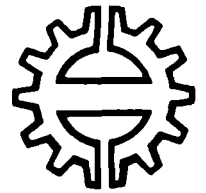


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Water Supply and Sanitation**

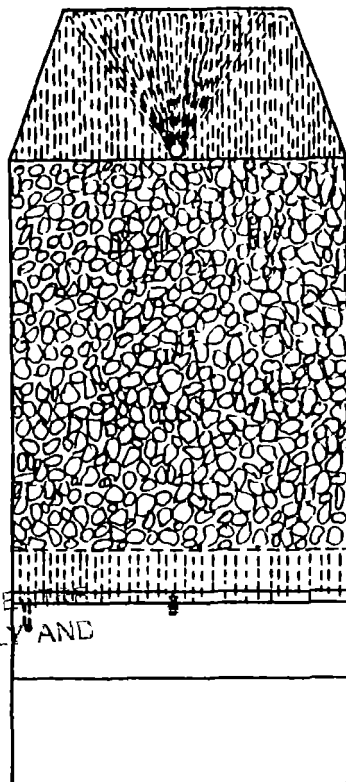
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Ashenafi Kibret

Dry Filter - Application in Water Treatment for Iron Removal



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**DRY FILTER - APPLICATION IN WATER
TREATMENT FOR IRON REMOVAL**

by

ASHENAFI KIBRET

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Thesis submitted to the
department of Civil Engineering,
Tampere University of Technology
in partial fulfillment of the
requirements for the degree of
Master of Science in Engineering

February 1986
Tampere, Finland

DRY FILTER-APPLICATION IN WATER TREATMENT FOR IRON REMOVAL

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ABSTRACT

Iron is objectionable constituent when present in drinking and process water more than the desired limit. Different iron removal methods and their modifications have been applied for iron removal. However, conventional methods are found expensive and there is an increasing tendency against the use of chemicals in water treatment.

Dry filter is one of the alternatives that can be applied for iron removal and the process uses the self-purification capacities of iron bacteria. These micro-organisms are found to adapt themselves to the prevailing conditions provided the environment is made favourable for them, such as ventilation, provision of surface media for their growth , raw water supply etc.

Investigations made on the pilot plants showed that iron removal process by dry filtration depends on the hydraulic load, filter depth, size of filter material, the development of the micro-organisms, and iron concentration in the raw water.

Dry filter does not only remove iron but it also removes manganese, ammonia, carbon dioxide and provides sufficient oxygen supply to the treated water. The results obtained from the test plants were not below the standard limits except from the full scale production plant. However, complete removal of iron by dry filter is feasible provided the best possible favourable combinations of the factors on which iron removal depends are found.

Micro-biological water treatment is certainly more appropriate to natural drinking water than production of chemically treated "water" in water treatment works. The biological process as to how these micro-organisms remove iron is still unanswered.

1 INTRODUCTION

Iron which is an essential element for plant and animal is a highly objectionable constituent when found in water supplies for either domestic or industrial use. When the water containing ferro iron is exposed to air or oxygen it oxidizes slowly forming objectionable colloidal precipitates. The presence of these precipitates in the water supply produces colour, turbidity and deposition. The deposits can cause staining of plumbing fixtures, interfere with laundering and with many industrial processes such as white paper-making, photography, dyeing, leather-tanning, food processing industry etc. (Weng 1984).

Accumulation of deposits in distribution systems is frequently responsible for creating ideal conditions for the growth of iron oxidizing micro-organisms such as clonothrix and crenothrix, the combined effect being the blockage of meters and valves as well as reduction of pipeline carrying capacity. Sloughing-off of these accumulations can cause taste and odour problems. Iron in high concentrations of several milligrams per litre will impart a metallic, astringent or medicinal taste.

The WHO international recommendations gives 0,3 mg/l of iron as a standard guideline value (WHO 1984). This value is fixed based on the aesthetic, taste and nuisance considerations.

Because of the problems associated with iron in water supplies and to attain the limited standard value treatment is often provided prior to distribution of water to consumers. The principle in removing iron is relatively simple, and it has been widely applied. In spite of its simplicity, conventional iron removal plants have proved expensive to construct and to run especially when the need of producing water free from iron increases and/or the iron concentration of the ground water rises.

In an effort to solve these problems and to meet the

increasing demand for better quality of water, various modifications of the conventional methods have been tried as well as entirely different methods.

Due to the expensiveness of conventional iron removing plants and to some extent low efficiency, alternatives are looked for among cheaper and more efficient methods. Nowadays the use of chemicals for the oxidation of iron in conventional methods is considered to be expensive and disadvantageous. This leads to an alternative to be sought especially to the use of natural methods for aeration and filtration. One of the natural methods of filtration that involves biological iron removal process and is cheaper than conventional plants is the dry filtration. In dry filter the raw water containing iron is spread over gravel material packed in a box and trickles through the filter material where iron is removed by different iron bacteria that develop through the process of filtration.

The application of dry filter is not widely known for the process of iron removal. Generally there are no definite design principles and the processes taking place in the filtration are not established. Intensive investigation of the dry filtration process has to be made in order to establish general design criteria and to explain the iron removal and other processes taking place in the filter. In this study the establishment of design criteria and analysis of the efficiency of dry filter for iron removal with respect to hydraulic load, iron concentration in raw water, the use of different sizes of filter gravel and filter depth are made. The development and type of micro-organisms which are responsible for the oxidation of iron are investigated. Generally, the effect of dry filtration on water quality improvements is examined.

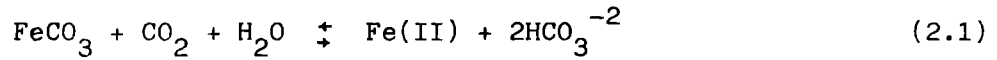
2 REACTION PROPERTIES OF IRON IN GROUND WATER

2.1 Origin of iron and its existence in groundwater

Iron is abundant in nature, composing about five percent of the earth's crust (Robinson 1967). It exists in many different chemical compounds and with the divalent ferrous form, Fe (II) and the trivalent ferric form, Fe (III). It is usually found in the form of oxides but carbonates and sulphides also occur. Minerals containing iron are like silicate minerals of igneous rocks, pyroxenes amphiboles and some micas generally contain iron. The various oxides containing iron are such as magnetite (Fe_3O_4), hematite (Fe_2O_3) and limonite ($2\text{Fe}_2\text{O} \cdot 3\text{H}_2\text{O}$). The sulphide and carbonate minerals which contain iron include pyrite (FeS_2) and siderite (FeCO_3) (Robinson 1967).

The presence of high concentration of iron is associated with deep well waters from shale, sandstone and other rocks. Iron is usually dissolved in the form of ferrous bicarbonate, but in some instances, mostly groundwaters, it may be found as higher oxides, in complex organic combinations, or, in rare instances, as the sulphides (Holden 1970).

The occurrence of iron in groundwaters is generally attributed to the solution of rocks and minerals. The solution of iron-bearing minerals is often due to the action of carbondioxide in groundwaters. Most of the carbondioxide is presumably generated by the bacterial decomposition of organic matter leached from the soil. This leaves significant amount of carbon dioxide and the water may be devoid of dissolved oxygen. The solution of the iron-bearing minerals may take place under anaerobic conditions in the presence of reducing agents (organic substances, hydrogen sulphide) capable of reducing the higher oxides of iron to the ferrous, Fe (II), states. Soluble ferrous iron may enter the groundwater through the reaction (Engelbrecht et al. 1967):



In an aqueous environment, both soluble and insoluble species of hydrolyzed iron may be present. The concentration of iron found in solution in natural waters is frequently limited by the solubility of its carbonates e.g. siderite (FeCO_3). Water of high alkalinity often, therefore, has lower iron content than waters of low alkalinity (AWWA 1971).

2.2 Chemical reactions

The chemical reactions involved in the solution and deposition of iron in natural ground water are readily reversible. The amount of Fe(II) or Fe(III) present in a given water is dependent on certain conditions of the water such as alkalinity, pH, carbon dioxide, dissolved oxygen content and concentrations of other minerals present (Robinson and Dixon 1968).

2.2.1 Fe(II) reactions

Fe(II) exists in natural groundwater mostly in anaerobic environment. In waters devoid of sulphides and carbonates, and when Fe(II) is dissolved under anaerobic conditions, the equilibria equations 1, 2 and 3 in table 2.1 will take place (Stumm and Lee 1960).

Table 2.1 Iron equilibria in water (Stumm and Lee 1960).

No.	Reaction	Equilibrium constant ¹⁾ 25° C	References
<i>Fe(II) Solubility</i>			
1	$\text{Fe}(\text{OH})_2(\text{s}) = \text{Fe}^{+2} + 2\text{OH}^-$	8×10^{-16}	[1]
2	$\text{Fe}(\text{OH})_2(\text{s}) = [\text{Fe}(\text{OH})]^{+} + \text{OH}^-$	4×10^{-16}	[1]
3	$\text{Fe}(\text{OH})_2(\text{s}) + \text{OH}^- = [\text{Fe}(\text{OH})_2]^-$	8.3×10^{-6}	[2]
4	$\text{FeCO}_3(\text{s}) = \text{Fe}^{+2} + \text{CO}_3^{-2}$	2.1×10^{-11}	[10]
5	$\text{FeCO}_3(\text{s}) + \text{OH}^- = [\text{Fe}(\text{OH})]^{+} + \text{CO}_3^{-2}$	1×10^{-5}	computed ²⁾
6	$\text{HCO}_3^- \rightleftharpoons \text{H}^+ + \text{CO}_3^{-2}$	4.8×10^{-11}	[10]
7	$\text{FeS}(\text{s}) + \text{Fe}^{+2} + \text{S}^{-2}$	6×10^{-16}	[39]
8	$\text{FeS}(\text{s}) + \text{OH}^- = [\text{Fe}(\text{OH})]^{+} + \text{S}^{-2}$	3×10^{-12}	computed
9	$\text{FeS}(\text{s}) + 3\text{OH}^- = [\text{Fe}(\text{OH})_2]^- + \text{S}^{-2}$	6.2×10^{-6}	computed
10a	$\text{H}_2\text{S}(\text{aq}) \rightleftharpoons \text{H}^+ + \text{HS}^-$	1×10^{-7}	[39]
10b	$\text{HS}^- \rightleftharpoons \text{H}^+ + \text{S}^{-2}$	1.3×10^{-13}	[39]
<i>Fe(III) Solubility</i>			
11	$\text{Fe}(\text{OH})_3(\text{s}) = \text{Fe}^{+3} + 3\text{OH}^-$	$\sim 10^{-36}$	computed
12	$\text{Fe}(\text{OH})_3(\text{s}) = [\text{Fe}(\text{OH})_2]^{+} + \text{OH}^-$	1.7×10^{-16}	computed
13	$\text{Fe}(\text{OH})_3(\text{s}) = [\text{Fe}(\text{OH})]^{2+} + 2\text{OH}^-$	6.8×10^{-22}	computed
14	$\text{Fe}(\text{OH})_3(\text{s}) = [\text{Fe}(\text{OH})_2(\text{d})]^{2+}$	2.9×10^{-7}	[2]
15	$\text{Fe}(\text{OH})_3(\text{s}) + \text{OH}^- = [\text{Fe}(\text{OH})_4]^-$	$\sim 10^{-6}$	estimated
<i>Complex Formation</i>			
16	$\text{Fe}^{+2} + \text{Cl}^- = [\text{FeCl}]^{+}$	~ 2.3	[8]
17	$\text{Fe}^{+2} + n\text{Cl}^- = [\text{FeCl}_n]^{2-n}$	—	[13]
18	$\text{Fe}^{+2} + \text{Cl}^- = [\text{FeCl}]^{+2}$	30	[40]
19	$[\text{FeCl}]^{+2} + \text{Cl}^- = [\text{FeCl}_2]^{+}$	4.5	[40]
20	$\text{Fe}^{+2} + n\text{Cl}^- = [\text{FeCl}_n]^{2-n}$	—	[40]
21	$\text{Fe}^{+2} + \text{SO}_4^{-2} = [\text{Fe}(\text{SO}_4)]^{+}$	1.5×10^4	[41]
22	$\text{Fe}^{+2} + \text{H}_2\text{O}_4^{-2} = [\text{Fe}(\text{H}_2\text{O}_4)]^{+}$	4.5×10^{10}	[42]
<i>Acidity</i>			
23	$\text{Fe}^{+2} + \text{H}_2\text{O} = [\text{Fe}(\text{OH})]^{+} + \text{H}^+$	5×10^{-9}	[1]
24	$\text{Fe}^{+2} + \text{H}_2\text{O} = \text{Fe}(\text{OH})^{+2} + \text{H}^+$	6.8×10^{-9}	[43]
25	$[\text{Fe}(\text{OH})]^{+2} + \text{H}_2\text{O} = [\text{Fe}(\text{OH})_2]^{+} + \text{H}^+$	2.6×10^{-6}	[43]
26	$[\text{Fe}(\text{OH})_2]^{+} + \text{H}_2\text{O} = \text{Fe}(\text{OH})_3(\text{s}) + \text{H}^+$	5.8	computed
<i>Polymerization</i>			
27	$2[\text{Fe}(\text{OH})]^{+2} = [\text{Fe}_2(\text{OH})_2]^{+4}$	30	[43]
28	$2\text{Fe}^{+2} + 2\text{H}_2\text{O} = [\text{Fe}_2(\text{OH})_2]^{+4} + 2\text{H}^+$	1.4×10^{-3}	[43]
<p>¹⁾ For most of the reactions listed, different authors have determined equilibrium constants, which differ slightly among each other. Other values may be found in [8].</p> <p>²⁾ Computed values have been obtained from other equilibrium constants given in this Table. For example, K_{18} can be computed from the equilibrium constants of reactions 11 and 24.</p> <p>³⁾ $[\text{Fe}(\text{OH})](\text{d})$ is dissolved molecular (undissociated) ferric hydroxide.</p>			

Using these equilibria equations and



where $K_w = [\text{OH}^-] [\text{H}^+]$

the activities of the different Fe(II) compounds can be plotted as a function of pH by taking the logarithms of

equation (2.2) and the equilibria equations in table 2.2. Figure 2.1 shows the solubility of $\text{Fe}(\text{OH})_2$ (s) in waters devoid of any carbonate or sulfur species (Stumm and Lee 1960).

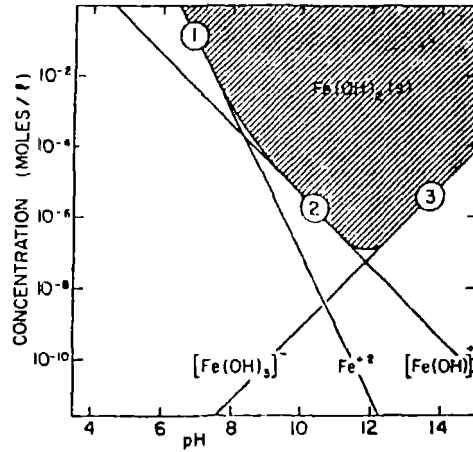


Figure 2.1 Solubility of $\text{Fe}(\text{OH})_2$ (s) in a non-carbonate, non-sulfide solution (Stumm and Lee 1960).

The solubility of Fe(II) cannot exceed the limits of the boundary defined by the shaded area in figure 2.1.

Figure 2.1, however, will not give a correct picture of the conditions in anaerobic groundwater where there are carbonate species and often sulfur compounds. Fe(II) reacts with carbonates and sulfides and form new compounds.

In natural waters, where only Fe(II) and carbonates are present the equilibria 4, 5 and 6 in table 2.1 will take place in addition to the equilibria 1, 2 and 3. Figure 2.2 shows the solubility of Fe(II) in a carbonate containing waters (Stumm and Lee 1960).

