

PURIFICATION OF WASTEWATERS CONTAINING ENDOCRINE DISRUPTING COMPOUNDS BY COAGULATION

RENÁTA MÉSZÁROS¹–SÁNDOR BÁRÁNY¹

The paper represents a review of literature data on treatment of wastewaters containing endocrine disrupting compounds (EDC), first of all estrogenic hormones, which have been detected in influents and effluents of sewage treatment plants (STP), surface and drinking waters in many countries. A short description of typical representatives of EDCs, their composition and sources of origin is given. The current data and results of EDC/PPCP (pharmaceutical and personal care products) removal during conventional water treatment in surface water treatment plants (SWTP) are summarized. Conventional SWTPs

typically treat water by coagulation using alum, ferric chloride, and/or synthetic polymers followed by flocculation, sedimentation, filtration, and disinfection. Alternative SWTP designs or modifications of conventional WTPs include additional water treatment processes, e.g., use of activated carbon, biofiltration, membranes, aeration, chemical softening, ultraviolet light irradiation. The efficiency of removal of EDCs and first of all of their most dangerous representatives bisphenol A and nonyl-phenol using coagulation by aluminium sulphate and polyaluminium chloride is considered in detail.

Keywords: endocrine disruptor; pharmaceutical; coagulation, water treatment; review.

Introduction

For over 70 years, scientists have reported that certain synthetic and natural compounds could mimic natural hormones in the endocrine systems of animals. These substances are now collectively known as endocrine-disrupting compounds (EDCs), and have been linked to a variety of adverse effects in both humans and wildlife. More recently, pharmaceuticals and personal care products (PPCPs) have been discovered in various surface and ground waters, some of which have been linked to ecological impacts at trace concentrations. The majority of EDCs and PPCPs are more polar compounds, than traditional contaminants and several have acidic or basic functional groups. These properties, coupled with the occurrence at trace levels create unique challenges for both removal processes and analytical detection. Reports of EDCs and PPCPs in water have raised substantial concern among the public and regulatory agencies; however, very little is known about the fate of these compounds during drinking and wastewater treatment [1].

Estrogenic hormones have been detected in influents and effluents of sewage treatment plants (STP) in many countries [2], surface water [3], as well as drinking water [4]. Considering the widespread occurrence and potential impacts of EDCs, it is highly important to remove them before discharge. The current data suggest that wastewater treatment processes have variable performance in removing EDCs.

¹ University of Miskolc, Institute of Chemistry
3515 Miskolc-Egyetemváros, Hungary
akmsab@uni-miskolc.hu
fkmmr@uni-miskolc.hu

It is generally accepted that the three major classes of endocrine disruption endpoints are estrogenic (compounds that mimic or block natural estrogen), androgenic (compounds that mimic or block natural testosterone), and thyroidal (compounds with direct or indirect impacts to the thyroid) [5]. The majority of research has focused on estrogenic compounds. The most frequently studied structures of EDCs can be seen on Figure 1.

