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Pretreatment of contaminated raw water by a novel double-layer biological aerated filter for drinking water treatment

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ABSTRACT

Polluted source water presents a challenge for the conventional water treatment process. Biological pretreatment would be helpful if feasible. To our knowledge, few literatures were reported about the pretreatment of polluted source water by biological aerated filter. A novel double-layer biological aerated filter (DLBAF) was devised using floating media (polyethylene) and sunken media (clinoptilolite) as the under layer and the upper layer media, respectively. The results indicated that DLBAF was efficient for ammonium, TOC and UV_{254} removal, with an average removal rate of 80.87%, 24.62%, 20.74%, respectively. Particle counts with sizes of 2~15 μm presented a removal rate above 55%, although the sizes distribution had not been altered. Microorganisms on the surface of the media were observed by scanning electrical microscope (SEM), and it indicated that there were different bio-film structures in the under and the upper media. The novel DLBAF got a simultaneous removal of organic matter and ammonium, and had a great potential for pretreatment of polluted drinking water resources.

Keywords: Polluted source water; Floating and sunken media; Clinoptilolite; Double-layer biological aerated filter; Biological pretreatment; Drinking water treatment

1. Introduction

With the drinking water sources gradually polluted, challenges for conventional treatment technology lie in ammonium and organic control, especially when the source water were contaminated with poor quality of domestic wastewater. Pre-treatment of feed water by chemical pre-oxidation has been widely reported as an effectively strategy for reducing organic loading of the following process [1]. Although chemical pre-oxidation could remove the organic matter or change the properties of the organic matter to benefit the subsequent process, the possible by-products may constitute the major issue of using chemicals, such as chlorine, ozone [2,3]. Moreover, a defect of presence of ammonium in raw water is its significant influence on the disinfection process and potential formation of hazardous nitrite in the distribution networks [4]. Biological pretreatment could remove organic matter and transform ammonium to nitrate if completely nitrification was achieved [5]. Water treatment by biological methods were known to be environment friendly and be demonstrated with little byproduct. Therefore, pretreatment of polluted source water by biological methods should be a trend for drinking water treatment.

Biological aerated filer (BAF) was one of the favorite biological methods in the field of water treatment, and could effectively reduce ammonium and organic matter through microbe mechanisms [6–9]. BAF has been demonstrated as an economic and efficient contact oxidizing process for its relatively small foot print and low cost [10,11]. Today, BAF has been widely studied and applied

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in wastewater treatment, and some other biological filtration methods had been successfully researched for drinking water treatment, but limited studies were reported to lend BAF as a pre-treatment method for drinking water production. Recently, simultaneous removal of organic matter, ammonium and manganese were achieved by BAF from drinking water [12]. And it was reported that when BAF was applied for drinking water, media types were important the performance of BAF for contaminant removal [13]. Among the reported literatures, floating media and sunken media was the main filter media used. The former was reported to have the advantage for suspended solids and organic matter removal in comparison with sunken carrier, but it is not proper for biofilm culturing due to its relatively low specific surface area [14,15]. Although the sunken media had been demonstrated to have the potential for ammonium removal and the smaller size of the sunken media would be beneficial [14,16]. However, the smaller the media size was, the more frequently should the filter be backwashed [17]. The frequent backwash would not only complicate the management of the BAF, but also influence the biofilm structure, thus influence the performance of the BAF.

Therefore, there should be a hypothesis that these two kinds of media with different physical and chemical properties would benefit each other and present stable operation as an efficient pretreatment for polluted source water. The objective of this test was to develop a Double-Layer BAF (DLBAF) and to evaluate its feasibility, which integrated two kinds of media into a single column. The performance of this DLBAF in terms of ammonium, organic matter and particulates removal has been investigated.

