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Transformation of an existing physicochemical plant for iron and manganese removal by the application of biological processes

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ABSTRACT

A system for removing iron and manganese in groundwater, based on biofiltration technology, was developed at the Center of Sanitary Engineering of the National University of Rosario, Argentina. This process has been successfully applied in several new water treatment plants in Argentina. This paper describes the transformation of an existing plant for iron and manganese removal in groundwater from physicochemical to biofiltration processes. In the existing plant, iron and manganese removal was performed by a lime decarbonation process with the addition of four chemical products. A study was conducted to determine the feasibility of its transformation using biofiltration. The project included physicochemical and microbiological characterization of the water to be treated, a survey of existing facilities, pilot plant tests, and the design and construction of modifications. The operation of the transformed plant started in October 2011, and after the initial period of start-up, the concentrations of iron and manganese in treated water were below the values stated in the regulations. With the new scheme the use of chemicals was avoided, the operation was simplified and as a consequence the operating costs were reduced significantly.

Key words | biofiltration, groundwater, iron, iron bacteria, manganese, physicochemical process

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INTRODUCTION

Iron and manganese 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 (Ingallinella *et al.* 2001).

Several processes are available for the removal of iron and manganese: ion exchange, water softening, oxidation by aeration, chlorination or ozonation followed by filtration, aerated granular filter, adsorption in coated sand beds, subsurface removal and biofiltration, among others (Sommerfeld 1999; Islam *et al.* 2010; Knocke *et al.* 2010; Chaturvedi & Dave 2012; Salem *et al.* 2012).

Biofiltration processes are based on the properties of ‘iron bacteria’ present in groundwater, that catalyze the oxidation of both metals. In nature, iron oxidizing bacteria (IOB) and manganese oxidizing bacteria (MnOB) are widespread (Mouchet 1992). Biofiltration processes have numerous advantages over conventional physicochemical processes to remove iron and manganese. The main advantage is that the use of chemicals for the removal of metals is not necessary, which implies a reduction in process complexity and low operating costs (Burger *et al.* 2008b).

In the literature, different full-scale biofiltration plants have been reported. The systems described consist mainly of pressure rapid filter in one or more stages, with previous or intermediate aeration and/or pH adjustment (Czekalla *et al.* 1985; Badjo & Mouchet 1989; Mouchet 1992, 1995; Bourgine *et al.* 1994; Hedberg & Wahlberg 1998; Gage *et al.* 2001; Štembal *et al.* 2004; Sharma *et al.* 2005; Burger

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et al. 2008b). Agreement exists among the researchers that the mechanisms involved in the removal of iron and manganese consist of a combination of physicochemical and biological processes (Burger et al. 2008b; Tekerlekopoulou & Vayenas 2008). The basic design of treatment water plants is not identical when iron and manganese are present and it is generally not possible to achieve simultaneous removal of Fe^{2+} and Mn^{2+} in a single reactor except in some cases of extremely low filtration rates (Mouchet 1992).

A process to remove iron and manganese present in groundwater that uses the technology of biofiltration was developed at the Center of Sanitary Engineering, Faculty of Sciences and Engineering of the National University of Rosario (Ingallinella et al. 2001, 2002; Pacini et al. 2003, 2005). The process has been successfully applied in water treatment plants from different localities of Argentina since 2002 (Pacini et al. 2006). The treatment line comprises a stage of aeration, followed by two filtration steps: a roughing prefiltration in a gravel bed and a conventional rapid filtration step in a sand bed. The main difference with the processes described in the literature is the inclusion of the step of roughing biofiltration which works at low velocities (2.0–2.5 m/h), allowing simultaneous removal of iron and manganese, depending on the relative concentrations of both metals (Pacini et al. 2006).

This paper describes the transformation of an existing physicochemical plant for iron and manganese removal in groundwater to a new plant based on biofiltration process. The plant mentioned is located in northern Santa Fe Province, Argentina, with a population of 15,000 inhabitants.

Description of the existing physicochemical plant

The existing treatment plant was installed in 1997 at a rate of operation between 100 and 150 m^3/h, depending on the seasons of the year. To remove iron and manganese, a lime softening process was used. Iron and manganese were removed by increasing pH with lime to precipitate both metals without previous oxidation.

Treatment steps in the original scheme consisted of the addition of lime, polyaluminum chloride to coagulate the precipitates formed, and the addition of a polyelectrolyte as a flocculation aid. The high pH of treated water required final adjustment by acidification with carbon dioxide. Thus, four chemicals were used in total.

A scheme of the physicochemical process of iron and manganese removal with the original design is shown in Figure 1.

The plant is supplied with groundwater from ten perforations with iron and manganese concentrations ranging from 0.10 to 1.60 mg/L and 0.25 to 0.57 mg/L, respectively.

