

Biological iron removal from groundwater: a review

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ABSTRACT

Iron is generally removed from groundwater by the process of aeration or chemical oxidation followed by rapid sand filtration. Different mechanisms (physicochemical and biological) may contribute to iron removal in filters but the dominant mechanism depends on the physical and chemical characteristics of the water and process conditions applied. Nowadays, there are increasing numbers of publications on methods of biological iron removal which are reported to be much more efficient and cost effective than conventional physicochemical iron removal. However, the biological iron removal mechanism is not fully understood and there are still controversies about whether the mechanism of iron removal in filters could be solely biological or whether the presence of iron oxidising bacteria supplements the physicochemical iron removal mechanisms under certain specific conditions. This paper reviews the theoretical background of biologically mediated iron removal, the process conditions required, the advantages and limitations of the method and a few case studies. A literature review revealed that biological iron removal is not suitable when pH and oxygen concentrations are high and/or NH_4^+ , H_2S and Zn are present. Physicochemical removal mechanisms can achieve the same removal efficiency under the conditions that are reported to be favourable for biological iron removal. Biological iron removal is likely to be supplementary to conventional physicochemical iron removal.

Key words | biological oxidation, filtration, groundwater, iron removal

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INTRODUCTION

Groundwater is the major source of drinking water throughout the world. Iron being the fourth most abundant element and the second most abundant metal in the Earth's crust (Silver 1993; WHO 1996), it is a common constituent of groundwater. There are no health-based guidelines for the concentration of iron in drinking water. However, it is undesirable as it causes several aesthetic and operational problems including bad taste, discolouration, staining, deposition in distribution systems leading to aftergrowth and incidences of high turbidity. Based on taste and nuisance considerations, the World Health Organisation (WHO) recommends that the iron concentration in drinking water should be less than 0.3 mg/l (WHO 1996). The EC directive recommends that the iron in water supplies should be less than 0.2 mg/l (EC 1998). In the Netherlands, several water supply companies are aiming at an iron level of

<0.05 mg/l in order to minimise the distribution system maintenance costs.

Several methods, namely oxidation–precipitation–filtration, lime softening, ion-exchange, sub-surface iron removal and membrane processes, have been employed for iron removal from groundwater. Stabilisation with phosphate or silicates is applied as well to avoid the oxidation or precipitation of iron. Among the different techniques mentioned above, aeration or chemical oxidation followed by rapid sand filtration is most widely used (O'Connor 1971; Wong 1984; Salvato 1992; Twort *et al.* 2000; Sommerfeld 1999). A sedimentation step sometimes precedes the rapid filtration for solid liquid separation, especially when iron concentration is very high, in order to reduce the load on subsequent rapid filters. Aeration–rapid sand filtration is the preferred method in developed as well as in developing

countries because, compared to other methods, this method is more economical, less complicated and generally avoids the use of chemicals, which is not usually welcome in the water industry.

Different mechanisms (physicochemical and biological) may contribute to iron removal in filters but the dominant one depends on the physical and chemical characteristics of the water and process conditions (Lerk 1965; Rott 1985; Hatva 1988, 1989; Mouchet 1992; Michalakos *et al.* 1997; Søgaard *et al.* 2000). In the commonly applied oxidation–precipitation–filtration method (physicochemical iron removal process) two mechanisms can be identified (Figure 1), namely:

- *Oxidation–floc formation (floc filtration)*, in which iron(II) is first oxidised to iron(III) by oxygen or chemical oxidant, which upon hydrolysis and agglomeration forms iron hydroxide flocs. These flocs are subsequently removed in rapid (sand) filters.
- *Adsorption–oxidation (adsorptive filtration)*, which involves the adsorption of iron(II) onto the surface of the filter media and its subsequent oxidation in the presence of oxygen or other oxidant to form a new iron oxide layer. This oxide layer (coating) enhances the adsorption and oxidation of iron(II) and facilitates the process. Oxidant concentration and pre-oxidation time have to be limited to avoid the oxidation–floc formation mechanism to occur.

The adsorption–oxidation mechanism of iron removal has several advantages, e.g.:

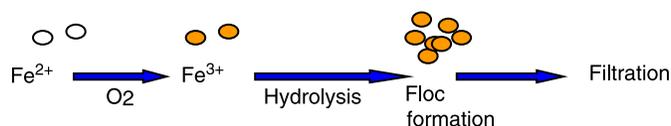
- it allows higher filtration rates and longer filter run lengths, since the specific volume of iron oxide adsorbed/precipitated on the sand grains is much lower than that of iron hydroxide flocs,
- its high filtrate quality.

