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Occurrence and removal of polymeric material markers in water environment: a review

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ABSTRACT

Alkylphenols (AP) and bisphenol A (BPA) are chemicals belonging to the endocrine-disrupting compound group. These compounds have a negative influence on the endocrine system of both humans and wildlife. Adverse effects include serious reproductive and developmental disorders, such as feminization of the population, especially in wildlife. AP and BPA are commonly used in household items and industrial applications or as monomers or additives in plastic production. Methods for AP and BPA degradation can be divided into four main groups. In membrane-type processes, the important factor is membrane selection. Good results were obtained with the application of dense nanofiltration or reverse osmosis membranes. The selection of bacterial or fungi species is crucial for biological treatment. *Sphingomonas* and *Pseudomonas* sp. are known for their good organic compound degradation capabilities. The oxidation process is based on the proper selection of an oxidation agent and catalyst when needed. Hydrogen peroxide and ozone are often mentioned in the literature. Common catalysts are UV radiation, Fe^{2+} ions, or oxides of tin, zinc, and titanium. In sorption processes, the selection of the sorbent is important. In the literature, natural materials like sewage sludge or river sediments are often mentioned. An important group is also the carbon-based sorbents like carbon nanotubes or activated carbon. The scope of this work is to present sources of AP and BPA in water matrices, as well as possibilities of AP and BPA removal along with important aspects of the processes.

Keywords: Alkylphenols; Bisphenol A; Occurrence; Degradation; Water; Wastewater

1. Introduction

Alkylphenols (AP), especially 4-tert-octylphenol (4tOP), 4-n-nonylphenol (4nNP) and bisphenol A (BPA), belong to the group of endocrine-disrupting compounds. This group of compounds resembles female reproductive hormones both in structure and properties [1–7]. Fig. 1 presents structures of 4nNP, 4tOP, BPA, and estradiol.

AP, especially 4tOP and 4nNP, are used in many branches of industry. The main application is the production of alkylphenol ethoxylates, substances used as industrial detergent [7]. The most popular nonylphenol ethoxylate represents 80% of the worldwide production of alkylphenol ethoxylates. The residual 20% is dominated by 4tOP ethoxylate [8–10]. The annual production of alkylphenol ethoxylate was equal to 500,000 tons in 1996 and decreased to 300,000 tons in 2006. It is estimated that 60% of the annual production ends up in the water environment [9–11]. Apart from being substrates for nonionic surfactants, AP is used in the production of personal care products, paints, detergents, herbicides, pesticides, emulsifiers, wetting and dispersing agents, antistatic agents, demulsifies and solubilizers.

The main application of BPA in the production of plastics like polycarbonates and phenolic and epoxy resins. The production of epoxy and polycarbonates equals 680,000 tons in 2006. Apart from being a monomer in the plastics industry,

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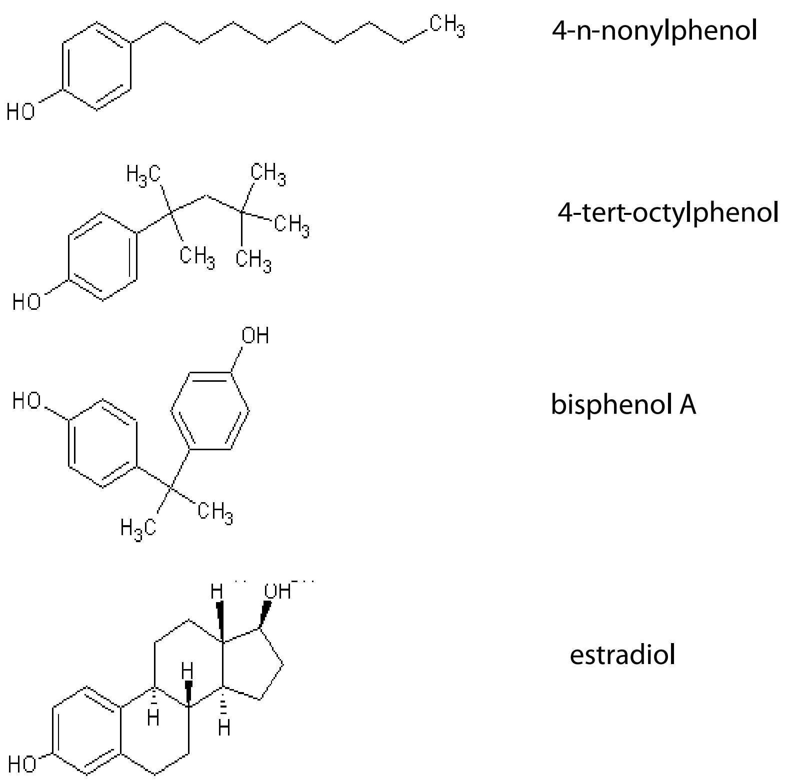


Fig. 1. Comparison of structures: 4-n-nonylphenol, 4tOP, BPA, estradiol (female reproductive hormone).

