

Counter-current/cocurrent dissolved air flotation system for algae-laden surface water treatment

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ABSTRACT

A novel countercurrent-cocurrent dissolved air flotation (CCDAF) was developed based on the mechanism of bubble-floc collision adhesion. The removal efficiency and mechanism of algae-laden and high-organic-substance surface water used the CCDAF was researched in this work. The results show that the average removal rate of algae by the CCDAF process was 96.50%. In addition, microcystin-LR, 2-methyl isobutyl alcohol (2-MIB), and geosmin (GSM) can be significantly removed in the flotation process and effectively avoid the release of intracellular substances of algae. The CCDAF has good removal efficiency of organic compounds simultaneously, and the average removal rates of COD_{Mn} , UV_{254} , and DOC were 37.6%, 46.3%, and 32.11%, respectively. The dominant mechanism of organic and algae removal in CCDAF's counter-current and cocurrent zone was discussed based on microscopic observation of bubble in different process areas. The experimental results provide an effective solution for algae-laden surface water treatment.

Keywords: CCDAF; Copolymerization DAF; Algae; Organics; Reservoir water

1. Introduction

The traditional coagulation and sedimentation water purification technology has a poor effect on high algae water treatment, such as cyanobacteria, and the removal rate of chlorella reaches only 58% [1–3]; and the algae cells are ruptured during the treatment to release soluble metabolites such as algal toxins and odor compounds [4]. In view of the low density and easy floating of algae [4], dissolved air flotation (DAF) has significant technical advantages in treating surface water with high algae removal efficiency and avoids the increase of releasing intracellular (or cell-bound) toxins during treatment and reduces the coagulation dose [5]. Algae, particles of raw water formed hydrophobic floc particles after coagulation and collided with and adhere to reflux dissolved water microbubble, then it rose to form scum and

was removed [6]. Hydrophobic flocculation particles with neutral or low surface potential are the key to flotation. Cyanobacteria in the surface of the electrical neutralization point, the removal efficiency of the highest, and it is also a sign of the coagulant effectively attached to the surface of the algae [7]. As the processing unit after coagulation, traditional DAF reduces the synergistic efficiency of coagulation and DAF. High collision frequency and the smaller bubble size, can significantly improve the collision probability of floc particles and bubbles to improve the adhesion rate to reduce the separation probability and the rate of rise, improve the algae removal efficiency, but also increase energy consumption. The produced microbubbles in the DAF could present variable and stable dimension, from 10 to 100 μm [8], and increase dissolved gas reflux ratio can effectively improve the floc particles and bubble collision frequency. Moreti [9] found that the best removal efficiency of first-stage DAF process is reflux ratio 30%, and more than 35% will destroy the floc structure.

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Based on preliminary experiments by the authors and considering the collision adhesion mechanism of microbubbles and floc-particles, and combined with the advantages of cocurrent and counter-current DAF processes, Counter-current/cocurrent DAF (CCDAF) was invented. In terms of contact zone structure, water flow characteristics, and the adhesion of bubble-floc, CCDAF appeared to be more optimized than DAF. In CCDAF, there is twice as much as dissolved air water and the microflocculation combines with DAF. Microbubbles are directly involved in the flocculation reaction process, which is the bubble-floc copolymer effect. The flocculation efficiency of the foam will be significantly improved, and the formation of the floc is stable and not easily desorbed [10]. The experimental results showed that the turbidity removal efficiency of the CCDAF process was significantly improved. In view of the characteristics of more complex water quality, it is necessary to study the characteristics of organic matter removal by the CCDAF process. In this paper, the water source of high algae and organic matter as the research object are used to (1) study the algae and organic matter removal efficiency in the CCDAF process; (2) study the morphology of algae and the changes of algal metabolites before and after the process; (3) study the removal efficiency and mechanism of organic matter with different forms and molecular weights; and (4) comprehensively evaluate the operating characteristics of CCDAF technology, thus providing technical support for the application of CCDAF technology.