2. Experimental

2.1. Experiment set-up

A cylinder made of plexiglass was adopted as the BAF reactor with the inner diameter and effective height at 30 mm and 2000 mm, respectively. The schematic diagram is depicted in Fig. 1. The roles in the column are following the arrangement in sequences from top to bottom as: 500 mm of clinoptilolite media, 200 mm of supported plate with scree at pore sizes of 2–6 mm and 500 mm of polyethylene media. Clinoptilolite is a kind of natural zeolite which could remove impurities from polluted water by ion-exchanging [18]. The characteristics of the two media are specifically illustrated in Table 1.

Up-flow mode was employed throughout the whole experiment, and on the bottom of this reactor, 200 mm of water distribution space was designed with the perforated aeration pipe and the perforated water distribution plate. The backwashing air and water inlets were also devised.

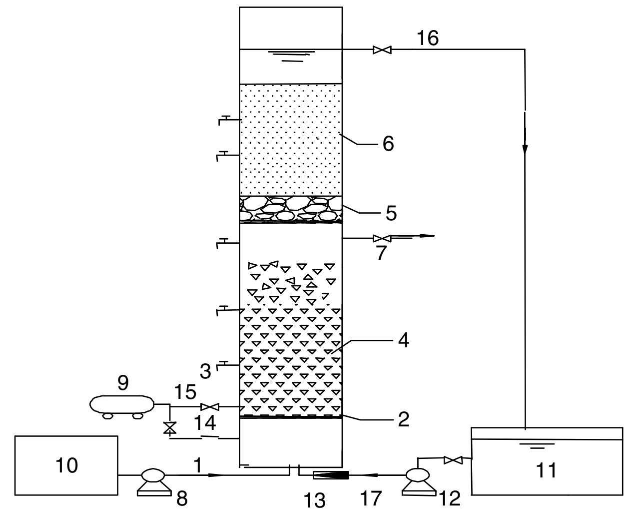


Fig. 1. Schematic diagram of experimental set-up.

Table 1
Properties of the floating and sunken media

	Floating media	Sunken media
Materials	Polyethylene	Clinoptilolite
Shape	Gear type	Irregular polyhedrons
Particle size (mm)	10 × 10	1~2
Specific Surface Area (m^2/g)	19.7378	1.7264
Density (g/cm^3)	0.949	1.75

2.2. Experimental procedure

Two stages consisted of this whole test. The each stage was summarized as follows with the identical operational conditions.

Stage one (20 d). Firstly, 7 d operation with raw water I was implemented for rapid adsorption and microbes culturing, because substrate concentration of the feed water may have a great influence on the nitrification rate and growth of the biomass in BAF media. A relatively high concentration of ammonia should be helpful for the nitrification biofilm formation. Transforming the feed water to raw water II on the 8th day was to make the reactor more accommodated with the real feed water.

Stage two (110 d). Long-term operation was to evaluate the performance and stability of this novel DLBAF.

According to our previous experiment (data not shown), the identical conditions for the two stages are optimized with Empty Bed Contact Time (EBCT) at 30 min, ratio of air to water at 1:1 and volumetric loading at 1.4 l/h.

2.3. Simulated polluted raw water

As stated above, drinking water sources are prone to be polluted by the domestic wastewater. To simulate this kind of contaminated feed water, local domestic wastewater (Harbin, China) was blended with the local tap water at a ratio of 1:15 (see raw water I in Table 2) or 1:35 (see raw water II in Table 2). The synthetic feed water was always stabilized overnight in the laboratory before use. The specific characteristics of the simulated polluted water were presented in Table 2.

2.4. Analytical methods

Feed water, effluent of the under layer and the upper layer were taken for water quality analysis. All of the water quality analysis was conducted following the standard methods. Ammonium was determined by the colorimetric methods using a spectrometer (UV721, HACH, USA); UV_{254} (pre-filtered through 0.45 μm membrane) was also determined by the spectrometer. Total organic carbon (TOC) was measured by a TOC analyzer (TOC-VCPN, Shimadzu, Japan). The particle size distribution was monitored with a particle counting system (HIAC Royco 9703 USA).

