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Assessment of the manganese content of the drinking water source in Yancheng, China

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ABSTRACT

Excessive intake of manganese can damage the nervous system of the human body. In August 2009, the manganese content of the drinking water source in Yancheng exceeded the national standard of drinking water source, which influenced the daily life of the local residents. The aim of this study was to investigate the factors leading to the manganese content of river water in Yancheng exceeding the national standard. To the data, the manganese content of surface water in Yancheng already met the national standard of drinking water source in September 2009, but the manganese content of river sediment was relatively high, especially in Mangshe River and Tongyu River. It was worthwhile to note that the soluble manganese content of the sediment in Mangshe River was even as high as 270 mg kg^{-1}, which suggested that the release of manganese from the sediment was the major cause of the pollution. The manganese content of the soil near the rivers was also determined, and the results indicated that the wastewater and waste slag discharged by the stainless steel factories nearby were the main pollution sources of manganese. Furthermore, the environmental factors affecting the release of manganese from the sediment were also investigated.

In recent 20 years, the fast economic development in China has brought serious environmental problems [1–3]. With continuously expanding scales of chemical, printing and dyeing industries, the wastewater discharged by these high-polluting industries become a major pollution source of the environment [4–6]. Because of the unbalanced economic development in different areas of China, the high-polluting industries have gradually been shifted from developed areas toward under-developed areas [7–9]. Jiangsu is one of the major provinces in China famous for its high economic development. However restricted by its historical and social factors as well as natural conditions, the north area of Jiangsu is backward in economic development. Thus, the high-polluting industries are shifted from the south area to the north of Jiangsu. With a population of around 8.1 million and about 150,000 ha of land, Yancheng is the largest city in the north area of Jiangsu. In August 2009, the manganese content of the drinking water source in Yancheng exceeded the National Standard for Drinking Water Quality (GB57492006), which influenced the life of the local residents. Many studies show that excessive intake of manganese can cause nervous-system damage, leading to Parkinson's disease, and also injure the artery breastwork and myocardium [10–12].

The goal of this study was to investigate what caused the manganese content of the river water in Yancheng inconstantly exceeding the national standard. To achieve this goal, we investigated three aspects as follows: (1) the manganese content of the drinking water source in Yancheng (Mangshe River, Tongyu River, Taidong River, and Chuanchang River), (2) the manganese content of the soil around the manganese-involved factories in Yangcheng, (3) the impact of the environmental factors on the release of manganese from the sediment. Based on the systematical investigation, we identified the factors that increased the concentration of manganese of the rivers in Yancheng.

1. Materials and methods

1.1. Sampling and preparation

All samples were collected in September 2009 according to the following methods:

(1) Twenty-two water samples (1000 mL) were collected from Mangshe River, Tongyu River, Chuanchang River, and Taidong River by using a sampler.
(2) Twenty-two sediment samples (1.0 kg) were collected from intertidal zone surface sediments (0–5 cm deep) from all four rivers by using a grab sampler in September 2009 (Fig. 1). In order to prevent the loss of soluble manganese during the stor-

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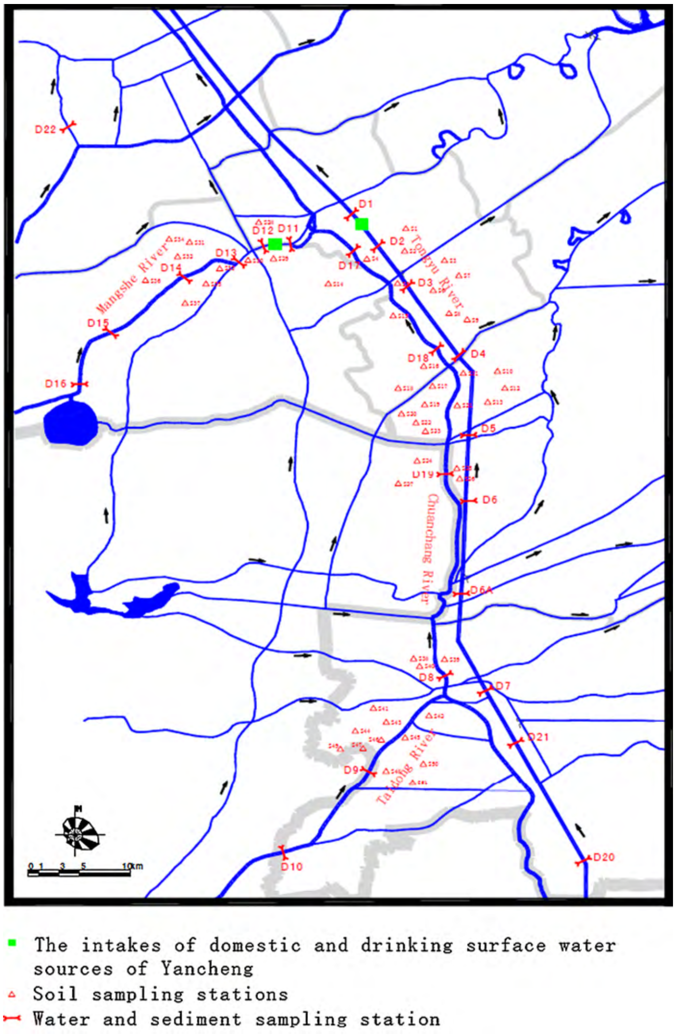

