Operational performance of biological treatment plant for iron and manganese removal

Dong Li, Jie Zhang, Hongtao Wang, Hong Yang and Baozhen Wang

ABSTRACT

The treatment plant was designed according to the theory of biological fixation and removal of manganese from water. The process included low level aeration followed by single stage filtration. The separated native iron and manganese oxidizing bacteria after multiplication were inoculated to the filtering medium alongside raw water. After 2–3 months of culturing, the filter exhibited strong manganese removal capability. Under the usual filtration rate conditions, the manganese concentration in the raw water was 0.575–3.05 mg l^{-1} and total iron (T-Fe) 0.01–0.5 mg l^{-1}; the manganese concentration in the filtrate was 0.05 mg l^{-1} and T-Fe in trace, which was superior to the Chinese National Standard for drinking water (Fe<0.3 mg l^{-1}, Mn<0.1 mg l^{-1}). The product water was stable and of high quality that completely meets the above-mentioned standard. The process can reduce investment costs by 50 million yuans (¥) and operation and maintenance cost by ¥12,000 per day compared with traditional processes.

Key words | biological fixation and removal of manganese, cost-effective process, low level aeration, simultaneous removal

Dong Li (corresponding author)
Hongtao Wang
Department of Environmental Science and
 Engineering,
Tsinghua University, Beijing 100084,
China
E-mail: *lidongzi76@hotmail.com*
Lidongzi@emails.bjut.edu.cn

Hong Yang
College of Architectural and Civil Engineering,
Beijing University of Technology,
100 Ping LeYuan,
Chaoyang, Beijing 100022,
China

Jie Zhang
Baozhen Wang
Water Pollution Control Research Center,
Harbin Institute of Technology,
66 West Dazhi Street,
Harbin 150006,
China

INTRODUCTION

Almost all current treatment processes are based on the theory of chemical contact oxidation for groundwater containing Fe^{2+} and Mn^{2+} (Trace Inorganic Substances Committee 1987; Li and Liu 1989). The removal of Fe^{2+} is efficient but the removal of Mn^{2+} is not satisfactory when the Fe^{2+} and Mn^{2+} containing groundwater is treated by some traditional processes (Pierre 1992). The product water is satisfactory when strong oxidants were used, but the cost of the oxidant is very high (Wang 1984; Knocke 1988; Ellis 2000). Some bacteria that oxidize iron and manganese have been reported in France and Egypt (Pierre 1992; Zakaria 2001). Up to now, about 18 genera listed in *Bergey's Manual* are found to have such activity including *Leptothrix*, *Crenothrix*, *Pedomicrobium*, *Gallionella*, *Metallogenium*, *Caulococcus*, *Kusnezovia*, *Thiobacillus ferrooxidans*, *Siderocapsa*, *Naumanniella*, *Ochrobium* and *Siderococcus*. Known as iron and manganese oxidizing bacteria, they are all aerobes; some can oxidize iron and manganese, while others oxidize iron or manganese (Buchanan & Gibons 1974). Applying the bacteria to remove iron and manganese from groundwater and identifying the perfect treatment process are the problems still to be solved.

Studies have been conducted on the application of iron and manganese oxidizing bacteria in filters in Austria, Australia and the UK (Bourgine *et al.* 1994; Talizawa *et al.* 2001; Mettler & Abdelmoula 2001). Iron oxidation was proved to be a function of the physics, chemistry and biology of the biological trickling filters (Dimitrakos 1997). Some results showed that iron and manganese oxidizing bacteria were in competition with nitrification bacteria for the dissolved oxygen (DO). The nitrification bacteria consume a great deal of DO which restricted the propagation of the iron and manganese oxidizing bacteria (Gouzinis & Kosmidis 1998; Vandenabeele 1995). Biological filters for iron and manganese removal have been adopted in plants in Finland. There, it was found that iron oxidizing bacteria cannot survive together with manganese oxidizing

bacteria because of their different living conditions. So iron and manganese removal can only be realized in two separate filters (Seppanen 1992). In Japan, iron and manganese can be removed by biological slow filters with a very low filtration rate of 10 m per day (Tamura *et al.* 1999; Kojima *et al.* 2000). Conventional processes including two-stage aeration and two-stage filtration are still applied in engineering with high operation and maintenance costs.

