



Removal of selected micropollutants from water by ultrasonication

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ABSTRACT

As a result of the urbanization process, an increase in population, and the development of analytical techniques, recently, many organic micropollutants are discharging and identified in aquatic systems. Moreover, many of them can be harmful to human health even at low concentrations. However, literature reports proved that traditional water treatment methods may be ineffective in the removal of organic micropollutants, thus they can migrate through the environmental matrices in an unchanged form for a long time. Consequently, innovative and effective techniques for water treatment should be developed to prevent the discharge of undesirable substances into the water and one of them is ultrasonication. This study aimed to determine the ultrasonication efficiency in the removal of selected organic micropollutants from water, namely bisphenol A, carbamazepine, 17- α ethinyloestradiol, pyrene, and triclosan. Research proved that sonication time, the addition of glass and sand particles and ozone addition had a significant impact on micropollutants degradation efficiency. Furthermore, in the research impact of water matrices in which micropollutants occur on degradation rate by ultrasonication was examined. To evaluate the process effectiveness, the gas chromatography method (GC-MS) preceded by solid phase extraction (SPE) was used. Moreover, Microtox[®] bioassay was implemented to evaluate the toxicity of the post-treated sample.

Keywords: Ultrasonication; AOPs; Water treatment; Micropollutants; Ecotoxicity; Ultrasound

1. Introduction

It is a well-known fact that organic micropollutants (OMPs) can enter the aquatic environment as a result of anthropogenic activity. The presence of this group of compounds in water raises concerns of many scientists due to its harmful impact on human health even at very low concentrations. In addition, the impact of some OMPs on living organisms has not been fully explained and examined yet. Furthermore, the maximum concentration of many OMPs in the environment is not regulated by existing guidelines and law regulations [1–3]. Thus, many of OMPs which are potentially harmful and biologically active in ecosystems were classified in the literature as compounds of emerging concern (CECs). OMPs include many groups of substances, for example, pharmaceuticals and personal care

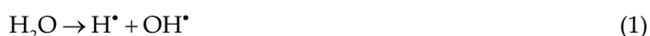
products (PPCPs), steroids and hormones, disinfection by-products (DBPs), pesticides, detergents, flame retardants, gasoline, plasticizers, and preservatives which can cause serious health problems [4–6]. For instance, literature data proved that bisphenol A (BPA) which was found in electronic devices, toys, CDs, books, plastic bottles, and other polymer material products can cause lung, prostate, and breast cancer. It can also affect immune function and the nervous system [7–12]. Carbamazepine (CBZ) as one of the PPCPs is used in various mental disorders, epilepsy, and pain treatment [13,14] and it has a very negative effect on the ecosystem [15–17]. The other PPCP is triclosan (TCS) which can disturb cell functioning, leads to cytotoxicity, it affects the reproductive systems and DNA stability [18]. Furthermore, it can be found in many daily use products, for example, detergents, soaps, deodorants, toothpaste,

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clothes, kitchenware, shave gels, skin cleaners, and other cosmetics [19–21]. Pyrene (PYR) presence in the environment is related to various natural and anthropogenic processes – primarily incomplete combustion of fuels but also volcano eruptions, fires, petroleum spills, incomplete combustion of fuels, and due to transportation systems. This one of more than 100 identified polycyclic aromatic hydrocarbons (PAHs) was proven to be mutagenic and toxic for humans and its transformation products can be carcinogenic [22–27]. 17 α -ethinyloestradiol (EE2) which is estrogen causes a reduction in fecundity and altered sex behaviors. EE2 is widely spread in the environment mainly due to a woman's excretion via urine and feces [28].

Furthermore, they were identified in many water matrices such as surface water, tap water, drinking water, and groundwater which provide evidence that classical treatment techniques can be low effective in CECs degradation [29–31]. One of the novel and effective techniques in water disinfection and wastewater treatment is ultrasonication. In this technology, sound waves with a frequency higher than 20 kHz are used (for environmental engineering purposes it most often ranges from 20 to 1,000 kHz) [32,33]. During intense ultrasound conditions, the formation, growth, and violent collapse of the bubble takes place which is called acoustic cavitation phenomena. In this process as a result of the periodic movement of water particles caused by the propagation of ultrasonic waves rarefaction, and compression phases can be distinguished. The first one is connected with negative pressure conditions and in this phase, the initial bubble can be formed (when the pressure reached a critical value). Then the bubble is growing until it reaches the critical size and subsequent collapse in positive pressure conditions (compression phase). The bubble implosion leads to the occurrence of extreme temperature and pressure values (5,000 K and 100 MPa, respectively) [8,34–36]. In addition, some hydrodynamical effects could appear, namely microjets, shear forces, and shock waves. During implosion, in the bubble collapse, the pollutants can be eliminated due to the pyrolytic process and reaction with generated hydroxyl radicals.

However, pollutants can be removed also at the bubble liquid interface, and in the liquid area by reacting with other oxidative species, for example, hydrogen peroxide [37–39]. The hydroxyl radicals are generated according to the formulas [40,41]:



The acoustic cavitation phenomena and related processes are illustrated in Fig. 1.

Although ultrasonic cavitation was found to be very effective in many harmful substances removal, there is a

tendency to combine this process with other treatment processes such as ozonation, UV irradiation, and hydrogen peroxide addition. Furthermore, many other diverse sonocatalysts and sonosensitizers can be used, for example, TiO₂, porphyrin, hypocrellin, ZnO, etc. [42–47].

