

Original Research

# EDTA Degradation by UV/Chlorine Treatment: Efficiency, Formation of Oxidation Byproducts and Toxicity Evaluation

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## Abstract

UV and chlorination are worldwide employed tertiary treatment for disinfection/oxidation of secondary effluents in sewage treatment plants. However, the combined UV and chlorination process (UV/chlorine) for removing non-biodegradable EDTA in secondary effluents is still unknown. In this study, we evaluated UV/chlorine for EDTA degradation and compared its formation of typical oxidation byproducts and toxicity change with chlorination. UV/chlorine showed synergistic effect on EDTA degradation, with removal efficiency of higher than 90% within 60 s at chlorine dosage of 0.30 mM, compared to UV irradiation of less than 2% and chlorination of about 65%. Increasing chlorine dosage from 0.050 to 0.30 mM and solution pH from 5.0 to 7.0 accelerated EDTA degradation. The coexisting bicarbonate and nitrate showed slight effects on EDTA degradation, but natural organic matter exhibited remarkable inhibition. As well, about 50%-80% of EDTA in realistic secondary effluent was removed by UV/chlorine. Three chlorinated byproducts including trichloromethane (TCM), dichloroacetic acid and trichloroacetic acid were formed in both chlorination and UV/chlorine, and UV/chlorine treatment slightly enhanced the production of TCM. The acute toxicities of treated wastewater by UV/chlorine to luminescent bacterium and rice seeds were greater than that by chlorination, which deserves extensive concerns in advanced treatment processes for secondary effluents.

**Keywords:** EDTA, UV/chlorine, secondary effluent, chlorinated byproducts, toxicity

## Introduction

Ethylenediaminetetraacetic acid (EDTA) is one of the generally used complexing agents in electroplating, detergent, and pulp and paper industries [1-3]. Its annual

consumption exceeded to around 10000 t worldwide, approximately half of which is employed in pulp and paper industry due to the production of totally-chlorine free pulp [3, 4]. Resulting from the discharge of industrial effluents, considerable EDTA enters into the receiving water due to its poor biodegradability and adsorptivity to activated sludge. Its widespread occurrence was found in secondary effluents from

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sewage treatment plants and in surface water, and its concentration varied from 10 to 35 mg/L and from 10 to 500  $\mu\text{g/L}$ , respectively [3-5]. Once EDTA enters the water environment, it tends to form chelate complexes with metal ions and thus enhances the migration of trace metals and radionuclides from contaminated sediments/soils or disposal sites to the surface water or groundwater as well as improves their bioavailabilities [6-8]. As a result, its potential risks to the environment and human health increased. Thus, the advanced treatment is highly desired to removal the residual EDTA in secondary effluents.

EDTA is resistant to conventional methods of water and wastewater treatment such as coagulation, precipitation, filtration and biological processes [4, 9, 10]. In recent years, advanced oxidation processes (AOPs) have been proposed to be capable of degrading EDTA in water, including Fenton oxidation [3, 11], electrochemical oxidation [1, 12], and photo-oxidation (e.g., UV/ $\text{H}_2\text{O}_2$ ) [13, 14]. Nevertheless, some inherent disadvantages hindered the above AOPs. In brief, Fenton reagent commonly employed in acidic pHs, but the complexation of EDTA with ferrous ions decreased the generation of hydroxyl radicals ( $\text{HO}\cdot$ ) even if EDTA could make Fenton oxidation broaden to neutral pHs. The accumulated intermediates (e.g.,  $\text{NH}_3$  and small organic acids) during ozonation of EDTA were resistant to be further oxidized by  $\text{O}_3$ , leading to an incomplete oxidation of EDTA. UV/ $\text{H}_2\text{O}_2$  was sensitive to the solution chemistry and required a long UV exposure time with a powerful output due to the low UV absorption coefficient of  $\text{H}_2\text{O}_2$  ( $19.6 \text{ M}^{-1} \text{ cm}^{-1}$  at 254 nm) [15].