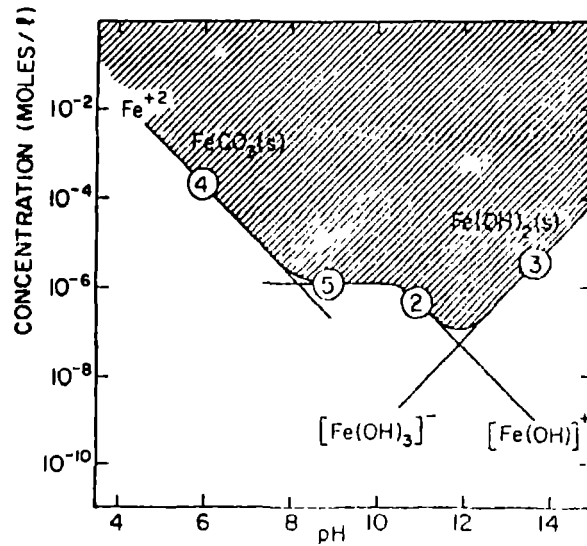


Figure 2.2 Solubility of Fe(II) in a carbonate containing water (alkalinity = 2×10^{-3} eg/l) (Stumm and Lee 1960).

From figure 2.2 it can be seen that at pH 10,5 FeCO_3 determines the solubility of Fe(II) while Fe(OH)_2 determines the solubility of Fe(II) for pH < 10,5 (Snoeyink and Jenkins 1980).

The other principal equilibria associated with Fe(II) in groundwater are sulfur compounds and the formation of complex ions and chelation (Robinson and Dixon 1968). Equilibria 7-10 in table 2.1 will occur between Fe(II) and species of sulfide system if available in groundwater. Figure 2.3 shows the solubility of Fe(II) in a carbonate and sulfide bearing water (Stumm and Lee 1960).

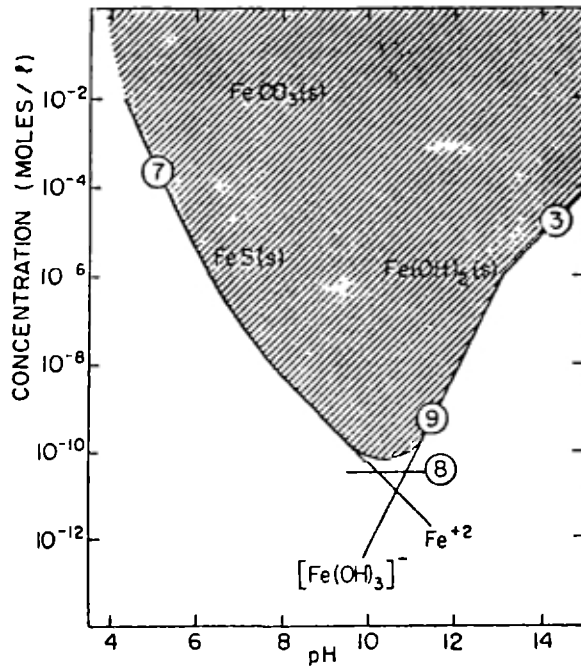


Figure 2.3 Solubility of Fe(II) in a carbonate and sulphide bearing water (Alkalinity = 2×10^{-3} eg/l; S(II) = 10^{-4} mol/l) (Stumm and Lee 1960).

2.2.2 Fe(III) reactions

On aeration or by the addition of oxidizing agents iron oxidizes from Fe(II) to the Fe(III) form and the Fe(III) forms an equilibria with H_2O . The most common species in the Fe(III) system is $\text{Fe}(\text{OH})_3(\text{s})$ which has a low solubility over pH range of 6-9, the most common cases in natural waters. The reactions which Fe(III) forms with H_2O are shown in table 2.1 equilibria 11-15. The solubility curves of Fe(III) in a non-carbonate, non-sulfide solutions is shown in figure 2.4 (Stumm and Lee 1960).

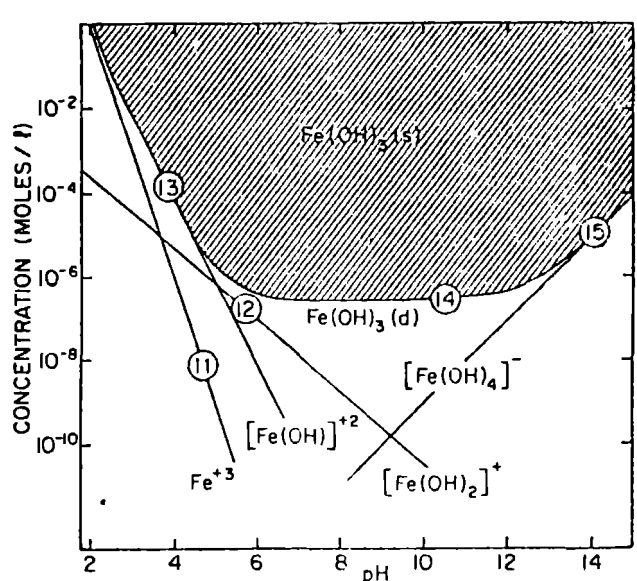


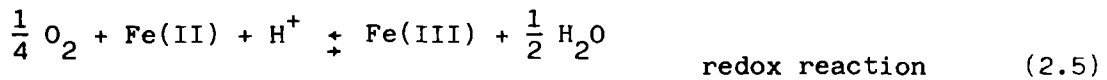
Figure 2.4 Solubility of Fe(III) in a non-carbonate, non-sulfide solution (Stumm and Lee 1960).

The low solubility of $\text{Fe}(\text{OH})_3$ over the wide range of pH is used as an advantage in conventional water treatment plants, where the precipitated $\text{Fe}(\text{OH})_3$ may be removed by filtration and/or sedimentation. However, Fe(III) can form complex compounds with hydroxides, orthophosphate, many organic bases etc. causing iron to remain in solution. The extent of complex formation is pH dependent, and, within pH range of natural waters, soluble or insoluble mixed Fe(III) complexes that may contain OH^- as well as other ligands can be formed (Stumm and Morgan 1970). Equilibria 16-22 in table 2.1 shows some examples of complex formation. The complex formations bring about, that the concentration of dissolved Fe(III) is higher than indicated in figure 2.4.

The incorporation of coordinating anions into basic precipitates not only alters the solubility relations but also strongly affects the colloid chemical properties of the dispersed phase. The complex formation will strengthen the colloid stability of the Fe(III) species, so they remain in colloid form, instead of combining as larger particles. Hence figure 2.4 does not precisely show the condition in natural waters but it gives a hint of the solubilities at different pH values.

2.2.3 Oxidation of Fe(II) + Fe(III)

Iron exists in natural groundwater in the soluble Fe(II) state where the water is devoid of oxygen. To remove this form of iron Fe(II) must be oxidized to Fe(III) form. This phenomenon occurs by a redox process where the oxidizing agents can be oxygen (O_2), permanganate (MnO_4^-), chlorine (Cl_2) or equivalent. Stoichiometrically, 1 mg/l of oxygen will oxidize 7 mg/l of Fe(II) (Engelbrecht et al. 1967). The reaction is as follows



As can be seen from the above reactions oxygen is reduced and Fe(II) is oxidized. If equation (2.4) is considered as reduction



which by the use of mass action law gives

$$\frac{[Fe(II)] [e^-]}{[Fe(III)]} = K \quad (2.7)$$

which is equal to

$$-\log [e^-] = \log K + \log \frac{[Fe(III)]}{[Fe(II)]} \quad (2.8)$$

and equation (2.8) can also be written as

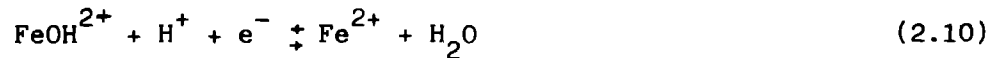
$$Pe = Pe^\circ + \log \frac{[Fe(III)]}{[Fe(II)]} \quad (2.9)$$

where P_e = the electron activity and is defined as $-\log [e^-]$

P_e° = the standard-electron activity and is equal to $\log K$.

In order to relate P_e° to $\log K$, K is defined in terms of the equilibrium constant of the reduction reaction (Stumm and Morgan 1970). P_e is a measure of the oxidizing or reducing intensity of a solution, i.e. how strong the solution is to oxidize or reduce other species. For instance, large positive values of P_e (low electron activity) represent strongly oxidizing conditions and small or negative values (high electron activity) correspond to strongly reducing condition (Stumm and Morgan 1970).

P_e° is the electron activity at $[Fe(II)] = [Fe(III)]$ or generally when the activity of the oxidant and the reductant are equal. Therefore, it can be said that P_e° is a comparative expression of the redox properties of the species. However, many redox equilibria are pH dependent as can be seen from the half reaction (reduction reaction).



from which P_e is calculated as

$$P_e = P_e^\circ + \log \frac{[FeOH^{2+}][H^+]}{[Fe^{2+}]} \quad (2.11)$$

This can be simplified as

$$P_e = P_e^\circ - pH + \log \frac{[FeOH^{2+}]}{[Fe^{2+}]} \quad (2.12)$$

The P_e° will not necessarily give a correct expression of the oxidizing or reducing intensities of the single species for most of the reactions in natural waters carried out at pH values around 7. Therefore, a constant $P_e^\circ (w)$ analogous to P_e° is introduced in which (H^+) and (OH^-) in the redox equilibrium equations are assigned their activities in neutral water. Values for $P_e^\circ (w)$ for 25°C thus apply to unit

activities of oxidant and reductant at pH = 7 (Stumm and Morgan 1970).

$P_{e^{\circ}}(w)$ is defined by

$$P_{e^{\circ}}(w) = P^{\circ} + \frac{nH}{2} \log K_w \quad (2.13)$$

where nH = the number of moles of protons exchanged per mole of electrons.

$nH = -1$ at pH = 7 in equation(2.13)

The $P_{e^{\circ}}(w)$ of different species can be compared directly and permits grading of different species in the order of their oxidizing intensity at pH = 7. Any species will tend to oxidize equimolar concentration of any other species having a lower $P_{e^{\circ}}(w)$ value. Figure 2.5 shows the comparison of $P_{e^{\circ}}(w)$ value. Figure 2.5 shows the comparison of $P_{e^{\circ}}(w)$ of different species (Harremöes et al. 1980/cited by Viegand 1984).

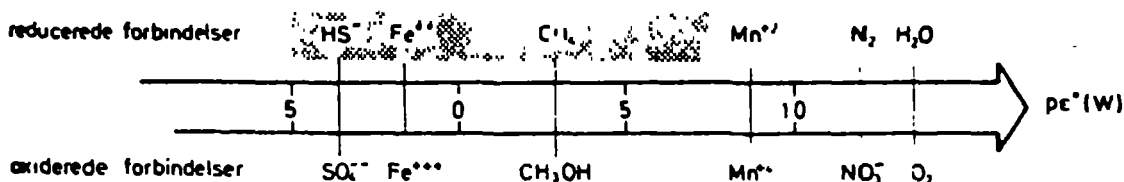


Figure 2.5 Comparison of the $P_{e^{\circ}}(w)$ of different species. Example; Fe^{3+} can in principle oxidize all the reduced species at left i.e. Hs^{-} . Fe^{2+} can in principle reduce all oxidized species at right, i.e. CH_3OH , Mn^{4+} , NO_3^{-} , O_2 (Harremöes et al. 1980/cited by Viegand 1984).

The quantity of $P_{e^{\circ}}(w)$ is a relative expression for the electron activity when all species other than the electrons are at unit activity. The electron activity can also be expressed in redox potential E_h .

$P\epsilon$ is related to E_h by

$$P\epsilon = \frac{F}{2,3RT} E_h \quad (2.14)$$

where F = Faraday's constant
 R = gas constant
 T = temperature in Kelvin scale.

By application of the redox potential equation (2.9) can be written as

$$E_H = E_H^\circ + \frac{2,3RT}{F} \log \frac{[Fe(III)]}{[Fe(II)]} \quad (2.15)$$

E_H can be measured in a well defined redox reactions in equilibria and together with pH give valuable information about the condition of the system. However, difficulties might occur with attempts to measure oxidation-reduction potentials in natural waters, where many redox systems are involved and often the systems are out of equilibria. In this kind of natural waters the measurement of redox potential must be done with care (Stumm and Morgan 1970).

A theoretical model useful in illustrating the various redox equilibria for iron is the E_h - pH diagram also known as the solubility-field diagram, which is computed from chemical equilibria data (Ghosh et al. 1966).

In table 2.2 the redox reactions of iron in a carbonate water are shown together with the activity equations (Ghosh et al. 1966).

Table 2.2 Fe-redox reactions and their activity equations in a carbonate bearing water (Ghosh et al. 1966).

No	Reaction ^a	Equilibrium Constant, 25°C	Reference
<u>Redox Reactions</u>			
13	$\text{Fe}^{++} + 2\text{e}^- = \text{Fe}$ $\text{Eh} = -0.440 + 0.0295 \log \text{Fe}$		Computed
14	$\text{Fe}^{+++} + \text{e}^- = \text{Fe}^{++}$ $\text{Eh} = 0.771 + 0.0591 \log \frac{a_{\text{Fe}^{+++}}}{a_{\text{Fe}^{++}}}$		Computed
	$\text{Fe(OH)}_{2(\text{s})} + \text{H}^+ + \text{e}^- = \text{Fe}^{++} + \text{H}_2\text{O}$ $\text{Eh} = 0.915 + 0.0591 \text{pH} + 0.0591 \log \frac{a_{\text{FeOH}^{++}}}{a_{\text{Fe}^{++}}}$	2.43 - 3.90	Computed
	$\text{Fe(OH)}_{3(\text{s})} + 3\text{H}^+ + \text{e}^- = \text{Fe}^{++} + 3\text{H}_2\text{O}$ $\text{Eh} = 1.06 - 0.1773 \text{pH} - 0.0591 \log a_{\text{Fe}^{++}}$	3.90 - 6.05	Computed
	$\text{Fe(OH)}_{3(\text{s})} + \text{CO}_3^{=} + 3\text{H}^+ + \text{e}^- = \text{FeCO}_{3(\text{s})} + 3\text{H}_2\text{O}$ $E_o = -0.672$	6.05 - 9.10	Computed
	$\text{Fe(OH)}_{3(\text{s})} + \text{H}^+ + \text{e}^- = \text{Fe(OH)}_{2(\text{s})} + \text{H}_2\text{O}$ $\text{Eh} = 0.271 - 0.0691 \text{pH}$	9.10 - ?	Computed
	$\text{Fe(OH)}_{2(\text{s})} + 2\text{H}^+ + 2\text{e}^- = \text{Fe} + 2\text{H}_2\text{O}$ $\text{Eh} = -0.0475 - 0.0591 \text{pH}$	9.10 - ?	Computed
	$\text{FeCO}_{3(\text{s})} + 2\text{e}^- = \text{Fe} + \text{CO}_3^{=}$ $E_o = -0.756$	6.05 - 9.10	Computed
<u>O₂ - H₂O System</u>			
	$\text{O}_2 + 4\text{H}^+ + 4\text{e}^- = 2\text{H}_2\text{O}$ $\text{Eh} = 1.229 - 0.0591 \text{pH} + 0.0148 \log (P_{\text{O}_2})$		Computed
<u>H⁺ - H₂ System</u>			
	$2\text{H}^+ + 2\text{e}^- = \text{H}_2$ $\text{Eh} = 0.0591 \text{pH} - 0.0295 \log (P_{\text{H}_2})$		Computed
^a $\text{Fe(OH)}_{2(\text{s})}$ = ferrous hydroxide in solid phase.			

