Endocrine Disrupting Compounds in Water Environment

Subjects: Environmental Sciences

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Endocrine disrupting compounds (EDCs) are a particularly dangerous group because they have estrogenic activity. Among EDCs, the alkylphenols commonly used in households deserve attention, from where they go to sewage treatment plants, and then to water reservoirs.

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1. Introduction

Environmental pollution has become a major challenge in recent years due to increasing population, urbanization, and industrialization [1]. The priority is to find the sources of EDCs and the routes by which they enter the aquatic environment.

EDC has been detected in a variety of fresh, brackish, and marine ecosystems. Due to their physical and chemical properties, EDCs can bioaccumulate, biomagnify, are persistent, and are very toxic to aquatic organisms, both for plants and animals [2].

These compounds are released into the environment from a variety of sources, primarily municipal and industrial waste, agricultural practices, animal waste, and sewage treatment plants (STP) [3]. Most of the packaging for food, cosmetic products, solvents, preservatives, and pesticides is also made of EDC-containing materials [2][4][5].

Long-term use of EDC, regardless of the concentration level, may accumulate in animals and be partially released into the environment through animal feces. In fish and the largest consumers of the food web, the rate of bioaccumulation is higher because most EDCs are lipophilic and concentrated in the fat of the consuming organisms [6]. Therefore, these substances will penetrate the food chain and ecosystems, potentially adversely affecting human health.

In agriculture, non-metabolized and non-degradable compounds in animal fertilizers still have active metabolites, reducing the quality of surface and groundwater and significantly affecting aquatic life. In water, EDCs undergo biodegradation and chemical and photochemical degradation, dilution, and sorption to sediments, which partially leads to their elimination from aquatic ecosystems [2][7][8][9].

2. Nonylphenol

The presence of nonylphenol in the environment is clearly correlated with anthropogenic activities such as sewage treatment, storage, and recycling of sewage sludge. NPEOs are used as non-ionic surfactants in industry (cellulose and paper, textiles, agriculture, metals, plastics, petroleum refining), in households in the form of detergents, solubilizers, and personal care products in non-EU countries [10][11]. Nonylphenol is a xenobiotic compound used in the production of antioxidants, additives to lubricating oils and in the production of ethoxylated surfactants nonylphenol, which is its main use (65%) [12].

As surfactants, NPs are used in cleaning agents, so their primary source in the environment are discharges of wastewater from industrial and municipal wastewater treatment plants (WWTPs), as well as land enriched with solid sewage or manure, runoff from pesticides and fertilizers on agricultural fields, and fodder livestock. Due to their cheapness, NPEO surfactants are used in various areas, for example in agricultural pesticides where surfactant is added to control the properties of pesticides [13][14].

Inadequately treated domestic sewage causes high concentrations of NP in the aquatic environment. The levels of NP depended on the size of NP discharges into the river, temperature, flow velocity, biodegradation, etc. About 60% of NP and its derivatives produced in the world ends up in water supply [15][16][17]. In addition, the presence of NP is observed in polyvinyl chloride (PVC), which can contaminate water passing through PVC plumbing [18].

Technical NP is the major form of NP that is released into the environment. Technical nonylphenol consists of a mixture of more than one hundred isomers that have an alkyl moiety attached at various positions on the phenolic ring, with para-substituted NP (4-NP). However, in the environment, the proportions of isomers may be different [13][19][20].

Due to its high hydrophobicity, resistance to biodegradation and low solubility, NP tends to accumulate in various environmental matrices [21]. NP can evaporate into the atmosphere from wastewater discharges, wastewater treatment plant (liquids and sludge) or heavily contaminated surface waters. NP binds to the aerosols generated by the wastewater treatment plant, leading to a reduction in air quality in the vicinity of STW. From the atmosphere, NP may re-enter aquatic and terrestrial ecosystems with rain and snowfall [12].

The concentration of nonylphenol in the surface layers of natural waters may decrease due to photolysis induced by sunlight [7][22]. Biodegradation of NPs is difficult due to physicochemical properties such as low solubility and high hydrophobicity. NP accumulates in environmental compartments that are characterized by a high content of organic substances, usually sewage sludge and river sediments. NP occurs in river waters in concentrations up to 4.1 µg/L and 1 mg/kg in sediments [15][16][23]. The concentration of NP in the water and sediments are shown in **Table 1**.

Table 1. The Bisphenol A (BPA) and nonylphenol (NP) concentrations in aquatic environments.

