

Removal of Manganese and Iron from Groundwater by using Chlorination and Rapid Sand Filtration: A Case Study

M. E. Sutharsan, S.P.S. Meegahakumbura and N.S. Miguntanna

Abstract: The average groundwater extraction from Murunkan well field, Mannar, is around 3.4 MCM/year, and it was triggered by extensive development projects and resettlements that took place in the last decade. Manganese and Iron variations in Murunkan groundwater reported during some periods created various operational and aesthetic issues in the potable water supply. This study is focused on treating iron and manganese in groundwater using chlorination and Rapid Sand Filtration (RSF) techniques. Pebble layers, height of 205 mm with the particle sizes ranging from 2-25 mm, were filled at the bottom of RSF model filter. Filter media, height of 700 mm with 0.45 mm effective size (D_{10}) and a uniformity coefficient (C_u) of 1.33, was placed above the pebble layers. The filter was operated continuously for 15 days, and influent and effluent water samples were collected from 15 trials. Filter backwashing was performed after every two trials to maintain a consistent filtration rate. Raw water pH values ranged from 6.74 – 7.48. Chlorine solution (1.08-1.26 ppm) was added at the inlet point, and a 2.88 m water column was maintained, allowing adequate mixing and oxidation time. The results revealed that average removals of iron and manganese as 60% and 73%, respectively, while maximum removals of 93% and 91% were observed for iron and manganese, respectively.

Keywords: Chlorine, Groundwater treatment, Iron removal, Manganese removal, Oxidation

1. Introduction

Water scarcity has become a serious issue currently all over the world, [1]. The dry zone of Sri Lanka is experiencing water deficiency for several months while the wet zone is experiencing an excessive amount of water. Pipe-borne water supply coverage of Northern Province is just over 12%, and most people in the Province merely rely on groundwater sources such as dug wells and tube wells to fulfil their daily water requirements [2]. Mannar district is located in the Northern Province of Sri Lanka where there are no perennial rivers. People in Mannar fulfil their drinking water needs entirely from groundwater sources as there are no reliable surface water sources which can be utilized for water supply. National Water Supply & Drainage Board (NWS&DB) of Sri Lanka provides pipe borne water to the residents in Mannar Town, Nanattan and Manthai West Divisional Secretariat Divisions of Mannar district from the Murunkan water supply scheme [15].

Six major types of groundwater aquifers are available in Sri Lanka and Murunkan aquifer is one of the most utilized aquifers in the dry zone, which is characterized as a deep confined aquifer of these sedimentary limestones and

sandstone formations [7]. The alluvial sandstone is vastly cracked and it splits up the geological formation into a series of secluded units, thus creating a set of separate groundwater sinks in the Miocene limestone belt of the North-Western coastal zone [7]. Groundwater intakes of NWS & DB in the Murunkan aquifer have been serving the people of Mannar district for a few decades, and currently, it supplies approximately 40% of the potable water demand of Mannar district. The average potable water extraction from Murunkan groundwater intake wells is around 3.4 MCM/year [15]. Although iron and manganese generally occur at concentrations less than 5 mg/l and 1 mg/l, respectively, in

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groundwater [4]. In the recent past, variation of manganese and iron was reported in Murunkan groundwater in certain periods of the year [15]. Iron and manganese do not pose any health hazards whereas they indeed create acceptability issues in drinking water [3,5]. Hence, secondary standards have been set for iron and manganese. The secondary (aesthetic) Maximum Permissible Limits (MPLs) of Sri Lankan drinking water standards (SLS 614: 2013) for iron and manganese are 0.3 mg/l and 0.1 mg/l, respectively. Iron and manganese variations of Murunkan groundwater, on few occasions, have not complied with SLS 614:2013 standards and created various operational, and aesthetic issues and customer dissatisfaction. Exceeding the recommended maximum permissible limits typically leads to taste and odour problems, stained water, tints on clothes, and bath ware fittings, discolouration of industrial products and clog pipes, and promotes the growth of gelatinous masses of iron bacteria. This will cause raising public complaints and general displeasure with the water utility. Hence, finding effective solutions for elimination of iron and manganese from the water extracted at Murunkan wells before distributing to customers is essential at the moment.

