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Study of removal effect on *Mesocyclops leukarti* with oxidants^{*}

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Abstract: *Cyclops* of zooplankton propagates prolifically in eutrophic waterbody and it cannot be exterminated by conventional disinfection process. The mutagenicity of *Mesocyclops leukarti* and its extermination with oxidants in a drinking waterworks in China were studied. Among five oxidants for use in bench-scale, chlorine dioxide is the most effective and the potassium permanganate is the weakest against *Mesocyclops leukarti* under the same conditions. Full-scale results showed that *Mesocyclops leukarti* could be effectively removed from water by 1.0 mg/L chlorine dioxide preoxidation combined with conventional removal physical process. After filtration, chlorite, a by-product of prechlorine dioxide, is stable at 0.45 mg/L, which is lower than the critical value of the USEPA. GC-MS examination and Ames test further showed that the quantity of organic substance and the mutagenicity in water treated by chlorine dioxide preoxidation are obviously less than those of prechlorination.

Key words:Oxidants, Chlorine, Chlorine dioxide, Mesocyclops leukarti, Preoxidation, Water safetydoi:10.1631/jzus.2006.B0171Document code: ACLC number: TU991

INTRODUCTION

Eutrophication of surface water is a significantly increasing worldwide problem (Ma and Liu, 2002). *Mesocyclops leukarti*, a kind of *Cyclops*, is a waterborne animal propagating prolifically in eutrophic water reservoirs or fresh water lakes in China. Some reservoirs such as Dahuofang Reservoir (Liaoning Province), East Taihu Lake (Jiangsu Province), Gaozhou, Qiyeshi and Xinfengjiang Reservoir (Guangdong Province), Miyun Reservoir (Beijing), etc. are sources of drinking water supply. The omnipresence of *Mesocyclops leukarti* in surface water is a currently growing problem in the production of drinking water.

In drinking water treatment, conventional process is still the main method for *Cyclops* removal. At present, in most waterworks, *Cyclops* is killed or inactivated by prechlorination and removed from water by water treatment process. For example, in Shijiazhuang waterworks located in North China, *Cyclops* is inactivated by prechlorination with 2.5 mg/L, there are still a few active *Cyclops* after filtration (Xing, 2004). *Cyclops* cannot be effectively inactivated by conventional disinfection due to its strong resistance to oxidation (Cui *et al.*, 2002). In addition, the mobility of *Cyclops* makes it easily penetrate sand filter and move into the clean water tank in waterworks, even municipal service pipe. In recent years, there were several contamination accidents with *Cyclops* appearing in municipal service pipe in Shijiazhuang, Tianjin and Binxian cities in China.

Adult *Cyclops* with body length of one to several millimeters that can be seen by naked eye brings diseases to consumers, as it is intermediate host of some diseases. For example, Guinea worm disease still affects a large number of people in rural areas in tropical countries. Olajide and Sridhar (1987) proposed that the disease could be effectively controlled by killing the intermediate host, *Cyclops*. Study of the problem revealed that the present solution is limited to conventional water treatment. Lupi *et al.*(1994) considered that flocculation and sand filtration processes were insufficient for removing zooplankton

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normally present in surface water, and nematodes and water fleas might still occur in treated water. It was reported that the inactivation of zooplankton was necessary to eliminate zooplankton in water treatment process and that rotifer abundance would be reduced from 20~300 ind./L to 0~5 ind./L in drinking water by oxidation with 2.5~3 mg/L of chlorine for 20 min (Bernhardt and Lusse, 1989). Mitcham *et al.*(1983) found that chloramine was more effective against Copepoda (*Cyclops*) but less effective against Cladocera (*Daphnia* and *Bosmina*).

Prechlorination to remove *Cyclops* may resolve the problem of drinking water security by increasing the halomethane content in treated water. It is essential to look for a more effective and safe removal technique. Laboratory experiments on inactivation effects of oxidants such as chlorine, chlorine dioxide, ozone, chloramine and potassium permanganate on *Mesocyclops leukarti* were conducted at the Binxian waterworks in China. After finding the optimum oxidant, full-scale use of chlorine dioxide preoxidation combined with routine clearing process for *Mesocyclops leukarti* removal was implemented. GC-MS examination and Ames test further revealed the kind and amount of organic substance and the mutagenicity in the treated water.