To allow a better and simple comparison, the removal rate for the DLBAF and the separate layers were conducted following the equations below:

$$R_{\text{total}} = \frac{C_{\text{raw}} - C_{\text{e-upper}}}{C_{\text{raw}}} \times 100\% \tag{1}$$

Table 2
Characteristics of the synthetic polluted source water

Items	Raw water I	Raw water II
Temperature (°C)	24 ~ 26	24 ~ 26
TOC (mg/l)	9.03 ~ 20.35	4.88 ~ 6.92
Ammonium (mg/l)	2.92 ~ 8.38	1.60 ~ 3.71
Nitrite (mg/l)	0.133 ~ 0.643	0.064 ~ 0.324
Turbidity (NTU)	29.64 ~ 104.43	25.50 ~ 85.62
UV254 (c/m)	0.231 ~ 0.434	0.112 ~ 0.167
Dissolved oxygen (mg/l)	2.02 ~ 5.87	2.12 ~ 6.34
pH	6.7 ~ 7.5	6.5 ~ 7.2

$$R_{\text{under}} = \frac{C_{\text{raw}} - C_{\text{e-under}}}{C_{\text{raw}}} \times 100\% \tag{2}$$

$$R_{\text{upper}} = \frac{C_{\text{e-under}} - C_{\text{e-upper}}}{C_{\text{raw}}} \times 100\% \tag{3}$$

$$R_{\text{upper}} = R_{\text{total}} - R_{\text{under}} \tag{4}$$

where, R_{total} is removal rate of contaminants by the whole DLBAF, R_{under} and R_{upper} are the removal rate by the under layer and the upper layer, respectively. C_{raw}, $C_{\text{e-under}}$ and $C_{\text{e-upper}}$ represent the concentration of contaminants in the raw water, effluent of the under layer and effluent of the upper layer, respectively. Note that the elapsed time for the passage of the flow from the inlet to the outlet was always brought into consideration whenever sampling and calculating the removal rate of contaminants.

3. Results and discussions

During the first stage, one week operation with raw water I was implemented, which theoretically could facilitate the mature of microbes for the media by the high substrate concentration. Naturally start-up mode was adopted for the operation of DLBAF. On the 8th day, feed water was replaced by raw water II. With time elapsing, removal of organic matter and ammonium increased. The BAF reached steady state after about 22 days' operation. With average hydraulic loading and organic loading rates at 47.5 m^3/m^3 filter per day and 279.78 g influent TOC/m^3 filter per day, respectively, the performance of DLBAF during the stable stage was presented below.

3.1. Removal of organic matter

TOC and UV_{254} are important parameters for monitoring the total organic matter and unsaturated organic matter in water. The selection of these two parameters is based on the knowledge that TOC and UV_{254} could reflect the total organic matter and dissolved organic matter in the water, respectively. The presence of organic matter in the drinking water could heavily affect the formation of disinfection by-products in the following distribution systems, thus it is very significant to evaluate the effect of DLBAF pretreatment on the organic matter removal [19]. The performance of DLBAF in terms of organic matter removal was illustrated in Fig. 2 and Fig. 3. As could be seen in Fig. 2, TOC was decreased from 5.89 ± 1.12 mg/l in the raw water to 4.44 ± 1.03 mg/l through the DLBAF treatment, with a total removal efficiency of 24.62%, and the contribution of under layer

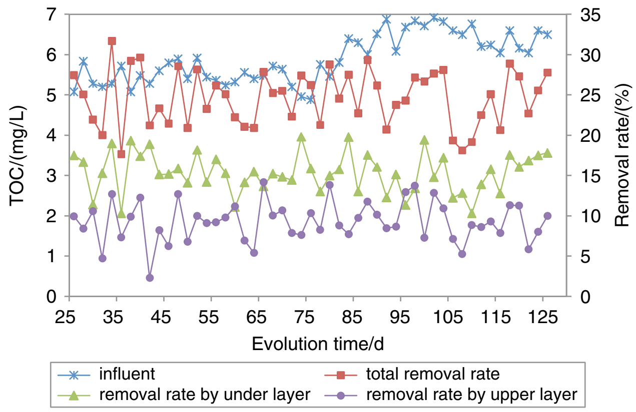


Fig. 2. Removal of TOC during the stable operation periods with operating conditions: EBCT 30 min, ratio of air to water 1:1 and volumetric loading 1.4 l/h.

