

Research on treating algae-polluted reservoir water by the process of pre-oxidation/dissolved air flotation/carbon sand filter

Yonglei Wang, Wenhao Wang, Ruibao Jia, Mei Li, Baozhen Liu, Kefeng Zhang, Wuchang Song and Junqi Jia

ABSTRACT

As the water diversion reservoir showed high organic matter and high algae in summer, the potassium permanganate pre-oxidation/dissolved air flotation/carbon sand double filter process was developed. The test results show that the optimum operation conditions of the combined process were as follows: the dosage of KMnO_4 was 0.3 mg/L, the dosage of polymeric aluminum ferric chloride (PAFC) was 3.0 mg/L (Al^{3+}), the reflux ratio was 10%, and the dissolved gas pressure was 0.3 Mpa. Taking Ji'nan Qeshan reservoir water algae pollution as the research object, the average removal rate of chlorophyll *a*, blue-green algae, turbidity, particle number and total organic carbon (TOC) reached 66.64%, 95.44%, 94.45%, 99.34% and 46.68%, respectively; the methylisoborneol (MIB) removal rate was 92.47%, the odor level decreased with process flow from raw water level 4 to effluent level 1.5, geosmin (GSM) dropped below the detection limit, and the total removal rate of trihalomethane formation potential (THMFP) was 33.56%. The effluent of the combined process meets the requirements of the *Hygienic Standard for Drinking Water* (GB5749-2006) after it is disinfected.

Key words | algae polluted reservoir water, carbon sand double layer filter, dissolved air flotation, potassium permanganate pre-oxidation

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INTRODUCTION

With increasingly serious groundwater pollution (Fan *et al.* 2016; Chai 2017; Liu *et al.* 2017), reliance on the lower reaches of the Yellow River as a source of drinking water has gradually increased. Due to the shallow water and the poor liquidity of the Yellow Reservoir, the water bodies show the characteristics of low temperature and low turbidity in winter and the characteristics of high algae and high organic matter in summer, and these characteristics pose a certain threat to the water quality of urban water supply. At present, most water plant processes are coagulation–sedimentation–filtration–disinfection conventional processes, and the effluent quality cannot always meet the requirements of *Drinking Water Health Standards* (GB5749-2006). Regarding the characteristics of water quality in Yellow River reservoirs, domestic scholars have carried

out a lot of research and engineering demonstrations (Lu *et al.* 2013; Song *et al.* 2015). Ruibao Jia and others took the '11th Five-Year Plan' national water pollution major science and technology special as an opportunity to carry out drinking water safety technology research and engineering demonstrations on the lower reaches of the Yellow River (Jia *et al.* 2012a, 2012b), and some water plants such as Jinan Yuqing water plant, Quehua water treatment plant, and Dongying Nanjiao water plant have undergone successful process transformations (Guo *et al.* 2012; Li *et al.* 2012). But algae, organic matter, smell and so on have been plaguing the operation of waterworks. Abeynayaka, Teixeira and others have found that air flotation has a strong removal effect on algae in water, and the combination of coagulation and air flotation

has better removal efficiency (Teixeira *et al.* 2010; Abeynayaka *et al.* 2016). Studies have shown that potassium permanganate pre-oxidation has a good removal effect on organic pollutants and odor (Chen 2017). In this study, the common problem of the water quality in the reservoir of the lower Yellow River in the Yellow River city was taken as the starting point from which to research the combined water purification process of the pre-oxidation of potassium permanganate/dissolved air flotation/charcoal filter. A pilot test study was conducted to optimize the process parameters and to investigate the adaptability of the combined process, so that after the disinfection of water it was possible to meet the *Drinking Water Health Standards* (GB5749-2006) requirements. This process is expected to improve on the conventional techniques of the water plant, which uses Yellow River Reservoir water as the water source, to provide technical support.

MATERIALS AND METHODS

Test device

The pilot test has a capacity of 5 m³/h, a pre-oxidized column before the addition of liquid KMnO₄, and oxidation contact for approximately 5 min, and the liquid coagulant polymeric aluminum ferric chloride (PAFC) was added before entering the mixing tank. The floc was flocculated after the flocculation basin into the flotation tank, by air contact, and after the separation of the effluent into the charcoal filter, water filtered through the charcoal water, the process

of which is shown in Figure 1. The design parameters of each unit of the test plant are shown in Table 1.