Although there are several methods available in practice for the removal of manganese and iron from the smaller scale of water treatment, very few studies have been done to remove iron and manganese from the larger scale water supply facilities. Even though the selected region is hydrogeologically significance, inadequate studies and investigations have been performed to date. Hence, finding efficient methods for the removal of manganese and iron from groundwater in the perspective of treating considerable high-water quantity, would be a sustainable solution for ensuring a customer-satisfied potable water supply service. Thus, the objective of the pilot study is to treat iron and manganese in groundwater using chlorination and Rapid Sand Filtration (RSF) techniques.

2. Literature Review

2.1 Groundwater Sources in Sri Lanka

Sri Lanka is a humid tropical island located in the equatorial region. The climatic and hydrogeological features of Sri Lanka depend on Indian Ocean monsoons which play a crucial role in the recharge of groundwater aquifers.

Sri Lanka principally lies beneath high-grade metamorphic rocks with distributed alluvial formations [6]. Groundwater is considered more consistent as the impact of chronological change is negligible. There are six main types of groundwater aquifers that have been recognized and categorized in the country considering their morphological and hydrogeological settings [7].

Although there are 103 river basins of different sizes scattered in the country, groundwater plays a significant part as a main potable water source, especially for the dwellings in the Northern Province. Sri Lanka experiences severe water shortages in the dry zone of the country for months while excess water is available in the wet zone [6].

Over 90% of the country lies beneath high-grade metamorphic rocks [8]. Thus, hard rock aquifers are more abundant. Deep confined aquifers are scattered along the North Western coastal plain [7].

There are eight isolated deep confined aquifers along the North Western coastal belt, namely, Palavi, Madurankuliya, Vanathavilluwa, Silawatura, Murunakn, Mulankavila, Paranthan and Mullaitivu [6]. They are the most utilized aquifers in the country so far where the surface water is scarce in the vicinity. Mannar Island is located in the arid region of Sri Lanka where there are no perennial rivers. The drinking water demand of Mannar district is solely catered by groundwater sources. Murunkan, which is identified as one of the major isolated hydrogeological basins in the Miocene limestone belt of the North-Western coastal zone, is dominant in providing potable water to the community.

2.2 Sources and Issues of Manganese and Iron

Iron (Fe) and Manganese (Mn) are similar and common heavy metals that can be observed in natural resources like soil, rock, and minerals. When water infiltrates through the earth's crust it liquefies iron and manganese [13]. Liquefied compounds reach groundwater eventually. Surface water is regularly mixed with air and oxidizes the available iron and manganese into non-soluble compounds. Hence, usually, surface water contains very less concentrations of iron and manganese. Other than the natural causes, anthropogenic activities such as industrial and agricultural disposals are major

causes of adding iron and manganese into water bodies. Dissolved Oxygen (DO) is the predominant factor that decides the degree of dissolving of iron and manganese in the water. In addition, the nature of water (alkalinity or acidity) will also trigger the liquefaction of iron and manganese.

When organic matter is present in groundwater, BOD requirement is higher, and available DO in the water is lesser, as the organic matter consumes DO for decomposition. Besides, DO content becomes significantly less as the aquifer runs deeper. Hence, iron and manganese are present in the dissolved form (as Fe^{2+} and Mn^{2+}). If groundwater is pumped out and reacts with air, Fe^{2+} will be oxidized to Fe^{3+} forming a reddish-brown precipitate while Mn^{2+} oxidized to Mn^{4+} forming a blackish precipitate as shown in Eq. (1). Besides the dissolved form, there are two more forms of iron and manganese present in water, namely, particulate and colloidal. Iron and manganese are oxidized in particulate form while very tiny particles are suspended in the water in colloidal form.

Table1 depicts the possible indications when iron and manganese are present in water in the above three forms.

Table 1 - Available Forms of Iron Manganese in Water and their Indications

Indication	Form of iron & manganese
Water is clear and red-brown or black particles appear as the water stands	Dissolved Iron (Fe^{2+}) & Manganese (Mn^{2+})
Water contains red-brown or black particles and settles out as the water stands	Oxidized Iron (Fe^{3+}) & Manganese (Mn^{4+})
Reddish or black colour that remains longer than a day	Colloidal iron & manganese

Dissolved iron or manganese is present, if red or black particles are settled after some time in the bottom of a water glass that is initially clear [13]. However, if water still has a black or red tint that lasts more than a day, it is apparent that colloidal iron or manganese is present in the water. Neither iron nor manganese in potable water causes a serious threat to the well-being of water users. However, there

could be a risk of causing health hazards if the users suffer from hemochromatosis [9].