Thus, this study aimed to examine the effect of the ultrasonication process on the removal of selected OMPs (BPA, CBZ, EE2, PYR, and TCS) from deionized water and surface water matrices. Moreover, the effect of ozonation, solid catalysts addition (sand and glass particles), and the influence of ultrasonication operational parameters on OMPs removal were also tested. In this paper, to evaluate the effectiveness of OMPs degradation, the gas chromatography method (GC-MS), and toxicological analysis of post-treated samples were performed using the Microtox[®] test.

2. Materials and methods

2.1. Sample preparation and reagents

The surface water samples analyzed in this paper were collected from the pond and canal in the rim part, around 15 cm below the surface water level. The pond and canal are located in southern Poland, in an urbanized area. Subsequently, 1 mg·L⁻¹ of BPA, CBZ, EE2, PYR, and TCS was added to the water matrices (pond, canal, and deionized water). These compounds provided by Sigma-Aldrich (Poznan, Poland) were analytical grade. In the study, the basic physicochemical parameters of the samples were determined. Turbidity was measured by using the HI-93414-02 Turbidity meter by HANNA Instruments Inc. Conductivity and pH were tested by the use of CPC – 505 device provided by Elmetron (Zabrze, Poland) while the total organic carbon (TOC) was measured using TOC – L Analyzer (Shimadzu, Kyoto, Japan). The properties of tested compounds are shown in Table 1.

2.2. Ultrasonication and ozonation processes

The ultrasonication process (US) was conducted in horn type ultrasonic device – Sonics VCX 500 provided by Vibracell Sonics, Sonics and Materials Inc., (Newtown Connecticut, USA). In the study, samples were sonicated in a 100 mL volume reactor with a constant frequency of 20 kHz by using a 13 mm diameter probe which can generate a maximum of 500 W of power and 114 μm of sonication amplitude. The sonication process was conducted in the range of 20%–80% of maximum amplitude at 1, 5, 10, 20, and 30 min of the treatment. To keep the constant temperature of the sample the cooling system was used. In the next stage of the study, sand particles (10, 20, and 50 g) and broken glass (20 g) were placed in the reactor. Subsequently, 1, 2, 5, and 10 mg·L⁻¹ of ozone were used, both as a single process and combined with 30 min ultrasonication at 80% of maximum amplitude. In this work, ozoner FM500 (WRC Multiozone, Gdansk, Poland) was used. The experimental setup used in the study is shown in Fig. 2.

2.3. Gas chromatography

The effectiveness of the ultrasonication and ozonation treatment was determined by using the 7890B gas

