

Treatment of Groundwater for the removal of Iron and Manganese From Groundwater wells of southern of Libya

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Abstract

The purpose of the study to investigate the impact of the presence of iron and manganese found in groundwater. The concentrations of both minerals as well as the location of the water source were identified along with its impact on water quality. Also, in this investigation a suitable method or technique for the removal of both iron and manganese is selected taking into consideration the local economic and environmental aspects. The removal will be accomplished by oxidizing both iron and manganese using aeration or using dissolved chemical oxidants converting them from soluble to insoluble precipitates. Precipitates of iron and manganese hydroxides are formed and removed from water through settling and filtration units. In this research the concentrations of iron and manganese were analyzed from groundwater aquifers of a number of towns in the southern part of Libya. These concentrations were compared to the local and international drinking water standards set by the World Health Organization (WHO). Some water samples reported have shown a wide difference in iron and manganese concentration and selected for treatment in this investigation. A complete treatment system has been designed to remove iron and manganese for the groundwater at Brak city of Alafia since the iron and manganese exceeds the limits. The oxidation of iron and manganese was done via aeration followed by flocculation and, settling, filtration and finally disinfection. At Alafia city, iron and manganese concentrations were 3.1mg/L, and 0.32mg/L respectively as compared to the standards set by the World Health Organization, 2004 for concentrations of 0.3 mg /L, 0.1 mg /L respectively, This process is believed to be very effective and economically feasible in the removal of both iron and manganese.

Key words: Iron, Manganese, Treatment, Aeration, settling, Filtration

1. Background

Iron (Fe) and manganese (Mn) are abundant elements in the earth's crust. They are mostly in oxidized state (ferric, Fe^{3+} , and Mn^{4+}) and are insoluble in natural waters. However, under reducing conditions (i.e. where dissolved oxygen is lacking and carbon dioxide content is high), appreciable amounts of iron and manganese may occur in groundwater. To forms soluble divalent ferrous (Fe^{2+}) and manganous (Mn^{2+}) ions (Shun Dan Lin, 2007a) . Natural sources of iron and manganese may include weathering of iron and manganese bearing minerals like amphibole, iron sulfide and iron rich clay minerals. In areas where groundwater flows through an organic rich soil, iron and manganese will also dissolve in the groundwater. Iron and manganese can also have anthropogenic sources including industrial effluents, landfill leakages and acid mine drainage. Well casing, pump parts, piping and storage tank can also contribute iron and manganese to groundwater (Nova Scotia, 2008). Generally iron and manganese cause many problems. However, in public water supplies they may discolor water, stain plumbing fixtures and laundry, and cause tastes and odors. Iron and manganese may also cause problems in water distribution systems because metal depositions may result in pipe encrustation and may promote the growth of iron bacteria which may in turn cause tastes and odors. Iron and manganese may also cause difficulties in household ion exchange units by iron and manganese. (Shun Dan Lin, 2007b). And the higher concentration of

these metals result in metallic taste of water, effect color and flavor of food and cause staining of different products like paper, cloths, and plastics. Iron and manganese in water are not a health risk but the manganese does not become a risk to the human health until reaching about 0.5 mg/L, according to a fact sheet released by the Connecticut Department of Public Health (2011), The same report states that high concentration of manganese can lead to toxicity of the nervous system over the course of many years causing a syndrome that resembles Parkinson's disease. There are several techniques have been applied to remove iron and manganese from groundwater. The most commonly used processes for physical/chemical removal of iron and manganese involve oxidation and precipitation following filtration methods include: aeration, chlorination or chlorine dioxide oxidation, potassium permanganate oxidation, advanced oxidation using ozone, and other physical/chemical methods for the removal of iron and manganese are also sometimes used either alone or in conjunction with oxidation and filtration processes : use of oxide-coated or catalytic media, lime softening and ion exchange. (HDR Engineering Inc., 2002).

2. Water analysis

Table (1) Iron and manganese concentrations in wells of different towns in the south of Libya.

sample	Murzuq (Aldesa)	Ubari (Ben haret)	Sebha (Alnaseriya)	Samno	Brak (Alafia)	Mansoura
Iron ppm	0.05	0.04	0.132	0.1	3.1	0.43
Manganese ppm	0.2	0.2	0.3	0.3	0.32	0.22

The samples were taken from groundwater and analyzed completely, the concentrations of iron and manganese were reported on Table (1). Such analysis were considered in the design and calculations of the iron and manganese treatment system.

