPEs' degradation of detergents during shelf time and develop a detergent using a natural surfactant (APG) to assess its possible substitution as a strategy to minimize exposure risks in humans and the environment (Fig. 1).

Materials and methods

Preparation of detergents

Two liquid detergents using NPE and APG, respectively (13%), were designed following the recommendations of ISO 6330:2012. Ten percent of fatty alcohol was added as a complementary surfactant. A builder (5%) was used for water softening and as a preservative to improve detergent performance (Lim et al. 2019). Enzymes (0.1%) were included to reduce the use of large quantities of chemicals and to improve cleaning capacity (Hasan et al. 2010). Finally, sodium hydroxide (0.9%) was used as an alkali to maintain pH and favor the cleaning processes (Chiplunkar et al. 2017). Solutions were stirred for 30 min at room temperature. On the other hand, thirty commercially available detergents were purchased in Mexico for further analysis.

Degradability in detergents

The biological oxygen demand (BOD) represents the oxygen consumed by the bacteria and microorganisms while the organic matter is decomposed under aerobic conditions. Therefore, NPE degradability in the designed detergents is determined by measuring the consumed oxygen following the International Standard of the Organization for Economic Cooperation and Development (OECD) 301D Closed Bottled Test. BOD measures were taken on days 0, 7, 14, 21, and 28 using a dissolved oxygen meter (MeterTo, model JPB-607 A). The flasks were previously prepared with 4% of the designed detergents and 1% of inoculum provided by the Electrolytic Zinc Refinery, Grupo México. Duplicate control blanks were prepared

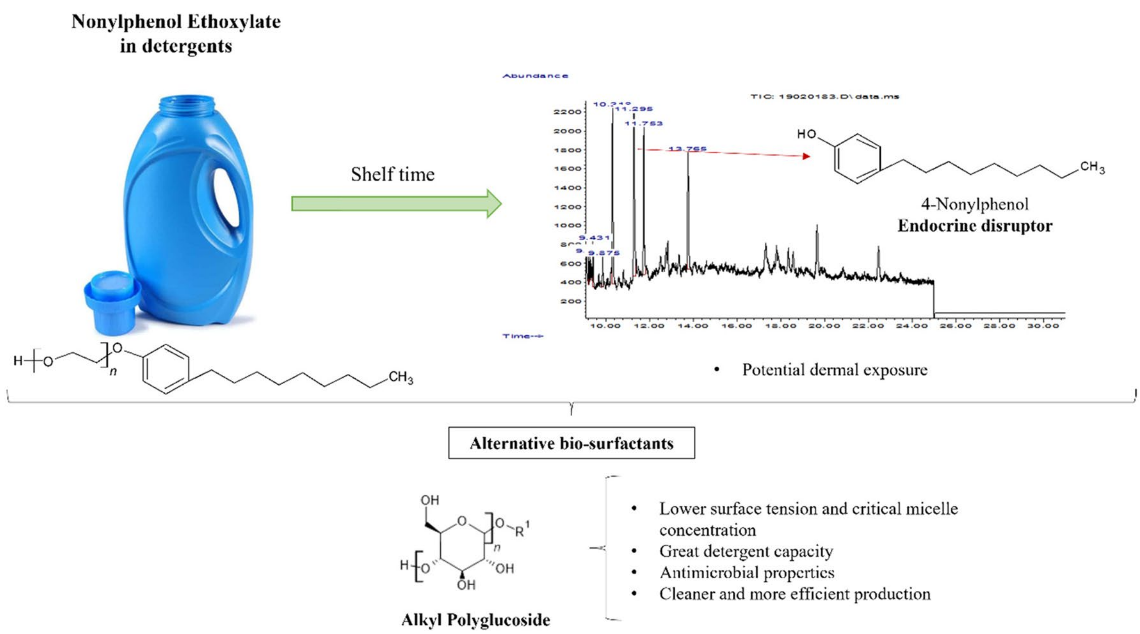

Fig. 1 Conceptualization of NP exposure from detergents and a perspective on their substitution