MATERIALS AND METHODS

Raw reservoir water

In Binxian waterworks located in northeast part of China (Heilongjiang Province), an eutrophic reservoir named Erlongshan is used as drinking water source, where the amount of *Cyclops* has greatly increased due to eutrophication. Microscopic observation results showed that *Cyclops* density is 10~24ind./L and that the species of *Cyclops* is *Mesocyclops leukarti*. The Erlongshan Reservoir (126°55'41'' E, 128°19'17'' N) has capacity of 9.4×10^6 m³ and highest water level of 191.5 m and control drainage area of 278.5 km².

At present, *Mesocyclops leukarti* in the waterworks is removed by prechlorination at dosage of 3.0 mg/L, which removes 80% of *Mesocyclops leukarti* from the water after filtration. The water treatment process used in the waterworks is shown in Fig.1.



Fig.1 The chart of drinking water treatment processes in Binxian waterworks

The raw water quality is listed as follows: average water temperature 15~19 °C, turbidity 10~30 NTU, COD_{Mn} 5.68 mg/L, pH 7.3~7.5, algae density $2.1 \times 10^6 \sim 2.6 \times 10^6$ cells/L and *Mesocyclops leukarti* density 20 ind./L.

Experimental equipment

Chlorine dioxide gas generator was manufactured by ourselves. Componential analysis of chlorine dioxide gas is given in Table 1. OF-3 type ozone generator was made by Shanghai Environmental Protection Company. XSB-1A inverted series biological microscope was made by Alltion Wuzhou Company. QP5050A type gas chromatograph-mass spectrometer was made by Shimadzu Instruments Factory. TA100 and TA98 strains without metabolic activation S9, were obtained from Harbin Medical University in China. pH and temperature measurements are conducted using WTW pH/Oxi 340i (made in Germany) with pH and temperature probe (pH-Electrode SenTix 4).

Table 1 Componential analysis of chlorine dioxide gas

Chemical composition	Chlorine dioxide	Chlorine	Chlorite	Chlorate
Percentage composition (%)	82.3	10.4	5.2	2.1

Methods

All analyses were performed according to the standard methods (Chinese EPA, 1997). Bench-scale study on the removal effect on *Mesocyclops leukarti* by five oxidants was in local waterworks lab. Full-scale study of water treatment process was carried out at the Binxian waterworks. *Mesocyclops leukarti* concentration was determined by microscopic count method, and the changes of *Mesocyclops*

leukarti number between raw water and treated water were investigated.

Chlorine dioxide was generated by mixing hydrochloric acid solution (23% weight) and sodium chlorite solution (25% weight) at weight ratio of 1:1. More than 300% stoichiometric acid is to improve the efficiency and lower Cl_2 , ClO_2^- and ClO_3^- production. Iodimetry was used for componential analysis of chlorine dioxide.

Ozone was produced in our laboratory from water cooled ordinary grade air using low frequency ozone generator (OF-3, Xingshen Environmental Facility Co. Ltd., Shanghai, China) operated with a dielectric tube outlet. Ozone output was 3 g/h and generation concentration was 18~20 mg/L. The ozone doses applied to the solutions were subsequently controlled by keeping the rate of gas flow constant and the contact time of ozone was quantified by the iodometry method.

Raw water and treated water by chlorine dioxide or chlorine preoxidation were sampled by 250 L, respectively. Organic substances in water samples were enriched with macroporous resin and then were eluted with about 60 ml of carbinol. The carbinol samples containing organic substances were concentrated to about 1 ml with K-D concentrator. The concentrated samples were examined in duplicate with gas chromatograph-mass spectrometer (GC-MS) and tested by Ames test procedure using TA100 and TA98 strains without metabolic activation S9 at increasing doses of concentration (corresponding to 1, 3, 5 and 7 L of equivalent volume per plate).

RESULTS AND DISCUSSION

Removal effect of five oxidants on *Mesocyclops leukarti*

The effects of five oxidants chlorine, chlorine dioxide, ozone, chloramine and potassium permanganate on *Mesocyclops leukarti* were assessed and compared in laboratory in order to determine the optimum oxidant for Binxian waterworks. The results of experiment are shown in Fig.2. Various dosages of five oxidants were added into a given volume of 10 L reservoir water with *Mesocyclops leukarti* density of 20 ind./L, pH of 7.4, and temperature of 18 °C.

