

Alkylphenols and bisphenol A in the aquatic environment and their leaching from plastic products – a review

Alina Pohl*, Michał Bodzek

Institute of Environmental Engineering, Polish Academy of Sciences, Poland 41-819 Zabrze, M. Skłodowskiej-Curie 34 Street, emails: alina.pohl@ipispan.edu.pl (A. Pohl), michal.bodzek@ipispan.edu.pl (M. Bodzek)

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ABSTRACT

Environmental pollution with plastics and plastic debris is one of the major issues in the world. In addition to the problem of microplastics formed from these wastes, additives leached from plastics are also an important aspect. This review paper pays particular attention to three phenolic compounds - bisphenol A (BPA), 4-n-nonylphenol, and 4-tert-octylphenol (4tOP) – used, among others, in the plastics industry. Numerous studies have shown that bisphenol A, 4-nonylphenol, and 4-tert-octylphenol are leached from plastic products, which is also an important source of their release into aquatic environments. The leaching process is influenced by external factors, including temperature, mixing, exposure to ultraviolet light, chemical or mechanical abrasion, and aging and brittleness of plastics. A significant amount of leaching tests are concerned on the migration of BPA from polycarbonate bottles into water. There is less comprehensive information on three phenolic compounds leached from different types of polymers. Moreover, despite the availability of several economic and ecological techniques for their removal from water and wastewater (including coagulation, adsorption, membrane processes, advanced oxidation), significant amounts are noted in some of the aquatic environments, which may cause harmful endocrine effects in organisms living in these ecosystems.

Keywords: Alkylphenols; Nonylphenols; Octylphenols; Bisphenol A; Aquatic environment; Leaching from plastic; Plastic additives; Remediation

1. Introduction

The most current scientific topics of recent years concern, among others, environmental pollution with microplastics, per- and polyfluoroalkyl substances, glyphosate, bisphenols, or substances disrupting the functioning of the endocrine system [1]. The last one, endocrine-disrupting chemicals (EDCs) can disrupt the endocrine system and thus cause harmful effects in both humans and wildlife [2]. The European Chemicals Agency (ECHA) has compiled a list of evaluations of endocrine disruptors containing 110 substances [3].

According to LaFleur and Schug [4] known and potential endocrine-disrupting substances in the environment come from different sources, for example, pharmaceuticals, personal care products, synthetic polymer compounds (xenoestrogens), pesticides, naturally occurring compounds in plants and animals, and inorganic and organometallic compounds. As the plastics industry is a dynamically developing branch of the economy, and the resulting plastic waste has become the fastest-growing stream of municipal waste in recent years, while polluting the environment on a huge scale [5,6], this review focuses on selected chemical additives used in the plastic industry that may act as estrogen mimics. Some plastic additives like plasticizers, stabilizers, or antioxidants exhibit estrogenic activity. Many industrial compounds that have shown this activity contain phenolic groups and are found in water samples [4]. Such

^{*} Corresponding author.

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compounds include alkylphenols among others (4-n-nonylphenol (4nNP), 4-tert-octylphenol (4tOP)) and bisphenol A (BPA) [7–10]. The use of some of them is limited by restrictions introduced by the European Commission, the United States Environmental Protection Agency, or health organizations such as the World Health Organization. Studies on the occurrence and fate of these compounds disrupting the functioning of the endocrine system of living organisms, derived from manufactured materials, is an important aspect of environmental protection.

2. General characteristics of alkylphenols and bisphenol A

Alkylphenols and their ethoxylates (APEOs) belong to the group of phenolic compounds. Alkylphenols are phenol-derivative compounds, in which one or more hydrogen atoms in the aromatic ring have been replaced by an alkyl group [11]. APEOs are non-ionic surfactants, consisting of branched-chain alkylphenols, which have been reacted with ethylene oxide, producing an ethoxylate chain [12]. Alkylphenols are used to produce alkylphenol ethoxylates [13]. The substances of the greatest practical importance among the discussed groups are nonylphenols (NPs), nonylphenol ethoxylates, octylphenols (OPs), and octylphenol ethoxylates [14]. Annual production of nonylphenols and octylphenols expressed in tonnes of the substance produced and/or imported in the European Economic Area for consumer use is 10,000–100,000 for both nonylphenols and octylphenols [15]. According to the research and markets website [16], the market for alkylphenol ethoxylates is expected to record variable growth trends in the long term. APEOs are unstable in the environment and undergo degradation processes, yielding metabolites that are generally more stable, and thus more persistent [17]. As a result of the decomposition of alkylphenol ethoxylates in the environment, NPs and OPs are formed, which are more toxic than the parent compound and can easily accumulate in biological tissues due to their lipophilicity [18,19]. It is estimated that more than half of the 300,000 tonnes of APEOs produced annually worldwide end up in the aquatic environment after wastewater treatment in the form of shortchain alkylphenol polyethoxylates, alkylphenol carboxylic acids and alkylphenols [2,19].