Coagulation conditions: the flocculant used was PAFC (the density was 1.2 g/cm³ and the mass fraction was 10%, Zibo Lujing), stirring rate of 50 r/min, reaction stirring rate of 15 r/min. Air flotation conditions: the average diameter of bubbles generated by the air flotation system was about 40–50 μm. The pressure of the gas soluble tank was 0.3 MPa, and the dissolved oxygen (DO) value was 11.30 mg/L. The releaser was a TS-1 releaser (manufacture by Tongji University).

Raw water quality

Raw water was from Jinan Queshan Yellow River Reservoir; during the test, the temperature of the raw water was 23–27.0°C, pH was 7.8–8.48, and the raw water quality indicators were those listed in Table 2. It can be seen from the table that the content of organic matter and algae in the raw water was relatively high, with blue-green algae up to 106 cells/L, indicating that the raw algae organic pollution was quite serious.

Analytical method

The test period was from August to October. During the test, samples were taken twice a day, one sample at a time, and a single sample was tested three times under the same conditions. In the test, the testing methods of COD_{Mn}, turbidity, PH, UV₂₅₄, ammonia nitrogen and other conventional indicators are as shown in the *Standard Methods for the*

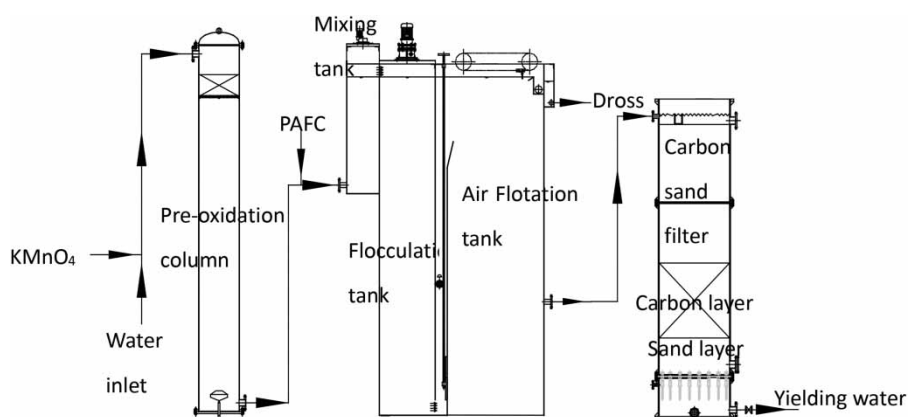


Figure 1 | Pilot test device of combined process.

Table 1 | Design parameters of the test plant

Unit	Design parameters	Remarks
Pre-oxidation column	Diameter 0.5 m, height 6.3 m, closed stainless steel equipment. Hydraulic retention time 5 min, processing capacity 15 m ³ /h	Can be pre-oxidation of potassium permanganate, chlorine/chlorine dioxide pre-oxidation, ozone pre-oxidation
Flotation tank	2.6 m × 0.8 m × 4.5 m, open stainless steel equipment. Air flotation unit: residence time is approx. 20 min	TS type release
Carbon sand filter	Activated carbon + quartz sand double filter Filter speed 8.5 m/h Recoil cycle 12–24 h	Activated: Particle size d0.5–d1.2, height 800 mm Quartz sand: Particle size d0.5–d1.2, height 400 mm

Examination of Water and Wastewater (APHA/AWWA/WEF 2005), and the specific parameters are provided with supplementary information (available with the online version of this paper). The number of particles was detected by a particle counter (IBR Versa Count, Hangzhou Green Cleaning).

Geosmin (GSM) and methylisoborneol (MIB) were detected by solid phase extraction and gas chromatography–mass spectrometry. The detection method was as follows: the water samples were filtered by an organic membrane of 0.25 µm, and then allocated to 20% sodium chloride solution and put into the corresponding device for testing.

