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Removal of iron and manganese using biological roughing up flow filtration technology

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Abstract

The removal of iron and manganese from groundwater using biological treatment methods is almost unknown in Latin America. Biological systems used in Europe are based on the process of double rapid biofiltration during which dissolved oxygen and pH need to be strictly controlled in order to limit abiotic iron oxidation. The performance of roughing filter technology in a biological treatment process for the removal of iron and manganese, without the use of chemical agents and under natural pH conditions was studied. Two pilot plants, using two different natural groundwaters, were operated with the following treatment line: aeration, up flow roughing filtration and final filtration (either slow or rapid). Iron and manganese removal efficiencies were found to be between 85% and 95%. The high solid retention capability of the roughing filter means that it is possible to remove iron and manganese simultaneously by biotic and abiotic mechanisms. This system combines simple, low-cost operation and maintenance with high iron and manganese removal efficiencies, thus constituting a technology which is particularly suited to small waterworks.

Keywords: Groundwater; Iron; Manganese; Iron oxidizing bacteria; Manganese oxidizing bacteria; Roughing filtration

1. Introduction

Iron (Fe) and manganese (Mn) cause aesthetic, organoleptic and operating problems when they are present in groundwater. These metals consume chlorine in the disinfection process and promote biofouling and microbiological induced corrosion in water networks.

In groundwater, Fe and Mn are present as Fe(II) and Mn(II). The processes available for their removal are either physico-chemically or biologically based. The advantages of biological treatments compared with conventional physico-chemical treatments can be summarized as follows: no use of chemicals, higher filtration rates, the possibility of using direct filtration and lower operation and maintenance costs (Mouchet, 1992).

Fe and Mn removal by biological processes are based on different stages of biofiltration where beds are colonized by Fe–Mn oxidizing bacteria. In nature, iron oxidizing bacteria (IOB) and manganese oxidizing bacteria (MnOB) are widespread. They are prevalent in groundwater, swamps, ponds, in the hypolimnion of lakes, in sediments, soils, wells and water-distribution systems. In the latter they can cause significant clogging problems (Ghiorse, 1984). These bacteria which are present in raw water can multiply in sand filters under

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appropriate conditions and are able to oxidize divalent ions Fe(II), Mn(II) and precipitate them under their oxided forms Fe(III) and Mn(IV).

IOB and MnOB, studied since the late eighteenth century, have been recognized for their ability to deposit Fe hydroxide or Mn oxide in structures outside their cells. Many researchers have studied the physiology of these microorganisms and the mechanisms involved (Van Veen et al., 1978; Ghiorse, 1984; Hallbeck and Pedersen, 1991; Corstjens et al., 1992; Søgaard et al., 2000). However, many aspects of Fe and Mn deposition are still poorly understood.

The main groups of IOB are (Czekalla et al., 1985; Mouchet, 1992):

- stalked bacteria, e.g. *Gallionella* sp., which are chemolithotrophic and microaerophilic,
- sheathed bacteria, e.g. *Leptothrix* sp., *Sphaerotilus*, etc., which are facultative autotrophic-heterotrophic,
- unicelular bacteria, e.g. *Siderocapsa*, *Siderocystis*, etc., which are heterotrophic and more difficult to recognize by microscopic observation than the previous ones.

With the exception of the stalked bacteria (*Gallionella*), which oxidizes only Fe, MnOB include the major groups reported for the IOB. Some of the species of the genera *Leptothrix*, *Crenothrix*, *Hyphomicrobium*, *Siderocapsa*, *Siderocystis*, and *Metallogenium* can even oxidize Fe or Mn indifferently. Other bacteria oxidize only Mn, e.g. *Pseudomonas manganoxidans* (Mouchet, 1992).

The bacteria involved in biological Fe and Mn removal need different pH and redox potential (Eh) conditions for each metal, as is shown in Fig. 1, a Pourbaix diagram.

IOB may be completely aerobic or microaerophilic, depending on the pH, whereas MnOB require a fully aerobic environment (DO > 5 mg/l) to precipitate Mn (Mouchet, 1992; Gislette and Mouchet, 1997).

Nowadays biological processes to remove Fe and Mn are widely used in Europe, and there are some treatment plants in the United States and Canada (Mouchet, 1995; Gage and Williams, 2001) but they are almost unknown in Latin America (Pacini et al., 2003). In general, patented systems used for the removal of these two metals include: initial aeration followed by rapid filtration for Fe removal and secondary aeration, pH adjustment and secondary rapid filtration for Mn removal (Czekalla et al., 1985; Mouchet, 1992; Gislette and Mouchet, 1997). The above-mentioned systems need sophisticated devices to control DO, pH and Eh in order to limit abiotic Fe oxidation. Because of differences in

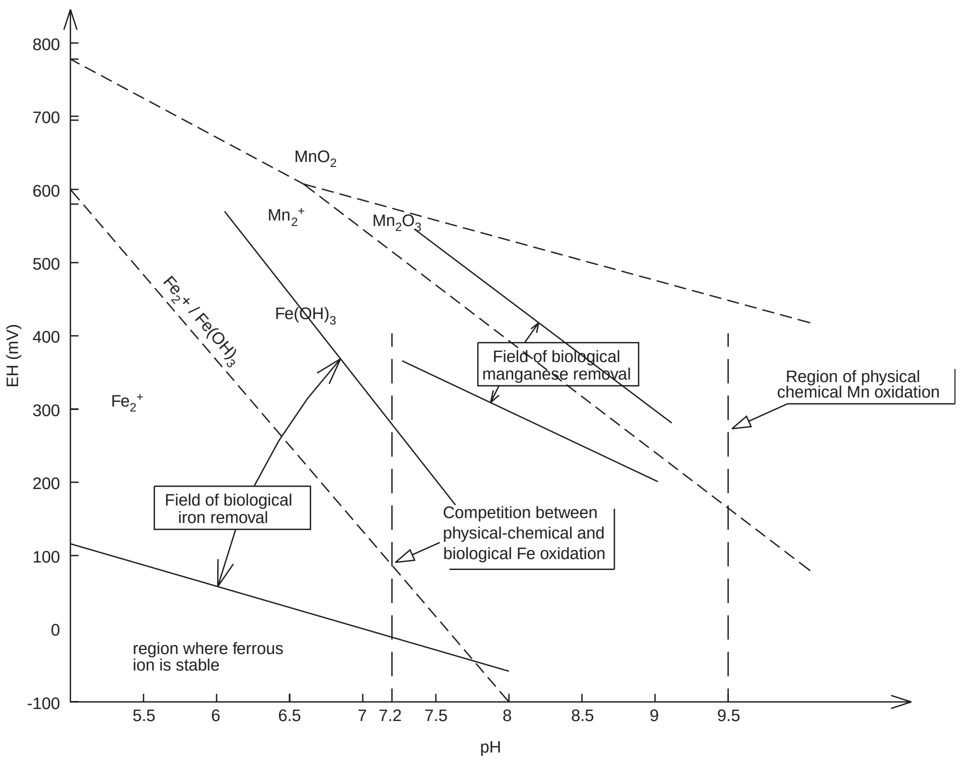


Fig. 1. Field of activity of Fe and Mn oxidizing bacteria in a pH-Eh diagram (Mouchet, 1992).