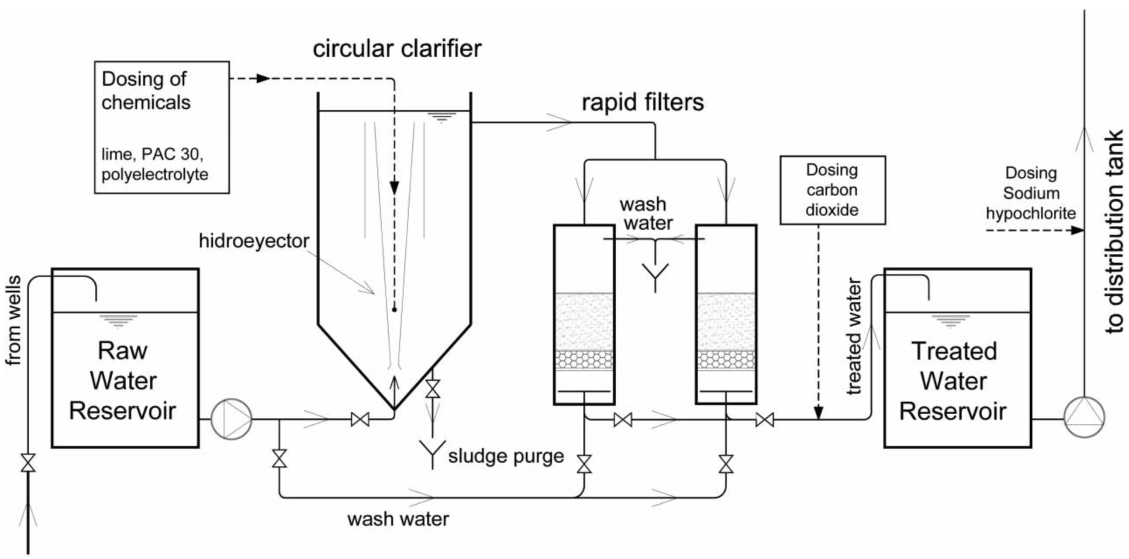

Figure 1 | Scheme of the treatment plant with physicochemical removal of Fe and Mn.

OBJECTIVE

The aim of this work was to upgrade the physicochemical existing water treatment plant into a biological plant for iron and manganese removal.

METHODS

The methodology used for transforming the existing plant into a biological one comprised the following steps:

1. Previous studies.
2. Proposal of modifications.
3. Start-up of full-scale plant after the implementation of proposed modifications.

Previous studies

Survey of existing facilities

A survey of the existing plant was performed in order to determine whether it could be used to implement the new process.

Physicochemical characterization of raw water

A preliminary study of raw water characterization was conducted collecting information, among other parameters, about the concentrations of total and ferrous iron and total manganese. In addition, determinations *in situ* of dissolved oxygen, pH, and redox potential were made.

Within the physicochemical characterization was included the determination of possible inhibitors of the activity of iron bacteria whose recommended limits are: zinc <0.10 mg/L, $SiO_2 < 50$ mg/L, in the case of biological removal of iron and NH_4^+ <0.10 mg/L for biological removal of Mn (Trauman 1997).

The analytical techniques used were as follows.

- pH and redox potential (Eh): electrometric method (HACH sensION1 Portable pH Meter, Model 50230 sensION electrode).
- Turbidity: Nephelometric method (HACH model 2100PTurbidimeter).
- Total Fe: Phenanthroline method, with the addition of sodium metabisulfite for total iron (HACH DR 4000 Spectrophotometer).
- Ferrous Fe: Phenanthroline method (HACH DR 4000 Spectrophotometer).
- Total manganese: PAN Hach Method 8149.
- Zinc (Zn): Atomic absorption spectrophotometric method.
- Total silica (SiO_2): Molibdosilicate method.
- Ammonia (NH_4^+): Nessler method.

Detection of iron bacteria in raw water

Given the difficulties involved in isolating IOB and MnOB in the laboratory, surface attachment collection methods (flow cell) described by Gariboglio & Smith (1993), Gariboglio (1996), and APHA-AWWA-WEF (2005) were utilized. The flow cell was used to collect bacteria in a raw water reservoir. Microscope slides from flow cell samples were examined with a trinocular microscope, Olympus BX40.

Standard test method for iron bacteria in water and water-formed deposits D932-85 (ASTM 2009) was used to identify the presence of iron bacteria in raw water. This test method covers the determination of iron bacteria by microscopic examination of its morphological characteristics. The method provides for the identification of the following genera of iron bacteria found in water and water-formed deposits: *Siderocapsa, Gallionella, Spherothilus, Crenothrix, Leptothrix*, and *Clonothrix*.

Pilot plant tests

A pilot reactor was installed beside the existing plant, to test the first stage of up-flow roughing filtration with the following objectives:

- To evaluate the possibility of using plastic rings as filter bed.
- To determine the possibility of using roughing filtration velocities higher than those adopted previously by the authors (from 2.5 to 5.0 m/h).