Fig. 1. Sampling stations in the area of Yancheng (September 28, 2009).

age and transportation, the sediment samples were carefully put into clean plastic vessels and stored at −20 °C prior to analysis. In the laboratory, sediment samples were thawed at room temperature, and air-dried.

(3) Nine sediment samples were collected from the sediments (0–50 cm deep) in those four rivers by using a sediment core sampler. In order to prevent the loss of soluble manganese during the storage and transportation, the sampled sediment cores were carefully put into clean plastic boxes and stored at −20 °C prior to analysis. In the laboratory, sediment samples were thawed at room temperature, and air-dried.

(4) Following the method as previously described by Chen et al. [13], 47 soil samples were collected from the 0 to 5 cm deep layer of the soil around the stainless steel factories in September 2009. Soil samples were carefully put into clean plastic vessels and stored at 4 °C prior to analysis. In the laboratory, soil samples were air-dried.

1.2. Quantification of manganese

1.2.1. Determination of the manganese contents of water samples

Each water sample was filtered with a cellulose membrane (0.45 μm). Then the manganese contents of filtered water samples were determined by using a flam atomic absorption spectrometer (FAAS, GBC Sens AA Dual, Australia).

1.2.2. Determination of total manganese contents of soil and sediment samples

Sediment or soil samples were digested in open PTFE® vessels (Jingcheng, Shanghai, China). One gram of each sediment or soil sample was digested with a mixed solution consisting of 8 mL of 65% HNO_3, 4 mL of 40% HF, 2 mL of 37% HCl, and 10 mL of H_2O. The digestion mixture was heated by a hot plate at 96 °C; after it boiled dry, its final volume was adjusted to 50 mL by adding water. Its manganese content was determined by the FAAS method following the procedure outlined by Lu et al. [2].

1.2.3. Determination of soluble manganese in soil and sediment samples

The sequential extraction of soluble manganese from soil or sediment samples was conducted according to the BCR method as previously described by Jan et al. [14]. Determination of soluble manganese of a sample was detailed below.

Step 1—Determination of exchangeable manganese in the extractable fraction (*F*(I)): Each soil or sediment sample (0.5 g) was mixed with 8 mL of 1 M magnesium chloride ($MgCl_2$) at pH 7.0. The mixture was continuously agitated at 120 r min^{-1} for 1 h at room temperature for extracting exchangeable manganese from the soil or sediment. The manganese content of the resulting extract was determined by using a FAAS.

Step 2—Determination of carbonated manganese in the non-extractable fraction (*F*(II)): The residue which had been extracted from Step 1 was mixed with 8 mL of 1 M sodium acetate (NaOAc; adjusted to pH 5.0 with acetic acid, HOAc). The mixture was incubated at room temperature for 5 h for leaching out manganese from carbonates. The manganese content of the leachate was determined by using a FAAS.

The soluble manganese content of a soil or sediment sample was calculated according to Eq. (1):

$$\mathrm{SW} = \mathrm{WF(I)} + \mathrm{WF(II)} \tag{1}$$

where SW is the content of soluble manganese of a sample (mg kg^{-1}), WF(I) is the content of exchangeable manganese of the sample (mg kg^{-1}), and WF(II) is the content of carbonated manganese of the sample (mg kg^{-1}).