Nonylphenols, one of the most commercially important groups of alkylphenols, are a group of organic compounds consisting of phenol linked to a 9-carbon linear or branched alkyl chain, which can be connected in different positions (ortho, meta, para). NP is not a single chemical structure, but a mixture of highly branched NPs [20]. Most of the literature data refer to linear (otherwise normal) nonylphenol, often para-substituted (i.e., 4-n-nonylphenol) [21]. 4-n-nonylphenol (4nNP) is a hydrophobic compound with a molar mass of 220.3 g/mol and an octanol-water partition coefficients (log K_{ow}) value of 5.76 [22]. It is poorly soluble in water and preferentially associated with suspended solids [23]. It is persistent in the aquatic environment, has a moderate bio-accumulative potential, and is extremely toxic to aquatic organisms [17].

The second group of the most important alkylphenols, that is, octylphenols, are derivatives of phenol containing 8 carbon atoms in the side alkyl chain [20]. Of the octylphenols, 4-tert-octylphenol (4tOP) dominates in the literature and environmental study [2,7–10,17,19]. The molar mass of this compound is 206.3 g/mol and the log- K_{ow} value is 4,12. Like 4nNP, it is also poorly soluble in water (5.11 mg/L at 25°C) [22] and tends to accumulate in living organisms, especially in fat tissue [24,25].

In addition to alkylphenols, this review also focuses on a third compound from the endocrine disrupting group, related to the plastics industry, namely bisphenol A (2,2'-(4,4'-dihydroxydiphenyl)propane). The molar mass of BPA is 228.3 g/mol, log K_{ow} 3.32, and the water solubility is 300 mg/L (at 25°C). Compared to the discussed alkylphenols, this compound is characterized by the lowest tendency to accumulate in living organisms (the lowest log K_{ow} value) and higher solubility in water [22,26].

Alkylphenols (including 4nNP, 4tOP) and BPA are mainly used in household applications (production of surfactants) and in the plastic industry as additives [9,27]. 4-nonylphenols and 4-octylphenols are used in the production of plastics such as high-density polyethylene (HDPE), polyethylene terephthalate (PET), and polyvinyl chloride (PVC). In addition to the use in plastic manufacture, they are also employed as substrates for non-ionic surfactants and in the production of textiles, paper, and agricultural chemicals [2,27]. In turn, BPA is one of the most important monomers used worldwide in the manufacture of epoxy resins, phenol resins, polycarbonates, polyesters, and also in flame retardants. Polycarbonate is used in reusable plastics, food packages, cans (baby bottles, drinking water bottles, etc.), electrical insulations, dental sealants, thermal papers, etc. Epoxy resins are often coated in food contact surfactants, electrical coils, PVC pipes [25,28,29]. BPA is present in electronic components, construction materials, drinking bottles, food containers, and medical devices [27,30,31].

3. Leaching of micropollutants from plastic products

Through the wide use of alkylphenols, including 4nNP and 4tOP, and bisphenol A in everyday plastic products, these compounds are leaching into the environment. The release of these substances into the environment including the aquatic environment occurs during manufacturing processes and by leaching from final products [32]. Sewage, including domestic and industrial sewage as well as urban and agricultural sewage, is mentioned as one of the main point sources of these endocrine-disrupting substances entering the aquatic environment [33]. The leaching of these substances from plastic materials, where additives like BPA are often not chemically bound with polymer, also has great significance [2,34,35].

Guart et al. [35] studied the potential of leaching of additives (phthalates, NPs, BPA) from plastics used in the water bottling sector (polycarbonate (PC), HDPE, low-density polyethylene (LDPE), PET, polystyrene (PS)) using three migration tests: (1) UNE-EN ISO 177 method - plastics and adsorbent (the food stimulant) are incubated at 40°C for 10 d, (2) an ultrasonic extraction of plastics in water at different times (1 g of plastic fragments with dimensions of 0.5–1 cm² were flooded with 100 mL distilled water and incubated in an ultrasonic bath for 5, 10, and 15 min), (3) UNE-EN 13130 method - also 1 g of plastic fragments with dimensions of

0.5–1 cm² and 100 mL distilled water were incubated in a water bath at 40°C for 10 d. The first method was used as a screening tool and was insufficient to identify specific compounds. For the third method, the obtained results were 0.332 μ g/dm² of PET, 0.579 μ g/dm² of HDPE, 0.694 μ g/dm² of PC for NPs, and 3.423 μ g/dm² of PC for BPA. It was noted that the most problematic compound was BPA.

A study by Cao and Corriveau [36] examined the migration rate of BPA from polycarbonate baby water bottles and reusable bottles into the water. After heating the bottles for 6 d at 70°C, the average migration rate of BPA was from 1.84 to 4.83 ng/cm²·h.

Kubwabo et al. [37] studied the leaching of BPA from plastic baby bottles and other polycarbonate bottles into water. The test was conducted at 40°C using water and 50% ethanol as solvents for 240 h. After this time the concentration of BPA ranged from 0.11 μ g/L (water, 8 h) to 2.39 μ g/L (ethanol, 240 h). The authors reported that migration of BPA from plastic products increased with temperature and incubation time. Several other studies have also shown BPA migrations from PC bottles into water [38–42].