Fig.2 shows that the inactivation effects of five oxidants on *Mesocyclops leukarti* strengthened gradually with increasing dosage and that there were obvious differences between the inactivation efficiency of different oxidants (Fig.2). Chlorine dioxide has the greatest inactivating effect on *Mesocyclops leukarti*, with the efficiency of potassium permanganate being the lowest under the same conditions.

For chlorine in Fig.2a, the inactivity efficiency could reach 80% and 90% after 30 min when adding dosage was 3.00 mg/L and 4.00 mg/L, respectively. Although good removal effect can be obtained if dosage of liquid chlorine increases to 5 mg/L or more, the negative effects resulting from the formation of by-products, like trihalomethanes and haloacetic acids, limit the use of prechlorination. Therefore, the maximal dosage of chlorine in this study was 4 mg/L due to human safety reason.

Chlorine dioxide has been reported to be more effective in inactivating pathogenic organisms including *Cryptosporidium parvum* (Liyanage *et al.*, 1996). In Fig.2d of this study, 100% of *Mesocyclops leukarti* could be inactivated by 1.5 mg/L of chlorine dioxide contact for 30 min. Unlike chlorine, chlorine dioxide cannot react with humic substances to form trihalomethanes (THMs) (Li *et al.*, 1996).

For ozone in Fig.2b, the results showed that the trend consisted of a rapid ozone consumption phase followed by a slower ozone decay phase. This is the same as the typical ozone decomposition trend in natural water (Park *et al.*, 2001). Raw water quality can influence ozonation reactions with the main parameters considered to be natural organic materials (NOM) (Laplanche *et al.*, 1995). One-hundred percent of *Mesocyclops leukarti* could be inactivated by 2.82 mg/L contact for 30 min. For potassium permanganate in Fig.2e, only 60% of inactivation effect was achieved by 2 mg/L contact for 30 min.

The chloramine in Fig.2c can achieve 95% effect of inactivation for 30 min with the chlorine:ammonia ratio=1:1 when the available chlorine is 3 mg/L. Some cities use chlorine for disinfection, and then later use chloramine to provide lasting disinfection as it is sent into pipes (Sung, 2002). Chloramines are weaker disinfectants than chlorine, but are more stable, thus extending disinfectant benefits throughout a water utility's distribution system. Generally chlor-



Fig.2 The inactivation effect of five oxidants on *Mesocyclops leukarti* (a) Cl₂; (b) O₃; (c) Chloramine; (d) ClO₂; (e) KMnO₄

amines are used for maintaining a disinfectant residual in the distribution system and are not used as the primary disinfectant (Kouame and Haas, 1991).

Among five oxidants, the rank of removal efficiency is: $ClO_2>O_3>chloramine>Cl_2>KMnO_4$. Thus, ClO_2 will be selected to further full-scale test.

It is worth noting that the rate of removing *Mesocyclops leukarti* was almost zero under condition of oxidant dosage of 1.0 mg/L and reaction time of 5 min. According to the raw water quality: COD_{Mn} 5.68 mg/L, algae density $2.1 \times 10^6 \sim 2.6 \times 10^6$ cells/L, these substances consumed oxidants, showing that mature *Mesocyclops leukarti* could not be effectively inactivated with lower dose of oxidants and shorter reaction time. So to inactivate or weaken *Mesocyclops leukarti* with adequate oxidants is a key factor. It showed that the NOM such as humic acids and fluvic acids in water react first with oxidants, and then react with *Mesocyclops leukarti*. So at the beginning of the experiment the removal efficiency was almost zero.

Dosage for removal of ClO₂ in full-scale study of *Mesocyclops leukarti*

The oxidant, chlorine dioxide, was added into the grid reaction tank together with coagulant, 15 mg/L of AlCl₃, and chlorine dioxide dosage ranged from 0 to 1.0 mg/L during the full-scale study. The results are shown in Fig.3 showing that the removal effects are strengthened gradually with the increase of ClO₂ dose. One-hundred percent of removal effect on treated water from sand filter can be achieved by 1.0 mg/L of ClO₂. In addition, 10% and 40% of removal rate may be achieved by sedimentation and filtration process under no chlorine dioxide dosing condition, respectively.

