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Effects of ultrasonication on the DBP formation and toxicity during chlorination of saline wastewater effluents ☆

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ABSTRACT

Chlorine disinfection of saline wastewater effluents rich in bromide and iodide forms relatively toxic brominated and iodinated disinfection byproducts (DBPs). Ultrasonication is a relatively new water treatment technology, and it is less sensitive to suspended solids in wastewaters. In this study, we examined the effects of ultrasonication (in terms of reactor type and combination mode with chlorination) on the DBP formation and toxicity in chlorinated primary and secondary saline wastewater effluents. Compared with the chlorinated wastewater effluent samples without ultrasonication, ultrasonic horn pretreatment of the wastewater effluent samples reduced the total organic halogen (TOX) levels in chlorination by ~30%, but ultrasonic bath pretreatment of the wastewater samples did not significantly change the TOX levels in chlorination, which might be attributed to the higher energy utilization and decomposition extent of organic DBP precursors in the ultrasonic horn reactor. Moreover, the TOX levels in the chlorinated samples with ultrasonic horn pretreatment (USH–chlorination), simultaneous treatment (chlorination+USH) and subsequent treatment (chlorination–USH) were also significantly reduced, with the maximum TOX reductions occurring in the samples with ultrasonic horn pretreatment. A toxicity index was calculated by weighting and summing the levels of total organic chlorine, total organic bromine and total organic iodine in each treated sample. The calculated toxicity index values of the chlorinated wastewater effluent samples followed a descending rank order of "chlorination" > "chlorination+USH" > "chlorination–USH" > "USH–chlorination", with the lowest toxicity occurring in the samples with ultrasonic horn pretreatment. Then, a developmental toxicity bioassay was conducted for each treated sample. The measured toxicity index values of the chlorinated wastewater samples followed the same descending rank order.

☆ This manuscript is dedicated to Prof. Michael Plewa of University of Illinois at Urbana-Champaign. His pioneering work on the toxicity evaluation of DBPs with *in vitro* bioassays has opened a new window for DBP studies and improved the safety of drinking water.

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Introduction

Seawater has been applied for toilet flushing in many coastal regions to reduce the dependence on freshwater resources (Ali et al., 2020; Li et al., 2018b; Tang et al., 2007; Mirti and Davies, 2005). However, such a practice introduces high levels of salts and bromide and iodide ions into the corresponding wastewater treatment system. For instance, the concentrations of bromide and iodide in the saline wastewater effluents in Hong Kong range from 20–31 mg/L as Br^- and 30–60 μg/L as $I^- + IO_3^-$ (Yang et al., 2015). To prevent pathogenic waterborne diseases, disinfection is an essential step in wastewater treatment before wastewater effluents are discharged into receiving water bodies (Grote et al., 2022; Le Roux et al., 2017; Dong et al., 2017a, 2017b). Chlorine is the most extensively used disinfectant for wastewater disinfection, due to its well-established application practices, broad-spectrum bactericidal potency and relatively low cost (Li et al., 2017a; Lee and von Gunten, 2010; Drinan and Spellman, 2012). However, the reaction of chlorine with effluent organic matter and inorganic ions unintentionally generates various halogenated disinfection byproducts (DBPs) (Xiao et al., 2022; Le Roux et al., 2017; Dong et al., 2017a; Tang et al., 2012; Krasner et al., 2009; Yang et al., 2005). Additionally, chlorination of water rich in bromide and iodide ions results in the formation of brominated and iodinated DBPs (Zha et al., 2014; Yang et al., 2014; Sun et al., 2009; Hua et al., 2006), which are substantially more toxic than their chlorinated analogues (Wawryk et al., 2021; Plewa et al., 2017; Dong et al., 2017b; Li et al., 2016; Liu and Zhang, 2014; Yang and Zhang, 2013; Wei et al., 2013; Richardson et al., 2007). Toxicological studies have shown that many halogenated DBPs can induce developmental toxicity and growth inhibition in aquatic organisms based on *in vivo* bioassays (Yang and Zhang, 2013; Liu and Zhang, 2014; Itoh et al., 2011). Accordingly, when halogenated DBPs in chlorinated saline wastewater effluents containing are discharged into receiving water bodies, they may cause adverse effects on the aquatic ecosystems or even public health.

Total organic halogen (TOX) has been widely used as a good surrogate and a collective parameter for the overall halogenated DBPs in a disinfected water sample (Kim et al., 2020; Zhu and Zhang, 2016; Kristiana et al., 2009; Simmons et al., 2002). Generally, TOX can be segmented into total organic chlorine (TOCl), total organic bromine (TOBr) and total organic iodine (TOI) (Chen et al., 2020; Kristiana et al., 2009; Hua and Reckhow, 2008; Zhang et al., 2000; Echigo et al., 2000). Studies have indicated that, for the same source water, the toxicity of the disinfected water is positively correlated with the TOX level in the disinfected water (Kristiana et al., 2020; Han and Zhang, 2018; Han et al., 2017; Lv et al., 2017; Yang et al., 2014; Pan et al., 2014; Echigo et al., 2004), thus the toxicity trend of a water sample that is disinfected under different scenarios can be well explained by the TOX trend of the disinfected water sample.