3. Remove of iron and manganese by aeration

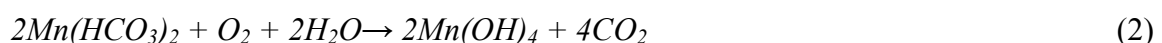
Aeration is used for the transfer of oxygen from the air into the water for the oxidation of iron and manganese to convert the iron and manganese from ferrous and manganous (soluble) forms to insoluble oxidized ferric and manganic forms (Thuresson, L. 1994). The most common aerators used in water treatment are : (1) gravity aerators (cascade, multi-tray aerators), (2) fountain aerators (spray aerators), (3) mechanical aerators, and injection or diffused aerators (Fahid Rabah, 2012).

3.1. Reactions of iron and manganese with oxygen

The following reaction describes the oxidation of ferrous iron by oxygen :



The following reaction describes the oxidation of manganous manganese by oxygen :



3.2. The conditions of iron and manganese oxygenation

At temperature 20°C and 1 atm for both reactions, oxygenation of iron with conversion 90% occur at $P_{O_2} = 0.2$ atm and adjust pH = 6.9 during 42 min. to conversion 90% of manganese occur at $P_{O_2} = 1$ atm, pH = 9.5 during 50 min. Three important assumptions of the proposed treatment system need to be considered namely: 1) the oxidation reaction are taking place at steady state

condition. 2) The reactors used are of type Continues Flow Reactor CFR. And 3) completely mixed contents of the continuous flow reactors.

3.3. Kinetics of iron oxygenation

At pH values greater than 5.5, the rate of oxygenation of Fe^{2+} iron was found to be first order with respect to Fe^{2+} and O_2 and second order with respect to OH^- ion (stumm and lee, 1961). As shown in Fig (1). Based on the experimental observations, the following expression was proposed Eq. (3) (John C. Crittenden *et al.* 2012) :

$$-r_A = -\frac{d[Fe^{2+}]}{dt} = k[Fe^{2+}][OH^-]^2 P_{O_2} \quad (3)$$

Table (2) parameters of kinetics of iron oxygenation.

Parameter	k $L^2/(\text{min.atm.mol}^2)$	$[Fe^{2+}]$ (mol/L)	$[OH^-]$ (mol/L)	P_{O_2} atm	$-r_A$ (mol/L.min)
value	1.887×10^{13}	5.55×10^{-5}	7.94×10^{-8}	0.2	1.322×10^{-6}

3.4. Kinetics of manganese oxygenation

Similar to Fe^{2+} , the rate of Mn^{2+} oxidation is dependent on P_{O_2} and $[OH^-]$ as shown in the Eq. (4). (John C. Crittenden *et al.* 2012) :

$$-r_A = -\frac{d[Mn^{2+}]}{dt} = k[Mn^{2+}][OH^-]^2 P_{O_2} \quad (4)$$

Where: k = reaction rate constant, P_{O_2} = partial pressure of oxygen $[Fe^{2+}]$, $[Mn^{2+}]$ = concentration of ferrous and manganous ions respectively, $[OH^-]$ = concentration of hydroxyl ions.

Table (3) parameters of kinetics of manganese oxygenation.

parameter	k $L^2/(\text{min.atm.mol}^2)$	$[Mn^{2+}]$ (mol/L)	$[OH^-]$ (mol/L)	P_{O_2} atm	$-r_A$ (mol/L.min)
value	20028841.5	5.828×10^{-6}	3.16×10^{-5}	1	1.165×10^{-7}