Trihalomethane formation potential (THMFP) was detected by gas chromatograph (6890N gas chromatograph (Agilent)). The detection method was as follows: the water sample was placed in the incubator at 25 ± 2°C for 5 days at pH 7.0 ± 0.2 (the free chlorine was 3–5 mg/L), then the content of trihalomethane was measured.

TEST RESULTS AND DISCUSSION

Optimization of operating parameters

Potassium permanganate dosage

A dosing pump was used to add KMnO₄ before the pre-oxidation column, the pre-oxidation time was 5 min, and the dosages were 0.1 mg/L, 0.3 mg/L, 0.5 mg/L, 0.8 mg/L, and

0 mg/L for comparison, and the resulting turbidity, COD_{Mn} and UV₂₅₄ removal effects are shown in Figure S1 (available with the online version of this paper).

The raw water was treated with the combined process. With the increase of KMnO₄ dosage, the turbidity and the removal rate of organic matter first increased and then leveled off, and when the dosage was 0.3 mg/L or higher, the removal efficiency index was basically constant, and the removal rate of COD_{Mn}, UV₂₅₄ and turbidity were 40.25%, 44.09% and 88.93%, increased by 20.52%, 20.17% and 13.88%, respectively over those of the no peroxidation values. KMnO₄ oxidation of organic matter in water resulted in a higher activity of hydrated manganese dioxide, with catalytic oxidation and adsorption affecting the hydrophilic properties of organic compounds, strengthening the coagulation effect, and encouraging the formation of dense flocs, thereby enhancing the removal of organic matter (Yang *et al.* 2006). When the dosage of KMnO₄ was 0.5 mg/L, the effluent of the flotation tank was slightly yellow, and turbidity was increased, which indicated that the KMnO₄ concentration was excessive. It was determined that the appropriate dosage of KMnO₄ for pre-oxidation was 0.3 mg/L.

Coagulant dosage of flotation unit

Coagulation conditions directly affect the air flotation effect, as excessive or insufficient coagulant dosage will affect the

Table 2 | Raw water quality during test period

Project	Turbidity (NTU)	UV ₂₅₄ (cm ⁻¹)	Ammonia nitrogen (mg/L)	COD _{Mn} (mg/L)	Chlorophyll (µg/L)	Blue-green algae (cells/mL)	Particle number (counts/mL)
Average	5.83	0.035	0.23	3.5	4.4	919	30,052
Range	2.49–7.42	0.023–0.047	0.126–0.335	2.43–4.06	3.90–5.60	838–1,028	34,591–88,627

floc and thus affect the air flotation effect (Edzwald 1995; Lundh *et al.* 2000).

The dosages of PAFC of 1.0 mg/L, 2.0 mg/L, 3.0 mg/L, and 4.0 mg/L (Al^{3+}) and the removal effect of air flotation on pollutants were examined (Figure S2, available online). With the increase of coagulant PAFC dosage, the removal rate of pollutants increased accordingly; when the dosage increased from 1.0 mg/L to 3.0 mg/L, the removal rate of turbidity increased from 30% to 77.01%, the removal rate of UV_{254} increased from 10.26% to 27.36%, and the removal rate of COD_{Mn} increased from 8.26% to 31.38%. When the dosage was more than 3.0 mg/L, the volume of floc was obviously increased, which caused the flotation strength of the flocs to be dependent on the carrier floc, and as the stability of the carrier floc was poor, the air flotation effluent effect declined. Therefore, for the coagulant PAFC, the suitable dosage is recommended as 3.0 mg/L (Al^{3+}).

Air flotation unit reflux ratio

The reflux ratio directly influences the treatment effect and economic performance of the air flotation process (Lundh *et al.* 2000). The reflux ratio is related to the pressure of the system, the bubble characteristics, dissolved gas pressure and the floc properties (Chen *et al.* 2009). In the feed water treatment, the reflux ratio is usually controlled at 5% to 15% (He *et al.* 2006). Optimizing the reflux ratio not only can improve the dissolved gas effect but can also control operating costs.

