



Manganese removal from the Qiantang River source water by pre-oxidation: A case study

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Abstract: We evaluated several different pre-oxidation treatments, namely the introduction of either potassium permanganate (KMnO₄), chlorine (Cl₂), or both to remove manganese (Mn) from the Qiantang River source water. Our results showed that Mn removal percentages were 12.7%, 71.0%, 17.4% and 58.7% when none of the oxidants, KMnO₄ only, Cl₂ only, or both oxidants were added, respectively. Furthermore, a field study showed that when the available Mn concentration in the source water was 0.14 mg/L, it could be reduced to less than 0.05 mg/L when a solution of KMnO₄ (0.47 mg/L) was added as the oxidant.

Key words: Pre-oxidation, Potassium permanganate (KMnO₂), Chlorine (Cl₂), Manganese (Mn) removal
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INTRODUCTION

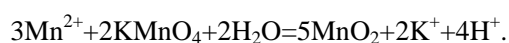
One of the many problems of using ground water is the need to remove manganese (Mn). Mn is a common element in ground water. It occurs naturally in rocks and soil and is a normal constituent of the human diet. Mn may become noticeable in tap water at concentrations greater than 0.05 mg/L by imparting a color, odor, or taste to the water. However, health effects from Mn are not a concern until concentrations are about 10 times higher. Exposure to high concentrations of Mn over the course of years has been associated with toxicity to the nervous system, producing a syndrome that resembles Parkinsonism. This type of effect may be more likely to occur in the elderly (Forstner and Wittmann, 1979; Nriagu, 1988; Kannan, 1995; Kang *et al.*, 2003).

Most source water used by the Hangzhou Water Group Company Ltd. originates from the Qiantang River. According to a recent investigation, the concentration of Mn in the river was often above 0.1 mg/L, which is the acceptable value of II in GB 3838-2002 Environmental Quality Standard for

Surface Water. Some literature showed that the removal of Mn by traditional treatment approaches (such as filtration, ion exchange, or aeration followed by filtration) is not ideal (Cherry, 1962; Choo *et al.*, 2005; Luan *et al.*, 2006). It is hard to reduce the Mn in the effluent water to the 0.1 mg/L level of the CJ/T206-2005 Water Quality Standards for Urban Water Supply or the 0.05 mg/L level of the Zhejiang Modern Water Plant Standard. Currently, the methods to remove Mn include: (1) contact oxidation—through aeration and Mn ore (Luan *et al.*, 2006); (2) biological methods—through bacteria in a biological filter (Tekerlekopoulou and Vayenas, 2007); (3) chemical Cl₂ oxidation—using strong oxidants, such as Cl₂, ozone, chlorine dioxide, and KMnO₄ to oxidize Mn²⁺ to manganese dioxide (MnO₂) that is then removed through sedimentation or filtration (Luan *et al.*, 2006; Roccaro *et al.*, 2007); (4) stabilization—adding chelant into the water to form MnO₂ chelate to prevent the decomposition of manganese dioxide (Wang *et al.*, 2004). Contact oxidation and biological methods require aeration equipment or reconstruction of the filter, and need a long time to

activate the filter media or grow enough bacteria, so they are not ideal in terms of either cost or time. Stabilization, by its very nature, cannot remove manganese. The effectiveness of abiotic homogenous Mn oxidation by oxygen is very slow at pH values below 9 (Stumm and Morgan, 1996). Thus, at pH values (6~8) usually encountered in most drinking waters, Mn will not be oxidized efficiently. As a result, Mn cannot be removed by simple aeration and precipitation. It is for these reasons that chemical oxidation is generally required to achieve precipitation of Mn in a reasonable time and at pH values common to water utility practice, i.e., between 6 and 8. It has been well established that KMnO_4 is an effective oxidant for dissolved Mn over a broad range of pH values (Knocke *et al.*, 1991; Mouchet, 1992; Katsoyiannis and Zouboulis, 2004). Thus, we decided to adopt the method of adding KMnO_4 to remove the Mn from source water.