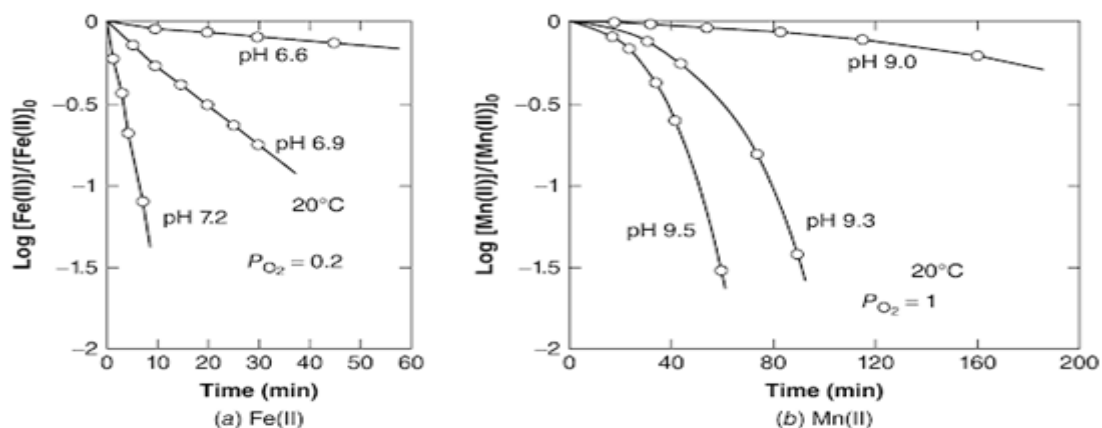


Figure (1).Oxygenation of iron(II) and manganese(II) in bicarbonate solution(After Fair *et al.*, 1971)

3.5. Mole balance at steady state

Volume of reaction calculated from mole balance of each aerator, at a steady state the mole balance equations Eq. (5) :

$$\text{Input} - \text{output} - r_A = 0 \quad (5)$$

$$Q_1 \cdot [Fe^{2+}]_0 - Q_1 [Fe^{2+}]_t - \frac{d[Fe^{2+}]}{dt} \cdot V_r = 0 \quad \text{For first reactor}$$

$$Q_2 \cdot [Mn^{2+}]_0 - Q_2 [Mn^{2+}]_t - \frac{d[Mn^{2+}]}{dt} \cdot V_r = 0 \quad \text{For second reactor}$$

Where : Q_1 , Q_2 = flow rate to aerator 1 and 2 respectively. $[Fe^{+2}]_0$, $[Mn^{+2}]_0$ = concentration of iron and manganese at time 0 = $[Fe^{+2}]_t$, $[Mn^{+2}]_t$ concentration of iron and manganese at time t. V_r = volume of reaction. $-d[A]/dt = -r_A$ = rate of reaction. Where $A = Fe^{2+}$ or Mn^{+2} .

Table (4) Parameters to calculate volume of reaction in aerator 1.

parameter	Q_1 (L/min)	$[Fe^{2+}]_0$ (mol/L)	$[Fe^{2+}]_t$ (mol/L)	$-r_A$ (mol/L.min)	V_r (L)
value	110	5.55×10^{-5}	5.55×10^{-6}	1.322×10^{-6}	4156

Table (5) Parameters to calculate volume of reaction in aerator 2.

parameter	Q_2 (L/min)	$[Mn^{+2}]_0$ (mol/L)	$[Mn^{+2}]_t$ (mol/L)	$-r_A$ (mol/L.min)	V_r (L)
value	327.5	5.828×10^{-6}	5.828×10^{-7}	1.165×10^{-7}	13894.5