1.3. The impact of the environmental factors on the release of manganese from the sediment and soil

1.3.1. The impact of humic acid on the release of manganese from the sediment

In summer, great quantities of biological materials, such as stems, branches and leaves, were washed into the rivers in Yancheng by rain. These materials could be degraded into humic acid [15]. The relationship between the concentration of humic acid and the release of manganese from sediment was analyzed according to the following method. Five conical flasks (250 mL) were used; 0.200 g of a dried sediment sample (from 0 to 10 cm deep layer of sediment) was added into each flask. Then 100 mL humic acid (Sigma Co., Inc., USA) solutions with different concentration were added into those flasks, respectively. The flasks were completely sealed and shaken in a G-25 model incubator shaker (New Brunswick Scientific Co., Inc., USA) at 120 r min^{-1}. The batch equilibrium test was continuously run over 72 h to ensure the release equilibrium of manganese being reached. Finally, the mixture in a flask was filtered with a cellulose membrane (0.45 μm), and the manganese content of the filtrate (C_e: mg L^{-1}) was determined by the FAAS. R_e (mg kg^{-1}), the release of manganese, was calculated

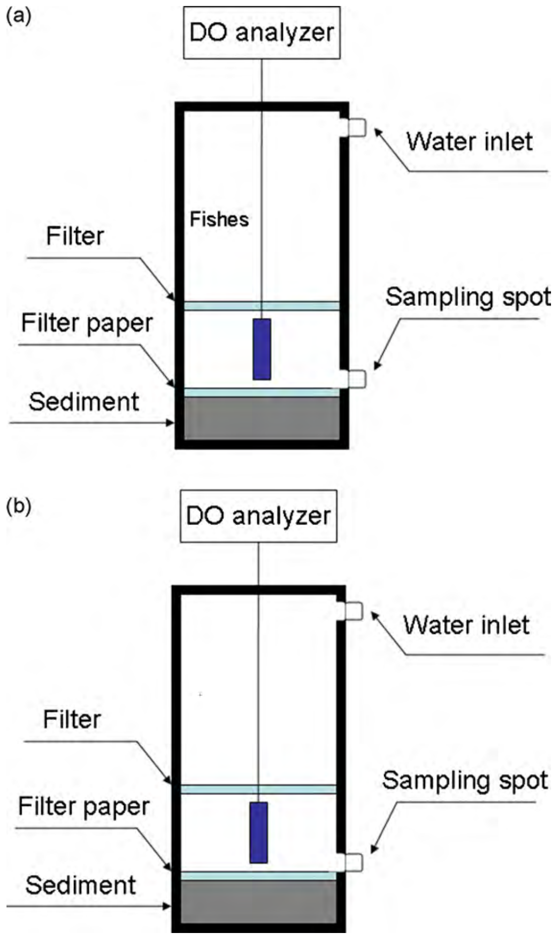

Fig. 2. (a) Container A and (b) Container B.

according to Eq. (2):

$$R_e = \frac{V \times C_e}{W} \tag{2}$$

where V is the volume of solution (L), and W is the weight of dry adsorbent (kg).

1.3.2. The impact of dissolved oxygen (DO) on the release of manganese from the sediment

Since Tongyu River is the largest drinking water source in Yancheng, the sediments in this river were analyzed. Containers A and B (Fig. 2)—each of them was 30.00 ± 0.50 cm long, 20.00 ± 0.50 cm high, and 100.0 ± 5.1 cm wide—were utilized in the following two experiments, respectively.

(1) Fifty liters of deionized water (DW) and 1000 g of a sediment sample (from 0 to 10 cm deep layer of sediment) were added into Container A maintained at 20 °C. There was fish culture in part of the rivers. In order to simulate this condition, one fish was kept in Container A. Every two days 20 mL of water was sampled from the bottom layer of the water in the container by a pipette, and then an equal volume of DW was added into the container. The manganese contents of water samples were determined according to method described in Section 1.2.1.
(2) Fifty liters of a humic acid solution at a concentration of 20 mg L^{-1} and 1000 g of a sediment sample (from 0 to 10 cm deep layer of sediment) were added into Container B maintained at 20 °C. The container was kept sealed. A 20 mL solution sample was collected from the bottom layer of the solution in the container daily and then an equal amount of the humic acid solution was added into the container immediately. The manganese contents of the solution samples were determined according to the method described in Section 1.2.1.

2. Results and discussion

2.1. The quality of the surface water in Yancheng

In August 2009, the concentration of manganese of the rivers was higher than 0.15 mg L^{-1}. But in September, as shown in Table 1, the manganese contents of the waters in Tongyu River, Mangshe River, Chuanchang River, and Taidong River water were lower than 0.1 mg L^{-1}, which indicated that the quality of the surface water in Yancheng already met the national quality standard of drinking water source.