to eliminate any potential bias or matrix effect. The chemical oxygen demand (COD) was calculated following the closed reflux method. The experiment was performed in triplicate to assure reproducibility; averages are reported. NPE degradation was confirmed by identifying and quantifying 4-NP with gas chromatography–mass spectrometry (GC–MS) following the method described previously by our research team. The linear range ($r^2 = 0.99$) of the calibration curve was from 0.5 to 50 μg L^{-1} and was determined from the average of five curves obtained for 3 days. Repeatability and reproducibility of the method were determined by evaluating different concentrations in triplicate on the same day and in duplicate on five different days. Blanks and calibration check samples were analyzed as quality and assurance control (QA/QC). The obtained values for detection (LOD) and quantification (LOQ) were 0.01 μg L^{-1} and 0.15 μg L^{-1}, respectively. Complete data can be consulted in the paper by our research group (Vargas-Berrones et al. 2020b). Also, directed monitoring of 30 samples of commercially available detergents (manufactured and sold in Mexico) was performed to detect NP after shelf time.

Detergent evaluation

The evaluation of the designed detergents was divided into three: physicochemical properties, detergency tests, and antibacterial activity assessment. Every determination was performed in triplicate to assure reproducibility; averages are reported.

Physicochemical properties

The following physicochemical properties were determined: surface tension, critical micellar concentration (CMC), and viscosity. Surface tension was determined following the Spanish Standard UNE 55–501-90 (Española 1990) using a test temperature of 20 °C and a tensiometer with a 4 cm diameter platinum foil (MGW-LAUDA Hucoa-Erlôss). Accordingly, CMC was calculated by measuring the surface tension of a concentration series (5%, 20%, 50%, and 100%); the surface tension changes strongly just before reaching the CMC. Viscosity was determined following the ASTM D-445–06 (Standard ASTM 2006) using a calibrated viscosimeter, a bath at a controlled temperature (± 0.02 °C), a glass thermometer accurately calibrated (± 0.02 °C), and a chronometer that allows lectures with 0.1 s of discrimination.

Detergency tests

Regarding the detergency tests, the foaming power was determined following the Spanish Standard UNE-EN 14371 (UNE Normalización Española 2005). A graduated test tube and a gear pump with an outlet flow of 0 L h^{-1} up to 280 L h^{-1}, which allows flow adjustment of 200 L h^{-1}, were used. The height of the foam produced in a 1 g L^{-1} solution at 50 ± 2 °C was determined at 30 s, 3 min, and 5 min. The wetting power by immersion was performed according to ISO 8022:1990 using a cotton woven disc (DIN 53901). The cotton discs were placed in solutions of a concentration series (2, 3, 4, and 5 g L^{-1}) to establish the wetting time (time that it takes to immerse). The cleaning performance (decontamination rate) was calculated following the ISO 6330:2012 using a front-loading horizontal rotating drum-type machine (Type A) and polyester fabric squares of 20 ± 4 cm by 20 ± 4 cm with a mass per area of 310 ± 20 g cm^{-3}, and individual weight of 50 ± 5 g (purchased from the Center for Test Materials B.V.). The decontamination rate was determined by the reflectance lecture with a photocolorimeter (GretagMacbeth-Color i5) before and after washing the test cloth materials with the test detergents (8 g L^{-1}).

Antibacterial activity assessment

The minimum inhibitory concentration (MIC) was determined to establish the antibacterial activity. MIC was measured following the turbidimetry methodology (ISO 20776–1) using sterile phosphate-buffered saline (PBS) as a liquid culture medium. Four different strains were evaluated to have a wide range of test organisms: (i) *Staphylococcus epidermis*, (ii) *Escherichia coli*, (iii) *Candida sp*., and (iv) *Pseudomonas aeruginosa* grown in agar coliform at 37 °C for 24 h. Results were obtained with a microplate reader Infinite M200, Tecan Software Magellan®.