In turn, Tisler and Christensen [43] conducted a series of experiments using reusable plastic bottles to check the migration of plastic additives to drinking water. The bottles with 750 mL of tap water were left at room temperature, without direct sun exposure. After 24 h, the authors determined the spectrum of compounds in the tested water samples, including 4-nonylphenol.

Staniszewska et al. [34] conducted research on the leaching of BPA from polycarbonate material and the leaching of 4tOP and 4NP from recycled tire granules, both into seawater. 100 mL of seawater collected from the Baltic Sea and 72 g of plastic materials were incubated at 20°C in the dark, shaken twice a day, for 60 d. The conducted experiment confirmed the leaching of the tested compounds from selected plastics to seawater. The highest observed concentrations were 370 ng/L for BPA and 2,700 ng/L for 4tOP and 4NP, respectively.

In another study, Suhrhoff and Scholz-Böttcher [44] performed experiments with 5 g of plastic fragments (polyethylene (PE), PVC, PS, PET), and 500 mL of deionized/salt water. All samples were incubated in the dark, at room temperature, for a maximum of 78 d. The researchers observed the release of plastic additives (phthalates, bisphenol A) in all cases. The highest concentrations of leached additives in salt water were found for PE and PVC. In addition, the authors observed that shaking increased the amount of the released additives. Moreover, the relationship between salinity and UV and the release of the tested compounds didn't show a clear tendency, as this process is different for each additive.

The leaching process is highly dependent on external factors like temperature and mixing but also depends on the aging and brittleness of plastics [45]. During the aging process of plastics, the internal polymer chain breaks and the specific surface area increases, which in turn stimulates the release of additives [46]. The leaching of additives from plastics is also affected by exposure to ultraviolet light, and chemical or mechanical abrasion [47]. In turn, according to Chen et al. [48], the release of EDC is slightly affected by salinity and dissolved organic matter content in the sample.

4. Fate and occurrence

After getting into the environment, alkylphenols (like 4NP and 4tOP) and bisphenol A, are determined in its various elements, such as water (surface water, freshwater, coastal and marine ecosystems), wastewater, bottom sediment, soils, atmosphere, and biota [8–10,19,28,49–57]. The aquatic environment is the element most exposed to these phenol derivatives [2].

Wastewater and leaching from plastic products are the two main point sources of the discussed phenol compounds. Alkylphenols flow into the marine water along with rivers and water purification plant waste disposal, harbour and marina pollution, and beach surface runoff [9]. BPA and alkylphenols can get into rivers mainly with the already mentioned sewage but also come from surface runoff from fields fertilized with activated sludge from wastewater treatment plants [58]. An important source of NP in the coastal zone in the summer season can be sunscreen creams and lotions, in which alkylphenols are used as emulsifiers [9]. The concentrations of these compounds in aquatic reservoirs may vary. Exemplary concentration ranges of 4nNP (or 4NP), 4tOP, BPA in water reservoirs around the world are listed in Table 1.

In Poland, information on the occurrence and distribution of 4-n-nonylphenol, 4-tert-octylphenol and bisphenol A in the aquatic environment is concentrated mainly in its northern part (studies of the Gulf of Gdansk, Southern Baltic) [7–10,54,57].

Lilja et al. [72] conducted research on a water sample collected in Gdynia (Gulf of Gdansk, north of Poland), the determined content of NP was 63 ng/L. Staniszewska et al. [9], investigating the waters of the Gulf of Gdansk and the tributary rivers, found that the concentrations of the discussed EDCs were similar to those in other regions of Europe and amounted from <5.0 to 277.9 ng/L for BPA, from <1.0 to 834.5 ng/L for 4tOP and from <4.0 to 228.6 ng/L for 4nNP. In other studies, Beck et al. [59] also tested water collected from the Baltic coast, but in Germany, concentrations of nonylphenol and 4-tert-octylphenol ranged from 2.5 to 13.8 ng/L and from 0.11 to 0.6 ng/L, respectively. Differences in the concentration of the discussed endocrine disturbing compounds in the water column of the Gulf of Gdansk were also studied by researchers. Staniszewska and Falkowska [54] found higher concentrations of 4nNP and 4tOP in the surface layer of water compared to the subsurface layer. In the case of 4tOP, it was from 13 to 45 times higher, and in the case of 4nNP, from 1.4 to 40 times higher, which may indicate the existence of external anthropogenic sources of these pollutants. The presence of these compounds in the surface layer of water may have an adverse effect on neuston, that is, organisms whose living environment is the water surface film [54]. However, higher concentrations of hydrophobic organic pollutants (such as nonylphenols, octylphenols, polychlorinated biphenyls, polycyclic aromatic hydrocarbons) are often observed in the bottom water layer and in bottom sediments. This is due to the hydrophobic properties of these substances, and the low oxygen content in these layers creates favorable conditions for bonding with organic matter [9,73–76].