Chemical oxidation using KMnO_4 has been widely used for treatment of pollutants in drinking water and wastewater applications for over 50 years. KMnO_4 has been used as an algacide and disinfectant to remove odour, iron and Mn (Humphrey, 1961; Cherry, 1962; Tang *et al.*, 2003). In recent years, great progress has been made in using KMnO_4 to remove organic matter, control by-products from chloridization, and help coagulation (Yuan B.L. *et al.*, 2005; Yuan D.Y. *et al.*, 2005; Tian *et al.*, 2006; Zhao *et al.*, 2006; Hu *et al.*, 2007; Li *et al.*, 2007; Yang *et al.*, 2007). In neutral pH solution, Mn^{2+} can be quickly oxidized to Mn^{4+} and the oxidation reaction can be written as follows:



Other reductants in the water can also consume some of the KMnO_4 . Chemical oxygen demand on Mn (COD_{Mn}) can be used to measure the content of the reductants.

In this study, we have compared and evaluated the Mn removal efficiencies using either KMnO_4 , Cl_2 , or both, and investigated the turbidity removal efficiencies under similar conditions.

MATERIAL AND METHODS

The water used in this experiment was the

Qingtai Water Plant's source water which was pumped or diverted directly from the Qiantang River. The water's quality is shown in Table 1. We carried out parallel experiments with four different treatments, namely, with no oxidants added, only KMnO_4 added, only Cl_2 added and both KMnO_4 , and Cl_2 added.

Table 1 Source water for experiments

Parameter	Value	Parameter	Value
Temperature ($^{\circ}\text{C}$)	12.5	Manganese (mg/L)	0.138
Ammonia (mg/L)	0.74	pH	7.36
Turbidity (NTU)	5.37		

A polyaluminum chloride (PAC) coagulant was added to 1000 ml of source water in a beaker. KMnO_4 , Cl_2 , or both were then also added at different concentrations. The solution was first quickly mixed at 150 r/min, then slowly stirred at 60 r/min, and finally set aside for 45 min. The supernatants were then taken for analyses, such as turbidity, pH, ammonia, manganese, total chlorine, free chlorine. The settled water was then filtered through a filter paper and the Mn concentration was measured.

In this study, the amount of Mn was measured by an atomic absorption method, the contents of ammonia and Cl_2 were determined by spectrophotometric methods, and turbidity was measured using a Hach 2100-N scattering turbidimeter (USA).

EXPERIMENTAL RESULTS AND ANALYSES

No oxidant

The dosage of coagulant added and the results of the experiments are shown in Table 2 and Fig.1. When no oxidant was added, the turbidity removal ratio could reach up to 68.7% with a coagulant dosage of 30 mg/L, but the removal efficiency of Mn was only 3.6%. Even after filtration, a removal efficiency of only 4.5% was achieved for Mn. The highest removal ratio was only 12.3% after coagulation and filtration. The results indicate that such a traditional treatment has little effect on Mn removal.

KMnO_4 as the only oxidant

Usually the turbidity after sedimentation in our water plant is below 4 NTU and as shown in Table 2, if the coagulant dosage is over 15 mg/L, the turbidity

can be controlled under 4 NTU. So a coagulant dosage of 15 mg/L was chosen in the current test. The results of the test are shown in Table 3 and Fig.2. In this experiment, the dosage of KMnO_4 was added at multiples of the original Mn concentration present in the source water. When the Mn in the source water was 0.138 mg/L and 0.414~0.552 mg/L KMnO_4 was added to the solution, which had 3~4 times of the original Mn concentration in the source water, the Mn removal ratio achieved the maximum 71% at the

coagulant (PAC) dosage of 15 mg/L. The Mn concentration in filtrated water could be reduced to under 0.05 mg/L, and the removal efficiency was much higher than the 12.3% achieved with no KMnO_4 added. When the dosage of KMnO_4 in the solution