Ultrasonication is a clean technology without adding any chemicals or generating any byproducts when used alone, and it has been gaining attention in water and wastewater treatment (Zhou et al., 2016; Naddeo et al., 2014; Mahvi, 2009). Researchers have found that a combination of chlorination and ultrasonication in water treatment is very effective in improving the disinfection efficiency (Luhovskyi et al., 2018; Zhou et al., 2016; Gao et al., 2014; Mahvi, 2009; Mezule et al., 2009; Blume and Neis, 2005). Although the operational cost of disinfection with ultrasonication was about 20% higher than that with UV light under the same disinfection efficacy, disinfection with ultrasonication was less sensitive to the level of suspended solids in waters than that with UV light (Lebedev et al., 2019; Jin et al., 2013; Lambert et al., 2010; Hulsmans et al., 2010; Gibson et al., 2009). When ultrasonication was applied simultaneously with chlorination to treat a secondary wastewater effluent, the disinfection efficiency was improved from 0.70- to 1.40-log, compared with chlorination alone (Blume and Neis, 2005). Most previous research on the combination of chlorination and ultrasonication has focused on disinfection efficiency (Luhovskyi et al., 2018; Ayyildiz et al., 2011; Hulsmans et al., 2010; Blume and Neis, 2005), little is known about the formation and toxicity of DBPs induced by the combination of chlorination and ultrasonication. Previous studies have shown that ultrasonication can break down large organic particles in wastewater effluent to smaller (aromatic) organic compounds and even further decompose them into aliphatic compounds, carbon dioxide and inorganic ions (Zhou et al., 2016; Rayaroth et al., 2016; Nagata et al., 2000; Jiang et al., 2002). Therefore, it is expected that ultrasonication could decrease the DBP level and toxicity in chlorinated wastewater effluents via two ways, i.e., by decomposing DBP precursors or by transforming intermediate aromatic DBPs into aliphatic ones. However, it remains unknown how to combine ultrasonication with chlorination in disinfecting saline wastewater effluents so that relatively low levels of DBPs and toxicity in the disinfected effluents can be achieved.

Accordingly, the present paper aimed to: (i) investigate the effect of ultrasonic reactor type on the formation of halogenated DBPs; (ii) compare the TOX formation in chlorinated saline wastewater samples without ultrasonication and with different combination modes of chlorination and ultrasonication; and (iii) study the effect of ultrasonication on developmental toxicity during chlorination of saline wastewater effluents.

1. Materials and methods

1.1. *Chemicals*

Ultrapure water (18.2 MΩ•cm) was generated by a Cascada I Laboratory Water Purification System (PALL, USA). A chlorine stock solution was prepared by diluting a reagent grade NaOCl solution (Sigma–Aldrich), and its concentration was determined by the DPD/FAS titration method (APHA et al., 2012). Chemicals, including sodium thiosulfate (1.0 mol/L), methyl *tert*-butyl ether, and methanol, were ordered from Sigma–Aldrich with reagent grade or higher purity.

1.2. *Wastewater sampling and characterization of the saline wastewater effluents*

Two saline wastewater effluent samples (without disinfection, 24 hr composite) were collected from a primary wastewater

Table 1 – Characteristics of the primary and secondary saline wastewater effluent samples.

	Primary effluent	Secondary effluent
pH[a]	7.12	7.22
Salinity (%)[a]	1.8	1.2
Dissolved organic carbon (mg/L as C)[b]	22.7	7.4
Ammonia (mg/L as N)[b]	24.9	0.8
Total suspended solids (mg/L)[c]	44.0	19.0

[a] The pH and salinity were measured with a pH meter (Thermo Scientific, Orion Star A111) and a portal refractometer, respectively.
[b] The concentrations of dissolved organic carbon and ammonia were measured with a total organic carbon analyzer (Shimadzu, Japan) and a flow injection analysis system (Lachat, 8500 Series), respectively.
[c] The data of total suspended solids was collected from the Hong Kong Drainage Service Department (HKDSD, 2017).

treatment plant and a secondary wastewater treatment plant (Appendix A Fig. S1). The collected samples were delivered to the laboratory immediately in an ice-cold condition and stored at 4°C in darkness to minimize changes in the constituents. Some basic characteristics of the two saline wastewater effluent samples are shown in Table 1.

1.3. *Chlorination/ultrasonication of the saline wastewater effluents*

Prior to the experiments, the wastewater effluent samples were brought to room temperature and passed through an 11-μm pore size filter to remove large particles. Aliquots of the filtered wastewater sample were disinfected by dosing 10 mg/L NaOCl as Cl_2 for a 30-min contact time. This chlorination condition was selected mainly because it could fulfill the disinfection goal (HKDSD, 2017). After the given contact time, the chlorine residual in each aliquot was dechlorinated with 0.1 mol/L sodium thiosulfate at 105% of the requisite stoichiometric amount for a 30 min dechlorination time (Pan et al., 2019).