Our earlier studies have shown that a biological process is also available for the removal of dissolved iron and manganese with the production of filtrate that is superior to that produced by other treatment processes (Pierre 1992; Zhang & Dai 1996). The removal of iron and manganese can be realized by one-stage weak aeration and one-stage filtration with a filtration rate of 8 m h^{-1} (Li *et al.* 2001). A full-scale biological treatment plant for iron and manganese removal was designed according to the theory of biological fixation and removal of manganese. As the oxidation reductive potential (ORP) of Mn^{2+} is high at pH around 7, the dissolved Mn^{2+} cannot be removed through aeration/oxidation unless the microbial community dominated by iron and manganese-oxidizing bacteria proliferate and reach a balance (Zhang *et al.* 1996). In the catalysis by the extracellular enzymes of manganese oxidizing bacteria, Mn^{2+} can be oxidized to insoluble Mn (IV), hydrated manganese oxide $MnO(OH)_2$. After sedimentation and adhesion to the media surface this is removed (Zhang *et al.* 1997). Although chemical oxidation of Fe^{2+} to insoluble $Fe(OH)_3$ is much easier than that of Mn^{2+} when exposed to air, autocatalytic oxidation of Fe^{2+} takes part in the metabolism of manganese oxidizing bacteria. Therefore, Mn^{2+} and Fe^{2+} can be simultaneously removed in one rapid filter. The colloid granules formed by ferric oxide before being introduced to the filter can be removed (Li *et al.* 2001). In the mature filter there is an abundant microbial community consisting of iron and manganese oxidizing bacteria and others, whose balance and stability is the key for effective oxidation of Mn^{2+}.

The operational performance of the water treatment plant (WTP) for iron and manganese removal is presented in this paper in detail.

Table 1 | Characteristics of the raw water

Parameter	Mean value	Parameter	Mean value
Temperature (°C)	9.00	Oxygen consumption (mg l^{-1})	0.56
pH	6.8	Chromaticity	< 5
Fe^{2+} (mg l^{-1})	0.15	Mn^{2+} (mg l^{-1})	1.5
Scent	ND	Coliform (CFU ml^{-1})	5

Groundwater characteristics

The groundwater is pumped from wells (depth 100 m, located in the southwest of Shenyang, Liaoning, China); each well has a flux of 3,000 m^3 day^{-1} and contains Fe^{2+} concentrations of 0.01–0.5 mg l^{-1} and manganese 0.575–3.05 mg l^{-1} at a temperature of 9°C. The concentration of iron and manganese fluctuates sharply in raw water. The groundwater characteristics are shown in Table 1. According to surveys carried out in different countries, iron and manganese often exist together in groundwater and the concentration of iron is often higher than that of manganese; this is in contrast to the findings here.

The design of the plant

The total treatment capacity of this WTP is 12×10^4 m^3 day^{-1} including filters, aeration tanks, clean water reservoir, backwash tower, pump house, iron sludge dehydrate room and transformer substation, office building, furnace. Silica sand was adopted as the filter media. The plan of this WTP is shown in Figure 1 and a flow chart shown in Figure 2. It is important to note that there is an overfall weir in the clean water reservoir which separates the backwash water and the chlorinated water as shown in Figure 2. The main treatment units and their parameters are shown in Table 2.

The characteristics of the process

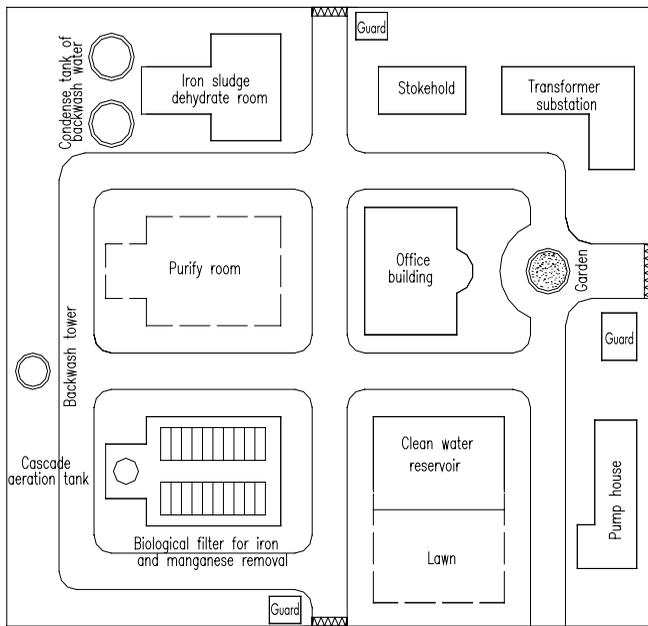The quantity of DO available to oxide Fe^{2+} and Mn^{2+} is finite in a biological filter. Low level aeration is adopted

which is realized by a cascaded aeration tank with a depth of 0.84 m. High level aeration is indispensable to develop pH and remove CO_2 in traditional processes, which needs a great deal of energy. CO_2 is the nutrient source for iron and manganese oxidizing bacteria such as *T. ferrooxidans* and *Gallionella*, so CO_2 removal and development of pH in a biological filter for iron and manganese removal is not necessary. According to chemistry reaction equations and aeration tests, it is enough to control DO at 3–4 mg l^{-1} for the biological process. The tiny colloid particles formed by Fe^{3+} easily penetrate the filter so the raw water is introduced to the filter as rapidly as possible after aeration in order to avoid the oxidation of Fe^{2+} prior to the filter.