BPA can be also found in a variety of consumer products [8,12,13].

AP and BPA have an adverse effect on both humans and aquatic biota. In humans, exposure to AP and BPA may lead to serious reproductive and developmental disorders as well as to cancer. Short-term results of exposure, especially to 4nNP, include eye and skin irritation, headaches, nausea, and vomiting. In marine biota, AP and BPA affect reproductive function and, apart from other effects, may cause feminization or even hermaphroditism in the population [12–17].

Due to adverse effects exerted upon both humans and aquatic organisms, some regulations were established in different countries to either forbid usage or determine its limits in the environment. Directive 2008/105/WE sets European Quality Standards (EQS) for both 4nNP and 4tOP as annual average (AA) and maximum allowable concentration (MAC). The values of these parameters are presented in Table 1.

The United States Environmental Protection Agency recommends a MAC for 4nNP equal to 7 μg/L. For BPA, the same agency established a limit of 100 μg/L for drinking water. Countries like Canada, Denmark, and France banned the usage of BPA in products designed for children aged 0–3 [13,15,18,19]. The scope of this work is to present sources of contamination as well as methods of AP and BPA removal from the water environment.

2. Sources and fate in environment

AP and BPA are introduced into the environment from different sources. Two of them are of greater significance.

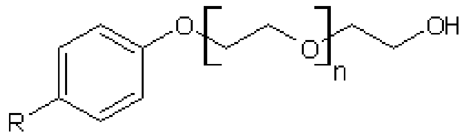
2.1. Wastewater and wastewater treatment plants

Due to the application of AP and BPA in different branches of industry as well as in common household applications, it is natural that municipal and industrial wastes will be an important source of contamination. In contrast to BPA, AP often enters both the municipal wastewater system and the environment in general in the form of the corresponding ethoxylates (Fig. 2) [6,13,20–23].

The literature on the subject provides data on the concentration of different alkylphenol polyethoxylates (APE) in surface waters and raw and treated wastewater. Wu et al. [21] provide results of APE measurement in four different wastewater treatment plants (WWTP) located in Hong Kong. The major compounds detected in sewage from the studied location were 4-n-nonylphenol-monoethoxylate (NPE1) and 4-n-nonylphenol-diethoxylate (NPE2) with concentrations of 3,548 and 3,288 ng/L, respectively. For 4-tert-octylphenol-monoethoxylate (OPE1) and 4-tert-octyl-phenol-diethoxylate (OPE2) the concentrations were detected in the range 1.1–58 μg/L. Loos et al. [24] delivers rich data from Belgian and Italian WWTP effluents, consisting of data measured for different length 4nNP ethoxylates and OPE2. The data ranges were from 1.3 μg/L to below detection limit (BDL) for NPE1, from 1 to 0.02 μg/L for NPE2, and from 0.08 to 0.81 μg/L for 4-n-nonylphenol-tri-ethoxylate (NPE3). Similar to [21], values for OPE2 were lower and ranged from 0.044 to 0.002 μg/L depending upon the WWTP. Ahel et al. [25] present results obtained for the sum of NPEs of different chain lengths measured in eight different WWTPs located in the canton of Zurich.

Table 1
EQS values for 4nNP and 4tOP

Compound	AA-EQS inland surface waters (μg/L)	AA-EQS other waters (μg/L)	MAC-ESQ inland surface waters (μg/L)	MAC-ESQ other waters (μg/L)
4nNP	0.3	0.3	2	2
4tOP	0.1	0.01	–	–



R: C $_9$H$_{19}$ 4-*n*-nonylphenol
R: 4 -(1,1′,3,3′-tetramethylbutyl) 4-*tert*-oktylphenol

Fig. 2. General form of alkylphenol polyethoxylate, *n* equals from 1 up to 19 units.

Results varied from 430 to 99 μg/L and 8 to 2 μg/L in primary and secondary effluents, respectively. Ding et al. [22] present data from Taipei WWTP and Laojie River contaminated with untreated municipal sewage. For Taipei WWTP, the data are as follows: 9.6, 15.3, and 5.4 μg/L for NPE1, NPE2, and NPE3, respectively. For Laojie River, the data are as follows: 10.3, 10.1, and 0.9 μg/L for NPE1, NPE2, and NPE3, respectively. Ferguson et al. [26] present data for the sum of NPEs of different chain lengths from influent and effluent of Yonkers (New York) municipal WWTP. The following data were obtained: 1,434 and 100.9 μg/L for influent and effluent, respectively.

In the environment, alkylphenol polyethoxylate undergoes degradation that results in AP as the end product. Fig. 3 presents the scheme of this process [6,10].

