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Enhanced treatment of polluted surface water from Yellow river (China) with biooxidation as pretreatment: Pilot scale studies

Xiangyang Song

Zhengzhou Water Supply Co., Zhengzhou, P.R. China
email: sxy@zzwater.com.cn

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ABSTRACT

The Yellow River in China is being polluted with artificial pollution, which brings great challenges to drinking water treatment plants (DWTP) along the Yellow River. The conventional treatment processes could not ensure satisfactory quality of drinking water, and innovative processes are crucial for the achievement of the newly issued drinking water standard (GB5749-2006). The DWTP in Zhengzhou City takes the reservoir water as source water, which has been suffering from algal bloom of late due to the pollution of the Yellow River. This study shows the feasibility of using biooxidation processes (Moving-Bed Biofilm Reactor (MBBR) and Vceramsite Biofilter (BF)) as pretreatment for enhancing surface water treatment. These two processes show effect in enhancing natural organic matter (NOM), and more significant effect was observed for NOM with lower molecular weight and higher biodegradability. BF shows higher capabilities of removing UV254 and controlling trihalomethane formation. MBBR and BF show effect in removing algae cells, and BF contributes to higher chlorophyll a removal (with average removal of 47.4%) than MBBR does. These two biofilm reactors show great potential of removing algae toxins (MCLR), which achieve removal rates of 56% and 63% respectively. The two processes are also observed to be effective in enhancing ammonia removal, which achieve removal rates of 61.6% and 68.0% respectively. MBBR and BF are effective in enhancing pollutants removal and are potentially feasible in drinking water treatment as pretreatment of conventional processes.

***Keywords*:** Yellow river; Moving-bed biofilm reactor; Vceramsite biofilter; Algae; Algal toxins; Ammonia

1. Introduction

The discharge of large-scale municipal and industrial sewerage into the Yellow River seriously deteriorates the source water quality for drinking water treatment. The newly issued Drinking Water Standard (GB5749-2006) promoted stricter standards, increasing water quality items and decreasing the corresponsive maximum contaminant levels (MCLs). The conventional water treatment process, which mainly aims to remove the pollutants such as turbidity, microorganisms, colloids, and heavy metals, could not achieve satisfactory water quality when the source water was severely polluted. The introduction of the enhanced treatment processes to remove these pollutants, which were difficult to remove through conventional processes, was the strategy to be adopted in the cities such as Zhengzhou City.

The source water from the Yellow River was pretreated in a reservoir through natural sedimentation, which decreased the turbidity from 500 NTU to 10 NTU. However, the accumulation and pre-sedimentation in the reservoir accelerated the proliferation of algae such as cyanobacteria and so on, which resulted in the algal bloom and the eutrophication in the reservoir. The levels of different pollutants such as algae, ammonia, toxins, waste and odor increased significantly, which were difficult to be removed through conventional

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processes. Additionally, the proliferation of cyanobacteria in drinking water sources is problematic as they can interfere with water treatment processes: go against the flocculation; react with coagulant because of tiny volume, disturbing the process of destabilization; and do harm to the hydroxylation of inorganic coagulant.

The introduction of pre-oxidation may be beneficial to the treatment of the polluted water from the Yellow River. In this study, the effectiveness of enhancing pollutants removal from the polluted Yellow River water, were investigated and compared between two kinds of bioreactors (i.e. MBBR and BF) based on the conventional treatment processes. Additionally, the operational parameters were also optimized through pilot scale studies.

2. Materials and methods

2.1. Experimental reactors and operational parameters

The MBBR processes included two series-wound reactors (Figure 1a), and the area of each reactor was 1 m^2 and the effective volume was 3 m^3. The carrier filled in MBBR was LT-style suspended air balls (the diameter of each ball was 100 mm, the air voids was about 87%), whose specific gravity was near to 1 and surface area was 360 m^2/m^3. 50% of the MBBR reactor was filled with suspended air balls. The influent flux was 4 m^3/h, and the aeration was continuously done to each reactor at the influent air flux of 0.5 m^3/h. The ratio of gas to water was 0.25:1, keeping the dissolved oxygen concentration above 5 mg/L all along. The sludge was discharged every 5 days.