Results and discussion

NPE degradability in-storage detergents

Table 1 shows the dissolved oxygen values and the chemical oxygen demand from the designed detergents stored at normal shelf conditions and demonstrates the decomposition of organic matter. Previous studies have proved that APE metabolites degrade more easily under aerobic conditions than under anaerobic conditions (Arora et al. 2023, Haq and Raj 2019, Khalil and Liu 2021). NPE initial biodegradability occurs rapidly by losing surfactant properties (Quiroga et al. 2019). Chemical and environmental factors like agitation, temperature, and the addition of yeast extracts affect and influence NPE biodegradation. Also, previous studies have demonstrated that aluminum sulfate, hydrogen peroxide, Pb, Cd, Cu, Zn, phthalic esters, and NaCl inhibit NP aerobic degradation (Mao et al. 2012). NP half-live in sewage, sludge, and sediments under aerobic conditions has been established from 1.1 to 99 days (Chang et al. 2005, 2008; Yuan et al. 2004), while under anaerobic conditions is from 23.9 to 69.3 (Chang et al. 2005). Aerobic degradation starts with the shortening of the EO chain and is completed with the diethoxylation (AP formation). The EO chain oxidation forms alkyl phenoxy ethoxy acetic acid and alkyl phenoxy acetic acid (Fig. 2). The three more reported intermediaries are APs (NP and OP), short-chain APEs with EO units (1 to 4), mainly APE 2, and a series of ether carboxylates including alkyl phenoxy acetic acid and alkylphenoxy ethoxy acetic acid (Bhandari et al. 2021; Ying et al. 2002). Previous studies have shown that even if 4-n-NP is persistent in the environment, it is readily biodegradable during activated sludge processes (Stasinakis et al. 2008). For example, 4-n-NP sorption on suspended solids was achieved with higher values of sorption coefficient at a sludge retention time (SRT) of 3 days, and the biodegradation was enhanced at SRT of 20 days (Stasinakis et al. 2010). Moreover, different methods like adsorption and degradation with fungi, bacteria, microalgae, and yeasts have been tested to remove NP. These organisms can use NP as a carbon and energy source for their growth and development (Bhandari et al. 2021). For sustainable bioremediation, a microbial approach is established as the most efficient, low-cost, and sustainable because physicochemical techniques present several drawbacks like high cost, toxic by-products, and the use of large quantities of solvents (Bilal et al. 2019; Różalska et al. 2015). However, conventional water treatment plants are ineffective for NP degradation (Lara-Moreno et al. 2022).

The degradation of NPE in the designed detergents under controlled conditions is proven by identifying and quantifying 4-NP using GC–MS. Concentrations of 4-NP in the test detergents increased throughout time, reaching 5 μg L^{-1} after 28 days (peak at 11.2 min) (Fig. 3); the other peaks observed may correspond to NP isomers (Lu and Gan 2014). Also, Table 2 shows that 36% of the commercially available detergents presented significant concentrations of 4-NP (from 0.87 to 134.80 μg L^{-1}), suggesting high levels of this pollutant when released into the environment. Therefore,

Table 1 Dissolved oxygen after *n* days in detergents stored at normal conditions

Sample	mg O_2 L^{-1} after *n* days 0	5	7	14	21	28	COD
DNPE	7.30	4.50	3.90	3.40	0.50	0.50	42.32
DAPG	7.00	3.80	3.50	3.50	0.50	0.50	20.84

[*] *DNPE* detergent with nonylphenol ethoxylate, *DAPG* detergent with alkyl polyglucoside, *COD* chemical oxygen demand