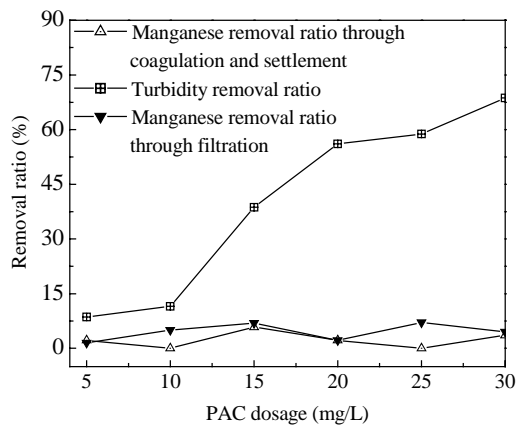


Fig.1 Removal of manganese and turbidity with no oxidant added

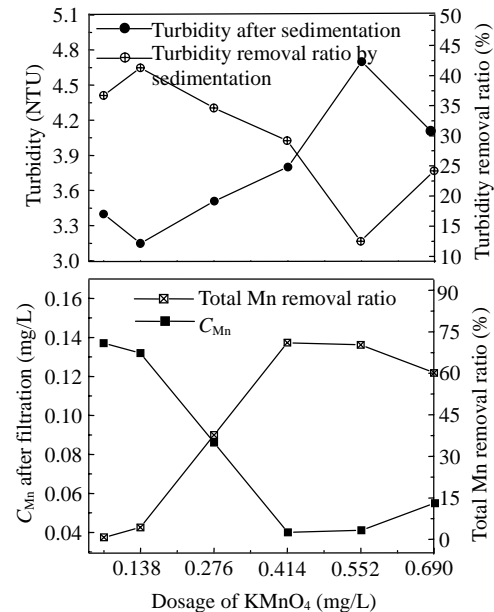


Fig.2 Test results—only KMnO_4 added

Table 2 Removal of manganese and turbidity with no oxidant added

Dosage of coagulant (mg/L)	pH	Tur1 (NTU)	R_{Tur1} (%)	C_{Mn1} (mg/L)	R_{Mn1} (%)	C_{Mn2} (mg/L)	R_{Mn2} (%)
5	7.32	4.91	8.6	0.135	2.2	0.133	1.5
10	7.22	4.75	11.5	0.140	0	0.133	5.0
15	7.17	3.29	38.7	0.130	5.8	0.121	6.9
20	7.15	2.36	56.1	0.135	2.2	0.132	2.2
25	7.13	2.21	58.8	0.141	0	0.131	7.1
30	7.10	1.68	68.7	0.133	3.6	0.127	4.5

Tur1: turbidity after sedimentation; R_{Tur1} : turbidity removal ratio by sedimentation; C_{Mn1} : Mn concentration after settlement; R_{Mn1} : Mn removal ratio through coagulation and sedimentation; C_{Mn2} : Mn concentration after filtration; R_{Mn2} : Mn removal ratio through filtration

Table 3 Test results—only KMnO_4 added

Dosage of KMnO_4 (mg/L)	pH	Tur1 (NTU)	R_{Tur1} (%)	Tur2 (NTU)	R_{Tur2} (%)	C_{Mn1} (mg/L)	R_{Mn1} (%)	C_{Mn2} (mg/L)	R_{Mn2} (%)	R_{Mn} (%)
0.069	7.22	3.40	36.7	0.23	93.2	0.145	-5.1	0.137	5.5	0.7
0.138	7.20	3.15	41.3	0.37	88.3	0.153	-10.9	0.132	13.7	4.3
0.276	7.16	3.51	34.6	0.42	88.0	0.178	-29.0	0.086	51.7	37.7
0.414	7.16	3.80	29.2	0.18	95.3	0.231	-67.4	0.040	82.7	71.0
0.552	7.19	4.70	12.5	0.28	94.0	0.281	-103.6	0.041	85.4	70.3
0.690	7.18	4.07	24.2	0.27	93.4	0.295	-113.8	0.055	81.4	60.1

Tur1: turbidity after sedimentation; R_{Tur1} : turbidity removal ratio by sedimentation; Tur2: turbidity after filtration; R_{Tur2} : turbidity removal ratio by filtration; C_{Mn1} : Mn concentration after settlement; R_{Mn1} : Mn removal ratio through coagulation and sedimentation; C_{Mn2} : Mn concentration after filtration; R_{Mn2} : Mn removal ratio through filtration; R_{Mn} : total Mn removal ratio

increased from 0.069 to 0.414 mg/L, the total Mn removal ratio increased from 0.7% to 71%. Further increasing the KMnO_4 dosage resulted in a decrease in the total Mn removal ratio, which is in accordance with the report by Roccaro *et al.* (2007) who found that a half stoichiometric dose (1.74 mg per 0.5 mg Mn) of an oxidizing agent (KMnO_4) provided better removal results than a stoichiometric dose.