The activity equations in table 2.2 can for a fixed pH be plotted in an activity diagram as shown in figure 2.6 (Stumm and Morgan).

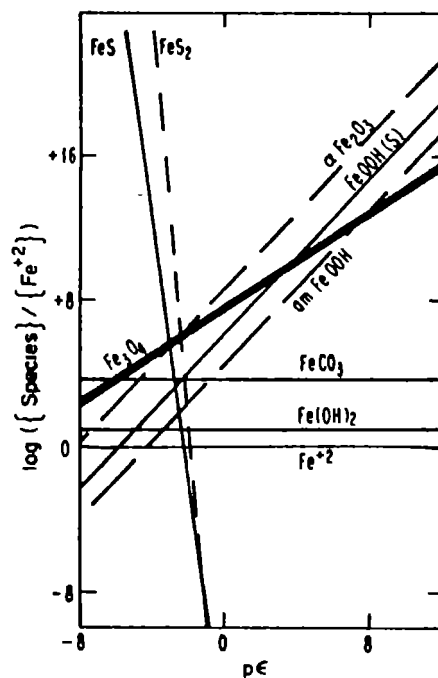


Figure 2.6 Activity diagram for a carbonate and sulfide bearing water. ($\text{pH} = 7$; $C_T = 10^{-3}\text{M}$, $\text{SO}_4^{-2} = 10^{-3}\text{M}$) (Stumm and Morgan 1970).

For a fixed Fe^{2+} activity the equation in table 2.2 can be plotted in a $E_h - \text{pH}$ diagram also known as stability field diagram. Care must be taken in the application of equilibrium data and $E_h - \text{pH}$ diagrams for the interpretation of the behaviour of materials like iron in natural waters due to comments given before about E_h measurement. Figure 2.7 shows the $E_h - \text{pH}$ diagram (Stumm and Lee 1960).

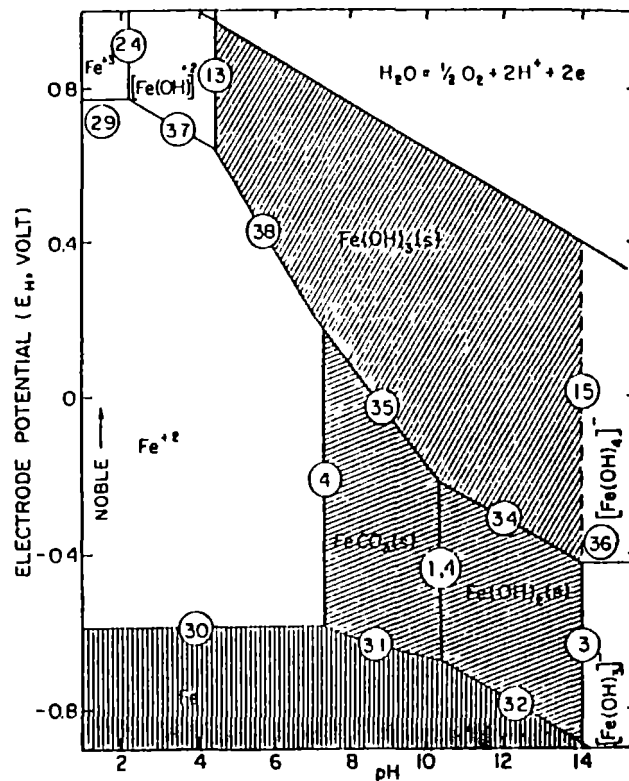


Figure 2.7 E_h - pH diagram. The shaded areas show the solid Fe-species. The lines denote the points at which the activities of soluble iron is 10^{-5} mol/l; alkalinity = 2×10^{-3} eg/l. (Stumm and Lee 1960).

E_h - pH diagram can be made for different activities of Fe^{2+} , but the purpose of the diagram is primarily to give a general view of the adjacent relations between iron species. Figure 2.8 shows E_h - pH diagram for different activities of soluble iron in the presence of carbonate species (Hem 1961).

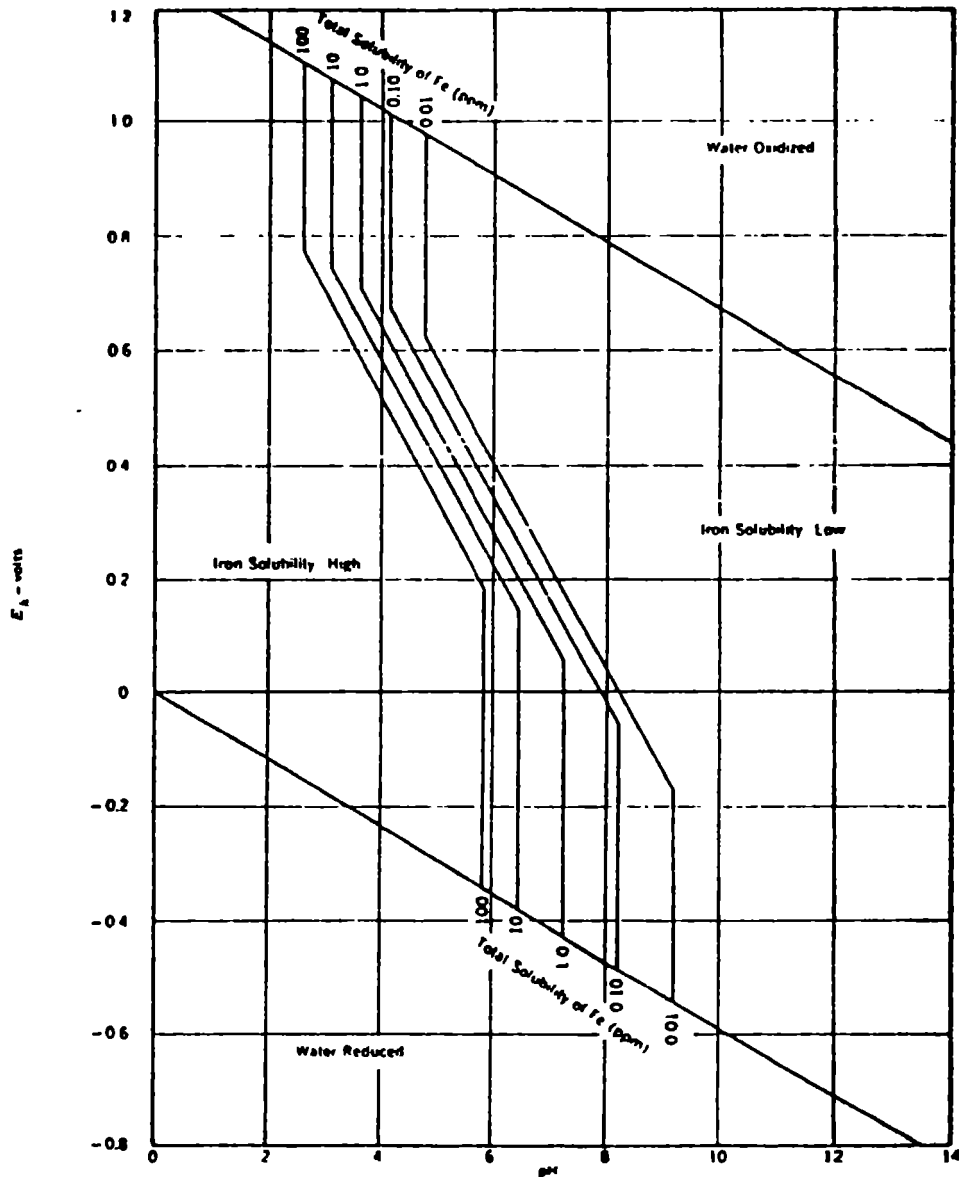


Figure 2.8 E_h - pH diagram for different activities of soluble iron in a carbonate bearing water $C_T = 1,6 \times 10^{-3}$ M (Hem 1961).

2.3 Biological Reactions

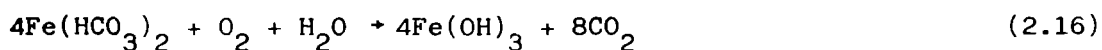
The aerobic as well as the anaerobic underground environment is populated by a large number of different physiological groups of bacteria. In a strongly reduced aquifer bacteria with a potential to reduce oxidized organic compounds, like nitrate, sulphate, manganese (IV) and iron (III) are found in higher concentrations than bacteria with an ammonia, sulphide, iron (II) and manganese (II) oxidizing potential. In a reduced aquifer also bacteria with an oxidizing potential are

detectable (Gottfreund et al. 1985). Recent investigations have also proved that iron bacteria were found in nearly all aquifers. The amount of either aerobic or anaerobic bacteria is found not to depend on the depth of the aquifer (Rott 1985). The development of these bacteria is supposed to be favoured whenever the redox state in the underground environment is changed (Gottfreund et al. 1985).

Iron bacteria due to their alkaline cell surface may support the adsorption and oxidation of iron in water works filters (Rott 1985). However, autotrophic iron bacteria are believed to oxidize Fe(II) to Fe(III) between pH-values of 0,5-3 and gain energy from this process. Above pH value of 4 different conditions should be considered whether iron appears in organic fixation or not. If the pH is raised iron is oxidized by means of chemical reaction and aerobic bacteria are able to gain energy, only if they anticipate the chemical oxidation (Rott 1985).

Other investigation revealed that Fe(II) can be oxidized biologically if pH is 5-6 and microaerophilic prevailed (Frischherz et al. 1985). Investigation on a biological pilot plant showed that, biological defferization developed at pH values of 6,3-6,4 (Richard 1978). Pure culture of Gallionella Ferruginea was also observed to develop in the pH range of 6,3 -6,6 (Kucera and Wolfe 1957).

Among the aerobic iron oxidizing bacteria Gallionella, Crenothrix, Siderocapsa, Leptothrix and Sphaerofilus are well identified. The latter two are heterotrophic requiring organic material as energy source while Crenothrix is facultative autotrophic. Gallionella is strictly autotrophic (Richard 1978). When these bacteria undergo an autotrophic metabolism, they draw their energy from the exothermic reaction (Richard 1978):



The stability of the various types of iron compounds formed in nature will determine the profile of the iron bacteria

selected. Ionic forms of iron can exist where the acidity is low and such soluble forms readily serve the thiobacillus type of iron-oxidizing organism as an energy source. An observation of the growth of *Sulfolobus* bacteria isolated from a sulfur-containing hot spring has been made at low pH values with the presence of Fe(II). Also Metallogenium type of bacteria has been isolated from an acid, iron-bearing stream. Figure 2.9 shows the domains of some iron compounds in terms of E_h and pH and the regions of selection of iron bacteria (Lundgren and Dean 1979).

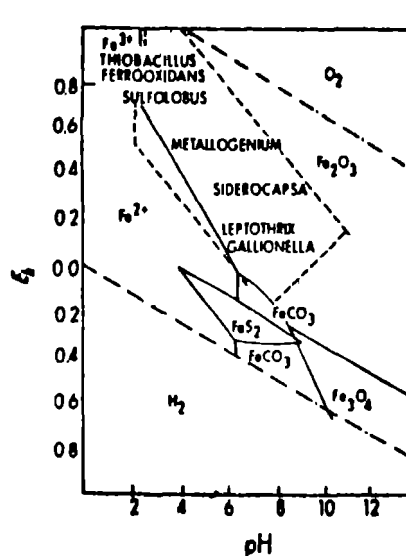


Figure 2.9 E_h -pH iron stability diagram showing the natural domains of the main groups of the iron bacteria (Lundgren and Dean 1979).

Iron carbonates and sulfides are substrates for *Leptothrix* and *Gallionella* organisms. Relatively low levels of these iron-containing compounds can serve as nutrients under conditions of neutral pH, low oxygen partial pressure and low E_h with a continuous iron supply necessary to support microbial growth. Other organisms that function within a short distance of the Fe(II)-Fe(III) borderline as shown in figure 2.9, are the *Siderocapsa* which oxidizes organic complexes of iron. The complexes exist as stable chelate compounds of reduced iron, resisting oxidation to ferric hydroxides. *Siderocapsa* utilizes the organic part of the complex for

nutrition, liberating the Fe(II). A rapid oxidation of the liberated iron occurs, resulting in the formation of ferric hydroxide precipitates which accumulate upon the surface of the cell (Lundgren and Dean 1979).

Subterranean groundwater treatment may also enable bacterial oxidation of Fe(II) complexes and precipitation of Fe(III) under aerobic conditions. The bacteria apparently are able to use iron in organic combinations. Gallionella, Chlamydo bacteria and Siderocapsa are observed (Rott 1985).

The favourable conditions of biological oxidation of iron in a particular pilot plant study was obtained at (Richard 1978):

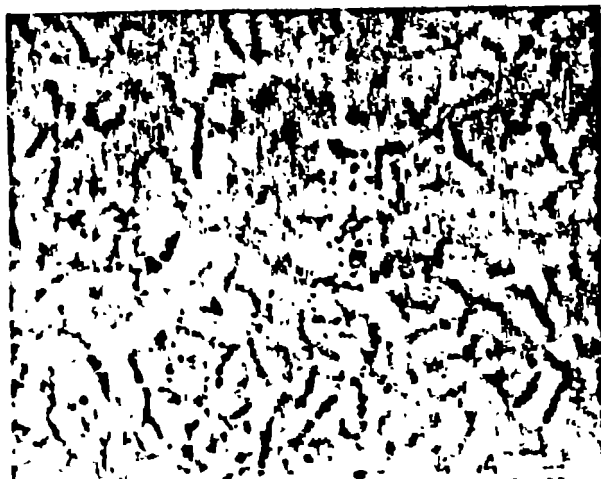
- dissolved oxygen concentration of 0,2 - 2,0 mg/l
- pH identical to raw water, 6,3 - 6,4.
- E_h of 40 - 200 m volts on average
- rH of 14 - 20.

There is not yet any definite principle regarding biological oxidation and precipitation by different iron bacteria. Also the role of the iron bacteria in the process is not well known. This is probably due to little or no experience in the field of biological water treatment application.

However, a recent study made on 15 biological water treatment works (rapid sand filters) in Federal Republic of Germany revealed that iron bacteria were responsible for the oxidation of Fe(II) (Czekalla et al. 1985). In this study the identification of iron oxides, in particular with biological structures was made by using different techniques. The study has been conducted in waters which represent different kinds of water characteristics with Fe(II) concentration varying from 0,05 mg/l to 16,5 mg/l and pH values of 6,5 to 7,9.

The results from this study showed that curved shadow like long thin rod-shaped bacteria were depositing slime (extracellular polymeric substances); which was termed as EPS. The EPS exhibited a high adsorption and oxidation of Fe(II) ions. The shadow structures were found to contain equal

proportions of Fe(II) and Fe(III) ions. Intensive iron oxidation took place on the shadow structures which were no longer seen clearly after certain time as they were completely converted by granular iron oxide hydrate. It was also observed that adsorption and oxidation of Fe(II) proceed at roughly the same time. Figure 2.10 shows the EPS iron oxidation process (Czekalla et al. 1985).



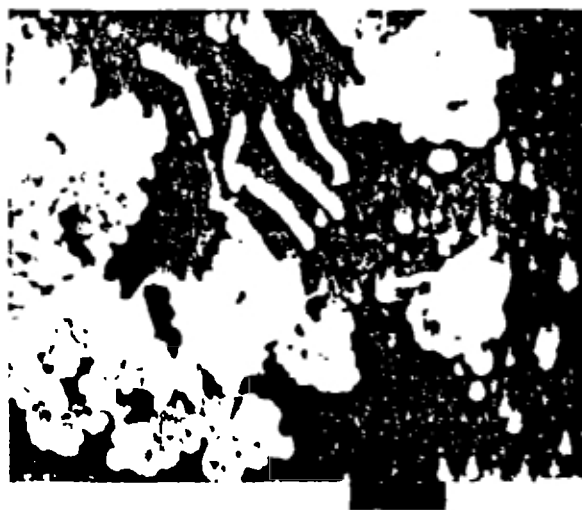
: Curved EPS slime structures produced by bacteria (20 hrs. time growth)



Iron oxidation (light refrangible ferric iron granules) starting at the slime structures (20 hrs.)