The pilot roughing filter consisted of a PVC pipe 0.315 m in diameter and 6 m in height. Two types of filter beds were tested: gravel and plastic rings, the latter with the aim of reducing the weight of the bed in the future roughing filter and trying to avoid structural reinforcements needed if gravel is used. Figure 2 shows a scheme of the pilot roughing filter that was operated with water from the raw water tank of the treatment plant.

Several configurations of filter bed were tested, including gravel, plastic rings, and combinations of both. In Figure 3, images of the materials used in the study are included. Table 1 shows different bed configurations, filtration rates used and frequency of washes.

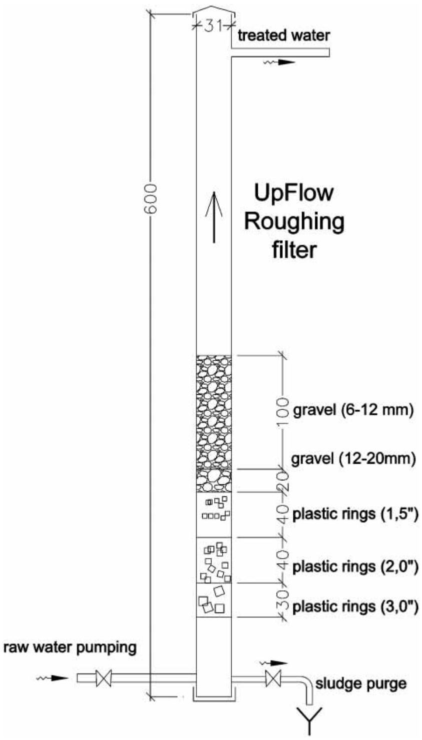

Figure 2 | Scheme of the pilot roughing filter and the filter layers (Steps 6 and 7, see Table 1).

As biological removal of iron is achieved in periods ranging from days to weeks, while manganese removal requires a maturing period between 2 and 6 months, the analysis of total iron concentration at the outlet of the reactor was chosen to check the removal efficiency of the different configurations.

Proposal of modifications of the existing treatment plant

The methodology for this stage was based on the results of previous studies in the pilot roughing filter. The final design of the modifications was done trying to take advantage of existing facilities.

RESULTS AND DISCUSSION

Previous studies

Survey of existing facilities

According to the dimensions of the reactors, the roughing filtration step could be implemented in the existing circular settler introducing the necessary modifications. Likewise, the final filtration step could be carried out in existing rapid filters without introducing any modification.

Physicochemical characterization of raw water

The results of total iron, ferreous iron, and total manganese concentrations in four samples of raw water are given in

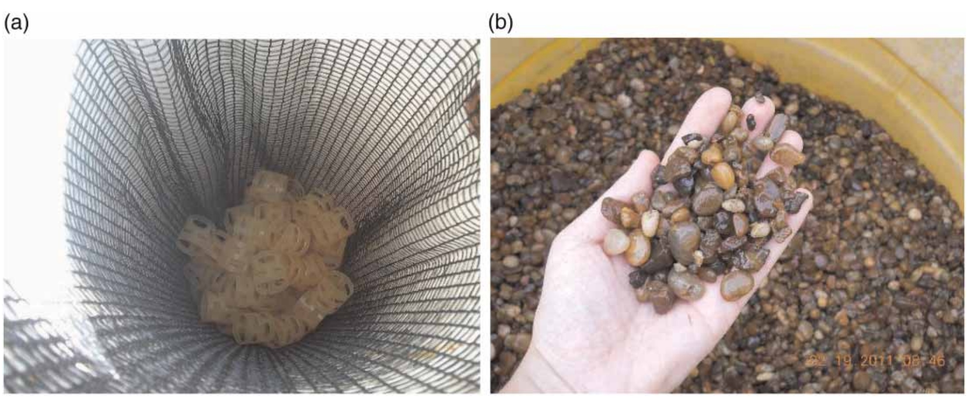

Figure 3 | (a) Plastic rings used in the pilot tests. (b) Filtering material (gravel) used in pilot roughing filter (Steps 5–7).

Table 1 | Trials configurations and filtration rates

Steps	Date	Filter media (height in meters)	Roughing filtration rates (m/h)	Frequency of washes
1	5/12/10 to 27/12/10	Plastic rings (pall type) (2.0 m)	4.50	Not washed
2	28/12/10 to 05/01/11	Plastic rings (pall type) (2.0 m)	4.50	Every 3 days
3	06/01/11 to 11/01/11	Plastic rings (pall type) (2.0 m)	2.75	Daily
4	11/01/11 to 21/01/11	Plastic rings (pall type) (3.0 m)	3.75	Daily
5	21/01/11 to 19/02/11	Gravel 6–12 mm (0.3 m-above)	3.75	Daily
		Plastic rings (2.0 m-below)		
6	20/02/11 to 17/03/11	Gravel 6–12 mm (1.0 m-above)	2.75	Daily
		Gravel 12–20 mm (0.2 m)		
		Plastic rings (1.1 m) only as support material (below)		
7	18/03/11 to 10/04/11	Gravel 6–12 mm (1.0 m-above)	4.50	Daily
		Gravel 12–20 mm (0.2 m)		
		Plastic rings (1.1 m) only as support material (below)		

Table 2. In Table 3, the physicochemical characteristics of two raw water samples are presented.