To investigate the effect of ultrasonic type on DBP formation, two typical ultrasonic reactors were selected, i.e., ultrasonic horn and ultrasonic bath. The ultrasonic horn (Sonics and Materials, VC 750), equipped with a probe of 1.27 cm in diameter, has an operating frequency of 20 kHz and a power output of 600 W. During the ultrasonication, the probe was submerged 5 cm into a 1000-mL wastewater sample that was stored in a 2000-mL glass bottle. To prevent the temperature increase of the wastewater sample, the ultrasonic horn was operated in a pulsed mode (on-time 25 sec and off-time 5 sec), which enabled the system to cool down during the off-time period. Additionally, the temperature of the wastewater sample was maintained at 25±2 °C with the aid of an ice bath (Fig. 1a). The ultrasonic bath (Branson, 3800 Series) has an operating frequency of 40 kHz and a power output of 110 W. The bath has the internal dimensions of 29 × 15 × 15 cm^3 (length × width × depth). Its internal body is made up of stainless steel and two transducers are located at the bottom of the bath (Fig. 1b). Ultrasonication was continuously supplied in the ultrasonic bath. The water level inside the bath was maintained by continuous circulation of water, and subsequently the temperature was also controlled at 25±2 °C. To reveal the combination effect of ultrasonication and chlorination, a 30-min ultrasonication with ultrasonic horn was applied in three combination modes with chlorination: 1) ultrasonication prior to chlorination ("USH–chlorination"), 2) applying ultrasonication and chlorination simultaneously ("chlorination+USH"), 3) chlorination followed by ultrasonication ("chlorination–USH"). A schematic diagram of sample treatment processes is illustrated in Appendix A Fig. S2.

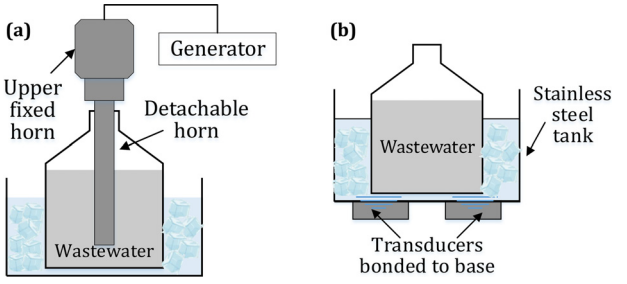

Fig. 1 – Schematic diagram of (a) an ultrasonic horn system and (b) an ultrasonic bath system.

1.4. *TOX analysis*

The TOCl, TOBr and TOI analysis was based on a previous study (Li et al., 2017a) with minor modifications. The modified procedure is given in Appendix A Text S1 and Fig. S3. Briefly, a 60-mL aliquot of a dechlorinated sample was acidified to pH 2 with nitric acid and passed through two consecutive activated carbon columns. The activated carbon columns were rinsed with a given volume of a KNO_3 solution, and the activated carbon in each column was subsequently combusted at 1000°C. The hydrogen halide and halogen gases were absorbed in 8 mL of ultrapure water. The concentrations of chloride and bromide ions in the absorption solution were measured with an ion chromatography system with a conductivity detector (Dionex, CA). The concentration of iodide ions in the absorption solution was measured following the method by Pan and Zhang (2013). Briefly, 3 mL of the absorption solution was acidified to pH < 3 with formic acid and sparged with pure nitrogen at 150 mL/min for 2 min. Then, the treated absorption solution was analyzed by ultra performance liquid chromatography/electrospray ionization-triple quadrupole mass spectrometry, whose operation parameters were set according to Pan and Zhang (2013).

1.5. *Comparative development toxicity bioassay*

An improved *in vivo* bioassay, using the sensitive embryo–larval stages of *Platynereis dumerilii*, has been successfully applied in determining the comparative developmental toxicity of individual DBPs and DBP mixtures (Han et al., 2017, 2021). This bioassay was adopted in this study for the developmental toxicity evaluation of chlorinated saline wastewater effluents without and with ultrasonication. The conditions for culturing *P. dumerilii* followed those in previous studies (Hutchinson et al., 1995; Yang and Zhang, 2013), with details