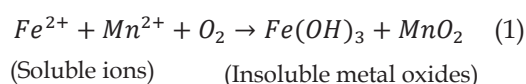
Though iron and manganese are not health hazardous, they create a lot of aesthetic problems at the household level when they reach consumers through the water supply. The presence of iron and manganese in water can cause tangible problems such as notable taste, odor, and stained colour [14]. Furthermore, it may lead to reddish-brown or brownish-black tints, mostly in laundry and bathware tools. In addition to the reddish-brown or brownish-black, slime is formed in toilet tanks by iron manganese bacteria. Iron manganese bacteria feed on iron and manganese and naturally occur in soil and shallow groundwater. Although these bacteria are not health threatening, they create a musty or swampy smell in the water.

2.3 Current Treatment Processes of Iron and Manganese

There are several methods in practice for removing iron and manganese, such as aeration followed by filtration, chemical oxidation followed by filtration, sequestration, and ion exchange water softeners. Sequestration is adding chemicals to water to minimize the problems due to iron and manganese without removing them from the water. Usually, polyphosphates are used and added to groundwater before contact with air or any other oxidants. Thus, it keeps iron and manganese in soluble form. Iron and manganese are not removed from groundwater by this method but are stabilized and dissolved [13, 14]. Hence, water will be clear and prevent staining laundry but the offensive metallic taste remains unchanged. Nevertheless, this process is not recommended when water is boiled. Therefore, sequestration using polyphosphate does not fit to treat potable water.

Ion exchange is also an effective method for the removal of iron and manganese when they present in low concentrations. The dissolved iron and manganese in water can be replaced with sodium ions and exchanged resin or zeolite [9].

Aeration followed by filtration is a quite popular method as it is economical. Dissolved iron (Fe^{2+}) and manganese (Mn^{2+}) ions will be oxidized by air to their insoluble forms $Fe(OH)_3$ and MnO_2 , respectively. The reaction that takes place is in the form of:



Chemical oxidation is effective when the water has higher concentrations of Fe^{2+} and Mn^{2+} according to the explanation of Mark et al. [14]. Chemicals such as Chlorine (Cl_2), Potassium permanganate (KMnO_4) and bleaching powder ($\text{Ca}(\text{ClO})_2$) can also be used as oxidants. Chlorine oxidizes iron best at a pH of 6.5 to 7.5, but manganese requires a pH higher than 9.5 for complete oxidation [14]. Chemical oxidation will also result in insoluble metal oxides which could be removed by filtration.

Various types of filter media are available and effectively used in removing iron and manganese. Manganese greensand, anthra/sand or iron-man sand, silica sand, electromedia, and ceramic are the commonly used filter media. Glauconite, which has ion exchange properties, is the active material in "greensand" [9].

As water infiltrates via the filter, dissolved iron and manganese are dragged out and oxidized to form metallic oxides. Thus, green sand is more effective than silica sand as a filter media.

Iron manganese deposits are built up over the filter media after running the filter. Then the system should be backwashed to flush out the deposits. Regular backwashing is critical for maintaining a consistent filtration rate.

Although slow sand filter (SSF) is more efficient for filtration and cost-effective, it has a very slow filtration rate of approximately $0.1\text{--}0.4 \text{ m}^3/\text{m}^2/\text{h}$ whereas the rapid sand filter (RSF) has a filtration rate of $5\text{--}15 \text{ m}^3/\text{m}^2/\text{h}$ [4].

2.4 Chemical Oxidation followed by Filtration

Chemical oxidation using oxidants such as chlorine (Cl_2), potassium permanganate (KMnO_4), and bleaching powder ($\text{Ca}(\text{ClO})_2$), followed by filtration is the common method used for manganese and iron removal as the process in the context of smaller scale water treatment. The reaction of oxidization of Fe^{2+} and Mn^{2+} is highly affected by the pH/Eh environment [10]. In addition, Labhasetwar [11], had compared the effectiveness of using chlorine and potassium permanganate (KMnO_4).