2. Materials and methods

2.1. Raw water quality

In this study, raw water taking from Jining region, the water receiving area of Shandong Province of South-to-North Water Diversion, as the research object. The raw water in this area mainly shows high algae and high organic matter, the total algal population is more than 10^6 – 10^7 cells/L, with Cyanophyta and Chlorophyta dominated. *Chlorella* and *Chlamydomonas* are dominant species with a small amount of *Microcystis aeruginosa*. Raw water temperature during the experiment ranged from 20°C to 27°C, pH ranged from 8.10

to 8.40, turbidity 13.3–25.2 NTU, DOC 6.95–7.78 mg/L, particulate matter 85,058–96,736 CNTs/mL, COD_{Mn} 5.65–7.26 mg/L, UV_{254} 0.145 ~ 0.152 cm^{-1} , and $\text{NH}_4^+\text{-N}$ 0.352 ~ 0.458 mg/L.

2.2. CCDAF system

The CCDAF system includes a cocurrent contact chamber, a counter-current contact chamber, and an air separation chamber (Fig. 1). Unlike traditional DAF technology, the contact chamber is divided into two stages: the collision contact chamber and the adhesive contact chamber. On the other hand, the dissolved air water is added in two instances. In the contact room, microbubbles and water reverse flow, completing a full collision with the suspended substance. The microbubbles are directly involved in the floc's condensation reaction, and they form larger flocs. Then, the effluent enters the adhesion contact chamber. In the adhesive contact chamber, microbubbles contact water flow in the same direction. The contact chamber's portion of the microbubbles flows into the adhesive contact chamber, increasing the concentration of microbubbles. This can improve the adhesion efficiency of the microbubbles and suspended substances in the adhesion contact chamber and form stable bubble-floc copolymerization floating to the flotation separation chamber.

2.3. Experimental design and process operation

The experimental setup consisted of front mixing, two-stage flocculation cell and CCDAF reactor (length-width-height 0.83 m × 0.1 m × 1.45 m). The test influent flow was 0.5 m^3/h . Before the dissolved air water refluxed to the CCDAF, removal for better flotation, raw water is mixed with coagulant and secondary flocculation tank, namely fast and slow flocculation, and test conditions based on previous research [10]. The coagulant is polyaluminum chloride and dosage is 5.0 mg/L. Coagulation rapid flocculation stage (G_1 value 52 s^{-1}) 4 min, slow flocculation (G_2 value 25 s^{-1}) 4 min. the HRT of CCDAF contact cell (including collision contact cell and adhesion cell) is 2 min. What's more, the velocity of collision contact cell and adhesion cell is 15–25 and 5–20 mm/s, respectively. The total reflux ratio of the

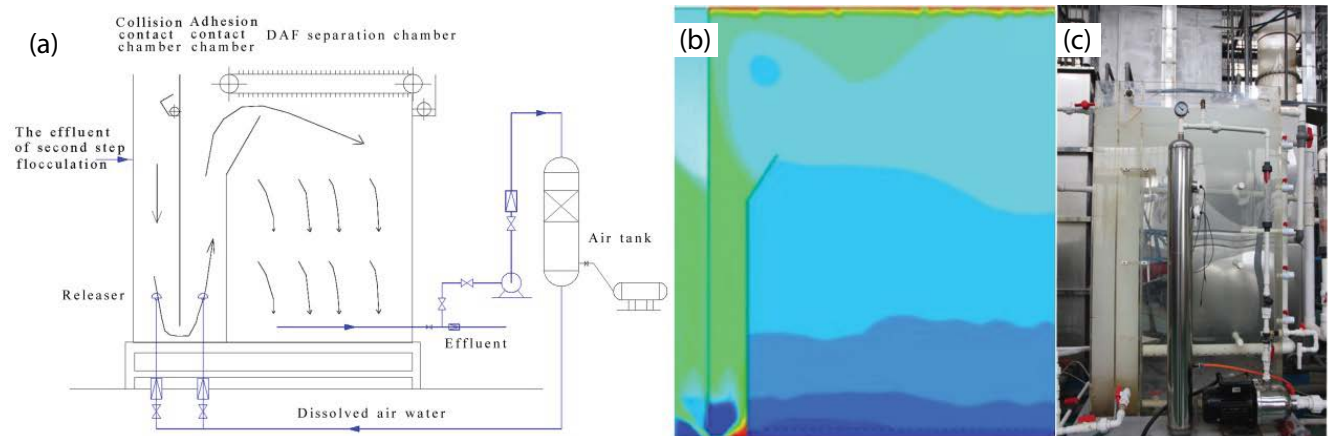


Fig. 1. Sketch map of CCDAF process (a: The sketch of CCDAF; b: Nephogram of microbubbles distribution in flotation tank; c: Photograph of CCDAF process).

dissolved air water is 12%, and the reflux ratio of collision and adhesion contact cell is 1/2.