The removal of *Mesocyclops leukarti* in water treatment process is the result of oxidizing inactivation by chlorine dioxide in combination with physically clearing process. The removal rate of *Mesocyclops leukarti* is 10% and 40% by sedimentation and filtration with no chlorine dioxide dosing, respectively. It means that *Mesocyclops leukarti* could



Fig.3 Removal effect of different chlorine dioxide dosage on *Mesocyclops leukarti*

be partially removed from water by single physically clearing process.

The vitality of *Mesocyclops leukarti* in raw water is found to be distinct, namely, the activity of larvae and senile *Mesocyclops leukarti* are obviously weaker than that of mature *Mesocyclops leukarti*. Therefore, part of weaker *Mesocyclops leukarti* may deposit together with the floc formed in flocculation process or be trapped by sand filter. However, the removal effect on *Mesocyclops leukarti* by single physically clearing process is finite because *Mesocyclops leukarti* with stronger vitality still might penetrate sand filter so further removal of it requires its inactivation by preoxidation with sufficient oxidant.

The above experimental results showed that *Mesocyclops leukarti* can be inactivated and its vitality greatly weakened with the increase of ClO₂ dosing. Similarly, larvae and senile *Mesocyclops leukarti*, are effectively removed from water by subsequent physical clearing processes. The results showed that the conventional clearing processes, such as sedimentation and filtration, play an important role in removing *Mesocyclops leukarti*, and that the inactivation of *Mesocyclops leukarti* by preoxidation with adequate available oxidant is essential for ensuring drinking water safety. *Mesocyclops leukarti* can be thoroughly removed from water by oxidant preoxidation in combination with physical clearing process, such as sedimentation and filtration.

By-products of ClO₂

Fig.4 shows that the concentration of residual prechlorine dioxide after filtration is constant at 0.15 mg/L and chlorite is 0.45 mg/L. According to rule of

USEPA, the concentration of residual chlorine dioxide and chlorite in drinking water must be controlled to less than 0.8 mg/L and 1.0 mg/L, respectively. Although chlorine dioxide volatile, it can still keep its persistent oxidation ability in water. In local waterworks, the water in the clean tank could keep residual chlorine in the range of standard with small dosage of post-chlorine. According to USEPA rules (USEPA, 1998), if the dosage of chlorine dioxide is under 1.4~2.0 mg/L, then the concentration of chlorite could keep lower than 1.0 mg/L.



Fig.4 The by-products in water after filtration with 1.0 mg/L prechlorine dioxide

Comparison of removal effects of two preoxidation processes

So 100% of *Mesocyclops leukarti* could be removed from treated water after filtration by 1.0 mg/L of ClO₂. At present, in local waterworks, *Mesocyclops leukarti* is removed by prechlorination process with 3 mg/L of chlorine. During the full-scale study, the removal effect on *Mesocyclops leukarti* by chlorine dioxide preoxidation with 1.0 mg/L of ClO₂ was compared with the existing prechlorination process with 3.0 mg/L of chlorine. The results are shown in Fig.5.

Fig.5 shows that the removal effect of chlorine dioxide preoxidation is better than that of prechlorination. For instance, *Mesocyclops leukarti* is thoroughly removed from waterbody by filtration with chlorine dioxide, whereas only 80% of removal effect is achieved with chlorine.

Mature *Mesocyclops leukarti* is hardly removed from water by sedimentation unless it is effectively inactivated by oxidation with oxidant. The removal result of sedimentation tank shows that *Mesocyclops* *leukarti* inactivation by chlorine dioxide is superior to that by chlorine. Fig.5 shows that 90% of removal rate in treated water after sedimentation is achieved by chlorine dioxide preoxidation, but for prechlorination only about 65% is achieved. The distinct difference in *Mesocyclops leukarti* inactivation efficiency of the two preoxidation processes may be the results of the body structure of *Mesocyclops leukarti* and the different performance of oxidant in oxidizing cellular tissue.