One-stage weak aeration and one-stage filtration were applied instead of two-stage strong aeration and two-stage
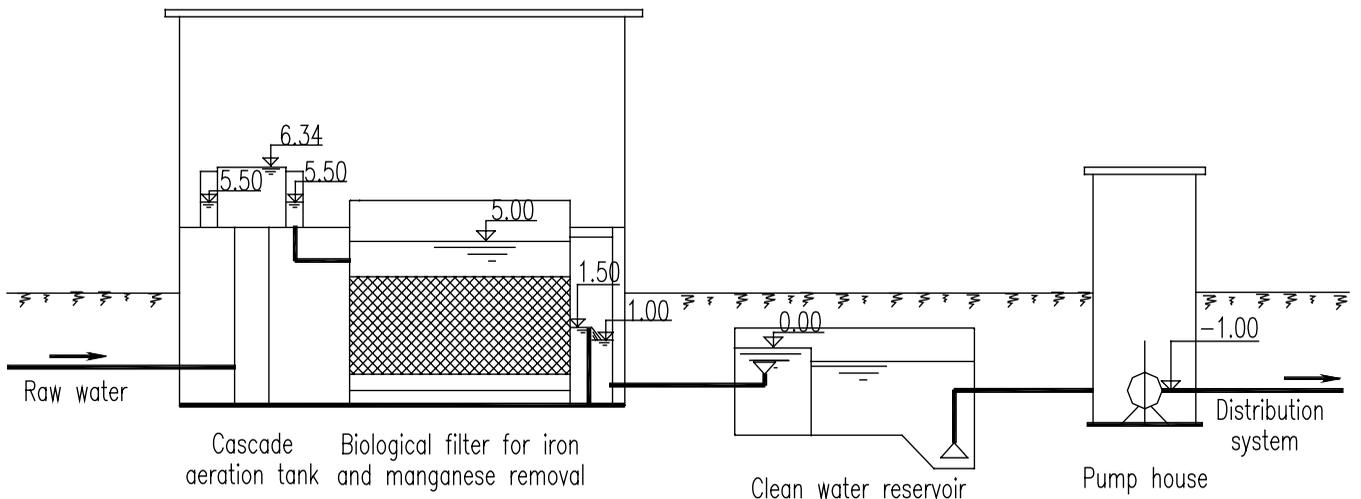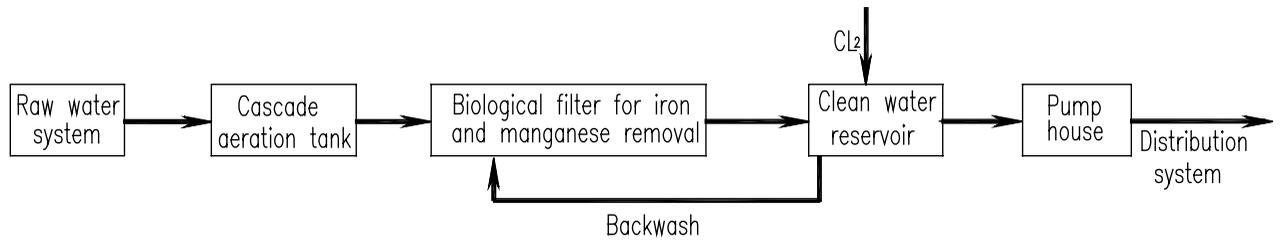

Figure 1 | Plan of the water treatment plant.

Figure 2 | Flow chart of the water treatment plant and water head loss between units.

Table 2 | Main treatment units

Units	Materials	Size (m)	Number	Units	Materials	Size (m)	Number
Cascade aeration tank	RC	Ø10.5 × 0.60	2	Backwash tower	RC	V = 540 m³ Height: 13.5m	1
Biological filter	RC	7.5 × 6.2 × 3.7	12	Pump house	RC	30 × 9	1
Clean water reservoir	RC	43 × 31 × 3.9	2	Iron sludge dehydrate room	RC	30 × 15	1

filtration. Traditional processes composed of many treatment units take up much land and require a large investment. The total investment would be ¥180 million based on the traditional process (two-stage strong aeration and two-stage filtration), while it is only ¥130 million when applying the biological process. Because a lot of the equipment (aeration equipment, treatment medicine introducing equipment) is not needed, operation and maintenance costs were also decreased. After inoculation of the bacteria into the filter, there is a low filtration rate, low intensity of backwash and long filtration period during the culturing period. Both Fe^{2+} and Mn^{2+} can be simultaneously removed when the filter becomes mature.

MATERIALS AND METHODS

All iron and manganese analyses were performed using an atomic absorption spectrophotometer (Lengguang 7520, Shanghai Analytic Instrument Co. Ltd, Shanghai). All the samples were quickly acidified (pH<2) with nitric acid to avoid oxidation prior to atomic absorption spectrophotometry. Before analysis the water samples do not need to be acid-decomposed. The detection limits of iron and manganese are 0.03 mg l^{-1} and 0.01 mg l^{-1}, respectively. For Fe, soluble Fe and total Fe (T-Fe) were detected; T-Fe is the sum of soluble Fe and suspended Fe. For Mn, only total Mn was detected, which is the sum of soluble Mn and suspended Mn. Detection of the bacteria was performed using a microscope (Olympus BX-41TF, Olympus Optical Co. Ltd, Japan).