When KMnO_4 was added, the removal ratio of turbidity was not greatly affected if the KMnO_4 dosage was less than the original Mn concentration in the source water. But the removal ratio of turbidity by sedimentation decreased when the dosage of KMnO_4 was increased.

Cl_2 as the only oxidant

There was ammonia in the source water, so when Cl_2 was added, it would react with ammonia first. If the Cl_2 dosage was not high enough, there would be no free Cl_2 in the water. Tests on Mn removal with only Cl_2 added were carried out with the coagulant (PAC) dosage set at 15 mg/L.

Table 4 and Fig.3 showed that the free Cl_2 and total Cl_2 content increased when the dosage of Cl_2 increased. However, the total Mn removal ratio did

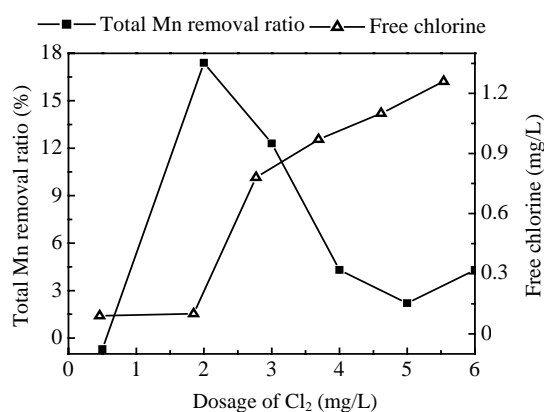


Fig.3 Test results—only Cl_2 added

not change proportionally. The highest removal ratio of Mn was 17.4% when the dosage of Cl_2 was 1 mg/L. Choo *et al.* (2005) also found that the Mn removal efficiency was not as effective as that for iron when only Cl_2 was added to the source water. Other constituents in source water were oxidized preferentially at the low Cl_2 dosage, and a relatively high Cl_2 dosage of about 2 mg/L was required to achieve a desirable Mn removal efficiency of greater than 40% (corresponding to the current Korean maximal contamination level of 0.3 mg/L). Though this figure is much higher than that found in our water system, it can still be seen that the Mn removal efficiency with only Cl_2 added was much lower than that with only KMnO_4 added (71% at a KMnO_4 dosage of 0.414~0.552 mg/L). Also, we found that the turbidity after sedimentation did not differ greatly whether or not Cl_2 was added.

Both KMnO_4 and Cl_2 as oxidants

To know whether adding Cl_2 would influence the efficiency of using KMnO_4 to remove Mn, we added both KMnO_4 and Cl_2 to the source water. Table 5 and Fig.4 show the results. We chose the dosage of KMnO_4 of 0.276 mg/L, twice the Mn concentration in the source water, because at this dosage the resulting Mn concentration after filtration can be controlled below 0.1 mg/L, a requirement of the CJ/T206-2005 Water Quality Standards for Urban Water Supply. The dosage of coagulant used was 15 mg/L.

As shown in Fig.4 and Table 5, the Mn removal efficiency decreased with the addition of Cl_2 compared with that when only KMnO_4 was used as the oxidant. For example, when only KMnO_4 was added at a dosage of 0.276 mg/L, the removal ratio of Mn was 37.7%, which decreased to 33.3% when Cl_2 was also added at a dosage of 2 mg/L. So, adding Cl_2 to the source water did not favor Mn removal with KMnO_4 .

To determine the influence of different KMnO_4

Table 4 Tests results—only Cl_2 added

Dosage of Cl_2 (mg/L)	pH	Ammonia (mg/L)	Tur1 (NTU)	C_{Mn1} (mg/L)	C_{Mn2} (mg/L)	R_{Mn} (%)	Free Cl_2 (mg/L)	Total Cl_2 (mg/L)
0.5	7.26	0.77	3.64	0.130	0.139	-0.7	0.09	0.19
1.0	7.12	0.73	3.20	0.126	0.114	17.4	0.10	0.52
2.0	7.08	0.66	3.28	0.137	0.121	12.3	0.78	1.03
3.0	7.01	0.58	3.43	0.140	0.132	4.3	0.97	1.79
4.0	7.00	0.52	3.31	0.140	0.135	2.2	1.10	>2.2
5.0	6.93	0.45	3.43	0.136	0.132	4.3	1.26	>2.2

Tur1: turbidity after sedimentation; C_{Mn1} : Mn after settlement; C_{Mn2} : Mn after filtration; R_{Mn} : total Mn removal ratio

dosages on Mn removal at a fixed Cl_2 dosage of 2 mg/L, we carried out the next test. The result can be seen in Fig.5.

