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Review

Ultrasonic treatment of endocrine disrupting compounds, pharmaceuticals, and personal care products in water: A review

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HIGHLIGHTS

- Removal of contaminants of emerging concerns in water was reviewed.
- Valuable information was provided for applications of ultrasonication in water treatment.
- Areas of future research for the removal of contaminants of emerging concerns are suggested.

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ABSTRACT

The presence of contaminants of emerging concerns such as endocrine-disrupting compounds (EDCs) and pharmaceuticals/personal-care products (PPCPs) is of concern because they are not completely removed during conventional water and wastewater (WW) treatment processes including coagulation/flocculation/sedimentation/filtration and biological activated sludge process. Recently, ultrasonic (US) treatment has been well-known as an advanced treatment process for the removal of complex inorganic and organic contaminants in water and WW. US treatment has shown substantial advantages, such as cleanliness, safety, energy savings, and negligible or no secondary pollution products. This review provides a summary of recent research on the removal of EDCs and PPCPs by US treatment and also provides information valuable for applications of US treatment in water and WW treatment. The removal of numerous EDCs and PPCPs of different classes was reviewed based on the current literature to (i) address key factors (water quality conditions (pH, temperature, background common ions, and promoters/scavengers), US frequency, power, and reactor type) influencing the sonodegradation of EDCs and PPCPs and their intermediates during US treatment, (ii) evaluate the effects of various catalysts and hybrid processes on sonodegradation, and (iii) discuss EDC and PPCP removal according to their properties. Additionally, areas of future research in US treatment for the removal of EDCs and PPCPs from water are suggested.

Abbreviations: AAP, acetaminophen; AC, activated carbon; ATZ, atrazine; BOD, biochemical oxygen demand; BPA, bisphenol A; CBZ, carbamazepine; CIP, ciprofloxacin; CNMs, carbon nanomaterials; CNTs, carbon nanotubes; COD, chemical oxygen demand; CyP, cumyphenol; DBS, dodecylbenzene sulfonate; DCF, diclofenac; DOC, dissolved organic carbon; E1, estrone; E2, 17b-Estradiol; E3, estriol; EC, electrochemical; EDCs, endocrine-disrupting compounds; EDSP, Endocrine Disruptor Screening Program; EE2, 17a-Ethinyl estradiol; GBs, glass beads; GOs, graphene oxides; IBP, ibuprofen; IPM, iopromide; MeOH, methanol; NPs, nanoparticles; NPX, naproxen; NTs, nanotubes; OXA, oxacillin; PAC, powdered activated carbon; PGRRP, planned groundwater recharge reuse project; PhACs, pharmaceuticals; PPB, propylparaben; PPCPs, pharmaceuticals/personal-care products; SDW, synthetic drinking water; S_H, Henry's low constant; SMX, sulfamethoxazole; SSWM, stainless steel wire mesh; SWNTs, single-walled carbon nanotubes; *t*-BuOH, *t*-Butanol; Ti-WM, Ti-wire mesh; US, ultrasonic; USEPA, The United States Environmental Protection Agency; UV, ultraviolet; WTPs, water treatment plants; WW, wastewater; WWTPs, WW treatment plants.

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Contents

1. Introduction

Many studies have shown the presence of contaminants of emerging concerns such as endocrine-disrupting compounds (EDCs) and pharmaceuticals (PhACs)/personal-care products (PPCPs), in wastewater (WW) effluents and various drinking water sources, where some EDCs and PPCPs may have ecological impacts even at very low concentrations (<$\mu g\,L^{-1}$) [1–7]. Stumm-Zollinger and Fair in 1965 and Tabak and Bunch in 1970 were the first to express concerns about the possible adverse effects of PhACs found in municipal WW, by revealing that several steroids were poorly removed by typical WW treatment processes [8,9]. Because the removal of EDCs and PPCPs can vary during water and WW treatment depending on their physicochemical properties, including size, charge, shape, hydrophobicity, p*K*a, and functional group, many studies have investigated the fate and transport of EDCs and PPCPs in water/WW treatment processes over the last decade [1,10–18].

