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Editorial

Removal of Organic Pollution in the Water Environment

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Abstract: The development of civilization entails a growing demand for consumer goods. A side effect of the production and use of these materials is the production of solid waste and wastewater. Municipal and industrial wastewater usually contain a large amount of various organic compounds and are the main source of pollution of the aquatic environment with these substances. Therefore, the search for effective methods of wastewater and other polluted water treatment is an important element of caring for the natural environment. This Special Issue contains nine peer-review articles presenting research on the determination and removal of environmentally hazardous organic compounds from aqueous samples. The presented articles were categorized into three major fields: new approaches to the degradation of water pollutants, new methods of isolation and determination of the emerging organic contaminants (EOCs), and the occurrence of EOCs in the water environment. These articles present only selected issues from a very wide area, which is the removal of organic pollution in water environment, but can serve as important references for future studies.

Keywords: emerging organic contaminants; water environment; EOCs determination; wastewater purification; advanced oxidation processes; electrochemical degradation; biosorption; liquid-liquid continuous extraction; fractional distillation

1. Introduction

Water is essential for life, and although approximately 70% of the Earth' surface is covered with water, only a small fraction (2.5%) is freshwater compatible with terrestrial life. Nowadays, a continuous increase in water demand is observed as a consequence of demographic growth, industry demand, and living conditions. At present, the societies of developed countries are aware of the importance of water quality, especially in western countries. It is a matter of concern that half of the European countries are already facing water stress. According to the European Environmental Agency Report, only around 40% of surface waters (rivers, lakes, and transitional and coastal waters) are in good ecological status or potential, and 38% are in good chemical status [1]. An intensive use of chemicals in everyday activities and unrestricted access to medicines has resulted in increased waste production and an intense emission of typical as well as new organic compounds into the surrounding environment. In recent years, the newly occurred compounds, called emerging organic contaminants (EOCs), are becoming more and more observable. This is very heterogeneous group of substances containing compounds from various chemical groups. They are created by natural as well as anthropogenic compounds with a presence that was not previously detected due to the lack of sensitive analytical methods, or their adverse health effects were not known. Their impact on living organisms is in general unknown, but the provided experiments have proved their negative influence on vitality, life span, and reproductive success [2,3]. Some of them exhibit disrupting endocrine effects or are suspected to cause it. This group of compounds is distinguished by a separate group named the endocrine disrupting compounds (EDCs) [4]. Their presence in the environment arouses special concern because they change the hormonal equilibrium not only of wild organisms but also of humans. The EDCs are very ubiquitous in every element of environment such as surface and ground waters, soil, and air. Despite their presence at low concentrations, they are considered as persistent due to their continuous delivery to the environment.

A number of EOC emission sources have been identified, but discharges of effluents from wastewater treatment plants (WWTPs) are considered as the main ones. These substances are present in all tested wastewater, both before and after the treatment process, usually in concentrations ranging from ng/L to µg/L [5]. The composition and concentration of EOCs in waters supplied to WWTP depend mainly on the socioeconomic characteristics of the population from which wastewater is collected. The concentration of EOCs in the effluents after the purification process depends on both the level of pollution of the incoming waters and the course of the purification process. The concentration ranges of selected groups of EOCs in raw and treated wastewater are presented in Table 1.

Table 1. Concentrations of compounds from the main groups of emerging organic contaminants (EOCs) recorded in urban wastewater and the efficiency of their removal in conventional wastewater treatment plants (WWTP) (based on ref [5–11]).

EOC Group	Concentration Range		Removal Range (%)
	Influent (ng/L)	Effluent (ng/L)	
Hormones	<MQL*– 670	<MQL−275	0–100
Plasticizers	<MQL−5850	<MQL−1840	32–100
Insect repellents	<MQL−42334	<MQL−1663	27–100
UV filters	<MQL−7800	<MQL−772	0–97.5
Surfactants	<MQL−8520	<MQL−3200	42–99
Antimicrobials	<MQL−8880	<MQL−5860	0–100
NSAIDs	<MQL−611000	<MQL−62000	0–100
Antibiotics	<MQL−303500	<MQL−37000	0–100

* MQL—Method Quantification Limit.