The test results show that when the dissolved gas pressure was approximately 0.3 Mpa, the micro-bubbles were small and dense, milky white, and the system ran stably. Therefore, the reflux ratio of the dissolved gas system under 0.3 Mpa is optimized, along with the air flotation process water inflow of $5 \text{ m}^3/\text{h}$ and PAFC dosage of 3.0 mg/L. For reflux ratios of 6%, 7%, 8%, 9%, 10%, 12%, and 14%, the removal efficiency of turbidity, UV_{254} and COD_{Mn} in the air flotation unit is shown in Figure 2(a).

As we can see from Figure 2(a), the removal rate tends to be stable or slightly decreased as the reflux ratio is increased to 10%. The effect of reflux ratio adjustment on turbidity was significant. When the reflux ratio was increased from 6% to 10%, the removal rate was increased by 21.7%.

The test results show that the proper increase of reflux ratio was beneficial for improving the treatment efficiency of the flotation tank. When the reflux ratio was increased to a certain extent, excessive micro-bubbles may increase and gather, and influence the flotation efficiency; moreover, the increase in reflux ratio will increase power consumption. Therefore, it was determined that the optimum reflux ratio of the flotation tank is 10%.

The effect on the removal of pollutants by this process

With a water flow rate of $5 \text{ m}^3/\text{h}$, the KMnO_4 dosage of 0.3 mg/L, the PAFC dosage of 0.3 mg/L (Al^{3+}), reflux ratio of 10%, and dissolved gas pressure of 0.3 Mpa, the carbon sand filter adopts the downward flow to complete the

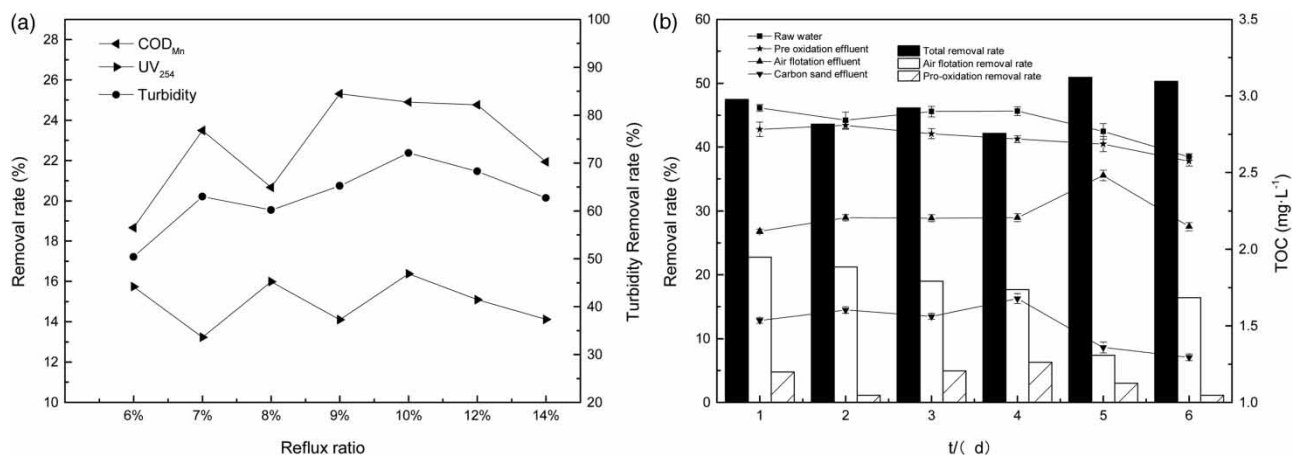


Figure 2 | (a) Removal efficiency of pollutants at different reflux ratios; (b) removal efficiency of TOC in each unit of the combined process.

process. The effects of the combined process on each removal index were investigated by trial.