3. Materials and Methods

Chemical oxidation using chlorine followed by RSF was adopted as the treatment method considering the cost-effectiveness, complexity

of the treatment process, filtration rate, high water quantity ($9,500 \text{ m}^3/\text{d}$), and spatial availability of the intake site. Consequently, the experimentally based study was implemented to investigate the iron-manganese removal efficiency using an RSF with chemical oxidation using chlorine.

3.1 Filter Arrangement and Selection of Filter Media

Filter model was constructed using a 160 mm nominal diameter uPVC pipe of 4 m length. Ordinary silica sand was sieved and a particle size distribution curve was obtained as depicted in Figure 1.

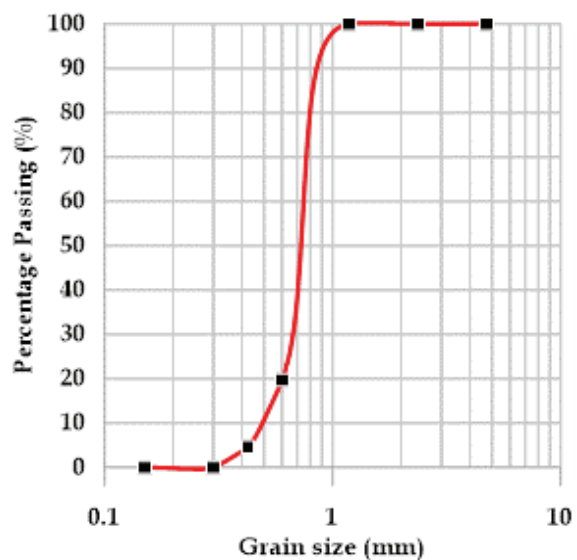


Figure 1 - Particle Size Distribution Curve of Filter Material

Pebble stones of 25 mm, 16 mm, 10 mm, and 2-5 mm size were placed as 75 mm, 50 mm, 30 mm, and 50 mm thick layers, respectively. The recommended limits for the effective size (D_{10}) and uniformity coefficient (C_u) of filter media in RSF is between 0.4-1.2 mm and 1.2-2.0, respectively [4]. Thus, filter media with 0.45 mm effective size (D_{10}) and a uniformity coefficient (C_u) of 1.33 was placed until it reached 700 mm height in the filter as shown in Figure 2.

The filter inlet, filter outlet, and backwashing inlet were connected by pipelines with valve arrangements as depicted in Figure 3. Raw water is conveyed from the collecting chamber where the water from all the boreholes is mixed in. The water column above the filter top was maintained at around 2880 mm in height in order to maintain an oxidation time at a minimum of 20 minutes as per the recommendations [11]. A separate pipeline was

provided from the chlorine tank to add chlorine for oxidation. Chlorine solution was added on top of the water column allowing the required oxidation time. Samples were collected from raw water and treated water for 15 no. of trials and laboratory tests were carried out to check the selected physical and chemical parameters based on standard methods of APHA [3]. Manganese (Mn^{2+}) and Iron (Fe^{2+}) concentrations of collected water samples were determined using a Spectrophotometer (HACH DR 5000). The pH values were measured using a pH meter (HACH pH1).

3.2 Oxidation using Chlorine

Oxidation contact time was maintained between 22.2-25.1 minutes during this study. The chemical oxidation contact time was kept to a minimum of 20 minutes for better oxidation [11]. More contact time shall be provided if the water contains colloidal manganese or colloidal iron.