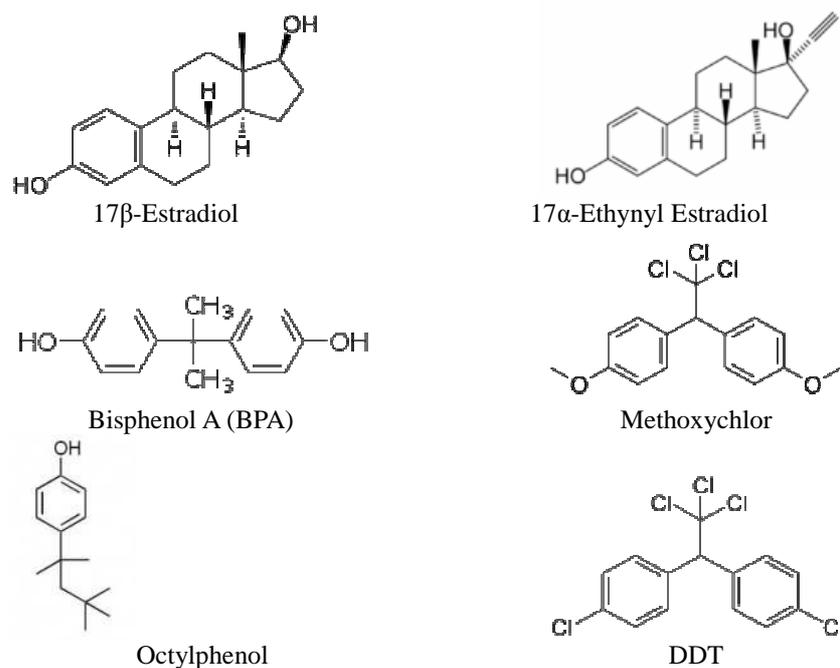


Figure 1. Structures of the most frequently studied estrogenic compounds

Endocrine disruption also can be caused by naturally occurring compounds. For example, estrogens from plant sources, known as phytoestrogens, have been linked to reproductive failures in animals since the 1930s [6].

Some scientists have suggested that some drinking water disinfection byproducts may act as EDCs.

Many of the thousands of anthropogenic chemicals currently released into the environment are endocrine-disrupting compounds (EDCs); a typical representative of them is bisphenol-A (see Figure 1). It has become one of the most dangerous pollutant in the environment within the past 80 years because of its presence in a multitude of products including food and beverage packaging, flame retardants, adhesives, building materials, electronic components, and paper coatings [7].

There are a number of papers as well as reviews devoted to problem of purification of drinking and industrial wastewaters from endocrine disrupting compounds. The most efficient methods are adsorption on activated carbon and oxidation by ozone and chlorine [8]. At the same time coagulation by multivalent salts hydrolysis products and/or adsorption

on freshly precipitated hydrolysis products particles could serve as an alternative or additional method of EDCs removal taking into account that coagulation is an obligatory stage of water conditioning in the majority of waterworks. Combination of coagulation with adsorption can give good results in the process of treatment of waters containing EDCs and PPCPs. Our paper gives a review of updated results in this area.

1. EDC/PPCP removal during conventional water treatment

Some of the most representative PPCPs found in Sewage Treatment Plants (STP) are antibiotics, lipid regulators, anti-inflammatory agents, tranquilizers, contrast media and contraceptives with very different chemical structures.

Most of these compounds come either from domestic sewage or from hospital as well as industrial discharges and enter municipal STPs. Modern STPs can effectively accomplish carbon and nitrogen removal, as well as microbial pollution control [9].

Recent studies have shown that the elimination of PPCPs in municipal STPs is often incomplete [10], with efficiencies ranging between 60% and 90% for a variety of polar compounds. Their removal can be attributed not only to biodegradation, but also to adsorption onto solid surfaces [11, 12]. As a consequence, significant fractions of PPCPs are discharged with the final effluent of the STP into the aquatic environment. A major factor influencing the efficiency of pollutants removal from water is their ability to interact with solid particles, both natural (clay, sediments, microorganisms) and added to the medium (active carbon, coagulants), because this facilitates their removal by physical-chemical (settling, flotation) or biological processes (biodegradation). However, compounds with low adsorption coefficients tend to remain in the aqueous phase, which favors their mobility through the STP and the receiving environment [13]. In this way, many PPCPs remain in the aqueous phase, such as the anti-inflammatories and the antibiotics, whereas some of them are adsorbed to the sludge, such as the estrogens [14].

The overall removal rates published in literature vary strongly. In Germany, the reported efficiencies range from 10% to 90% depending on the nature of the compound [10]. In Brazil, the removal efficiencies for pharmaceutical polar compounds vary from 12% to 90%, where the efficiencies obtained in activated sludge processes were higher than those of obtained in biofilters [15]. Another study, carried out in the USA concluded that many PPCPs (around 80%) were removed [16]. In all these cases, the removal includes both degradation and adsorption and the difference between the two mechanisms has not been assessed yet. In the case of polar compounds, such as carboxylic acids, for which the adsorption effects are expected to be very low, the main mechanism of elimination is attributed to biodegradation.