The sketch map of the BF reactor is presented in Figure 1b. The BF reactor was made of plexiglass (the height was 3 m, and the diameter was 0.5 m). Porous ceramics (the diameter of ceramic was from 2 to 5 mm) was filled in the reactor to 2 m. The influent flux was 1 m^3/h, and the ratio of gas to water was also kept at 0.25:1. The dissolved oxygen concentration was also higher than 5 mg/L.

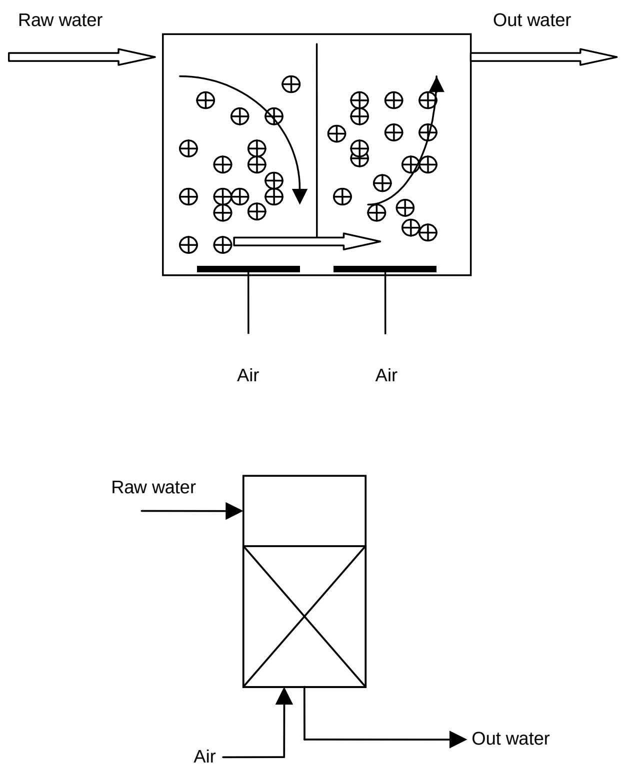

Fig. 1. Two reactors sketch map.
(a) MBBR, (b) BF

2.2. Experimental conditions

The raw water in this study, which has been pretreated by two stage grit chambers, was introduced from the influent ditch of the Shiyuan Water Works. The pilot scale study was carried out in 2004 (spaning 7 months), and the biofilms in these two reactors had well formed on the carriers and operated for about 4 months during the start-up of these processes. The temperature of the influent water was from 5 to 30°C during this study, and the pH was in the ranges of 7.8 to 8.3.

2.3. Analyses

Permanganate consumption (COD_{Mn}) was determined according to the Chinese standard methods [1]. DOC was measured with a Shimadzu-5000 TOC analyzer.

The parameters of AOC and BDOC were employed in this study to investigate the effects of these two bioreactors on the bio-stability of drinking water. The first method, the AOC bioassay, is one in which the growth of a test organism(s) is correlated with the concentration of BOM. The second method, the BDOC assay, consists of measuring the consumption of DOC through the ability of a mixed microflora to catabolize organic carbon to carbon dioxide and/or new biomass. The AOC bioassay was carried out according to an amendment of APHA 9217. *Pseudomonas fluorescens* strain P-17 was inoculated into the pasteurized sample water and incubated at 22°C for 2 d. After the number of P-17 colonies was counted, sample water was pasteurized again to kill the P-17. The water was inoculated with the Spirillum strain NOX and incubated at 22°C for the next 3 d. The value was calculated by multiplying the number of colonies by the yield rate of each strain. BDOC was measured with the method established by

Servais [2]. Enough raw water was collected and some raw water was filtered through a 2-mm polycarbonate membrane filter. The filtrate was used as inoculum for BDOC analysis. The residual raw water sample was filtered through a 0.45 mm polycarbonate membrane filter, the first 100–200 mL filtrate was discarded for avoiding possible contamination by the filter, then the sequential filtrate was collected and poured into prepared 500 ml vials. Inorganic nutrients were added to the vials with prepared amount. Then 5 mL of the inoculum was added to every vial. Incubation was conducted in the dark at 20°C for 10 days. The value of BDOC was defined as the difference between original DOC and final DOC.