Fig.5 Comparison of removal effect on *Mesocyclops leukarti* by two different processes

In contrasted to common bacteria or virus, Mesocyclops leukarti has a special surface structure consisting of seven layers cell tissue such as bottom membrane, epithelium, calcific layer, etc. The body surface provides Mesocyclops leukarti stronger protection against oxidation. Mesocyclops leukarti cannot be effectively inactivated unless the oxidant destroys its surface structure by oxidation or directly penetrates through it into the body to oxidize inner protein to lose enzyme activity and not participate in the activities of the oxidation-reduction system. Therefore, higher oxidizability of oxidant is the key to thoroughly inactivate Mesocyclops leukarti. For chlorine in ClO₂, oxidation number is expressed as +4. According to counting, it contains 263% of available chlorine, i.e. oxidizability of ClO₂ is about 2.5 times as high as that of chlorine (Huang et al., 1997). Therefore, the ability of chlorine dioxide to oxidize Mesocyclops leukarti body structure is more effective than that of chlorine. In addition, after dissolving in water, 100% of ClO₂ exists in molecular state not reacting with water molecular like chlorine, which makes ClO₂ contact Mesocyclops leukarti surface

easily and permeate through it into inner body to destroy inner cellular protein in order to inactivate *Mesocyclops leukarti*. The above advantages of chlorine dioxide not only result in a better inactivation of *Mesocyclops leukarti* but also lower required amount of it than chlorine.

Influence of preoxidation on organic substances and mutagenicity

GC-MS examination and Ames test were carried out with raw water sample and two water samples treated by preoxidation in order to evaluate the safety of the drinking water. The GC-MS examination results are shown in Fig.6 and Table 2. The Ames test results are shown in Table 3.



Fig.6 Graphic representation of total organic ions of (a) raw water, (b) water sample treated by prechlorination and (c) water sample treated by chlorine dioxide preoxidation

	Raw water –		Water treated by preoxidation			
Organic species			Chlorine		Chlorine dioxide	
	Amount	Peak area	Amount	Peak area	Amount	Peak area
Paraffin	22	50052134	19	83418846	14	38552821
Olefin	4	3703129	0	0	1	722801
Halogenated hydrocarbon	2	5619919	13	65706374	3	24916046
Area	2	6571797	3	5218775	3	5853788
Heterocyclic compound	2	7668507	1	1626622	1	775786
Nitrogen compound	11	64235860	4	26415371	1	36663305
Benzene	7	236040538	5	108419603	6	134638566
Alcohol, aldehyde, etc.	21	386596361	31	333345213	12	81050299
Total	71	760488245	76	624150804	41	323173412

Table 2 Analysis of GC-MS results of three water samples

 Table 3 Changes of TA98-S9 and TA100-S9 dose-response relationship

Water sample	Dosago (L/plata)	MR		
	Dosage (L/plate)	TA98-S9	TA100-S9	
Raw water	1	0.67	1.01	
	3	0.66	1.28	
	5	1.15	1.51	
	7	1.75	1.66	
Prechlorination	1	0.87	0.95	
	3	1.13	1.55	
	5	2.54	2.23	
	7	2.81	2.82	
Chlorine dioxide preoxidation	1	0.84	1.33	
	3	1.06	1.44	
	5	1.56	1.77	
	7	2.10	2.07	

GC-MS examination

Compared with the amount of halogenated hydrocarbon in raw water (Table 2), it increases to 13 species in water treated by prechlorination and the total amount of organic substance increases to 5 species. Fig.6 shows that the amount of organic species occurring in retention time between 20 min and 30 min accounts for 44.4% of total amount. Moreover, the spectrum peak intensity is stronger and the area of the peak in the whole scanning area is larger than that of the water sample treated by chlorine dioxide preoxidation.

The amount of organic substance decreases to 41 species by chlorine dioxide preoxidation, which is equal to 57.5% of that in raw water (Fig.6). Species of 85.4% is low boiling point and micromolecule organic substances, which occurs in retention time

before 20 min. In addition, the number of halogenated hydrocarbon is only 3 species in water treated by chlorine dioxide preoxidation.

The total amount of organic substance resulting from chlorine dioxide preoxidation was decreased from 71 species to 41 species in raw water, only about half that from prechlorination, 76 species. Especially, the matters harmful to people's health, halogenated hydrocarbon, were only 3 species in water treated by chlorine dioxide preoxidation compared with 13 species from prechlorination. In addition, most matters produced by chlorine dioxide preoxidation are low boiling point and micromolecule organic substances such as paraffin, alcohol, etc., which account for almost 85.4%. But for prechlorination, about 55% of matters are macromolecule organic compounds such as halogenated hydrocarbon with high boiling point.