As can be seen in Table 3, the concentrations of potential inhibitors for iron and manganese removal (Zn, SiO_2 are lower than the recommended value (Trauman 1997).

Redox potential varied between 230 and 260 mV, pH ranged from 6.8 to 7.0, and dissolved oxygen concentration range was between 2.60 and 5.30 mg/L, depending on the level of water in the raw water tank. The measured values of redox potential and pH were suitable for the installation of a biological process (Mouchet 1992).

Detection of iron bacteria in raw water

Slides from flow cell with 7 days of exposure in the raw water tank were observed microscopically. The abundant presence of stalked bacteria (*Gallionella)* was detected (Figure 4) and there was a low presence of filamentous bacteria (Figure 5).

Table 2 | Total Fe, Fe^{2+}, and total Mn of water raw water reservoir

Date	Total Fe (mg/L)	Fe^{2+} (mg/L)	Total Mn (mg/L)
28/10/10	1.20	0.45	0.30
26/11/10	1.20	0.70	0.28
11/01/11	1.10	ND	0.46
19/02/11	1.25	ND	0.37

Table 3 | Physicochemical characterization of raw water

Parameter	Units	28/10/2010	26/11/2010
Color		4	<2
Turbidity	(UNT)	3.8	1.9
pH		7.0 (24 °C)	7.1 (27 °C)
Total solids	(mg/L)	940	1,000
Conductivity	(μS/cm)	1,600	1,700
Total hardness	($CaCO_3$ mg/L)	225	230
Calcium	(Ca^{2+} mg/L)	62	59
Magnesium	(Mg^{2+} mg/L)	17	21
Total alkalinity	($CaCO_3$ mg/L)	180	270
Chloride	(Cl^- mg/L)	200	260
Sulfate	(SO_4^{2-} mg/L)	170	200
Nitrite	(NO_2^- mg/L)	<0.005	<0.005
Nitrate	(NO_3^- mg/L)	<1	<1
Amonia	(NH_4^+ mg/L)	<0.05	<0.05
Total arsenic	(As mg/L)	<0.02	<0.02
Fluoride	(F^- mg/L)	0.30	0.40
Zinc	(Zn mg/L)	<0.05	–
Silica	SiO_2 (mg/L)	–	40

Pilot plant tests

Table 4 presents the average concentrations of total iron in water at the pilot roughing filter outlet at various steps of operation.

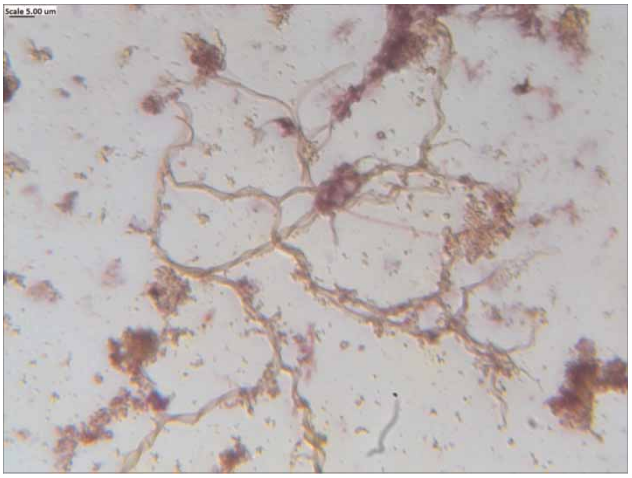

Figure 4 | Microscopic observation of stalked bacteria (*Gallionella)* in raw water (bar 5 μm).

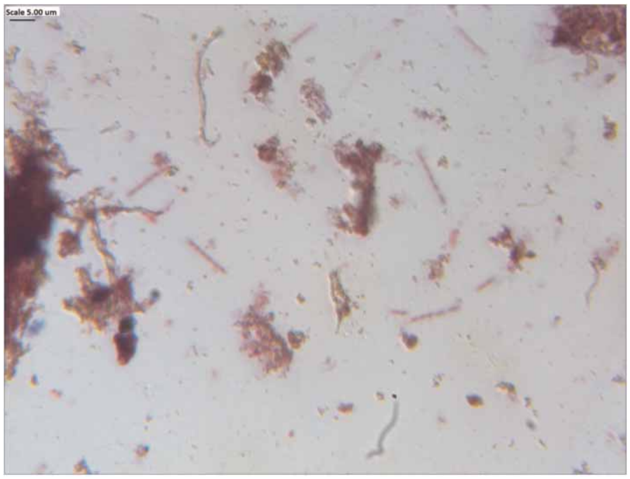

Figure 5 | Microscopic observation of filamentous bacteria in raw water (bar 5 μm).