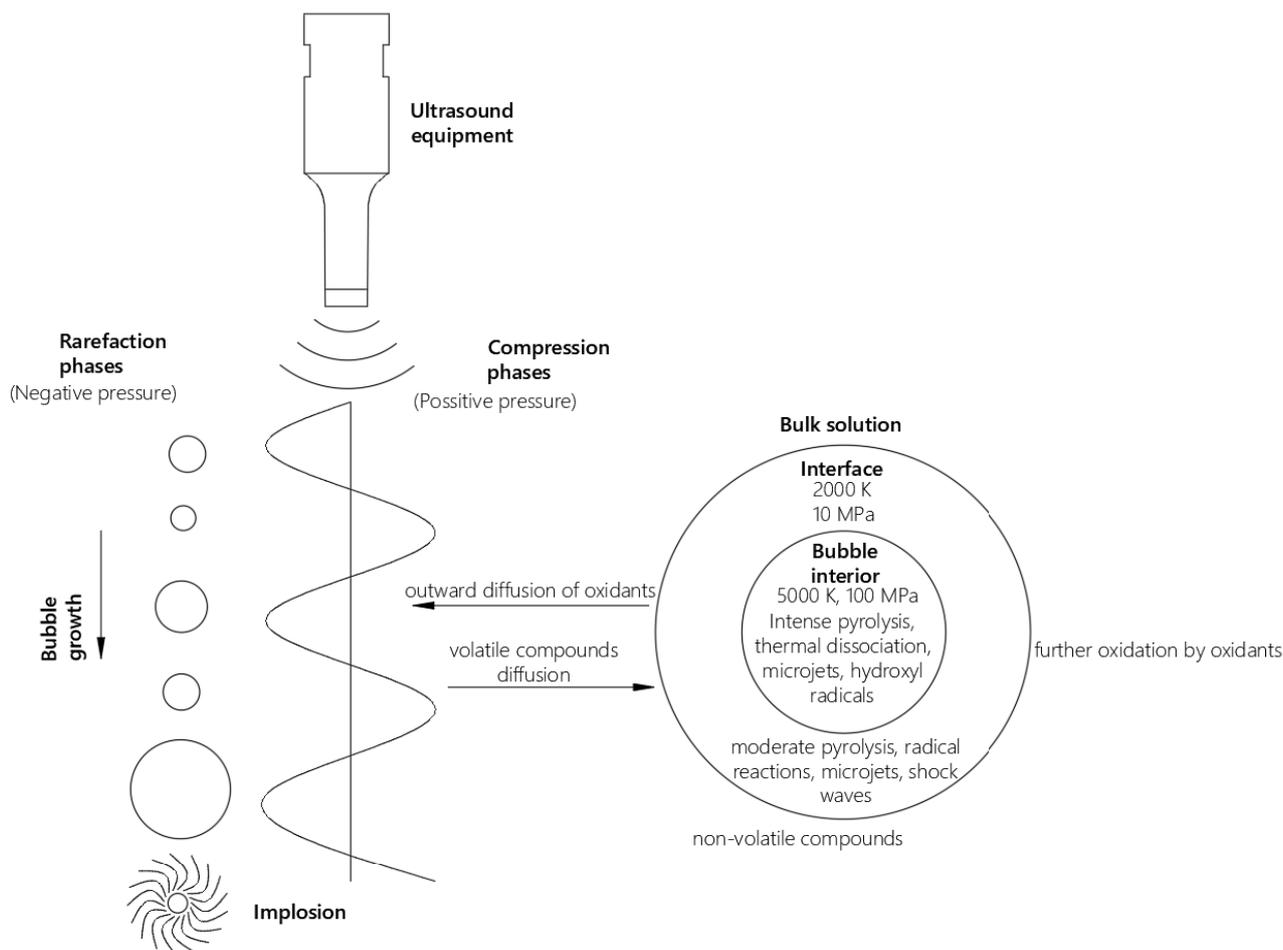


Fig. 1. Acoustic cavitation phenomena and related mechanisms [38,39].

chromatograph (GC) provided by Perlan Technologies (Warsaw, Poland). In GC-MS analysis, helium was used as the carrier gas at the flow rate of 1.1 mL/min. The temperature of the ion trap and ion source was set to 150°C and 230°C, respectively. The injector temperature was 250°C and the oven temperature varied from 80°C to 300°C. The capillary columns (0.25 μm thickness SLBTM – 5 ms 30 m \times 0.25 mm) were obtained from Sigma-Aldrich (Poznan, Poland). Limit of detection (LOD) for BPA, CBZ, EE2, PYR, and TCS was equal to 0.05, 0.20, 0.15, 0.03, 0.22 $\text{ng}\cdot\text{L}^{-1}$, respectively. The recovery of tested compounds varied from 97% to 99%.

To make the samples matrices to a suitable form, and to improve the quality of chromatography analysis, the Solid Phase Extraction (SPE) technique was applied by using SupelcleanTM ENVITM – 18 tubes with an Octadecylsilane (C_{18}) as a cartridge bed. In the conditioning process, 5 mL of methanol (MeOH) and pure water were used, while during elution 5 mL of dichloromethane (DCM) was applied. The sample flow was set to 1 mL/min. while the drying time in the vacuum was equal to 5 min. The SPE process was carried out under a negative pressure of 5–10 kPa. The OMPs elimination degree after treatment was evaluated based on the peak areas which were compared with the corresponding data from the calibration process.

2.4. Toxicity analysis

The toxicity assessment of the samples after selected processes was conducted in Microtox[®] bioassay according to the Microtox Omni system procedure in Microtox 500 analyzer (Warszawa, Poland). In this test, highly sensitive to toxic substances luminescent bacteria *Aliivibrio fischeri* and based on its light emission intensity, toxicity effect can be evaluated. In the study, average toxicity after 5 and 15 min of bacteria exposition to the post-treated sample was studied and the results were classified according to toxicity classes shown [49].

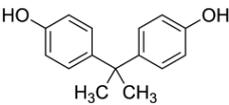
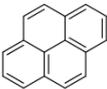
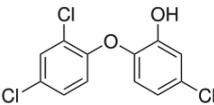
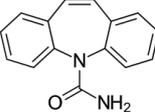
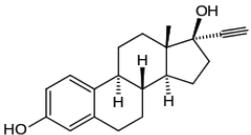
2.5. Calculations

In this paper, acoustic pressure P_a was calculated as follows [50]:

$$P_a = \sqrt{2\rho cI} \quad (5)$$

where ρ – water density (1,000 kg/m^3), c is the sound speed in the water (1,500 m/s), I – ultrasonic intensity calculated as acoustic power P divided by tip area of ultrasonic transducer A .

Table 1
Properties of tested compounds [48]

Property	Compound name				
	BPA	PYR	EE2	CBZ	TCS
Molecular formula	$C_{15}H_{16}O_2$	C_6H_{10}	$C_{20}H_{24}O_2$	$C_{15}H_{12}N_2O$	$C_{12}H_7Cl_3O_2$
Molecular weight, $g \cdot mol^{-1}$	228.29	202.25	296.4	236.27	289.5
Solubility in water, $mg \cdot L^{-1}$	300 ¹	0.135 ¹	11.3 ²	18 ¹	10 ³
Octanol-water partition coefficient $\log K_{ow}$	3.32	5.18	4.14	2.45	4.76
CAS No.	80-05-7	129-00-0	57-63-6	298-46-4	3380-34-5
Structural formula					