The different reaction mechanisms of organic substances reacting with chlorine dioxide and chlorine cause diverse results in GC-MS examination. Free radical oxidation reaction occurs when chlorine dioxide reacts with organic substances. The macromolecule organic compounds are oxidized into oxocompounds and micromolecule substances, in which some volatile compounds, even CO2 are formed (Benjamin et al., 1986). Moreover, the relatively stronger oxidizability of chlorine dioxide may easily cause the oxidation of organic substance to such products as those with low boiling point and minimal molecular weight and short retention time in the graphic result of GS-MS examination. When chlorine oxidizes organic substance, electrophilic substitution reaction results in the formation of more mutagenic halogenated hydrocarbons such as trichloromethane with high boiling point and macromolecule organic compounds with long retention time (Reckhow *et al.*, 1990; Långvik and Holmbom, 1994). In addition, many intermediate products such as aldehyde may be produced during chlorination due to no thoroughly oxidizing organic substances with chlorine. It results in the total amount of organic substance by prechlorination being more than that treated by chlorine dioxide preoxidation. Therefore, it can be concluded that the drinking water quality by chlorine dioxide preoxidation is better than that by prechlorination.

Ames test

The mutagenicity of concentrated water samples before and after preoxidation is expressed by MR value (rate of mutagenicity). The result with MR value greater than 2 is used considered as having mutagenic activity. The results of Ames test are shown in Table 3.

The results of raw water have not behaved mutagenic activity under any dose condition and the MR value of water sample by prechlorination is higher than that by chlorine dioxide preoxidation at the same doses.

TA98 strain is used to detect frame-shift point mutations and TA100 strain is applied to detect base pair substitution mutations. Cl₂-treated water shows mutagenicity with TA98 strain (MR=2.54) and TA100 strain (MR=2.23) at lower doses (5 L equivalent/plate), whereas ClO₂-treated water might have lower mutagenic activity detectable only at the highest doses (7 L equivalent/plate). And the MR value with TA98 and TA100 are 2.1 and 2.07, respectively. Thirteen species of halogentated compounds are formed during prechlorination because of electrophilic substitution reaction between chlorine and organic substances. And their special chemically active group, halogen with relatively strong electrophonic effect, reinforces molecule polarity to react with biologic enzyme system, while the mutagenic action is attained by a series of chemical reactions such as DNA base substitution. During chlorine dioxide preoxidation, less halogentated compounds (only one species) are formed due to different reaction mechanism that is oxidation-reduction reaction, not electrophilic substitution reaction. Hence. mutagenicity of ClO2-treated water was obviously lower than that of Cl₂-treated water. In addition, the

mutagenicity of ClO₂-treated water occurs at the doses of 7 L equivalent/plate, which was different from some previous Ames test results (Fiessinger, 1991; Huang and Li, 1998), probably because of impurities produced by chlorine dioxide gas generator (Table 1). It can be concluded that chlorine accounts for 10.4% of the total chlorine dioxide content, resulting in the mutagenic activity analogous to the result of prechlorination. This can be improved by enhancing chlorine dioxide gas purity by using advanced generator.

CONCLUSION

Among five oxidants used in the bench scale test, chlorine dioxide had the most inactivation effect on *Mesocyclops leukarti* and the efficiency of potassium permanganate was the lowest under the same conditions. One-hundred percent of *Mesocyclops leukarti* could be inactivated by 1.5 mg/L of chlorine dioxide contact for 30 min. Chlorine dioxide preoxidation has very good inactivation effect on *Mesocyclops leukarti*.

In full-scale study, 100% of removal effect in treated water from sand filter can be achieved by using 1.0 mg/L of ClO₂. *Mesocyclops leukarti* can be effectively removed from water by chlorine dioxide preoxidation combined with conventional physical processes such as filtration or sedimentation. The existing process of prechlorination cannot achieve the same removal effect as chlorine dioxide.

Chlorite, the by-product of ClO_2 , is constant at 0.45 mg/L after filtration, which is lower than the critical value of USEPA.

GC-MS examination and Ames test further showed that the sort and amount of organic substances in treated water after chlorine dioxide preoxidation were evidently less than those of prechlorination, and that the mutagenicity of drinking water was greatly reduced. The advantage of chlorine dioxide disinfection may be further improved by enhancing chlorine dioxide gas purity (>95%).

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