Environment	BPA	NP	Location	References
Surface water	nd–376.6	117–865	China,	[24][25]

Environment		BPA	NP	Location	References
				Pearl River Delta	
		15.80–110.38	nd–104.02	China, Honghu Lake	[26]
		16.3–30.1	-	Poland, Vistula river	[27]
		0.71–47.40	-	Philippines, Laguna Lake	[28]
Groundwater		nd–35.54	5,6	China	[29][30]
		1–629	-	Poland	[27]
Wastewater treatment plants	IW	nd–5927.5	nd–9560	Poland	[27]
	EW	nd–190	nd–9560		
	IW	234.6–1527.1	519.6–4183.4	China	[31]
	EW	3.1–623.6	13.4–471.6		
Seawater		10.6–52.3	22–201	Greece, Thermaikos Gulf, Northern Aegean Sea	[32]
		240	730	Japan, Tokyo Bay coastal area	[33]
Sediment	W	7.02–13.95	-	China, Erhai Lake	[34]
	S	11.09–63.46	-		
	W	-	75.4	Germany, Luppe River	[35]
	S	-	22,342.6		
	W	nd–37,000	-	Indonesia, Mahakam River	[36]
	S	nd–952.6	-		
	W	<1.3–5.2	nd	New Zealand, Lyttelton Harbour	[37]
	S	<0.4–9.9	nd		
	W	5.0–277.9	4.0–1721	Poland, Gulf of Gdańsk (southern Baltic Sea)	[38]
	S	2.64–60.20	0.08–1001		

The presence of NP in surface waters is correlated with anthropogenic activity and wastewater discharge from STW, industrial plants, municipal wastewater treatment plants, and rainwater discharges [12][40]. NP concentrations in rivers are subject to seasonal fluctuations with higher concentrations in summer due to increased microbial

Environment	BPA	NP	Location	References
Drinking water	6–53	-	Poland	[27]
	2.6–6.2	1.7–3.9	Serbia, rural area	[39]
	2.5–35.6	1.2–7.9	Serbia, urban area	[39]

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Groundwater is of particular interest as it accounts for about twenty percent of the world's freshwater supply and is extremely susceptible to contamination by various pollutants as a result of urban activities. NP concentrations in groundwater were incredibly low [44][45][46]. The microbial decomposition of NP in aquifers is limited due to the low temperature conditions prevailing there: low temperature, low C, and low O_2 content). The processes controlling the entry of pollutants into groundwater are sorption and biodegradation [12][15].

nd—not detected; IW—influent; EW—effluent; W—water; S—sediment.

Ethoxylates nonylphenol (NPEO) ends up in significant amounts in wastewater treatment plants, where they biodegrade into several by-products, including nonylphenol, which is more resistant than the parent compound [12][47]. NP is the major degradation product, does not undergo further transformation, and is highly adsorbed in the sludge; therefore, it is often found in higher concentrations in wastewater than in tributaries [12][47][48]. In most cases, it is the strong sorption of pollutants, and not the microbiological activity, which limits the rate of biodegradation [49]. On the rate of biodegradation NPEO, the main sources of NP in WWTPs, is affected by many factors e.g., temperature, NP isomer present in the environment, oxygen availability, pH and additives of yeast extracts, surfactants, aluminum sulphate, acetate, pyruvate, lactate, manganese dioxide, iron chloride, sodium chloride, hydrogen peroxide, heavy metals, and phthalic acid esters [50][51][52].

Conventional physicochemical wastewater treatment methods have not been shown to be effective in removing endocrine disruptors such as NPs due to their low molecular weight [1][53]. Novel purification techniques, including advanced oxidation methods, UV [54], adsorption (powdered activated carbon (PAC), and granular activated carbon (GAC)), ion exchange and membrane filtration (ceramics, polymers, and zeolites) are still under analysis [55][56].

One of the other methods of NP removal is to use cells and enzymes. Tanghe et al. in 1999 first described the *Sphingomonas* strain that degraded NP [57]. Since then, microorganisms involved in the biodegradation of NPs in the aquatic environment have been discovered (*Sphingobium, Pseudomonas, Pseudoxanthomonas, Thauera, Novosphingonium, Bacillus, Stenotrophomonas, Clostridium, Arthrobacter, Acidurvorax, Rhizobium, Corynebacterium, Traynebacterium, Rhodococcus, Candida, Phanerochaete, Bjerkandera, Mucor, Fusarium and Metarhizium*) [13][52][58][59][60][61]. NP-degrading microorganisms were isolated from municipal sewage treatment plants from sludge, sewage sludge and activated sludge under aerobic and anaerobic conditions. Appropriate pH value, temperature, and the level of aeration of sewage sludge contribute to the increase of microbiological activity, and thus increase the degradation of NP.

Anaerobic degradation of NP has only recently been shown. Half-lives of anaerobic degradation ranged from 23.9 to 69.3 days. The rate of anaerobic degradation of NP was enhanced by increasing the temperature and adding yeast extract or surfactants [15][43][62]. Bacterial strains are effective in improving NP biodegradation and short-chain fatty acid accumulation. The species *Propionibacterium*, *Paludibacter*, *Proteiniphilum*, *Guggenheimella*,

Lactobacillus, *Anaerovorax* and *Proteiniborus* correlated with the short-chain fatty acids synthesized as a result of NP degradation [59][63].