Table 4 | Average concentrations of total Fe in the pilot roughing filter outlet and efficiencies

Steps	Average total Fe (mg/L)	Average removal efficiencies (%)	Standard deviation removal efficiencies (%)	N (number of samples)
1	0.59	51	8 (32–64)	16
2	0.48	60	10 (47–70)	8
3	0.49	59	14 (41–74)	4
4	0.47	64	11 (41–83)	16
5	0.45	63	5 (51–73)	26
6	0.23	81	5 (68–86)	26
7	0.18	87	3 (76–91)	23

The conclusion after analyzing the results of pilot plant tests was that the most suitable configurations for the biological roughing filter bed were those used in Steps 6 and 7 (layer of gravel). These configurations had greater efficiency in the removal of total iron with less variation. In these configurations, the layer of plastic rings only served as supporting the gravel layer. The higher efficiency obtained could be attributed to the greater surface area of the gravel layer (about 600 m^2/m^3) versus that offered by plastic rings (120–150 m^2/m^3). Greater biofilms were developed on the gravel and, therefore, there were more sites where iron and manganese could be bio-oxidized and precipitated. Moreover, in Step 7, roughing filtration rate was higher than usually applied in treatment plants already projected by the authors (Pacini *et al*. 2006), and removal efficiencies of total iron were not affected.

Project of modifications

Taking into account the results obtained in previous studies, the transformation project was developed trying to use most of the existing facilities. The sequence adopted was the following: aeration, up-flow roughing filtration in a gravel bed, and rapid sand filtration without the addition of chemicals (Figure 6).

The treatment plant was taken out of operation in late August 2011 and several changes were made as explained below.

A perforated tray aerator with plastics rings was placed before the raw water tank (Figure 7(a)).

The transformation of existing circular settler in an up-flow roughing filter included the following actions: dosing system and chemical mixing were eliminated (Figures 7(b) and 8), structural reinforcement was implemented, and a steel false bottom was built. The inside of the reactor was coated with plastic reinforced with glass fiber.

A layer of gravel of 1 m in height and 6–12 mm grain size was placed above a supporting bed of gravel 12–50 mm grain size (Figure 9(a)). Plastic rings were not used (as in Steps 6 and 7 of pilot roughing filter) because of the high cost and probably because they could not hold their shape over time due to the significant weight of the upper bed of gravel. Two bottom drain valves (250 mm in diameter) were added for cleaning the roughing filter by bottom discharges.

An inspection of rapid filter beds was conducted to determine the existence of mud balls. During the inspection

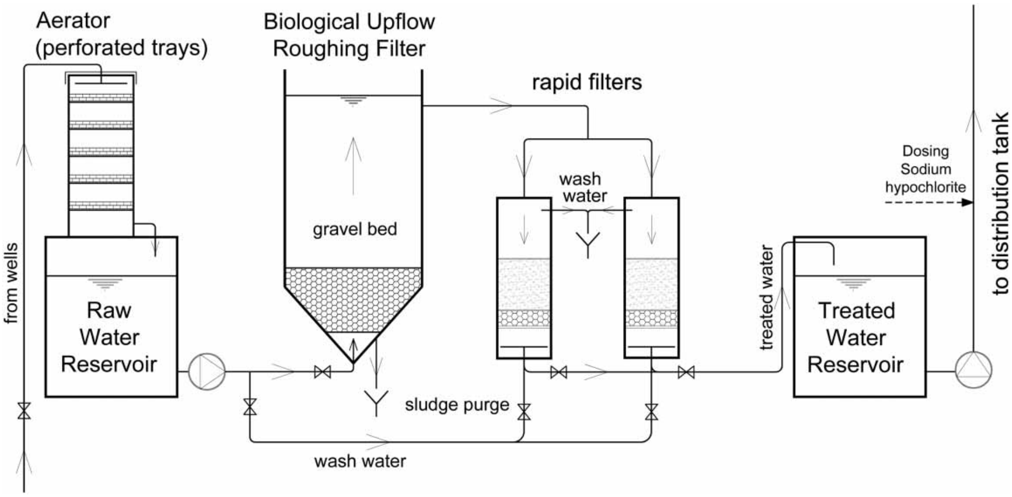

Figure 6 | Scheme of biological upgraded water treatment plant for Fe and Mn removal.