2.2. The manganese content of the river sediment

Though the manganese content of the river water reached the national standard of drinking water source in September 2009, those rivers might still be contaminated. As shown in Table 2, the average manganese contents of the sediments in Tongyu River, Mangshe River, Chuanchang River, and Taidong River ranged 600–1050 mg kg^{-1}, 750–1550 mg kg^{-1}, 600–850 mg kg^{-1}, and 650–700 mg kg^{-1}, respectively. The background manganese content of the soil in China ranges from 180 mg L^{-1} to 1156 mg L^{-1} [16]. Thus, the manganese content of the sediment in Tongyu River reached the highest value of the national background value, and that in Mangshe River was 50% higher than the national average. It is worthwhile to note that Tongyu River and Mangshe River are main drinking water sources in Yancheng. It is well known that the chemical form of manganese is one of the factors determining the release of manganese from sediment [17–19]. As shown in Table 2, soluble manganese contents of the sediments in the four rivers were higher than 270 mg kg^{-1}, and that in Mangshe River even attained to 420 mg L^{-1}. Such a high content of soluble manganese of the sediment was very likely to cause an increase in manganese content of river water.

The above analyses evidently indicated that the release of manganese from the sediment was one of the important factors leading to the manganese content of the drinking water source in Yancheng exceeding the national standard. Usually, two factors may result in a high manganese content of river sediment. The one is that the sediment has a high background manganese content; the other is that the river has been polluted by manganese-involved factories. The manganese contents of the different layers of the sediment (0–50 cm in depth) were determined. For the sediments in Tongyu River and Mangshe River, the manganese contents of their surface layers were higher than those of the bottom (Fig. 3a and b), which indicated that these two rivers were polluted by manganese recently. In contrast, for the sediment in Chuanchang River, the manganese content of its surface layer was lower than that of the bottom (Fig. 3c), which indicated that the river had been polluted by manganese a long time ago. For the sediment in Taidong River, its manganese content peaked in the 5–10 cm deep layer and decreased with increasing depth of layers (Fig. 3d). This result indicated that the background manganese content of the sediment in Taidong River was low, but the river was constantly polluted by manganese.

2.3. The manganese content of the soil near the stainless steel factories around the rivers

To find out the pollution source, soil samples were collected from the soil near the stainless steel factories around the rivers,

Table 1
Water quality data of major rivers in Yancheng (September 28, 2009).

Sample number	River name	Monitoring station	Surface layer water (mg L^{-1})			Bottom layer water (mg L^{-1})			Average value (mg L^{-1})		
			Nitrogen	COD_{Mn}	Manganese	Nitrogen	COD_{Mn}	Manganese	Nitrogen	COD_{Mn}	Manganese
D1	TongYu River[a]	East of the city waterworks	0.72	4.5	0.085	0.78	4.5	0.079	0.75	4.5	0.082
D2		Yanqu Bridge	0.65	4.6	0.089	0.62	4.6	0.061	0.64	4.6	0.075
D3		Qinyuan Ferry	0.66	4.7	0.066	0.58	4.6	0.068	0.62	4.7	0.067
D4		Doulong Hill	0.53	4.3	0.087	0.61	3.8	0.072	0.57	4.1	0.080
D5		Qizhao Sluice	0.90	3.6	0.051	0.87	3.5	0.050	0.89	3.6	0.051
D6		Baiju Zhen	0.82	3.4	0.043	0.82	3.3	0.059	0.82	3.4	0.051
D6-A		Caoyan Bridge	0.83	3.5	0.065	0.79	3.7	0.066	0.81	3.6	0.066
D7		South of the fertilizer plant	0.80	3.8	0.033	0.77	3.8	0.055	0.79	3.8	0.044
D20		Liangduo Zhen Bridge	1.02	4.4	0.030	0.98	4.4	0.032	1.00	4.4	0.031
D21		Guben bridge	1.25	4.2	0.045	1.38	4.0	0.044	1.32	4.1	0.045
D8	Taidong River	Baiju Zhen	0.67	3.0	0.019	0.70	2.5	0.019	0.69	2.8	0.019
D9		South of Guangshan Zhen	0.64	3.1	0.020	0.69	3.0	0.018	0.67	3.1	0.019
D10		Taidong Bridge	0.72	3.0	0.018	0.77	3.3	0.014	0.75	3.2	0.016
D11	Mangshe River[a]	Tongyu North Bridge road	0.57	4.4	0.023	0.57	4.2	0.054	0.57	4.3	0.039
D12		Fenggang Bridge	0.56	4.4	0.030	0.62	4.5	0.034	0.59	4.5	0.032
D13		Jingkou Bridge	0.59	4.5	0.033	0.53	4.5	0.057	0.56	4.5	0.045
D14		Mangshe Yanqu Bridge	0.66	4.4	0.045	0.64	4.5	0.054	0.65	4.5	0.050
D15		The 331 Bridge	0.46	4.2	0.017	0.49	4.2	0.025	0.48	4.2	0.021
D16		Dazong Lake Exit	0.53	4.0	0.010	0.53	4.0	0.006	0.53	4.0	0.008
D17	Chuanchang River	Huangtu Ditch	0.50	4.8	0.049	0.49	4.7	0.041	0.50	4.8	0.045
D18		South of Kaifa Bridge	0.60	4.8	0.074	0.52	4.7	0.078	0.56	4.8	0.076
D19		North of Doulong Hill	0.66	4.0	0.070	0.72	3.8	0.083	0.69	3.9	0.077