The fate and effectiveness of removing organic pollutants are closely related to their physicochemical properties (Henry's constant (H), n-octanol/water partition coefficient (K_{ow}), sorption coefficient (K_D), partition coefficient between soil organic carbon and water (K_{OC})) [12]. Traditional municipal WWTP applied two stages of treatment: mechanical and biological; the third stage with advanced technologies is rarely used. The purpose of the mechanical stage is to remove suspended matter by filtration, sedimentation, and flotation processes. In this stage, the adsorption of pollutants on the suspension particles and absorption in the fats present in the wastewater takes place. During mechanical treatment, hydrophobic compounds (log $K_{ow} > 3$) undergo partial removal from wastewater, while the efficiency of hydrophilic compound expurgation is very low [13,14]. Biological treatment is usually carried out using the conventional activated sludge (CAS) method under aerobic and anoxic conditions. Removal of organic pollutants at this stage is associated with biodegradation (biotransformation) and sorption on activated sludge [12]. Biodegradation can occur through metabolism or co-metabolism. In order for metabolism to be possible, compounds must have low toxicity, and their concentration must be high enough to support the life processes of microorganisms [15,16]. Co-metabolism is the degradation of organic pollutants by microorganisms when using other substances as a source of nutrients [17]. It is the basic mechanism of biodegradation of EOCs due to their very low concentrations in wastewater. Acid and lipophilic compounds are biodegradable to a much greater extent than polar, neutral, and basic compounds [18]. Removal of organic pollutants from municipal wastewater can also take place by means of volatilization, i.e., transformation from the form dissolved in water to gaseous. The intensity of this process depends on the H value and WWTP operating conditions (intensity of aeration and mixing of wastewater, temperature, and atmospheric pressure).

Conventional wastewater treatment methods are usually not sufficiently effective in removing EOCs. There is demand for the use additional processes like precipitation and chemical coagulation, flocculation, desorption, neutralization, and reverse osmosis for more thorough purification. Unfortunately, these methods, in many cases, are not sufficiently effective in eliminating pollution, or the economical side of carrying out prevents them from being used on a larger scale wastewater treatment. Additionally, the application of physico-chemical processes causes a transfer of EOCs from the water phase to the receiving material or solid phase, which are new wastes, the management of which creates new environmental problems. Another approach is applied by the so called advanced oxidation processes (AOPs) [19]. They are based on the oxidation of organic pollutants by strong oxidants, mainly hydroxyl radicals generated by ozone, hydrogen peroxide, and others [19]. The best oxidation results are achieved in synergistic processes using systems consisting of two or three components, e.g., H_2O_2/UV or O_3/H_2O_2/UV [19,20]. The use of such systems improves the cleaning effects by increasing the efficiency of mineralization of EOCs and reducing the amount of products of incomplete oxidation [21].

2. Overview of the Special Issue

The Special Issue consists of nine papers describing a wide spectrum of research related to the removal of environmentally significant pollutants from aqueous samples and their determination in various matrices. The presented articles can be classified into three broad thematic sectors related to the topic of the special issue: new methods of isolation and determination of compounds from the EOCs group [22–24], occurrence of EOCs in the water environment [24,25], and new approaches to the degradation of significant water pollutants or their removal [26–30].