: Slime surface completely covered by granules of ferric iron (44 hrs.)

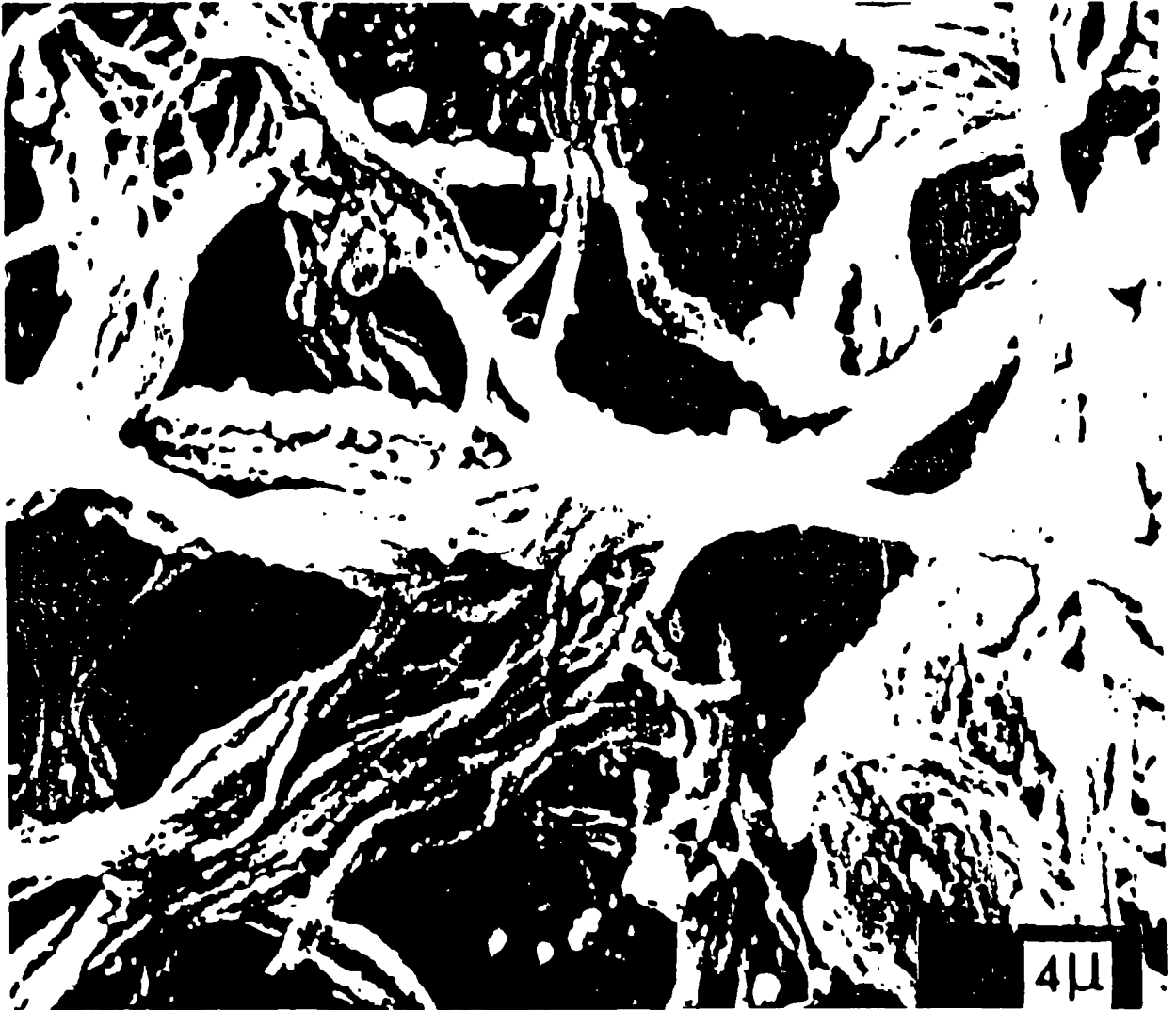


SEM micrograph showing iron oxides and bacteria (26 hrs.)

Figure 2.10 The EPS iron oxidation (Czekalla et al. 1985).

The other type of bacteria identified during the same study was the Gallionella iron oxidizing bacteria. Their bacteria was characterized first by forming ferric iron stalks, which consist of extremely fine ferric iron fibres. They were formed

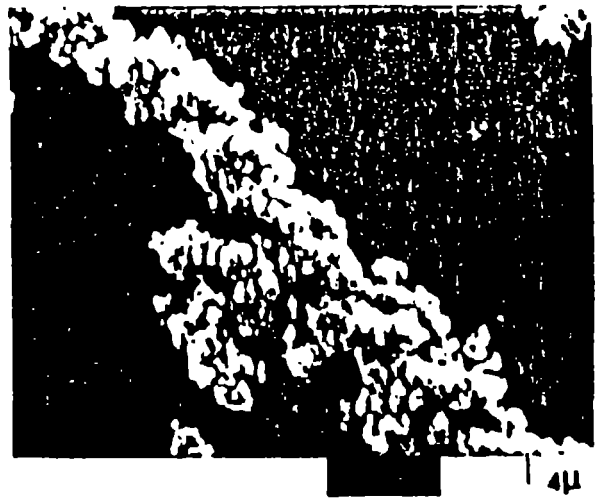
by intracellular iron oxidation, where by the ferric iron produced by the cell was excreted in the form of these fibres. This primary biological iron oxidation of the excreted ferric iron structures with their large surface area was often succeeded by a further secondary iron oxidation which can completely cover the original structures. It was also found that the total stalk length of this bacteria in 1 cm³ of aqueous backwashing material was 4,6 km, which was found to be within the range of ferric iron stalk production in pure cultures of these micro organisms (Czekalla et al. 1985). Figure 2.11 shows the Gallionella iron oxidation process (Czekalla et al. 1985).



: Gallionella ferric iron stalks settling on filter grain, individual ferric iron fibers visible in the twisted stalks (SEM micrograph)



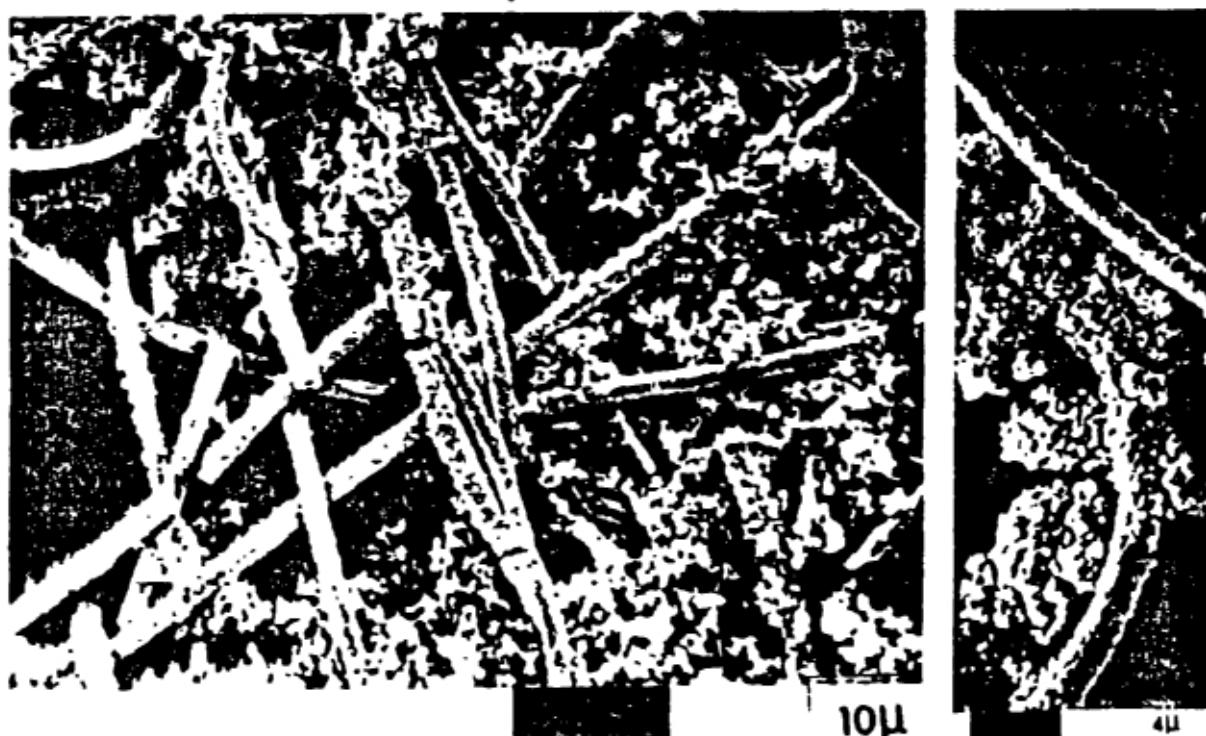
: The backwashing material in the light microscope (bar = 20µm)



: Heavily secondary oxidized Gallionella stalk almost hiding the biological structure (waterwork Hamburg Bostelbek, SEM micrograph)

Figure 2.11 The Gallionella iron oxidation (Czekalla et al 1985).

Also the oxidation of iron by the *Leptothrix ochracea* type of bacteria was observed. This type of bacterial iron oxidation was characterized by a thick mesh of ferric iron sheaths of the micro-organism. The sheath surface was somewhat smooth and it also induced additional iron oxidation. Secondary biological iron oxidation can completely cover the original biological structure. The raw water in which *Leptothrix ochracea* iron oxidation took place differ from raw water with *Gallionella* iron oxidation in its hardness. It was somehow indicated that *Gallionella* iron oxidation required a higher calcium concentration (about 70 mg/l) than *Leptothrix ochracea* iron oxidation (Czekalla et al. 1985). The requirement of calcium by (*Gallionella* was also stated during cultivation of the pure culture of this bacteria (Wolfe 1958). Figure 2.12 shows the *Leptothrix ochracea* iron oxidation (Czekalla et al. 1985).



: *Leptothrix ochracea* iron sheaths, right: heavily oxidized sheaths at higher magnification (backwashing material, SEM micrograph)

Figure 2.12 The *Leptothrix ochracea* iron oxidation (Czekalla et al. 1985).

Another type of iron oxidizing bacteria observed was the *Toxothrix trichogenes*. This bacteria was characterized by typical curved iron oxide structures, which were formed extremely fine, hair pin-like fibres (Czekalla et al. 1985). They were formed by gliding bacterial trichomes, also by the excretion of extra cellular polymeric slime substances. Therefore, this process can be viewed as being closely related to EPS iron oxidation. The difference was that the typical fibre iron oxide structures possessed a high degree of stability and their characteristic form was sheath like curved shape. It was also subjected to subsequent secondary biological iron oxidation through the formation of minute granular ferric iron particles. Figure 2.13 shows the *Toxothrix trichogenes* iron oxidation (Czekalla et al. 1985).



Typical curved hairpin like ferric iron structures of *Toxothrix trichogenes* on the exposed glass slide (in situ ongrowth experiment, SEM micrograph)

Figure 2.13 The *Toxothrix trichogenes* iron oxidation (Czekalla et al. 1985).

In another study made in Austria on a particular dry filtration and flooded filtration pilot plants the presence of extracellular polymeric structures, which was responsible for

the oxidation of iron, was found in isolated pure cultures (Frischherz et al. 1985).

Opinions existing so far tend to differ substantially regarding the biology and physiology of iron oxidizing micro-organisms. This is due to the different views that are taken of the role which physiological and ecological factors play in producing the oxidation reactions. In connection with drinking water treatment, biological methods have been conceived and discussed to some extent in some literatures (Frischherz et al. 1985/Richard 1978). In spite of its long tradition, this type of method still presents a number of unanswered questions, mainly in respect to the quantitative and qualitative development of bacteria in the course of filtering process.

It should be mentioned that the utilization of iron oxidizing bacteria requires optimization of their technological environment and proper handling to ensure high efficiency, whatever these conditions may be.

3 APPLICATION OF IRON REMOVAL PROCESSES - PRINCIPLES AND EFFICIENCY

3.1 Conventional iron removal plants

Presently the main principles of iron removal are chemico-physical methods and to certain extent biological-catalytic techniques. The process of iron removal by conventional methods is mainly divided into three basic categories ie. oxidation of Fe(II), precipitation of the oxidized Fe(II) and removal of the precipitates by sedimentation and/or filtration. Some of the most common combination of different iron removal processes used for different water characteristics are (Cox 1969):

1. Aeration, sedimentation and filtration - for waters containing iron in the absence of appreciable concentrations of organic matter.
2. Aeration, contact oxidation, sedimentation and filtration - for waters where iron is loosely bound to organic matter but no excessive carbon dioxide or organic-acid content.
3. Aeration and contact filtration - where iron is bound to organic matter but no excessive organic-acid content.
4. Contact filtration - where iron is bound to organic matter but no excessive carbondioxide or organic - acid content.
5. Aeration, chlorination, sedimentation and sand filtration - where iron is loosely bound to organic matter.
6. Aeration, lime treatment, sedimentation and sand filtration - where iron is in combination with organic matter, organic acids.
7. Lime treatment, sedimentation and sand filtration - for soft well water devoid of oxygen, containing iron as ferrous bicarbonate.

When dissolved Fe(II) is exposed to air or oxidizing agent oxidation begins, and a transformation into its stable state of Fe(III) takes place. Obviously, oxidation is a method of changing soluble iron to an insoluble state rendering it filterable. The oxidation process can be accomplished by (Cheremisinoff et al. 1976):

- Aeration (O_2 as oxidant)
- Hypochlorites
- Chlorine
- Chlorine dioxide
- Potassium permanganate

Aeration

Aeration is the addition of oxygen to groundwaters to oxidize dissolved iron and manganese to facilitate the removal. It is also commonly referred to as air oxidation. Aeration can be accomplished by different methods and the most common used in groundwater treatment is the gravity aeration which include weir aeration, cascades aeration, and stacks of perforated pans or tower cascades (Pöpel 1976).

The hydraulic load of 0,01 m/s - 0,07 m/s for oxidation efficiency of iron from about 75 % to 90 % is recommended with decreasing hydraulic load. The air to water flow ratio, RQ, generally decreases more than proportionally to the increase of the hydraulic load. The most economical air to water flow ratio is from 60 to 20 (Pöpel 1976).

During aeration, water is exposed to air which oxidizes the iron. The rate of reaction depends on pH; it is more rapid at higher pH values. After aeration, depending on raw water characteristics (Wong 1984) a detention time of several hours may be necessary to remove insoluble iron precipitates. Sometimes sedimentation tanks with sludge collection and removal facilities are used instead of a simple retention tank if the total iron concentration is high (Wong 1984). After the water is aerated and provided with certain detention time, the removal of the rest of the precipitates can be accomplished by

gravity or pressure filters. Air oxidation in some situations is a satisfactory although a relatively slow process (Shrode 1972). Simple aeration is generally ineffective in oxidizing the last undesirable traces of soluble iron. When high concentrations of manganese are present, and when the iron exists as complexes or chelates, aeration is generally ineffective. The investment costs involved have also precluded its use in many systems (Cheremisinoff et al. 1976).