After getting into the water, due to their physical and chemical properties, 4nNP, 4tOP, BPA can be sorbed on the

Concentration range of selected endocrine-disrupting chemicals in surface water (μ g/L) around the world

Area of study	BPA	4nNP/4NP	4tOP	References
The Gulf of Gdansk, Poland	bdl-0.278	0.001-0.834	0.004-0.229	[9]
The coast of the Baltic Sea, Germany	_	0.0025-0.0138	0.00011-0.006	[59]
North Sea, German Bight	-	0.09–1.4	0.013-0.3	[50]
Surface water, Denmark	bdl-330	-	-	[30]
Rivers flowing into Lake Biwa, Tokyo, Japan	-	110-3080	bdl-90	[60]
Thermaikos Gulf, Greece	0.011-0.052	bdl	0.002-0.018	[61]
St. Lawrence River, Canada	-	bdl	bdl-0.013	[62]
The Great Lakes, Canada	-	bdl-0.92	bdl-0.47	[62]
The Pearl River Delta, South China	0.017-0.234	0.034-3.152	0.003-0.122	[63]
Xiangjiang River, China	0.006-3.079	bdl-0.013	bdl-0.156	[64]
Rivers in Romagna area, Italy	bdl-0.244	bdl-0.135	bdl	[65]
Zumbro River Watershed, USA	_	-	0.075-11	[66]
Ogun and Ibeche Rivers, Lagos, Nigeria	bdl	43.9–79.4	57.1-68.6	[67]
The Kaveri River, India	6.6–136	bdl-2200	bdl-16.3	
The Vellar River, India	2.8-6	22.4-158	-	[68]
The Thamirabarani River, India	9.8–36	bdl-1455	-	
Yellow River, China	_	0.05-0.17	-	[69]
Lake Moon, China	-	2.4-26.4	0.04-0.68	[70]
Llobregat River, Spain	-	0.02–0.53	bdl-0.88	[71]

bdl - below detection limit

bottom sediments and bioaccumulated in biota, ultimately damaging human health [7,27].

According to Koniecko et al. [7] concentrations of 4NP in sediments in the inflowing river to the Baltic Sea ranged respectively from <LOQ to 4.93 ng/g·d.w. (d.w. - dry weight) (Gizdepka River), <LOQ-2.21 ng/g·d.w. (Kacza River), <LOQ-4.44 ng/g·d.w. (Vistula Swibno) and for 4tOP, respectively 0.55-47.12 ng/g·d.w. (Gizdepka River), <LOQ-8.51 ng/g·d.w. (Kacza River), <LOQ-23.43 ng/g·d.w. (Vistula Swibno). For samples from the coastal station of the Gulf of Gdansk maximum concentrations of 4NP and 4tOP in sediments were respectively 13.56 and 48.88 ng/g·d.w. In the case of an open water station, it was 249.08 ng/g·d.w. for 4NP and 20.47 ng/g·d.w. for 4tOP. The authors noted that the concentrations in bottom sediments were comparable to other European coastal regions, but much lower than the concentrations of these compounds in marine sediments from Southeast Asia. Lubecki and Kowalewska [77] studied, among others, BPA concentrations in the bottom sediments of the Gulf of Gdansk and the Szczecin Lagoon, the maximum concentrations were 32.0 ng/g·d.w. for costal samples and 10.7 ng/g·d.w. for samples from the lagoon. In other studies, on bottom sediments from the southern Baltic carried out by Staniszewska et al. [57] the highest mean concentration of BPA was 11.26 ng/g·d.w. for sediments of river estuaries into the sea and the lowest mean concentrations of BPA was 5.73 ng/g·d.w. for coastal sediments.

5. Remediation of alkylphenols and bisphenol A from water environment

During the last two decades, alkylphenols and alkylphenol polyethoxylates, as well as bisphenol A as priority

hazardous substances present in aquatic compartments, have become a relevant research topic for scientists interested in water engineering [78,79]. This research is focused on finding technologies that are efficient, economically viable, technologically simple, and without environmental impact. It is indicated that these compounds can be removed from an aqueous environment by coagulation, adsorption, membrane processes and advanced oxidation.

5.1. Coagulation-flocculation

Coagulation–flocculation is used for the removal of particulate matter, colloids as well as some dissolved substances. Matamoros and Salvadó [80] evaluated the micropollutants removal with the coagulation/flocculation method in a lamellar clarifier used for secondary effluent treatment. The removal rates ranged from imperceptible elimination up to 50%, among which relatively high rates (20%–50%) were observed for octylphenol at pH = 7–8. Asakura and Matsuto [81] pointed out that treatment of landfill leachate by coagulation and sedimentation was not able to remove bisphenol A, but assured much higher removal rates of nonylphenol (90%), due to their high K_{ow} (4–6).