2.2. Polymeric materials

AP and BPA that originate from polymeric material production may take a form of additives or components of additives. For example, 4nNP is used to produce tris (nonylphenyl) phosphate [27–29]. Polymerization reactions are often uncompleted, leaving unreacted monomers and some other ingredients. BPA, for example, is used as either a monomer or an intermediate in the production of the following resins and polymers: polycarbonate, epoxy resins, polysulfone, polyacrylate, polyetherimide, unsaturated polyester, and phenolic resins. Additives or unreacted monomers are commonly not chemically bonded with polymer itself. This leaves the possibility of migration of plastic-associated contaminants (AP, BPA) into the environment. Migration commonly takes place in amorphous regions of polymers, and its rate is dependent upon factors like concentration gradient, pore size and contaminant molecule size (molecular weight) [18,30–36]. Due to porosity, especially in amorphous fragments in polymer structures, plastic debris and microplastics can absorb and accumulate organic contaminants like 4nNP even up to several orders of magnitude more than in the surrounding medium [36,37]. Hirai et al. [32] present results of the analysis of BPA, octylphenol, and sum of all 4-nonylphenols in plastic debris collected from the open ocean and both remote and urban beaches. For some of nonylphenols, values range from 0.3 to 3,940 ng/g, and for octylphenol the values were lower and range from 0.1 to 153 μg/g. BPA in most samples was detected in trace concentration below 0.1 ng/g. Another important factor is the amount of plastic debris, especially microplastics in surface waters. Table 2 presents the results of microplastic content found in the literature on the subject.

Studies from all over the world indicate the presence of AP and BPA in aqueous matrices. In Poland, a series of studies were conducted by scientists from the University of Gdańsk [14,44]. The subject of the studies was the Gulf of Gdańsk along with the Vistula River estuary and some smaller rivers like Kacza, Plutnica, and Gizdebka. Several sampling points were established in the Gulf of Gdańsk. Dębogórze, Mechelinki and Swarzewo were selected due to proximity of WWTP, and other points like Orłowo, Sopot or Gdańsk harbor were selected as potential sources of phenol derivatives. In samples from April 2011, the highest concentration of 4nNP was recorded around the Sopot sampling point: 172.1 ng/L in the bottom layer. The highest 4tOP concentration was found around the Vistula estuary: 65.9 ng/L in the subsurface layer. The largest concentration of BPA was found at GN station located at some distance from the coastal area: 67.7 ng/L in the bottom layer. In a sampling campaign conducted from spring of 2011 till autumn 2012, samples were taken from the surface microlayer from water from the Gulf of Gdańsk. The average values measured are as follows: 193.2, 128.1, and 127.7 ng/L for BPA, 4tOP, and 4nNP, respectively. Warta River was the subject of studies by Zgoła-Grześkowiak [45]. In the course of these studies, the results were for 0.1 μg/L for 4tOP concentration and BDL for 4nNP. Table 3 presents the results of studies of BPA and AP published in the literature on the subject from studies around the world.

3. Removal techniques

3.1. Membrane-based techniques for removal of AP and BPA

Membrane-based processes, especially nanofiltration and reverse osmosis, are commonly used in water and wastewater