UV/chlorine is the latest advanced oxidation process being considered as an alternative to UV/ $\text{H}_2\text{O}_2$  with the higher UV absorption of  $\text{HClO}/\text{ClO}\cdot$  ( $59/66 \text{ M}^{-1} \text{ cm}^{-1}$  at 254 nm) [16, 17]. Recently, UV/chlorine has been developed to be capable of degrading micropollutants and persistent contaminants, such as pharmaceuticals and personal care products [15, 18], pesticides [19, 20], as well as taste and odor compounds [21]. Moreover, either UV or chlorination is worldwide employed for disinfection and/or oxidation of secondary effluents in sewage treatment plants. So the combination of both for removing the residual organic matters in the secondary effluents can be very easy to realize. The two main species including  $\text{HO}\cdot$  and chlorine radical ( $\text{Cl}\cdot$ ) formed from the photolysis of  $\text{HClO}/\text{ClO}\cdot$  are responsible for decontamination in UV/chlorine [16, 17].  $\text{HO}\cdot$  reacts very rapidly with EDTA at near diffusion-controlled rates [22]. Similarly,  $\text{Cl}\cdot$ , an selective oxidant with oxidation potentials of 2.47V, reacts rapidly with compounds containing amino and/or carboxylic group, such as aniline and benzoic acid, at rate constants of  $\sim 10^{10} \text{ M}^{-1} \text{ s}^{-1}$  [16, 23]. EDTA is rich in carboxylic and/or amino group, implying that  $\text{Cl}\cdot$  has potential in degrading EDTA. To the best of our knowledge, the removal of EDTA by UV/chlorine has not been reported.

In addition, the generation of chlorinated by-products is the issue of common concern when chlorine is used for oxidation or disinfection in drinking water treatment and wastewater reclamation, and examples of chlorinated byproducts are trihalomethanes (THMs) and haloacetic acids (HAAs) [24-26]. Recently, many studies examined the formation potential of chlorinated byproducts in UV/chlorine use, but the results were diverse. None of chlorinated byproducts was detected in the degradation of caffeine and N,N-diethyl-3-methylbenzamide by UV/chlorine [20]. UV/chlorine reduced the formation of THMs compared to chlorination in the degradation of some pharmaceuticals, e.g., diatrizoate and carbamazepine [27, 28]. However, some known chlorinated byproducts, e.g., THMs and HAAs, were significantly enhanced when the UV/chlorine process was employed for degrading ibuprofen, trimethoprim, chloramphenicol and natural organic matter [15, 18, 29]. Based on the above controversies, it is necessary to analyze the generation potential of chlorinated byproducts in the treatment of EDTA-containing wastewater using UV/chlorine. Also, considering the production of chlorine-containing intermediates and typical chlorinated byproducts identified in most studies, evaluation of the risk caused by UV/chlorine is of great significance in defining its environmental safety in the long run.

The objective of this study was firstly to investigate the performance of EDTA degradation by UV/chlorine. Effects of chlorine dosage, solution pH, initial concentration of EDTA and coexisting substances on EDTA degradation were explored. A radical scavenging experiment was designed to determine the possible contribution of the reactive species to EDTA degradation. Furthermore, its applicability in treatment of the realistic effluents from domestic sewage treatment plant was demonstrated. Finally, the formation of representative chlorinated byproducts before and after treatment were evaluated.

## Materials and Methods

### Materials

The main reagents employed in this study were all of ACS grade from Sigma-Aldrich, including ethylenediaminetetraacetic disodium salt ( $\text{Na}_2\text{EDTA}$ , 99%), humic acid (99%), ferric nitrate (99.5%), disodium hydrogen phosphate ( $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$ ), sodium dihydrogen phosphate ( $\text{NaH}_2\text{PO}_4 \cdot 2\text{H}_2\text{O}$ ) and tert-butyl alcohol (TBA, 99.5%). Sodium hypochlorite ( $\text{NaOCl}$ ) solution was obtained from Aladdin. All the reagents used were at least of analytical grade and the solutions were prepared with Milli-Q water (Millipore,  $18.2 \text{ M}\Omega \cdot \text{cm}$ ). The luminescent bacterium T3 spp was purchased from Institute of Soil Science, Chinese Academy of Sciences.

## Experimental Procedures

The photochemical experiments were carried out in a magnetically-stirred cylindrical borosilicate glass reactor (volume of 500 mL and 60 mm in diameter) with a quartz tube in the center. A low-pressure mercury UV lamp (Shanghai Jiguang Special Lighting Appliance Factory, 28 W, 254 nm) was employed. The solution temperature was kept at  $25 \pm 0.2^\circ\text{C}$  using a thermostat system. The solution pH was buffered using 10 mM phosphate solution. Various dosage of NaClO solution was spiked into 500 mL solution containing 0.05 M EDTA. Samples were withdrawn at predetermined time intervals and rapidly quenched with  $\text{Na}_2\text{SO}_3$  solution (1.0 M).

The acute toxicity analysis experiment was carried out using the luminescent bacterium T3 spp in the logarithmic growth phase and the acute toxicity was evaluated by the relative luminescence (%). Another toxicity assay was carried out by immersing rice seeds (20 of each) in solutions before and after chlorination and UV/chlorine treatment. Length and fresh weight of rice seedling and roots were recorded after a 7-day incubation of rice seeds in the test solution at  $25^\circ\text{C}$  [30]. To confirm the experimental repeatability, the batch experiments were conducted in triplicate.