5.2. Adsorption

Adsorption, due to its relatively low cost, can be used to removal a lot of micropollutants from water and wastewater. The process can be carried out using typical activated carbon or activated carbon coming from natural sources [82]. Hernández-Leal et al. [83] demonstrated high removal of various micropollutants (bisphenol A and nonylphenol) during powdered activated carbon treatment with initial compound concentrations of 100-1,600 µg/L, at a dose of 1.25 g/L and a contact time of 5 min. The authors evaluated also the treatment effectiveness of aerobic effluent externally contaminated with organic micropollutants (0.1-10 µg/L) in a granulated activated carbon column. The removal rates for all the compounds were generally high (>67%), of which nonylphenol to more than 50%. Mesoporous carbon has an ordered pore structure with a large volume and adsorption surface area which is an advantage over typical activated carbon. Liu et al. [84] and Yuan et al. [85] conducted adsorption studies of nonylphenol ethoxylates on mesoporous carbons with a pore size of 2.8-3.0 nm. For initial concentrations of 107-530 mg/L, the adsorption rates were 923, 720 and 463 mg/g. Numerous studies demonstrated that eco-friendly cyclodextrin polymers are a promising alternative in the treatment of wastewater containing a mixture of emerging trace substances [78,86]. 1 g of material was able to adsorb 85% of alkylphenol ethoxylate from the solution containing 200 ppm of non-ionic surfactant [78]. Such materials could be used in batch or column adsorption experiments with surprisingly similar performances and can be easily regenerated. In recent years, potential low-cost adsorbents based on zeolites, clays, graphene/graphene oxides, nanomaterials, and composite materials have been extensively developed, demonstrating excellent adsorptive bisphenol A exclusion performance from aqueous solutions [79]. For instance, Vakili et al. [87] used crosslinked chitosan/zeolite as a fixedbed column for the adsorption investigation of different organic micropollutants including bisphenol A from wastewater. They reported that the bisphenol A removal capacity and removal efficiency of the chitosan/zeolite were 1.4 mg/g and 89%, and these parameters are constant in at least seven adsorption/desorption cycles. Modified zeolite with cetyltrimethylammonium bromide shows increased adsorption efficiency of bisphenol A amounted to 37.8 mg/g [88].

Phenolic xenoestrogens (octylphenol, nonylphenol, bisphenol A and bisphenol F) can be removed from water by means of membrane processes, mainly by nanofiltration (NF), reverse osmosis and ultrafiltration (UF). Both, the retention coefficient and the rate of adsorption of xenoestrogens strongly depend on the type of removed compound (Table 2) as well as on a membrane type [79,89–92]. During NF of water containing a mixture of xenoestrogens changes in volumetric permeate flux (J_{ν}) were not observed, and relative volume permeate flux (α) was close to 1. The high

retention of octylphenol and nonylphenol in the range from 61 to 73% was observed for SF-10 and DS-5-DK membranes, while in the case of bisphenol A DS-5-DK (69%) and MQ-16 (75%) membranes were more sufficient. With the increase of the concentration of xenoestrogens in water, the decrease of retention was observed as a result of the progressive saturation of the membrane surface with these compounds. Bisphenol A removal was studied with a nanofiltration membrane and a maximum rejection of 90% was obtained [91].

In Khazaali et al. [93] study, BPA was removed from aqueous solutions using a low-pressure reverse osmosis system. The results showed a maximum rejection of 87.34% for a 50 mg/L feed concentration at 408.1 kPa and pH 8. Nasseri et al. [94], using polysulfone/graphene oxide nanocomposite membranes, reported 93% removal of bisphenol A under standard experimental conditions. Zahari et al. [95] developed nanofiber polyvinylidene fluoride (PVDF) nanocomposite membranes with various fillers, including polyvinylpyrrolidone (PVP), activated carbon (AC), and MnO₂, for the removal of bisphenol A from wastewater. They indicated that in a single-layer filtration, the PVDF/PVP, PVDF/ PVP/AC, and PVDF/PVP/MnO, membranes showed 100% bisphenol A removal efficiency. For UF membranes, at concentrations of the compounds in deionized water at the level of 40 µg/L, the removal of bisphenol F, bisphenol A, 4-tert-octylphenol and 4-nonylphenol for GM membranes was up to 19%, 67%, 28%, and 52%, respectively, while for the more compact DS-GE membrane, it was up to 72%, 56%, 88% and 100%, respectively [92]. Romero-Reves and Heemstra [96] developed functionalized polyethylene glycol/poly(methyl methacrylate)-based ultrafiltration membranes and demonstrated the capability of bisphenol A removal in monocomponent and multicomponent filtration systems. It was found that at the optimized ultrafiltration experiments, the membranes showed 87% bisphenol A attachment capacity in a monocomponent filtration system (initial bisphenol A concentration: 200 nM). In a multicomponent filtration system, the aptamers-functionalized membranes could selectively attach 14 nmol bisphenol A from a mixture of bisphenol A, atrazine, and microcystin-LR (total initial concentration: 200 nM).

5.3. Advanced oxidation processes

Advanced oxidation processes (AOPs) are novel technologies for water and wastewater treatment. AOPs are

Table 2

Comparison of the retention coefficients and adsorption rates of phenolic xenoestrogens and relative permeate flux of the investigated nanofiltration membranes (Osmonics)

Compound	Nanofiltration membrane type					
	SF – 10	DS – 5 – DK	MQ - 16	DS – 51 – HL		
	Retention/adsorption (%)					
4-tert-octylphenol	71.8/52.2	72.7/51.4	47.0/17.9	48.0/19.5		
4-nonylphenol	60.5/68.0	70.3/68.0	50.0/52.5	43.5/57.3		
Bisphenol A	45.8/52.5	68.8/69.3	75.0/75.0	60.7/33.4		
Bisphenol F	-	75–78	-	-		
Relative permeate flux, α	0.92	0.99	0.99	0.98		

based on transient *in-situ* generation of highly reactive oxidants and radicals (mainly OH[•], O_2^{--} , H_2O_2 , O_3), which cause mineralization of organic compounds, water pathogens, including alkylphenols and bisphenol A [78,79,97,98].