In conventional iron removal plants both of these physicochemical removal mechanisms occur simultaneously. The dominant mechanism under given conditions depends on the water quality and process conditions applied; specifically, low oxygen concentration and a short pre-oxidation time in the supernatant of the filters reduces the role of the “oxidation–floc formation” mechanism and as a consequence the “adsorption–oxidation” mechanism will be dominant. At high pH values, the rate of oxidation of iron(II) is higher which is in favour of the “oxidation–floc formation” mechanism. This paper focuses on the role of the biological iron removal mechanism.

BIOLOGICAL IRON REMOVAL

Biologically mediated oxidation and removal of iron has been reported in rapid sand filtration of groundwater (Frischherz *et al.* 1985; Czekalla *et al.* 1985; Bouwer & Corstjens 1993; Badjo & Mouchet 1989; Hatva 1989;

Oxidation-floc formation mechanism



Adsorption-oxidation mechanism

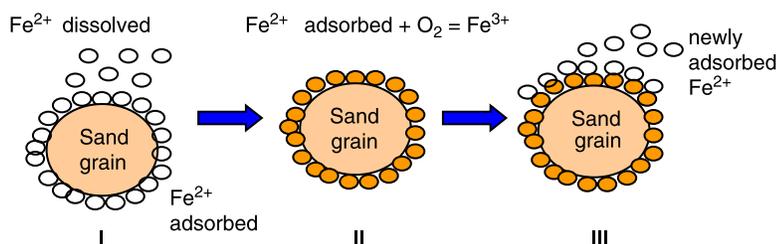


Figure 1 | Physicochemical iron removal mechanisms

Mouchet 1992; Bourguine *et al.* 1994; Tyrell 1997; Tyrell *et al.* 1998). Biological iron removal is attributed to the activities of microorganisms. These microorganisms are reported to have the unique property of causing oxidation and precipitation of dissolved iron under pH and redox potential (*Eh*) conditions that are intermediate between those of natural groundwater and those required for conventional (physical–chemical) iron removal (Wolfe 1964; Mouchet 1992).

Mouchet (1992) defined the field of activity of these iron bacteria, which straddles the theoretical boundary between the fields of Fe^{2+} stability and the formation of iron hydroxides as defined by thermodynamic analysis of the electrochemical equilibria (Figure 2). This is related to the fact that iron bacteria relying on iron(II) oxidation as a source of energy are gradient organisms, signifying that they are not likely to develop under strongly reducing or strongly oxidising conditions, but rather at the point where they have a source of both ferrous ions and air (Wolfe 1964; Trembley *et al.* 1998). By aerating the raw water and increasing the dissolved oxygen concentration, the redox potential of raw water is increased in the pH–*Eh* range where biological iron oxidation is expected to take place predominantly (Mouchet 1992).

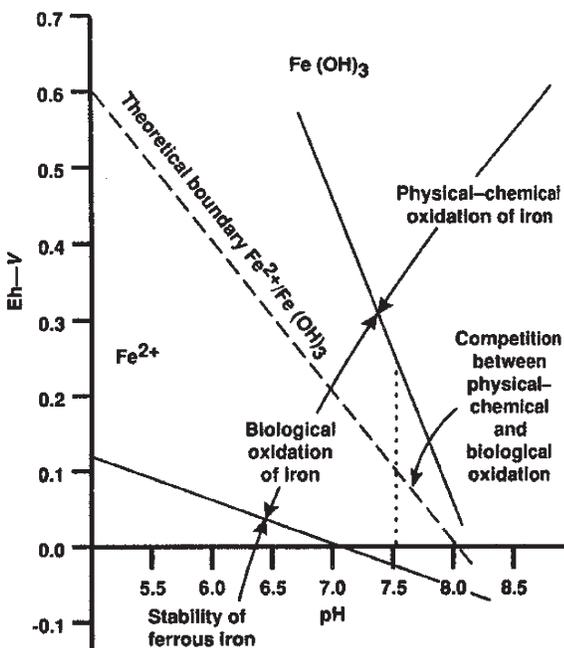


Figure 2 | Field of activity of iron bacteria (Mouchet, 1992)