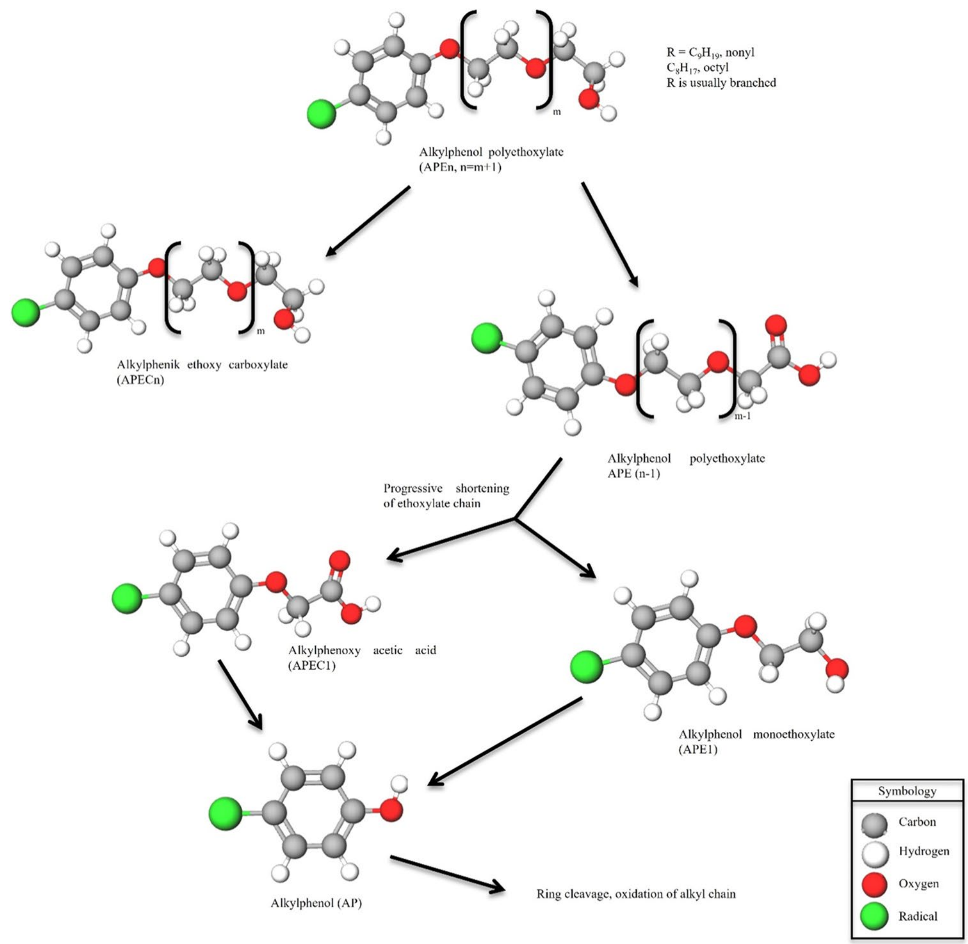

Fig. 2 Estimated aerobic NPE degradation route (Ying et al. 2002). Ying G-G, Williams B, Kookana R (2002): Environmental fate of alkylphenols and alkylphenol ethoxylates—a review. Environment International 28, 215–226