Some bacteria and fungi produce laccase, a multifunctional enzyme. Laccases are well-known enzymes that have found application in bioremediation, both as free and immobilized enzymes. Thanks to their broad range substrate specificity, laccases are able to remove xenobiotics, including EDCs. Therefore, the white rot fungi (WRF) are a promising tool in the above-mentioned elimination of EDCs during wastewater treatment processes [64]. The advantage of fungi over bacteria in lignin mineralization results from the production and secretion of laccase-non-specific enzyme outside the cell, which gives fungi access to non-polar and insoluble substances, operation over a wide range of temperatures and pH values, and the developing fungal hyphae also make it possible to reach contaminants inaccessible to bacteria [64][65][66]. The laccase from WRF like *Pleurotus eryngii*, *Trametes versicolor* or *Phanerochaete chrysosporium* can adhesion to alkylphenols, because these compounds have functional groups such as amino, hydroxyl, or alkyl groups in its chemical structure, acting as electron donors for oxygenases [64][67][68][69][70][71].

3. BPA

BPA is a monomer in the production of polycarbonate plastics and epoxy resins, and as an additive in the production of PVC coatings is added to various plastics as a plasticizer. These materials are used in food storage containers, water, baby bottles, food, and drink cans. Under the influence of elevated temperature, BPA may migrate from containers to food and beverages [4][72]. BPA-derived monomers, especially bis-GMA (methacrylate bisphenol–glycidyl) are used in dental materials, from where they can be released [73].

The release of BPA to the aquatic environment takes place in several ways, including from production plants, from wastewater as a result of incomplete treatment, or in physicochemical and biological processes in treatment units, from leachate from landfills, as well as from leaching from discarded BPA-based products [24][74][75][76][77]. The problem is also sludge from recycled paper, the production of which uses BPA as a reactive agent. These sediments are used as fertilizers in agriculture, eventually contaminating the groundwater with BPA [78]. BPA also enters groundwater through its release to landfill leachate [44][79][80][81][82].

The results showed that the leaching of BPA from polyethylene microplastics (LDPE) and polycarbonate (PC) was 2.68 µg/g and 14.45 µg/g, respectively [83]. Studies have shown that diffusion of BPA in the environment is related to the hydrolysis of PC: processing time for polycarbonate bottles in the presence of water is only a few (3–7) years [84].

BPA concentrations in surface water vary depending on the location, sampling period and depth of sampling. Studies in which both water and sediment were collected indicate significantly higher BPA concentrations in the sediments than in the upper water column, which is related to the slowing down of biodegradation processes in anaerobic environments [79][85][86][87]. Rivers in Europe and North America with higher BPA concentrations detected are commonly associated with production facilities [84][86]. Although BPA dissolved in surface water has a short half-

life due to photo- and microbial degradation, its metabolites can persist much longer [88]. The BPA metabolites can be toxic for aquatic organisms [76]. Like BPA, its metabolites are xenoestrogens, e.g., 4.40-dihydroxy-methylstilbene and 4-methyl-2,4-bis (4-hydroxyphenyl) pent-1-ene (MBP), whose estrogenic activity levels exceed BPA levels respectively 40- and 300-fold [89][90].

Observed BPA concentrations in oceans and estuaries are low compared to freshwater systems (**Table 1**). It is related to the fact that sewage from municipal and mixed municipal-industrial wastewater treatment plants is the main source of environmental BPA, and its runoff goes to rivers [74][85][91][92][93][94]. BPA is leached faster in marine systems than in freshwater systems [95][96], and microbial degradation may be slower [97][98]. Moreover, the bioavailable fraction of dissolved BPA may increase with salinity [96].

The poor biodegradability of BPA in nature leads to the contamination of surface and groundwater. Researchers can remove BPA using the following methods: photodegradation (TiO_2) is the most commonly studied and uses photocatalyst, adsorption (natural adsorbents, carbon and graphene, clay, nanomaterials, and composite materials), biodegradation with the use of microorganisms, or phytoremediation [9][99][100][101][102].

The main source of microorganisms in biodegradation is activated sludge from WWTPs. *Bacillus thuringiensis*, *Pseudomonas putida* YC-AE1, *Sphingomonas paucimobilis* FJ-4, *Lactococcus lactis*, *Bacillus subtilis* and many more bacteria are capable to use BPA as substrate in their metabolism [84][103]. Among fungi, BPA degradation was observed in *Saccharomyces cerevisiae* and WRF [104][105]. WRF converts BPA into a much less reactive substance in enzymatic oxidation. The oxidized form of BPA does not bind to ERα dependent estrogen receptors. This may be due to laccase oxidation of both BPA hydroxyl groups. The product obtained by laccase catalyzed oxidation of BPA is 2,2-bis (4-phenylquinone) propane [106].

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