3. Results and discussions

3.1. The effectiveness of organics removal through MBBR and BF

Figure 2 shows the comparision between the removal of organic matter (i.e. COD_{Mn} and TOC) by the bioreactors as MBBR and BF, through the long-term experiments spanning about seven months. The COD_{Mn} concentrations in the raw water were in the ranges of 3.28 to 4.81 mg/L, with the average value of 4.10 mg/L. It was observed in that MBBR and BF exhibited similar potential of removing COD_{Mn} (Fig. 2a). The COD_{Mn} concentrations in the MBBR effluent were from 2.50 to 4.33 mg/L (with the average value of 3.63 mg/L), which

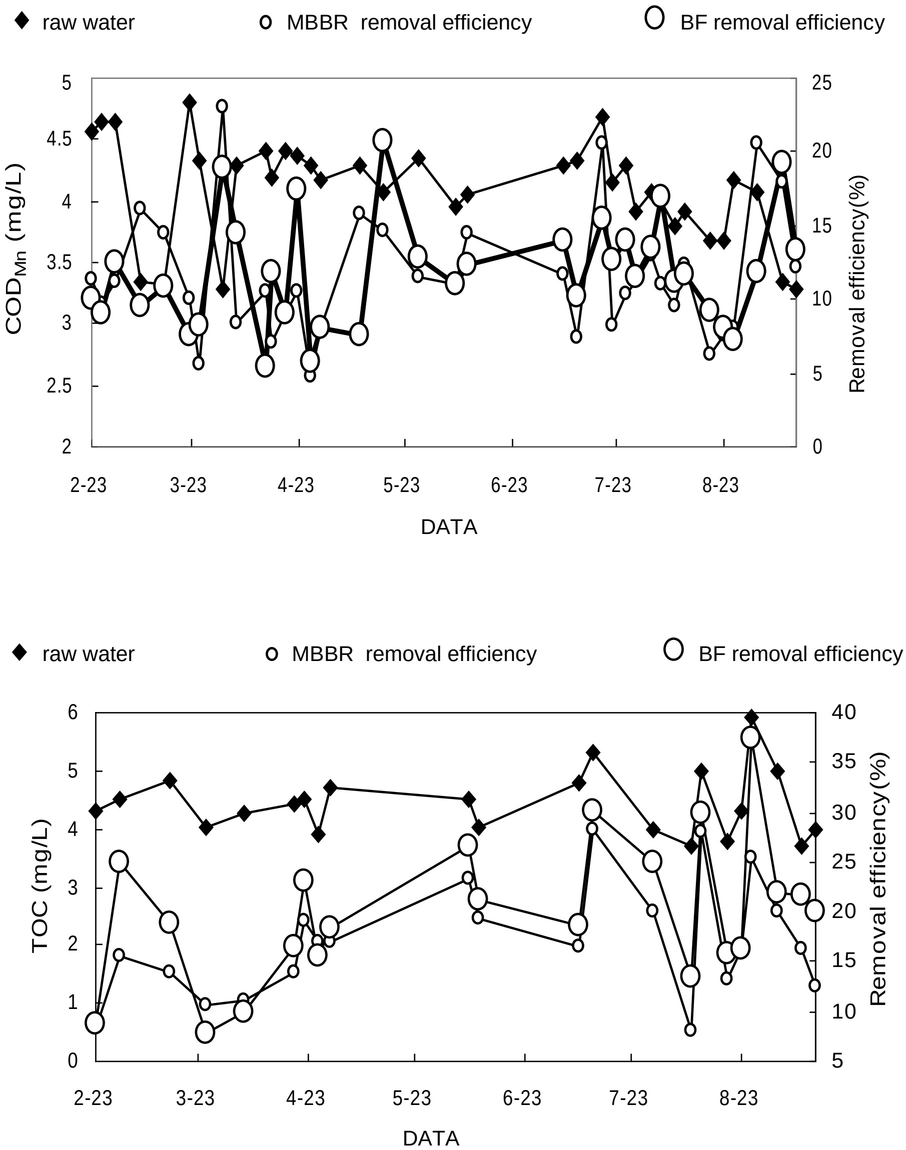

Fig. 2. The removal of organics matter (i.e. COD_{Mn} and TOC) by the bioreactors as MBBR and BF. (a) COD_{Mn}, (b) TOC.

corresponded to the COD_{Mn} removal efficiency from 4.67 to 23.1% (with the average value of 11.53%). Similarly, the COD_{Mn} concentrations in the BF effluent were from 2.66 to 4.16 mg/L (with the average value of 3.62 mg/L), which corresponded to the COD_{Mn} removal efficiency from 5.45 to 20.8% (with the average value of 11.67%).