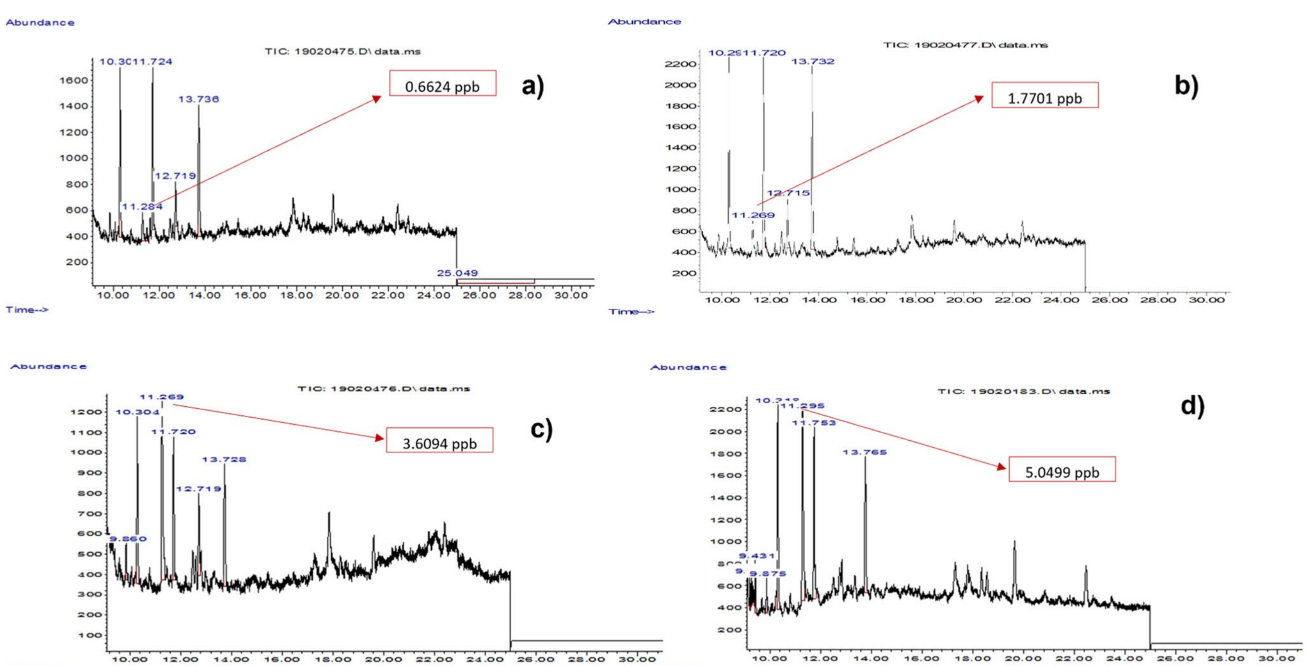

Fig. 3 Formation of 4-NP during OECD 301 D experiment in test detergents stored at normal conditions after **a** 7 days; **b** 14 days; **c** 21 days; **d** 28 days

Table 2 Concentrations of 4-NP in commercial detergents

Sample number	Surfactant type	Concentration, μg L^{-1}
1	Non-ionic	134.80
2	Anionic and non-ionic	0.8739
3	Non-ionic and anionic	< LOD
4	Anionic and amphoteric	106.99
5	Anionic and amphoteric	30.50
6	Non-ionic	12.85
7	Anionic	37.77
8	Non-ionic and anionic	24.86
9	Anionic	< LOD
10	Anionic	< LOD
11	Anionic	< LOD
12	Anionic	< LOD
13	Anionic	< LOD
14	Anionic	< LOD
15	Anionic	< LOD
16	Anionic	< LOD
17	Anionic	< LOD
18	Non-ionic	114.87
19	Anionic and non-ionic	79.65
20	Anionic, non-ionic, and amphoteric	12.16
21	Anionic	< LOD
22	Anionic	< LOD
23	Anionic	< LOD
24	Anionic	< LOD
25	Anionic	< LOD
26	Anionic and non-ionic	< LOD
27	Anionic	< LOD
28	Anionic and non-ionic	4.75
29	Anionic	< LOD
30	Anionic	< LOD

**LOD* limit of detection, *LOQ* limit of quantification

concentrations of 4-NP above the Water Framework Directive of the European Union (2 μg L^{-1}) (European Union 2013) and the EPA recommendation (6.6 μg L^{-1} for surface water and 1.7 μg L^{-1} for saltwater) (EPA 2005) are expected. It is critical to acknowledge that concentrations of 4-NP were found in the commercially available detergents that include non-ionic surfactants in their ingredients list, while 4-NP was not found in detergents with anionic and amphoteric surfactants as ingredients. Even when the labeling does not openly include NPE as an ingredient, NPE has been recognized as the predominant APE and the most used for over five decades due to their efficiency and cost-effectiveness in detergent formulation (Jawaid et al. 2024). Currently, detergents are being designed using mostly anionic surfactants, although there is no regulatory framework on the use of NPE in detergents in developing countries (i.e., Mexico). This behavior could be due to the increasing trend of environmental and social responsibility to minimize endocrine disruptor occurrence. Therefore, cost-effective, high-performance, and environmentally friendly alternative development are paramount for the prompt implementation by industries.

The following limitations have been considered when interpreting the results of this study: (i) the concentration of 4-NP increases over time until day 28 as the standard establishes; thus, it is crucial to analyze if there is still an increment in the 4-NP concentration throughout time considering that shelf life of a detergent is of 12 to 24 months (Kübelbeck et al. 2018; Ramnani et al. 2005; Riedler et al. 2003); (ii) 4-NP was the only isomer considered in this study because it is the most toxic and reported (Calafat et al. 2005; Lara-Moreno et al. 2022); however, NP is a mixture of approximately 20 isomers with intermediaries structures so, the identification of other isomers is needed. Also, each isomer presents a different degradation range, and only a few persist after 45 days (Li et al. 2018b; Metcalfe et al. 2022; Zhang et al. 2023); (iii) NPE biodegradability in detergents during shelf time was proved applying the OECD protocol; however, NPE degradation was not calculated. Future research is suggested to establish biodegradability percentages according to common commercial detergent formulations associated to environmental and health implications.