Hypochlorites

This is applied as either sodium or calcium salt hydrolyzed to hypochlorous acid which is a stronger oxidant than the molecular oxygen used in aeration. This enables the soluble iron to be oxidized more rapidly and completely (Cheremisinoff et al. 1976). This process, however, increases the possibility of forming chloroderivatives of organic compounds which cause taste and odor problems as well as imparting a chlorinous taste to water. Hypochlorous acid does not efficiently oxidize organically bound iron (Shrode 1972).

Chlorine

Like the hypochlorites, chlorine must first react with water to form hypochlorous acid, the active oxidizing agent. Thus chlorine is subject to the same limitations as hypochlorites (Shrode 1972).

When hypochlorites or chlorine are used as the oxidizing agents process equipments needed are chemical feed system, sometimes a small retention tank and pH adjustment system as well as equipments for removal of precipitates. Because the oxidation is often slow and incomplete, as for the case of manganese oxidation, the process is normally applied only to iron-bearing waters unless an unusually long contact time is provided.

Chlorine dioxide

This is a strong oxidizing agent which will rapidly oxidize soluble iron. Chlorine dioxide reacts with iron more rapidly than chlorine, but it is not used very widely. This is due to the high cost of chlorine dioxide and the difficulty in the determination of the dosage and handling on site (Shrode 1972). Chlorine dioxide can not be transported, hence it must be prepared at site by means of ClO_2 -generator from sodium chlorite and hydrochloric acid.

Potassium permanganate

The use of potassium permanganate for the removal of iron is very effective because the reactions are complete, rapid and only require a minimum quantity of chemical. Theoretically 1 mg/l of potassium permanganate will oxidize 1,06 mg/l of Fe(II), provided that no other contaminants causing a potassium permanganate demand are present. Potassium permanganate will rapidly oxidize organically bound iron (Shrode 1972). It can be used in either gravity or pressure filtration water treatment system.

Technical considerations

The choice of an oxidizing agent is not only limited to the question of the removal of iron and manganese but also depends on general usefulness and the effects on other problems, such as turbidity, odour etc. Also the sequence of chemical addition, if more than one type of chemical is used in a water treatment, must be done based on some general observations and to obtain the required reaction time. For instance, potassium permanganate should ideally be added early in the system so that its reaction is virtually completed before the addition of chlorine, coagulants and other treatment chemicals.

The different types of iron oxidizing methods are usually followed by sedimentation and/or filtration. The overall diameter of iron precipitates varies generally from 5-100 μm ($5 \cdot 10^{-4}$ - $1 \cdot 10^{-2}$ cm) (Olson and Twardowski 1975). As the specific gravity of iron particles is small the settling velocity is low, may be 10^{-4} - 10^{-2} cm/s, and hence proper considerations must be made when designing sedimentation tanks for iron removal.

In certain instances, the use of slow sand filter is justified and may yield satisfactory results, but it cannot be effectively used for the removal of large amounts of iron from water due to rapid clogging. Filters often increase in efficiency due to the encrustation of the grains of the medium with higher oxides of iron which act as catalysts (Holden 1970). The development of biological growths, including iron bacteria, on the surface layers of slow sand filters, also play an important role in the removal of iron by such units (Holden 1970).

More often, rapid filters, either gravity or pressure filters, are applied after oxidation as a final treatment step with or without sedimentation (or flotation). Type and dimensioning of filters depends on treatment prior to filtration, the lower suspended solid concentration is, the longer filter run and finer filter material can be applied.

3.2 Other iron removal methods

3.2.1 Vyredox method

This method was first developed in Finland and now used in some other countries like Sweden, Netherlands etc. In this method Fe(II) iron is oxidized in the ground to the insoluble Fe(III) form (Hallberg and Martinell 1976):

The Vyredox method achieves a high degree of oxidation in the aquifer around the well. In other words, the E_h and pH are kept so high that the iron is precipitated and retained in the strata. Figure 3.1 shows the transition between the oxidized and reduced states of iron and manganese (Techlinger et al. 1985).

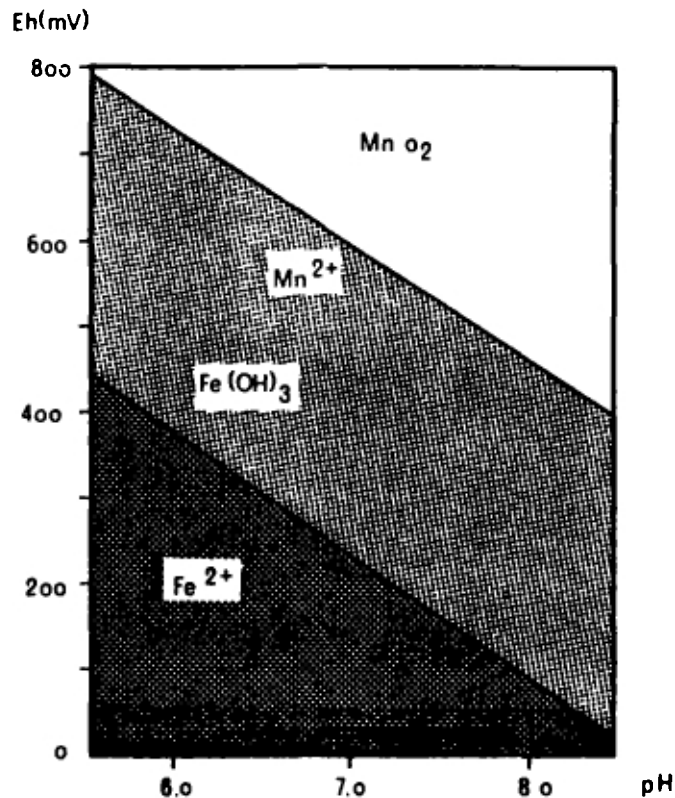


Figure 3.1 Dissolved Fe(II), Mn(II) and precipitated Fe(OH)₃, MnO₂, iron and manganese as a function of the variables pH and E_h (Techlinger et al. 1985).

In figure 3.1 the lower of the two sloping lines show the balance between Fe(II) and Fe(III). At every point on the line the concentration of the two states is equal. For all values of pH and E_h under the line, the concentration of Fe(II) is greater than the concentration of the Fe(III). To remove both iron and manganese both the pH and E_h must be raised above those presented by the upper line. The lines can vary depending on the concentration of other substances (Techlinger et al. 1985).

This method fits in with the needs of iron and manganese oxidizing bacteria. Iron is precipitated first, in the zone

furtherst from the well. The number of living bacteria increases, and so does the number of dead bacteria. The organic matter contained in some of them follows the water flowing towards the well. It becomes a source of carbon for the bacteria that preferentially oxidize manganese. This process takes place nearer the well, where the E_h is higher, that means, the method provides favourable condition for removing iron first, and then manganese (Hallberg and Martinell 1976). The bacteria which oxidizes iron and manganese develop if oxygen is present in the water in small amounts (Grombach 1985).

The introduction of air or oxygen into the aquifer is accomplished through vertical boreholes by injecting oxygen saturated and iron free water. The injection of preaerated or oxygen saturated water is through a number of injection wells situated in a ring around the supply well or through the supply well itself (Techlinger et al. 1985).

Continuous pumping from the supply well is not possible even if injection wells are available. Pumping from the supply well during injection of oxygenated water would cause a direction of groundwater flow straight to the supply well and an equal distribution of the oxygenated water in the oxygenated zone would be impossible (Rott 1985). In general the method works as shown in figure 3.2 according to the following procedures (Hallberg and Martinell 1976).

- a) The pump in the supply well needing a Vyredox treatment is stoped
- b) Water is pumped up from another supply well through the aerator in which the water is disgassed to remove the carbondioxide, methane, etc. and enriched with oxygen.
- c) It is then forced back into the well according to point (a) and also in the surrounding injection wells. This normally goes on for 20 hours.

- d) A four-hour contact time follows.
- e) Purified ground water can then be pumped up out of the treated supply well and transformed directly to the mains.
- f) After a time, normally 2-4 weeks depending on the actual circumstances, the uprising water will begin to show slight traces of iron and manganese. This means that the well has to be treated over again according to point (a) to (f).

When wells are operated in this way the following conditions prevail around the well (Hatva et al. 1973):

- with the artificial oxidation, a steady gradient is formed in the ground around the zone of the supply well, where iron is precipitated and absorbed in the soil on the oxidated side and soluble on the reduced side.
- the gradient zone can be found in the observation (injection) wells by means of E_h and pH measurement.
- filamentous iron and manganese bacteria (Gallionella, Leptothrix) are found abundantly in the gradient zone.

Figure 3.2 shows principles of the Vyredox method (Hallberg and Martinell 1976).

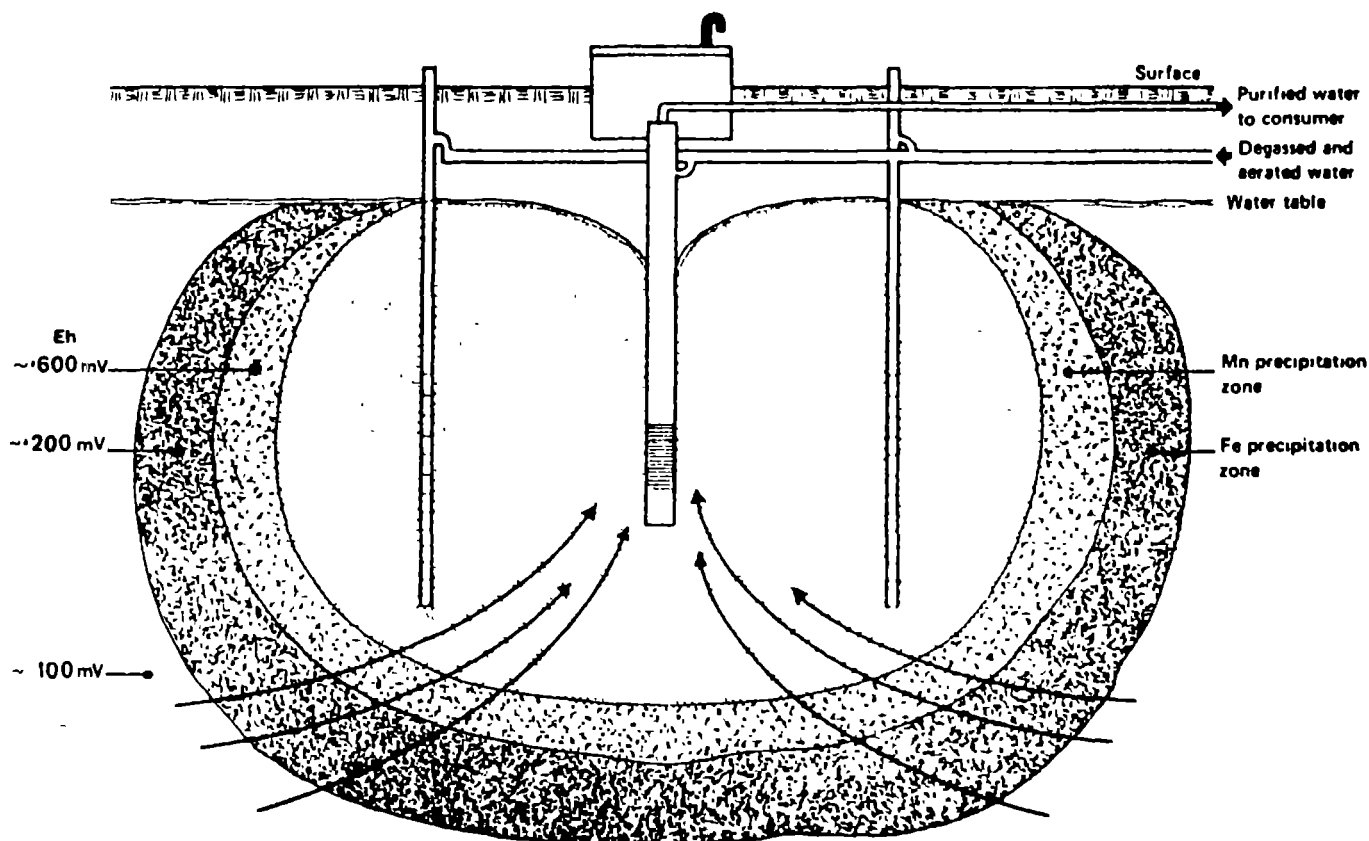


Figure 3.2 Principles of the Vyredox method (Hallberg and Martinell 1976).

During the injection of oxygen-rich water the dissolved oxygen is consumed by all oxidizable compounds. Due to the consumption of the dissolved oxygen, the injected water will spread further from the injection well than the injected oxygen. The degree to which the oxygen front lags behind the injection front depends on the quantity of oxidizable compounds in the solid phase of the aquifer (Beek 1985).

At the beginning of each abstraction, a part of the oxygen injected, i.e., the quantity which is still present in solution between the oxygen front and the well, will be pumped back. During further abstraction the iron hydroxides and manganese oxides will adsorb Fe(II) and Mn(II) dissolved in the slowly moving ground water. In the same manner in which oxygen front remains behind the injection front during the injection, during abstraction the Fe(II) and Mn(II) fronts will lag behind the abstraction front due to interaction with

the iron hydroxides and manganese oxides developed. The Fe(II) and Mn(II) front will advance slowly through the zone in which the oxides formed during the injection are present. As soon as one or both of these fronts begin to approach the well, the concentration of iron and/or manganese in the water abstracted will increase and, once again, oxygen-rich water will need to be injected. During this injection of oxygen-rich water the adsorbed Fe(II) will be converted to Fe(III) and Mn(II) to Mn(IV) and so the process continues (Beek 1985). Fe(II) is more strongly adsorbed than Mn(II); thus, a special separation takes place between the two. Iron hydroxide will form furthest from the well, manganese oxide will accumulate between the well and the iron hydroxide formed. Figure 3.3 shows schematic overview of the processes occurring in the aquifer during iron and manganese removal (Beek 1985).

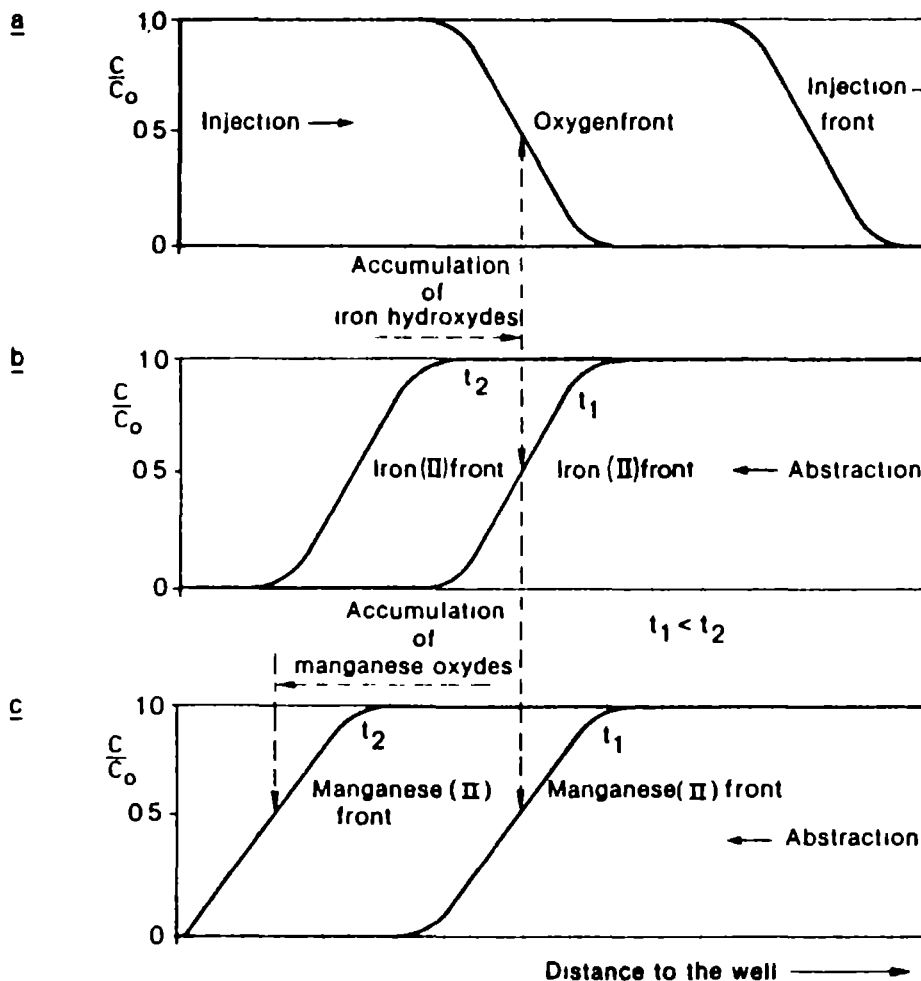


Figure 3.3 Schematic overview of the processes occurring in the aquifer during iron and manganese removal (Beek 1985).