## Analytic Methods

Solution pH was monitored using a pH meter (PB-10, Sartorius). EDTA concentration was measured by a slightly modified HPLC method developed by Nowack [31] and the detailed procedures followed as: 1 mL samples were evaporated to dryness in an oven at  $90^\circ\text{C}$ . Then, 1 mL formate buffer (5 mM sodium formate and 15 mM formic acid in water, pH 3.3) was added to dissolve the residues. Afterwards, 20  $\mu\text{L}$  of 20 mM Fe(III) solution was added and the solution was thermostated at  $90^\circ\text{C}$  with water bath for 1 h. After the solution cooled to room temperature, 40  $\mu\text{L}$  of 40 mM tetrabutylammoniumbromide (TBA-Br) solution was added. Finally, the formed Fe(III)-EDTA complex was separated on C 18 column (4.6 mm $\times$ 100 mm, 5  $\mu\text{m}$  particle size, Agilent) with formate buffer (1 mM TBA-Br, 5 mM sodium formate and 15 mM formic acid, pH 3.3) and methanol (90:10, v/v%) as the eluent at a flow rate of 1.0 mL/min. The detection wavelength was set at 258 nm. The concentration of total organic carbon (TOC) was determined by TOC-V CPN analyzer (Shimadzu, Japan).

Trichloromethane (TCM), dichloroacetonitrile (DCAN) and trichloroacetonitrile (TCAN) were extracted using methyl tert-butyl ether (MTBE), and analyzed by Gas Chromatography supplemented with Electron capture detector (GC-ECD), which referred to the standard method of USEPA Methods 551.1 [32]. Dichloroacetic acid (DCAA) and trichloroacetic acid (TCAA) were extracted with MTBE, methylated with acid methanol and measured by GC/ECD, which was

carried out according to standard method of USEPA Methods 552.3 [33].

## Results and Discussion

### Efficiency of EDTA Degradation by UV/Chlorine

The efficiencies of EDTA degradation by UV irradiation, chlorination and UV/chlorine processes under the same operating conditions were shown in Fig. 1a). Less than 2 % of EDTA degradation was observed by single 254 nm irradiation within 180 s, coincident with previous reports [34]. EDTA degradation by chlorination increased with the increase of reaction time, and about 80% of EDTA was degraded within 180 s. However, UV/chlorine significantly accelerated the degradation of EDTA, with about 80% EDTA degradation within 15 s and more

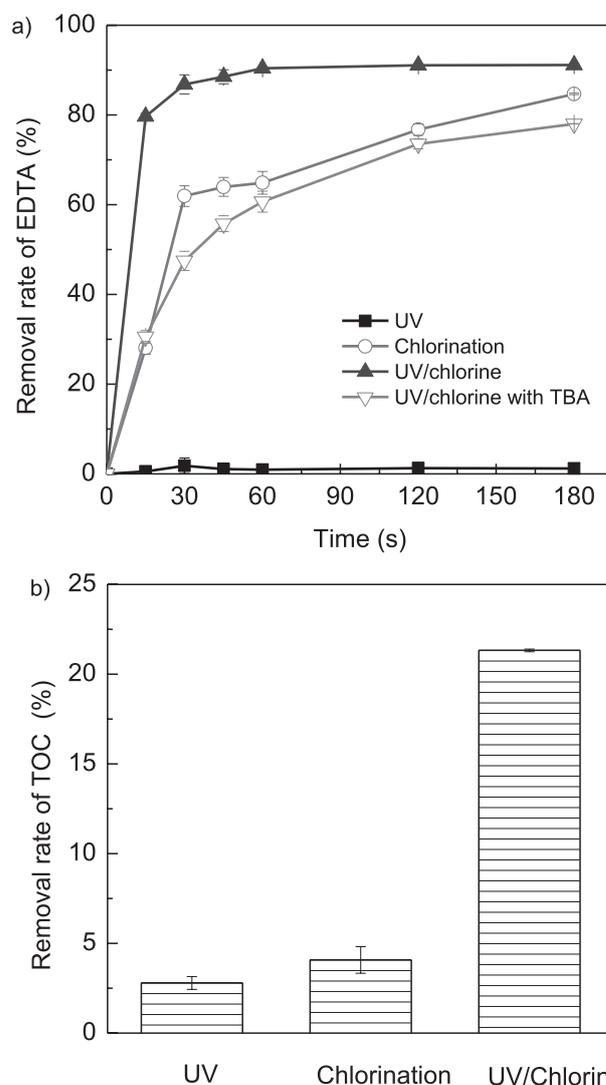


Fig. 1. Comparison of EDTA degradation a) and TOC removal b) by UV photolysis, chlorination and UV/chlorine. Conditions:  $25^\circ\text{C}$ , pH 7.0,  $[\text{NaClO}]_0 = 0.30 \text{ mM}$ ,  $[\text{EDTA}]_0 = 0.050 \text{ mM}$ , and  $[\text{TBA}]_0 = 10 \text{ mM}$ .

than 90% within 60 s. In general, EDTA degradation by UV/chlorine was higher than the sum of that by the two individual processes. Thus, UV/chlorine showed synergistic effect on EDTA degradation.