Several significant works demonstrated that ozonation efficiently reduced the concentrations of many trace organic compounds present in secondary wastewater effluents, including pharmaceuticals and alkylphenols through direct and indirect oxidation reactions [78]. Ning et al. [99] showed that the ozonation process was an appropriate tool for the degradation of nonylphenol and octylphenol. The dose of ozone required for the complete degradation of these substances was increased, but a usual dose of 12 mg/L was sufficient for degradation [100]. The study carried out on the removal of nonylphenol from wastewater treatment plants showed that ozonation is an economically viable solution as a tertiary step and a promising technology to reduce pollutants emissions [101]. A drawback of ozonation is the insufficient mineralization of pollutants. The combination of ozonation with homogeneous with other techniques such as ultrasonic/UV radiation, hydrogen peroxide, or other hybrid methods can increase the production of hydroxyl radicals can be one of the solutions [102–104].

5.4. Fenton/Fenton-like processes

Fenton/Fenton-like processes, thanks to the formation of hydroxyl radicals by the reaction within hydrogen peroxide and ferrous ion, have broadly been employed for the degradation of aqueous bisphenol A owing to easy operation and superior catalytic activity [105]. For instance, Guo et al. [106] prepared iron nanoparticles from grapeseed extracts and applied them as a heterogeneous Fenton-like catalyst for the degradation of bisphenol A with an efficiency amounted to 96.4% at pH range 3–11. The degradation mechanism consisted of synergistic effects of adsorption and oxidation with hydroxyl radicals to form the bisphenol A intermediates, followed by mineralization to CO₂ and H₂O. Zhang et al. [107] used a multi-component catalyst, β -cyclodextrin (β-CD) and reduced graphene oxide (rGO) co-modified Fe_3O_4 as a synergistic catalyzer for bisphenol A removal. The removal of BPA amounted to 78.2% after 5 cycles, indicating its excellent stability and reusability. Although electro-Fenton is popular for the removal of pollutants from wastewater, only one paper was found for alkylphenols and their ethoxylates. Oturan et al. [108] compared the treatment of mature landfill leachates (>10 y) by electro-Fenton and electrooxidation for the removal of 55 pollutants with poor degradability, including alkylphenols and their ethoxylates. Both electrochemical processes were efficient for the removal of alkylphenols and their ethoxylates: 85%-100% by electrooxidation and 96%-100% by electro-Fenton.

5.5. Photocatalytic degradation of nonylphenols

Photocatalytic degradation of nonylphenols in aqueous solutions in the presence of $\text{TiO}_{2'}$ as a catalyst, degrades them via radicals generated after irradiation. Intermediate products are hydroxylated mono-, alkylphenols and di-ethoxylates, and other lower molecular weight products [78]. The proposed model involves the oxidation of the aromatic ring

and ethoxylate chains, followed by the oxidation of longer aliphatic chains, resulting in the formation of intermediates such as ethylene glycols and various acidic compounds, and finally almost quantitative conversion to CO_2 [78]. Musaev et al. [109] have studied the photocatalytic degradation of two commercial alkylphenol ethoxylates, IGEPAL® CO-520 (a nonylphenol ethoxylate with 5 mol of ethoxylates) and TritonTM X-100 (an octylphenol ethoxylate with 9.5 ethylene oxide units) using PEG-templated TiO₂ and SiO₂-TiO₂ nanocomposites. The results showed that photodegradation initially occurs by cleavage of the ethoxylate chain and shorter-chain alkylphenol ethoxylates result.

In recent years, numerous endeavors have been made to develop highly efficient photocatalysts for the degradation of aqueous bisphenol A [110]. TiO₂-based photocatalysts have been extensively studied for the degradation of a wide range of aqueous pollutants, including bisphenol A, owing to their low toxicity, superior effectiveness, and photochemical stability [111]. Sambaza et al. [112] used polyaniline-enfolded TiO, nanorods as photocatalysts for the photocatalytic degradation of bisphenol A, achieving a degradation rate of 99.7% under ultraviolet light radiation and optimized experimental conditions. In addition, they reported extraordinary reusability of the polyaniline/TiO, catalyst following the achieved 80% degradation capacity in the fifth consecutive degradation cycle. In another report, Tang et al. [111] used an anatase TiO₂/MIL-101(Cr) nanocomposite for photocatalytic bisphenol A degradation in wastewater and obtained under ultraviolet light irradiation and optimum experimental conditions, achieving a degradation rate of 99.4%. The TiO₂/MIL-101(Cr) nanocomposite shows excellent stability and recyclability in four consecutive cycles, with no significant loss in photocatalytic degradation activity. Wang et al. [113] developed a TiO₃/aspartic acid-β-cyclodextrin/reduced graphene oxide composite and used it as a photocatalyst for the degradation of bisphenol A in contaminated water. They stated that the bisphenol A degradation efficiency of the composite reached 85.6% under ultraviolet light irradiation. Furthermore, the composite demonstrated excellent stability and reusability in five repetitive cycles of degradation experiments.