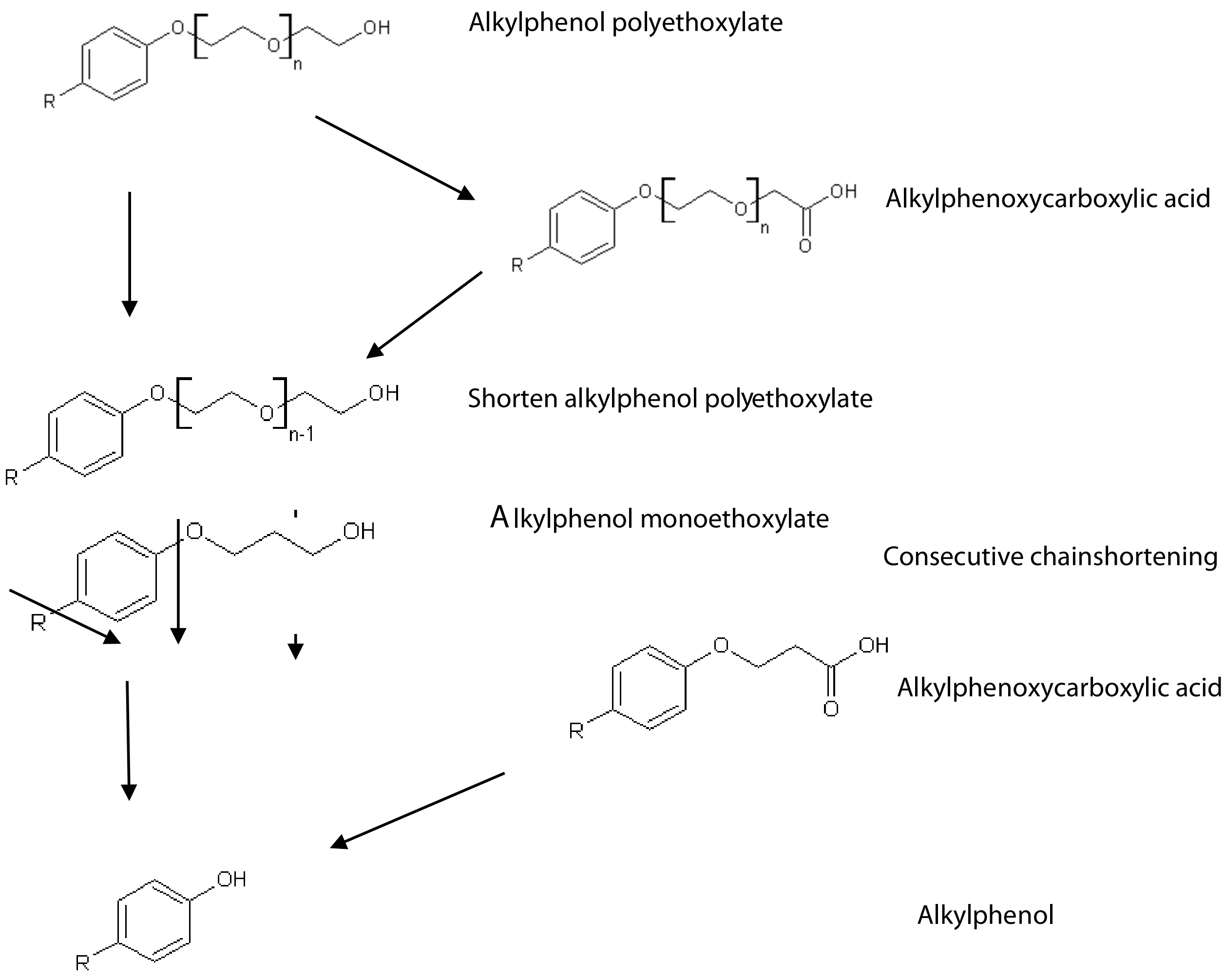


Fig. 3. APE degradation mechanism.

Table 2
Microplastic content in surface waters around a world

Area of study	Microplastic content (items/m^3)	Identified plastics	Ref.
Northwestern Pacific Ocean	0.030	PP, PS, PET, PE	[38]
Bering Sea	0.091	PP, PS, PET, PE	
Chukchi Sea	0.23	PP, PS, PET, PE	
Chabahar Bay	0.49	PE PP PS PET PVA	[39]
Faafu Atoll, Maldives	0.26	PE PP PET PU PS	[40]
Northern coastal waters of Surabaya, Indonesia	0.485	PS PE PP PU PET PBD	[41]
Buqu River	967 ± 141	PET PE PP PS PA	[42]
Naqu River	817 ± 589	PET PE PP PS PA	
Lhasa River upstream Lhasa city	683 ± 354	PET PE PP PS PA	
Lhasa River downstream Lhasa city	700 ± 94	PET PE PP PS PA	
Nyang River upstream Nyingchi	483 ± 118	PET PE PP PS PA	
Nyang River downstream Nyingchi	517±24	PET PE PP PS PA	
Baltic Sea around Stockholm	3.11–7.73	PP PE PS	[43]
Baltic Sea outer Stockholm Archipelago	0.19–1.13	PP PE PS	

Abbreviations: PP–polypropylene, PS–polystyrene, PET–polyethylene terephthalate, PE–polyethylene, PVA–polyvinylalcohol, PU–polyurethane, PBD–polybutadiene, PA–polyamide