Conventional surface water treatment plants (SWTPs) typically treat water via coagulation using alum, ferric chloride, and/or synthetic polymers followed by flocculation, sedimentation, filtration, and disinfection. Conventional SWTPs achieve high removals of pathogens and other biological particles, and modest removal of dissolved organic carbon (DOC, 1 to 10 mg/L). In the United States, chlorine and chloramines are typically used for disinfection, while ozonation is more commonly practiced in European countries. Alternative SWTP designs or modifications to conventional WTPs sometimes include additional water treatment processes (e.g., adsorption on activated carbon, biofiltration, membranes, aeration, chemical softening, and ultraviolet light irradiation).

By comparing the influent and effluent estrogen concentrations, authors [2] concluded that the removal efficiency for estrone (E1) and 17 β -estradiol (E2) were 87 and 61%, respectively. In a study of 20 STP in Sweden, the average removal efficiency was 81% by activated sludge treatment and only 28% by solid supported bacterial treatment. Therefore it is essential to install additional treatment processes after the secondary treatment step, to ensure a more complete removal of such compounds.

The attenuation of PPCPs in receiving waters and in drinking water treatment plants also has been demonstrated in several studies and has been reviewed recently [17]. For example, the attenuation of several PPCPs was observed as secondary wastewater effluent was infiltrated into groundwater [18] and during river bank filtration [17]. In contrast to the efficient removal of many PPCPs observed in saturated and unsaturated groundwater, conventional drinking water treatment plants do not appear to remove PPCPs well. For example, clofibric acid has been detected at concentrations as high as 270 ng/L in drinking water from Berlin [19]. Of 47 wastewater tracers and EDCs analyzed, 15 were detected in raw drinking water (river water) samples, and 14 in finished drinking water samples from Atlanta [20].

2. Chemical precipitation processes

Metal salts (aluminum sulfate, ferric chloride) and softening chemicals (calcium oxide, sodium carbonate) are commonly added to destabilize particles present in water or to precipitate new particles (coagulation), aggregate particles (flocculation), and improve settling characteristics of particles (clarification). Sand filtration is commonly used after clarification for additional particle removal. Natural organic matter and EDCs or PPCPs may adsorb to particles in water and metal hydroxide particles formed during coagulation. Furthermore, chemical precipitation can remove moderately hydrophobic organic contaminants that have a strong affinity for adsorbed natural organic matter [1].

However, very little is known about the association of EDCs or PPCPs to particles present in water treatment systems. Therefore, it is difficult to make a priori predictions about the removal of EDCs or PPCPs during chemical precipitation processes.

An insight to the problem can be obtained by considering the behavior of pesticides, herbicides, and polycyclic aromatic hydrocarbons during chemical precipitation processes. Partitioning of organic compounds onto particles can occur through several mechanisms.

Some of the more hydrophobic compounds are present in wastewater effluents, but may not occur in raw drinking water supplies [20]. Some surfactants include both polar and non-polar moieties. The partitioning of octylphenol in English river sediments was the highest to the clay and silt fraction of the sediments, suggesting a hydrophobic interaction with organic carbon as well as surface area associated with the clay and silt particles [21]. As a result, only a few compounds (e.g., bisphenol A, E2, 17- α ethinylestradiol, octylphenol, polycyclic aromatic hydrocarbons) could be associated with organic phases of particles in drinking water treatment plants.

In addition to hydrophobic partitioning, organic contaminants can be adsorbed to particles by interactions of polar functional groups with charged particles and mineral surfaces by complexation or ion exchange. For example, the observed sorption of many polar pharmaceuticals (e.g., tetracycline) to soil and sediment is considerably stronger than predicted by hydrophobic interactions alone [22]. Such interactions could be particularly

important in drinking water treatment, where mineral oxides provide a relatively high density of surface functional groups that could interact with polar pharmaceuticals. For example, approximately 25% of the nonylphenols in groundwater were removed during coagulation with alum [23]. Therefore, it is possible that some polar EDCs or PPCPs could be removed during coagulation.