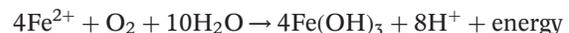
Mechanism and process conditions

The exothermic oxidation of iron(II) can be catalysed by some bacteria due to the oxidation–reduction enzymes that they excrete (flavins); trivalent iron rendered insoluble in hydroxide form is then stored in the mucilaginous secretions (sheaths, stalks, capsules, etc.) of these bacteria. The organisms responsible for this phenomenon belong to the genera *Gallionella*, *Leptothrix*, *Crenothrix*, *Clonothrix*, *Siderocapsa*, *Sphaerotilus*, *Ferrobacillus* and *Sideromonas* (Wolf 1964; Degremont 1991; Corstjens 1993). These iron-oxidising bacteria are widespread and are prevalent in groundwater, ponds, hypolimnion of lakes or impoundments, sedimentary deposits and soil. Two mechanisms of bacterial oxidation have been reported (Czekalla *et al.* 1985; Bourguine *et al.* 1994):

- (i) intracellular oxidation by enzymatic action of autotrophic bacteria (*Gallionella* and *Leptothrix ochracea*),
- (ii) extracellular oxidation by the catalytic action of polymers excreted by iron bacteria (*Gallionella*, *Leptothrix*, *Crenothrix*, *Clonothrix*, *Sphaerotilus* and *Siderocapsa*).

In anaerobic groundwater, iron is mainly present in soluble forms of iron(II). Physicochemical removal processes require aeration of this water in order to sufficiently raise the redox potential of the water so that the dissolved iron(II) present will be converted into insoluble oxidized forms (Stumm & Lee 1961; Sung & Morgan 1980).

The metabolic activities of iron bacteria are not fully understood but it is believed that the same oxidation of iron is carried out by some variation of the physical–chemical reaction:



It is believed that the energy that is released by this reaction allows the bacteria to reduce and assimilate the carbon from CO_2 , thereby synthesising their own nutrients. However, oxidation of ferrous ions does not furnish much energy on a molar basis and about 600 mol of Fe^{2+} is needed to assimilate 1 mol of carbon (Viswanathan & Boettcher 1991; Mouchet 1992). This implies that large amounts of

iron have to be oxidised to satisfy the energy requirements for the growth of iron oxidising bacteria.

Gage *et al.* (2001) stated that, whatever the metabolic pathway for the iron oxidation reactions, the biological process is catalytic in nature and causes a rapid oxidation. The red insoluble precipitates formed are all slightly hydrated iron oxides that, beneficially, are more compact forms than the precipitates formed when using physical–chemical processes. This feature partially explains the greater iron retention capacity between backwashes of biological filters when compared to physical chemical treatment filters. It is likely that in these “physical–chemical filters” the oxidation–formation mechanism is dominant as the observed effect of a greater iron retention capacity can also be attributed to the adsorption–oxidation mechanism.

Iron bacteria are generally robust and, because of the variety of species involved, one type or another is able to thrive under most environmental conditions. A pH of 6–8 is required for their activity. However, at a pH above 7.2, biological processes will compete with conventional (physical–chemical) processes. The optimum temperature typically ranges from 10–15°C for *Gallionella ferruginea* and 20–25°C for the *Sphaerotilus–Leptothrix* group (Mouchet 1992). The biological oxidation process is, however, inhibited in the presence of H₂S, chlorine, NH₄⁺ and some heavy metals (Twort *et al.* 2000). Mouchet (1992) pointed out that water applied to the biological filter must contain <0.01 mg H₂S/l. Furthermore, it was reported that the iron removal rate decreased by 50% in the presence of a zinc concentration of 0.45 mg Zn/l and there was total inhibition of the treatment when the zinc concentration reached 1 mg Zn/l.

Morris & Siviter (2001) mentioned that the biological iron removal process is not suitable for operation where there are rapid or large flow variations because the bacterial population needs to adjust over a period of time to the new conditions. Constant flow and loading provide the optimum operating conditions for biological iron removal plants. They added that the following criteria are normally required for biological iron removal to be effective:

Dissolved oxygen	< 1 mg/l (< 10% saturation)
pH	5.5–7.5
Redox potential	> 100 mV
Iron(II)	0.1–10 mg/l

Iron(III)	Negligible
Inhibitory materials (e.g. H ₂ S, NH ₄ ⁺ , Zn)	Negligible

However, it is to be noted that, in the presence of oxygen, iron(II) is always partly oxidised in the supernatant and in the filter bed. So there will always be some iron(III) present when iron(II) is removed in the filter in the presence of oxygen.