2.4. Analytical methods

The pH, NTU, and UV_{254} were measured using pH tester (PHS-3D), light scattering turbidity meter (TSZ-400A), and ultraviolet visible spectrophotometer (TU-1810), respectively. Measurement of algae cell count was used microscopic counting method, and the spectrophotometer method was used to measure chlorophyll A. Algal toxins (extracellular microcystin-LR (MC-LR)) were measured using HPE-HPLC. Gas chromatography was used to measure 2-methyl-isoborneol (2-MIB), geosmin (GSM), and THM formation potential (THMFP). The molecular weight distribution was measured using cup ultrafiltration. DOC, particle counting and fluorescence characteristics were measured using total organic carbon analyzer, particle counter and fluorescent photometer (TOC-VCPH-CPN, GREAN IBR versa count, and F-7000 fluorescent photometer). COD detection used potassium permanganate method according to the water and waste water detection and analysis methods [11].

3. Result and discussion

3.1. Performances of algae removal

In this research, the original algae count was 2350–6750 million cells/L and the algae count of CCDAF's effluent was 83–210 million cells/L. The average removal rate was 96.50%, as shown in Fig. 2. Comparing with CCDAF, the average removal rate of algae in C/F/DAF was only 92% and this in conventional DAF was about 93.7% according to Teixeira [2,5]. The average size of *M. aeruginosa* cells was 1–10 μm [4,7]. In the CCDAF, the size of microbubbles was between 30 and 50 μm [12]. Because of the negative charge on the surface of the algae cell membrane, there was a water layer strongly attached to the cell membrane [7]. After coagulation, the charged and hydrated layer was weakened, and the positively charged algal flocs were formed. There was negative charge on the surface of the microbubble, and the surface potential was $-30 \sim -50 \text{ mV}$ [12,13]. Therefore, microbubbles could adsorb positively charged substances in water, and the

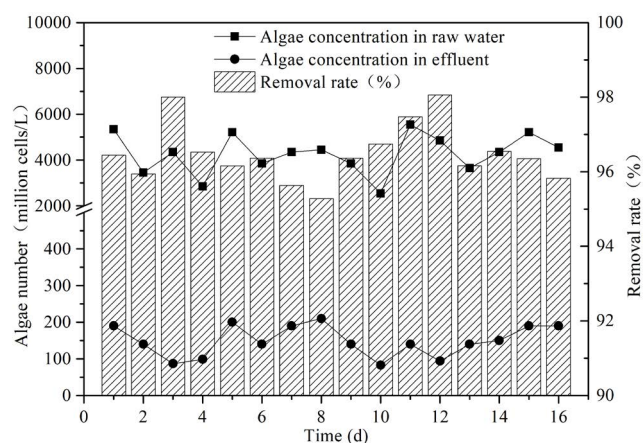


Fig. 2. The algae removal of CCDAF process.

algae could be attached to the bubble or flock [8,14], which improved the removal efficiency of algae after the collision and copolymer of bubble-floc.

The morphology of algae in the water before and after the CCDAF process was studied by using a transmission electron microscope and scanning electron microscope. As shown in Fig. 3(b), the morphology of algae did not change after the CCDAF process, and they had both globular, smooth cell walls. In other words, the algae cell was not destroyed. In addition, the inside of algal cells showed no cavitation after CCDAF [15]. This demonstrated that there was no loss of outflow in algal cells. Figs. 3(c) and (d) clearly show the external morphology of algae. The algae cell is smooth, full, and has no deformation. Therefore, the morphology of algal cells remained intact. In summary, the internal material and cell wall structure of algae were not destroyed. Thus, there was no significant effect on the morphology of algae after CCDAF treatment.