¹ – at 25°C, ² – at 27°C, ³ – at 20°C

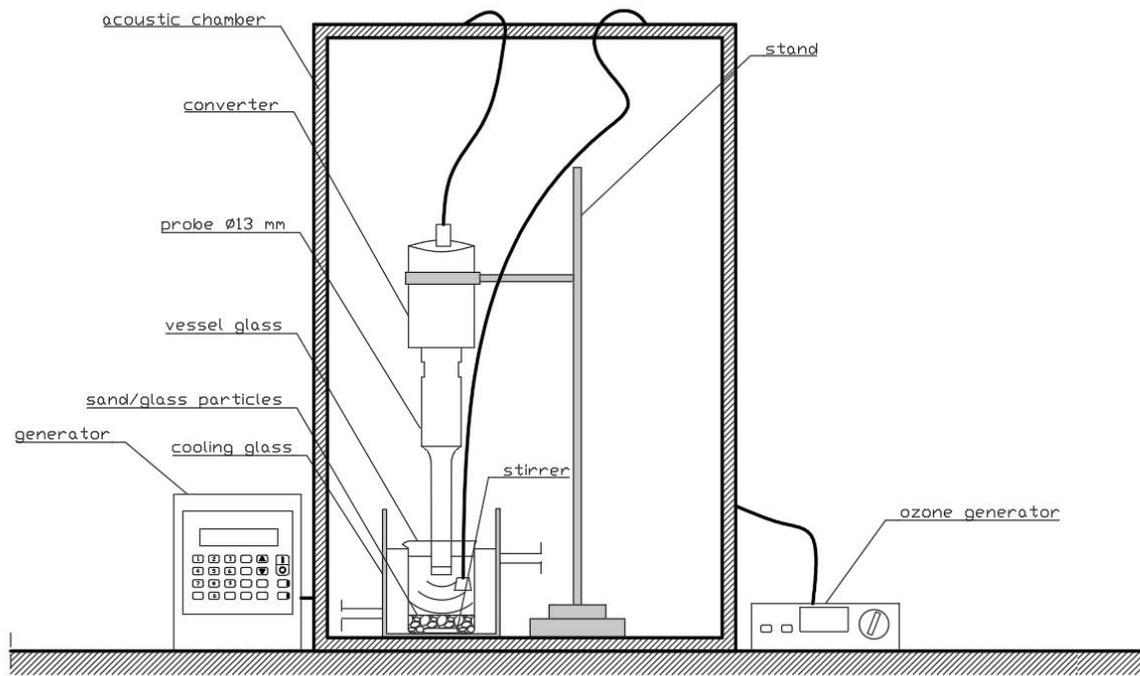


Fig. 2. Experimental setup used in the study (own investigation).

While acoustic power is expressed as the ratio of sonication energy E and sonication time t [51]:

$$P = \frac{E}{t} \quad (6)$$

Furthermore, based on the experimental data, the degradation process followed pseudo-first kinetics order, and the reaction constant k was calculated from Eq. (3).

$$\ln\left(\frac{C}{C_0}\right) = -kt \quad (7)$$

where C , C_0 – the concentration of pollutants at a given time t , and at the initial time, respectively.

3. Results

3.1. Effect of sonication time and power on micropollutants removal

The effect of ultrasonication time and power on BPA, CBZ, EE2, PYR, and TCS degradation rate is shown in Figs. 3–7. At this stage separate solutions of micropollutants in deionized water were used.

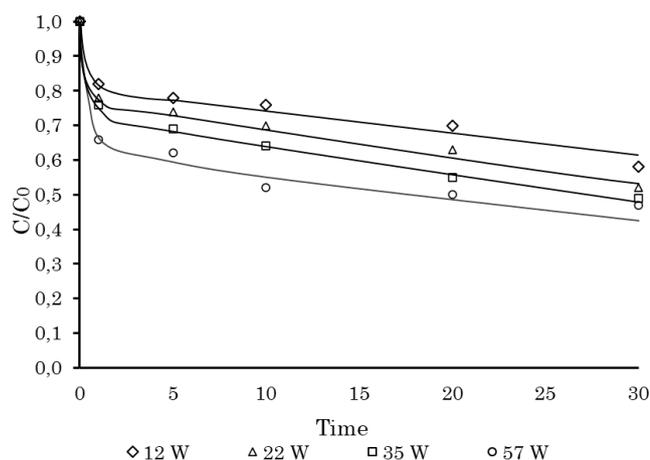


Fig. 3. Effectiveness of ultrasonication in TCS removal at different acoustic power, C – concentration of the compound after treatment, C_0 – initial concentration of the compound (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, matrix: deionized water containing 1 mg·L⁻¹ TCS).

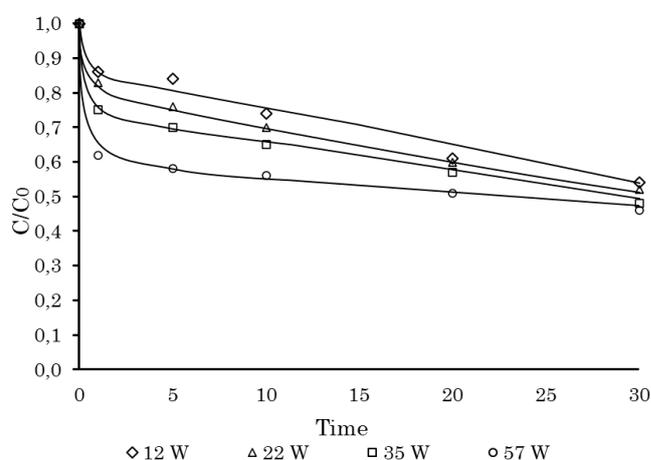


Fig. 5. Effectiveness of ultrasonication in EE2 removal at different acoustic power, C – concentration of the compound after treatment, C_0 – initial concentration of the compound (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, matrix: deionized water containing 1 mg·L⁻¹ EE2).