It takes time for the oxidizing zone to become fully efficient. The cycle of withdrawing groundwater, aerating and injecting it, must be repeated many times. Figure 3.4 shows how the efficiency increases with the number of cycles (Hallberg and Martinell 1976).

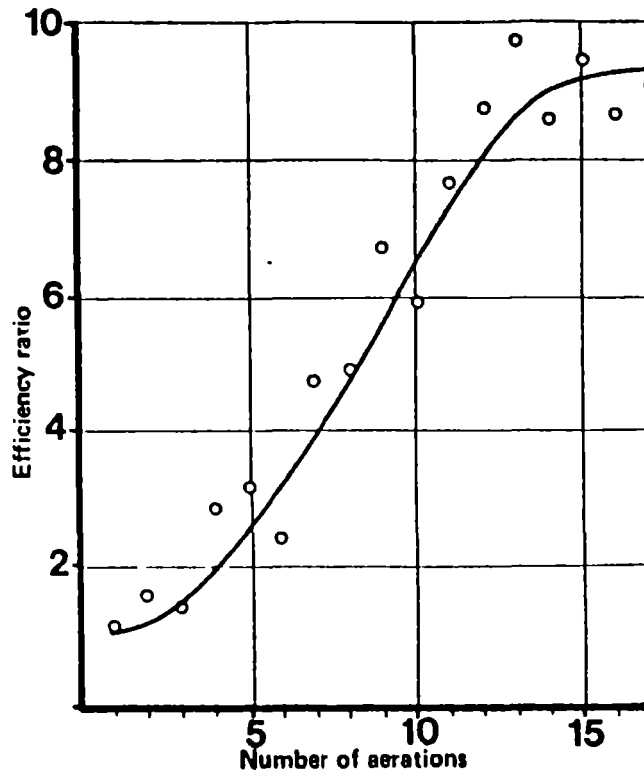


Figure 3.4 The efficiency ratio of a Vyredox plant increasing with the number of aerations (Hallberg and Martinell 1976).

In this process the precipitation of iron takes place in the ground and in most cases it is believed that the deposits of the precipitates will not reduce or block the permeability of the aquifer (Hallberg and Martinell 1976). However, since this method is quite new and the process of precipitation takes place in the ground, more investigation might be required to know its exact effect on the permeability of the aquifer.

3.2.2 Overland flow

Overland flow can also be applied in water treatment as iron removal process. To maximize the iron removal overland flow must accomplish two objectives (Zirschky and Carlson 1984).

1) Oxidation of Fe(II) to Fe(III)

2) Sedimentation-filtration of the resulting iron precipitate

The source of the groundwater and the degree of pretreatment before overland flow will determine the influent Fe(II) concentration. The DO (dissolved oxygen) concentration and pH are the two variables of primary concern in evaluating the potential of overland flow for Fe(II) oxidation. To increase the oxidation rate, the pH and DO concentration of the groundwater must be increased. The DO concentration is increased by natural gas transfer (Zirschky and Carlson 1984). Overland flow provides a large area through which gas transfer can occur. Therefore, the potential for the transfer of oxygen into solution and the dissolution of CO₂ from solution to occur should be relatively high. As the pH value and DO concentration of water increase, the Fe(II) oxidation rate also increases. Thus, by providing a large surface area for gas transfer, overland flow promotes the oxidation of Fe(II) (Zirschky and Carlson 1984).

Sedimentation or filtration is the other aspect of overland flow that must be successful for good iron removal. Theoretically, clarification is optimized by the use of shallow sedimentation basins, and an overland flow slope is, in effect, a shallow sedimentation basin. Particles need only settle a short distance before intercepting the slope, thereby being removed. The vegetative cover intercepts many of the particles, providing additional suspended solids removal. Care must be taken, however to avoid scouring the slope.

The information as to how the overland flow functions is very scarce. However, the overall iron removal efficiency of the overland flow, as it was applied in Salo water treatment in Finland, was found to be more than 90 %. The quality of the water was not affected significantly with respect to the drinking water characteristics (Zirschky and Carlson 1984). Figure 3.5 shows the plan view of the overland flow of Salo water treatment plant (Finland) (Zirschky and Carlson 1984).

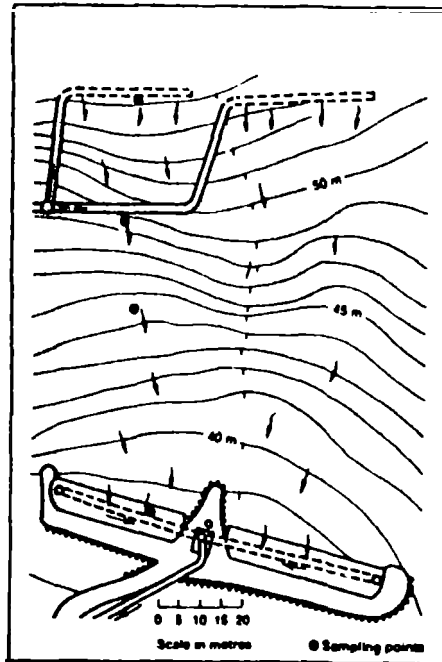


Figure 3.5 Plan view of overland flow of Salo, Finland, water treatment system (Zirschky and Carlson 1984).

The disadvantage of overland flow iron removal process might be external contamination of the water, although it was stated that the quality was not affected significantly. The other aspect is that iron bacteria might participate in the process, as the system seems to create favourable condition for them to grow.

3.2.3 Miscellaneous iron removal methods.

Other iron removal principles include contact beds, ion exchange method, diatomite filters, and some other methods which are rarely applied.

Contact beds

The purpose of contact bed is to facilitate oxidation of iron or manganese through the catalytic action of previously precipitated oxides of these minerals on the gravel or ore. The common ore used in this process include manganese ore and pyrolusite, which is anoxide of manganese (Cox 1969). As

iron-bearing water passes through such a filter, the iron comes in contact with the medium, and oxidizes to form insoluble ferric iron. The resulting precipitates are then trapped in the filter bed. As the precipitates build up in the filter, it will eventually clog up and, must be cleaned by backwashing. Backwashing will be required more or less frequently, depending on the size of the filter, the amount of iron in the supply and the quantity of water used. As contact beds depend on the catalytic action of oxidized iron or manganese deposits on the filter or on the effect of oxidizing nature, the beds should never be washed clean, but deposits should be removed at intervals (Leher et al.1980).

When the water is devoid of dissolved oxygen, the medium participates directly in the reaction. In this instance, the higher oxides of manganese release oxygen to the adsorbed iron, causing oxidation and precipitation. Periodic regeneration of the bed with potassium permanganate is necessary to maintain an adequate supply of the manganese oxides on the coating (Leher et al. 1980).

Ion exchange method

The basic principle of the ion exchange process involves physical contact between mineral ions in the water with more active ions located on a natural or artificial media (resin). The most common media include manganese zeolites, carbonaceous zeolites or artificial plastic resins. Upon contact, the mineral ions in the water replace ions (usually sodium) present on the exchange resin within the reactor. When the sodium ions on the resin have been depleted the resin is recharged with a salt brine solution that reverses the above reaction (Furgason and Day 1975). Ion-exchange softeners should be considered for hard waters containing iron and/or manganese, provided that the raw well water is devoid of oxygen as the process removes only soluble iron and manganese together with calcium and magnesium (Cox 1969). The exchange resin is often adversely affected by insoluble iron and manganese compounds. These compounds tend to physically bind themselves to the resin and resists removal by conventional

regeneration processes. This condition becomes more prevalent with prolonged usage and eventually lowers the exchange capacity of the resin (Furgason and Day 1975).

This process should not be used, due to the above mentioned reasons, when the iron and manganese content exceeds 0,5 mg/l for each 17 mg/l of hardness, upto a maximum of 10 mg/l. This establishes the lower limits of hardness as 340 mg/l for an iron content of 10 mg/l (Cox 1969). Water softeners should be applied to remove iron when the water is clear when drawn and no iron bacteria are present (Leher et al. 1980).

Diatomite filter

The filter media used in diatomite filter is manufactured from diatomaceous earth deposits which consist of the siliceous fossil remains of dead diatoms. The processed materials have unique structural properties which make them particularly suited for use as filter media in water filtration (AWWA 1971).

Present experience relates principally the diatomite filters to turbidity removal from surface water but experience is growing with installations designed for iron and manganese removal from groundwaters. The method in iron and manganese removal employs calcined magnesite (MgO) and diatomaceous earth which are fed to a rapid mixing tank. In the tank 5 to 10 minutes of contact time is provided during which Fe(II) and Mn(II) are oxidized in contact with the MgO. Thereafter, the water is filtered through a diatomaceous earth filter. An advantage of this system is that the filter media is discarded after each filtration cycle, thereby avoiding any accumulation of precipitates or bacterial growth on the filter media. The filter is backwashed after the filter cake is removed (AWWA 1971).

4 DESCRIPTION OF DRY FILTER

The application of dry filter in groundwater treatment is mostly for the removal of iron, manganese, ammonia and some other contaminants. Information regarding the processes involved in dry filtration on iron removal are very scarce. This is perhaps because dry filters have not been traditionally used for this purpose.

4.1 Principles of iron removal

Dry filter is a gravity filter except that the filter media is not submerged in water and hence the name "dry filter" is given. The overall removals of iron and other impurities associated with the process of dry filtration are brought about by a combination of the different filtration phenomena and greatly by biological activity (Huisman 1977).

The principle involved in the dry filter was first explained to be of catalytic action. This theory assumes that through the higher velocities in the filterbed with turbulent flow, the thickness of the layer of liquid adhering to the surface of the crystals of the filter material diminishes. The electric double layer which surrounds every grain may then lose a great number of positive ions, supposing that the first adhering layer consists of negative ions. It is also explained that, there is an equilibrium for every rate of flow between the number of ions being carried along with the stream and the number of ions caught again in the series of ions of the electric double layer at the vacant spots. As these spots are being taken up by higher-valent ions, or ions from equivalent series in the periodic system, such as Fe(II), Fe(III) and Mn(II); these will be bound more strongly than the lower valent ions or those from the principal series of the periodic system. Gradually the lower-valent ions will therefore be replaced by higher-valent ones. It is also stated that the speed of the reaction being directly proportional to the concentration of the reacting ions the reaction at the boundary of solid and liquid will be much higher than in the centre of the stream, and higher for turbulent than for

centre of the stream, and higher for turbulent than for laminar flow (Kooijmans 1955).

On the other hand iron removal principle in dry filter is revealed to be more of biological phenomena. Micro-organisms naturally present in the raw water or purposely added to it are adsorped on the filter material, where they multiply selectively using available iron and manganese ions. In dry filter the water is not completely saturated with oxygen, therefore, favourable condition for iron and manganese bacteria is created (Grombach 1985).

If oxygen is scarce but not totally absent, which is quite often the case, a special kind of microbe develops. This obtains energy for its metabolism from the oxidation of dissolved iron and manganese to insoluble salts. Its effect is known to many water works as an objectionable clogging of groundwater filters and boreholes by the precipitation of brown iron and black manganese (Grombach 1985). The other advantage of dry filter is that the presence of air in the pores of the filter bed increases the actual velocity at which the water moves down. This means stronger cross-currents and a greater chance for Fe(II) and Mn(II) ions and suspended particles to come into contact with the filter media and thereby to the micro-organisms and also to the catalytic surface action of the filter materials which promotes filtration efficiency. This is the reason why dry filtration is also preferred when the presence of organic matter prevents spontaneous deferrisation (Huisman 1977).

The principle of iron removal by dry filter is confirmed to be more of biological process on a pilot plant study made in Austria (Frischerz et al. 1985). In this study microbiological investigations showed that the micro-organisms contained in the untreated water occupied the available filter surface within short time after the supply of the raw water to the filter. On studies of the micro-organisms extracellular polymer structures were found in isolated pure culture. These micro-organisms responsible for extra cellular polymers were found to be time dependent and hence the maturation of the

filter. Manganese depositing bacteria were found only sporadically at the beginning of the filtration run, their number rose along with increasing age of the dry filter to form certain regular percentage (5 to 12 %) of the biological slime (Frischherz et al. 1985). It was also suggested, that in the upper portion of the dry filter where the greater part of the iron mud is deposited, the activity of the micro-organisms, although their presence has been confirmed, is strongly affected (Frischherz et al. 1985).

The iron removal process was mainly determined by three factors (Frischherz et al. 1985):

- configuration and area of the filter surface as held-fast for a biocenosis.
- residence time of the water
- usable volume of voids for iron deposition.

The filtration rate applied varies from 3 to 10 m/h depending on the concentration of iron in the raw water. In waters with very low iron concentrations, (for instance 0,2 mg/l of Fe) matured dry filters can operate at higher filtration rates (up to 60 m/h) without causing any break through of iron (Frischherz et al. 1985).

The start up behaviour of the dry filter was also investigated during the pilot plant study in Austria (Frischherz et al. 1985). Accordingly, in a dry filter supplied at a rate of 3 m/h, iron removal was seen to occur immediately after the beginning of the test, which was suggested to be of a chemical nature. But micro-aerophilic were found in practically all the samples observed. Manganese removal or ammonium oxidation did not take place during the same time as iron removal. It was shown that there was a time lag between ammonium oxidation and manganese removal. Figure 4.1 shows the start up behaviour of a dry filter; elimination of Fe, Mn and NH_4 - oxidation (mg/l). The numbers 1 - 5 show the maturing phases of the new dry filter with respect to the elimination of the shown constituents.

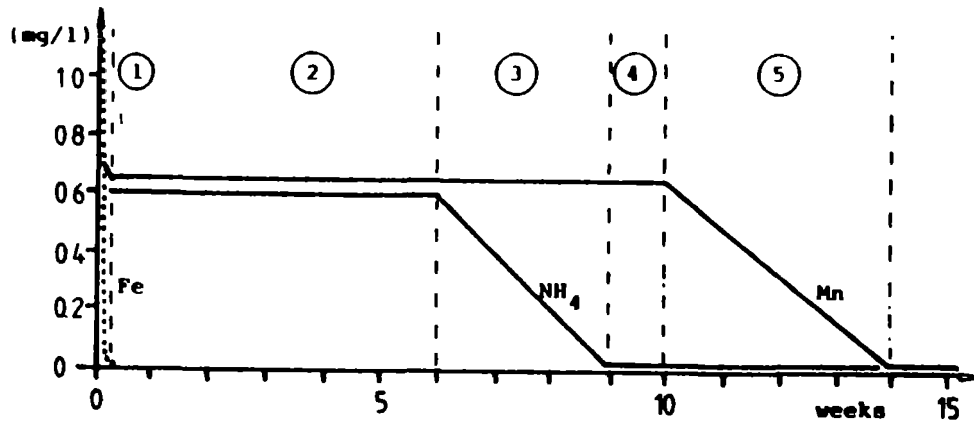


Figure 4.1 Start-up behavior of a dry filter; elimination of Fe, Mn and NH₄-oxidation (mg/l) (Frischherz et al. 1985).