The water purification mechanism of the traditional DAF is mainly the collision and adhesion of raw water on the injected microbubbles [13]. Whereas, the crucial differences of CCDAF lie in the dissolved gas and the graded water reflux, which can shorten the primary flocculation time and promote microbubbles to directly participate in the coagulation process [16]. Then, with the complete combination of microbubbles and large floc grains, copolymerized floc bubbles are generated in the middle of algae floc grains, as shown in Figs. 4(b) and 5, and form a “copolymerization air flotation” (Fig. 5(d)). Hence, the cohesion of bubbles in the CCDAF process can develop thoroughly. On the other hand, the bubbles can firmly adhere to algae floc grains, so that the flocculated particles with air are stable enough to remain floating in the flotation process [17]. During the copolymerization process of the CCDAF, the dissolved air water is injected after initial coagulation, and the main coagulation mechanisms are destabilized and electrically neutralized. During the

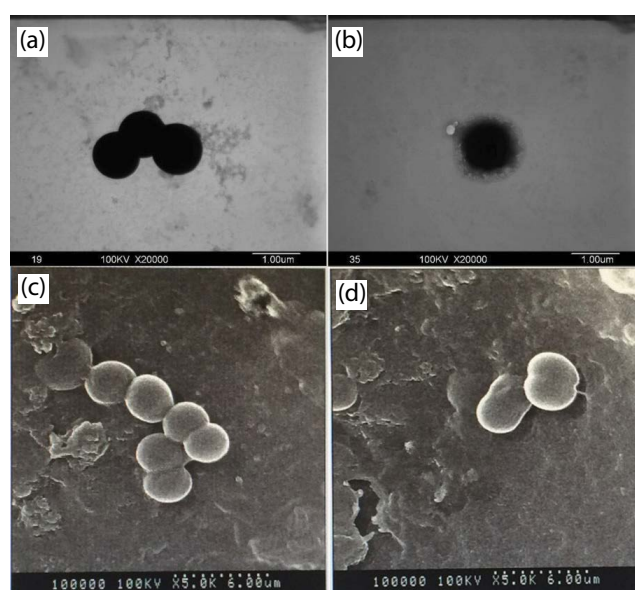


Fig. 3. TEM (a: before, b: after) and SEM (c: before, d: after) images of algae before and after CCDAF.

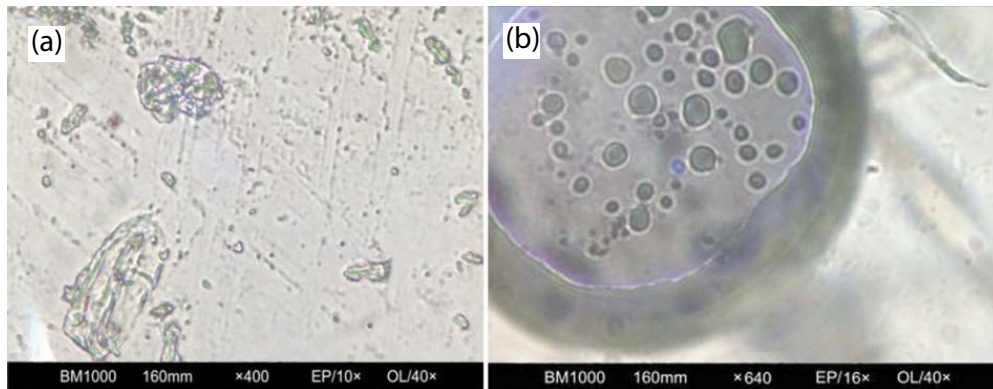


Fig. 4. Microscope observation of bubble-flocs copolymerization (a. Micromorphology of floc; b. Copolymer floc).