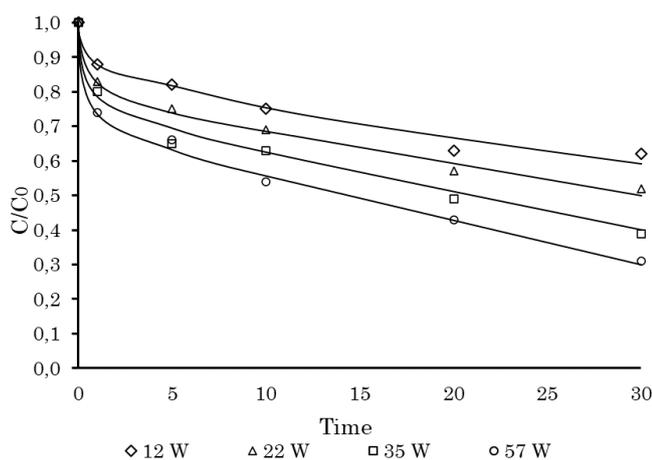


Fig. 4. Effectiveness of ultrasonication in CBZ removal at different acoustic power, C – concentration of the compound after treatment, C_0 – initial concentration of the compound (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, matrix: deionized water containing 1 mg·L⁻¹ CBZ).

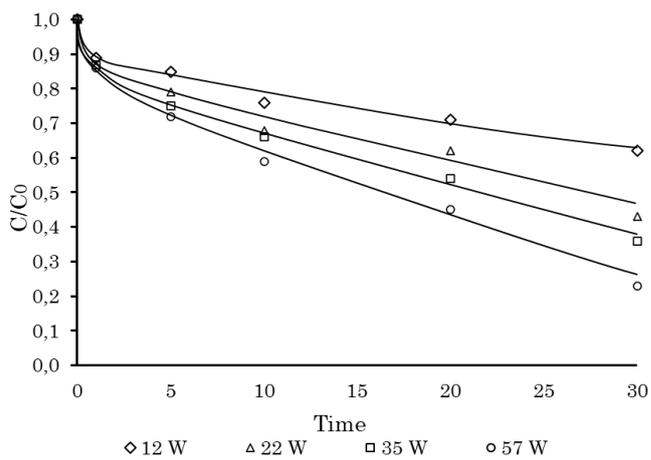


Fig. 6. Effectiveness of ultrasonication in PYR removal at different acoustic power, C – concentration of the compound after treatment, C_0 – initial concentration of the compound (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, matrix: deionized water containing 1 mg·L⁻¹ PYR).

Research revealed that the highest degradation rate was obtained in the case of PYR and BPA (77% and 71%, respectively) during 30 min sonication at the highest intensity. It may be related to their relatively low molecular weight and thus higher possibility to enter inside the acoustic bubble. This study provided further evidence that the removal rate was proportional to the energy delivered to the liquid for all micropollutants. Moreover, ultrasound treatment was found to be effective even at the lowest intensity. The average degradation rate of micropollutants after 30 min at 9 W/cm² was 39%. The reaction constant at 43 W/cm² varied from 0.11 (CBZ solution treatment) to 0.23 (BPA solution treatment). The calculated parameters of acoustic cavitation are shown in Table 2.

Obtained relation between degradation rate time and power is common in the literature data [45,47,52]. Acoustic power is a parameter proportional to the ultrasound intensity, acoustic amplitude, and acoustic pressure thus more bubbles and more oxidants can be generated according to the Eqs. (1)–(4).

It was estimated [53] that at 20 kHz frequency ultrasonication minimum intensity in water to induce cavitation phenomena is 1 W/cm², which is a much lower value than the lowest intensity value used in this study (9 W/cm²). However, it was proved that an excessive number of cavitation bubbles can limit the propagation of ultrasonic waves and influence the degradation effectiveness [54].

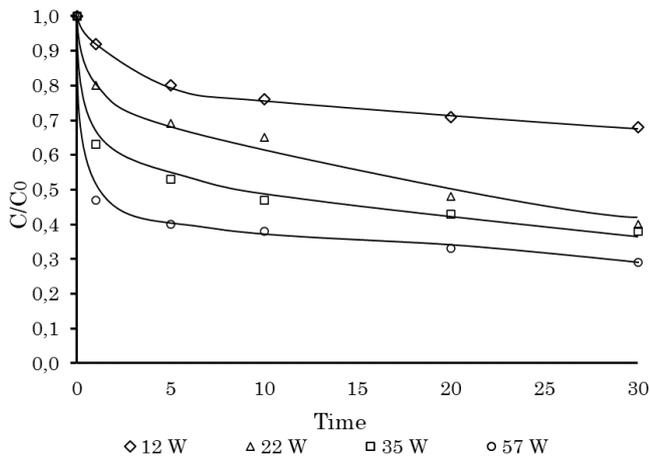


Fig. 7. Effectiveness of ultrasonication in BPA removal at different acoustic power, C – concentration of the compound after treatment, C₀ – initial concentration of the compound (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, matrice: deionized water containing 1 mg·L⁻¹ BPA).

Table 2
Parameters of ultrasonic treatment

Solution	P (W)	I (W/cm ²)	P _a (MPa)	k (1/min)
TCS	12	9	0.52	0.06
	22	17	0.71	0.08
	35	26	1.02	0.09
	57	43	1.14	0.13
CBZ	12	9	0.52	0.05
	22	17	0.71	0.07
	35	26	1.02	0.08
EE2	57	43	1.14	0.11
	12	9	0.52	0.05
	22	17	0.71	0.06
PYR	35	26	1.02	0.09
	57	43	1.14	0.14
	12	9	0.52	0.04
BPA	22	17	0.71	0.06
	35	26	1.02	0.06
	57	43	1.14	0.07

3.2. Effect of water matrix

The samples collected from the pond were characterized by a pH value of 8.1, while conductivity and turbidity were equal to 5.2 mS·cm⁻¹ and 13.5 NTU, respectively. The pH value, turbidity, and conductivity of water from the river were 8.0, 17.0 NTU, and 5.9 mS·cm⁻¹, respectively. It was highlighted in the literature data that water matrix can highly influence the sono-chemical degradation efficiency of

micropollutants [55]. For instance, Seymour and Gupta [56] indicated an increase in the removal rate of OMPs in water matrice containing sodium chloride at 20 kHz frequency sonication while Prado et al. [57] showed an increase in TCS sonodegradation from seawater, urban run-off water, and influent wastewater comparing to deionized water.