EDCs and PPCPs are possible contaminants of drinking water, particularly when WW is reused. The Endocrine Disruptor Screening Program (EDSP) was established for EDCs by the United States Environmental Protection Agency (USEPA) in 1998, which classifies compounds that could potentially interfere with the endocrine system and determines the effects of these compounds [19]. The EDSP suggested that both human and wildlife influences be evaluated, and that estrogen, androgen, and thyroid end points should be examined. In drinking or natural water, no federal regulation currently exists for PhACs; an evaluation of a PhAC associated with ecological testing is required by the US Food and Drug Administration if the environmental concentration in water is expected to exceed 1 $\mu g\,L^{-1}$ [20]. Only a few EDCs and PPCPs (erythromycin, 17b-estradiol (E2), estriol (E3), 17a-ethinyl estradiol (EE2), and estrone (E1)) are currently listed in the USEPA's "Drinking Water Contaminant Candidate List 4" [21]. The State of California has examined the potential impacts of EDCs and PPCPs for indirect potable reuse of municipal WW effluent: "The recycled water for EDCs and PPCPs identified by the Department will be monitored each year by the planned groundwater recharge reuse project (PGRRP), based on the PGRRP engineering report review and the affected groundwater basins" [22].

As mentioned previously, the removal of EDCs and PPCPs can vary depending on their physicochemical properties and treatment techniques in water treatment plants (WTPs) and WW treatment plants (WWTPs). Many studies have shown that EDCs and PPCPs are removed to different degrees by coagulation/flocculation/sedimentation/filtration [23–25], chlorination [24,26], activated carbon (AC) adsorption [27–29], carbon nanomaterials (CNMs) (*e.g.*, carbon nanotubes (CNTs) [30–32] and graphene oxides (GOs) [33]), membrane filtration [34–36], ozonation [24,37], ultraviolet (UV) irradiation [38–40], ultrasonication [41–43], and biological processes [4,10,44]. Unlike widely known advanced oxidation techniques such as ozonation (O_3/H_2O_2), photocatalysis (UV/TiO_2), ultraviolet (UV/H_2O_2), and Fenton/photo-Fenton, ultrasonic (US) treatment has recently been recognized as an advanced treatment process for the removal of complex inorganic and organic contaminants in water and WW [42,45–47]. The sonodegradation process is based on water sonolysis, which produces H_2O_2, $OOH^{\cdot}$, $OH^{\cdot}$, $OH_2^{\cdot}$, and $H^{\cdot}$ through the nucleation/growth/collapse of cavitation bubbles in water due to the high pressure and temperature caused by ultrasound waves [48]. US treatment has shown substantial advantages, such as cleanliness, safety, energy saving, and negligible or no secondary pollution products [49,50].

Over the last decade, many studies have reported the removal of EDCs and PPCPs by US treatments. Thus, a comprehensive review of EDC/PPCP removal by US treatment is important, because sonodegradation of EDCs and PPCPs is influenced significantly by their unique properties, including size, shape, p*K*a, functional groups, and hydrophobicity (usually determined in terms of the octanol-water partition coefficient, K_{OW}), as well as water quality and ultrasonication conditions. The primary objective of this review is to combine current knowledge of US treatment of EDCs and PPCPs in water and WW, and identify knowledge gaps and limitations to highlight future research areas based on the most recent and relevant studies. In particular, this review aims to (i) address key factors (water quality conditions (pH, temperature, background common ions, and promoters/scavengers), US frequency, power, and reactor type) influencing the sonodegradation and intermediates of EDCs and PPCPs during US treatment, (ii) evaluate the effects of various catalysts and hybrid processes on sonodegradation, and (iii) discuss EDC and PPCP removal with regard to their properties during US treatment.