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An integrated view of the intimate coupling UV irradiation and algal treatment on antibiotic: Compatibility, efficiency and microbic impact assessment

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ABSTRACT

The aim of this study is to investigate the UV and algal contribution in an intimate coupling UV irradiation and algal treatment to remove two commonly used antibiotics, cefradine and amoxicillin (AMX), including the compatibility of UV wavelength and the algal species, the removal efficiency evaluation and the microbic impact assessment. The green algae *Chlorella pyrenoidosa*, *Selenastrum capricornutum* and *Scenedesmus obliquus* achieved a satisfactory growth capacity and played a dominant role during the treatment. The optimal application involved the UV-irradiation at 365 nm combined the green algae *S. obliquus*. After 24 h, the excellent removal efficiency (99.84%) was obtained after the treatment. Our results indicated that the green algae performed a satisfactory growth capacity under the UV irradiation and played a dominant role for the biodegradation of the target antibiotics and the UV irradiation has been viewed as trigger for the algal treatment. Compared with the traditional biotechnology, the advantages of the intimate coupling treatment included high removal efficiency, energy conservation during the treatment, and low environmental impact after the treatment.

1. Introduction

Because of the widely application and low removal efficiency, antibiotics are frequently detected in surface water, ground water, drinking water, wastewater, and soil [1]. Although antibiotics were detected at a relevant low level (ng/l or μg/l), the presence of the compounds may induce resistance in bacterial populations, which cause non-effective therapy of the corresponding diseases [1] and finally harm the human health [2].

Generally, the common antibiotics treatment technologies include bio-degradation, chemical degradation and physical degradation. Biological treatment such as activated sludge is a widely used technology in current Sewage Treatment Plants (STPs). However, due to the impact of antibiotics on bacteria, the removal efficiency of the activated sludge treatment on antibiotics is still unsatisfactory. Additionally, the sewage receives bacteria which exposed to antibiotics in a relevant long-term, therefore, the selective press also leads to several bacterial strains easily acquire resistance against the antibiotics impact and release their antibiotic resistance genes (ARGs). For example, the occurrence of antibiotic resistance in *Acinetobacter* isolates to AMX, chloramphenicol and rifampin significantly increased by 29.2%, 43.8% and 21.4%, respectively [3]. On the other hand, due to the excellent removal capacity, Advanced oxidation processes (AOPs), such as photolysis, hydrogen peroxide and Ozone, have been increasingly applied to obtain a high treatment rate and rapid treatment time [4]. However, the high requirement of catalyst dosage, high electrical power consumption and the secondary pollution should not be ignored [5]. For example, although 73.07% of cefradine was removed after a 0.237 h of UV-irradiation, the toxic of the by-products after the UV-treatment were 1.04 times higher than that of antibiotic itself [6]. The removal efficiency of Sulfamethoxazole (SMX) was enhanced after a 60 min ozonation, while the toxicity slightly increased after the stage of ozonation [7] and the toxicity of Sulfamethazine (SMT) was also improved after the photo-Fenton process [8]. Thus, to overcome the above problems and to obtain an efficient and economical treatment, the combination of chemical and biological processes as a potential alternative has been developed. Compared with the individual treatment, the chemical treatment step involved in the combination has been applied

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to improve the biodegradability of the target compounds by resulting in the intermediates, which have been easily degraded in the subsequent biological treatment step [9].

The green algae was not the target organism of the antibiotic. Compared with activated sludge, the algae have great abundance of biomass in aquatic environments [1] and also performed high tolerance to antibiotics even at a relevant high concentration (mg/l) [10]. Apart from this, microalgae have been considered as a potential source for the production of biodiesel, which could to reduce the industrial surpluses after the treatment [11]. There was good application of green algae to treat the antibiotic amoxicillin, which integrated with Fenton process [12]. Similarly, the combination of UV-irradiation and algal process as a potential choice also could perform the distinct dominance. For one thing, UV-irradiation could be a large-scale effluent treatment option to decompose the antibiotics but the process cannot transform all antibiotics into harmless compound(s) or CO_2 directly [13]. For another, microalgae is a good choice in reducing the toxicity of the parent compounds and the corresponding by-product after the preceding chemical process [14–16]. Thus, there was good application of the algal combination treatment to treat antibiotic. The published results indicated that the removal rate of norfloxacin was improved 4 times after an algae and UV-irradiation combined treatment [17].