Medium A: 0.8 g peptone, 0.2 g yeast extract, 0.1 g $CaCl_2$, 0.2 g $MnSO_4 \cdot H_2O$, 0.1 g K_2HPO_4, 0.2 g $MgSO_4 \cdot 7H_2O$, 0.2 g $NaNO_3$, 0.1 g $(NH_4)_2CO_3$, 1,000 ml H_2O, pH 6.8–7.2

Isolation of the bacteria

All the vessels were sterilized before the test. Sterilized distilled water was used as diluent. The method of sequential grads dilution was adopted. 1 ml raw water from the aeration tank was diluted 10_1–10_6 fold, then 0.1 ml of diluted sample was added to the plates with medium A, and incubated at 25°C for 15 days. Round brown colonies, which amount to 50% of the total colonies on plates with medium A, were found after 15 days. The colonies can be regarded as having manganese oxidizing activity if the colour of the colonies changed to blue when TMPD(2,2,4-tetramethy-1,3 pentandiol) solution was injected into it. Five colonies were chose at random and inoculated into medium A. After shaking the culture for 7–10 days (100 rpm, 25°C), the sedimentation was removed by centrifuge at 100g for 10 minutes at 4°C, the upper clear solution was centrifuged at 3000 × g for 10 minutes at 4°C, and the sedimentation was suspended with Tris-HCl buffer (10 mmol l^{-1}, pH 7.2) and centrifuged again. This operation was done twice. Suspended with Tris-HCl buffer (10 mmol l^{-1}, pH 7.2) when the temperature of the buffer was near to 4°C, then the concentration of bacteria was controlled to $OD_{600} = 1.0$. A sample of 10 ml of such suspended bacteria was taken. $MnSO_4$ powder was injected into it until the manganese concentration reached 20 mg l^{-1}. It was
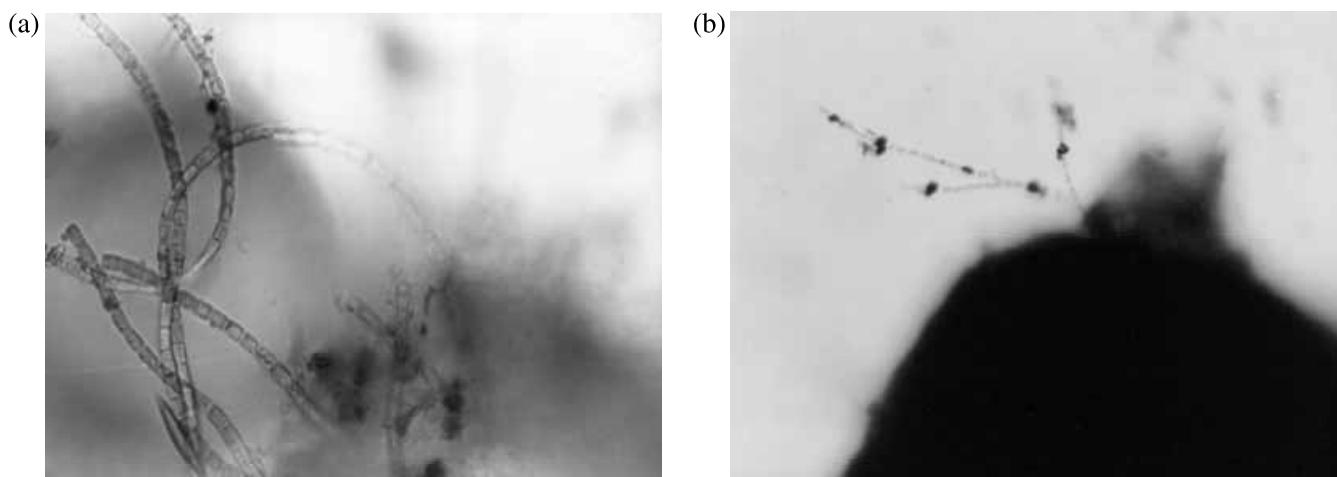
Figure 3 | Iron and manganese oxidizing bacteria (a) in the plastic container; (b) on the surface of the sand.

incubated in a shake bed for 12 hours and after another 12 hours the manganese concentration was measured. This was 24 hours from the beginning of inoculation to the first detection. The bacteria were removed prior to atomic absorption spectrophotometry. From detection results we knew that the bacteria could oxidize Mn^{2+} at the mean rate of 80 nmol day^{-1}.

$$V = 10 \times (20 - X)/55$$

where V is the mean rate of Mn^{2+} oxidation (nmol per 24 hours) and X is the manganese concentration at the 24th hour (mg l^{-1}).