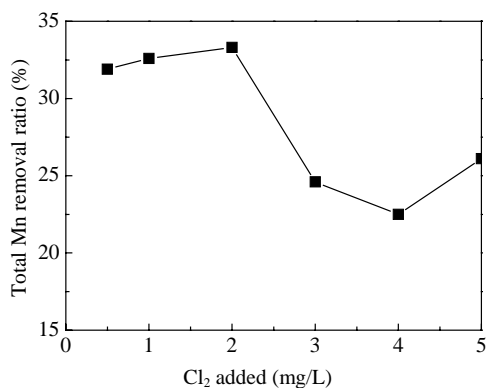


Fig.4 Influence of Cl_2 on KMnO_4 removing Mn

Table 5 Influence of Cl_2 on KMnO_4 removing Mn

Dosage of Cl_2 (mg/L)	pH	C_{Mn1} (mg/L)	C_{Mn2} (mg/L)	R_{Mn} (%)	Total Cl_2 (mg/L)
0.5	7.25	0.180	0.094	31.9	0.32
1.0	7.24	0.162	0.093	32.6	0.68
2.0	7.14	0.186	0.092	33.3	1.30
3.0	7.09	0.173	0.104	24.6	2.03
4.0	7.01	0.182	0.107	22.5	>2.2
5.0	6.97	0.180	0.102	26.1	>2.2

C_{Mn1} : Mn after settlement; C_{Mn2} : Mn after filtration; R_{Mn} : total Mn removal ratio

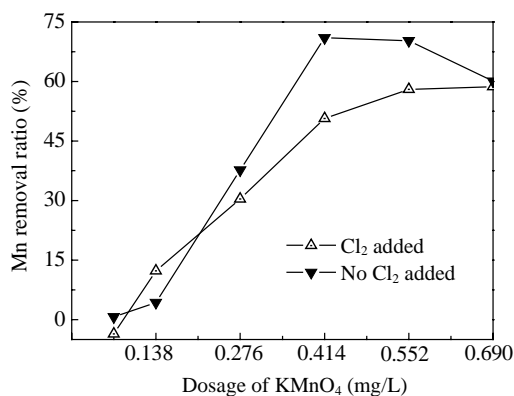


Fig.5 Influence of different dosages of KMnO_4 on Mn removal at a fixed Cl_2 dosage of 2 mg/L

It can be seen that after adding 2 mg/L Cl_2 to the source water, the Mn removal efficiency increased with the increasing dosage of KMnO_4 , achieving a maximum of 58.7% when the dosage of KMnO_4 was 0.552~0.690 mg/L, 4~5 times that of the original Mn concentration in the source water, lower than the 71% Mn removal efficiency with only KMnO_4 added at a

dosage of 0.414~0.552 mg/L, 3~4 times the original Mn concentration in the source water. So, if Cl_2 was added when using KMnO_4 to remove Mn from source water, it would not only demand more KMnO_4 but also decrease the efficiency of KMnO_4 in removing Mn.

Fig.6 shows the Mn removal of the four different treatments. Tests showed that the Mn removal efficiency was enhanced when oxidants were added. The turbidity removal ratio was also related to the oxidants added and their dosages. Comparing adding both KMnO_4 and Cl_2 with adding KMnO_4 alone, we found that the former decreased the Mn removal efficiency. The maximum Mn removal efficiency was 12.3%, 71%, 17.4% and 58.7% when no oxidants, KMnO_4 only, Cl_2 only, and both KMnO_4 and Cl_2 were added, respectively. When the original Mn concentration was 0.138 mg/L in the source water, the resulting Mn concentration in filtered water decreased to less than 0.05 mg/L when only KMnO_4 was added at a dosage of 0.414~0.552 mg/L, 3~4 times the original Mn concentration in the source water.