However, it was indicated that BF showed higher potential of removing TOC than MBBR. The TOC concentrations in the source water were from 3.70 to 5.90 mg/L (with the average value of 4.43 mg/L). MBBR contributed to the TOC removal efficiency from 6.97 to 28.30% (with the average value of 16.96%), and the residual TOC concentration were from 3.10 to 4.40 mg/L (with the average value of 3.66 mg/L). In comparison, the TOC removal efficiency in BF unit was from 7.76 to 37.29% (with the average value of 20.09%).

In bioreactors, the organics were removed through the main mechanisms such as bio-assimilation, biodegradation, and adsorption. The aeration supplies the oxygen (DO), which was necessary for the growth and propagation of microorganisms in the bioreactor units. Additionally, the carriers in these reactors provide the medium to support and accelerate the formation and regrowth of biofilms, which increases the bacterial quantity for the degradation of organic matters. Additionally, the biofilms would also supply the bio-secretion during bacterial metabolization, acts as bio-flocculants and prove beneficial to the removal of organic matter and particles[3].

3.2. The removal of AOC and BDOC by MBBR and BF—organic characteristics effects

The parameters of COD_{Mn} and TOC could only provide information on the total organic concentration, failing to supply information on the characteristics of organics speciation, functional characteristics, and bioavailability. However, the organics characteristics plays significant role in the organics removal, especially for the bio-reactors such as MBBR and BF. The parameters as assimilable organic carbon (AOC) and biodegradable dissolved organic carbon (BDOC), which were indicative on the bioavailability of organics, provided important information on the organics removal by bioreactors.

Table 1 compares the effectiveness of removing AOC and BDOC between MBBR and BF. As indicated, the concentrations of AOC and BDOC were 0.276 mg/L and 1.57 mg/L, respectively. Both MBBR and BF exhibited promising potential of removing AOC and BDOC, and decreased the concentrations of AOC and BDOC to 0.158 mg/L, 0.104 mg/L and 0.542 mg/L, 0.213 mg/L. Comparatively, BF obviously showed higher capability of removing AOC and BDOC. As for AOC and BDOC, the average removal efficiency was 62.3% and 86.4% in BF unit, and 42.8% and 65.5% in MBBR, respectively.

The porous ceramics in BF showed good surface characteristics for the formation and regrowth of biofilms, such as coarse surfaces, high BET surface areas, micro- and meso-pore structures. The high biomass and its high bio-reactivity were important for the removal and degradation of organics in the system. The biofilm on the porous ceramic surfaces, together with the bio-flocculation, bio-adsorption, and filtration reactions contributed to the removal of organic colloids [4]. Comparatively, the carriers in the MBBR reactors, together with the biomass on the surfaces of carriers, were fluidized by the influent and air aeration, which inhibited the effects such as bio-flocculation, bio-adsorption, filtration, and bio-assimilation.

Natural organic matter (NOM) can be divided into two fractions: biodegradable, and refractory. Numerous methods have been developed to quantify the biodegradable fraction of organic matter in water, from which two major established methods exist today for the measurement of biodegradable organic matter (BOM). AOC refers to a fraction of the total organic carbon (TOC), which can be utilized by specific strains or defined mixtures of bacteria, resulting in an increase in biomass concentration that is quantified. AOC typically comprises of just a small fraction (0.1–9.0%) of the TOC [5], and represents the most readily degradable fraction of BDOC/BOM. However, BDOC is the difference between initial DOC of the water sample and the minimum DOC observed during the incubation period of 28 days for suspended indigenous bacteria or 5–7 days for bacteria attached to sand. The BDOC concentration represents the fraction of DOC that is both mineralized and assimilated by heterotrophic flora, determined as the difference between the initial DOC concentration and the minimum DOC concentration observed during the incubation period. Joret et al. (1991) suggested that BDOC values represent

Table 1
Effectiveness of removing AOC and BDOC between MBBR and BF.