NP has been widely reported in water samples (Babay et al. 2008; Belmont et al. 2006; Bina et al. 2018; Félix–Cañedo et al. 2013; Jardim et al. 2012; Martín et al. 2014; Martinez and Peñuela 2013; Montagner 2011; Vargas-Berrones et al. 2020a); nonetheless, no information about NP in detergents was found when the literature review was carried out; only an investigation by Cheng and Ding (2002) reported NPEOs in detergents with concentrations from 0.2 to 21% in 41% of 90 household detergents analyzed. The presence of NP in detergents may represent potential risk effects for human health, mainly through dermal contact. Previous studies have demonstrated NP dermal absorption in humans and animals with effects like skin irritation (Talmage 2020); however, exposure through detergents has not been studied before because, to date, exposure through food and water intake has been the main and only route analyzed (Chang et al. 2019; Osimitz et al. 2015; Raecker et al. 2011).

Detergent performance

Some of the following results have been previously shared in a preprint (Bernal-Jácome et al. 2023). The detergent with APG (DAPG) showed better results regarding surface tension and CMC. The DAPG presented lower surface tension (25% lower than the detergent with NPE); therefore, the CMC of DAPG was achieved using only 20% of the surfactant in the solution, while the detergent with NPE (DNPE) presented this point at 50%. Previous studies have demonstrated that lower surface tension values benefit soil removal (Mousavi and Khodadoost 2019). Also, lower CMC implies that less detergent is needed to achieve the maximum micelle formation, improving cleaning processes and minimizing economic and environmental costs (Singh et al. 2018). These results are similar to previous studies where APG significantly reduces surface tension and CMC in detergents, allowing better penetration of the detergent in textiles and achieving better soil removal (Karthick et al. 2018; Pukale et al. 2017). On the other hand, the viscosity of the detergent with NPE (DNPE) and with APG is 445.74 and 20.38 mpoise, respectively, meaning that DAPG presents a viscosity 22 times lower than DNPE. Detergents with high viscosity hinder production processes and kinetically limit water solubility (Calero et al. 2017); hence, DAPG offers a cleaner and more efficient production.

DAPG presents higher foam formation (550 ml of height in 5 min) than DNPE (153 ml of height in 5 min). This behavior is expected because compounds with lower surface tension usually favor foam generation due to the energy decrease of the molecular interface (Chiplunkar et al. 2017). However, previous studies have shown that APG foam height decreases as the alkyl chain length increases, and increases depending on the concentration in the solution. Therefore, APG could be selected according to the desired foam stability (Chiplunkar et al. 2017; El-Sukkary et al. 2008). In addition, natural antifoaming agents may be used in the production processes to control foam generation (Schilling and Zessner 2011). Unstable and poor foam formation is desirable because foam blocks the oxygen-mass transformation from air to water, affecting organisms (Effendi et al. 2017).

The wetting time of both detergents decreased as the concentration increased. The wetting power of DNPE at 5 g L^{-1} was 37 s, and the DAPG was 163 s. These values represent 4.4 times higher wetting power for the detergent with NPE related to soil removal efficiency (Moulay et al. 2005), assessed through the decontamination rate test; results are shown in Table 3. Several stains, like grass, rice starch, pigment, oil, wine, cocoa, and others, present similar decontamination rates with both detergents, though each detergent works better in different stains; however, DAPG presents a better decontamination rate in 64% of the samples tested. APG's capability as a surfactant was confirmed; nonetheless, proportions in formulations need to be considered and assessed accordingly to their use. These results are similar to previous studies where APG in detergent formulations shows a better decontamination rate in different fabric types (Karthick et al. 2018; Pukale et al. 2017). Other natural surfactant assessment is recommended; for example, methyl ester sulfonate (MES) has proven stable in hard water and meets cleaning standards (Tai et al. 2018).