Culture and harvest of the bacteria

Brown colonies were transferred to test tube slants with medium A and incubated at 25°C for 10 days.

Domestication and multiplication of the bacteria

After culturing the bacteria were inoculated into a large plastic container (80 l) and the fresh underground water was introduced into it from the bottom of the container. An aeration fitting was put in to keep the bacteria in a separate state. Weak aeration and low flow rate were controlled avoiding the loss of bacteria to the overfall. The water conditions in the plastic container can be considered as the same as those of the filter environment.

After 2 weeks' domestication and multiplication of the bacteria, the concentration of bacteria reached 1.5×10^9 CFU ml^{-1} (most probable number, MPN) in the plastic container. The iron and manganese oxidizing bacteria in the plastic container are shown in Figure 3(a). About 2,400 l of such bacteria solution was inoculated into each filter (filter depth: 3.7 m, filter bed depth: 0.9 m, filter area: 7.5 m 6.2 m) on 9 September 2001, when low filtration rate (4 m h^{-1}), low intensity of backwash (10 l m^{-2} s^{-1}), long backwash period (48 hours) and short backwash duration (3 minutes) were adopted. All the filter beds are of the same depth (0.9 m). The maturation of the filter involves the multiplication of the microbial community dominated by iron and manganese oxidizing bacteria with intense activity. With the increase of the bacterial population in the filters, the ability to remove iron and manganese is developed. At the end of October, the filters showed a satisfactory performance and then the operational conditions were set at: filtration rate (8 m h^{-1}), intensity of backwash (15 l m^{-2} s^{-1}), backwash period (36 hours) and backwash duration (6 minutes). In one backwash period, the pressure drop is about 1.5–1.8 m. The iron and manganese oxidizing bacteria on the surface of the filtration sand are shown in Figure 3(b).

The concentration of bacteria reached 3×10^6 CFU ml^{-1} on the surface of the filter medium.

A coarse sand medium with diameter 0.9–1.0 mm was adopted in filter 2 in order to compare with the mixed sand medium with diameter 0.5–1.2 mm in filter 1 before inoculation. The uniformity coefficients (P60/P10) and effective sizes (P10) of the filter media in filter 1 and 2 are 1.11 and 0.9 and 1.33 and 0.6, respectively. All the other conditions were the same, such as filter bed depth (0.9 m), filter size, filtration rate, intensity of backwash, the method of inoculation and so on.

Figure 4 | Variation of T-Fe in filtrate and raw water of filter 1.

RESULTS AND DISCUSSION

The bacteria collected from the water supply system were known as native bacteria. The native bacteria include many species. After isolation the bacteria that related to iron and manganese oxidation were inoculated into the culture medium. During the culturing period, the domestication and multiplication of iron and manganese oxidizing bacteria are the key steps. Although the population of the bacteria in culture medium is large after culturing, it is still small for the whole filter. So the multiplication of bacteria in a short time is necessary. After that they were inoculated into the filter. After multiplication for a certain period, say 1–2 months depending on water temperature and characteristics, the filter became mature and exhibited stable performance with high removal efficiency for both Mn and Fe, and started to produce clean filtrate.

The stability of the microbial ecosystem needs to be maintained by adequate populations of some species, especially the iron and manganese oxidizing bacteria as the dominant species. Otherwise, the populations of the dominant species will be lost and the ecosystem will be changed. The bacteria not only adhere to the surface of sand, but are also present in the iron sludge in the filter spaces, whose biochemical oxidation capacity is important to the whole maturated filter. Therefore, it is necessary to maintain a low backwash intensity during the culturing period. The stability of the microbial community requires many operational parameters to be controlled, such as the
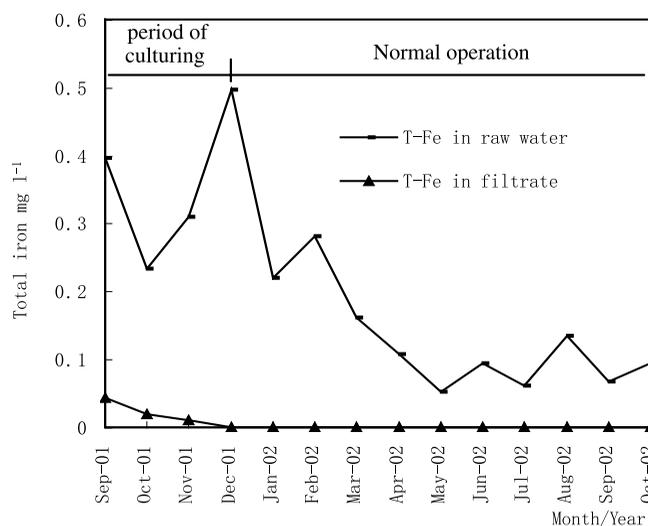
characteristics of raw water, in particular the concentration of Fe^{2+}, backwash intensity, period and duration of backwash. If the raw water contains no Fe^{2+}, the balance of the microbial community with iron and manganese oxidizing bacteria present as the dominant species would be destroyed, and the oxidation activity for Mn would be weakened and even lost (Zhang *et al.* 2001). After the maturation of the filter, stable operation was maintained by the adjustment of suitable filtrate rate, filtration period and backwash intensity and duration as important technical/economic parameters.