	Raw water	MBBR out water	MBBR removal efficiency(%)	BF out water	BF removal efficiency(%)
AOC(μg/L)	276	158	42.8	104	62.3
BDOC(mg/L)	1.57	0.542	65.5	0.213	86.4

10–30% of the total dissolved organic carbon content of drinking water [6]. AOC and BDOC have often been measured separately as indicators of bacterial regrowth, or together as indicators of bacterial regrowth and disinfection, by product formation potential, respectively [7].

The removal of AOC and BDOC were important for the water stabilization and bacterial regrowth control in the drinking water distribution system. An absence of biodegradable organics after water treatment to limit bacterial regrowth has been recommended in the literature [8]. In this study, it was observed that the MBBR and BF reactors exhibited good potential of removing AOC and BDOC, which was beneficial to the increase of water stability and the control of bacterial growth in the distribution systems of Zhengzhou City.

3.3. The control of disinfection by-products formation by MBBR and BF

The organics in the source water acted as DBPs precursors, and would produce carcinogenic DBPs during subsequent chlorination. The decrease of NOM concentration and the transformation of NOM species would change the DBPs formation potential in the effluent.

Figure 3 compares the effectiveness of controlling THMs formation between MBBR and BF. The THMs formation potential (THMsFP) was from 24.20 to 37.80 μg/L (with the average value of 31.67 μg/L). Results indicated that the THMsFP of the MBBR effluent were from 19.1 to 31.6 μg/L (with the average value of 24.47 μg/L). The MBBR unit decreased the THMsFP by 22.07%. BF showed higher potential of controlling subsequent THMs formation than MBBR, and the percent of THMsFP decrease was from 16.81 to 52.38% (with the average value of 35.08%) as for the BF effluent.

Combining the results in Figure 2, it was observed that the removal efficiency of organics (i.e. COD_{Mn}, TOC), for a specified bioreactor, was lower than the corresponsive THMsFP decrease. Young et al. also reported that the biological activated carbon (BAC) unit contributed to the total DOC removal efficiency of 13 to 25%, and the corresponding THMsFP removal efficiency of approximately 20 to 33% [9]. It was noted that the organics in the source water, which was expressed as COD_{Mn} and TOC, was the DBPs precursors during disinfection. Consequently, it was inferred that the bio-oxidation process in MBBR and BF units was inclined to remove the organics with higher THMsFP. Former studies also indicated that the bio-oxidation processes contributed to the removal of organics species with lower molecular and higher THMsFP.

3.4. The removal of algae and toxins by MBBR and BF

As for a drinking water source which was characterized by eutrophication, the enhanced removal of algae and its bio-secretim (e.g. toxins) was of critical importance. Figure 4 compares the effectiveness of removing algae between MBBR and BF. The total algal counts in the raw water were in the ranges of 0.79 to 4.33 million counts per liter. BF led to the average algae removal of 47.3%, which was slightly higher than that of MBBR (46.3%).

The efficiency of removing algae from bioreactors has also been investigated before. Wu and Wang have investigated the effectiveness of 3 different kinds of bio-oxidation processes for algal removal before. As reported, the aerated submerged biological filter gains a steady removal rate for the gross algae, an average of about 70%; for the air-lift biological contact oxidization reactor equipped with elastic YDT as the media and the direct micropore aerated biological reactor, the average algal removal rates are 60.2% and 51.6% respectively

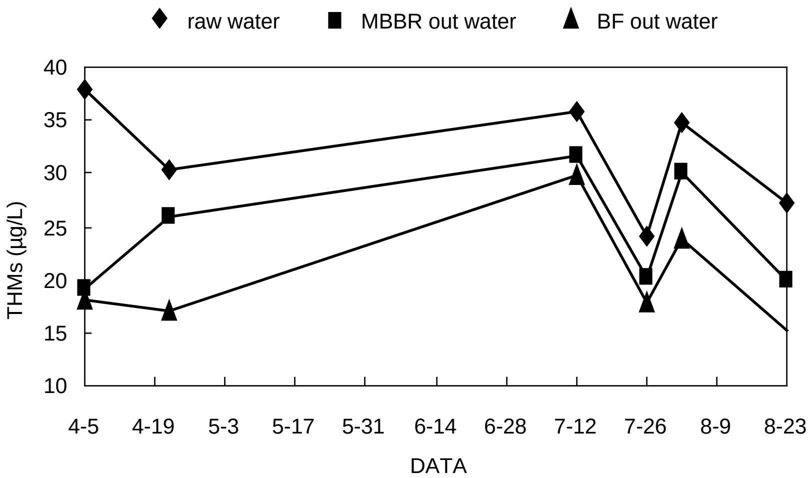

Fig. 3. The effectiveness of controlling THMs formation between MBBR and BF.