[a] The main drinking water source of Yancheng.

and their manganese contents were determined. As shown in Fig. 4a, nearly 34% of soil samples had extremely high manganese contents, and the manganese contents of 17% of the samples were about twofold of the national standard. Furthermore, soluble manganese contents of nearly 90% of the samples were higher than 100 mg kg^{-1} (Fig. 4b). It is known that soluble manganese in soil can be easily leached out by rain, seeping into groundwater or rivers to pollute the water or sediment. We found stainless steel factories near the rivers, and some of the factories did not treat the wastewater and residue effectively. Therefore, we infer that the acidic pickling wastewater and waste slag discharged by the stainless steel factories were the main pollution sources of this water contamination accident.

2.4. *The impact of humic acid on the release of manganese from the sediment*

Since the manganese content of the river water seasonally fluctuated, it was necessary to investigate the effect of the environmental factors on the release of manganese from the sediment. As shown in Fig. 5, increasing the concentration of humic acid of the water led to more manganese released from the sediment [20,21]. This result suggests that the release of manganese from the sediment correlates with the concentration of humic acid of water. It can be explained that natural organic acids adsorb manganese and form complex in water, which can increase the release of manganese from sediment. Since Mangshe River and Tongyu River are

Table 2
The manganese content in the major rivers sediment (September 28, 2009).

Sample	River name	Sampling station	Total manganese contents (mg kg^{-1})	Soluble manganese contents (mg kg^{-1})
D1	TongYu River[a]	East of the city waterworks	966	327
D2		Yanqu Bridge	912	314
D3		Qinyuan Ferry	1002	375
D4		Doulong Hill	878	286
D5		Qizhao Sluice	859	322
D6		Baiju Zhen	740	273
D6-A		Caoyan Bridge	865	295
D7		South of the fertilizer plant	700	301
D20		Liangduo Zhen Bridge	561	225
D21		Guben bridge	576	238
D8	Taidong River	Baiju Zhen	585	254
D9		South of Guangshan Zhen	571	285
D10		Taidong Bridge	649	198
D11	Mangshe River[a]	Tongyu North Bridge road	967	448
D12		Fenggang Bridge	771	301
D13		Jingkou Bridge	1510	625
D14		Mangshe Yanqu Bridge	903	430
D15		The 331 Bridge	1095	408
D16		Dazong Lake Exit	715	315
D17	Chuanchang River	Huangtu Ditch	832	327
D18		South of Kaifa Bridge	723	211
D19		North of Doulong Hill	796	175

[a] The main drinking water source of Yancheng.