Concentrations of iron (Fe^{2+}, T-Fe) and manganese (T-Mn) in raw water and filtrate were measured daily; the data shown in Figures 4–7 were monthly averaged values to indicate the performance over the long period of operation. The performance of filter 1 in the period of culturing and the following 1-year period of normal operation in terms of Fe and Mn removal is shown in Figures 4 and 5, respectively. The figures show the variation of Fe and Mn concentrations in the raw water and filtrate of filter 1 during the operational period with sharp fluctuations of Mn and Fe^{2+} concentrations in the raw water: Mn 0.575–3.05 mg l^{-1} and Fe^{2+} 0.01–0.5 mg l^{-1}. In the early period of operation, Mn concentration in the filtrate was very high. With the increase of the biomass and activity through culturing, the quality of filtrate
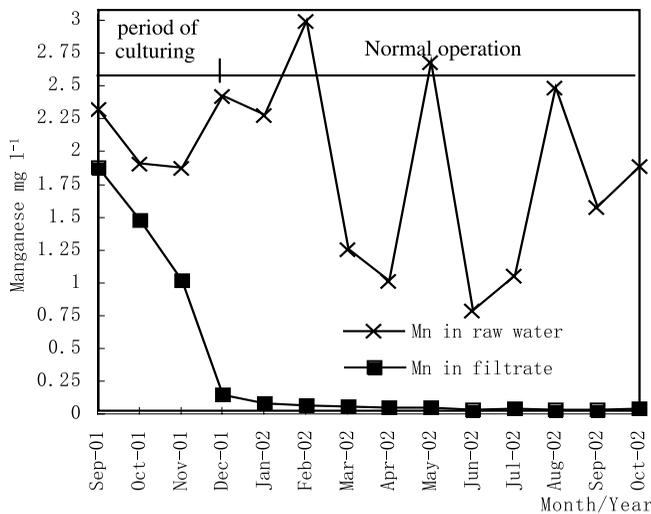
Figure 5 | Variation of manganese in filtrate and raw water of filter 1.

Figure 6 | Variation of T-Fe in filtrate and raw water of filter 2.
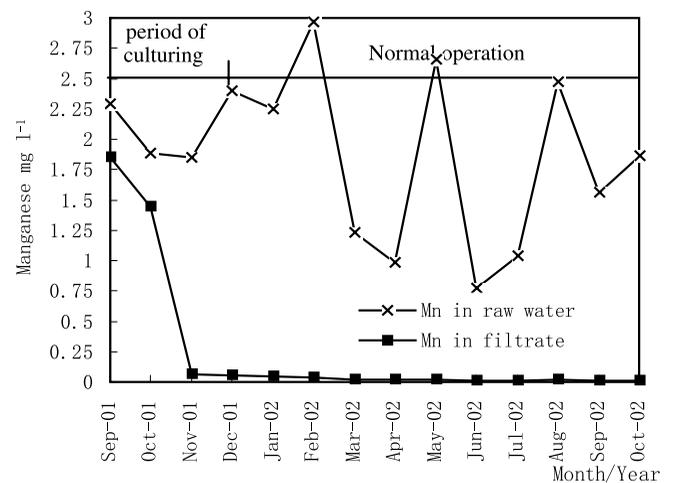
Figure 7 | Variation of manganese in filtrate and raw water of filter 2.

gradually improved. On 25 September, filter 1 exhibited the ability to remove manganese. Two months later, Mn in the filtrate had decreased to 0.12 mg l^{-1}, which showed that the filter had matured. However, it was still not stable because of the fluctuation of filtrate quality to some extent. Three months later Mn in the filtrate was less than 0.05 mg l^{-1}, which meets the Chinese national standard for drinking water: Fe<0.3 mg l^{-1}, Mn<0.1 mg l^{-1} (*Analytical Method* 1998).