The inefficient degradation of EDTA under irradiation was due to the low photo-activity of EDTA. Chlorination is used to degrade organic pollutants through the oxidation of free chlorine [35]. Hypochlorous acid or hypochlorite can react with an amino group in EDTA [35, 36], leading that a part of EDTA was removed during chlorination. The degradation of complexed EDTA with metals in water was demonstrated to be primarily attributed to  $\text{HO}\cdot$  and  $\text{Cl}\cdot$  generated in UV/chlorine process [17]. Hence, the enhanced degradation of EDTA by UV/chlorine was possibly ascribed to the oxidation of  $\text{HO}\cdot$  and  $\text{Cl}\cdot$  formed from the photolysis of chlorine. To verify this point, the effect of the addition of tert-butanol (TBA), a good scavenger for  $\text{HO}\cdot$  and  $\text{Cl}\cdot$ , on EDTA degradation was examined. TBA greatly inhibited EDTA degradation, and EDTA degradation in presence of TBA approached to that by chlorination. This result suggested that  $\text{HO}\cdot$  and  $\text{Cl}\cdot$  were responsible for the enhancement on EDTA degradation. Besides, TOC removal during UV irradiation, chlorination and UV/chlorine processes were analyzed, and the results were shown in Fig. 1b. Less than 5% of TOC removal was observed in the processes of UV irradiation and chlorination, whereas about 20% of TOC was removed by UV/chlorine, indicating that acetate group in EDTA molecule was destroyed. Obviously, the involvement of  $\text{HO}\cdot$  and  $\text{Cl}\cdot$  helped the occurrence of a decarboxylated process of EDTA degradation.

### Effects of Chlorine Dosage, pH, EDTA Concentration and Coexisting Substances

Chlorine dosage determines the production of reactive species, affecting the degradation of EDTA by UV/chlorine. As shown in Fig. 2, increasing chlorine

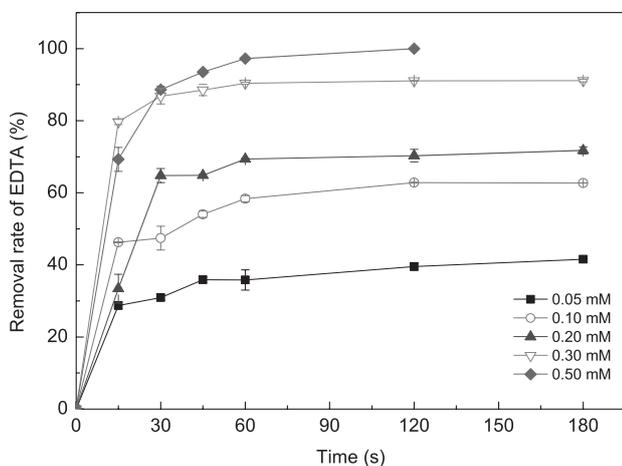


Fig. 2. Effect of chlorine dosage on EDTA removal by UV/chlorine. Conditions: 25°C, pH 7.0, and  $[\text{EDTA}]_0 = 0.050$  mM.

dosage promoted the removal efficiency of EDTA. As the chlorine dosage increased from 0.050 to 0.30 mM, the removal rate of EDTA increased from 40% to 91% within 60 s. When the chlorine dosage increased to 0.5 mM, an almost complete degradation of EDTA was achieved within 60 s. Apparently, the increase of chlorine dosage improved the production of  $\cdot\text{OH}$  and  $\cdot\text{Cl}$ , and thus accelerated the degradation efficiency of EDTA.

Solution pH is a significant factor on the efficiency of UV/chlorine process, and thus the influence of pH on EDTA degradation was investigated at initial pH ranging from 5.0 to 9.0 (Fig. 3). Increasing initial pH from 5.0 to 7.0 led to an obvious increase in the degradation efficiency of EDTA. Within 60 s, 70~92% of EDTA was degraded at initial pH ranging from 5.0 to 8.0, while the highest removal efficiency of EDTA (higher than 90%) was achieved at initial pH of 7.0. However, further elevating pH to 9.0 resulted in a significant deceleration of EDTA removal, with 25% reduction within 120 s. This was possibly caused by the higher radical scavenging rate in alkaline conditions than those in circumneutral conditions [37, 38].