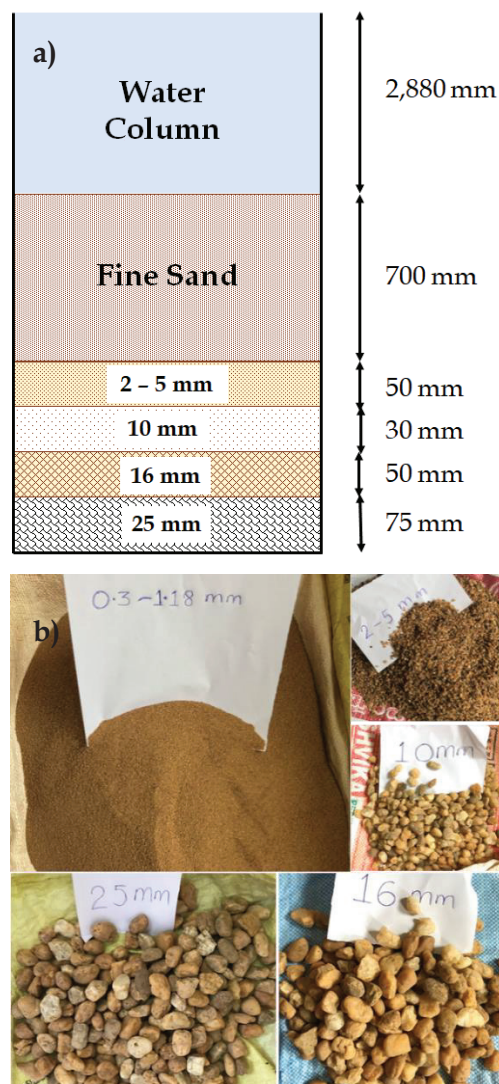


Figure 2 - Filter Model a) Filter Layers
b) Filter Media

Chlorine solution which has an average concentration of 1.18 mg/l was added on top of the water column and oxidation time was maintained between 22.2-25.1 minutes.

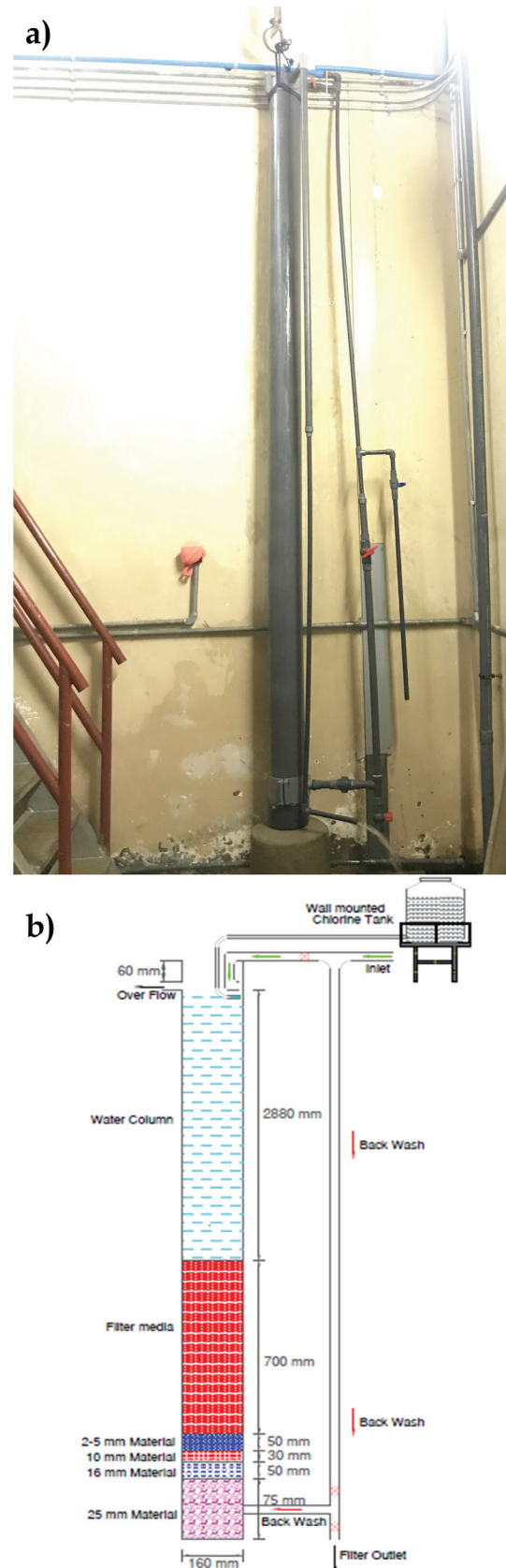


Figure 3 - a) Filter Model, b) Schematic Diagram of Filter Model Arrangement

The oxidation time and concentration of chlorine for 15 trials are tabulated in Table 2. The recommended filtration rate for RSF is 5-15 m³/m²/h [4] and a constant filtration rate of 7.46 m³/m²/h was maintained during the experiment. Nevertheless, the clear water head loss was found to be 3.05 m whereas the recommended head loss is up to 2.5 m [4]. The filter model was in operation continuously throughout the day for 15 consecutive days to evaluate the actual performance of the filter when using this methodology. The pre-chlorination and oxidation processes took place above the filter media within the filter model. Untreated water and treated water samples were safely collected in containers for 15 trials.