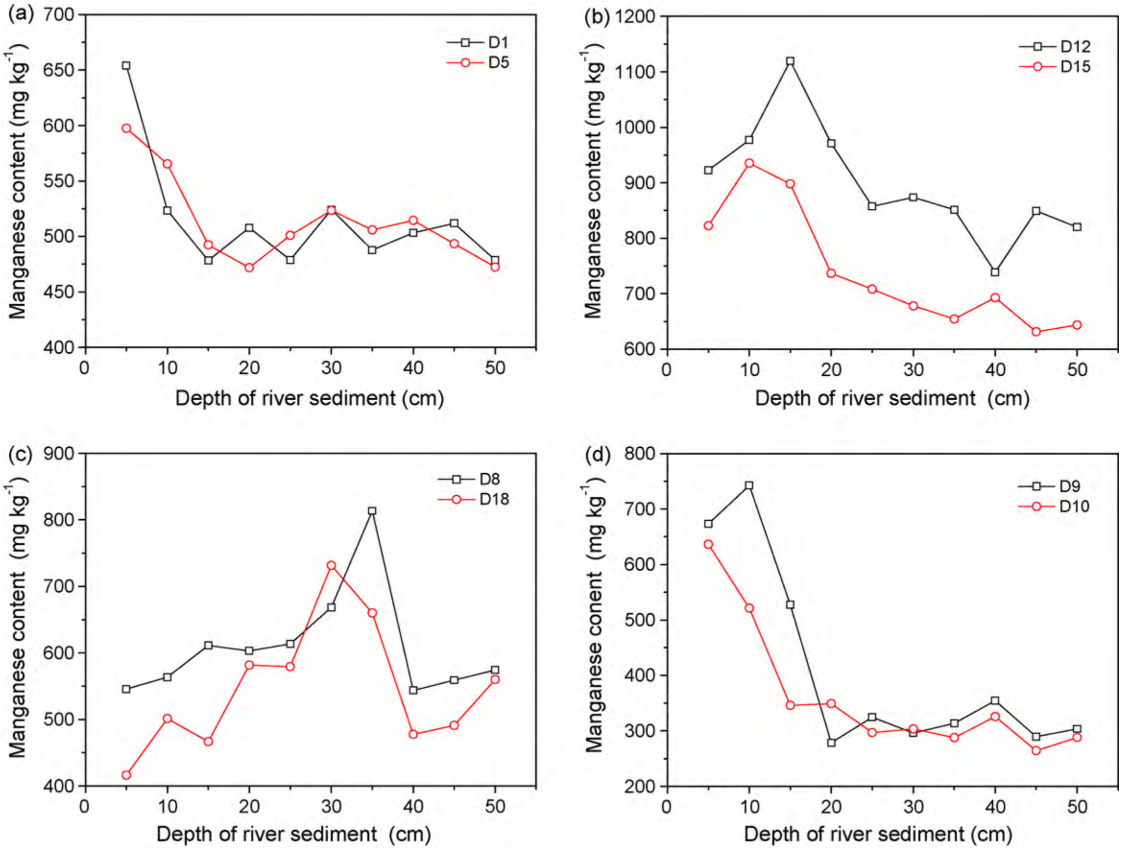

Fig. 3. The manganese content of the river sediment: (a) Tongyu River, (b) Mangshe River; (c) Chuanchang River; and (d) Taidong River.

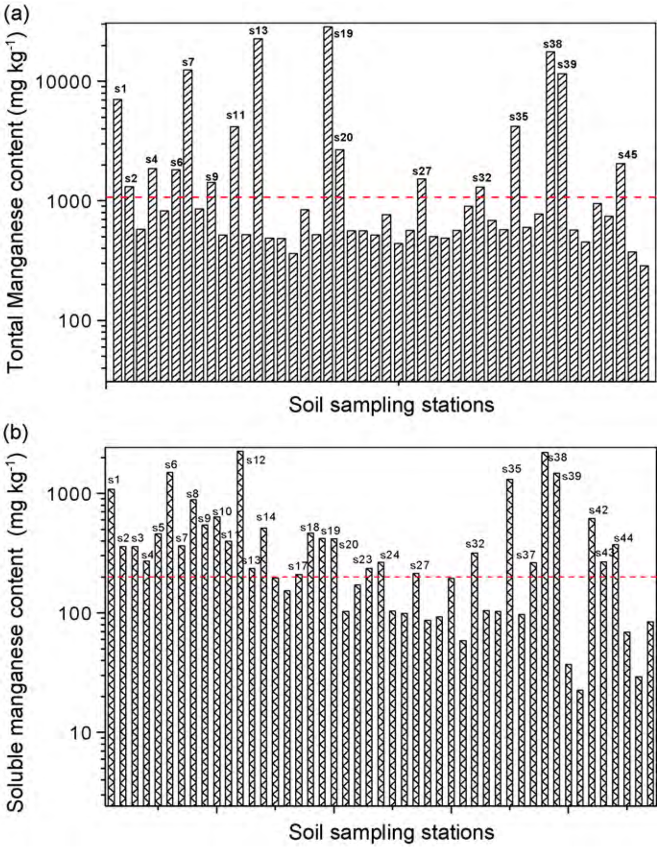

Fig. 4. The manganese content of the soil: (a) total manganese content and (b) soluble manganese content.

the major drinking water sources of Yancheng, higher amounts of manganese released from the sediments in these rivers (Fig. 5) had a significant impact on the drinking water quality in Yancheng. In summer, amounts of straw were washed into the river by the rain, and the straw could be degraded into humic or phytic substance. As a result, the concentration of seasonal organic matter in rivers, such as humic substance, was comparatively high in summer. Therefore, we infer that a seasonal increase in dissolved organic matter (DOM) was another factor causing an over-high manganese content of the surface water in Yancheng. This can be explained by the chelating between the DOM and manganese [22].