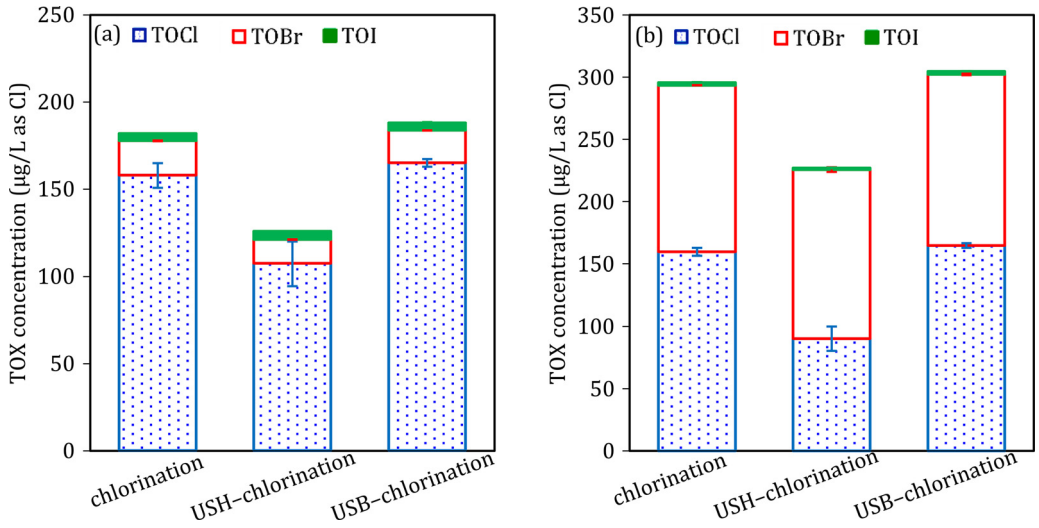

Fig. 2 – TOCl, TOBr and TOI concentrations in chlorinated (a) primary and (b) secondary saline wastewater effluent samples without ultrasonication, with ultrasonic horn pretreatment, and with ultrasonic bath pretreatment. Each error bar indicates the standard deviation of triplicate measurements.

as described in Appendix A. The procedures for concentrating the samples and conducting the developmental toxicity bioassay are also given in Appendix A.

2. Results and discussion

2.1. *Effect of ultrasonic type on the DBP formation in chlorinated saline wastewater effluents*

Fig. 2 shows the TOCl, TOBr and TOI concentrations in the chlorinated primary and secondary saline wastewater samples without and with ultrasonic pretreatment. TOCl was the main component of TOX formed in the primary wastewater samples, while both TOBr and TOCl were the main components of TOX formed in the secondary wastewater samples. Compared with the chlorinated secondary wastewater sample, a relatively lower TOBr concentration and a relatively higher TOI concentration in the chlorinated primary wastewater sample were observed, which might be due to the higher ammonia concentration in the primary wastewater effluent sample (24.9 mg/L as N). When 10 mg/L of chlorine was dosed to the primary wastewater effluent sample, monochloramine formed quickly and became the major disinfectant (Li et al., 2018a, 2017b); In contrast, when 10 mg/L of chlorine was dosed to the secondary wastewater effluent sample that contained a very low level of ammonia (0.80 mg/L as N), breakpoint chlorination should occur and the remaining free chlorine became the major disinfectant (Ding et al., 2013). It has been reported that the formation of TOBr was favored in chlorination than in chloramination, and the formation of TOI was favored in chloramination than in chlorination (Zhu and Zhang, 2016; Yang et al., 2015).

Fig. 2a shows the TOCl, TOBr and TOI concentrations in the chlorinated primary wastewater effluent samples without ultrasonication, with ultrasonic horn pretreatment, and with ultrasonic bath pretreatment, which were labeled as "chlorination", "USH–chlorination", and "USB–chlorination", respectively. Compared with the chlorinated primary effluent sample without ultrasonication, a remarkable TOX reduction of 31.6% was observed in the chlorinated primary effluent sample with ultrasonic horn pretreatment; However, the TOX level increased slightly by 3.4% in the chlorinated primary effluent sample with ultrasonic bath pretreatment ($p > 0.10$, not significant statistically). Fig. 2b shows a similar trend for the chlorinated secondary effluent samples. Compared with the chlorinated secondary effluent sample without ultrasonication, a significant TOX reduction of 23.2% was observed in the chlorinated secondary effluent sample with ultrasonic horn pretreatment, while a slight TOX increase of 3.0% was found in the chlorinated secondary effluent sample with ultrasonic bath pretreatment ($p > 0.10$, not significant statistically).

The different effects of ultrasonication on the DBP formation might be attributed to the different degrees of organic particle decomposition induced by the two ultrasonic reactors, which might be ascribed to their different designs and energy utilization. It has been reported that the pyrolysis reaction in cavitation bubbles can result in an extremely high energy density and an extremely high local temperature (up to 5000°C) (Islam et al., 2019; Rayaroth et al., 2016). During ultrasonication, some large organic particles in wastewater effluents could be broken down to small organic particles, which could be further decomposed to aromatic compounds (e.g., DBP precursors) via the high local temperature. Besides, free radicals (e.g., hydroxyl radical) can be generated from water molecules during the cavitation (Rayaroth et al., 2016). The aromatic compounds are generally susceptible to hydroxyl radical attack, and then they are cleaved to low molecular weight aliphatic compounds via decarboxylation, demethylation, and deamination (Lei et al., 2019; Rayaroth et al., 2016). Further reactions could lead to a certain degree of mineralization during ultrasonication. Accordingly, a general decomposition and degradation pathway of organic particles in wastewater effluents by ultrasonication was proposed as illustrated in