Table 4 presents MIC results for each detergent considering the four strains evaluated. Values show that microbial growth has been inhibited since time cero in the four strains studied. However, it is crucial to consider lower concentrations (< 10%) to establish the minimum concentrations in which the detergent presents antimicrobial properties. These results are similar to others previously reported in which APG demonstrates appropriate antimicrobial properties

Table 3 Results of detergency test for detergents with NPE and APG

No	Product code	Description	DNPE Percentage of decontamination rate	DAPG Percentage of decontamination rate
1	C-BC-01	Tea for medium/high temperatures	4.09	0.24
2	C-BC-02	Coffee	3.23	0.81
3	C-BC-03	Tea for low temperatures	4.98	1.19
4	C-S-01	Blood (aged)	8.05	8.43
5	C-S-02	Cocoa	4.92	6.68
6	C-S-03	Wine (aged)	2.74	3.40
7	C-S-08	Grass	21.23	19.69
8	C-S-12	Black currant juice	16.33	23.88
9	C-S-15	Bill(blue)berry juice	14.79	24.74
10	C-S-28	Rice starch, colored	17.28	17.78
11	C-S-44	Chocolate drink, pure	1.87	6.49
12	C-S-103	Wine, not aged	0.94	11.46
13	C-05	Blood, milk, ink	0.34	0.50
14	C-12	Pigment, oil, low milk content	9.98	6.32

Table 4 Antimicrobial properties results of detergents with NPE and APG

			DNP			DAPG		
	Time (seg)	Strain response	10%	30%	50%	10%	30%	50%
S. epidermis	0	0.121	0.082	0.098	0.093	0.115	0.14	0.178
	7167	0.122	0.083	0.131	0.103	0.102	0.136	0.168
	14,335	0.127	0.085	0.129	0.103	0.103	0.136	0.167
	21,503	0.286	0.093	0.142	0.115	0.105	0.137	0.167
	28,671	0.717	0.093	0.144	0.114	0.105	0.137	0.167
	36,262	1.282	0.092	0.145	0.114	0.106	0.137	0.167
E. coli	0	0.126	0.088	0.083	0.108	0.106	0.135	0.192
	7167	0.127	0.097	0.091	0.109	0.093	0.13	0.182
	14,335	0.218	0.104	0.09	0.108	0.094	0.131	0.181
	21,503	0.666	0.11	0.091	0.107	0.095	0.132	0.182
	28,671	1.004	0.119	0.09	0.107	0.096	0.134	0.183
	36,262	1.208	0.115	0.091	0.107	0.098	0.135	0.183
Cándida	0	0.150	0.091	0.094	0.093	0.115	0.142	0.193
	7167	0.150	0.09	0.109	0.122	0.101	0.137	0.184
	14,335	0.151	0.094	0.112	0.121	0.103	0.138	0.184
	21,503	0.155	0.095	0.116	0.114	0.106	0.14	0.185
	28,671	0.255	0.096	0.116	0.114	0.109	0.142	0.186
	36,262	0.725	0.096	0.117	0.114	0.112	0.143	0.186
Pseudomonas aeruginosa	0	0.119	0.119	0.13	0.091	0.113	0.14	0.188
	7167	0.121	0.124	0.149	0.095	0.1	0.135	0.173
	14,335	0.254	0.123	0.151	0.096	0.104	0.139	0.174
	21,503	0.873	0.126	0.15	0.095	0.105	0.141	0.174
	28,671	1.125	0.128	0.149	0.093	0.106	0.142	0.175
	36,262	1.266	0.127	0.15	0.094	0.107	0.142	0.176

without toxicity problems (Boonprakobsak and Rachtanapun 2022, Smułek et al. 2021).