Compared with the sequential combined treatments, the intimate coupling of chemical and biodegradation has been viewed as a novel alternative [18]. First of all, we should oxidize the organics only to the degree that they are easily biodegraded and not overly oxidized which would waste energy and/or oxidizing chemicals. Additionally, it may difficult to achieve the special conditions which was directly fit to the subsequent biological treatment. Thus, the aim of the present study was to support an intimate coupling of UV irradiation and algal treatment. Three species of microalgae (*Chlorella pyrenoidosa*, *Selenastrum capricornutum* and *Scenedesmus obliquus*) and four kinds wavelength of the medium pressure mercury lamps (185, 254, 280 and 365 nm) were first introduced to provide the optimum algal species and wavelength to obtain the best treatment efficiency. Cefradine and AMX, two widely used antibiotics, were selected as the target compounds. Conventional biological processes are usually suspected to contribute to antibiotic-resistant bacteria selection, whether the intimate coupling of UV irradiation and algal treatment exerted a similar selective pressure should be determined. Thus, the bacteria *E.coli* and *S.aureus* were used to assess the bacteriostatic level of the target antibiotic after the intimate coupling treatment. Additionally, previous studies also indicated the impact of antibiotics on the non-target organisms [19,20]. Thus, to exclude the possibility that the toxicity of the target antibiotics increased after UV irradiation or other chemical degradation [21,22], the aquatic impact of the target antibiotics and the corresponding effluent after the intimate coupling treatment was also assessed using rotifers as the test organism. Thus, our present study provided an integrated view of the intimate coupling UV irradiation and algal treatment on antibiotic, involving compatibility, efficiency and the microbic impact assessment.

2. Materials and methods

2.1. Test materials and culture conditions

Three algal species *S. obliquus*, *S. capricornutum* and *C. pyrenoidosa* were obtained from the Institute of Hydrobiology of the Chinese Academy of Sciences. The culture condition was maintained at 25 ± 1 °C in illumination incubator with a 12:12 h light/dark scale, using BG-11 medium. *E.coli* and *S.aureus* used in our study were donated by The College of life Science and Technology, China Pharmaceutical University. The bacterial culture condition was 37 °C in a bacteriological incubator with the beef extract peptone medium. The freshwater rotifer *B. calyciflorus* was captured from a pond on Jiangning campus of China Pharmaceutical University. The population of the rotifers was cloned from a single female and maintained in our laboratory for 3 months. The rotifers' culture condition was maintained at 25 ± 1 °C on a 12:12 h light/dark cycle in the illumination incubator with 2000 lx light. EPA medium was applied for the rotifer culture and the green alga *C. pyrenoidosa* was used as food. The medium and food has been renewed daily.

Table 1
Experimental design using three species of microalgae and ultraviolet radiation with mercury lamps four kinds of wavelength.

Antibiotic	Algal species	UV wavelength (nm)			
		185	254	280	365
Cefradine	*S. obliquus*	A1c	A2c	A3c	A4c
	S. capricornutum	B1c	B2c	B3c	B4c
	C. pyrenoidosa	C1c	C2c	C3c	C4c
Amoxicillin	*S. obliquus*	A1a	A2a	A3a	A4a
	S. capricornutum	B1a	B2a	B3a	B4a
	C. pyrenoidosa	C1a	C2a	C3a	C4a

Table 2
Market in the bacteriostatic test.

Bacteria species	Treatment group	Algal treatment time (h)		
		0	24	Combined with UV treatment
E. coli	Alga + AMX	E-A-0	E-A-24	E-A-UV-24
	Alga + Cefradine	E-C-0	E-C-24	E-C-UV-24
	Alga + Blank	–	E-Alga	–
	NS	–	E-Control	–
S. aureus	Alga + AMX	S-A-0	S-A-24	S-A-UV-24
	Alga + Cefradine	S-C-0	S-C-24	S-C-UV-24
	Alga + Blank	–	S-Alga	–
	NS	–	S-Control	–

2.2. Chemical and analytical methods

The antibiotic cefradine (95.48% purity) and AMX (95.78% purity) were purchased from Yabang investment holding group CO., LTD. The actual concentrations of the target compounds were determined by high performance liquid chromatograph (HPLC, LC-10AT, SHIMADZU). Two antibiotics were separated and determined with an Inertsil ODS column (4.6 mm × 150 mm, 5 μm). The mobile phase of the antibiotic as follows: water-methanol-3.86% sodium acetate-4% acetic acid (682:300:15:3) for cefradine and 0.05 mol/l of monopotassium phosphate (2 mol/l of KOH adjust pH to 5.0)-acetonitrile (97.5:2.5) for amoxicillin. All detections were performed by UV absorption at the wavelength of 254 nm and the Quantization was used using external standards which were based on peak areas. The limit of detection (LOD) and limit of quantitation (LOQ) of the analytical methodology were determined based on 3.3 times and 10 times the standard deviation of the background noise, respectively.

2.3. Experimental set-up

The green algae were normally cultured to reach the logarithmic growth phase before the treatment in BG-11 media at 25 ± 1 °C and 4000 lx illumination. First of all, the algae was mixed with cefradine and AMX into a photo reactor (250 ml), respectively. The final algal density and the concentration of the two antibiotics in the treatment were 10×10^6 cells/ml and 100 mg/l, respectively. The intimate coupling UV irradiation and algal treatment on the target compounds was performed using different algal species and UV wavelength (185, 254, 280 and 365 nm), respectively (see Table 1). After a 5 min (0.083 h) UV-irradiation the fluorescent lamps were shut down while we have the