As shown in Fig. 4, efficient degradation of EDTA was obtained at initial concentration of EDTA of 0.025 mM and the chlorine dosage of 0.20 mM, with the removal efficiency of 96% within 180 s. However, the increased concentration of EDTA reduced the degradation of EDTA. The concentration of the formed  $\text{HO}\cdot$  and  $\text{Cl}\cdot$  depended on the chlorine dosage. Thus, the declining degradation of EDTA was resulted from the deficiency of sodium hypochlorite at the higher initial concentration of EDTA.

Effects of coexisting substances including bicarbonate ( $\text{HCO}_3^-$ ), nitrate ( $\text{NO}_3^-$ ), and natural organic matter (NOM) on EDTA degradation were investigated, and the results were illustrated in Fig. 5. Negligible

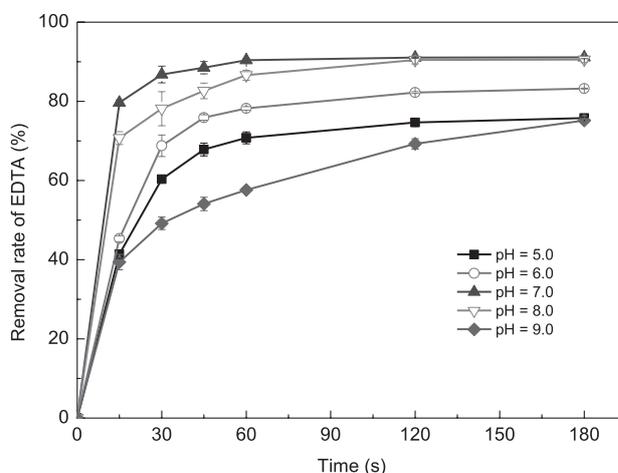


Fig. 3. Effect of pH on EDTA removal by UV/chlorine. Conditions: 25 °C,  $[\text{EDTA}]_0 = 0.050$  mM, and  $[\text{NaClO}]_0 = 0.30$  mM.

effect on EDTA degradation was observed by the addition of 15 mg/L  $\text{NO}_3^-$ . The presence of 150 mg/L  $\text{HCO}_3^-$  showed a slight influence on EDTA degradation, with about 10% reduction. Humic acid was used as a representative compound of NOM, and the addition of 6.0 mg C/L NOM significantly inhibited EDTA degradation, with reduction of about 30%. As we know, NOM acts as UV inner filter, which can also scavenge radicals [17, 39]. An obvious decrease in EDTA degradation in the presence of NOM was possibly caused by the inner filter effect of UV as well as radical scavenging effect by NOM. Thus, the coexisting NOM would be a significant factor on decontamination from sewage effluents during the photolysis of chlorine.

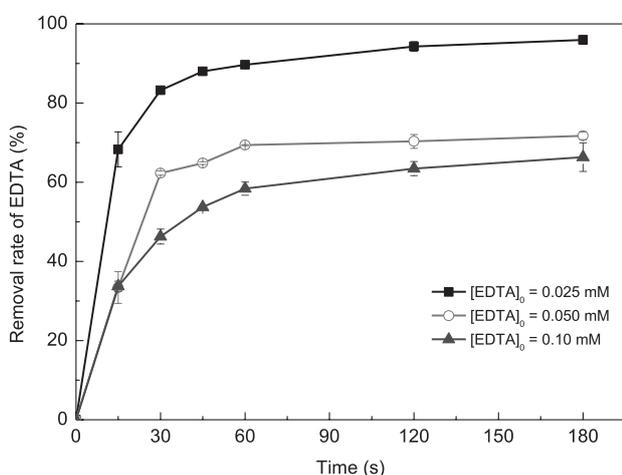


Fig. 4. Effect of initial concentration of EDTA on EDTA degradation by UV/chlorine. Conditions: 25°C, pH 7.0, and  $[\text{NaClO}]_0 = 0.20$  mM.