2.1. New Methods of EOCs Isolation and Determination

Mielech-Łukasiewicz and Starczewska [22] proposed a new electrochemical method for determining the pharmaceutical residues in aqueous samples. The target pharmaceuticals were two compounds from the fungicide group: itraconazole and posaconazole. Cyclic voltammetry and square wave voltammetry with the use of boron-doped diamond (BDD) electrode were used for determining the properties of analytes and in their analytical characterization. The developed method is simple, fast, and sensitive, and its significant advantage is that there is no need to isolate the analytes from the matrix before the determination. The research carried out for river water and tap water samples indicate that the proposed method can be used in the analysis of environmental samples as an alternative to chromatographic methods, which are most often used in EOC determination in natural waters and wastewater [5]. Hryniewicka et al. [23] described the use of high-performance liquid chromatography with ultraviolet detection (HPLC-UV) for the determination of two pharmaceuticals, budesonide and sulfasalazine, in water and wastewater. For the isolation of target compounds from aqueous samples, dispersive liquid-liquid microextraction (DLLME) followed by solidification of floating organic droplet (SFOD) was used. The paper presents the optimization of extraction parameters, such as the type of extraction solvent, pH, and sample ionic strength as well as extraction time. Analysis carried out for spiked samples of river water and municipal wastewater confirmed the usefulness of the proposed method in aquatic environment research. A new, simple, and sensitive method for determination of three hormones (β-estradiol, estrone, and diethylstilbestrol) and ten other compounds from the EOC group (diclofenac, triclosan, propylparaben, butylparaben, benzophenone, 3-(4-methylbenzylidene)camphor, N,N-diethyltoluamide (DEET), bisphenol A, 4-t-octylphenol, and 4-n-nonylphenol) was proposed by Kotowska et al. [24]. Isolation of analytes by ultrasound-assisted emulsification microextraction with solidification of floating organic droplet (USAEME-SFOD) was done simultaneously with derivatization with acetic anhydride to enable determination of EOCs using gas chromatography mass spectrometry (GC-MS). Good accuracy and precision as well as high sensitivity of the developed method enabled its use for natural water samples.

2.2. Occurrence of EOCs in the Environment

In addition to the new method, Kotowska et al. [24] present the results of the determination of thirteen EOCs in groundwater collected at municipal solid waste (MSW) landfill sites and in groundwater from wells distant from sources of pollution. Ten compounds were detected in groundwater from MSW monitoring wells. Five compounds were detected in shallow groundwater wells (depth: 3–8 m), and two compounds in deep drilling wells (depth: 15–46 m). Ferrari et al. [25] described in their paper a well-documented study of the occurrence and concentration in aquatic sediments of ten congeners of polybrominated diphenyl ethers (PBDEs). PBDEs belong to the group of flame retardants and are used for reducing the risk of fires. These compounds are persistent organic pollutants from the EOC group and are toxic to living organisms, including humans. In the presented study, the isolation of PBDEs from sediment samples was done by ultrasound-assisted solvent extraction followed by gas chromatography with electron capture detection (GC-ECD). Six out of ten target compounds were detected in sediment samples taken from Guarani Aquifer in Brazil in concentrations ranging from 0.24 to 2.7 ng/g. According to the authors, pollution of the examined water reservoir with compounds from the PBDEs group is associated with improper management of solid municipal waste.