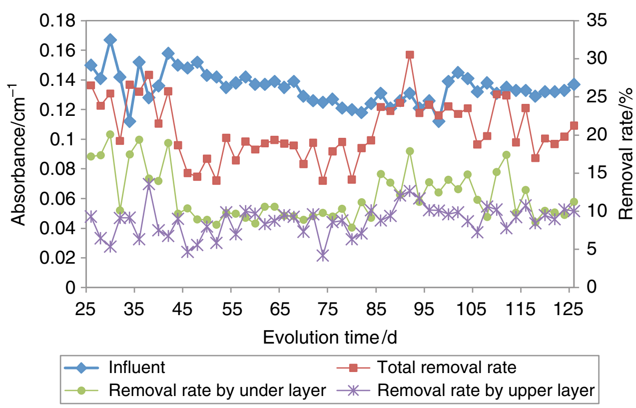


Fig. 3. Removal of UV_{254} during the stable operation periods with operating conditions: EBCT 30 min, ratio of air to water 1:1 and volumetric loading 1.4 l/h.

and upper layer for TOC removal averaged at 15.34% and 9.28%, respectively. Moreover, Fig. 3 showed that DLBAF could achieve 20.74% removal of UV_{254}, with the separate removal at 11.97% and 8.77% for the under and upper layer, respectively. As UV_{254} was determined after filtration by 0.45 μm membrane, it could indirectly represent the dissolved organic matter in the water samples.

Effective removal of TOC and UV_{254} indicated that DLBAF had a great potential for not only the total organic matter, but also the dissolved organic matter. Obviously, the under layer contributed more removal of organic matter than the upper layer, this was in accordance with that organic matter removal would decrease with the increasing height in the single layer BAF [6].

During the operation periods, the presence of the supporting plate between the two layer of media not only served as the support of the upper layer, but also induced the under layer into two kinds of status. That is, with naked eyes, it was found that the part near the supported plate was pressed due to the aeration and hydraulic lift forces, and the part farer from the supported plate appeared to be a relatively suspended or fluidized status. The former would be beneficial for the attachment and growth of bio-film due to its better rejection capacity of solids, while the latter would get more oxygen transferring due to the fluidized media [20]. Moreover, these two statuses were capable of interchanging repeatedly rather than standing alone. On the other hand, DO measurement stated that the average concentration of DO in the inlet of the reactor was 11.5 mg/l, and the DO concentration insured the necessity of biodegradation capacity for microbes. Therefore, the under layer presented a higher removal organic matter than the upper layer.

3.2. Variations of nitrogen

Removal of ammonium during the pre-treatment process would significantly benefit the subsequent water treatment process. Profiles about the variations of nitrogen were illustrated in Fig. 4.

As could be seen, high and ideal removal of ammonium was observed during the test period, with the total removal rate at 80.87%, while that for under layer and upper layer was 50.98% and 29.89%, respectively. The simulated raw water had concentration of ammonium at 2.54 ± 1.02 mg/l, while that was reduced to 0.46 ± 0.22 mg/l after the treatment of DLBAF.