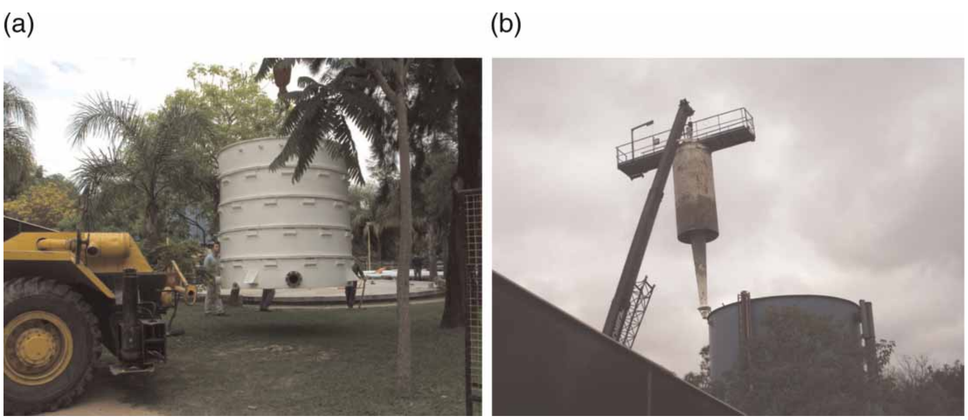

Figure 7 | (a) Incorporation of perforated trays aerator. (b) Retirement of hydro-ejector (incoming system of mixing chemicals at existing circular settler).

it was found that the mantles were in good condition. It was observed that sand grains were coated with MnO_X precipitates (Figure 9(b)). The existing sand bed was not replaced. Gauges were incorporated to measure pressure drops.

The entire project includes the construction of a settler to recover wash water with a total volume of 160 m^3 and the construction of two sludge drying beds of 25 m^2 in total area. These facilities will be constructed in a second stage.

Start-up of the modified plant

Operation of the modified plant began on October 31, 2011, and at present is treating 100 m^3/h in winter and 150 m^3/h in summer. Removal efficiencies of total Fe and total Mn obtained in the up-flow roughing filter and rapid filter during the first 20 months of plant operation are shown in Figures 10 and 11, respectively.

The percentage of removal efficiency was calculated as:

$$\left(\frac{C_{\text{inlet}} - C_{\text{outlet}}}{C_{\text{inlet}}}\right) \cdot 100$$

where C_{intet}: concentration of total Fe or total Mn at the system inlet, C_{outlet}: concentration of total Fe or total Mn at the system outlet.

Figure 10 shows the total iron removal efficiency. The maturation time for iron removal was very short. Concentration of iron in treated water was 0.02 mg/L within 24