Despite the predictions, available data from pilot and full-scale water treatment plants indicate that removal will be modest at best. Neither lime softening nor alum coagulation (conventional or enhanced dosages ranging from 6 to 18 mg/L) demonstrated atrazine removal. Coagulation/flocculation/sedimentation with alum and iron salts or excess lime/soda ash softening did not result in significant removal of antibiotics (i.e. sulfachlorpyridazine, sulfadimethoxine, sulfamerazine, sulfamethazine, sulfathiazole, and trimethoprim). In another study, ferric chloride precipitation did not remove several pharmaceuticals frequently detected in surface waters (diclofenac, carbamazepine, bezafibrate). Certain pesticides were poorly removed by coagulation and 50% of the pyrene, fluoranthene, and anthracene were removed through hydrophobic interactions. On the basis of predictions of hydrophobic interactions and results of full-scale measurements, it was concluded that EDCs and PPCPs not associated with colloidal or particulate material will most likely be poorly removed during coagulation [1].

Park [24] studied the effects of aluminum chloride on activated sludge performance and removal of 17- α ethinylestradiol, four 10 L activated sludge reactors that only varied in influent aluminum concentrations were operated. Aluminum was added in 0.1, 0.5, 1.5, and 2.5 mg/L concentration to the influent wastewater.

The laboratory reactor study has shown that aluminium in influent wastewater exhibits significant positive impact on the bioflocculation of activated sludge. A correlation between effluent suspended solids, particulate-colloidal biopolymers, and 17- α ethinylestradiol concentrations were established. The following conclusions are drawn from this study:

- Higher Al-fed activated sludge led to better biomass settling and dewatering properties.
- Higher Al-fed systems resulted in lower effluent, total suspended solids (TSS) and lower particulate-colloidal biopolymers in the effluent.
- Higher Al-fed activated sludge resulted in better removal of 17- α ethinylestradiol.
- Alum plays very important role in activated sludge bioflocculation and it also enhances the removal of EDCs in wastewater treatment systems [24].

Xiaoying discussed [25] the coagulation characteristics of bisphenol-A with polyaluminum chloride (PACl- Al_{13}) as coagulant, examined the impact of coagulation pH, PACl- Al_{13} dosage, total organic carbon and turbidity on bisphenol-A removal, and analyzed the possible dominant mechanisms in water coagulation process.

Formation and performance of flocs during coagulation processes were monitored using photometric dispersion analyzer. When the concentration of humic acid matters and turbidity was low in the solution, the experimental results showed that the removal of bisphenol-A increases and subsequently decreases with the PACl- Al_{13} dosage increasing. The optimal PACl- Al_{13} dosage was found at bisphenol-A/PACl- Al_{13} = 1:2.6(M/M) in the experimental conditions. Results show that the maximum bisphenol-A removal efficiency occurred at pH 9.0 due to the adsorption of bisphenol-A onto Al_{13} aggregates rather than the charge neutralization mechanism by polynuclear aluminous salts in the solution. The humic

acid matters and kaolin in the solution have significant effect on bisphenol-A removal with PACl-Al₁₃ in the coagulation. The bisphenol-A removal will be weakened at high contents of humic matters. The removal rate of bisphenol-A increased and subsequently decreased with the turbidity increasing.

To characterize the relationship between coagulant dosage and settling performance (turbidity/TOC removal) the floc formation rates were measured using PDA. The content of bisphenol-A in the tested water was 5.0 mg/L. The effect of coagulant dosage on the bisphenol-A removal was examined at 5, 7, 9, 10, 11, 13, 15, 17.5 and 3.37 mg/L of total organic carbon, and pH 7.0 [25].

As shown in Figure 2, an increase in PACl-Al₁₃ dose from 1 to 13 mg/L, improved the bisphenol-A removal in the settling process. The maximum bisphenol-A removal efficiency was 26.8%, and it was reached at zeta potential close to the isoelectric point of hydrolysis product particles. The results indicate that the dominant removal mechanism at pH 7.0 is the charge neutralization of large negatively charged natural colloids by positively charged hydrolysis product particles of hydrolyzing salts. This promotes agglomeration of destabilized colloids [25]. The mechanisms of interaction of aluminium salts hydrolysis product particles with different contaminants and efficiency of water treatment by hydrolysing salts are discussed in detail in our review [30].