Organics

The total organic carbon (TOC) of raw water was in the range 2.60–2.92 mg/L, and the TOC of the effluent of the combined process was 1.29–1.68 mg/L; the average removal rate was 46.76%. The results of the experiment show that the combined process was good for the removal of organics (Figure 2(b)). The pre-oxidation of KMnO_4 enhanced the removal efficiency of organics (Chen & Yeh 2005). On the one hand, due to the strong oxidative nature of KMnO_4 , the large organic molecules were decomposed into small molecular organic matter, which changes the chemical molecular structure of the original organism, and these small molecular organics produced by oxidative decomposition were more easily adhered to by micro-bubbles. On the other hand, the intermediates, which were oxidized by KMnO_4 , were partially biodegradable and thus easily removed by the subsequent adsorption and biodegradation by activated carbon. The air flotation unit removed the colloidal and suspended organic matter in water by exploiting micro-bubble flotation. The activated carbon filter material in the carbon sand filter can play an important role in the adsorption and biodegradation of organic matter. After air flotation, the water had better dissolved-oxygen conditions for carbon biodegradation, and enhanced its biodegradability.

Number of particles

During the test, the number of raw water particles was 34,591–88,627 counts/mL, the removal efficiency of the combination was stable between 99.00% and 99.56% (mean value of 99.34%), the suspended particulate matter of raw water was heavy elements in the range of 2–5 μm , and the removal of particulate matter mainly occurred in the air flotation unit,

as small air bubbles can effectively adhere to the particles. The removal of particulate matter by potassium permanganate is mainly due to its coagulation and own oxidation. In addition, the carbon sand filter unit contribution is also more obvious, due to the large specific surface area of activated carbon, and there are many oxygen groups on the carbon surface, due to the force of chemical bonds, and the suspension particles have a certain adsorption capacity. In addition, after the operation of activated carbon for a period of time, a biofilm grows on the surface of the activated carbon, which has the function of adhesion to particles, and also enhances the retention ability of the filter layer.

Odorous substances

During the test period, the odor produced by the reproduction of blue-green algae was mainly caused by the actinomycetes and algae in the raw water (Shen 2010).

The odor of raw water was analyzed, the analysis of odor and taste showed four levels of earthy odor, the content of MIB was approximately six times the national standard for drinking water, and the content of GSM was not in excess of the standard. The effect of the combined process on the removal of the odorant is obvious (Table 3), with the units decreasing step by step; odor level decreased from 4 in the original water down to 1.5 in the effluent, GSM in the flotation unit fell below the detection limit, the removal rate of MIB was 92.47% (pre-oxidation, air flotation, carbon sand filtration removal of 21.67%, 35%, 35.8%, respectively), and the effluent GSM met the *Hygienic Standard for Drinking Water* (GB5749-2006) limit requirements (reference limit of 0.00001).

Although KMnO_4 will destroy the algal cells and release odorous substances, the KMnO_4 's strong oxidation can decompose odorous substances, and the intermediate product of KMnO_4 (manganese dioxide) complex, adsorption and removal, can cause the odorous substances in the

Table 3 | Removal efficiency of odorous substances by the combined process

Project	Raw water	Pre-oxidation effluent	Flotation effluent	Carbon sand filter effluent	National standard/reference value (GB5749-2006)
Odor level analysis	Earthy odor, 4 grade	Earthy odor, 3.2 grade	Geosmin, 2.9 grade	Geosmin, 1.5 grade	no
GSM (mg/L)	$6 \times 10^{-6} \pm 0.023$	$5.8 \times 10^{-6} \pm 0.026$	$<5 \times 10^{-6}$	$<5 \times 10^{-6}$	0.00001
MIB (mg/L)	$6 \times 10^{-5} \pm 0.019$	$4.7 \times 10^{-5} \pm 0.021$	$2.6 \times 10^{-5} \pm 0.028$	$4.5 \times 10^{-6} \pm 0.025$	0.00001

Table 4 | Removal efficiency of THMFP in each unit of the combined process

Sampling port	Concentration ($\mu\text{g/L}$)			
	Chloroform	Bromodich	Dibromoch	Tribrometh
Raw water	49 ± 0.021	41.1 ± 0.019	33 ± 0.023	4 ± 0.023
Pre-oxidation	43 ± 0.022	38.6 ± 0.023	31.9 ± 0.025	3.6 ± 0.027
Air flotation tank	36 ± 0.019	36.3 ± 0.022	26.9 ± 0.026	3 ± 0.022
Carbon sand filter	28 ± 0.028	31.2 ± 0.021	23 ± 0.024	2.5 ± 0.026

water to further reduce (Zhao 2008). In addition, both GSM and MIB are cyclic alcohols, which are semi-volatile organic compounds, which can be removed by air flotation, and the surface of the activated carbon filter in the carbon sand filter has a large porosity, which can also effectively remove the odorous substances.