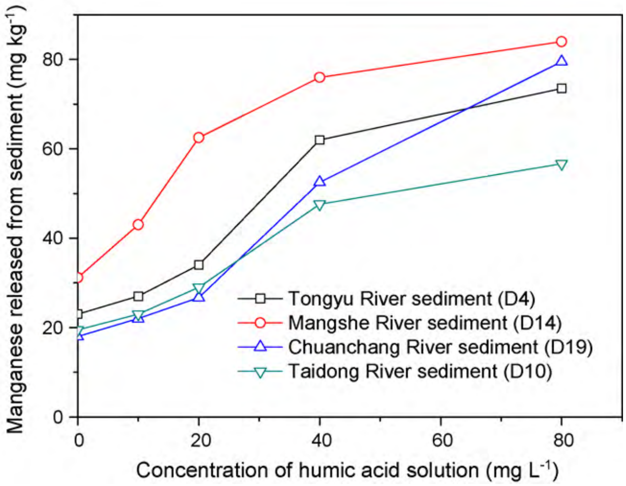

Fig. 5. The impact of humic acid on the release of manganese from the river sediment.

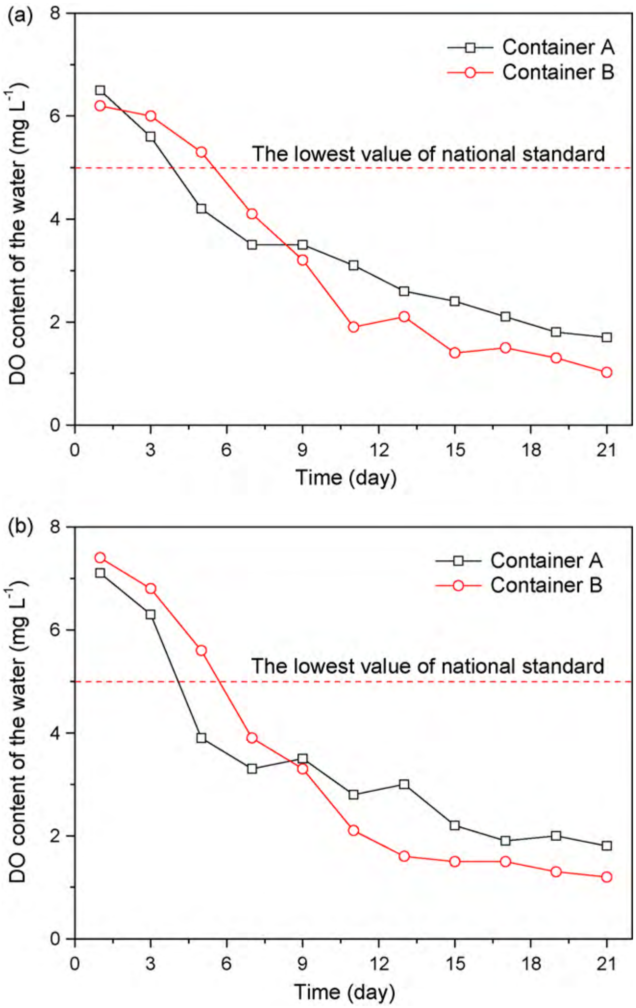

Fig. 6. DO content of the water over time: (a) the sediment sampled from Mangshe River and (b) the sediment sampled from Tongyu River.

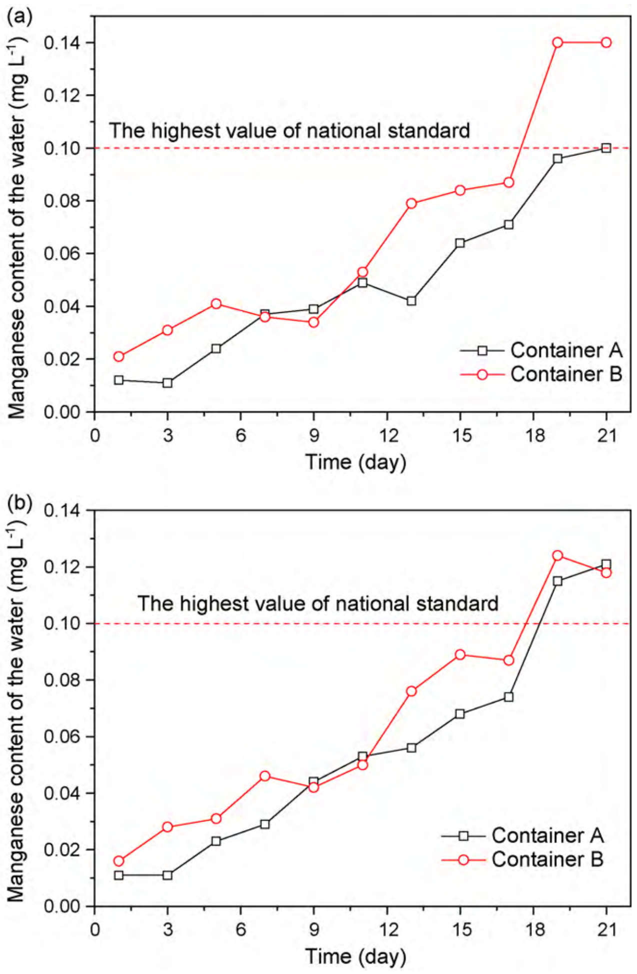

Fig. 7. The manganese content of the water over time: (a) the sediment sampled from Mangshe River and (b) the sediment sampled from Tongyu River.