optimum conditions for the biological removal of Fe and Mn, (see Fig. 1), when rapid filters are used, it is not possible to carry out removal of Fe and Mn simultaneously in one step, except when very low velocities are used. In several regions of northern Europe, slow sand filter processes including one or two pre-treatment steps are also applied (Hatva, 1988; Wahlberg and Hedberg, 1997).

Roughing filtration technology is a filtration process through a coarse medium using low filtration rates. It is mainly used as pretreatment in order to retain solid matter before slow filtration (Wegelin, 1988; Boller, 1993). This process has been used successfully as pretreatment to remove turbidity, being subsequently followed by slow sand filtration (Ingallinella et al., 1992, 1998).

Given the high solid retention capability of roughing filtration, this process was considered likely to be an appropriate treatment for the removal of Fe and Mn from groundwater by means of biological processes. The system proposed in this paper comprises: aeration, up flow roughing filtration and final filtration. At first, slow sand filtration as final filtration was tried out (Phase I). On completion of the Phase I trials, it was proved that simultaneous removal of Fe and Mn could be achieved. The processes were carried out at natural pH and Eh, without using special devices to control DO. In order to extend the field of application to larger water flows, the slow filtration process was then replaced by a rapid filtration process in another series of experiments (Phase II). Also the following issues were studied: the performance of the system with two different compositions of natural waters, two types of aerators, different granulometric configurations and different operating parameters. The aim of this paper is to present the main results of the two pilot plant tests.

2. Materials and methods

2.1. Pilot plants descriptions

Pilot plant tests were carried out using groundwater with two different compositions, the first containing Fe and Mn, whereas the second contained only Mn. Both pilot plants were installed in the province of Santa Fe, Argentina, the first in the city of Avellaneda being monitored from April 2000 to December 2003. The second pilot plant was located in Las Garzas and was operated over a period of six months during the second half of 2001. The main physico-chemical characteristics of the raw waters are summarized in Table 1.

The pilot plant in Avellaneda was operated in two phases. During Phase I, the processes used were: aeration (trickling filter), up flow roughing filtration and slow sand filtration. In Phase II the final step was replaced by rapid filtration.

Las Garzas pilot plant comprised the following processes: aeration (holed trays) followed by up flow roughing filtration. No final filtration step was used because these trials were focused on the roughing filter performance.

Design and operative parameters were chosen based on previous research and literature data. The optimal roughing filtration rate, for surface water, ranged from 1 to 1.20 m/h, according to Ingallinella et al. (1992). However, pretreatments used for the biological removal of Fe and Mn were able to operate at higher velocities (2–3 m/h) (Hatva, 1988).

During the cleaning process of the roughing filter, the water stored inside the filter was drained out at high drainage velocities. Wegelin (1988) recommended a high initial filter drainage velocity from 60 to 90 m/h be used. Two different filter media were tested: Type 1 medium

Table 1
Quality of raw water used in pilot plant trials

Parameters	Units	Avellaneda				Las Garzas			
		Minimum	Average	Maximum	*N*	Minimum	Average	Maximum	*N*
pH		7.0	7.1	7.3	22	6.8	7.1	7.4	32
Redox potential (Eh)	mV	325	361	391	11	389	423	505	19
Dissolved oxygen	mg/l	4.6	6.0	7.4	37	1.1	1.30	1.9	10
Turbidity	NTU	1.02	1.55	2.15	5	0.46	0.65	0.90	31
Color	mg/l(Pt/Co)	12	15	20	5	1	4	11	30
Total Fe	mg/l	0.19	0.29	0.44	41	0.09	0.12	0.37	32
Ferrous Fe	mg/l	0.05	0.13	0.22	41	0.03	0.02	0.06	18
Total Mn	mg/l	0.18	0.24	0.37	36	1.08	1.44	1.83	32
Dissolved Mn	mg/l	0.15	0.21	0.29	26	1.00	1.35	1.76	10
Hardness	$mgCaCO_3/l$	254	265	275	5	*	*	*	*
Alkalinity	$mgCaCO_3/l$	259	279	294	5	*	*	*	*
Conductivity	μS/cm	838	940	1104	41	*	*	*	*

N: Number of observations. *Not measured.

was selected according to Ingallinella et al. (1992), and Type 2 medium was tried out in order to optimize the cleaning process.

During Phase I, slow filtration rates were 0.20–0.35 m/h, the filter medium being sand of 0.15–0.30 mm in diameter. During Phase II, rapid filtration rates of 12 and 15 m/h were used (Trauman, 1997). Furthermore, combinations of two backwashing rates (25 and 50 m/h) with two backwashing times (10 and 20 min) were tried out. The main specifications and design parameters of the pilot plants are described in Fig. 2. The operational parameters and the granulometric configuration of the