At higher PACl-Al₁₃ dosage, the interaction between voluminous aluminium hydroxide precipitates and natural colloids is known as sweep coagulation. The particles are destabilized by adsorption on aluminium hydroxide sols and are enmeshed in sol aggregates, promoting the formation of relatively large, quickly sedimenting flocs. Thus, Bisphenol-A, which is not adsorbed initially, can be removed by enmeshment in the flocs [25].

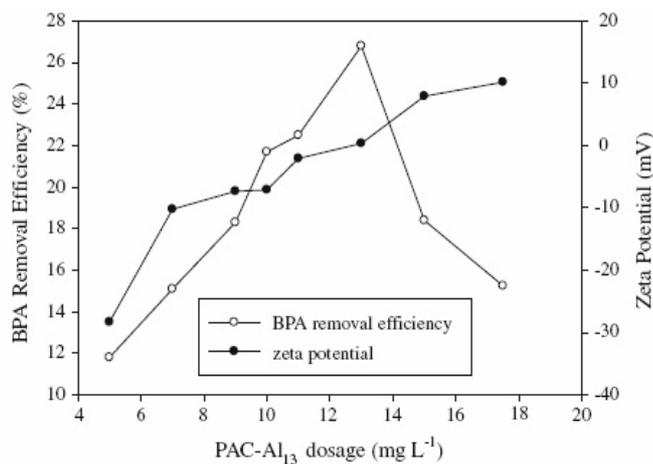


Figure 2. Effect of PAC-Al₁₃ dosage on BPA removal efficiency and zeta-potential of 5 mg/L BPA solution (TOC = 3,36 mg/L, pH = 7) [25]

Amirtharaja and Tambo [26] reported that the zeta potential of properly destabilized particles usually ranges between -4 mV to $+3$ mV. For these reasons it can be supposed that the optimum PACI- Al_{13} dosage will correspond to that at which zeta potential of particles will be approximately zero. As shown in Figure 2, the bisphenol-A removal increased with the increase of coagulant dosages before reaching the optimal dosage, which was found at bisphenol-A/PACI- Al_{13} = 1:2.6(M/M) In these conditions zeta potential remained negative but gradually approached zero. When zeta potential was reversed, the bisphenol-A removal decreased due to the re-stabilization of particles [25].

It has been shown that the flocs size vs. time increased as the PACI- Al_{13} dosage increased from 5 to 13 mg/L (Figure 3). Higher dosage did not significantly increase the rate of floc formation. The floc growth follows the classical pattern composed of a short induction period followed by a rapid growth phase until the steady state floc size is reached. These data correlate with the results presented in Figure 2, which indicates that the mechanism might be re-stabilization of colloidal particles when over dose of coagulant was added [25].

3. Removal of low level contaminants by coagulation

Paper [27] describes the removal efficiency of bisphenol A and nonylphenol present in waters at low level, by conventional water treatment methods – coagulation/flocculation/sedimentation, oxidation-chlorination and ozonation and adsorption on powdered activated carbon (PAC) and granular activated carbon. Coagulants tested were alum, poly-aluminum hydroxychloride (PACI), poly-aluminum silicate sulfate (PASS), poly-aluminum chloride silicate (PACS), and $Fe_2(SO_4)_3$. Results are presented in Table 1.