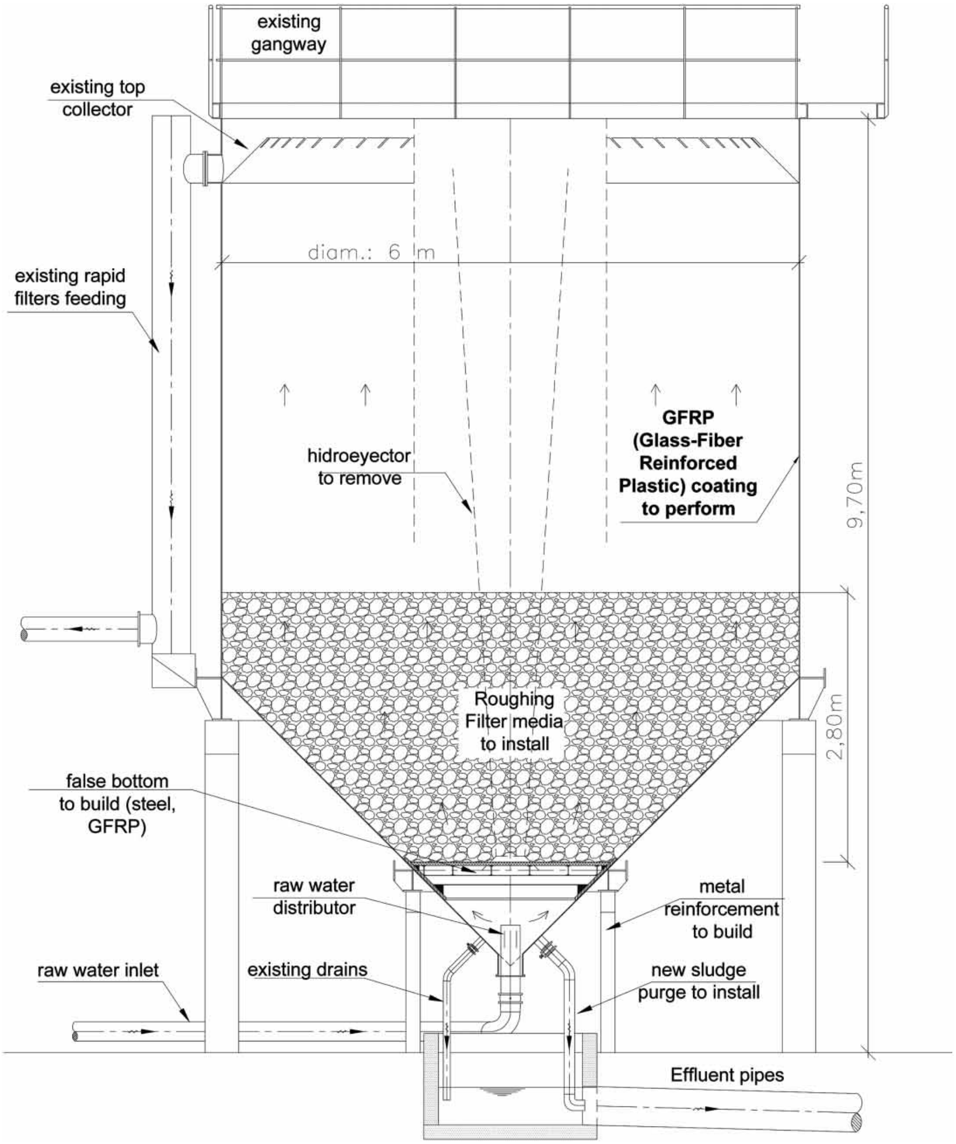

Figure 8 | Changes made in the existing circular settler to transform it in an up-flow roughing filter.

hours of operation. Concentration of Mn in treated water was 0.045 mg/L after 15 days of operation (Figure 11). The mentioned values were below those recommended in current legislation in Argentina (0.10 mg/L for iron and 0.05 mg/L for manganese).

The maturation time for Mn removal is also very short when compared with those times reported in the literature. According to studies (Mouchet 1992; Ingallinella *et al.* 2001, 2002; Pacini *et al.* 2005) the maturation time, by natural spontaneous process, required to achieve maximum removal of Mn by a biological process ranges from 2 to 6 months because manganese specific bacteria need an acclimation period greater than iron bacteria.

Inoculation experiences using backwash water or support material from other well-established filters previously used for simultaneous iron and manganese removal to reduce the start-up period of new filters are reported in the literature (Hope & Bott 2004; Štembal *et al.* 2004; Li *et al.* 2005; Burger *et al.* 2008a, b). Štembal *et al.* (2004) reported that the removal of manganese began approximately 2 weeks after the

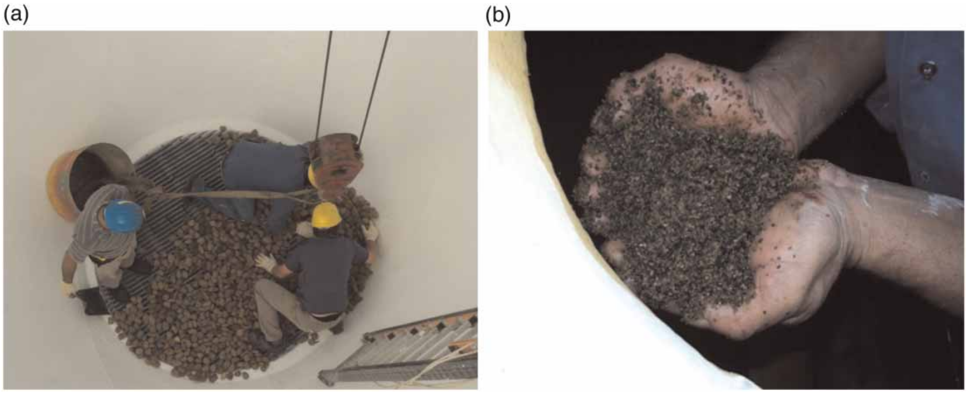

Figure 9 | (a) Placing gravel in roughing filter (previous circular settler). (b) Filtering material covered with manganese precipitates in rapid filter.

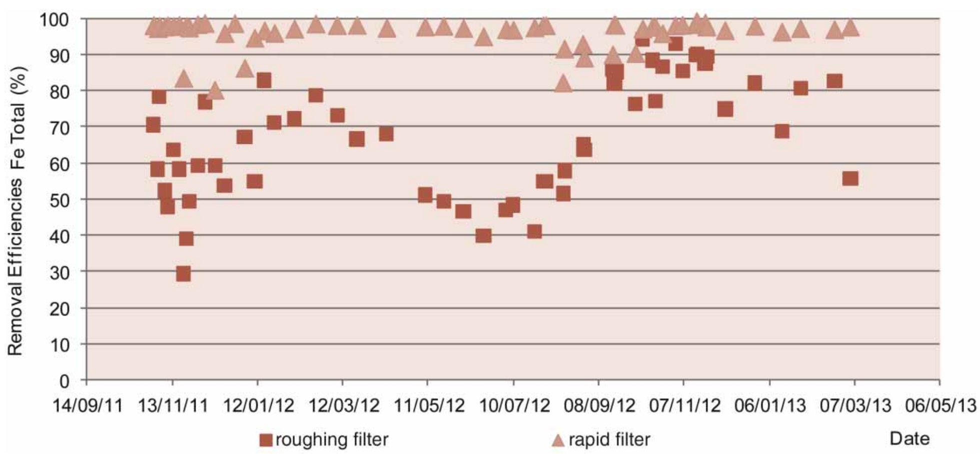

Figure 10 | Total Fe removal efficiencies at the outlet of each stage of treatment plant.