The data on the performance of filter 2 during the period of culturing and the following 1-year period of normal operation are shown in Figures 6 and 7, during which time T-Fe was present in the filtrate in trace amounts and Mn concentration was less than 0.05 mg l^{-1}. The variation of Mn removal efficiency of filters 1 and 2 with operation time is shown in Figure 8 and Figure 9, respectively. It is clear that the maturation time of the coarse sand medium with diameter 0.9–1.0 mm is shorter than that of the mixed sand medium with diameter 0.5–1.2 mm and the removal efficiency of filter 2 is superior to that of filter 1, which showed that the uniform coarse sand medium easily adapted to biological removal of iron and manganese. This is ascribed to the development of a deeper effective biological filtering bed and the enhancement of the effective biomass in the filtration space in the uniform coarse sand medium compared with the mixed
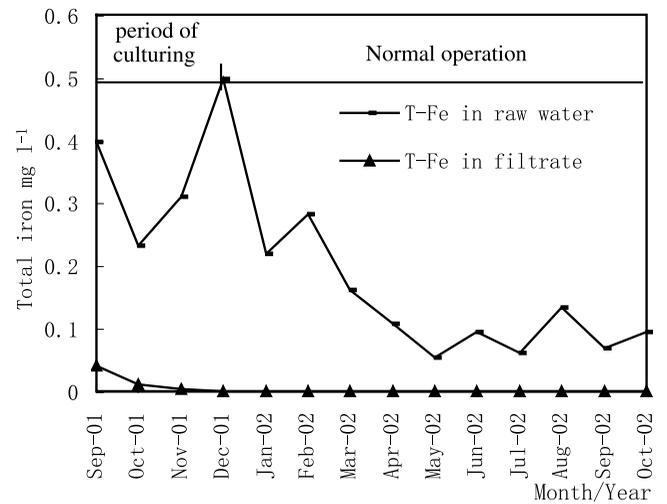
sand medium bed, which led to the improvement of removal capability. Owing to this phenomenon, the coarse sand medium overcame the disadvantage that the removal of Fe^{2+} and Mn mainly takes place on the surface of filter media, which resulted in the decrease of water head loss and the filtration period being prolonged to 52 hours. Iron and manganese can be removed equally and simultaneously along the filter bed, so the cementation of iron sludge on the surface of the filter media was also alleviated and then followed by a complete backwash.
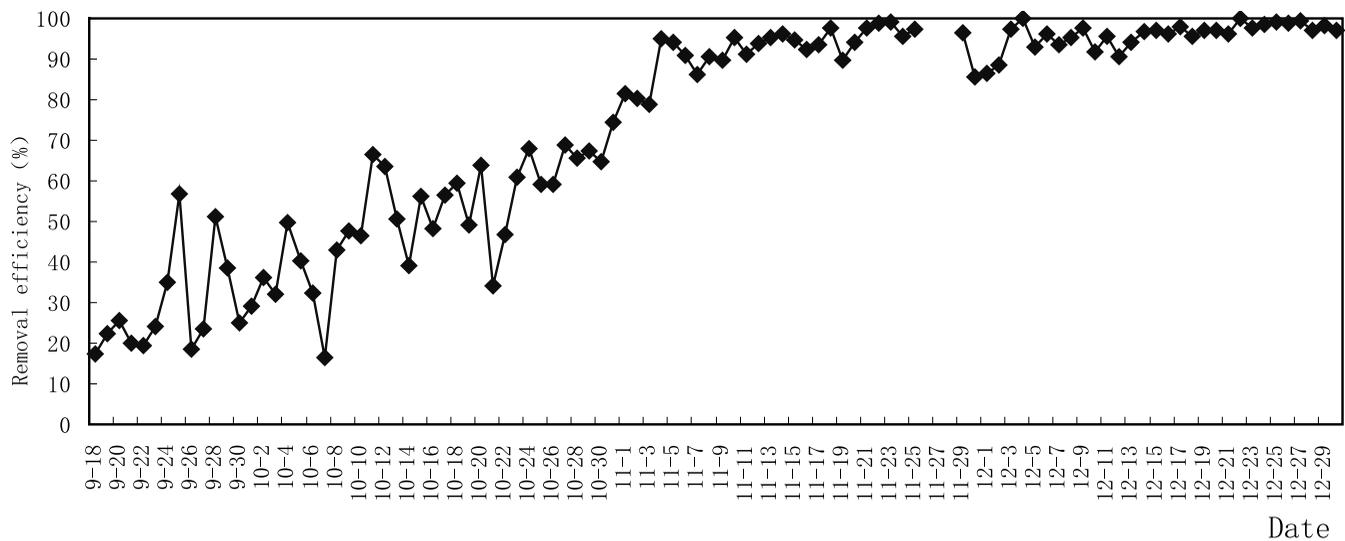
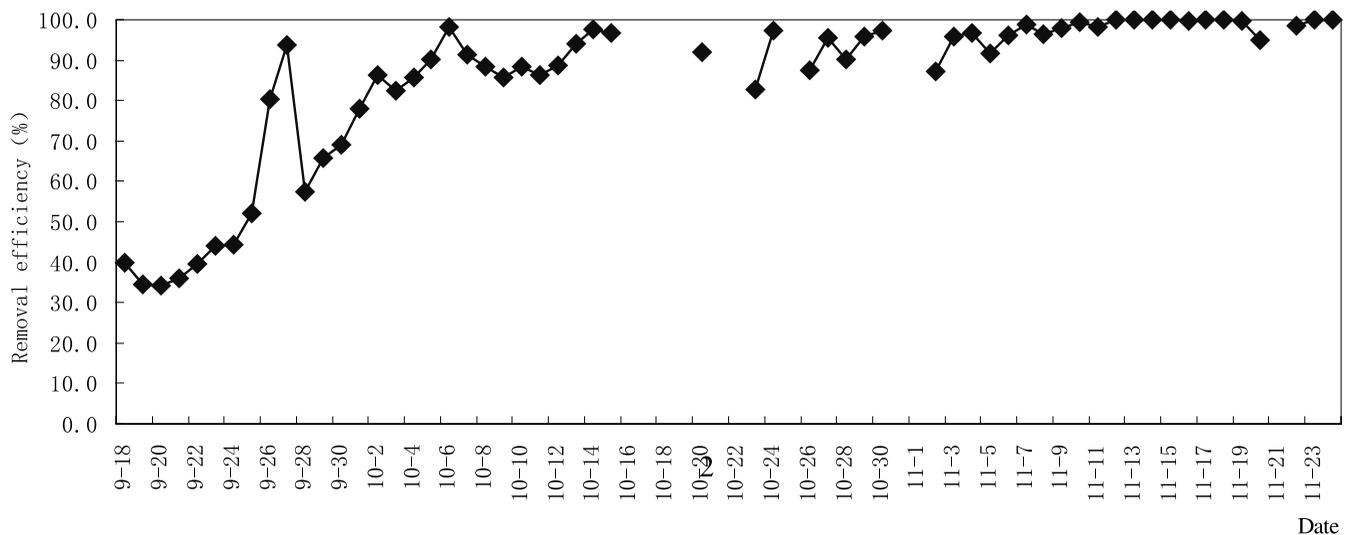**Figure 8** │ Variation of Mn removal of filter 1 with operation time.