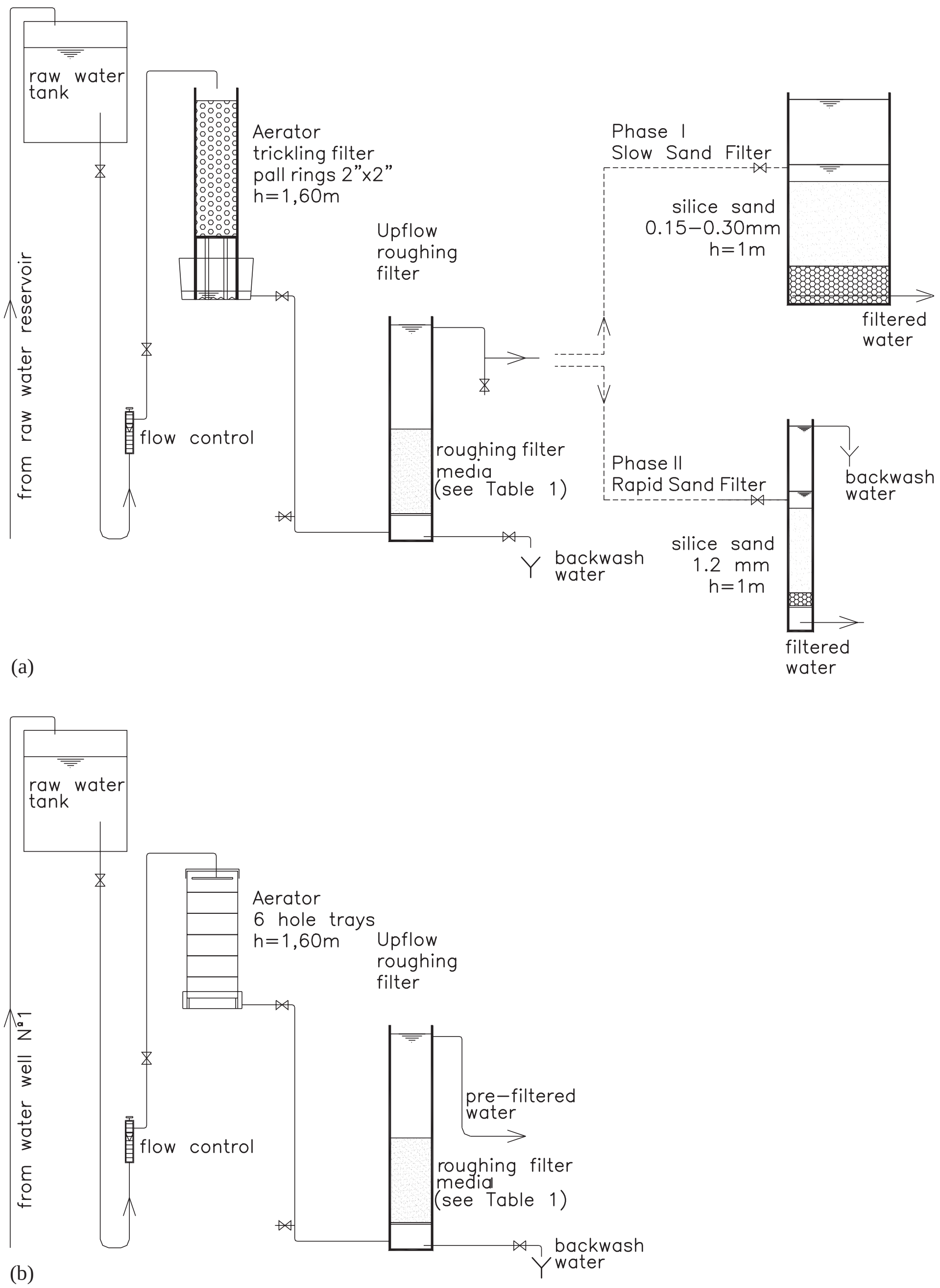


Fig. 2. (a) Avellaneda Pilot Plant, (b) Las Garzas Pilot Plant.

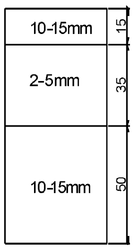
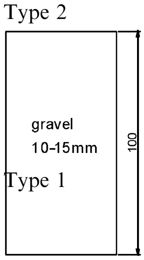
roughing filter used in the different filter runs are shown in Table 2.

2.2. Sampling and analytical methods

Raw water and the effluent of each process were sampled once a day, 1–3 days per week. The following parameters were analyzed: pH and Eh (pH-meter HACH sension1), DO (DO-meter Hanna HI9143), and turbidity (Turbidimeter Lamotte 2008). Total and ferrous iron were determined by the phenanthroline method, with the addition of sodium metabisulfite for total iron (Spectrophotometer HACH DR 4000). Total and dissolved manganese were determined by the PAN Hach Method 8149; for dissolved manganese the samples were filtered by 0.45 μm membrane (Spectrophotometer HACH DR 4000). Water flow, head loss and environment temperature were also registered.

Given the difficulties involved in isolating IOB and MnOB in the laboratory, surface attachment collection methods (flow cell) described by Smith (1992) and Gariboglio (1996), were utilized. The flow cells was composed of a cylindrical chamber with 8 microscope slides inside through which water flowed (2 l/h). The flow cell was used to collect bacteria in wells, in the raw water reservoir and in treated water during a period of 7–15 days in each site.

Table 2
Pilot plants operative conditions

Trials	Aerator (type)	Roughing filter		Final filtration		Filtration rate, m/h (Backwash rate, m/h)		
		Filter media	Runs	Runs	Type	Aerator	Roughing filter	Final filtration
Avellaneda Pilot Plant Phase I (26/04/00 03/09/01)	Trickling filter	Type 1 Gravel	N°1 (11 weeks)	N°1 (24 weeks)	Slow sand filter	1	1 (90)	0.20
			N°2 (13 weeks)			2	2 (90)	0.35
			N°3 (5 weeks)	N°2 (11 weeks)		1	1 (90)	0.20
			N°4 (6 weeks)			2	2 (90)	0.35
			N°5 (7 weeks)	N°3 (11 weeks)		2.5	2.5 (90)	0.45
			N°6 (4 weeks)					
			N°7 (7 weeks)	N°4 (13 weeks)				
			N°8 (6 weeks)					
Avellaneda Pilot Plant Phase II (22/10/01 14/07/03)	Trickling filter	Type 1	N°1 (9 weeks)	N°1–N°2 (16 to 29 days)	Rapid sand filter	2	2 (90)	12
			N°2 (22 weeks)	N°3 to N°8 (20 to 30 days)		2	2 (90)	12
			N°3 (7 weeks)	N°9 to N°11 (6 to 12 days)		2	2 (90)	12
			N°4 (4 weeks)	N°12 to 20 (limit to 7 days)		2–2.5	2–2.5 (90)	12–16
		Type 2 	N°5 (8 weeks)	—	—	2	2 (90)	—
			N°6 (8 weeks)					
			N°7 (8 weeks)					
Las Garzas Pilot Plant (11/06/01 24/12/01)	Holed trays	Type 1	N°1 (17 weeks)	—	—	2-1	1.5–0.8 (100)	—
			N°2 (9 weeks)			1	0.8 (150)	—

Microbiological samples at different filter beds were also collected using mini-flow cells that consisted of a plastic basket containing 4 microscope slides. Samples of fresh backwash sludge and raw water reservoir sediments were collected directly.

Slides from flow cell and mini-flow cells and sludge samples were examined with a trinocular microscope, Olympus BX40. Keys given in the ASTM Standard Test Method for Iron Bacteria (ASTM, 1997), Standard Methods (APHA, 1995), Methods for the Examination of Water and Associated Materials (Environmental Agency, 1998) and papers by different authors were used for the identification of bacteria (Van Veen et al., 1978; Czekalla et al., 1985; Spring, 2002).