4. The processes description

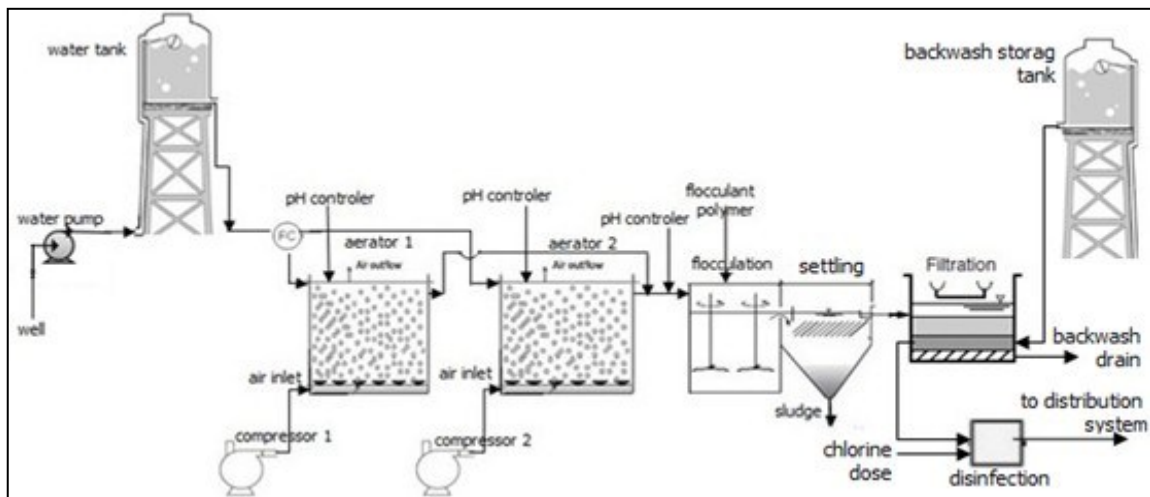


Figure (2) Proposed process flow diagram.

As shown in figure (2). The water flows from the water tank to the flow controller (FC) to divide the flow into two flows. The first flow enters the first aerator with the air entering from the bottom of the aerator by the air compressor 1, with the addition of the pH controller to be 9.6, and the second flow enters the second aerator with the addition of a pH controller , to be 9.5 use (soda ash or caustic soda to rise pH, sodium bisulphate to reduce pH). with the air flow from the bottom of the aerator by air compressor 2. The water exits from the two aerators combine together with the pH

adjustment to enter flocculation unit and add polymer to increase sediment density and facilitate settling. The water with large particles (flocs) directly enters to the settling unit (lamella clarifier) will correspond to the settling of the flocs. The water produced from settling unit contains small solid particles that did not precipitate entering the filtration unit to remove these solid particles. After this process, inject the water with the disinfectant and the treated water into the distribution system.

4.1. Aeration

Iron and manganese react in water with oxygen to form iron and manganese sediments. This process is performed in two aerators. In the first aerator, 90% of the iron is oxidized with pH which is controlled for iron oxidation, and the manganese is not oxidized in this aerator. In the second aerator, 90% of the manganese is oxidized with pH which controlled for manganese oxidation. The iron is almost completely oxidized because the pH is high in this aerator. This process produce water with iron and manganese (soluble) concentrations = 0.07mg/L, 0.09 mg/L respectively.

4.2. Flocculation

The water out of the aeration units contains solid particles of iron manganese hydroxide are very small (initially $< 2.5\mu\text{m}$ diameter) and have low density, and these particles have a very slow settling velocity. This problem can often be solved by adding polymers as flocculant and gentle stirring to produce larger and denser floc (J. Poul Guyer *et al.* 2016). anionic polyacrylamide or polysulfonate use as flocculant to binding with 90% of small particles at relatively low dosing concentration from 0.1 - 0.5 mg/L with slow mixing time 180 sec and $G = 50 \text{ s}^{-1}$ and $Gt = 9000$ (Hermann H. Hahn, 2000).

4.3. Settling

To sedimentation the flocs produced after flocculation process. The lamella clarifier (inclined plate clarifier) designed to high removal efficiency and short settling time. The principle of shallow depth sedimentation has been extended to design of parallel plate system, sometimes referred to as lamella clarifiers. Clarifiers using the plate system are usually purpose built to take advantage of the high settlement rates which can be obtained and the greater density of sludge provided. They require an efficient flocculation stage which is critical for successful operation. The flocculated water enters at the base of lamella plates and travels upward between the plates counter current to settled moving down (Ratnayaka *et al.* 2009).

4.4. Filtration

The smaller size particle present in the water cannot be settled in through the settling tanks. The filtration unit is introduced and designed to remove such small size particles by applying a rapid sand filter. The rapid sand filter contains dual media (anthracite, sand media). The rapid-filtration cycle consists of two stages: (1) a filtration stage, during which particles accumulate, and (2) a backwash stage, during which the accumulated material is flushed from the system. During the filtration stage, water flows downward through the filter bed and particles collect within the bed. The filtration stage typically lasts from 1 to 4 days and the backwash typically lasts 10 to 15 min, and backwash velocity 0.762 to 1.219 m/min. (Kerry J. Howe, *et al.* 2012a).