Table 3
Concentration of BPA and selected AP in environmental samples worldwide

Area of study	BPA	4nNP	4tOP	Ref.
Tama River, Tokyo, Japan	11.3 ng/L	14.9 ng/L	2.5 ng/L	[1]
Ogun River	ND	48.8–72.3 ng/L	57.13–68.3 ng/L	[2]
Ibeche River	ND	43.9–79.4 ng/L	66.2– 68.6 ng/L	
Manfouah WWTP	3.853 µg/L	–	–	[3]
Wadi Hanifa WWTP	3.628 µg/L	–	–	
King Saud University WWTP	4.672 µg/L	–	–	
Al-Hofuf WWTP, Al-Ahsa	3.180 µg/L	–	–	
Wadi Al-Araj WWTP, Taif	6.679 µg/L	–	–	
Galindo WWTP	88±16 ng/L	–	ND	[5]
Estuary of Bilbao	347±1 ng/L	–	ND	
Aire WWTP, Geneva, Switzerland	–	12.6 µg/L	204 ng/L	[6]
Kaveri River, India	6.6–136 ng/L	ND–2,200 ng/L	ND–16.3 ng/L	[8]
Vellar River India	2.8–6 ng/L	22.4–158 ng/L	ND–16.3 ng/L	
Tamiraparani River, India	9.8–36 ng/L	ND–1,455 ng/L	ND–16.3 ng/L	
Guadalquivir, Spain	–	1.1±0.2 µg/L	9±1 µg/L	[9]
Guadiana, Spain	–	0.9±0.3 µg/L	0.8±0.1 µg/L	
Linares WWTP effluent, Spain	–	41±6 µg/L	4.1±0.6 µg/L	
Andujar WWTP effluent, Spain	–	23±4 µg/L	8±1 µg/L	
Mengibar WWTP effluent, Spain	–	24±5 µg/L	8.3±0.4 µg/L	
Bailen WWTP effluent, Spain	–	20±4 µg/L	8±1 µg/L	
Coruna industrial zone, Spain	0.035 µg/L	ND–0.059 µg/L	ND–0.11 µg/L	[12]
Sembawang Park Singapore coastal area	0.04 µg/L	1.03 µg/L	–	[16]
Punggol Singapore coastal area	0.19 µg/L	1.63 µg/L	–	
Pasir Ris Singapore coastal area	0.04 µg/L	0.37 µg/L	–	
Changi Singapore coastal area	0.05 µg/L	0.32 µg/L	–	
Jurong Pier Singapore coastal area	0.17 µg/L	2.4 µg/L	–	
Tuas Jetty Singapore coastal area	0.04 µg/L	2.76 µg/L	–	
Warta River, Poland	–	ND	0.1±0.01 µg/L	[45]
Fujima River	0.03 µg/L	ND–0.3 µg/L	ND–0.09 µg/L	[46]
Shiratori River	ND	0.02–0.1 µg/L	ND	
Sapporo City WWTP, Japan	–	ND	914 ng/L	[47]

ND– not detected (below limit of quantification)

treatment to remove a large variety of organic and inorganic contaminants [48]. Membrane processes can be characterized by several parameters.

3.1.1. Operating pressure

Pressure difference is a driving force because the membrane process retention coefficient and permeation rate depend upon it. The literature on the subject provides some data on the operating pressure. Agenson et al. [48] conducted their lab-scale tests at two different pressures, namely 0.1 and 0.3 MPa. A higher pressure was applied by Jung et al. [49]. Depending upon the membrane, it was either 1 or 0.5 MPa. The operating pressure of 1 MPa was also applied in a process presented in [50].

3.1.2. Contact angle

The contact angle is a measure to determine hydrophobicity of a membrane. Studies conducted by Wintgens et al. [51] present the results of studies governing nonylphenol retention and hydrophilicity of different membranes. Studies conducted on 11 different nanofiltration membranes indicate that retention of nonylphenol increases with an increase of hydrophilic character (thus with a decrease of contact angle). This behavior was explained by assuming the solution diffusion model of nonylphenol transportation through the membrane. Wintgens et al. [51] stated that although nonylphenol flux is stable, the water permeation coefficient increases with decreasing contact angle (thus with an increase of hydrophilic character), which leads to increase of nonylphenol retention.

3.1.3. Solute parameters

Dudziak et al. [52] present results of rejection depending upon solute particle physico-chemical parameters. For reverse osmosis processes (DS–3–SE membrane), the statistical analysis provides that the dipole moment of the particle is the most important parameter influencing rejection. For nanofiltration membranes (DS–5–DK), statistical analysis indicates the importance of the logK_{ow} coefficient (a measure of hydrophobicity of compound). Kimura et al. [53] present the result of studies of a reverse osmosis process for organic compound removal on two membranes, namely XLE and SC-3100. Obtained results indicate that the most important factor influencing the rejection of the solute compound is the size of the compound (expressed as molecular weight), especially for the XLE membrane. A similar dependence was found by Dudziak at al. [52] for a loose GE nanofiltration membrane. Several studies present the results of the rejection coefficient for different membranes. Often these studies were conducted at a laboratory scale. These results are presented in Table 4.

One of the many applications of membrane technology is a membrane bioreactor (MBR) used for wastewater treatment. A MBR is typically composed of two parts. The first part, a biological one, is responsible for the biodegradation of wastewater compounds. The second part, a membrane part, is used for further removal of contaminants from the first part. A large variety of membranes can be used for this process. The literature on the subject proposes polymeric as well as metallic or ceramic membranes [55]. Cases et al. [56] present the results of a comparison between the removal efficiency of conventional WWTP based on an activated sludge process with pilot-scale MBR. Results of this comparison indicate that higher removal efficiency for nonylphenol is obtained for the MBR process with a hollow-fiber membrane. For 4tOP and BPA, the removal efficiency was similar regarding the process or membrane applied. Similar studies were conducted by Clara et al. [57].

3.2. Biologically based techniques for AP and BPA removal

The literature on the subject provides different data on various bacteria and fungi able to degrade AP and BPA. The species mentioned most often are *Pseudomonas*, *Bacillus*, *Sphingomonas*, and *Klebsiella* sp. of different strains [57–63]. From fungal species, the literature often mentions different strains of *Fusarium falciforme* and *Fusarium* species. Cabana et al. [64,65] present a result of studies upon nonylphenol and BPA degradation by laccase enzyme, obtained from white-rot fungi *Coriolopsis polyzona*. Studies indicate that free laccase can degrade 100% of BPA in 4 h and 80% of nonylphenol technical mixture in 1 h. Laccase in immobilized form (cross-linked aggregate) can degrade 95% of BPA in 2.5 h. Table 5 presents different bacteria and fungi able to degrade BPA and AP.