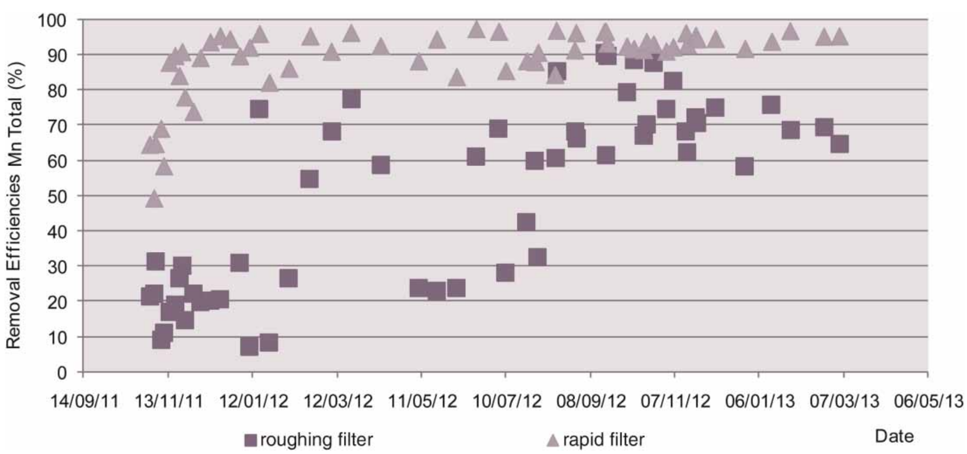

Figure 11 | Total Mn removal efficiencies at the outlet of each stage of treatment plant.

initial inoculation of the filter with sludge from another plant. During the inoculation period, the plant mentioned worked with 50% of the designed hydraulic load.

In this case, as was mentioned, high levels of Mn removal were reached in 15 days without inoculation and working at full hydraulic load. This was attributed to the fact that the sand of rapid filters was not replaced and the grains were coated with MnO_X precipitates, which acted as catalysts in the oxidation/precipitation of Mn (Stumm & Morgan 1996).

It is known that highly effective manganese removal could be achieved by maintaining an active manganese dioxide (MnO_X(s)) surface on the filter media that adsorbs soluble Mn. However, this process requires the presence of free chlorine to regenerate the media (Knocke *et al*. 2010). On the other hand, Burger *et al*. (2008b) established that physicochemical mechanisms of Mn removal do take place in the form of adsorption of Mn^{2+} to Mn oxides present in biological filters. The authors mentioned indicate that further research should address the potential pathways and factors affecting biological oxidation or adsorption in biofiltration systems, and the potential role that MOB has in allowing the system to achieve autocatalytic operation without the addition of strong oxidants.

Figure 11 shows that manganese removal efficiency in the roughing filter increased with time. A similar behavior was observed during pilot plant tests performed previously by the authors (Pacini *et al*. 2003). A noticeable decrease in removal efficiency of iron and manganese in the roughing filter during winter months was detected, although total removal efficiency did not change.

The amount of sludge generated before transformation was estimated at about 275 g/m^3 (total fixed solids per m^3 of water produced). After transformation, this amount was reduced to 2 g/m^3.

The operation costs of the transformed plant decreased significantly because the use of chemicals was eliminated (saving 7,000 US$/month, October 2011), the raw water used for washing was reduced by 69% (before transformation 540 m^3/week were used and after decreased to 170 m^3/week), and the man-hours assigned to the tasks of operation plant were also reduced by 25%. In total, it is estimated that the operating cost per m^3 of water produced was reduced by 60% (from 0.46 US$/$m^3$ to 0.28 US$/$m^3$). These results are consistent with those reported by Mouchet (1995), who noted that operating costs were reduced by 50–80% when plants were transformed from physicochemical to biological processes. The investments needed to implement the modifications in the existing plant were about 60,000 US$ (October 2011), corresponding to 25% of the cost of a new biofiltration plant. It was estimated that the upgrade investment was amortized in 1 year.

CONCLUSIONS

We demonstrate the advantages of transforming physicochemical plants into biological plants for iron and manganese removal if the physicochemical and microbiological characteristics of raw water are adequate as follows:

- Inoculation with sludge of existing treatment plants was not necessary because in 15 days the desired removal was achieved in the second stage of filtration (rapid filters), probably because the original sand with deposits of MnO_X was not removed.
- The operating costs were significantly reduced, the operation of the plant was simplified, and the risks involved in handling chemicals were avoided.
- The environmental impact of sludge disposal was minimized as no chemicals were used.
- Transformation of plants from physicochemical to biological treatment could be accomplished by taking advantage of existing installations without major investment.

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