ZnO is a semiconductor that has been extensively studied as a photocatalyst for the degradation of aqueous bisphenol A [114]. For instance, Shekoohiyan et al. [115] developed a CuO/Fe₂O₂/ZnO composite for the visible-light-promoted photocatalytic degradation of bisphenol A. The bisphenol A exclusion capacity of the composite reached 97.5% after 120 min of visible light irradiation under optimized experimental conditions. Kamaraj et al. [116] fabricated ZnO nanoparticles (10-50 nm) from cactus pear fruit peel (CPFP) infusions for the use of photocatalytic degradation of bisphenol A under sunlight irradiation. The result showed almost complete mineralization of bisphenol A at the standard experimental conditions, and ZnO nanoparticles showed >90% bisphenol A removal efficiency in five repeated photocatalysis cycles. Ruan and Hu [117] prepared catalyst C_3N_4 –Zn/Bi₂WO₆ for the degradation of bisphenol A, which achieved up to 93% efficiency.

Furthermore, in recent years, many researchers have developed bismuth and silver-based semiconductors for the photocatalytic degradation of bisphenol A [118]. For instance, Wang et al. [119] synthesized S-doped bismuth oxybromide nanosheets, which were used as a photocatalyst for the degradation of bisphenol A under visible-light irradiation at $\lambda \ge 420$ nm. It was found that the nanosheets degraded 87%–92% of bisphenol A within 180 min of the visible-light irradiation.

5.6. Biological degradation

The results of studies reported in the literature indicate the possibility of degradation as well as complete mineralization of nonylphenols with the participation of bacteria under aerobic and anaerobic conditions. Sphingomonas strains are capable of metabolizing isomers of nonylphenols in the absence of other carbon and energy sources. Also, some fungi can use nonylphenols as their only carbon source. Biodegradation with Sphingomonas leads to 1,4-dihydroxybenzene, while decomposition under the influence of fungal enzymes produces several or more metabolites including 4-hydroxybenzoic acid and hydroxyphenylpropionic acid. Biodegradation studies of ¹⁴C isotope-labelled 4-nonylphenol using G. simplex fungi and Sphingomonas bacteria confirmed the possibility of total mineralization [120]. Recent studies have also reported the removal, biosorption, and biodegradation of alkylphenols by microalgae [121]. For example, Zhou et al. [121] studied the removal of nonylphenol and octylphenol by the freshwater green microalga Scenedesmus obliguus in cultures exposed to different concentrations of alkylphenols. The dissolved concentrations for alkylphenols in the medium decreased rapidly: nonylphenol and octylphenol were rapidly eliminated with efficiencies of 70.9%-90.4% for nonylphenol and 37.8%-82.2% for octylphenol, depending on the initial concentration. Similar findings were previously reported by Gao et al. [122] and Wang et al. [123] for other algal species, Chlorella fusca and Microcystis aeruginosa, respectively.

Elimination of aqueous bisphenol A using biological methods, that is, using microalgae and microorganisms/ fungi and their enzymes under both free and immobilized conditions has also been recognized as a promising technique due to their higher efficiency and eco-friendly nature [124]. Bisphenol A biodegradation involves the formation of nontoxic metabolites with less hazardous effects than bisphenol A, as well as complete biomineralization into water and carbon dioxide. However, the intermediates formed, namely p-hydroxyacetophenone and hydroquinone, are sometimes more hazardous than bisphenol A [125]. Several bacteria found in wastewater treatment plants, river water, and soil are capable of readily degrading bisphenol A [126]. For instance, the bacterial family Sphingomonadaceae has been identified for its high bisphenol A degradation capacity in bisphenol A-contaminated solutions and metabolizes this compound using its source of energy and carbon [127]. Also, the isolated Pseudomonas putida strain YC-AE1 from the contaminated soil of Guangdong, China, was reported to efficiently and rapidly biodegrade aqueous bisphenol A [128]. Zühlke et al. [129] and Jia et al. [130] isolated Bacillus amyloliquefaciens bacterial strain and Sphingobium species (YC-JY1) from sewage sludge and investigated their biodegradation capacity for bisphenol A. They reported that bacteria and YC-JY1 strain degraded 77% and 100% of bisphenol A, respectively.

6. Toxicity and restrictions

Accumulation of alkylphenols and bisphenol A in aquatic ecosystems may have a negative impact on the organisms living in them. The presence of these compounds in bottom sediments may affect benthic organisms, especially silteating, at a low level of the trophic chain [7]. Alkylphenols are also very toxic to vertebrates, invertebrates, and algae. Chronic exposure to these compounds has been proven to cause estrogenic effects in fish, birds, and mammals [7,131]. Negative and harmful effects on organisms need not be caused by high EDC concentrations in environmental matrices. Even trace amounts can interfere with the activity of natural hormones. Due to the ability of these compounds to bioaccumulate and biomagnify, organisms at the highest trophic level are also exposed to their negative impact [132].