With respect to the under layer light carrier, the initial operation demonstrated that this kind of carrier itself could not remove ammonium before acclimatization. As no chemicals were introduced into this process, removal of ammonium by the under layer should be attributed

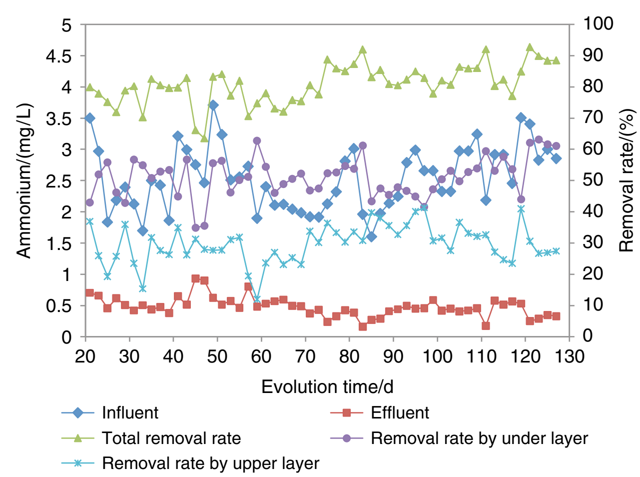


Fig. 4. Variations of ammonium during the stable operation periods with operating conditions: EBCT 30 min, ratio of air to water 1:1 and volumetric loading 1.4 l/h.

to the biodegradation mechanism of the microbes. DO concentration was important for ammonium removal [21], and as previously stated, the DO concentration is sufficient for the growth of ammonium oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB).

Literatures reported that zeolite has the potential for ammonium adsorption. Strong adsorption and ion exchange capacity was beneficial for its application in water treatment, so ammonium removal by zeolite should be attributed to adsorption, ion exchange and biological degradation [22]. After certain time of running, zeolite would lose ion exchange rate by 25%–30%, but it would not affect the adsorption capacity of zeolite [23]. In the stable operation periods, although the feed concentration of ammonium changed in the ranges of 2.92–8.38 mg/l, the effluent ammonium had always been kept near 0.5 mg/l. This should be very significant for the application of BAF, because actually the feed water could fluctuate and thus influence the biological effect especially in drinking water production.

In fact, complete nitrification included two steps and needed two kinds of bacteria. AOB was needed to oxidize ammonium to nitrite, and then NOB was needed to oxidize nitrite to nitrate. During the experimental periods, no accumulation of nitrite was observed, which indicated that DLBAF could keep stable running in response to the fluctuation of feed ammonium concentration, and complete nitrification had always been accomplished within the test ammonium concentration.

3.3. Removal of particulates

The presence and various concentrations of particulates had a great influence on the conventional water treatment process, such as coagulation and flocculation. Moreover, particulates have been demonstrated to be related to the pathogen in the water, and removal of particulates is often significant for evaluating the treatment process. To get a more sound consolidation of the feasibility of DLBAF, particulates size distribution (sizes ranging between 2 μm and 15 μm) was determined.

The total particulates in the influent and effluent were 34439.6 counts/l and 14492.2 counts/l, respectively. Note that no significant differences were found between the sizes distributions of the influent and effluent (see Fig. 5). The pressed status of floating media should be responsible for mainly removal of particulates.

Generally, the upper clinoptilolite layer, which has a smaller particle size than the under layer, was prone to be blocked by the suspended particulates. In this test, the blockage of upper clinoptilolite layer was ideally controlled via the rejection of suspended solids by the under layer. This phenomenon was very critical for the practical operation of smaller size filter, and successfully operation of DLBAF demonstrated that backwashing could be minimized if selection of filter size was appropriate.