The oxidation of ammonium to nitrite and nitrate started after longer time operation (more than six months). After oxidation of ammonium, one week later the complete demanganisation lasted another four weeks. After the end of the 14-week start up period, the oxidation rate of the dry filter remained unchanged. This can be seen from figure 4.1. Tests using other filtration rates showed these to have little impact on the time pattern, whereas the ultimate iron, manganese and ammonium concentrations reached at the outlet were greatly dependent on filtration rate.

4.2 Constructional details

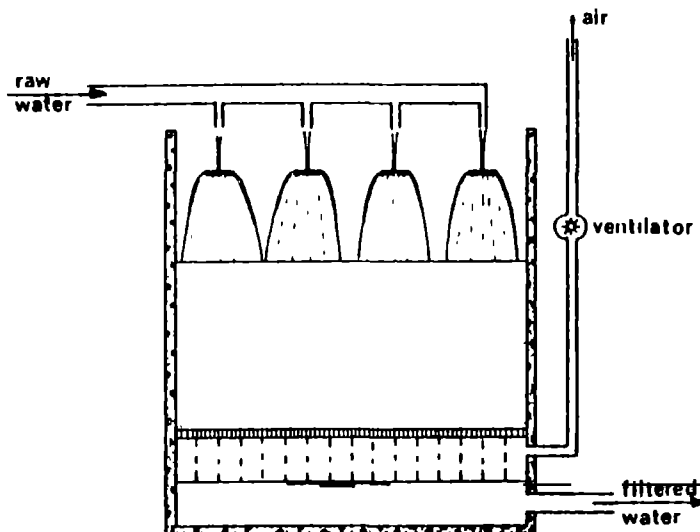
4.2.1 Water inlet

The raw water is sprinkled through fixed-nozzle distribution system or through a perforated pipe. The fixed-nozzle distribution system consists of a series of spray nozzles located at the points of equilateral triangles covering the filter bed. A system of pipes placed in the filter distributes the raw water uniformly to the nozzles. Special nozzles having flat spray patterns (Dresden type) are commonly used, and the

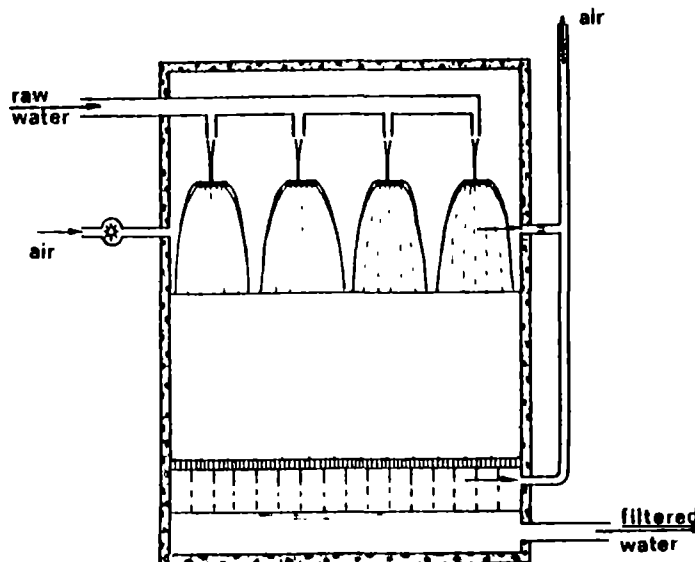
head is varied systematically so that the spray falls first at a maximum distance from the nozzle and then at a decreasing distance as the head slowly drops.

4.2.2. Filter box

Dry filters can be constructed as open or closed filters (Kooijmans 1955). Figure 4.2 shows dry filters with open and closed construction system (Huisman 1977 a).



a) open construction



b) closed construction

Figure 4.2 Dry filter with open construction, (a), and with closed construction, (b) (Huisman 1977).

Construction materials for dry filter can be concrete, ply wood, steel bars etc. provision of sufficient slots is very essential in the case of open construction. In closed construction, supply of air can be provided by a simple house ventilator or by section under the filter bed (Kooijmans 1955). The depth of the filter depends on the iron concentration of the raw water and can be above 1,5 m.

Shallow tank is provided at the bottom of the filter box for collecting the filtered water. A small gap is provided between the top surface of the filtered (collected) water in the basin and the bottom of the filter box to allow ventilation. In addition to filtered water collection the basin might serve as a sedimentation basin. However, care must be taken when determining the surface loading due to the different characteristics of iron flocs from other particles.

The supernatant water from the collection basin is taken by a pipe to clear water storage tank or to a secondary treatment process depending on the iron content of the effluent. It can also be collected by weir constructed around the collection basin and lead to the required pipe. The basin can be constructed from steel plates or reinforced concrete.

4.2.3 Ventilation system

Natural ventilation can be enough for dry filter provided that the open area of the slots are adequate to allow enough air. The most important effect of the air introduction into the filter is the stimulation and maintenance of biological activity. In the dry filter the raw water percolates downward through the filter bed, accompanied by an equal or a bit smaller or larger amount of air from which the oxygen consumed is replenished immediately allowing the removal of the required constituents (Huisman 1977). The close contact of the water with both the filter material and the air is conducive to various desired processes in the dry filter such as elimination of iron and manganese, oxygen enrichment, removal of humates, ammonium oxidation, mechanical deacidification, desorption of gases etc. (Frischherz et al. 1985).

For open construction type of dry filter the atmospheric air can flow through the porous media of the filter. If the filter material is packed in casing without any sufficient slots or perforations of the filter box, ventilation has to be provided by additional airblower.

4.2.4 Filter media

The ideal filter medium is a material that has a high surface area per unit of volume, low in cost, a high durability and does not clog easily. Suitable material is generally a locally available crushed stone or gravel graded to a uniform size within the range required. Other materials such as traprock and crushed foamed slag have also been used. For deferrisation the use of broken material is supposed to be advantageous as the process involves removal of positively charged particles, i.e. for catalytic action that may take place in the filter (Huisman 1977). Stones small in size do not provide sufficient pore space to permit free flow of raw water. Clogging of the media and "ponding" inside the filter or at the surface will result. Large diameter stones avoid the ponding problem but have relatively small surface area per unit volume; thus they cannot support large biological population. Size uniformity is a way of ensuring adequate pore space. The sizes of the material so far used in dry filter varies greatly. According to available information and observations made the variation is from

1-2 mm (Greene 1970); 2-3,5 mm (Kooijmans 1955), 4-10 mm and 10-20 mm (Frischherz et al. 1985) and 40-50 mm as in Raahe.

4.3 Filter Cleaning

The operating conditions of the dry filter such as filter medium, particle size, filtration rate and height of filter are governed by the iron concentration and the resulting deposits. The suspended matter (if any) together with iron precipitates and biological iron sludge is deposited in the available pore space on the filter gravel. The surface area of the filter gravel available for such purpose depends on the size of the material and available pore space (Huisman 1977).

If the filter is sufficiently porous, there is plenty of room between the gravel for the precipitated iron and manganese, so that danger of clogging is small.

The length of one run of the dry filter depends on the filtration rate when smaller sizes of filter material are used (4-20 mm) (Frischherz et al. 1985). If larger sizes (40-50 mm) are used or smaller filtration rate (less than 6 m/h) is applied, the end of the filter run cannot be marked by the passage of iron through the filter or by clogging of the filter by iron deposits. For filtration rates greater than 6 m/h flow washes out the iron mud and requires subsequent removal of washout (Frischherz et al. 1985). Although in some cases the backwashing is processed in dry filters, there is not as such filter resistance phenomenon. Washing is required if there is ponding or clogging of the filter by iron deposits.

5 PILOT PLANT STUDIES

5.1 Objectives for the study

The application of dry filter for iron removal from groundwater is believed to be of biological process. Studies have been made in order to identify the type of micro-organisms involved in the process and some design criteria for their development and applications of dry filtration system are proposed. The criteria so far established tend to differ. The difference is probably due to the following reasons.

- As the process involves micro-organisms and the characteristics of the raw water are different; the adaptation of the micro-organisms to the different conditions give different results.
- The investigations made so far were based on different situations. The use of different sizes of filter material, the variation in iron concentration of the raw water, the arrangement of ventilation system and some other environmental factors affect the results and hence different criteria were obtained.

In order to develop general criteria for the design and application of dry filter intensive investigation must be done on different types and sizes of filter materials, with different iron concentrations and other factors required for the process. The purpose of this study was to establish certain design criteria for the following conditions under this particular study.

- iron removal efficiency with respect to surface hydraulic load
- to identify the type and development of the micro-organisms
- influence of filter depth on iron removal

- the effect of dry filtration on general water quality
- the effect of difference in iron concentration in raw water to removal efficiency for the same sizes of filter material
- iron removal efficiency with respect to different sizes of filter material.

Other conditions such as ventilation system, raw water distribution on the surface of the filter and filtered water collection methods were proposed.

5.2 Test plants

The experimenta was conducted on two pilot plants and one full scale plant. Alterations on the full scale plant have not been possible but follow up of the process was made. The full scale plant is situated in Raahe and was put into operation in late 1984. The pilot plants were situated in Haukipudas and Siikajoki, all in Finland. Separate description of each plant is given below.

Haukipudas:

The pilot plant in Haukipudas was put into operation on 16/09/85 and run up to 13/12/85. The plant was constructed of column of rigid PVC pipe diameter of 400 mm. The height of the pipe column was 2,0 meter and filled with natural stones of sizes 40-50 mm. The depth of the filter material was 1,6 m and it was increased to 1,8 m after 2 months of filter run. The water was sprayed over the filterbed by shower-spray nozzle. Raw water was supplied from the actual inlet of the main water treatment plant and due to the plants operation mode, supply was interrupted for 4 to 6 hours a day. The surface area of the filterbed was 0,126 m². Air was provided by slots of 5x10 cm sizes provided around the pipe column. Later on additional aeration system was provided by a small diameter flexible plastic pipe connected to the air pipe line of the main plant. Figure 5.1 shows the pilot plant in Haukipudas.



Figure 5.1 The pilot plant made of PVC pipe in Haukipudas

Raahе:

The plant in Raahе is a full scale plant dimensioned for capacity of 500 m³/d and consisting dry and slow sand filters. The surface area of the dry filter is 7,5 m² and height is 1,8 m. The size of the filter material was 40-50 mm of crushed stones. Raw water was sprayed over the filter by nozzles provided with flat spray patterns. The plant was housed. However, since the house was not heated and free circulation of air was existing, freezing effect on sidewalls of the filterbox prevailed. The type of construction material was similar to the pilot plant in Siikajoki except that filtered water was collected into a small settling basin prior to the slow sand filter unit.

Siikajoki:

The pilot plant in Siikajoki was constructed in such a way that it can also be used as a production plant after the completion of this study. The plant was put into operation on 16/09/85 and continued after the study period. The total surface area of the plant was 3,5 m² and height of 2,0 m. The net filter depth at the beginning was 1,8 m and latter increased to 2,0 m. The filter box was constructed of flat-grid steel bars with holes all over the surface. Under the filter box was provided a small water collection chamber constructed of steel plates. An outlet pipe was connected to one end of the collection chamber. Figure 5.2 shows the schematic flow through process of the Siikajoki pilot plant.

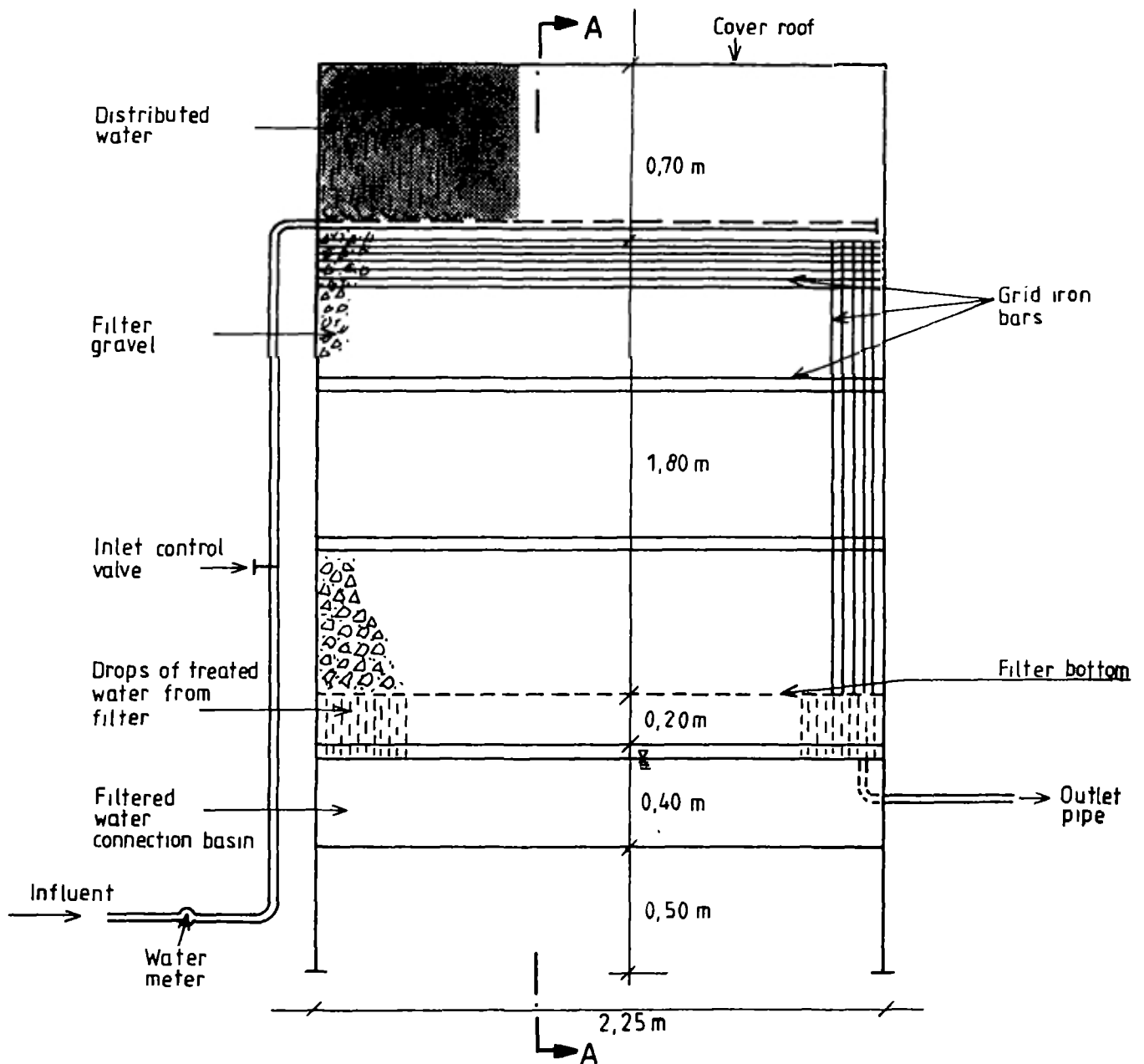


Figure 5.2 Schematic presentation of flow through process in dry filter of Siikajoki pilot plant (Note drawing in not to scale).