3. Results and discussion

3.1. Avellaneda pilot plant

3.1.1. Microbiology observations

Before starting up the operation of the pilot plant, a great number of IOB (especially *Gallionella)* and precipitated Fe material were detected in the wells and in the water reservoir (Fig. 3, left).

During the operation of the pilot plant, slides from flow and mini-flow cells located in different points, were examined microscopically. The following bacteria were detected:

- A great number of *Gallionella* and precipitated ferric material in the water entering the aerator.
- A great number of *Gallionella* and sheathed bacteria, possibly *Leptothrix* (both with a large amount of ferric material on their stalks and sheaths) and unicelular bacteria, regularly aligned, inside the roughing filter (Fig. 3, right).
- A great number of *Gallionella* and sheathed bacteria, possibly *Leptothrix*, inside the slow sand filter (Phase I) and in the rapid sand filter (Phase II).
- A short number of stalked and sheathed bacteria in the water leaving the system during Phase I and II.

These observations might suggest that most of the microorganisms present in the raw water colonized the different filter beds but did not enter the filtered water, in a similar pattern to the results reported by Hatva (1988).

Microscopic observation of backwash sludge of roughing filter and rapid filter showed the presence of large numbers of *Gallionella* inside large flocculent precipitates of Fe and Mn. Sheathed bacteria, detected in miniflow cells, were not however observed in fresh sludge samples, probably because they were masked by the precipitates.

3.1.2. pH, Eh, DO and temperature

Mean values of pH and Eh in different runs during Phase I are shown in Fig. 4. Similar results were found during Phase II.

As for pH, Eh and DO analyzed in raw water, it was found that their values corresponded to those present in a competition zone between biotic and abiotic Fe oxidation mechanisms. However, regarding Mn removal, pH and Eh values clearly corresponded to those present in a zone where biotic oxidation mechanisms take place (see also Fig. 1).

In general, pH in raw water (Phase I and II) showed little variation from neutral value but increased to a mean value of 7.7 after aeration, probably due to CO_2 liberation in the aeration process and its consumption by autotrophic bacteria metabolism (*Gallionella*). A slight increase of pH was detected in the following steps.

An increase of Eh values in the water leaving the aeration device and a slight increase in the other stages of the process were observed. These variations registered throughout the different stages are similar to those observed by Hatva (1988), who reported increases in Eh values from +261 to +479 mV in a biological treatment system composed of a dry contact filter, a wet contact

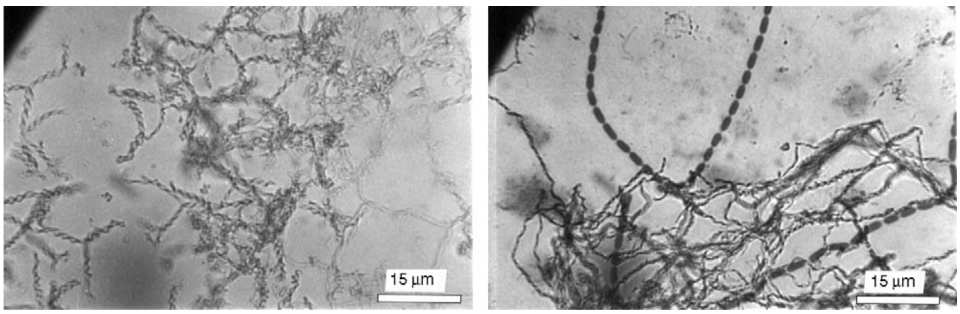


Fig. 3. (Left) *Gallionella* in raw water, (right) *Gallionella* and sheathed bacteria in roughing filter.

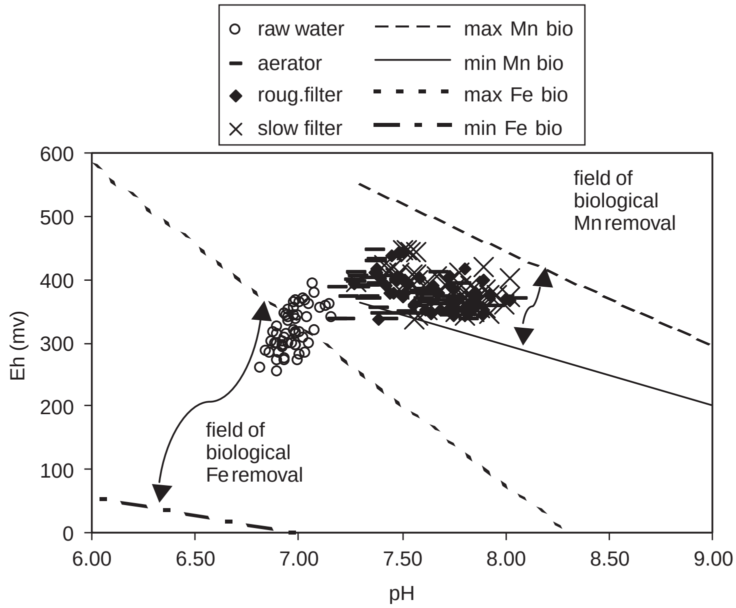


Fig. 4. pH-Eh Diagram, Phase I, Avellaneda Pilot Plant.

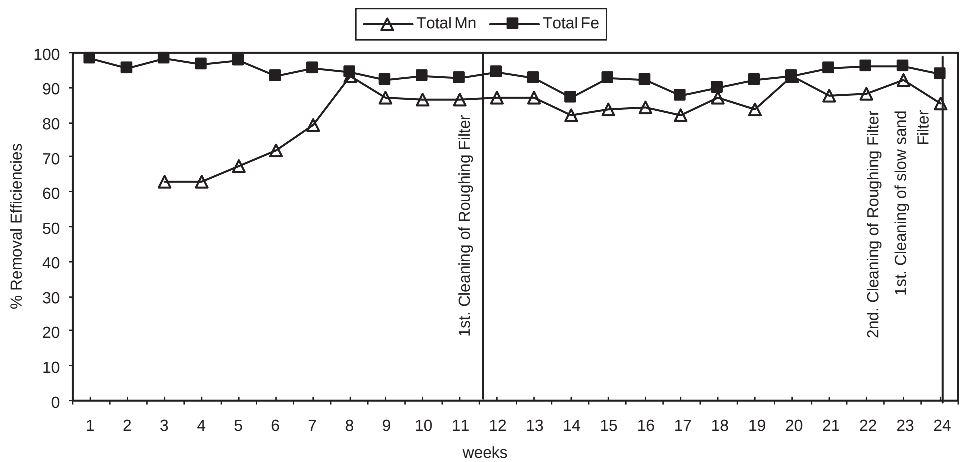


Fig. 5. Total Fe and total Mn removal efficiencies during Run 1, Phase I, Avellaneda Pilot Plant.

filter and a slow sand filter. Mean DO value was 6 mg/l in raw water because of the aeration produced during the filling-up of the reservoir, reaching 8.5 mg/l at the outlet of the aeration device. During Phase I, the temperature was also measured, with mean values between 15 °C and 24 °C being registered. Mouchet (1992) reported optimum temperature ranges from 10 °C to 15 °C for *Gallionella* and 20 °C–25 °C for the *Sphaerotilus–Leptothrix* group.