4.5. Disinfection

Disinfectant doses depend on whether the disinfectant is being used for inactivation, residual maintenance, or both. When chemical disinfectants are added to water, some of the chemical will be consumed during rapid oxidation of reduced compounds in the water, this consumption is known as the initial demand. Once the initial demand has been satisfied, additional chemical addition leads to

a residual concentration in the water. (Kerry J. Howe *et al.* 2012b). Chlorine is an effective disinfectant where water is not turbid. use chlorine dose to oxidize the remaining iron and manganese (0.07 mg/L, 0.09 mg/L) respectively, 0.63 mg/L of chlorine to oxidize 1 mg/L of iron, and 1.29 mg/L to oxidize 1 mg/L of manganese, the amount of chlorine to oxidation of iron and manganese = 0.16 mg/L, and destroys most recognized pathogenic microorganisms, there should be a chlorine residual of 0.2 to 0.5 mg/L. or by use any other disinfectant like ozone or UV (Anon, 1996).

5. Iron and manganese plant component design units and operation

5.1. Aeration units

5.1.1. Flow rates in aeration units

The population of Brak (Alafia) is about 4500 inhabitants. And the amount of water consumed per person is about 140-150 liters per day. The amount of water consumed in this town = 630000 liters per day used this value as the water flow rate used in this process.

$$Q = 140 \times 4500 = 630000 \text{ L/day} = 437.5 \text{ L/min} = 0.4375 \text{ m}^3/\text{min} = 26.24 \text{ m}^3/\text{h}.$$

Assume 25% of water flow rate in aerator 1, and 75% in aerator 2

$$Q_1 = 0.25 \times Q = 110 \text{ L.min}^{-1}.$$

$$Q_2 = 0.75 \times Q = 327.5 \text{ L.min}^{-1}.$$

Where: Q = water flow rate, Q_1 = flow rate to aerator 1, Q_2 flow rate to aerator 2.

5.1.2. Theoretical Oxygen and air required

The concentration of iron and manganese = 3.1 mg/L, 0.32 mg/L respectively, in theory, 0.14 mg/l of oxygen is needed to oxidize 1mg/L of iron and 0.29 mg/L of oxygen for each mg/L of manganese (Shun Dan Lin, 2007c). The initial oxygen level in waters with iron and/or manganese present is typically zero, and oxygen residual about 5 mg/L.

$$\text{The total amount of oxygen required} = 3.1 \times 0.1432 + 0.32 \times 0.2912 + 5 = 5.5268 \text{ mg/L}.$$

Air density 1.2047 g/L. at temperature 20°C and pressure 1atm, air contains 21% oxygen, each liter of air contains $0.21 \times 1204.7 = 252.9 \text{ mg/L}$ oxygen.

$$\text{The amount of air required to aerator 1} = (Q_1 \times 5.5268) / 252.9 = 2.4 \text{ L/min by compressor 1}.$$

$$\text{The amount of air required to aerator 2} = (Q_2 \times 5.5268) / 252.9 = 7.15 \text{ L/min by compressor 2}.$$

The compressed air under pressure of (35 to 70 kN/m²) (NPCS Board of Consultants. At el. 2009) with oxygen transfer efficiency from the compressed air is in the range of (10-15%).

5.1.3. Technical design of aerators

Assume the volume of reaction = 60% of aerator tank volume, and calculated the parameters of design by following equations:

$$V = L \times W \times h \tag{6}$$

$$A_x = W \times h \tag{7}$$

$$v_f = Q_i/A_x \tag{8}$$

Where: $Q_i = Q_1$ or Q_2 , V = volume of aerator, L = length of aerator, W = width of aerator. h = height of aerator, A_x = front area of aerator, v_f = flow velocity.

Table (6) Parameters of aerators design.

parameter	$V(m^3)$	$L(m)$	$W(m)$	$h(m)$	$A_x(m^2)$	$v_f(m/s)$
Aerator 1	8	2	2	2	4	4.58×10^{-4}
Aerator 2	20	5.25	2	2	4	1.364×10^{-3}

5.2. Lamella clarifier design

Parameters for the Lamella clarifier that is proposed for the settling operation is shown on table. (7,8). The design equations for the lamella clarifier are shown through equations (9 to 15). Detailed design parameters for the lamella clarifier are shown on figure. (3) (David W. Hendricks, 2006), (Thomas E. Wilson, 2005):

$$A_{pt} = Q/V_s = n.A_p = n.L.W.\cos \theta \quad (9)$$

$$h = L.\sin \theta \quad (10)$$

$$A_{sp} = (\cos \theta) / d \quad (11)$$

The ratio of the total projected area to the water surface area is approximately 8:1.