The concentration of iron and manganese and pH value were measured for both untreated and treated water samples. Water quality tests were performed based on APHA [3], at Regional Laboratory, National Water Supply & Drainage Board, Vavuniya, Sri Lanka.

Table 2 - Oxidation Time and Chlorine Concentration used for Trials

Trail No	Oxidation time (min)	Chlorine Concentration (mg/l)
1	22.7	1.26
2	23.2	1.16
3	22.7	1.14
4	23.2	1.09
5	22.2	1.15
6	22.4	1.18
7	22.9	1.15
8	24.8	1.23
9	22.3	1.08
10	22.7	1.20
11	22.3	1.15
12	25.1	1.26
13	22.8	1.17
14	22.2	1.24
15	22.6	1.23

4. Results and Discussion

4.1 Chemical Characteristics of Groundwater before and after Filtration

The iron concentration of raw water samples ranged between 0.00 – 1.47 mg/l (SD = 0.47) and 13% of raw water samples exceeded the maximum permissible limit specified in the SLS 614:2013. The iron concentration was in the range of 0.00-1.0 mg/l (SD = 0.28) in treated water samples. Typically, iron stains in the plumbing fixtures and laundry at a

concentration above 0.3 mg/l. The maximum manganese concentration was noticed to be less than the maximum iron concentration. Manganese concentration of raw water was found between 0.08-0.72 mg/l (SD = 0.16) while treated water values ranged between 0.01-0.27 mg/l (SD = 0.09). The maximum removal efficiency of iron and manganese was found to be 93% & 91%, respectively. Further, iron removal was observed in 60% of the samples, whereas removal of manganese was observed in 73% of the samples.

It is observed that, in the first two trials, iron and manganese concentrations in the treated water sample were higher than the concentration in the raw water sample. The dissolving of inorganic solids and ions from filter media may be the possible reason for the increasing manganese and iron existence in the treated water sample for the first couple of days. All the raw water samples are slightly alkaline, yet pH values were compiled with SLS 614:2013 standards. Table 3 describes the observed values of pH and iron-manganese concentrations for 15 trials. Although the existence of manganese and iron in water is not health hazardous, it may create aesthetic problems at the household level. According to the SLS 614:2013 standard, the secondary (aesthetic) maximum permissible levels for manganese and iron are 0.1 mg/l and 0.3 mg/l, respectively, in potable water and these limits are considered to be safe limits. The maximum removal of iron is 93% (Iron concentrations in raw water and treated water are 0.27 mg/l and 0.02 mg/l, respectively) while the maximum removal of manganese is 91% (manganese concentrations in raw water and treated water are 0.13 mg/l and 0.01 mg/l, respectively). Figure 4 depicts the variation of manganese and iron concentrations for 15 trials. It is critical to consider how pH affects the elimination of iron and manganese.

4.2 Principle of Chlorination

Iron and manganese removal incorporates in two stages: (a) an oxidation process - insoluble precipitates are formed by the dissolved manganese and iron oxidation (b) a solid separation process-removal of the precipitated material from the water stream. Chlorination plays a significant role during the above first stage. Chlorine has the potential of killing iron bacteria and any other disease-causing bacteria that may be present in water, while chemically oxidizing the iron or manganese forming a non-soluble particle. The oxidation of

manganese and iron is governed by contact time, chlorine concentrations, and pH value [11]. Hypochlorous acid (HOCl) and hydrochloric acid (HCl) form when chlorine reacts with water by a reversible hydrolysis reaction. Hypochlorous acid is a weak acid and may dissociate into hypochlorite ions (OCl⁻) and hydrogen ions (H⁺). Both hypochlorous acid and hypochlorite ions are included in the free available chlorine which can react with after chlorination occurs. The recommended residual chlorine concentration should be 0.5-0.8 mg/l when treated water reaches the clear water tank [4]. Further, there is a potential to form trihalomethanes (THM) when sufficient levels of chlorine are in contact with organic matter. THM formation becomes disinfectant limited, within the network, when the free chlorine residual typically drops to 0.3 mg/l [4].

dissolved ions and organic compounds which may be present in water.