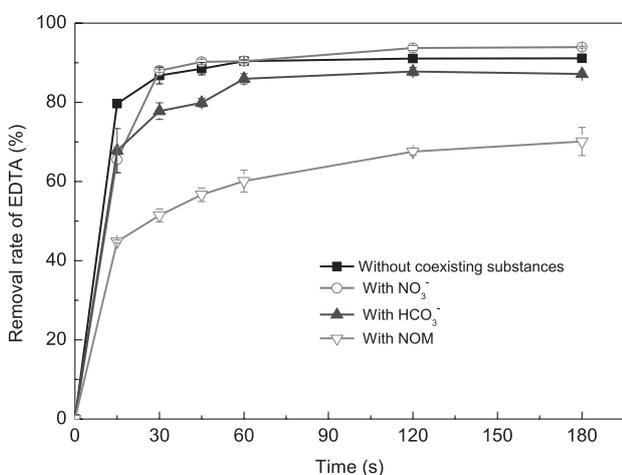


Fig. 5. Effects of coexisting substances on the degradation of EDTA by UV/chlorine. Conditions: 25°C, pH 7.0,  $[\text{NaClO}]_0 = 0.3$  mM,  $[\text{EDTA}]_0 = 0.050$  mM,  $[\text{NO}_3^-]_0 = 15$  mg/L,  $[\text{HCO}_3^-]_0 = 150$  mg/L, and  $[\text{NOM}]_0 = 6.0$  mg C/L.

## EDTA Degradation in Realistic Effluents after Biotreatment

The applicability of UV/chlorine process was further verified to treat a secondary effluent sampled from a domestic sewage treatment plant locating in Wenzhou (Zhejiang Province, China), where 0.050 mM EDTA was added. Its water quality was listed in Table 1. As shown in Fig. 6, EDTA degradation in the realistic wastewater was obviously inhibited, with about 40~50% reduction compared with that in simulated solution at the chlorine dosage of 0.30 mM. This was caused by the inner filter effects of UV light absorbers ( $\text{UV}_{254} = 0.041$   $\text{cm}^{-1}$ , Table 1) and radical scavenging by co-existing DOC, NOM and bicarbonate/bicarbonate, etc. [17, 39]. The inhibition effect by co-existing substances could be reduced by increasing the reaction time or the chlorine dosage. About 15% increment of EDTA degradation was obtained by extending the reaction time to 10 min at the chlorine dosage of 0.30 mM. As well, about 80% of EDTA degradation was achieved by increasing the chlorine dosage to 1.0 mM.

## Formation of Oxidation Byproducts

The concentrations of three typical THMs including TCM, DCAN and TCAN, and two haloacetic acids (HAAs) containing TCAA and DCAA were measured after 36 h chlorination and 5 min UV/chlorine treated EDTA-containing water. As presented in Fig. 7a), three major oxidative byproducts (OBPs) containing TCM, TCAA, and DCAA was detected, while the other OBPs were not found because their concentrations in this work were below the detection limit. It was observed that the concentration of produced THMs (about 0.5  $\mu\text{g/L}$ ) was much lower than HAAs (>4  $\mu\text{g/L}$ ). The formation of TCM in UV/chlorine higher than chlorination was consistent with the results of some

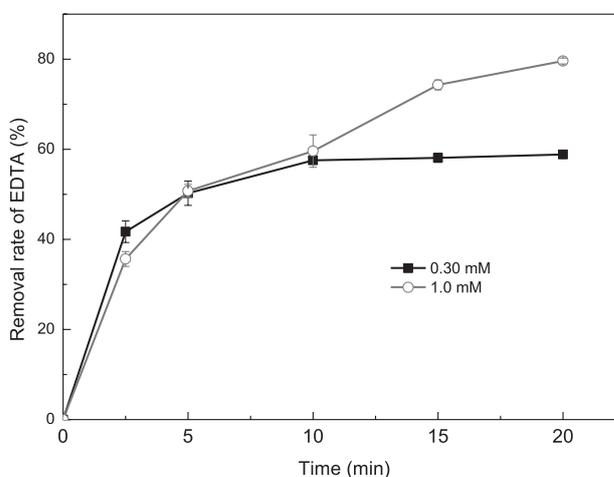


Fig. 6. EDTA degradation in realistic effluents from sewage treatment plant. Conditions: 25°C, pH 7.6,  $[\text{NaClO}]_0 = 0.30$  mM or 1.0 mM,  $[\text{EDTA}]_0 = 0.050$  mM.

Table 1. The properties of the tested secondary effluent.

pH	DOC (mg/L)	UV <sub>254</sub> (cm <sup>-1</sup> )	NO <sub>3</sub> <sup>-</sup> (mg/L)	HCO <sub>3</sub> <sup>-</sup> (mg/L)	EDTA (mM)
7.6	6.0	0.041	9.0	155	0.050