2.5. The impact of dissolved oxygen (DO) on the release of manganese from the sediment

As shown in Fig. 6(a and b), the DO content of the water decreased over time. In contrast, the manganese content of the water obviously increased over time, and it even exceeded 0.1 mg L^{-1} on the 21st day of the experiment (Fig. 7(a and b)). These results indicated that the manganese content of the water rose as its DO content declined. This might be due to the mechanism that in the absence of oxygen, part of the oxide manganese in the sediment is reduced into soluble manganese, which is released into the water [23]. In summer, great quantities of stems, branches, and leaves were washed into rivers by rain; and these biological materials were then degraded into the DOM. The reactions of degrading the DOM consumed the DO; consequently, the DO content of the river water declined. According to the monitored data (Tables 3 and 4), the DO contents of the waters in Mangshe River and Tongyu River were low in July 2009, which was beneficial to reducing oxide manganese into soluble manganese. As a result, more soluble manganese was released from the sediment into the river water, leading to an increase in its manganese content in August 2009. Therefore, compared to other seasons, the river water had a higher manganese content in summer because it had a lower DO content at that time.

Table 3
The average DO content (mg L^{-1}) in the water of Tongyu River from January 2009 to September 2009[a].

Monitoring time	The average DO content in the water of different monitoring stations of Tongyu River (mg L^{-1})											
	Fuanliang Bridge	Guben Bridge	The south of fertilizer factory	Beihai Bridge	Tongyu Ferry	Caoyan Bridge	Tanxiang Zhen	Chenbei Bridge	Chengdong waterworks	Fuyang Bridge	Tongyu Bridge	Hongyu Zhen
January	7.3	6.5	7.9	8.2	8.1	10.8	9.0	12.9	11.9	11.7	11.8	9.2
March	6.7	6.8	6.0	6.5	6.4	7.8	9.5	10.4	9.2	9.5	9.2	8.5
May	5.2	3.0	5.2	5.0	5.1	5.3	5.1	7.2	5.6	6.9	7.5	9.9
July	4.1	3.1	4.3	5.1	5.9	3.8	1.63	5.1	2.3	5.1	5.2	5.7
September	4.5	3.7	5.4	5.8	5.2	5.7	5.1	8.4	5.1	8.3	7.2	6.1

[a] With the help of Jiangsu Province environment monitoring stations.

Table 4
The average DO content ($mg\,L^{-1}$) in the water of Mangshe River from January 2009 to September 2009[a].

Monitoring time	The average DO content in the water of different monitoring stations of Mangshe River ($mg\,L^{-1}$)					
	Dazong Lake	Fenggang Bridge	Mangnan Yao Ferry	Mangshe Yanqu Bridge	Jingkou Bridge	Tongyu North Bridge road
	10.1	8.8	10.7	9.4	10.3	8.9
January	9.8	9.9	9.8	9.7	10.1	9.4
March	5.1	9.4	5.0	7.2	8.6	6.3
May	4.5	6.2	2.9	4.1	5.2	3.0
July	5.6	5.7	5.2	4.4	4.9	5.9

[a] With the help of Jiangsu Province environment monitoring stations.

3. Conclusions

By systemically investigating the manganese content of the drinking water source in Yancheng, we draw the following conclusions:

(1) In September 2009, the manganese contents of the waters in Tongyu River, Mangshe River, Chuanchang River, and Taidong River were all below 0.1 $mg\,L^{-1}$, meeting the national standard of drink water source.

(2) The manganese content of the sediment in Tongyu River reached the highest value of the national background value, and that in Mangshe River was even 50% higher than the upper limit of the national standard. The high manganese content of the river sediment resulted from the wastewater and waste slag discharged by the stainless steel factories nearby. Therefore, the release of manganese from the polluted sediment and soil around the factories caused an over-high manganese content of the surface water in August 2009.

(3) The environmental factors were also responsible for the seasonal increase in the manganese content of the river water. In summer, a low DO concentration and a high DOM concentration in the river water resulted in more soluble manganese released from the polluted sediment into the river water.

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