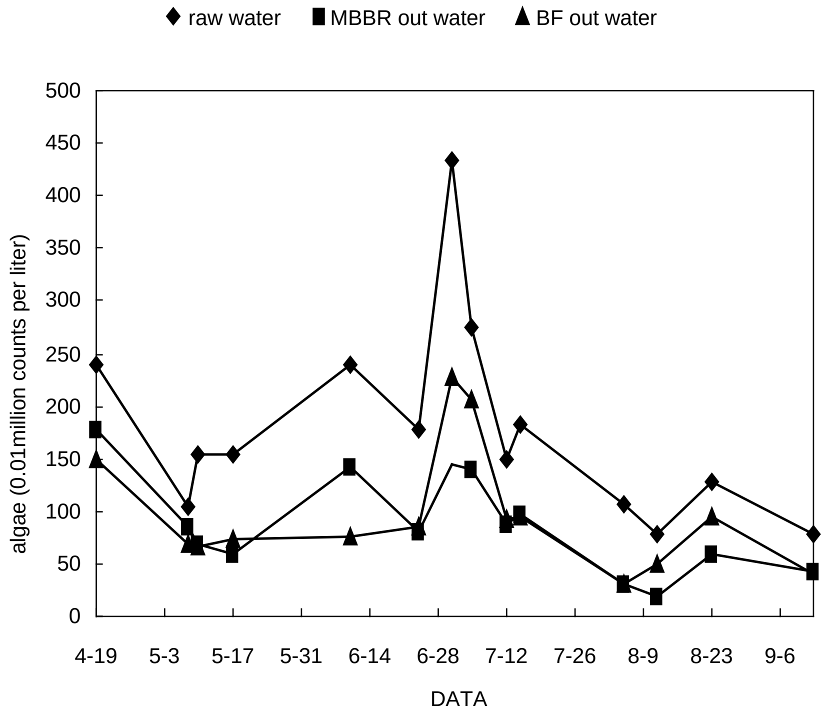

Fig. 4. The effectiveness of removing algae between MBBR and BF.

within the beginning stage of the experiment, but gradually rises to more than 70% (on an average) during the later stages of the experiment with the increase of biofilm depth. They ascribed the substantial algal removal ways to be the effects of bio-flocculation, adsorption, detachment, and sedimentation by the biofilm [10]. Shaher et al. also indicated the effective removal of algae by a filter made out of chopped wheat straw [11].

MBBR and BF also showed promising removal potential for algal toxins (i.e. Microcystin-LR), which contributed to the MCLR removal of 56% and 63%, respectively. The mechanisms of toxins removal in bioreactors, such as MBBR and BF, have been rarely investigated till now, and may be ascribed to the bio-chemical and physio-chemical reactions involved in these processes. Lv et al. studied the degradation of microcystins RR, YR, and LR and also of *Micocystis viridis* by using batch biofilm reactors, reporting that the oxic reactor was much more effective than the anoxic reactor for the degradation of *Micocystis viridis* and microcystins. In the oxic reactor, the concentrations of RR, YR, and LR decreased from 363.4 μg/L, 178.3 μg/L, and 116.1 μg/L to 50.2 μg/L, 35.9 μg/L, and 15.0 μg/L in 12 h. The microcystins removal was higher than 90% after 24 h, and was below the detection limit after 73 h. It was inferred that the biofilms in the MBBR and BF and the aeration played significant roles in the removal of toxins in the source water.

4. Conclusions

From results presented above, major conclusions as follows may be stated:

1. MBBR and BF show satisfactory potential of removing different pollutants, such as organics, algae, toxins, AOC, and BDOC in source water, and could effectively decrease the THMsFP.
2. MBBR and BF show different behaviors of enhancing the removal of the pollutants noted above, and this is ascribed to the difference in the reactor style, carrier, biomass community composition, the diffusing efficiency of substrate and dissolved oxygen.
3. The bioreactors as MBBR and BF may be employed as pretreatment of the conventional water treatment processes in Zhengzhou City, aiming to enhance the removal of different pollutants in the polluted Yellow River.

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