3.2.1. Degradation pathway and metabolites

Alkylphenol's degradation and pathway are dependent upon active microorganisms responsible for degradation. Tuan et al. [59] and Rajendran et al. [68] proposed that the primary degradation product of AP is alkyl catechol. This was confirmed for different *Pseudomonas* strains as well as for *Candida rugopelliculosa*. Tuan at al. [59] stated that further degradation leads to the formation of either long-chain alkyl 2-hydroxymuconic semialdehyde or long-chain alkyl cis, cis-muconate. This pathway is referred to as phenolic ring hydroxylation.

The second proposed pathway is called the alkyl chain oxidation pathway. This pathway was proposed by Rajendran at al. [68] and Różalska et al. [69]. The pathway

Table 4
Rejection of BPA and AP in membrane processes

Membrane	Operating pressure (MPa)	BPA rejection (%)	4nNP rejection (%)	4tOP rejection (%)	Ref.
UTC 60	0.3	47	–	–	[48]
NTR 729 HF	0.3	99.8	–	–	
UTC70	0.3	99.8	–	–	
ES10C	0.3	>99.9	–	–	
LF10	0.3	>99.9	–	–	
NTR 729 HF	1	–	95	–	[49]
AD SWRO	0.1	>98	–	–	[50]
CE BWRO	0.1	10–40	–	–	
XLE BWRO	0.1	>98	–	–	
NF 270	0.1	80	–	–	
NF 90	0.1	98	–	–	
BW 30	0.1	>98	–	–	
DS–3–SE	2	97±4.4	94±2.1	91±2.3	[52]
DS–5–DK	2	69±0	80±9.1	83±9.4	
GE	2	61±8.7	68±14	63±15	
XLE	0.5	63	–	–	[53]
SC 3100	0.5	18	–	–	
X20	1.03 (150 psi)	96.1±1.5	–	–	[54]
TS80	1.03 (150 psi)	25.6±8.7	–	–	
NF270	0.34 (50 psi)	4.6±6.5	–	–	

Table 5
Degradation of BPA and AP in biological processes

Organism	BPA (%)	4nNP (%)	4tOP (%)	Ref.
Bacillus sp. CYR2	–	57±6	38±4	[58]
Pseudomonas sp. SU1	51.7	–	–	[61]
Klebsiella sp. NE2	51	–	–	
Klebsiella sp. SU5	57	–	–	
Pandoraea sp. HYO6	51.3	–	–	
Alcaligenes sp. OTI7	54.7	–	–	
Enterobacter sp. HI9	60.7	–	–	
Serratia sp. HI10	59.3	–	–	
Pseudomonas sp. NAR11	56.7	–	–	
Pseudomonas sp. FU12	57.7	–	–	
Bacillus NO13	56	–	–	
Bacillus NO15	59	–	–	
Enterobacter HA18	68.3	–	–	
Pseudomonas sp. SU19	53.7	–	–	
Pseudomonas sp. FU20	55.3	–	–	
Rhizobium WZ1	–	79.9	–	[66]
Sphingobium WZ2	–	88.3	–	
Fusarium falciforme RRK20	–	–	73	[67]
Fusarium falciforme RRK08	–	–	70	
Fusarium sp. RRK09	–	–	69	
Fusarium falciforme RRK10	–	–	65	
Fusarium oxysporum species complex RRK18	–	–	65	
Aspergillus fumigatus RRK07	–	–	52–56	
Aspergillus fumigatus RRK15	–	–	52–56	
Trichoderma longibrachiatum RRK01	–	–	52–56	
Trichoderma asperellum RRK14	–	–	52–56	

starts from hydroxylation or carboxylation at the terminal carbon atom of an alkyl chain that leads to its shortening. Its final degradation product is 4-hydroxybenzoic acid. This pathway was tested for two different types of fungi, namely *Gliocephalotrichum simplex* and *Candida rugopelliculosa RRKY5*, with nonylphenol as the degraded molecule.

Vijayalakshmi et al. [60] identify BPA degradation products like phenol, acetophenone hydroquinone, and p-hydroxybenzoic acid. Zhang et al. [63] propose several degradation pathways dependent upon the degrading organism. Although pathways differ, two of the resulting end products are the same regardless of the pathway, namely hydroquinone and p-hydroxybenzoic acid. Kolvenbach et al. [62] present the pathway for further hydroquinone degradation by *Sphingomonas* sp. strain TTNP3. This pathway includes 4-hydroxymuconic semialdehyde, maleylacetate, and 3-oxoadipate.