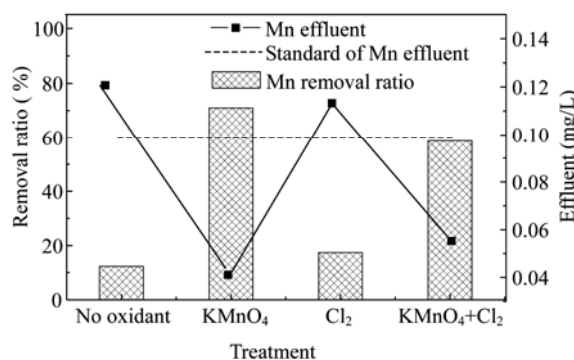
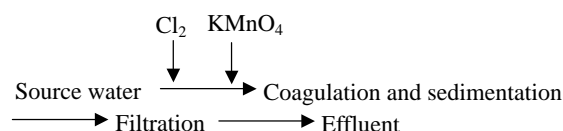


Fig.6 Mn removal under different treatments

FIELD STUDY OF USING KMnO_4 AND Cl_2 TO REMOVE Mn AND ORGANISMS

Since November, 2006 the Mn concentration in the source water of the Qiantang River has become higher and higher. In light of the limitations of traditional treatment methods in removing Mn, water plants in Hangzhou City that use the Qiantang River as the water source all added KMnO_4 to the water to enhance the removal efficiency of Mn. We took the Qingtai Water Plant as an example to carry out our field study. The treatment process of the Qingtai

Water Plant is coagulation→sedimentation→filtration. Cl₂ was chosen as the disinfectant, which was added before coagulation. Due to the high concentration of ammonia present in the source water, the Cl₂ added existed mostly as chloramines. KMnO₄ was added after pre-chlorination and before coagulation. The process flow was as follows:



Influence of different KMnO₄ dosages on contaminant removal efficiency

The relationship between the KMnO₄ dosage and contaminant removal efficiency is shown in Table 6 and Fig.7.

Table 6 Contaminant removal ratio at different KMnO₄ dosages

KMnO ₄ dosage (mg/L)	C _{Mn,inlet} (mg/L)	C _{Mn,outlet} (mg/L)	R _{Mn} (%)	C _{COD_{Mn},inlet} (mg/L)	C _{COD_{Mn},outlet} (mg/L)	R _{COD_{Mn}} (%)
0	0.12	0.08	33.0	3.45	2.17	35.6
0.30	0.12	0.07	45.0	3.41	2.29	44.0
0.40	0.17	0.10	41.0	3.50	2.73	21.0
0.47	0.14	<0.05	64.3	3.81	2.39	20.8
0.57	0.16	0.06	62.5	3.65	2.32	18.9

C_{Mn,inlet}: inlet Mn concentration, mg/L; C_{Mn,outlet}: outlet Mn concentration, mg/L; R_{Mn}: Mn removal ratio, %; C_{COD_{Mn},inlet}: inlet COD_{Mn} concentration, mg/L; C_{COD_{Mn},outlet}: outlet COD_{Mn} concentration, mg/L; R_{COD_{Mn}}: COD_{Mn} removal ratio, %

At almost the same concentrations of Mn and organisms, the Mn removal ratio increased from 33% to 45% and the organism removal ratio increased from 35.6% to 44% when KMnO₄ was added at a dosage of 0.3 mg/L. In the same Mn source water, the removal ratio of Mn increased when the dosage of KMnO₄ increased. When the Mn in the source water was 0.14 mg/L, a dosage of KMnO₄ of 0.47 mg/L, which was 3.4 times the original Mn concentration in the source water, could limit the Mn concentration in the effluent to under 0.05 mg/L. This was consistent with the jar test controlling the dosage of KMnO₄ to be under 0.05 mg/L.

If the Mn concentration in source water is high, a low KMnO₄ dosage will lower the organism removal efficiency of the system. The KMnO₄ reacts not only

with Mn but also with organisms, so at the same Mn concentration in the source water, the dosage of KMnO₄ should be increased if the level of organism contamination in the source water is high.