Nevertheless, according to the study results of Labhasetwar [11], iron and manganese removal was not remarkable at lower doses of chlorine. The removal efficiency of iron and manganese could be increased by increasing chlorine concentration, but it is most important to consider the residual chlorine concentration THMs are potentially carcinogenic to humans. Granular Activated Carbon (GAC) filters are required for the effective removal of THMs.

4.3 Backwashing

After the first two trials, the filtration rate reduction was observed and the turbidity of the

Table 3 - Summary of observed Values for 15 trials

Trial No	pH @ 25 °C		Fe Concentration (mg/l)		Mn Concentration (mg/l)	
	Raw water	Treated water	Raw water	Treated water	Raw water	Treated water
1	7.24	7.25	0.02	0.03	0.09	0.12
2	6.89	6.92	0.01	Undetected	0.11	0.12
3	6.86	6.94	0.01	Undetected	0.12	0.02
4	6.85	6.98	0.01	Undetected	0.12	0.03
5	7.22	7.22	Undetected	0.04	0.10	0.26
6	7.22	7.21	0.06	0.03	0.11	0.10
7	7.22	7.19	0.06	0.05	0.11	0.04
8	7.08	7.17	0.27	0.02	0.15	0.08
9	7.36	7.41	Undetected	0.04	0.11	0.08
10	7.48	7.53	0.08	0.06	0.13	0.01
11	7.14	7.20	Undetected	0.04	0.22	0.15
12	6.74	7.10	Undetected	0.31	0.08	0.16
13	7.37	7.39	0.99	1.00	0.32	0.27
14	7.33	7.35	0.07	0.05	0.22	0.09
15	7.44	7.37	1.47	0.46	0.72	0.27

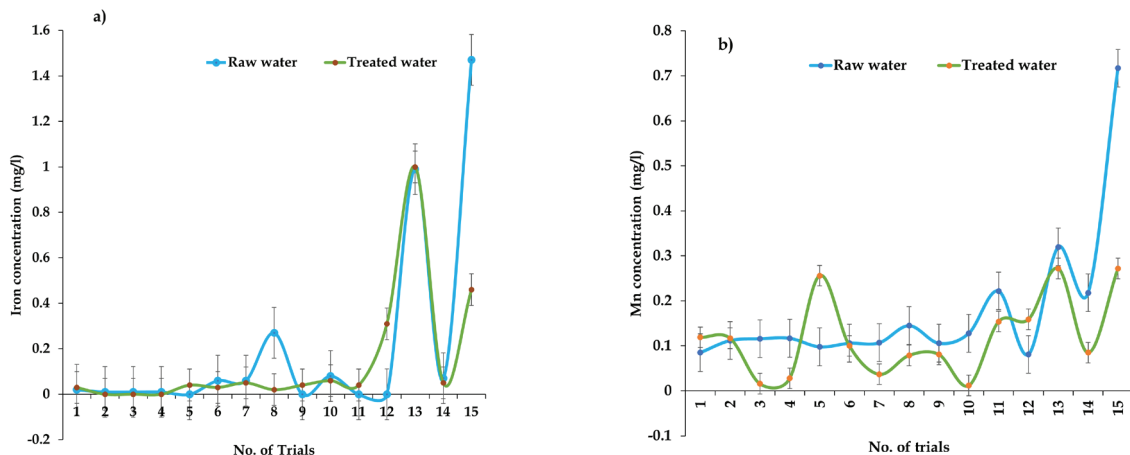


Figure 4 - Variation of Ion Concentration a) Iron, b) Manganese

filter outlet was increased. This may be due to the clogging of the filter pores which can lead to a drop in the filter efficiency. The filter inlet valve was closed and it was allowed to flow water upward through the filter column and it removed the clogged particles and cleared the pores. The backwashing flow rate was maintained at around 0.6-0.8 m³/h and backwash time was maintained at around 5 minutes. This agrees with the recommended minimum backwashing time of 5 minutes [4]. Thus, regular backwashing is essential for higher removal efficiency. As depicted in the Figure 3, in this experiment, backwashing was done after every two trials (after conducting trials nos. 2, 4, 6, 8, 10, 12, & 14). The results show substantial removal of manganese in the treated water after the backwashing in most cases. Manganese removal efficiency is increased by frequent backwashing.