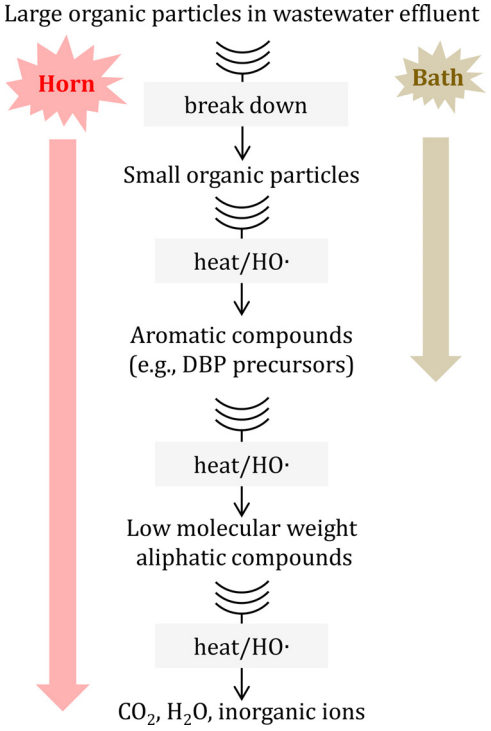

Fig. 3 – Proposed decomposition and degradation pathway of large organic particles in wastewater effluents by ultrasonic horn and ultrasonic bath treatment.

Fig. 3. For the ultrasonic horn, since the ultrasonic generator directly contacted the wastewater, the ultrasonic energy could be utilized to a higher extent by wastewater (Fig. 1a), and thus some large organic particles could go through the proposed pathway of decomposition and degradation. As a result, the ultrasonic horn might reduce the level of DBP precursors and eventually reduce the formation of halogenated DBPs in the subsequent chlorination (Fig. 2). For the ultrasonic bath, since the ultrasonic generator did not directly contact the wastewater (Fig. 1b), some energy could be absorbed by the water in the bath, leading to relatively low ultrasonic energy utilization by the wastewater. Thus, large organic particles in wastewater effluents might be broken down to aromatic DBP precursors only and their further decomposition might be limited. As a result, more DBP precursors could form in the ultrasonic bath treatment and eventually increase the DBP formation in the subsequent chlorination (Fig. 2). To verify the proposed pathway, the UV absorbance at 254 and 272 nm of the primary effluent sample treated with ultrasonic horn alone or ultrasonic bath alone was measured. The UV_{254} and UV_{272} absorbance values of the primary effluent sample treated with ultrasonic horn alone were 0.193±0.002 and 0.157±0.002, respectively; and the UV_{254} and UV_{272} absorbance values of the primary effluent sample treated with ultrasonic bath alone were 0.200±0.001 and 0.160±0.001, respectively. A higher UV absorbance value indicates a higher aromaticity level of the sample. The one-tailed *t*-statistical significance analysis was conducted per Li et al. (2017a); The analysis showed that for either the UV_{254} or UV_{272} value, the UV absorbance of the primary effluent sample treated with ultrasonic horn was significantly lower than that treated with ultrasonic bath ($p < 0.10$, Appendix A Table S1), suggesting that a higher portion of aromatic compounds might be decomposed to non-aromatic ones (aliphatic compounds or even inorganic compounds) in the effluent sample treated with ultrasonic horn than in the effluent sample treated with ultrasonic bath. Previous studies have demonstrated that aromatic compounds are good precursors of aromatic halogenated DBPs, which are intermediate DBPs and may further decompose to aliphatic halogenated DBPs (Hu et al., 2022; Han et al., 2021; Jiang et al., 2020; Nihemaiti et al., 2017; Chuang et al., 2015; Zhai and Zhang, 2011).

2.2. *Effect of ultrasonic mode on the DBP formation in chlorinated saline wastewater effluents*

As stated above, the ultrasonic horn performed better in decomposing aromatic compounds than the ultrasonic bath, so it was selected to further investigate the effect of ultrasonic mode on the DBP formation in chlorinated saline wastewater effluents. Fig. 4 shows the TOX concentrations in the chlorinated effluent samples without ultrasonic horn treatment (i.e., "chlorination"), and with ultrasonic horn pretreatment (i.e., "USH–chlorination"), simultaneous treatment (i.e., "chlorination+USH"), and subsequent treatment (i.e., "chlorination–USH").

Fig. 4a shows the results of primary effluent samples. Compared with the primary effluent sample treated by chlorination alone, the TOCl levels decreased significantly in the samples treated by chlorination in combination with ultrasonic horn treatment. Because TOCl was the main component of TOX, the TOX concentration in the chlorinated primary effluent sample with ultrasonic horn treatment was significantly lower than that without ultrasonic horn treatment. The TOX concentration in the chlorinated primary effluent sample was 184 μg/L as Cl, while the combination of USH–chlorination, chlorination+USH or chlorination–USH decreased the TOX concentration to 126, 168 or 137 μg/L as Cl, respectively. The maximum reduction of TOX was 32%, which occurred in the primary effluent sample with USH–chlorination.