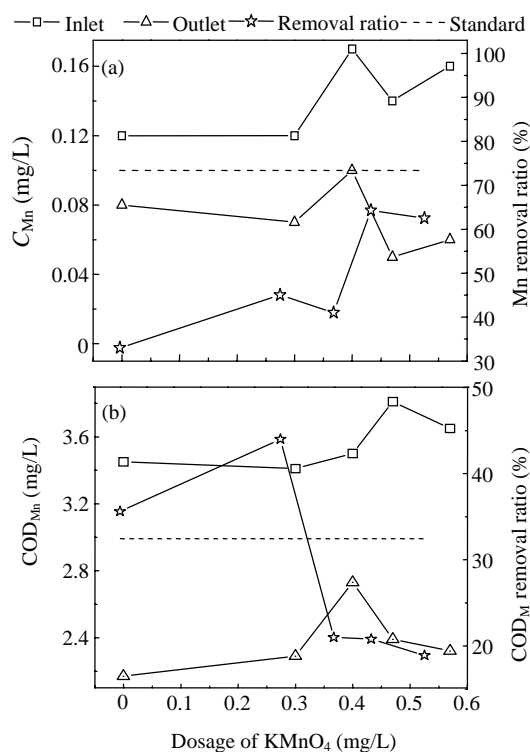


Fig.7 Removal ratios of Mn and COD_{Mn} and effluent concentration after different treatments

Influence of a fixed KMnO₄ dosage on the Mn removal ratio under different contaminant concentrations

At the same KMnO₄ dosage of 0.3 mg/L, the Mn and COD_{Mn} removal ratios are shown in Table 7. The Mn removal ratio increased from 36.4% to 44% when the Mn concentration increased from 0.11 to 0.18 mg/L in the source water, whereas the organism removal ratio decreased correspondingly from 34.7% to 17.2%.

CONCLUSION

(1) From the jar tests, it can be seen that the highest Mn removal ratios were 12.3%, 71.0%, 17.4% and 58.7% when none of the oxidants, only KMnO₄, only Cl₂, or both KMnO₄ and Cl₂ were added, respectively. When the Mn concentration was

Table 7 Removal ratio of Mn and COD_{Mn} at the same KMnO₄ dosage of 0.3 mg/L

No.	C _{Mn,inlet} (mg/L)	C _{Mn,outlet} (mg/L)	R _{Mn} (%)	C _{COD_{Mn},inlet} (mg/L)	C _{COD_{Mn},outlet} (mg/L)	R _{COD_{Mn}} (mg/L)
1	0.11	0.07	36.4	3.52	2.21	34.7
2	0.16	0.1	37.5	3.47	2.89	24.6
3	0.18	0.1	44.0	3.52	2.57	17.2

C_{Mn,inlet}: inlet Mn concentration, mg/L; C_{Mn,outlet}: outlet Mn concentration, mg/L; R_{Mn}: Mn removal ratio, %; C_{COD_{Mn},inlet}: inlet COD_{Mn} concentration, mg/L; C_{COD_{Mn},outlet}: outlet COD_{Mn} concentration, mg/L; R_{COD_{Mn}}: COD_{Mn} removal ratio, %

0.138 mg/L in the source water, the Mn in filtered water decreased to less than 0.05 mg/L with only KMnO₄ added at a dosage of 0.414~0.552 mg/L, 3~4 times the original Mn concentration in the source water. Cl₂ is not as effective as KMnO₄ in removing Mn. The Cl₂ requirement has been found to be in excess of the stoichiometric requirement. This is likely to be because of Cl₂ demand by organic compounds. The additional introduction of Cl₂ decreased the overall removal efficiency of KMnO₄ in removing Mn.

(2) Our field study demonstrated that when the Mn concentration in source water was 0.14 mg/L, a KMnO₄ dosage of 0.47 mg/L, which was 3.4 times the original Mn concentration in the source water, could control the Mn concentration in the effluent to be under 0.05 mg/L. This was similar to the result from the jar test in which the dosage of KMnO₄ was controlled to be 0.414~0.552 mg/L, 3~4 times the original Mn concentration in the source water. The Mn removal ratio increased with the increasing Mn concentration in the source water, whereas the organism removal ratio decreased. While KMnO₄ dosing is very effective, if the source water Mn level fluctuates significantly, adjusting the permanganate dosing according to Mn levels may be operationally difficult. Nonetheless, slight overdosing of permanganate (up to 0.1 mg/L) has been found not to cause any adverse effects.

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