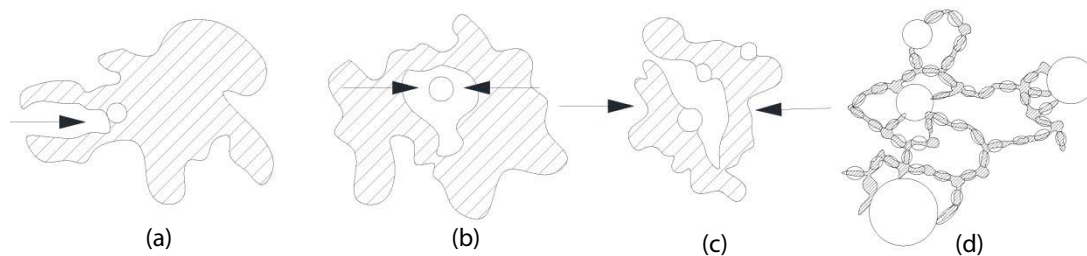


Fig. 5. Three kinds of formation of microbubbles (a, b, and c) caught in the algae flocs and copolymerization of microbubbles with flocs (d).

first recycle of the dissolved air water, the primary mechanism of microbubbles adhering to particles is the collision and adhesion between microbubbles and flocs. After the secondary injection of dissolved air water, the main clarification mechanisms include the collision and adhesion between microbubbles and flocs, the adsorption bridging and the sweep flocculation of bubble-flocs and floc-microbubble particles (as shown in Fig. 5).

3.2. Algae toxin removal

Algae toxins released by cyanobacteria have attracted wide attention [16], among which the most widely distributed and the most toxic is microcystin (Microcystin, referred to as MCs) [18]. Microcystin-LR (MC-LR) is documented as the most common microcystin toxic [19]. To investigate the effect of air floatation on algae and the release of intracellular substances, the research studied the release and removal of MC-LR from the algae by CCDAF treatment. As shown in Fig. 6, the extracellular MC-LR increased from 0.3 $\mu\text{g/L}$ to 0.4 $\mu\text{g/L}$. Mixing and two-stage mechanical flocculation existed among the three-stage mixing in the CCDAF process. Because of this, a small amount of intracellular MC-LR was released, increasing extracellular MC-LR. SEM showed that the algal cells were not damaged and were still intact. However, high ionic salt was dosed in water in the coagulation stage, leading to algal aggregates disintegration, cell dehydration and contraction in hypertonic conditions. On the other hand, mechanical agitation also caused algae cells to disperse. Therefore, the extracellular MC-LR concentration increased slightly, which was caused by the release

of intracellular MC-LR in the dehydrated cells under the action of coagulant. The coagulant does not cause algal cell disruption.

The flotation is a physical process: microbubbles and algae floc adhesion removal are physical adhesion. Therefore, it will not cause the deformation and destruction of algal cells. In the CCDAF process, the bubble-flocs were removed by floating and partially removed MC-LR in the meantime. The effluent content of MC-LR decreased from 0.4 to 0.2 $\mu\text{g/L}$, which accords with the Hygienic standard for drinking water of China (GB5749-2006). On the other hand, the concentration of extracellular MC-RR and MC-YR in the

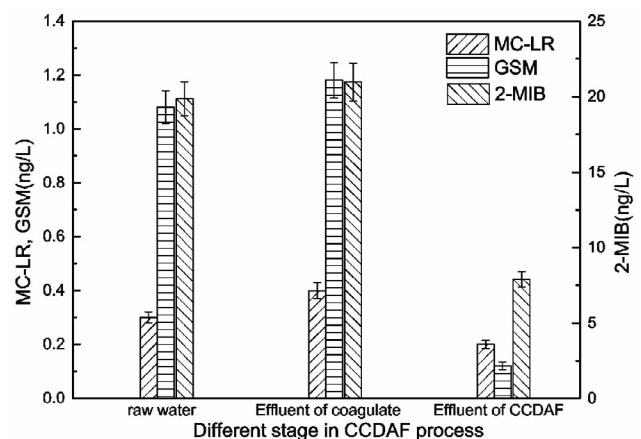


Fig. 6. Removal rule of extracellular MC-LR, 2-MIB, and GSM.

raw water and the CCDAF's effluent were detected, which were less than the detection limit of 0.0004 mg/L.

3.3. Odor removal

It is difficult to remove the odorous substances produced by algae via the conventional water purification process [19]. In a surface water source polluted by algae, such as reservoir water, the breeding of actinomycetes and algae in water is the origin of water odor. The GSM and 2-MIB released by actinomycetes and algae are the main substances that produce the odor.