THMFP

Trihalomethanes (THMs) have a high carcinogenic, teratogenic and mutagenic risk, and they are one of the chemical classes most frequently detected in drinking water disinfection by-products (Wang & Liu 2002). THMFP is the natural organic matter that can react with chlorine to produce THMs. If THMFP is removed before chlorination, the amount of THMs produced can be effectively controlled.

During the experiment, the effluent THMFP content of each unit is shown in Table 4. The combined process has a good removal effect for THMFP with a total removal rate of 33.36%. The removal rates of THMFP by air flotation and carbon filtration were 11.72% and 13.77% respectively, and the removal rate by pre-oxidation was lower and was 7.87%. There was no significant change in THMFP components in each process unit.

Table 5 | Removal rates of non-regular indices by the combined process

Project	Water inlet			Yielding water			Average removal rate (%)
	Maximum	Minimum	Average	Maximum	Minimum	Average	
Chlorophyll <i>a</i> ($\mu\text{g/L}$)	5.6	3.9	4.4	1.5	1.4	1.5	66.64
Blue-green algae (cells/mL)	1,028	838	919	54	29	42	95.44
Particle number (counts/mL)	34,591	88,627	30,052	296	123	201	99.33

The reason why the combined process removes THMFP is as follows. First, because KMnO_4 selectively destroys the active sites on the THMFP molecule, the ability to generate new organics is poor (Fang 2010). Second, the oxidized disinfection by-product precursor is mostly low molecular weight organic matter, for which activated carbon has excellent adsorption properties.

Algae

During the experiment, the contents of chlorophyll *a* and blue-green algae were in the ranges 3.9–5.6 mg/L and 838–1,028 cells/mL (Table 5), respectively, and the removal rate of chlorophyll *a* was in the range 61.20–73.25% (mean value of 66.64%) and the removal rate of blue-green algae was in the range 94.39–97.01% (mean value of 95.44%). The combined process has a high removal effect on algae, mainly due to the efficient removal of algae by air flotation; in addition, the strong oxidation ability of KMnO_4 can directly kill algae, and the strong oxidizing property destroys the organic layer of the algae, which causes the oxidant to destroy the cell enzyme system (Shuang et al. 2011).

Technical and economic indicators

From the economic point of view, the indicators of the combined process are better than the traditional process. Detailed information is in the supplementary information (available with the online version of this paper).

CONCLUSIONS

- (1) The optimal operation conditions of pre-oxidation/dissolved air flotation/carbon sand filter are as follows: the suitable dosages of KMnO_4 pre-oxidant and coagulant

PAFC are 0.3 mg/L and 3.0 mg/L (Al^{3+}) respectively, and the pressure of dissolved gas is 0.3 Mpa, and the reflux ratio is 10%. The effluent concentration of TOC is 1.29–1.68 mg/L (total removal rate is 46.68%), the turbidity of effluent is 0.30–0.42 NTU (total removal rate is 94.45%), and the average removal rates of particle number, chlorophyll *a* and cyanobacteria are 99.34%, 66.64% and 95.44% respectively.

- (2) The combined process can effectively remove odor and the potential of trihalomethane formation. The smell of effluent is 1.5, the removal rate of MIB is 92.47%, and GSM is below the limit of detection in the air flotation unit. The total removal of THMFP by the combined process is 33.36%, and the THMFP was mainly removed in the air flotation and carbon filtration units.
- (3) In the combined process, KMnO_4 pre-oxidation can enhance the coagulation effect, effectively oxidizing organic matter and algae; after flotation, dissolved air and water provide better dissolved oxygen conditions for biodegradation of the carbon layer, and enhance their biodegradation function. The combined process can enhance the removal of pollutants from high algae and high organic matter, and has good application value in the treatment of algae-polluted reservoir water.

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