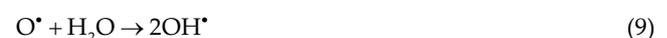
As shown in Figs. 8–12, in the present study, degradation rates of OMPs generally decrease in the order: pond matrix, canal matrix, and deionized water matrix.

After 30 min of the treatment maximum removal rate was equal to 81% in the case of PYR and pond water matrix. The average removal of all OMPs after 30 min sonication was 69%, 68%, and 62%, respectively for the canal, pond, and deionized water matrices. Enhancement of the removal rate of OMPs during sonication in natural water matrices may be attributed to the presence of organic and inorganic species which can act as extra nuclei for the acoustic cavitation process. Moreover, the presence of some compounds could enhance the degradation by facilitating the migration of pollutants inside or near the acoustic bubble e.g. as a result of the so-called salting out effect. In addition presence of some particles may be related to the partial adsorption of the solutes. However, there was no significant difference in efficiency between the pond and canal water matrice which might be related to their similar parameters and chemical composition [57–59].

3.3. Removal of OMPs using ozonation and ultrasonication treatment

In previous stages of the study maximum degradation of OMPs was obtained at the maximum power and intensity, thus the combined ozonation and ultrasonication (O₃/US) treatment was conducted at 57 W power. As shown in Fig. 13, ozone assistance during the sonication process leads to an increase in the OMPs removal efficiency. The addition of 10 mg·L⁻¹ O₃ in O₃/US treatment resulted in a 24% average efficiency improvement compared to 30 min ultrasonication used as a single process. Similarly to a single US process, the maximum degradation rate during O₃/US was obtained in the case of PYR (93%) which was the highest degradation rate in this study. In all experiments, ozonation as a single process had the lowest efficiency. Furthermore, US treatment was more or equally effective in OMPs elimination than 10 mg·L⁻¹ O₃ addition even at 1 min sonication time.

Degradation of OMPs by ozonation can occur as a result of oxidation by OH^{*} radicals produced during the decomposition of O₃ or by electrophilic OMPs attack by ozone in molecular form. Moreover, ozone has a stronger oxidation-reducing potential (2.07 V) than oxygen and even hydrogen dioxide. The synergic enhancement of effectiveness observed in this study could be attributed to the improved thermolytic decomposition of O₃ and subsequent increase of OH^{*} according to the following equations [29,60]:



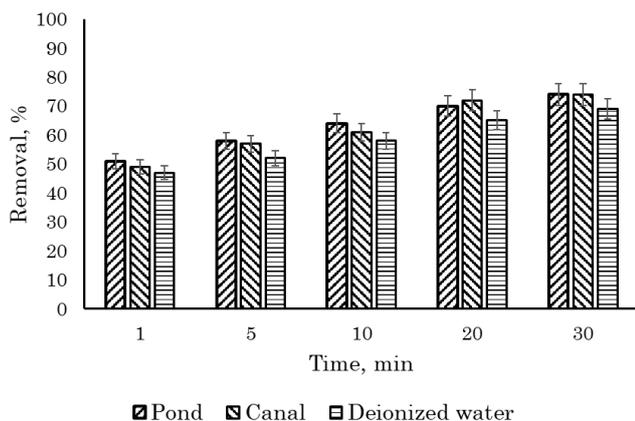


Fig. 8. Effectiveness of ultrasonication in BPA removal from water (volume of the treated sample 100 mL, pH not regulated, temperature 25°C, power 57 W, 20 kHz frequency, matrice: canal, pond and deionized water containing 1 mg·L⁻¹ OMPs).

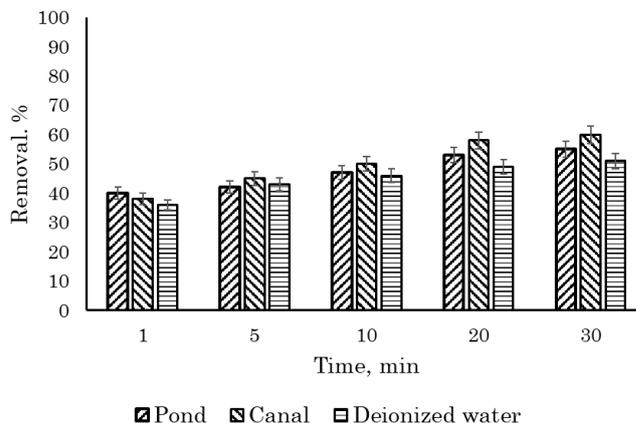


Fig. 10. Effectiveness of ultrasonication in TCS removal from water (volume of the treated sample 100 mL, pH not regulated, temperature 25°C, power 57 W, 20 kHz frequency, matrice: canal, pond and deionized water containing 1 mg·L⁻¹ OMPs).