3.3. Oxidation methods for BPA and alkylphenol degradation

In general, oxidation processes are radically based. Different agents can be applied as the source of radicals. The sources mentioned most often in the literature are hydrogen peroxide [70–73], ozone [73–77] and persulfate [76,78,79]. However, other agents like manganese dioxide [80] are possible. Other methods of obtaining radicals for this process are sonification, electrooxidation or hydrodynamic cavitation. All these mentioned processes are based on hemolytic cleavage between the O–H bond in the water molecule according to Eq. (1) [79,81].

$$H_2O \xrightarrow{X} OH^\bullet + H^\bullet + Y \tag{1}$$

X: ultrasonic energy, electrochemical potential, cavitation; *Y*: heat, electrons.

3.3.1. Catalysis

To improve the degradation rate or to activate the oxidation agent, different catalysts are applied. A common pair is hydrogen peroxide or persulfate with UV radiation. Reactions (2) and (3) present the activation for this process [70–72,76,78].

$$H_2O_2 \xrightarrow{hv} 2OH^\bullet \tag{2}$$

$$S_2O_8^{-2} \xrightarrow{hv} 2SO_4^{\bullet-} \tag{3}$$

Apart from UV radiation, the literature often mentions Fe^{2+} ions or oxides of tin, zinc and titanium.

Eqs. (4) and (5) present the activation reaction for hydrogen peroxide and persulfate catalyzed by Fe^{2+} ions [78,83,84].

$$H_2O_2 + Fe^{+2} \xrightarrow{hv} HO^{\bullet} + HO^{-} + Fe^{3+} \tag{4}$$

$$S_2O_8^{-2} + Fe^{+2} \xrightarrow{hv} SO_4^{\bullet -} + SO_4^{2-} + Fe^{3+} \tag{5}$$

Molkenthin et al. [71] present results for BPA degradation with hydrogen peroxide as the oxygen source combined with UV radiation and Fe^{2+} ions as a dual catalyst system. Similar studies were conducted by Wu et al. [82]. However, in this process, 4tOP undergoes degradation with Fe^{3+} ions as the catalyst. In both cases, with the increase of catalyst concentration, the reaction rate constant values increase.

3.3.2. Effect of pH

The effect of pH is strictly dependent upon the degradation agent used in the process. Dulov et al. [70] and Neamțu et al. [85] present results indicating that the nonylphenol degradation process is faster in the alkaline medium. In both examples, an increase in pH values leads to an increase in the reaction rate constant. This phenomenon was explained based on the molar adsorption coefficient. Czaplicka et al. [86] obtain a similar dependence upon pH for 2,3,4,5-tetrachlorophenol photodegradation. The reaction rate constant increases almost three times with a change in pH from 2 to 11.

Somewhat different effects are observed when hydrogen peroxide, persulfate or ozone is used to degrade the target molecule. Kusvuran et al. [74] observe the highest degradation rate for BPA at pH 3. A similar effect is observed for BPA degradation in a persulfate hydrodynamic cavitation system [79]. The highest reaction rate constant, 12.3×10^{-3} [1/min], is observed at pH 6, in contrast to 3.2×10^{-3} [1/min] at pH 12. A decrease in reaction rate with an increase of pH value is also observed by Lu et al. [80]. Studies on 4tOp and 4nNP reduction with MnO_2 as the degrading agent indicate that MnO_2 reduction potential decreases with an increase of pH, which leads to a drop of the reaction rate. Wu et al. [82] study 4tOP photodegradation enhanced with Fe(III) ions. In this case, the reaction rate drops with increase of pH. This is explained because, in lower pH (3.5), the amount of OH· is highest, which leads to the maximal reaction rate possible.

Oxidation processes provide a faster alternative to biological processes. Table 6 presents data concerning degradation degree as well as oxidation methods for AP and BPA degradation.

3.4. Sorption processes for AP and BPA removal

The primary concern in sorption studies is sorbent selection. In most cases, sorbents can be divided into two groups. The second group consists of natural materials like, for example, river sediments and activated sludge or some mineral materials like kaolinite or gibbsite [88–90]. The second group consists of materials either made or modified by humans, for example, carbon-based materials and nanomaterials or some modified materials like chitosan [91–93].

3.4.1. Natural materials

The most often used sorbent materials mentioned in the literature are soils of different parameters, sewage sludge, and peat and river or marine sediments [89,94–97,103]. Apart from the previously mentioned, mineral sorbents like kaolinite, gibbsite, Na-montmorillonite, perlite, vermiculite, and limestone are found in the literature [97–100]. Apart from the materials mentioned above, Björklund et al. [97] used some tree bark or modified sawdust for experiments.

3.4.2. Man-made or modified materials

Carbon-based materials play an important role in this group. Several studies present results of sorption studies with the application of different carbon materials like microporous carbon, carbon nanotubes, and activated carbon powder [91,92,101]. Other possibilities are materials like chitosan derivatives [93].