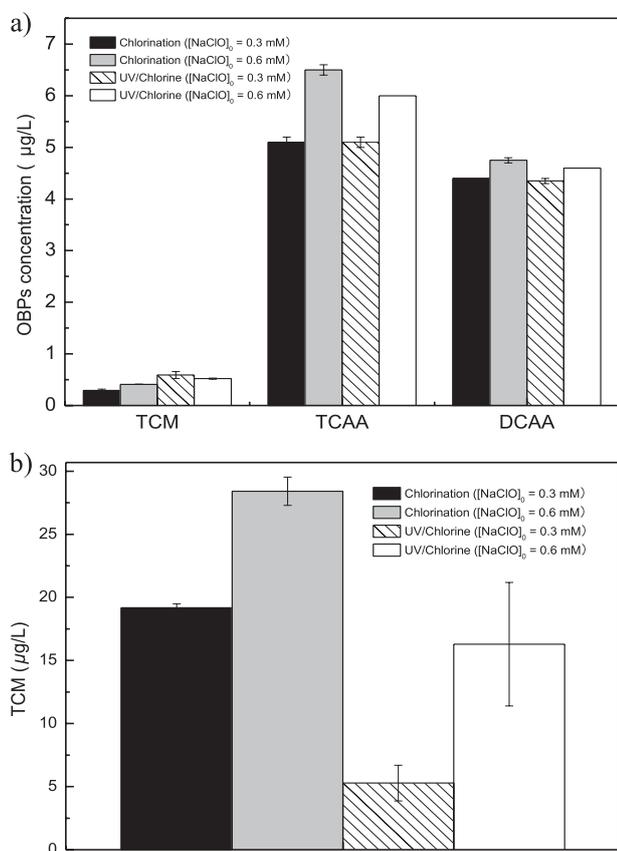


Fig. 7. Formation of OBPs after chlorination and UV/chlorine oxidized EDTA in simulated solutions a). Formation of TCM during treatment of EDTA in realistic effluent by chlorination and UV/chlorine b). Conditions: 25 °C, pH 7.0, [EDTA]<sub>0</sub> = 0.050 mM, and [NaClO]<sub>0</sub> = 0.30 mM.

researchers [15, 18]. The higher concentration of TCM produced in the UV/chlorine process could be explained that decarboxylation in the UV/chlorine process promoted the formation of OBPs with smaller molecular weight [18]. TCM formation increased with increasing concentrations of chlorine dosage under chlorination while it decreased in the UV/chlorine experiments, which could be explained by the reactions between the produced radicals (e.g., •OH and •Cl) and TCM [27, 28]. However, TCM formed in secondary effluents after chlorination and UV/chlorine showed opposite results that the formation of TCM in UV/chlorine was lower than chlorination (Fig. 7b). The complex composition in the actual wastewater containing a variety of organic substances and bicarbonate scavenged free radicals such as •Cl, thereby inhibiting the production of OBPs.

The two HAAs, DCAA and TCAA, increased as the chlorine concentration increased whatever the

chlorination process or the UV/chlorine process, and TCAA increased faster than DCAA in these two technologies. Besides, the concentrations of both HAAs detected in the UV/chlorine process were lower than the chlorination.

### Toxicity Evaluation

Considering that the discharge of treated wastewater into the surrounding environment might have a certain impact on the growth of organisms, two toxic biological tests (luminescent bacterium and rice planting tests) were conducted to evaluate the toxicity changes before and after treatment using UV/chlorine. As shown in Table 2, three controls (blank, chlorination and UV/chlorine) all had low relative luminescence and the results were consistent with other studies [26]. In chlorination and UV/chlorine treatments, disappointingly, the relative luminescence decreased to 15.21% and 11.65% respectively, which could be possibly caused by the formed OBPs. The lower relative luminescence in the UV/chlorine process compared to chlorination was probably due to the higher concentration of TCM formed during the former process. And, consistent with the formation of TCM in the realistic effluent, the toxicity after chlorination was higher than UV/chlorine.

As shown in Table 3 and Fig. 8, both simulated solution and actual effluent treated by UV/chlorine exhibited significant inhibition on the growth of rice seeds, especially on roots of rice seeds. The slightly higher toxicity to rice seeds after UV/chlorine treatment than chlorination also could be explained by the production of more OBPs in the UV/chlorine process. Similar results were observed in the treatment of NOM and phenacetin by UV/chlorine [40, 41]. The cytotoxicity of the treated NOM by UV/chlorine was about 1.7 times that of chlorination [40]. In the degradation of phenacetin by UV/chlorine and chlorination, the acute toxicity to luminescent bacterium Q67 increased [41]. Thus, considering that the toxicity

Table 2. Relative luminescence of luminescent bacterium before and after chlorination and UV/chlorine treatment (25°C, pH 7.0, [EDTA]<sub>0</sub> = 0.050 mM, and [NaClO]<sub>0</sub> = 0.30 mM).

Treatment	Simulated solution	Realistic effluent
Blank control	44.94%	16.18%
Chlorination	15.21%	17.00%
UV/chlorine	11.65%	19.35%

Table 3. Shoot and root lengths (cm) of seed germination in the simulated solutions (a) and actual effluents (b) treated by chlorination and UV/chlorine.