$$A_s = A_{pt}/8 \quad (12)$$

$$V_o = Q/A_s \quad (13)$$

$$Q_p = \frac{Q(W.\frac{s}{\sin \theta})}{A_s} \quad (14)$$

$$V_p = \frac{Q_p}{W.d} \quad (15)$$

Where: Q = Water flow rate, L = Length of plate, S = Spacing between plates, h = Height of plates, x = Thickness of plate, r = Horizontal spacing between plates, A_p = The projected area, A_{pt} = Total projected area, d = Vertical spacing between plates, n = Number of plates, A_s = water surface area. V_o = over flow rate to lamella, V_s = the settling velocity, A_{sp} = Specific surface area, V_p = mean velocity within. plate cell, Q_p = the flow rate of plate.

Table (7) parameters of lamella clarifier design.

parameter	θ	$L(m)$	$W(m)$	$H(m)$	$X(mm)$	$S(mm)$	$r(mm)$	$d(mm)$	n
Value	55	3	1.5	2.45	0.7	45.8	80	114	7 plates

Table (8) parameter of lamella clarifier design.

parameter	$Q(m^3/h)$	$Q_p(m^3/h)$	$A_p(m^2)$	$A_{pt}(m^2)$	$A_{sp}(m^2)$	$A_s(m^2)$	$V_o(m/h)$	$V_s(m/h)$	$V_p(m/h)$
Value	26.24	1	2.58	18	4.12	2.25	11.66	1.5	14.55

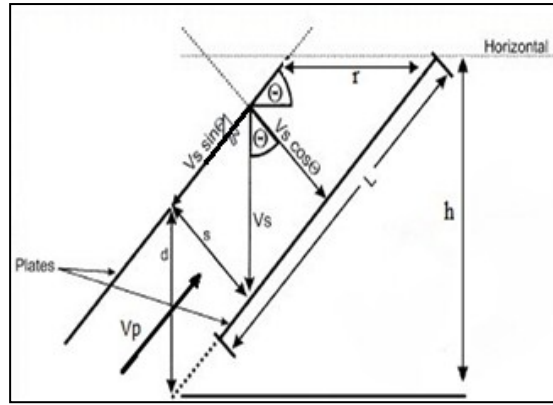


Figure (3). Plates design of lamella clarifier.

5.3. Rapid filter design

The parameters of a rapid filter design which shown in Table (9).calculated by following equations

$$A_f = Q/v_f \quad (16)$$

$$A_f = (\pi/4) \times d^2 \quad (17)$$

$$V_f = (\pi/4) \times d^2 \times h_f \quad (18)$$

Where: Q = The flow rate, v_f = Filtration rate, A_f = Filter surface area, d_f = Diameter of filter, h_f = Height of filter, V_f = Volume of filter.

Table (9) parameters of rapid filter design.

Parameter	Q(m ³ /sec)	v_f (m/sec)	A_f (m ²)	d_f (m)	h_f (m)	V_f (m ³)
Value	7.28×10^{-3}	4.06×10^{-3}	1.79	1.22	1.83	2.127

5.3.1. Volume of each media

Assumed the diameter of particle of media = effective size of particle, and the volume of each media calculated by following equation:

$$V_m = (\pi/4) \times d^2 \times L \quad (19)$$

Where: L = depth of media, ES = effective size, V_m = Volume media, UC = Uniformity coefficient.

Table (10). Parameters of filter media.

parameter	L (m)	ES (mm)	UC	V_m (m ³)
Anthracite	0.610	1.0	1.700	0.713
sand	0.205	0.5	1.600	0.953

5.3.2. Head loss for fixed bed flow

The head loss of each type of media was calculated according to the Kozeny equation shown below (Shun Dan Lin, 2007d) :

$$h = \frac{L.k\mu(1-\varepsilon)^2}{g\rho\varepsilon^3} \cdot \left(\frac{V}{A}\right)^2 \cdot v_f \quad (20)$$

$$\frac{V}{A} = \frac{6}{\phi.d} \quad (21)$$

Where: k = dimensionless coefficient, v_f = filtration rate, A = the grain surface area, V = the grain volume, ε = filter porosity, ϕ = shape factor, μ = dynamic viscosity, ρ = water density, h = head loss in clean filter.