4.4 Variation of pH

The pH value of treated water is observed to be higher than that of raw water in 12 out of 15 trials, as depicted in Figure 5. Further Fe²⁺ & Mn²⁺ ions have acidic properties in the presence of water. The increase in pH value in treated water samples indicated a decrease in Fe²⁺ and Mn²⁺ concentrations in water through the oxidation process to Fe³⁺ and Mn⁴⁺ [12]. The maximum pH value was recorded in trial 10. Consequently, the lowest manganese concentration in effluent and highest manganese removal were also recorded in trial 10. Hence, it is apparent that an increase in pH indicates the increasing removal efficiency of manganese and iron ions in water.

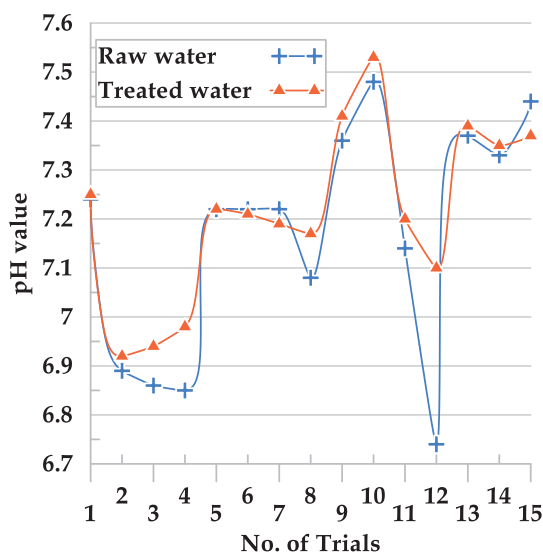


Figure 5 - Variation of pH

Figure 4 clearly shows the reduction of iron and manganese after the treatment process. Iron concentration reduction was observed in 9 out of 15 trials while manganese concentration reduction was observed in 11 out of 15 trials.

But it is noted that, for some trials, although the iron and manganese concentrations are reduced after the treatment, still the concentration values exceed the maximum permissible limits of SLS Standards. Raw water pH values ranged from 6.74 - 7.48 as depicted in Figure 5.

The best pH range for the oxidation of iron is between 6.5-7.5 while a pH value higher than 9.5 is required for manganese to get complete oxidation [11]. Hence, chlorination may not be effective for higher levels of manganese.

Nevertheless, Scherer and Ph [9] reviewed the ideal rate of oxidation of manganese and iron by chlorination at a pH of about 8.5 and 8.0, respectively. The maximum pH observed in raw water was 7.48 and that may be the possible cause for less iron manganese removal rate. Sodium carbonate (washing soda) can be used with chlorine to create an alkaline environment for a higher removal rate.

5. Conclusions and Recommendations

As a pilot study, a filter model was erected and raw water was conveyed through the filter model after oxidizing by chlorination, and effluent was collected at the bottom and tested for iron, manganese, and pH. Based on the results of laboratory tests, there was a considerable reduction in manganese and iron in the treated water sample by using the treatment process of chlorine oxidation followed by Rapid Sand Filtration.

The findings also suggest that iron removal is 60% and manganese removal is 73% for 15 no. of trials by using the filter media with the effective size (D₁₀) and uniformity coefficient (C_u) of 0.45 mm and 1.33, respectively. The maximum removal of iron and manganese was found to be 93% and 91%, respectively.

There is a positive correlation between pH values and removal efficiency. It was revealed that backwashing enhanced the treatment plant's efficiency. Future studies shall be

carried out using filter media with different effective sizes and uniformity coefficients to find the suitable particle distribution of the filter media for the efficient removal of iron and manganese.

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