As proposed above, a large portion of DBP precursors could decompose by the ultrasonic horn pretreatment, resulting in the lowest level of DBP precursors and then the lowest level of DBPs in the effluent samples treated with USH–chlorination. During the simultaneous treatment with chlorination and ultrasonication (i.e., chlorination+USH), some DBP precursors might be decomposed by ultrasonication, while some other organic molecules might be activated by ultrasonication to become DBP precursors. It has been reported that free radicals were formed during ultrasonication (Nie et al., 2021; Rahman et al., 2020; Gibson et al., 2008). The formed radicals, especially hydroxyl radicals, could react with aromatic compounds in the effluent to form phenolic compounds (Nie et al., 2021; Rahman et al., 2020). Due to the electron dominating property of phenolic compounds, chlorine substitution tends to occur on the benzene ring, resulting in the formation of halogenated DBPs (Jiang et al., 2020). Therefore, the effluent sample treated with chlorination+USH resulted in a higher level of TOX than that treated with USH–chlorination. For the subsequent ultrasonic horn treatment (i.e., chlorination–USH),

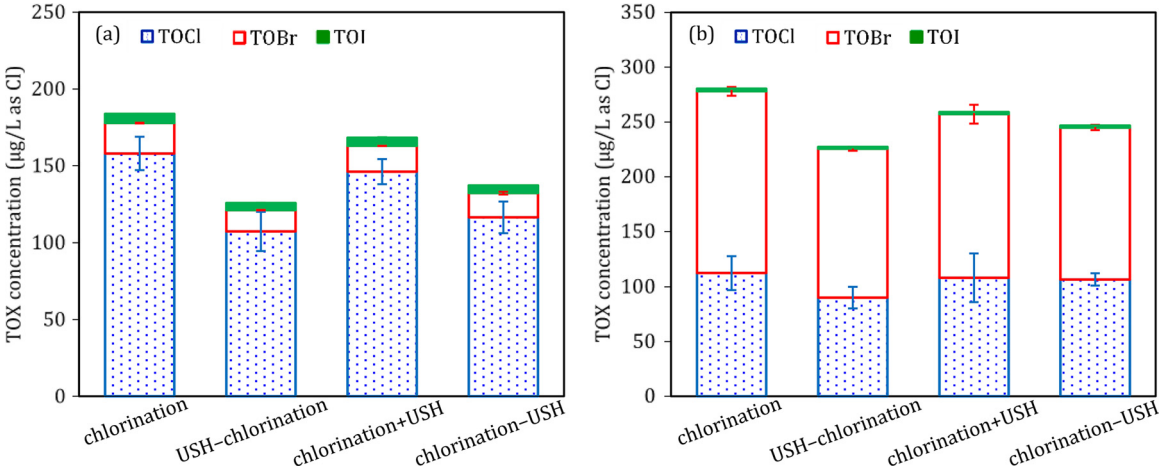

Fig. 4 – TOCl, TOBr and TOI concentrations in chlorinated (a) primary and (b) secondary wastewater effluent samples without ultrasonic horn treatment, and with ultrasonic horn pretreatment, simultaneous treatment and subsequent treatment. Each error bar indicates the standard deviation of triplicate measurements.

it had no chance to affect DBP precursors, but it might decompose the already-formed DBPs in chlorination and transform them into organic acids, carbon dioxide and inorganic ions. Therefore, a higher reduction of TOX was observed in the effluent sample treated with USH–chlorination than in the effluent sample treated with chlorination+USH. Fig. 4b shows a similar trend for the secondary effluent samples. Compared with chlorination alone, the levels of TOCl and TOBr reduced remarkably when chlorination was combined with ultrasonic horn treatment. The TOX concentration in the chlorinated secondary effluent sample was 280 μg/L as Cl, while the combination of USH–chlorination, chlorination+USH or chlorination–USH decreased the TOX concentration to 227, 259 or 247 μg/L as Cl, respectively. The maximum reduction of TOX was 19%, which occurred in the secondary effluent sample also with USH–chlorination. As discussed in Section 2.1, DBP precursors could be minimized in the ultrasonic horn pretreatment owing to its higher energy utilization. As a result, the chlorinated effluent sample with the ultrasonic horn pretreatment formed the lowest level of TOX.

For the same source water, the toxicity of the disinfected watert has been reported to be positively correlated with the TOX level in the disinfected water (Han et al., 2017; Lv et al., 2017; Pan et al., 2014; Echigo et al., 2004). Richardson et al. (2007) reviewed the toxicity data and found that iodinated and brominated DBPs were averagely 450 × and 125 ×, respectively, more toxic than their chlorinated analogues. Zhu and Zhang (2016) proposed a toxicity index by weighting and summing the levels of TOCl, TOBr and TOI in a disinfected water sample. To be specific, they assigned the toxicity potency weighting of TOCl, TOBr and TOI (all in μmol/L) as 1, 125 and 450, respectively, and then they calculated the toxicity index of the disinfected water sample as "1 × [TOCl] + 125 × [TOBr] + 450 × [TOI]". With that, the calculated toxicity index values were 146, 108, 127 and 119 for the chlorinated primary effluent samples without ultrasonic horn treatment, and with ultrasonic horn pretreatment, simultaneous treatment and subsequent treatment, respectively; the calculated toxicity index values were 612, 496, 549 and 509 for the secondary effluent samples of "chlorination", "USH–chlorination", "chlorination+USH" and "chlorination–USH", respectively. For either the primary or secondary effluent, the calculated toxicity index values of the four chlorinated effluent samples followed a descending rank order of "chlorination" > "chlorination+USH" > "chlorination–USH" > "USH–chlorination", indicating that the wastewater effluent that was pretreated with ultrasonic horn (i.e., USH–chlorination) formed the lowest toxicity of the DBP mixture during chlorination.