3.1.3. Startup period

Fig. 5 shows total Fe and total Mn removal efficiencies during the startup period (Phase I). From the beginning of the operation and throughout the entire trial the Fe removal efficiency reached 90%. Regarding Mn, there was an initial period of eight weeks in which its removal efficiency rose from 63% to over 90%. The removal efficiency was calculated as $(C_{\text{inlet}} - C_{\text{outlet}}/C_{\text{inlet}})100$, where C_{intet}: concentration of total Fe or total Mn at the system inlet and C_{outlet}: concentration of total Fe or total Mn at the system outlet.

This behavior coincides with the findings of Seppänen (1988) who concluded that the time necessary for Mn bacteria development is longer than that needed by Fe bacteria and can even take several months.

3.1.4. Total Fe and Mn

Total Fe and Mn concentrations in the water leaving each step of each run during Phase I (see Table 2), after startup period, are shown in Fig. 6.

Total Fe concentration at the aerator outlet varied significantly due to the detachment of Fe precipitates developed in the plastic carrier. However, total Fe values at the roughing filter outlet showed less variation and mean values decreased throughout the successive runs. Mean total Fe concentration at the slow sand filter outlet was 0.03 mg/l throughout the whole of the Phase I trials showing little variation, in spite of the variation of raw water quality. This concentration was much lower than the recommended limit prescribed by current regulations (0.10 mg/l).

In general, total Mn concentration at the aerator outlet showed less variation than that registered in total Fe concentration, as may be observed from Fig. 6. During the fourth run, the detachment of precipitates in the aerator did not mean an increase in total Mn concentration in its outlet. As from the second run, an average of 0.05 mg/l total Mn was reached at the roughing filter outlet, which is the recommended value in the regulation in force. Mean total Mn concentration at the slow sand filter outlet was 0.025 mg/l as from the second run.

Removal efficiencies were on average 95% for total Fe and 88% for total Mn, over the whole period after start-up. As can be observed, the behavior of the roughing filter was steady and the removal efficiency gradually improved throughout the course of the subsequent runs. The efficiency was not affected either by successive hydraulic cleanings, shutdowns of the pilot plant or by increasing the filtration rate.

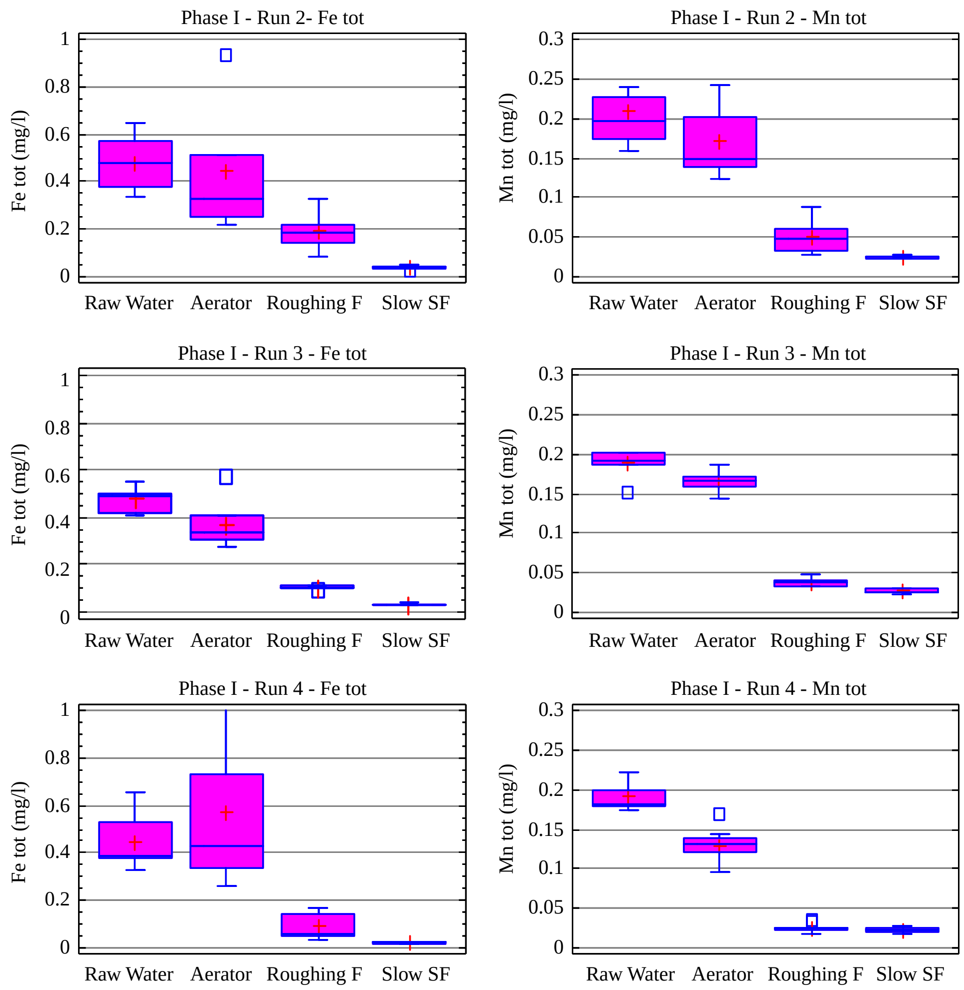


Fig. 6. Total Fe and Mn in the outlet of each stage, Phase I, Avellaneda Pilot Plant. Box and whisker diagrams. Horizontal lines: first quartile, median value and third quartile. Cross: arithmetic mean. Bars: 1,5 interquartile range. Squares: atypical value.

Total Fe and Mn concentrations at the outlet of each step in each roughing filter run during Phase II are shown in Fig. 7.