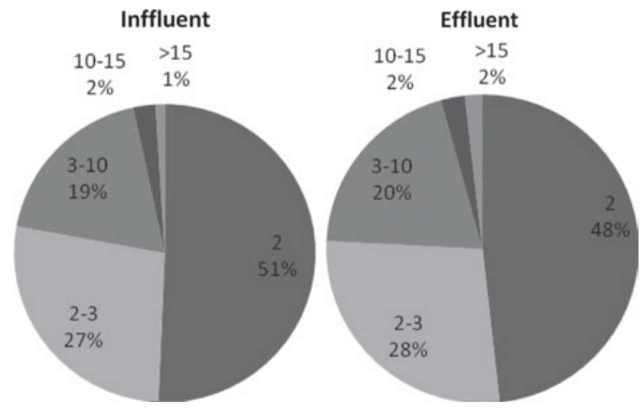
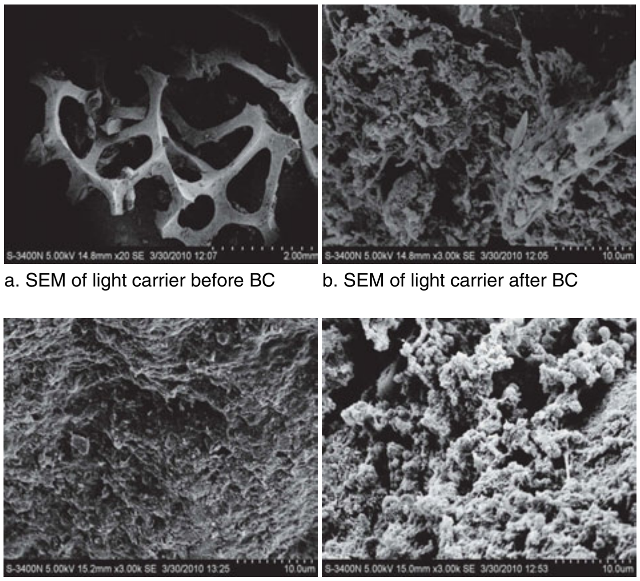


Fig. 5. Distribution of different sizes (μm) of particulates. Total inffluent number and effluent number of particulates were 34439.6 counts/l and 14492.2 counts/l.

3.4. Microscopic observation of the media

Ideal contaminant removal was achieved as stated above. At the end of this experiment, media samples were carefully taken out for SEM analysis. It is clearly to see the smooth appearance on the surface of the virgin polyethylene (Fig. 6-A), while loose biofilm structure (Fig. 6-B) with filamentary bacteria and other microorganisms was found on the media at the end of the test. While for the upper layer zeolite, virgin surface of zeolite presented a relatively dense structure with different sizes and various shapes of pores (Fig. 6-C). Denser biofilm structure was observed after acclimatization on the surface of zeolite. The abundant microorganisms observed on the surface of the media should be accounted for the removal of organic matter and ammonium.

As stated in Section 3.1, the under layer polyethylene appeared to be in a fluidized status, so the biofilm would be always impacted by the influent and the aeration force, which made the surface biofilm structure a little looser. As the upper layer had a smaller diameter and no apparent turbulence happened here, the biofilm appeared to be denser after a long-term operation. This was in accordance with the phenomena that the under layer could reject more suspended solids to insure the efficiency of the upper layer.



a. SEM of light carrier before BC
b. SEM of light carrier after BC
c. SEM of clinoptilolite before BC
d. SEM of clinoptilolite after BC

Fig. 6. Comparison of the carriers before and after biofilm culturing (BC).

4. Conclusions

Based on the hypothesis that integration of floating media (polyethylene) and sunken media (zeolite) would achieve stable operation, the performance and mechanism of the established DLBAF were evaluated. The operational conditions were Empty bed contact time of 30 min, ratio of air to water of 1:1 and volumetric loading of 1.4 l/h, it could be concluded as follows.

As a pre-treatment method, DLBAF could achieve an average removal rate at 24.62% for TOC, ranging from 17.65% to 31.71%, and an average removal rate at 20.74% for UV_{254}, ranging from 14.00% to 30.53%. Besides, DLBAF also presented an ideal removal of ammonium, average at 80.87% and ranging from 63.54% to 92.71%. It was found that DLBAF could reduce the mass of particulates, but not changing the size distribution of solids.

The two different media were believed to play synergistically. Experimental results also suggested that the under media could benefit the upper layer by reducing particulates, and the upper layer had the potential for ammonium shock load.

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