2.3. *Developmental toxicity of the chlorinated saline wastewater effluents without and with ultrasonication*

To verify the calculated toxicity index values of the chlorinated saline wastewater effluents without and with ultrasonication, the developmental toxicity bioassay was conducted with the sensitive embryo–larval stages of *P. dumerilii*. Fig. 5 shows the concentration factor (CF)–response curves of the developmental toxicity of chlorinated effluent samples without and with precedent, simultaneous and subsequent ultrasonic horn treatments. This figure included four charts representing the toxicity tests for the primary and secondary effluent samples conducted on two different days. As shown in Fig. 5a and b, the normal developmental percentage of the chlorinated primary effluent sample at CF < 1 × was close to the seawater control; it decreased dramatically as the CF increased; and it reached 0% at CF 3.5 × . Similar trends were observed for chlorinated secondary effluent samples as shown in Fig. 5c and d. The normal developmental percentage of the chlorinated secondary effluent sample at CF 20 × was close to the seawater control, and then it decreased dramatically in the CF range from 20 × to 80 × . All the four charts indicated that the development toxicity of the four chlorination scenarios followed a descending rank order of "chlorination" > "chlorination+USH" > "chlorination–USH" > "USH–chlorination", which was consistent with the rank order of the calculated toxicity index values (in Section 2.2).

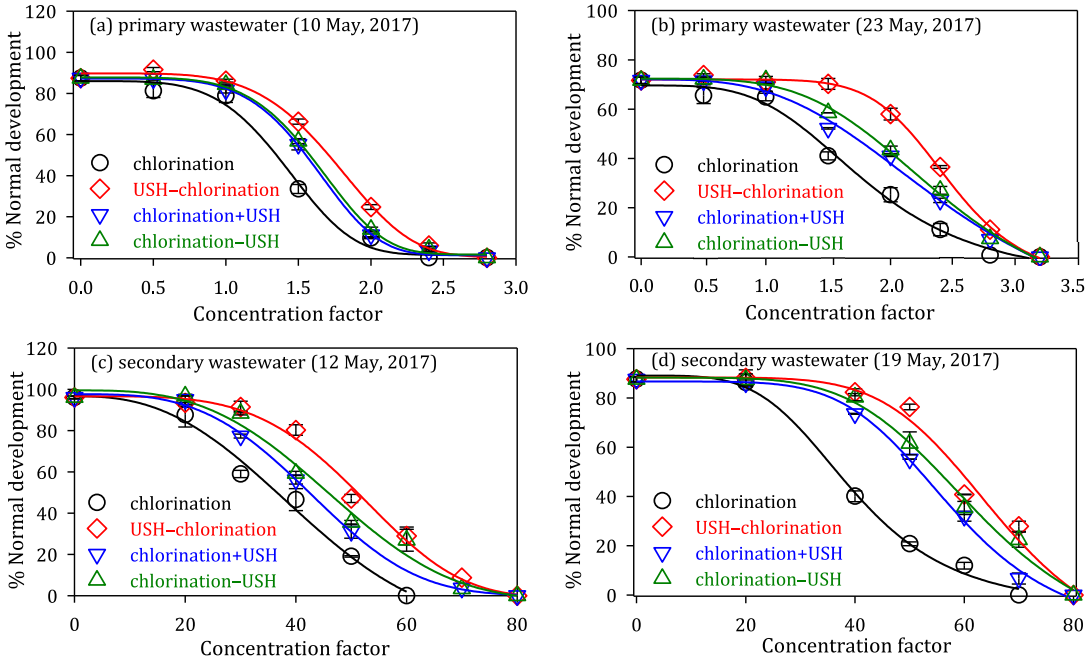

Fig. 5 – Developmental toxicity of the chlorinated (a, b) primary and (c, d) secondary saline wastewater effluent samples without and with ultrasonic horn treatment. Different charts represent toxicity tests conducted on different days. Each datum presents the mean of duplicate measurements and the difference between the mean and the measured value.

Table 2 – Developmental toxicity of the chlorinated primary and secondary wastewater effluent samples without and with ultrasonic horn treatment.