3.4.3. Sorption kinetics

Liao et al. [95] present some kinetic study results. Experimental data for NP sorption are fitted to four different models and correlation coefficients (R^2) were compared. The two-compartment first-order model provided the best fit to experimental data for all studies of sorption materials. Zhou et al. [96] present the results of kinetic studies for all used materials. Studies indicate that, regardless of the material, BPA removal follows pseudo-second-order kinetics. Similar results are obtained for sorption studies of nonylphenol on marine sediments presented in [89]. In this study, pseudo-second-order kinetics provide better agreement with experimental data.

3.4.4. Sorption isotherms

The Freundlich isotherm was selected as the most suitable for most cases regarding sorption material or sorbed molecule [89,90,92,98,101–103]. In studies [98], it is determined that the Langmuir isotherm provides better agreement with experimental data for all studied compounds except nonylphenol. However, the negative value of the $q_{\max}$ parameter for most compounds in the Langmuir equation indicates that this equation is insufficient to explain the sorption process.

3.4.5. Effect of pH

For the sorption of BPA, no significant changes were detected at pH ranges from 2 to 8 [88,90,92,]. Zhou et al. [96] study BPA sorption of some natural materials and establish that at pH 8 and above, sorption decreases with pH increase. This phenomenon is explained based on the pKa value for BPA (9.9–10). Zhou et al. state that at high pH, BPA turns into anionic form, which causes electrostatic repulsion between the negatively charged sorbent and the sorption material surface. Similar behavior is observed by Joseph et al. [92]. Joseph et al. study the sorption of BPA on single-walled carbon nanotubes. As in the previous example, electrostatic repulsion is suspected to decrease removal. Clara et al. [90] present data for BPA sorption on sewage sludge. At pH 9

Table 6
Degradation degrees of AP and BPA in oxidation processes

Degradation agent	Time (min)	BPA (%)	4nNP (%)	4tOP (%)	Reference
$H_2O_2/Fe^{2+}/UV$	40	–	90	–	[70]
UV/Fe^{2+}	90	100	–	–	[71]
UV–C photolysis	120	52	–	–	[76]
H_2O_2/UV–C	20	100	–	–	
S_2O_8/UV–C	10	100	–	–	
$Na_2S_2O_8/UV/Fe^{2+}$	120	97	–	–	[78]
Ultrasound-assisted electrochemical oxidation	30	90	–	–	[80]
Photodegradation Fe(III)	60	–	–	80	[81]
ZnO/UV/NaOCl	60	99.9	–	–	[82]
SnO_2/UV/NaOCl	60	94.3	–	–	
TiO_2/UV/NaOCl	60	98.1	–	–	
MnO_2	90	–	92	84	[86]
Photocatalytic degradation/TiO_2	45	–	–	91.2	[87]

and above, the desorption process is observed, and at pH 12 almost all sorbed BPA is released. Bouki et al. [102] study sorption of nonylphenol on activated sludge. Results indicate that the desorption process starts above pH 11. This behavior is explained based on nonylphenol pKa values (10.28). At pH 11 and higher, nonylphenol becomes ionized, hence more hydrophilic, which can cause migration to the water phase.

4. Conclusions

AP and BPA are in an important group of compounds due to their adverse effects on both human and wildlife health, especially when it comes to reproduction issues. Another important factor is the worldwide occurrence of AP and BPA. Scientists from all over the world provide results of BPA and AP in different water-based matrices like wastewater, river, lakes, seas, and oceans. These results indicate that present methods of wastewater treatment are insufficient and have room for further improvement. In the course of this paper, methods of BPA and alkylphenol removal were divided into four different categories. Membrane processes are commonly used to remove a wide range of inorganic pollutants from drinking water [48]. So, it is only natural to apply this process to remove AP and BPA. Studies conducted indicate that the highest results are obtained for nanofiltration membranes [48–54]. The reported retention reaches almost 100%. However, the vast majority of studies are conducted at the laboratory scale. In biological methods of BPA and AP degradation, removal reaches up to 90% [66] depending upon the microorganism. An important drawback is the time of the process, which varies for 50 min [65] up to 14 d [69]. There is also the case of metabolites and excessive biomass formation in the course of the process. Advanced oxidation processes pose some alternatives to previously mentioned methods. In this case, the process lasts about 60 min [81,82]. In some cases, the time is reduced to 10 min [76]. Also, AP BPA removal varies from 80% to almost 100% [70,71,76,78, 80–82,86–87]. The drawback of this approach is the use of external reagents. The absorption process gives the opportunity to apply natural materials as sorbents for AP and BPA removal like, for example, soil, sediments of different origin, peat or sludge [89,94–98,104]. An important drawback of the presented methods is scale. The vast majority of studies are conducted at the laboratory scale. Only a few are conducted in pilot plans. To improve water quality, further studies should take into consideration the possibility to realize processes at a large plant scale. The second field of studies is the development of new compounds that are less toxic and can substitute AP and BPA.

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