Table (11) parameters of head loss in clean filter

Parameter	L (m)	g (m/s ²)	k	μ (kg/s.m)	ε	d (m)	ϕ	h (m)	ht (m)
Anthracite	0.61	9.81	6	9.982×10^{-4}	0.4	0.001	0.7	0.104	0.805
Sand	0.205	9.81	5	9.982×10^{-4}	0.4	5×10^{-4}	0.7	0.701	

5.3.3. Head loss for fluidized bed

The head loss of each type of media was calculated according to the Richardson Zaki equation shown below (Shun Dan Lin, 2007e) :

$$h = \frac{Le(1-\varepsilon_e)(\rho_s - \rho)}{\rho} \quad (22)$$

$$Le = L \left(\frac{1-\varepsilon}{1-\varepsilon_e} \right) \quad (23)$$

$$\varepsilon_e = \left(\frac{v_b}{v_s} \right)^{0.22} \quad (24)$$

Where: v_s = terminal velocity. Le = expanded bed depth. ε_e = expanded bed porosity. ρ = water density. ρ_s = particles density. v_b = backwash velocity through dual media. h = head loss during filter backwash. ht = total head loss.

Table (12) Parameters of head loss during filter backwash.

parameter	v_b (m/s)	v_s (m/s)	ε_e	ρ_s (kg/m ³)	Le (m)	h (m)	ht (m)
Anthracite	0.01524	0.081	0.69	1600	1.18	0.219	0.422
Sand	0.01524	0.077	0.7	2650	0.41	0.202	

6. Discussion

The aeration method to remove both iron and manganese from groundwater does not have any impact on environment, health and risks compared to iron and manganese oxidation methods where chemicals Treatment are involved. In this process, when the pH is raised to 9, all organic compounds are oxidized in groundwater such as phenols. The advantages of this method are that

they do not need oxidizing chemicals, and partly removes CO₂, H₂S and CH₄ if presented in water. The oxidation by aeration is ineffective in cases of where low pH and high concentration of iron and manganese or when the iron is associated with organics.. Iron and manganese oxidation using aeration techniques usually employed different means of introducing air into water such as cascade aeration, multi tray or plate aeration, surface aeration and diffused aeration. Diffused aeration was found very effective in our proposed treatment system. The rate of aeration depends upon contact time, transfer efficiency, and the concentration of iron and manganese present.

Iron and manganese both react with oxygen but the manganese reaction with oxygen is very slow and needs high pH 9.5 or more during long time. Iron is oxidized at pH 7.2 during 15 minutes. In this investigation it is highly recommended to treat iron and manganese in two stages. The first stage, the pH need to be adjusted using sodium bisulfate i.e. lowered to a value of 6.9 in order to remove iron effectively. In the second stage, the pH has to be raised to a value of 9.5 using caustic soda (sodium hydroxide) in order to effectively remove manganese.

7. Conclusions

- Aeration of GW can effectively remove iron and manganese to a level accepted by local and international standards.
- When manganese is available in GW at higher concentrations, it is important that the water treatment process is well monitored to ensure the removal of Fe and Mn are within the acceptable levels.
- Manganese requires larger amount of oxygen for oxidation during aeration as compared to iron.
- Iron is easily oxidized than manganese during aeration.
- Longer retention time may be required for the removal of manganese if aeration and filtration are applied in the water treatment process.
- The pH adjustments need to be carefully monitored after oxidation of iron and manganese because pH may affect the floc formation and removal during the water treatment.
- The pH value of the effluent treated GW need to be adjusted to a level (6 – 8), below this level is believed to cause iron and manganese flocs formed to re-dissolve in water.

8. Recommendations

- Proposed GW treatment system for iron and manganese removal need to be tested through a pilot scale study before any field application.
- Further studies need to be conducted on GW that contains higher levels of Fe and Mn, Ammonium salts and natural organics matter because might have dissolved oxygen deficit.
- An economic study for the proposed GW treatment system for iron and manganese removal is essential for field applications in order to get cost of treatment per cubic meter.

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