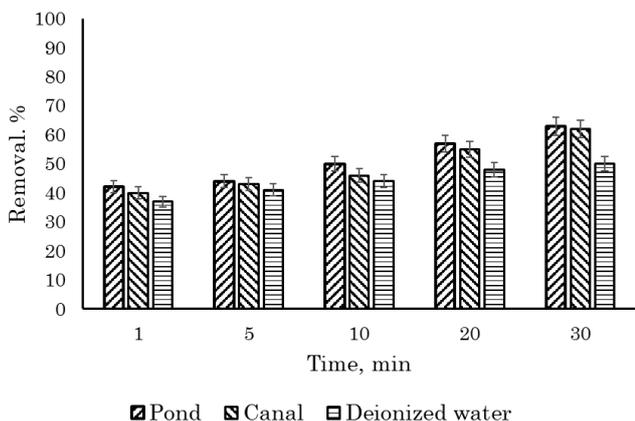


Fig. 9. Effectiveness of ultrasonication in EE2 removal from water (volume of the treated sample 100 mL, pH not regulated, temperature 25°C, power 57 W, 20 kHz frequency, matrice: canal, pond and deionized water containing 1 mg·L⁻¹ OMPs).

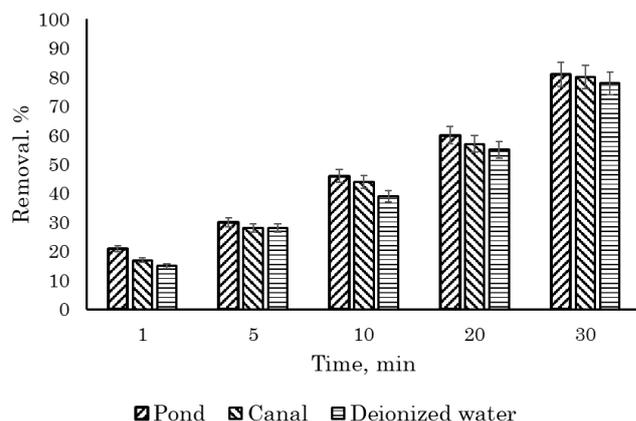


Fig. 11. Effectiveness of ultrasonication in PYR removal from water (volume of the treated sample 100 mL, pH not regulated, temperature 25°C, power 57 W, 20 kHz frequency, matrice: canal, pond and deionized water containing 1 mg·L⁻¹ OMPs).

3.4. Sand and glass particles effect on OMPs removal efficiency

In ultrasonic cavitation solid particles are used as supportive materials to accelerate the degradation rate of pollutants. As heterogeneous catalysts, their presence in the solution increases the number of cavitation bubbles by acting as additional nuclei [61]. Moreover, the presence of solid particles leads to the reduction of the cavitation threshold, an increase in the liquid temperature, and an increase in hydrodynamic effects resulting in oxidants formation enhancement [62,63]. In the study, the addition of solid particles (sand and glass) increased the OMPs degradation rate (Fig. 14). The ultrasonication effectiveness was slightly proportional to the dosage of catalyst (increased dosage of solid particles resulted in more acoustic bubbles. Maximum removal degree was obtained while 20 g glass particles were added to the treated solution. In that case, effectiveness varied from 58% to 86%. Furthermore, the glass catalyst was more effective than sand particles at the same dosage. Obtained relation might be related to

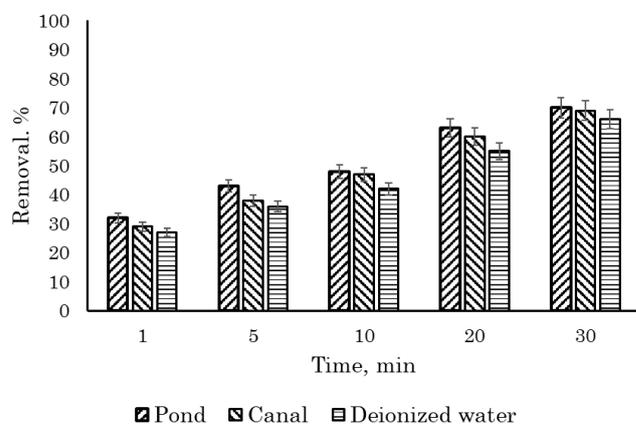


Fig. 12. Effectiveness of ultrasonication in CBZ removal from water (volume of the treated sample 100 mL, pH not regulated, temperature 25°C, power 57 W, 20 kHz frequency, matrice: canal, pond, and deionized water containing 1 mg·L⁻¹ OMPs).

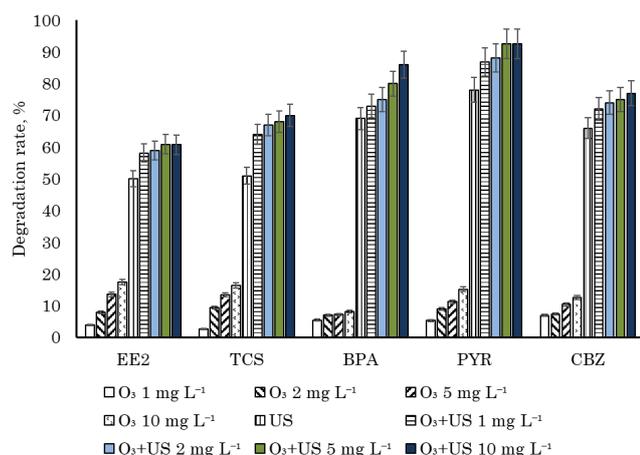


Fig. 13. Decomposition of BPA, CBZ, EE2, PYR, and TCS in ozonation, ultrasonication, and ultrasonication combined with ozonation processes (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, the dosage of O₃: 1, 2, 5, 10 mg·L⁻¹, 30 min sonication time, 57 W power, matrice: deionized water containing a mixture of 1 mg·L⁻¹ BPA, CBZ, EE2, PYR, and TCS).