2.3. New Approaches to Degradation of Water Pollutants

The removal of platinum and palladium from environmental samples by biosorption on fungi *Aspergillus* sp. and yeast *Saccharomyces* sp. was described by Godlewska-Żyłkiewicz et al. [26]. The introduction of these metals into the aquatic environment is mainly associated with the production, use, and recycling of automotive catalysts. Optimization of biosorption parameters such as solution pH, biosorbent mass, and contact time of the solution with the extraction medium was performed to determine the conditions in which the sorption efficiency is highest. The sorption kinetics was tested, and the Langmiur and Freundlich adsorption isotherms were used for interpretation of the process equilibrium. The research conducted shows that the proposed microorganisms can be successfully used to remove platinum and palladium from contaminated waters and industrial waste. Ambauen et al. [27] investigated electrochemical oxidation of the organic model pollutant salicylic acid. Two anode materials, platinum and boron doped diamond, were used along with chloride and sulfate electrolytes. The work presents a detailed kinetics analysis and identification of oxidation process products. Studies have shown that the products of salicylic acid electrochemical oxidation are hydroxylated and chlorinated intermediates, and the dominance of one of the forms depends on the composition of the electrolyte used. The best results of electrochemical degradation of salicylic acid were achieved where the combination of BDD electrode and chloride electrolyte was used, and the worst results were achieved when a platinum electrode was placed in the same electrolyte. A very low oxidation efficiency of the test compound was observed when the sulfate electrolyte was used in combination with both BDD and platinum electrodes. Karpinska et al. [28] describes detailed studies on the kinetics of the degradation processes of doxazosin (DOX) under the influence of sunlight in environmental conditions and some advanced oxidation processes (AOPs). Doxazosin is a biologically active compound used for the treatment of some prostate complaints and hypertension. The authors checked DOX photochemical behaviors and stated that it is a photoliable compound and its degradation is a result of a direct photolysis. Its $t_{1/2}$ in the presence of a natural matrix lasted from 1 h 30 min to 40 min depending on the chemical composition of the samples of surface water. The studies on DOX behavior under the influence of examined chemical and photo-chemical processes (UV/H_2O_2, Fenton and photo-Fenton process, and $SO_4{\cdot}^-$ radicals) were performed. It was stated that $SO_4{\cdot}^-$ radicals are most efficient and caused DOX degradation in a very short time. The application of a new photocatalyst for the degradation of a selected organic compound was proposed by Regulska et al. [29]. The authors examined the photochemical properties of crystalline $NiAl_2O_4$ decorated with graphene quantum dots. They characterized morphology and structure of a synthetized composite using thermogravimetric methods as well as spectral techniques (XRD, ATR-FTIR, SEM, EXD, and UV-Vis diffuse reflectance spectra). Its photocatalytic activity in ratio to chosen model pollutants (rhodamine B, quinolone yellow, eriochrome black, methylene blue, phenol, and thiran) was studied. It was stated that newly obtained material exhibits photocatalytic activity under the influence of visible light. The detailed mechanism of its operation was proposed and discussed. Its efficiency strongly depends on the presence of electron and hole scavengers and the chemical properties of adsorbed organic compounds. The above articles [27–29] concern the application of chemical processes for removal of organic pollutants. Another approach to wastewater cleaning was proposed by Mendoza et al. [30]. The authors focused on the problem of cleaning wastewater generated by the petroleum industry. They proposed the use of continuous liquid-liquid extraction with dichloromethane ($CLLE_{DCM}$) and high-power fractional distillation (HPFD) to resolve this problem. The efficiency of $CLLE_{DCM}$ and HPFD was examined individually and in combination: $CLLE_{DCM}$-HPFD and HPFD-$CLLE_{DCM}$. The chemical parameters of wastewaters were checked. It was stated that all processes remarkably improved the quality of the samples used. The greatest achievements were obtained by HPFD.

3. Conclusions

The presented Special Issue concerns the problem of the appearance of new organic pollutants in surface water bodies. At present, it is obvious that the chemical composition of the surface water is the result of industrial, agriculture, as well as domestic activities of human population. As it mentioned in the introduction, the wastewater treatment plants have been identified as the one of the main sources of organic pollutions. Thus, much more effort should be made for the improvement of wastewater cleaning processes. The authors involved in the preparation of this Special Issue described the results of examinations, at a laboratory scale, of the efficiency of chemical as well as physical processes for the removal or degradation of selected model pollutants. However, it should be noted that extension of the proposed processes for technological scale requires intense additional studies. The environmental studies, especially those concerning the determination of trace impurities, require effective isolation and concentration procedures. The reagents used for this purpose should meet the requirements of green chemistry. The DLLME-SFOD as well as USAEME-SFOD procedures described in this Special Issue seem to be proper for environmental studies as they are effective and environmentally friendly. Another approach is based on the use of BDD electrodes for direct determination of the target analyte in environmental samples. The described method allowed an assay of examined pharmaceuticals without their isolation from liquid samples.

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