	A	B	C	D
Shoots (a)	33.09±1.62	18.76±1.23	16.68±1.21	21.92±0.51
Roots (a)	41.97±0.50	4.25±0.94	3.16±0.01	8.61±0.50
Shoots (b)	48.22±2.37	32.72±0.81	16.53±0.13	28.73±0.47
Roots (b)	47.23±4.69	31.62±1.62	3.15±0.07	6.82±0.96

A: blank control; B: 0.05 mM EDTA solution; C: treated solution by UV/chlorine; D: treated solution by chlorination

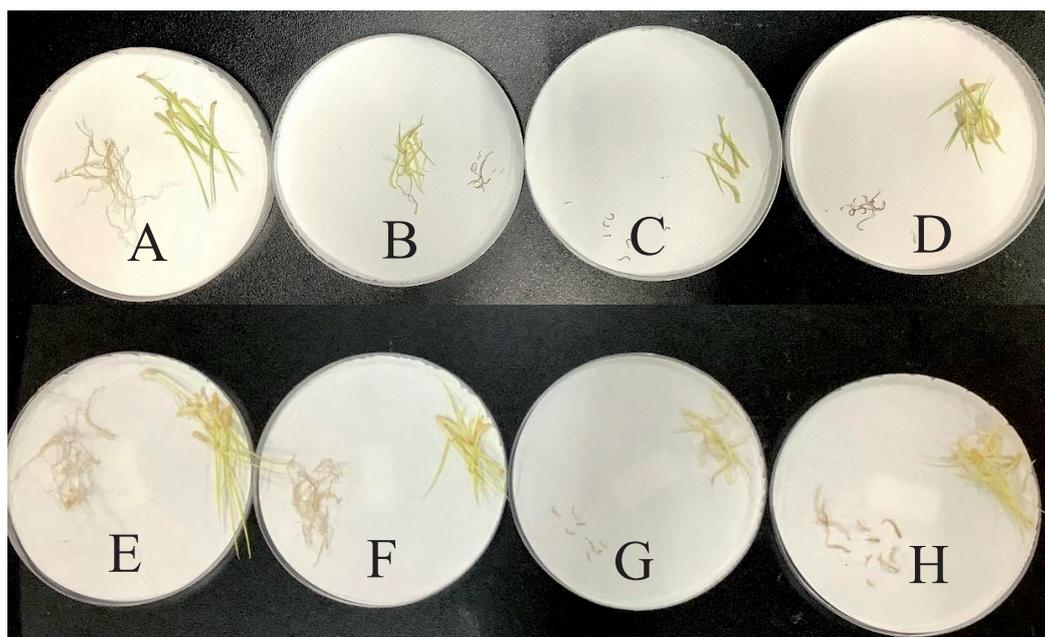


Fig. 8. Photos of rice roots and shoots after 7-days incubation in the simulated solution and actual effluent before and after chlorination and UV/chlorine treatment.

A: ultrapure water; B: 0.050 mM EDTA solution; C: treated solution by UV/chlorine; D: treated solution by chlorination; E: realistic effluent; F: realistic effluent containing 0.050 mM EDTA; G: treated effluent by UV/chlorine; H: treated effluent by chlorination.

generated along with UV/chlorine oxidation, the optimization of this process to reduce the production of OBPs is still needed.

### Conclusions

UV/chlorine process showed synergistic effects on EDTA degradation comparing to UV irradiation or chlorination alone. An almost complete EDTA was degraded within 60 s at the initial EDTA of 0.050 mM and the chlorine dosage of 0.50 mM. EDTA degradation was promoted by the increasing chlorine dosage and the decreasing initial concentration of EDTA. The increasing initial pH of the solution from 5.0 to 7.0 accelerated EDTA degradation, but the further rising pH to 9.0 showed the inhibition effect. EDTA degradation was less affected by co-existing  $\text{NO}_3^-$  and  $\text{HCO}_3^-$ , but significantly inhibited by NOM.  $\text{HO}\cdot$  and  $\text{Cl}\cdot$  formed in UV/chlorine largely accounted for the

enhanced degradation of EDTA, as supported by the radical scavenging experiment. In addition, UV/chlorine could remove about 50%~80% EDTA from realistic secondary effluent at the chlorine dosage ranging from 0.3 to 1.0 mM. Three major OBPs including TCM, TCAA and DCAA were detected in both chlorination and UV/chlorine, and the slightly higher concentration of TCM produced in UV/chlorine than chlorination was observed. Both simulated solution and actual effluent treated by UV/chlorine showed significant inhibition on luminescent bacterium and the growth of rice seeds.

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### Conflict of Interest

The authors declare no competing financial interest.

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