Figure 9 │ Variation of Mn removal of filter 2 with operation time.

Some studies on the engineered bacteria have been conducted (Piao *et al*. 1998). Because of the differences in weather and geological conditions, the quality of groundwater is not the same everywhere. In the open environment the engineered bacteria and microbial community must adapt to the specific environment in which they live. The native bacteria have the superiority from this point. Whether the engineered bacteria that are cultured in the lab can survive well in the open environment in a short time is uncertain. It is obvious that it will take some time to obtain a large number of iron and manganese oxidizing bacteria from the native bacteria. Beyond question, the culturing of the engineered bacteria with strong propagation capability and adaptability directly affects its application and value in engineering.

Our early studies showed that during the maturation of the filter, some other bacteria also proliferate such as the *Nitrosomonas* and the nitrobacteria. Their growth influences the concentration of NH_4^+, NO_2^- and NO_3^- in the product water. For some pollutants such as organics, NH_4^+ and NO_2^-, the biological filter demonstrates high removal efficiency (Zhang *et al.* 1996). Some groundwaters contain other heavy metal ions such as Pb^{2+}, Hg^{2+}, Cu^{2+} and Cr^{2+} besides Fe^{2+} and Mn^{2+}. Their potential for harm is greater than that of iron and manganese for human society. Research on the simultaneous removal of these poisonous ions has not been conducted. Research into limitations for removal of Fe^{2+} and Mn in raw water by biological filtration is going on.

CONCLUSIONS

The biological filter for iron and manganese removal was first applied in this WTP. The theory of biological fixation and removal of manganese was confirmed by monitoring the performance of the full-scale plant. To date, a manganese concentration in the filtrate of 0.05 mg l^{-1} and trace amounts of T-Fe have been achieved, which is superior to the Chinese National Standard for drinking water (Fe $<$ 0.3 mg l^{-1}, Mn $<$ 0.1 mg l^{-1}). The population of coliforms is less than 3 CFU l^{-1}, and the total bacterial population is 0 CFU l^{-1}. The technology for biological removal of iron and manganese is technically feasible.

Simultaneous removal of iron and manganese from groundwater was achieved in one biological filter for iron and manganese removal. Although there was a high concentration of Mn (3.05 mg l^{-1}) and a low concentration of Fe (0.01 mg l^{-1}) in the raw water, the quality of the filtrate was excellent. This shows that the treatment technology has a broad flexibility. Research into limitations to the removal potential is going on.

The simple aeration unit can generate enough DO for the biological filter's needs, so it is not necessary to remove CO_2 and develop pH by high-level aeration. Compared with the traditional process, one aeration tank, the manganese removal filter and some other equipment were not needed, so capital investment can be saved and the economic benefit can be increased.

Uniform coarse sand medium has more merits compared with the mixed sand medium for biological filters for iron and manganese removal.

Bacteria with the Fe^{2+} and Mn^{2+} oxidizing activity have long been recognized. However, the taxonomy, genetics and physiology of the bacteria are poorly understood. Progress in these areas will advance the efficiency of iron and manganese removal from groundwater.

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