(a) Chlorinated primary effluent sample	Test conducted on 10 May, 2017				Test conducted on 23 May, 2017			
	EC_{50}±SD[a]	Toxicity index[b]	Regression coefficient	Statistic value[c]	EC_{50}±SD	Toxicity index	Regression coefficient	Statistic value
Chlorination	1.42±0.01	704	0.992	–	1.72±0.08	581	0.993	–
USH–chlorination	1.77±0.03	565	0.999	$p<0.05$	2.39±0.03	418	0.999	$p<0.05$
Chlorination+USH	1.62±0.03	617	0.999	$p<0.05$	2.05±0.01	488	0.995	$p<0.05$
Chlorination–USH	1.65±0.01	606	0.999	$p<0.05$	2.16±0.03	463	0.997	$p<0.05$
(b) Chlorinated secondary effluent sample	Test conducted on 12 May, 2017				Test conducted on 19 May, 2017			
	EC_{50}± SD	Toxicity index	Regression coefficient	Statistic value	EC_{50}±SD	Toxicity index	Regression coefficient	Statistic value
Chlorination	37.2±1.1	26.9	0.988	–	39.7±1.3	25.2	0.997	–
USH–chlorination	51.7±0.3	19.4	0.994	$p<0.05$	61.6±0.2	16.2	0.983	$p<0.05$
Chlorination+USH	42.7±0.1	23.4	0.999	$p<0.05$	54.6±0.7	18.3	0.997	$p<0.05$
Chlorination–USH	45.9±0.8	21.8	0.987	$p<0.05$	58.1±1.3	17.2	0.994	$p<0.05$

[a] The standard deviation of duplicate measurements for each chlorinated saline wastewater sample.
[b] The toxicity index value is the reciprocal of the EC_{50} value × 1000.
[c] Chlorinated saline wastewater effluent samples with ultrasonic horn treatment were compared to those without ultrasonic horn treatment per EC_{50} values.

A regression analysis was performed to obtain the EC_{50} value for each treated effluent sample. The EC_{50} values and the regression coefficients are summarized in Table 2. The regression coefficients for all the curves ranged from 0.983 to 0.999. To determine whether the EC_{50} values of chlorinated effluent samples without and with ultrasonic horn treatment were significantly different, one-tailed *t*-statistical significance analysis was conducted according to a previous study (Li et al., 2017a). The results (Table 2) indicated that, for either the primary or secondary effluent, the developmental toxicity of the chlorinated effluent sample treated by chlorination in combination with any ultrasonic horn treatment was significantly lower than that treated by chlorination alone.

The measured toxicity index of each sample was obtained as the reciprocal of the EC_{50} value × 1000 (Pan et al.,

2014) and listed in Table 2. The developmental toxicity rank order of the four chlorination scenarios was "chlorination" > "chlorination+USH" > "chlorination–USH" > "USH–chlorination", which matched well with the rank order of the calculated toxicity index values (in Section 2.2). Compared with the chlorinated primary effluent samples without ultrasonic treatment (i.e., "chlorination"), the developmental toxicity of the chlorinated samples with ultrasonic horn pretreatment, simultaneous treatment, and subsequent treatment (i.e., "USH–chlorination", "chlorination+USH" and "chlorination–USH", respectively) decreased by 20%–28%, 12%–16%, and 14%–20%, respectively. Also, compared with the chlorinated secondary effluent samples without ultrasonic treatment, the developmental toxicity of the "USH–chlorination", "chlorination+USH" and "chlorination–USH" samples decreased by 28%–36%, 13%–27%, and 19%–32%, respectively. Therefore, the ultrasonic horn treatment significantly reduced the developmental toxicity of both chlorinated primary and secondary effluent samples.

3. Conclusions

Effects of the ultrasonic reactor type and mode on the DBP formation and toxicity in the chlorinated primary and secondary saline wastewater effluents were evaluated.

Compared with the chlorinated wastewater effluent samples without ultrasonication, TOX was reduced by about 30% in the chlorinated samples with ultrasonic horn pretreatment, but it was slightly elevated in the chlorinated samples with ultrasonic bath pretreatment (not significant statistically). This could be ascribed to the higher energy utilization and decomposition extent of organic DBP precursors in the ultrasonic horn reactor than in the ultrasonic bath reactor.

Compared with the chlorinated wastewater effluent samples without ultrasonication, the TOX levels in the chlorinated samples with ultrasonic horn pretreatment (USH–chlorination), simultaneous treatment (chlorination+USH) and subsequent treatment (chlorination–USH) were reduced by 19%–32%, 8%–9% and 12%–26%, respectively, with the maximum TOX reductions occurring in the samples with ultrasonic horn pretreatment.

A toxicity index was calculated by weighting and summing the levels of TOCl, TOBr and TOI in each treated sample. For either the primary or secondary effluent, the calculated toxicity index values of the four chlorinated effluent samples followed a descending rank order of "chlorination" > "chlorination+USH" > "chlorination–USH" > "USH–chlorination", indicating that the wastewater effluent that was pretreated with ultrasonic horn formed the lowest toxicity of the DBP mixture during chlorination. Moreover, a developmental toxicity bioassay was conducted for each treated sample. The measured toxicity index values of the four chlorinated wastewater effluent samples followed the same rank order of "chlorination" > "chlorination+USH" > "chlorination–USH" > "USH–chlorination".

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2022.05.029.

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