Advantages and limitations of biological iron removal

Mouchet (1992) reported the marked improvement in performance by converting conventional iron treatment plants to biological ones. The primary advantages associated with this process were reported to be high filtration rates (10–70 m/h), high retention capacity (1–5 kg Fe/m²), elimination of chemical reagents, flexibility of operation, and reduced capital and operating costs. According to Mouchet (1992) a shift from physicochemical to biological precipitation of iron can increase the water treatment plant capacity substantially and reduce operation costs by up to 80%. However, high iron concentrations in the influent may cause breakthroughs, as the rate of absorption by the bacteria may not be high enough to match the supply rate.

Gage *et al.* (2001) reported that biological iron and manganese removal systems could have the following advantages:

- smaller plants because of higher applied filtration rates (sometimes in excess of 50 m/h versus 10–15 m/h) or because aeration and filtration can take place simultaneously in the same vessel;
- longer filter runs because of iron retention in the filter due to the formation of more dense precipitates and the use of a more coarse media;
- denser backwash sludge that is easier to thicken and dewater;
- higher net productions due to less water being required for backwashing and being able to use raw water for the backwash;
- require no chemical addition;
- no deterioration of water quality over time;
- lower capital and operating costs through the elimination of chemicals, less frequent backwashing, fewer components, etc.

The main disadvantage of this method is the long maturation time before full efficiency is achieved; perhaps 50–60 days for a new filter and 5 days after a 2-month shutdown (Stevenson 1997). The other shortcomings of this method include:

- not suitable for all types of groundwater;
- anaerobic conditions may develop in the filter bed, thus converting back iron(III) to iron(II), resulting in an elevated iron concentration in the filtrate;
- increased sludge production and backwash water with filter ageing;
- need for two filtration stages to remove iron and manganese as the required redox potential conditions for iron and manganese oxidising bacteria are very different;
- ineffective in the presence of ammonia (NH_4^+) and inhibiting substances like H_2S and Zn (Twort *et al.* 2000; Stevenson 1997; Gage *et al.* 2001).

Comparison of iron removal mechanisms and case studies

Czekalla *et al.* (1985) studied the performance of 15 groundwater treatment plants in Hamburg, 3 reservoir water treatment plants in the Harz Mountains, together with a groundwater treatment plant and 2 test filtration plants in Braunschweig, Germany. They analysed the backwashing materials and microbial settlements in the filter and reported that bacterial species of *Gallionella*, *Leptothrix*, *Toxothrix*, *Metallogenium*, *Hyphomicrobium*, *Siderocapsa* and *Siderocytis* were responsible for the oxidation of iron and manganese in water. It was concluded that rapid sand filters in waterworks not treated by oxidising agents are biotechnological plants and the resident bacteria require optimal care (i.e. physicochemical conditions) to operate efficiently, as in the case of any living system.

Smith & Smith (1994) compared the efficiency of physicochemical and biological iron removal processes for three raw waters with iron levels of 1.8 mg/l, 2.4 mg/l and 2.9 mg/l during the entire filter cycle. Performances of the filters were compared in terms of flow rates, filter run length, backwash water savings and water quality. It was found that “biological filters” can operate at higher filtration rates and

have longer run lengths without appreciable head loss. The biological filter not only met the 0.3 mg/l iron standard faster than the existing physicochemical plant after the backwash, but also reached a lower filtrate iron concentration. In all the cases, the iron concentrations in the filtrate of biological filters were non-detectable (<0.05 mg/l). Furthermore, since the biological filters were using untreated water for backwashing and backwashed at lower rates, substantial water savings were realised.

Michalakos *et al.* (1997) studied the removal of iron in pilot-scale trickling filters and compared the performance of the filters in the presence and absence of iron bacteria. The process conditions applied in this study were: temperature = 28°C, pH = 7.0–7.3, $\text{O}_2 = 8$ mg/l and Fe = 1–17 mg/l. A start-up time of approximately 10 days was necessary for the biological oxidation to begin. In order to assess the filter performance with physicochemical iron oxidation alone, the same set of experiments were repeated after the filter was disinfected with 5% NaOCl to exclude bacterial growth. It was concluded that the existence of iron bacteria in the filter dramatically improved the filter efficiency under the same operating conditions. For the same filtration rate, the biological oxidation allowed treatment of almost double the iron concentration than physicochemical oxidation alone.