The behavior of the aeration and roughing filtration processes during Phase II was similar to those observed in Phase I. In the third run there were peaks of total Fe at the outlet of the aerator (1.4 mg/l) which were reflected in total Fe concentration at the outlet of the roughing filter. Nevertheless total Fe concentration at the rapid filter outlet was around 0.012 mg/l in all runs. As for total Mn removal, the performance of the roughing filter was more constant than for Fe removal, with Mn mean values that ranged from 0.019 to 0.013 mg/l at the system outlet. Mean removal efficiencies were on average 94% for total Fe and 92% for total Mn.

3.1.5. Ferrous iron and dissolved manganese

During Phase I, ferrous iron and dissolved manganese were measured in the water leaving each step (see Fig. 8).

Ferrous iron concentration in the raw water was about 40% of total Fe due to the high level of aeration which occurred during the filling up of the raw water reservoir. A significant reduction of ferrous iron concentration was registered in the aerator, due probably to a combination of physico-chemical oxidation and biological oxidation taking place in the biofilms which developed on the plastic carriers. This is in agreement with other authors' findings (Hatva, 1988; Seppänen, 1988). On the contrary, a negligible oxidation of Mn was observed in the aeration step and the dissolved manganese concentration was on average 85%

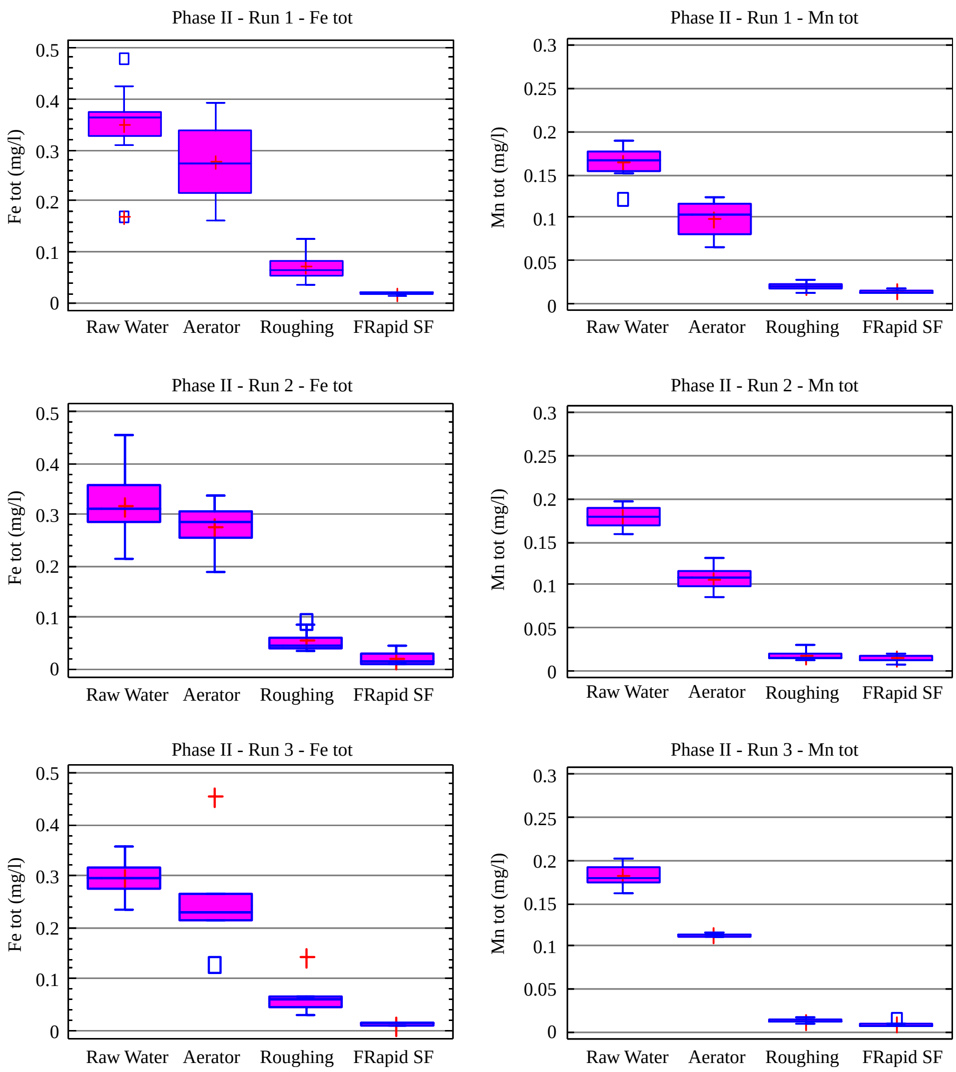


Fig. 7. Total Fe and Mn at the outlet of each stage, Phase II, Avellaneda Pilot Plant. Box and whisker diagrams. Horizontal lines: first quartile, median value and third quartile. Cross: arithmetic mean. Bars: 1,5 interquartile range. Squares: atypical value.

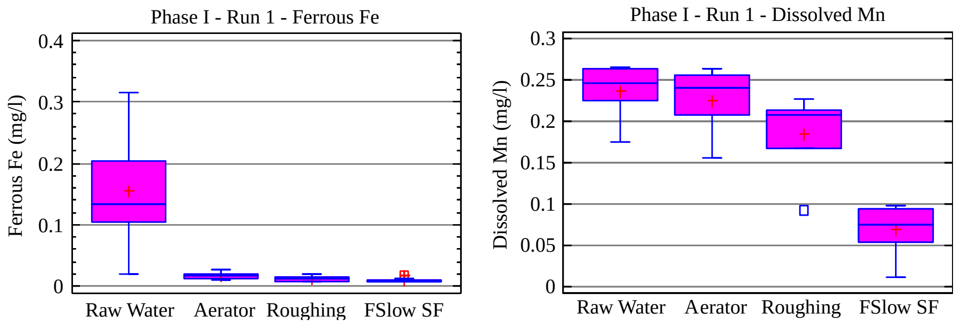


Fig. 8. Ferrous iron and dissolved manganese, Avellaneda Pilot Plant. Box and whisker diagrams. Horizontal lines: first quartile, median value and third quartile. Cross: arithmetic mean. Bars: 1,5 interquartile range. Squares: atypical value.

of the total Mn. This result is due to the fact that abiotic oxidation of Mn by DO is very slow at pH < 9.6 (Stumm and Morgan, 1996).