Humans are mainly exposed to endocrine-disrupting chemicals such as alkylphenols and bisphenol A mainly by the intake of contaminated foods and drinking water [17,133]. The nonylphenols and octylphenols added as additives to plastic packaging may migrate into products, including foods and drinking water constituting a source of human exposure. Other routes include the use of personal hygiene products, detergents, or spermicides in contraceptives [134]. Nonylphenol was detected in human biological samples such as breast milk, blood, and urine [17]. After entering the human body, phenol derivatives can imitate the effects of 17 β -estradiol (steroid sex hormone) [7] causing cancer of the sex organs: testicles, prostate, and breasts. In animals, the effects of BPA, NP, and OP disrupt the endocrine system, causing feminization or hermaphroditism [9,135,136].

Due to their endocrine effects, phenol derivatives have been classified by the European Chemicals Agency (ECHA) as substances of particular concern [137]. The first restrictions on the use of BPA in consumer products date back to March 2011. Currently, the European Union (EU) prohibits the production, transport, and sale of baby bottles containing this compound [25,137–139]. Under Regulation 10/2011/EU [140], the European Union allows the use of BPA in materials that have contact with food, but in a limited amount (up to 0.05 mg/kg). This compound is also approved for use in food contact materials in other countries such as USA and Japan [132,141]. In the case of nonylphenols and their ethoxylates, the first restrictions in the European Union regarding the reduction of these compounds date back to 2003. For octylphenols, there are no restrictions on their consumer use [132].

Nonylphenols and octylphenols are listed among the priority substances in the field of water policy in the Water Directive of the EU (Directive 2000/60/EC) [142]. Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy [143] sets environmental quality standards for surface waters at the level of 0.3 μ g/L for nonylphenols, including 4t-OP as average annual concentrations in inland surface waters. For drinking water, The United States Environmental Protection Agency recommends a maximum allowable concentration of 4nNP and BPA of 7 and 100 μ g/L, respectively [2,25]. While the EU Directive 2020/2184 of 16 December 2020 on the quality of water intended for

human consumption [144] it follows that the World Health Organization recommended that three representative substances (bisphenol A, nonylphenol, β -estradiol) as reference factors for assessing the presence of endocrine disruptors and the effectiveness of their removal, with concentrations of 0.1 µg/L for bisphenol A and 0.3 µg/L for nonylphenol.

7. Summary and conclusions

We are now living in the plastic world which is a serious and global problem. But apart from the problems and threats posed by environmental pollution with plastics and their degradation particles (microplastics, nanoplastics), plastic additives are also a big issue. Considering that some of them are non-permanently bound to the polymer matrix and can migrate from the plastic to the external environment, exacerbates the problem. Leaching additives from plastic products is one of the ways through which bisphenol A and alkylphenols can get into the environment, including the aquatic environment. Studies have shown that these compounds can be leached into water, for example, from products such as polycarbonate water bottles, recycled tire granules, plastic packaging made of HDPE, LDPE, PET, PS, PE, PVC. However, there is a lack of studies in which experiments were conducted for each type of polymer and all three discussed substances. Given the increased use of plastics in the packaging sector, leaching testing allows to determine the extent of migration of these endocrine disruptors which, if ingested continuously, may have effects on human health. Therefore, the leaching mechanism of BPA, 4tOP, 4nNP used as additives in the polymer industry needs to be further investigated.

Some technologies, characterized by efficiency, profitability, simplicity and environmental friendliness, like coagulation, adsorption, membrane processes, advanced oxidation, and biological degradation, can be used to remediate these compounds from the aquatic environment. Up to 50% of octylphenol can be eliminated during coagulation/flocculation. In the case of nonylphenol, it can be up to 90%. In adsorption, the efficiency depends primarily on the adsorbent used and its properties, ranging from 50% to 90%. In the case of membrane processes, such as nanofiltration, reverse osmosis, and ultrafiltration, the efficiency of removing micropollutants, including BPA or alkylphenols, strongly depends on the type of removed compound as well as on a membrane type and its separation properties. Depending on the mentioned factors, up to 100% removal of these compounds is observed, which shows that membrane techniques are highly effective in removing endocrine active compounds, such as BPA, OP, and NP, from water and wastewater. Other promising methods include AOPs: ozonation, Fenton/Fenton-like processes, or photocatalytic degradation. When listing the methods of remediation of phenol-derivative compounds from the water environment, it is impossible not to mention biological methods that are particularly environmentally friendly. The effective use of bacteria, fungi, and algae for the degradation or complete mineralization of nonylphenols or bisphenol A has already been confirmed by researchers.

Limiting the amount of these pollutants in the aquatic environment is important due to the endocrine properties of these compounds, affecting the proper functioning of the endocrine system of living organisms. EDC concentrations do not have to be high to cause negative and harmful effects on organisms. Trace amounts can also affect the activity of natural hormones, posing a risk to aquatic organisms and, due to their ability to bioconcentrate and biomagnify, ultimately also to humans. It is also important to monitor their levels in the environment and fate, especially in aquatic environments, to avoid long-term impacts on living organisms.

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