As shown in Fig. 6, the odorant is also a metabolite of algae. Therefore, GSM and 2-MIB release and the removal rules were basically consistent with MC-LR. At the coagulation stage, the odor substance in the algal cells, such as 2-MIB and GSM, was released. 2-MIB increased from 19.866 to 20.972 ng/L and GSM increased from 1.08 to 1.18 ng/L. The concentration of 2-MIB and GSM was decreased after the CCDAF process, and the effluent water concentration was 7.9 and 0.12 ng/L, respectively. MIB removal was better than conventional DAF according to Elías-Maxil et al. [12]. In conclusion, coagulation caused the release of intracellular olfactory substances, which were partially removed by CCDAF. This phenomenon is the same as the removal mechanism of MC-LR.

As seen from the earlier analysis, algal metabolites (MC-LR, GSM, and 2-MIB) significantly decreased after CCDAF treatment. Thus, CCDAF is effective in avoiding the release of intracellular algal material, and it has a significant algae removal effect. Also, CCDAF, compared with other processes to remove algae, showed a significant advantage. This is also a significant advantage of CCDAF algae removal compared to other algae removal processes.

3.4. Performances of organic compounds removal

In this study, the COD_{Mn} , UV₂₅₄, and DOC of raw water were 5.65–7.26 mg/L, 0.145–0.152 cm^{-1} , and 6.95–7.78 mg/L, respectively. The COD_{Mn} , UV₂₅₄, and DOC of CCDAF's effluent water were 3.37–4.12 mg/L, 0.0751–0.0806 cm^{-1} , and 4.21–4.82 mg/L, respectively. The removal rates of COD_{Mn} , UV₂₅₄, and DOC were 37.6%, 46.3%, and 32.4%, respectively (Fig. 7). In addition, the removal rates of COD_{Mn} , UV₂₅₄, and DOC were 36.1%, 44.0%, and 32.2%, respectively (Table 1), during the conventional DAF running alone with closed the releaser of CCDAF collision contact chamber. Compared with the conventional DAF, the removal rates of CCDAF increased by 4.1%, 5.0%, and 0.58%, respectively, and the removal was excellent.

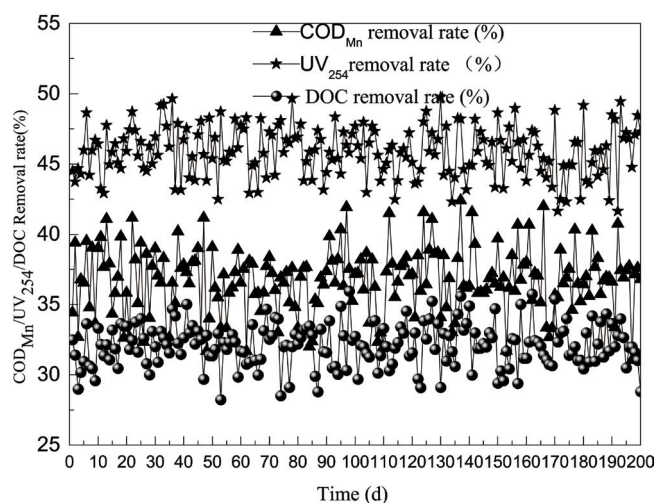


Fig. 7. The organic matter removal of CCDAF process.

Further analysis showed that the macromolecule organic matter, attached and wrapped to the colloidal particles, could be destabilizing coagulation and polymerization in the coagulation stage. Then, the matter could be attached to the surface of the alum. Finally, it was removed by the adhesion of microbubbles. COD_{Mn} reflects organics, including suspended-state, colloidal and dissolved organic matter. The DAF process removes suspended and colloidal substances. Therefore, the suspended-state and glue posture organisms will be removed easier than small-molecule organic matter [20,21].

The test results can also be explained by the thermodynamics theory of foam flocculation adhesion. For hydrophobic substances, microbubbles are easily adhered to the surface of the substance and floated. Microbubbles are hydrophobic and have a tendency to reduce their interface energy, making it easier to adsorb hydrophobic substances [13]. For hydrophilic substances, the attraction to water is greater than the attraction to microbubbles, resulting in microbubbles close to these substances squeezing the surface of the hydration film. This results in poor adhesion.