3.1.6. Turbidity

Average turbidity at the outlet of the system during Phase I ranged from 0.4 to 0.6NTU. At the outlet of the aeration device a high variability in turbidity was registered due to detachment of precipitates. Average turbidity at the outlet of the system during Phase II ranged from 0.2 to 0.4NTU. High variability in turbidity at the outlet of the aerator was detected, similar to Phase I, with peaks of 20NTU. In the following stages (roughing filter and rapid filter) turbidity was stable and the overall Fe and Mn removal efficiency was not affected by the above-mentioned peaks.

Both biofilms and Fe precipitates were accumulated onto the aerator contact material. When they reached a certain size they were detached and transported by the water, increasing turbidity and Fe concentration in the water at the outlet.

The trickling filter aerator was highly efficient in reaching the expected DO level. However, due to the problems outlined above, the authors concluded that another type of aerator, with less contact material, might be more suitable for the purpose of reducing detachments (see 3.2).

3.1.7. Filtration runs

3.1.7.1. Roughing filter runs. During Phase I, no criteria were found for the selection of the optimal run duration of the roughing filter, since no breakthrough was observed, (i.e. the filtered water quality was not affected). Hence, a period of 30–90 days was empirically chosen. In Phase II, it was decided that the run be limited to reach a maximum value of head loss (22 cm). Two piezometers were installed. After the first cleaning, (see Fig. 9), a recovery of head loss around 15 cm was observed, but subsequent cleanings produced only a poor recovery of head loss.

At the end of these trials the filter bed was removed. In the intermediate layer (see Table 2) mudballs and cementation between gravel grains, especially in the interface with the bottom layer were observed. These were undoubtedly due to deficient cleaning of the filter bed.

It was concluded that the criteria adopted to select the optimal run duration were not suitable and that more trials would be required to find the optimal combination between the frequency of roughing filter washing, drainage velocity and its granulometric configuration.

Due to the clogging problems described above, the filter medium Type 1 was subsequently replaced by a uniform medium (Type 2). Using the new medium, three roughing filter runs were then performed using 2 m/h as the filtration rate and a duration run of 60 days.

Fe removal efficiencies registered ranged from 60 to 85%, values which were lower than those obtained using the filter medium Type 1, but still high. The lowest efficiency value corresponded to the detachments produced in the aerator. Fe average values at the outlet of the roughing filter were between 0.05–0.10 mg/l, reaching maximum values of 0.20 mg/l. Mn removal at the outlet of the roughing filter ranged from 75 to 94%, similar to that obtained with the filter medium Type 1. Due to the higher solid retention capacity of this roughing filter bed, head loss values were lower than 5 mm, hence it was concluded that the Type 2 medium was more suitable than Type 1.

3.1.7.2. Slow sand filter runs. The first slow sand filter run, during Phase I, was of 24 weeks whereas the following runs were from 10 to 12 weeks. At the top of the filter medium, mudballs within a layer of 10 cm were observed after long shutdown periods, due probably to the significant level of hardness of the water (see Table 1).

3.1.7.3. Rapid sand filter runs. During Phase II, runs of rapid filter based on different operative parameters were evaluated. In all trials washing time was 10 min. When

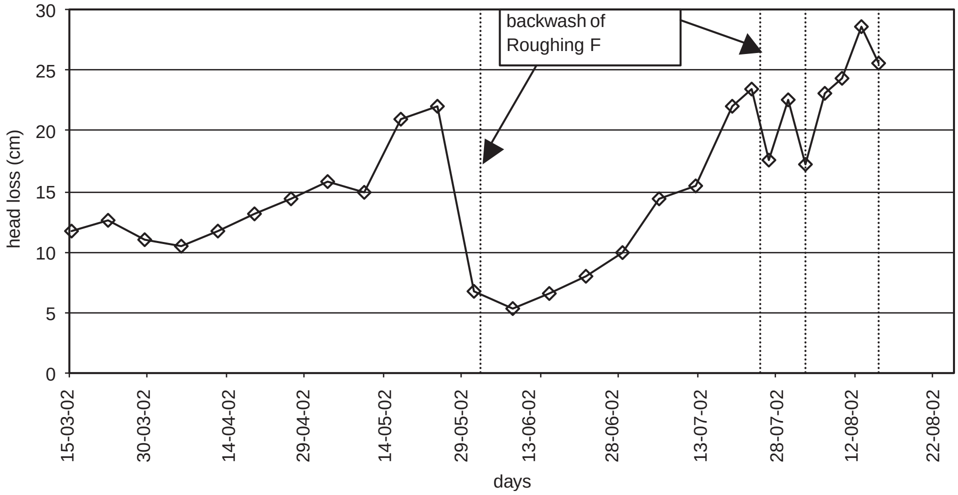


Fig. 9. Roughing filter head loss, Phase II, Avellaneda Pilot Plant.

50 m/h was used as the backwashing rate, the duration of the runs was on average 29 days. However, when reduced to 25 m/h, the backwashing was not effective enough and the length of the runs became successively shorter ending up with cycles of 6 days.

An experiment was designed (Pacini et al., 2003) to evaluate the optimal combination of the operative parameters for the rapid filter. The following parameters were selected: filtration rates of 12 and 15 m/h, backwashing rates of 25 and 50 m/h and backwashing times of 10 and 20 min. Finally, it was concluded that the best combination for the system tried out was: a filtration rate of 12 m/h, a backwashing time of 10 min and a backwashing rate of 50 m/h.

3.2. Las Garzas pilot plant

3.2.1. Microbiological observations

Using the methodology described above, a high concentration of sheathed bacteria, possibly Lepthotrix, was detected in the well water.

During the operation of the pilot plant, a microscopic examination of slides placed in mini-flow cells inside the roughing filter was carried out. A small number of *Gallionella*, and a large number of sheathed bacteria, possibly of the *Leptothrix genus,* were detected.

In all cases, the oxidized Mn, possibly deposited as MnO_2, was observed either as black precipitate coating the free cells, or as pustules on the sheaths of the filamentous bacteria.

3.2.2. pH, Eh and DO

In Fig. 10, pH and Eh registered during operation of the pilot plant in Las Garzas are shown. It may be observed that these values were within the field of biological Mn removal.

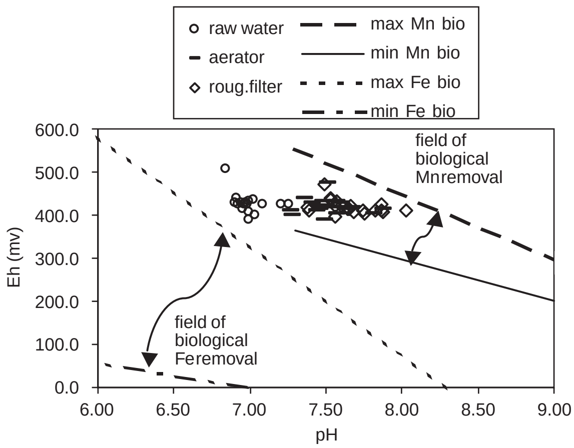


Fig. 10. pH-Eh diagram. Las Garzas Pilot Plant.