To our knowledge, there are no studies regarding 4-NP identification and quantification in detergents and the formulations of APG detergent as a direct substitute for NPE. This study introduces the first evidence that NPE degrades under normal storage conditions in commercial detergents. Also, the first comparison between NPE and APG as a surfactant in detergents is presented. The feasibility of APG as a sustainable substitute for an emerging pollutant and prohibited compound in several countries (NPE) was demonstrated. The quantity of non-ionic surfactants used in detergent industries is not explicit; however, previous studies have found concentrations of NPE in detergents from 0.2 to 21% (Huang et al. 2014). The NPE market is estimated to be worth $ 928.89 million by 2027, registering a growth rate of 7.3% from 2020 to 2027 (Research DBM 2020), and the growing demand for NPE in the detergent, fertilizer, and paint industries will continue to increase. Global cleaning product market growth will achieve a rate of $ 59.5 billion by 2025 with an annual growth rate of 4.8% (Markets and Markets 2023). Thus, the environmental risks associated are hard to predict; the countries without regulations (Latin American countries) are the most likely to be affected. The chemical industry represents one of the most important economic sectors worldwide, and the introduction of new technologies is constantly increasing (Pearce and Tombs 2019). Accelerated industrialization and economic development have caused exponential population and urbanization growth (Chen 2018; Zafra et al. 2015). This situation has led to higher consumption of natural resources and environmental pollution (Li &Yu 2011; Nations 2015; Zhang et al. 2015), for example, detergent consumption. In 2017, more than sixty billion dollars were used for detergent production (Giagnorio et al. 2017), and the surfactant market represented thirty-three million dollars in 2016 (Karray et al. 2016). Even though these potential risks and adverse effects in the industry are well known, it is still a challenge to persuade the industries to replace conventional surfactants due to the costs that this may represent (up to 50 times higher than synthetic surfactants) (Farias et al. 2021).

Conclusions

Water quality and availability have deteriorated due to anthropogenic activities, population growth, industrialization, and urbanization. Among the pollutants, detergents represent a critical source of pollution with associated health and environmental risks. Therefore, efforts in investigation and strategies to obtain a responsible approach to the use and handling of NP and its ethoxylate in detergents are required to promote consciousness of the impact of NP as a pollutant. This study proved the degradation of NPE and formation of 4-NP in detergents during shelf time, presenting an exposure pathway that has not been explored. Concentrations of 4-NP up to 5 μg L^{-1} were found after 28 days in the designed detergents, and 36% of the commercially available detergents tested presented concentrations above the European Union and the EPA regulatory framework. Thus, a detergent was developed using APG to substitute NPE. The detergent with APG presented lower surface tension and CMC, showing a better penetration of the detergent in the fabrics tested. Also, better detergent capacity in 64% of the stains tested and effective inhibition of bacterial growth were demonstrated. Overall, results show that APG is an adequate substitute for NPE. This substitution allows cleaner and more efficient production, resulting in lower economic and environmental costs. Countries without regulation in this regard are reluctant due to the monetary costs that the substitution of NPE represents; however, the environmental cost associated has not been balanced with the ecological and human risks. The green technology developed proved the multiple benefits that natural surfactants offer in terms of availability, performance, and environmental health. It is paramount to promote awareness of the negative impacts of endocrine disrupters. Also, environmental regulations and continuous monitoring are recommended to address the contamination in ecological receptors that could develop into significant effects on human health. The results of this study provide new and valuable information to establish a basis for further studies and for industries to improve and implement greener detergent production.

Author contribution All persons who meet authorship criteria are listed as authors, and all authors certify that they have participated sufficiently in the work to take public responsibility for the content, including participation in the concept, design, analysis, writing, or revision of the manuscript. Sampling, analytical methods, and manuscript writing by Luis Armando Bernal-Jácome; analytical methods by Juan Manuel Izar-Landeta and Luis Fernando González-Salazar; conceptualization and funding by Rogelio Flores-Ramírez; conceptualization, analytical methods, writing—reviewing, and manuscript editing by Karla Ximena Vargas-Berrones.

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Data availability Data available on request from the authors.

Declarations

Ethics approval The paper reflects the authors' own research and analysis in a truthful and complete manner.

Consent to participate and for publication All persons who meet authorship criteria are listed as authors, and all authors consent to participate and publish in the work to take public responsibility for the content.

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