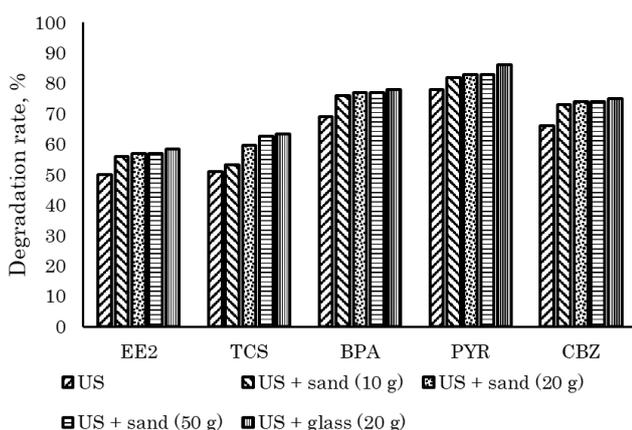


Fig. 14. Decomposition of BPA, CBZ, EE2, PYR, and TCS in ultrasonication and ultrasonication combined with sand and glass addition (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, the dosage of sand: 10, 20, 50 g, the dosage of glass: 20 g, 30 min sonication time, 57 W power, matrice: deionized water containing a mixture of 1 mg·L⁻¹ BPA, CBZ, EE2, PYR, and TCS).

the more irregular shape of the glass particles and thus generation of more bubbles and enhanced hydrodynamical effects. The average enhancement of the process after the addition of 50 g sand was equal to 13% compared to 30 min sonication at maximum power used alone. However, the addition of heterogenous catalysts was slightly less effective than the addition of 10 mg·L⁻¹ of O₃.

3.5. Toxicity assessment

Based on the Microtox[®] bioassay performed in this study it can be concluded that after ultrasonication treatment

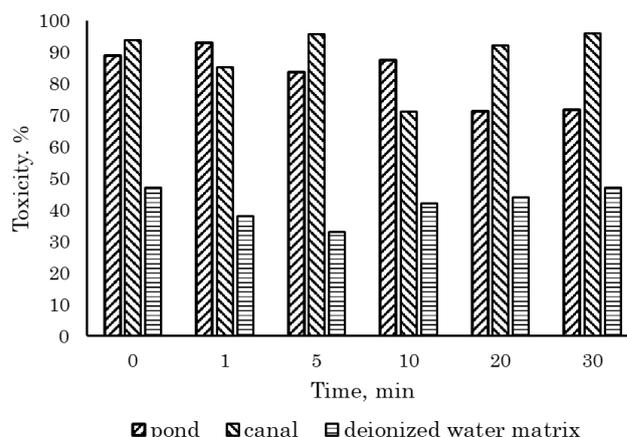


Fig. 15. Toxicity effect measured in Microtox[®] after sonication (volume of the treated sample 100 mL, pH = 7.0, temperature 25°C, 20 kHz frequency, 57 W power, matrice: pond water, canal water, and deionized water containing a mixture of 1 mg·L⁻¹ BPA, CBZ, EE2, PYR, and TCS), an average of 5 and 15 min of bacteria exposure time.

toxicity of the post-treated sample was not significantly reduced. The results of the toxicity assessment are given in Fig. 15.

It can be seen that the toxic effect varied from 71 to 93, from 71 to 96, and from 33% to 47% for pond, canal, and deionized water, respectively. However, after 30 min sonication of pond water toxicity effect changed from highly toxic to toxic. Worthnothy, initial toxicities of water from the pond and the canal were significantly higher than the toxicity of deionized water containing OMPs. Moreover, results indicated that the TOC value of the post-treated samples was not reduced significantly (results not shown). Obtained relation may be associated with the formation of decomposition by-products which can be potentially toxic to living organisms. Furthermore, formatted intermediates can be even more harmful to human health than the initial compound. Nowadays, the problem of water toxicity after AOPs treatment attracts the attention of many scientists which confirms the need and importance of extensional ecotoxicological analysis [27,64–69].

4. Conclusions

To sum up, the increase of harmful substances in water as a result of anthropogenic activity necessitates the search for alternative methods of water treatment. This work showed that ultrasonication can be very effective in that field. As expected, the degradation rate of OMPs increased as the treatment time and ultrasound power increased. Furthermore, the effectiveness of the acoustic cavitation process can be enhanced by combining ultrasonication with ozonation treatment or the addition of solid particles to the solution. It was noted that the removal rate of OMPs was proportional to the dosage of ozone and the dosage of the glass and sand particles. Moreover, this research showed that the water matrix had a slight impact on sonication effectiveness and the degradation rates of OMPs generally increase in the order: deionized water, canal water, and pond water. Furthermore,

O₃/US process was slightly more effective than usage of ultrasonication with sand and glass particles. Although the removal rate of OMPs during ultrasound treatment was high it should be pointed out that the toxicity of the post-treated samples was relatively high thus some intermediates could be generated. Therefore, ultrasound treatment can be a very effective technique, however further detailed analysis is needed to be concentrated on generated by-products and the impact of the post-treated sample on the environment.

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