DO average values were 1.30 mg/l in raw water, 8.10 mg/l at the aerator outlet and 7.40 mg/l at the roughing filter outlet.

3.2.3. Startup period

Total Mn removal efficiency increased linearly reaching 95% in the eighth week (see Fig. 11-a), with a top value of 98% during startup period.

Likewise, in the case of the Avellaneda trials, the period needed to reach the maximum total Mn removal efficiency was 8 weeks, which is in accordance with the findings of several other researchers (Hatva, 1988; Gislette and Mouchet, 1997).

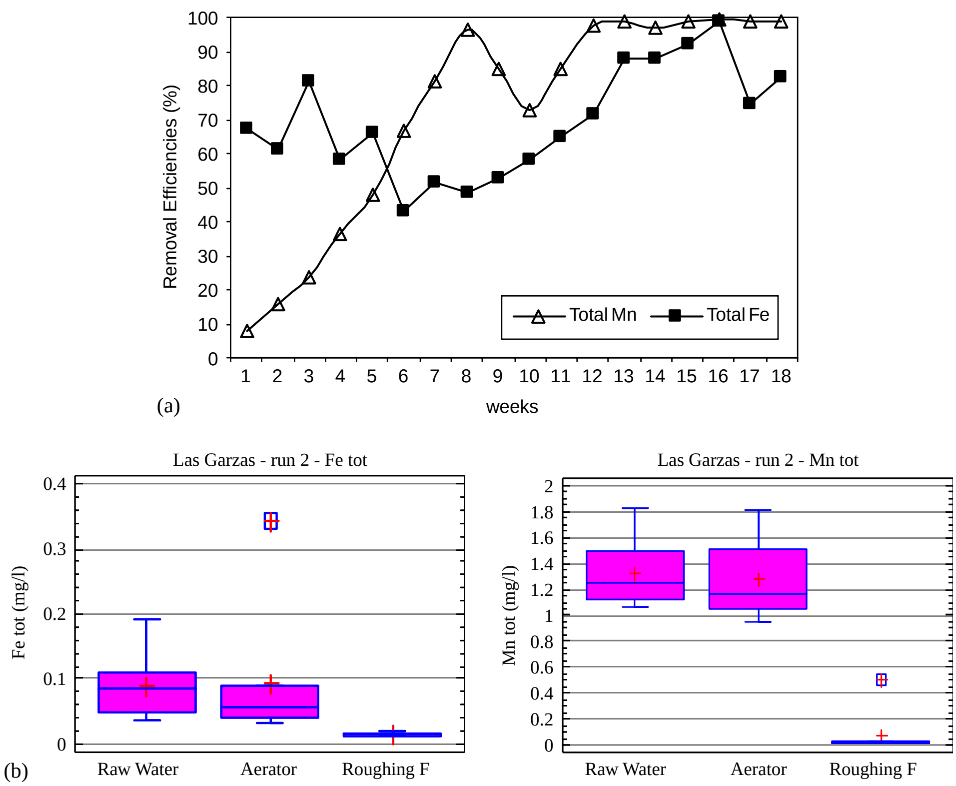


Fig. 11. Las Garzas Pilot Plant: (a) Total Fe and Mn removal efficiencies; (b) Total Fe and Mn at the outlet of each reactor after startup period (Box and whisker diagrams). Horizontal lines: first quartile, median value and third quartile. Cross: arithmetic mean. Bars: 1,5 interquartile range. Squares: atypical value.

3.2.4. Total Fe, total Mn and turbidity

Total Mn average concentration at the outlet of the roughing filter was 0.020 mg/l and total average Fe concentration was 0.015 mg/l (see Fig. 11b).

Average turbidity in treated water was less than 0.5NTU. At the outlet of the holed tray aerator a lower variability in turbidity than in the Avellaneda trials was observed, with peaks of 2.5NTU. The tray aerator reached a better performance than the aerator used in the Avellaneda trials.

3.2.5. Ferrous iron and dissolved manganese

The concentration of dissolved manganese after the aeration stage was 95% of the total Mn, whereas the Fe was completely oxidized, similar to what was registered in the Avellaneda trials.

The high degree of dissolved manganese removal in the roughing filtration stage could well be attributed to biological processes since pH was much lower than that required for abiotic processes.

3.2.6. Roughing filter runs

The first run was finished in 17 weeks with no variation in treated water quality recorded. Since the sludge from the first backwashing contained a very low concentration of suspended solids, it was decided that the drainage rates be increased from 100 to 150 m/h. Likewise, in the second backwashing only a small amount of sludge was obtained, in spite of the backwash rate having been increased. The grains of gravel samples from the roughing filter bed were covered with black deposits, unlike the brownish ones observed on gravel grains from the Avellaneda roughing filter, which were principally composed of Fe deposits. Further investigations are needed to find out the suitable drainage rates and roughing filter runs for waters containing only Mn.

4. Conclusions

- Total Fe and total Mn removal efficiencies obtained by this system, which functions under natural conditions and without using any chemical agents, were very high, between 85 and 95%.
- Not only the high concentration of IOB observed in the raw water reservoir and in the aeration process, but also pH, Eh and DO values, suggest that Fe oxidation at these points of the treatment line could be attributed to a combination of biotic and abiotic

mechanisms. The Fe removal percentage of each mechanism is difficult to determine and was not the object of the present study.

- The concentration of dissolved manganese at the start of the treatment line in both pilot plants and the pH, Eh and DO results, combined with the detection of MnOB, go to indicate that the high Mn removal efficiency obtained was probably largely due to biotic processes.
- Up flow roughing filtration plays a significant role in the treatment line since a very high metal removal efficiency was achieved, removing simultaneously Fe and Mn in one step without requiring any sophisticated control of pH, Eh and DO. This represents an interesting option to existing patented systems of double rapid biofiltration.
- The following design parameters are recommended for the concentrations of Fe and Mn used in the trials.
  - Filtration rates: 2.5 m/h for roughing filtration, 0.45 m/h for slow sand filtration and 12 m/h for rapid sand filtration.
  - Filter runs: 30 days for roughing filter, 90 days for slow sand filtration and 15–30 days for rapid sand filtration.
  - Filter media: gravel 10–15 mm for roughing filtration and sand 1.2 mm for rapid filtration.

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