There are several patented and trademark biological iron removal systems now available on the market. Degremont (1991) has proprietary filters for biological iron and manganese removal, namely Ferazur and MagnazurTM, respectively. Anglian Water has developed “Bioferro”, a biological process for the removal of iron from groundwaters. Biological iron removal has also been reported to be employed for removing iron from handpump water supplies (Tyrrel 1997; Tyrrel *et al.* 1998) and in slow sand filters (Hatva 1988, 1989; Seppanen 1992).

It is to be noted that there is some contradiction among the claims of different researchers regarding the oxygen concentration in water in “biological iron removal”. Some reported that for biological iron removal the oxygen concentration in the water should be low (Mouchet 1992; Morris & Siviter 2001) while others claimed to have achieved biological iron removal even with high concentration of oxygen at near neutral pH (Michalakos *et al.* 1997; Czekalla *et al.* 1985; Hatva 1988, 1989; Seppanen 1992).

Table 1 summarises some water quality and process parameters of the few “biological iron removal” plants reported in the literature. This shows that the method has been applied in different parts of the world for a wide range of iron concentrations (up to 16 mg/l), filtration rates (8–30 m/h) and temperatures (11–31°C).

DISCUSSION

It is still debated whether “near neutral pH bacteria” actually oxidise iron(II) and grow autotrophically or merely deposit iron in an oxidised form. Ehrlich (2002) stated that “microbes can promote iron oxidation, but this does not mean that the oxidation is always enzymatic. Because ferrous iron has a tendency to auto-oxidize (without bacteria or catalyst) in aerated solution at pH values above 5, it is difficult to demonstrate enzyme-catalyzed iron oxidation in near-neutral air-saturated solutions. At this time the most extensive evidence for enzyme-catalyzed iron oxidation in air-saturated solution by bacteria has been amassed at pH values below 5”.

In iron bacteria other than the acidophile species, autotrophy using ferrous iron as a source of energy has not been conclusively demonstrated (Hughes & Poole 1989). The bacteria do, however, process iron intracellularly. Environments high in iron(II) will lead to elevated iron levels inside the cell. The cells have mechanisms to remove such unwanted ions. Therefore, oxidation of iron(II) may be purely to detoxify their intracellular environment rather than to create energy. The cell membrane and extracellular polymers do provide many sites for the adsorption of iron(II) ions. Once adsorbed the ions may undergo the oxidation reaction. This leads to the formation of the characteristic sheaths often cited as evidence for biological iron removal. It appears that bacteria can act as a pathway for iron(II) oxidation either via adsorption onto the cell membrane or via oxidation to detoxify the intracellular environment. It cannot be established if such mechanisms provide a significant removal capacity (Hughes & Poole 1989).

Søgaard *et al.* (2000) proposed that a probable explanation for the increased precipitation rate of iron in biological filters relative to physicochemical filters could be that precipitation of iron hydroxides takes place in contact

with exopolymers of the bacterium *Gallionella ferruginea*. The exopolymers acts as a catalyst for the oxidation process of iron and prevent re-dissolution of iron hydroxides in spite of the oxic/anoxic conditions found in the biological filters. Furthermore the fact that the iron-precipitating bacterium *Gallionella ferruginea* is chemolithotropic with respect to the oxidation of iron(II) is not proven.

The reported increased efficiency and advantages of biological iron removal can also be achieved if the mode of operation of iron removal filters is switched from predominantly floc filtration (oxidation–floc formation) to predominantly adsorptive filtration (adsorption–oxidation) (Sharma *et al.* 2001). The commercially available catalytic filter media BIRM (a federally registered trademark of Clack Corporation, USA) also claims similar advantages as that of biological iron removal reported by Mouchet (1992), Gage *et al.* (2001) and others. Therefore, it is still to be answered what is the clear advantage of using bacteria for iron removal in place of physicochemical processes, as iron bacteria may not be available in all groundwaters.