3.5. Removal efficiency of THMFP

Chlorine is commonly used as a water supply plant disinfectant. Natural organic matter (NOM) is the main precursor of chlorine disinfection byproducts contained in effluent water. The addition of chlorine reacts with NOM to produce trihalomethanes (THMs) and halogen acetic acids (HAAs). THM is one of the most important disinfection byproducts

Table 1
The efficiency removal of organic compounds in CCDAF and DAF

Organic testing index	CCDAF		DAF	
	Concentration of effluent	Removal rate (%)	Concentration of effluent	Removal rate (%)
COD_{Mn} (mg/L)	3.37–4.12	37.6	3.61–2.63	36.1
UV ₂₅₄ (cm^{-1})	0.0751–0.0806	46.3	0.0812–0.0451	44.0
DOC (mg/L)	4.21–4.82	32.4	4.71–5.27	32.2

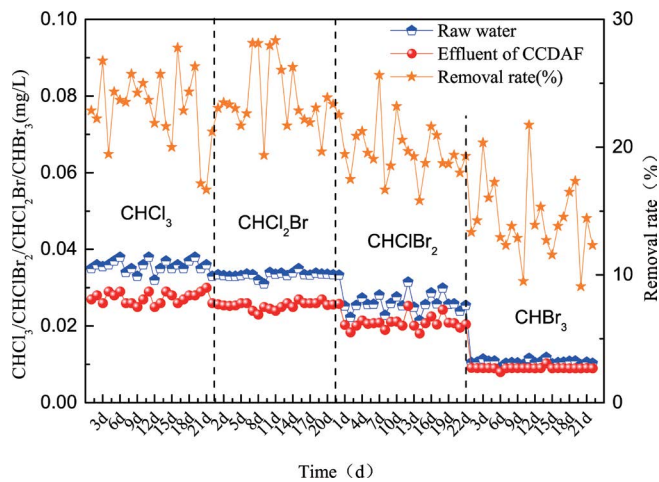


Fig. 8. THMFP removal rate of CCDAF process.

and has been identified as a “three-cause” substance. Algal organic matter also produces THMs and HAAs during chlorine disinfection [22]. It is a greater risk to drinking water safety [23].

THMs are the generic name of CHCl_3 , CHCl_2Br , CHClBr_2 , and CHBr_3 , and their precursors are mainly NOM. The traditional process for NOM removal capacity was limited. Therefore, when dealing with micropolluted water with NOM, the water still contains more THMs. This creates a large THMFP, and a large number of THMs will be produced after chlorine disinfection [24]. Therefore, improved ability to remove THMFP in the water treatment process can reduce the amount of THMs produced by chlorine disinfection. The effect of CCDAF process on THMFP is shown in Fig. 8. The average removal rates of CHCl_3 , CHCl_2Br , CHClBr_2 , and CHBr_3 were 23.75%, 23.04%, 19.68%, and 14.14%, respectively.

4. Conclusions

- The average removal rate of algae by the CCDAF process was 96.50%. The algal cell substance did not flow out, and the cell wall remained intact; the morphology of algae did not change significantly after the CCDAF treatment. The MC-LR, 2-MIB, and GSM can be significantly removed by the CCDAF process and the effluent water concentrations of these were 0.2 $\mu\text{g/L}$, 7.9, and 0.12 ng/L , respectively. Compared with other algae removal processes, the CCDAF process significantly removed algae at the same time, effectively avoiding the release of algae intracellular material.
- CCDAF was better at removing macromolecules, hydrophobic organic matter and THMFP. The average removal rates of COD_{Mn} , UV_{254} and DOC were 37.6%, 46.3%, and 32.11%, respectively, and the effect of CCDAF on removal of organic compounds was $\text{UV}_{254} > \text{COD}_{\text{Mn}} > \text{DOC}$. Organic adhesion mechanism of the CCDAF process is the charge neutralization desorption coagulation and surface adsorption.
- In the collision zone of the CCDAF process, the main adhesion mechanism of the flotation is the adhesion, collision and copolymerization between microbubbles

and floc particles; during the secondary reflux, the main adhesion mechanisms include the collision adhesion between the microbubbles and floc particles, the adsorption bridging and the sweep flocculation of floc-microbubble particles.

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