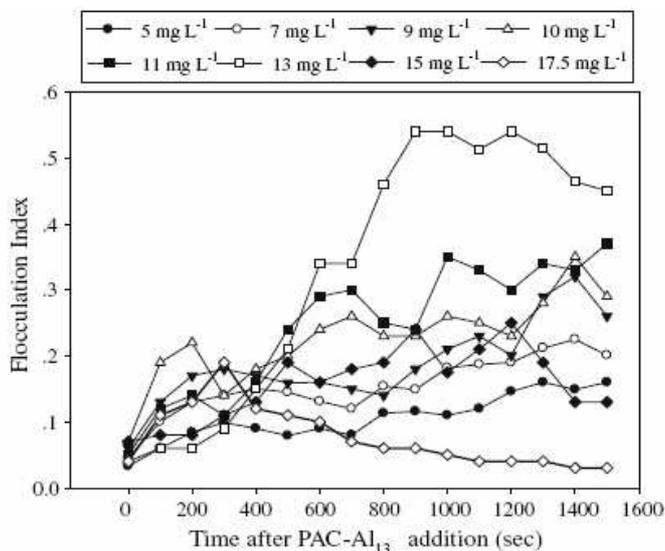


Figure 3. Flocculation index vs time for eight PAC- Al_{13} dosages [25]

Table 1

Removal of bisphenol-A and nonylphenol by coagulation, flocculation and sedimentation [27]

Chemical	Initial conc.	alum 50 (mg/L)	PACI 45 (mg/L)	PASS 45 (mg/L)	PACS 30 (mg/L)	Fe ₂ (SO ₄) ₃ 45 (mg/L)
Bisphenol-A (µg/L)	1	0.93	0.96	0.94	0.96	0.94
Turbidity (NTU)	5.1	0.76	0.64	0.74	0.6	0.69
DOC (mg/L)	2.16	1.53	1.64	1.59	1.62	1.67
Nonylphenol (µg/L)	4.5	4.56	4.35	4.43	4.5	4.4
Turbidity (NTU)	9.4	0.82	0.7	0.76	0.63	0.77
DOC (mg/L)	2.25	1.62	1.5	1.58	1.6	1.57

It can be seen that the turbidity index dropped from 5.1–9.4 to 0.60–0.82 nephelometric turbidity units (NTU) and dissolved organic carbon (DOC) concentrations decreased from 2.16–2.25 to 1.5–1.67 mg/L. Despite the fact that DOC decreased by 26–30%, removal efficiencies of nonylphenol and bisphenol were only 4–7% and 0–3%, respectively.

Spiked EDCs did not appear to be adsorbed onto natural organic matter completely during 30 minutes of coagulation/flocculation/sedimentation. The type of coagulant used did not affect the EDC removal efficiency significantly. Chemical precipitation using iron and aluminum salts was found to be ineffective in removing estrogen [28] and pharmaceuticals [29].

Conclusions

1. Endocrine disrupting compounds (EDC) have been detected in influents and effluents of sewage treatment plants (STP), surface and drinking waters in many countries.
2. The most efficient methods of removal of EDCs from waters are adsorption on activated carbon and oxidation by ozone and chlorine. At the same time coagulation by multivalent salts hydrolysis products and/or adsorption on freshly precipitated hydrolysis products particles could serve as an alternative or additional method of EDCs removal taking into account that coagulation is an obligatory stage of water conditioning in the majority of waterworks. Combination of coagulation with adsorption can give good results in the process of treatment of waters containing these contaminants.
3. Elimination of Pharmaceutical and Personal care products (PPCPs) in municipal Sewage Treatment Plants (STPs) can reach 60%–90% depending on the polarity of the compounds to be removed. Their removal can be attributed to biodegradation, and also to adsorption onto solid surfaces.
4. The removal efficiency of bisphenol A and nonylphenol, as the most dangerous representatives of EDCs, by coagulation using aluminum sulphate and poly-aluminum chlorides has been studied in a number of works. The conventional

coagulation/flocculation water treatment process had very low removal efficiencies for bisphenol A (0–3%) and nonylphenol (4–7%).

5. The degree of EDCs removal from waters with relatively high contents of pollutant in the presence of big doses of poly-aluminum-chlorides can reach 26–28%. The mechanism of removal is “sweep” coagulation, i.e. entrapment of contaminants between the freshly precipitating hydrolysis product particles $[\text{Al}(\text{OH})_3]$, i.e. by their enmeshment in the forming flocs.

Acknowledgements

The described work was carried out as part of the TÁMOP-4.2.1.B-10/2/KONV-2010-0001 project in the framework of the New Hungarian Development Plan. The realization of this project is supported by the European Union, co-financed by the European Social Fund.

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