In reality the boundary between physicochemical and biological iron removal is not well defined (Degremont 1991). Mouchet (1992) pointed out that the domain of effective biological iron removal is narrower than the field of existence of iron bacteria, as shown in Figure 1. Iron removal from anoxic groundwater by adsorption onto new or iron oxide coated filter media is possible in the pH–*Eh* range of iron bacteria (Sharma *et al.* 1999, 2001), which indicates that the process is not necessarily biological. In many iron removal plants and pilot studies, nearly complete removal of iron has been seen to occur immediately after the beginning of a filter run or test. The fact that iron was removed at the very beginning of the filtration process strongly suggests that this process is predominantly a physicochemical reaction. Furthermore, in the presence of oxygen iron(II) oxidation starts immediately (after the aeration step), implying that biological iron removal might be only supplementary to the conventional oxidation–floc formation method of iron removal.

It has been reported that in some plants the removal efficiency has increased after the development of bio films or “coatings” on the surface of the filter media (Mouchet 1992; Bourguin *et al.* 1994). This improved efficiency can also be explained by the increased adsorption of iron(II) onto

Table 1 | Summary of biological iron removal treatment plants

Treatment plant	Fe (mg/l)	Mn (mg/l)	pH	Eh (mV)	Temp. (°C)	Filtration rate (m/h)	Capacity (m ³ /d)	Media	Reference
Grove, Nottingham, England	0.16		7.7		13.3	30	6700	Sand 8/16 grade	Morris & Siviter (2001)
Astrup Waterworks Esbjerg, Denmark	11–13	0.45–0.7	6.5–6.9	–60		11.6	6000–7200	Quartz 2–5 mm	Nielsen (1996); Søgaard <i>et al.</i> (2000)
Grindsted, Denmark	3		7.3	–60		8	7200–9600	Quartz 2.4–4 mm	Søgaard <i>et al.</i> (2000)
Dalstrops Waterworks, Sweden	0.05	0.42	8						Hedeberg & Wahlberg (1998)
Saint Hill Water Treatment Plant, England	2.5–4.0		6.7–6.8		11	26	6500		Bourgine <i>et al.</i> (1994)
Lome, Togo	0.8–1.1		6.1–6.5		30–31		44000	Sand, effective size 1.35 mm	Badjo & Mouchet (1989); Mouchet (1992)
Barrie Ontario Ferazur, Canada	0.5	<0.05				22			Gage <i>et al.</i> (2001)
Waterloo, Quebec, Canada	0.6–1.71	0.26–0.45				12	4128		Williams (2002)
South France	3.8		6.5 – 7.0	290		30	1000	Effective size 1.18 mm	Tremblay <i>et al.</i> (1998)

the surface of iron oxide coatings formed on the filter media (Sharma *et al.* 1999) or the increased rate of iron(II) oxidation in the presence of an iron oxide coated surface (Tamura *et al.* 1976; Tufekci & Sarikaya 1996). Indeed, controversy surrounds the current debate as to whether the mechanism of iron removal in filters is physicochemical or biological in nature (Czkella *et al.* 1985; Mouchet 1992; Søgaard *et al.* 2000). However, it is to be noted that the adsorption–oxidation mechanism of iron removal can achieve the same iron removal efficiency and has the same advantages under the same process conditions that are favourable for biological iron removal.

SUMMARY AND CONCLUSIONS

A review of the available literature on biological iron removal points to the following findings:

- Iron in groundwater could be removed by biological oxidation under specific water quality and process conditions. It is not possible to employ the biological iron removal method for all groundwaters, especially when pH and oxygen concentrations are high and/or NH_4^+ , H_2S and Zn are present.
- Biological iron removal is very sensitive to process conditions (e.g. temperature, filtration rate, oxygen concentration) and will not be suitable when there is rapid or large variation in the flow and water quality.
- Biological iron removal is not necessarily the sole iron removal mechanism in the filters. Physicochemical mechanisms like oxidation–floc formation and adsorption–oxidation (adsorptive filtration) can achieve the same removal efficiency under the same conditions, so it is not clear which mechanism is dominant.
- The presence of iron oxidising bacteria in iron removal filters is not unlikely to increase the efficiency of iron removal in filters. They could contribute by catalytically oxidising iron(II) to iron(III) if the rate of oxidation of iron(II) is very slow in water. This type of iron oxidation is only possible if the iron is entering the filter bed in iron(II) form. However, in the presence of oxygen iron(II) oxidation starts immediately (after the aeration step) in the supernatant and filter bed. Moreover the adsorption–oxidation mechanism will occur in the filter

bed as well, implying that biological iron removal is likely to be supplementary to conventional physico-chemical iron removal.

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