Tests were carried with crushed stone of diameter 40-50 mm and later replaced by similar type of stones of diameter 10-20 mm. The raw water was pumped through a pipe provided by a water meter. The water was sprayed over the filterbed through a perforated pipe nozzles. The perforations were made in such a way that a central pipe can supply water all over the filterbed surface. The water coming through the perforated nozzles was sprayed upwards and collided with the cover roof,

where it breaks into smaller droplets thereby spreading over a larger area. After the water was sprayed over the area it starts to percolate through the filter media. In this filter intermediate sampling pipes were inserted into the filter body at depths of 0,6 and 1,2 meters initially from top and 0,8 and 1,4 meters latter after the increment of the whole filter depth by 0,2 meters. The plant was first put into operation in open air but latter on due to freezing problem isolation tent and electrical heating was constructed. Figure 5.3 shows the distribution of the raw water on the filter surface.

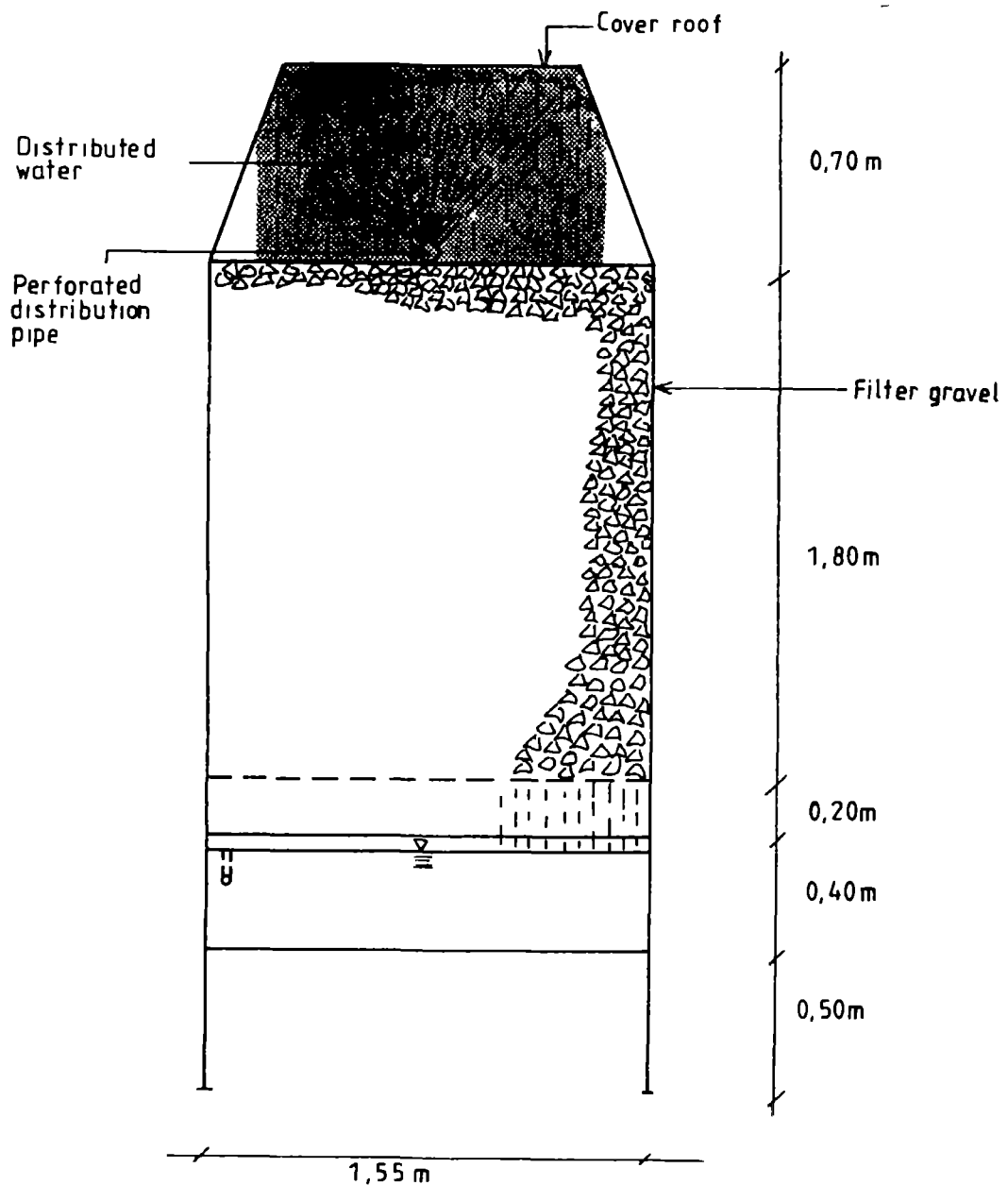


Figure 5.3 Section A-A of figure 5.2 showing the water distribution pipe and the cover roof breaking the water into smaller droplets (Note the drawing is not to scale).

5.3 Sampling and analysis procedures

Samples were taken from raw and treated waters. In Siikajoki samples were also taken from intermediate sampling pipes installed on the filter body of the test plant.

Fresh samples were transported to the laboratory, the time difference between the collection and analysis of the samples was usually less than half a day.

Parameters such as pH, temperature and redox potential were measured at site during sampling. Other parameters analysed during the study were manganese, ammonia, hardness, alkalinity, carbondioxide and dissolved oxygen. Iron was analysed by spectrophotometric method. All other parameters, including iron, were analysed according to the standard methods 14th edition as modified to Finnish laboratory standard.

5.4 Raw water characteristics

The raw water characteristics in the three places differ from each other. Iron concentration in the raw water of Haukipudas was from 16 mg/l to 20,6 mg/l, in Raahe from 0,43 mg/l to 2,43 mg/l, and in Siikajoki from 7,4 mg/l to 12 mg/l. Table 5.1 shows the average raw water quality parameters of Haukipudas, Raahe and Siikajoki plants.

Table 5.1 Average raw water quality parameters in Haukipudas, Raahe and Siikajoki plants during the study.

Average raw water quality parameters	units	Name of the plant		
		Haukipudas	Raahe	Siikajoki
pH	-	6,90	6,60	6,70
temperature *	°C	4,50	5,50	3,50
dissolved oxygen	mg/l	0,00	4,40	0,00
ammonia	mg/l	0,23	0,02	0,16
hardness	mg/l	52	37	36
	as(CaCO ₃)			
manganese	mg/l	0,56	0,14	0,31
total iron	mg/l	17,80	1,00	10,40
carbon dioxide	mg/l	41,00	19,90	30,40
alkalinity	meg/l	1,07	0,51	0,61
redox potential	mVolt	141	250	182
ferrous iron	mg/l	17,8	0,52	10,30

* is affected by the seasonal temperature. During the study the seasons were autumn and winter.

5.5 Development and type of bacteria involved

Spots of reddish-brown sediments started to appear on the filter material after few weeks of plant operation. These spots were supposed to be the beginning of the development of iron bacteria responsible for differisation. In both pilot plants this stage was accompanied by start up in the reduction of iron concentration in the filtered water.

The appearance time of the spots and their development patterns in the two pilot plants have not followed the some sequence. In Haukipudas pilot plant the spots were observed on the third week after the plant was put into operation. The development of the spots was seen to start from the bottom of the filter. More over, the development was very slow and has

not completely covered the upper part of the filter till the end of the study period. In order to facilitate the development, supply of air has been provided from the bottom upwards through the filter. This was done because it was assumed that the slots on the pipe might not be enough to supply sufficient airflow. This assumption was made depending on the amount of dissolved oxygen available in filtered water. However, no change has been obtained after air ventilation was applied.

In Siikajoki pilot plant the reddish-brown spots started to appear on the second week after the filter was put into operation. On the third week almost the entire filter surface was covered by iron biomass. In this pilot plant the spots appeared at random all over the filter body both when the different sizes of filter material were put into operation. Figure 5.4 shows the formation of iron biomass on the dry filter in Siikajoki pilot plant.



Figure 5.4 Formation of iron biomass on the dry filter of Siikajoki pilot plant.

The plant in Raahе was in operation already when this study programme was started. Therefore the sequence of the development of the biomass was not followed from the beginning. However, at present the whole filter was covered by the iron biomass.

Samples of iron biomass were taken from all the plants to analyse and identify the presence and type of iron bacteria. The samples were collected by scraping off the biomass from the filter stones. Three samples were taken from each plant at three different places, i.e. from the top, middle and bottom parts of the filter unit.

5.6 Hydraulic load and contact time

As the raw water trickles through the filterbed a thin film of water is formed on the surface of the filter stone. The thickness of the thin film of the water covering each filter stone depends on the hydraulic load and hence the iron removal efficiency of the plant is affected.

The flow was altered from time to time depending on the conditions required both in Haukipudas and Siikajoki. The alteration of flow was usually done from higher to lower or visaversa by observing the results of iron removal obtained from the initial hydraulic loading.

The effect of washing down the iron deposits from the filter has been observed for a short time when the flow was changed from smaller to higher amount. This gives brownish colour to the effluent water, but stabilizes within very short time.

The amount of flow in Raahe was not changed except one time, because the plant was in full production and alteration was not possible.

Water meter was provided in Siikajoki pilot plant. The hydraulic load applied in Siikajoki was from 0,6-4,5 m/h and in Haukipudas from 0,8-5,7 m/h. The hydraulic load applied to the dry filter in Raahe was from 1,4-2,2 m/h.

Attempts have been made to measure the time taken by the water to pass through the filter. This was done by opening the inlet pipe valve and counting the time taken by the water from the time of opening the inlet valve until it reaches the collection basin at the bottom of the filter. The measurement of the contact time made in Siikajoki has shown that the water takes about 45 seconds to pass through the filter material of sizes from 40-50 mm with a depth of 2,0 meters. This was done only once at hydraulic load of 4,5 m/h (full capacity of the plant). Repeted measurement has to be made for different hydraulic loads and over a longer period of time in order to

obtain accurate results. This is because the contact time can depend on the hydraulic load and also the amount of iron deposits in the filter can affect the accuracy of the measurement. Table 5.2 shows the hydraulic load applied and other conditions of the test plants. Test 1 in table 5.2 is the test carried over the study period while tests 2 and 3 are tests carried every four hours of 3 days each using 40-50 mm and 10-20 mm filter gravel at Siikajoki pilot plant.

Table 5.2 Hydraulic load applied and other conditions of the test plants.

Name of the test plant	Filter depth (m)	Size of gravel (mm)	Surface area (m ²)	Hydraulic load (m/h)
Haukipudas	1,6-1,8	* 40-50	0,126	0,8 - 5,7
Raahe	1,8	40-50	7,5	1,4 - 2,2
Siikajoki				
test 1	1,8	40-50	3,5	0,6 - 4,5
test 2	2,0	40-50	3,5	0,4 - 3,7
test 3	2,0	10-20	3,5	0,5 - 3,8

* natural stone (not crushed as in the other plants).

6 RESULTS AND DISCUSSION

6.1 Iron removal efficiency and hydraulic load

The experimental results for iron removal and water quality improvement by dry filter were obtained from the study made for three months time in Haukipudas and Raahe and for four and half months time in Siikajoki plants. The effluent iron concentrations obtained from Haukipudas and Siikajoki pilot plants during the experiment were still higher than the maximum limits given by WHO (as 0,3 mg/l). The results of the effluent iron concentration obtained from Raahe was below the standard limit in most cases.

The iron removal efficiencies of each plant were different from one another and this could be due to the difference in iron concentration and characteristics of the raw water and some other factors in the operation conditions. Equations for fit in curves for iron removal and efficiency were developed using small statistical computer for the results from Haukipudas and Siikajoki.

Haukipudas

The results of the analysis of iron as obtained during the study of the pilot plant is presented in appendix 1 and plotted in figure 6.1.

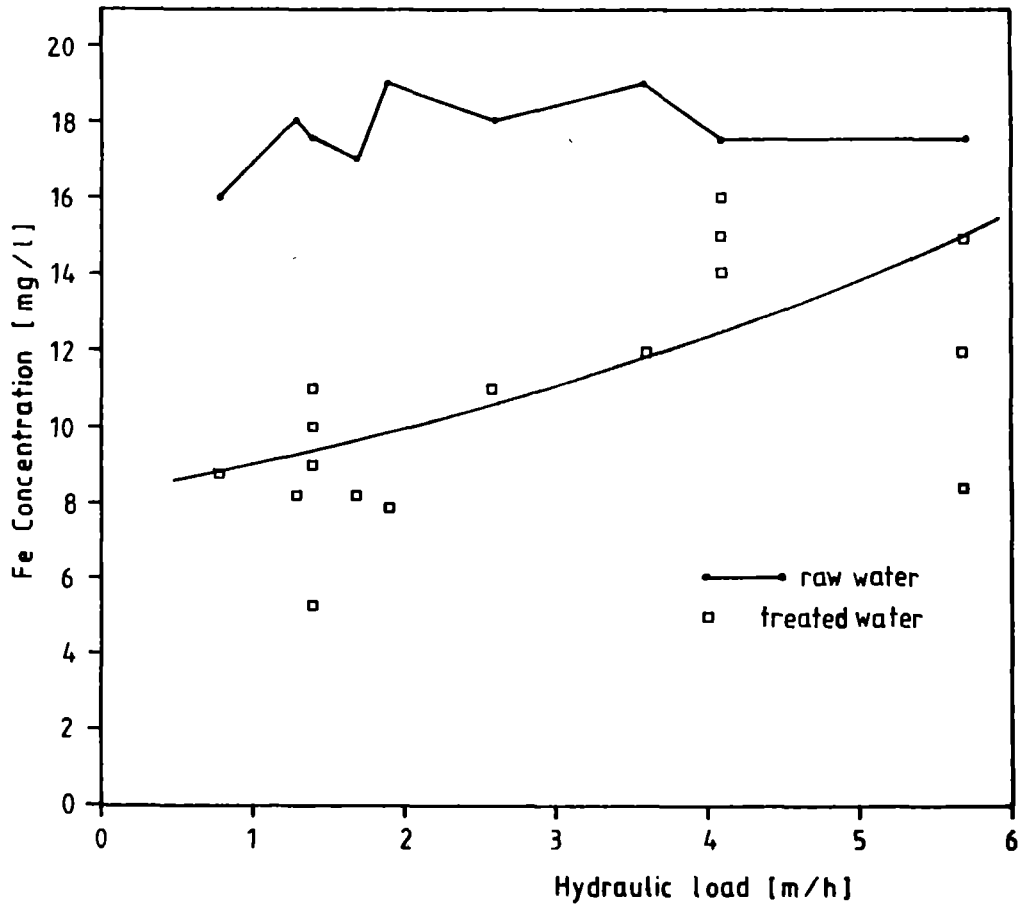


Figure 6.1 Iron concentration in raw and treated water as a function of hydraulic load from Haukipudas pilot plant.

The iron concentration in treated water after the dry filter shows decreasing trend as the hydraulic load decreases. Fit in curve for the results of the iron concentration in treated water was made. The curve exhibits exponential function characterized by the equation:

$$Fe = 8,16e^{0,11q} \quad (6.1)$$

where Fe = effluent iron concentration in mg/l
 q = hydraulic load

Correlation coefficient for equation (6.1) is 0,6.

Equation (6.1) gives an idea how the change in hydraulic load affects the iron concentration in the effluent. The relationship between effluent iron concentration and hydraulic load might remain the same, but the position of the curve changes under different conditions e.g. design and construction of the filter, longer time functioning after starting the operation etc. Theoretically equation (6.1) exhibits the minimum effluent iron concentration of 8,16 mg/l, however, smaller concentration up to 5,3 mg/l under hydraulic load of 1,4 m/h was obtained. The solid iron escaping through the filter was measured after filtering the effluent water through a 0,45 μ m filter paper and solid iron concentration was found to vary from 1,0-5,0 mg/l.

Iron removal efficiency of dry filter also depends on the hydraulic load. Removal efficiency of the filter was increased with lower hydraulic loading as shown in figure 6.2.

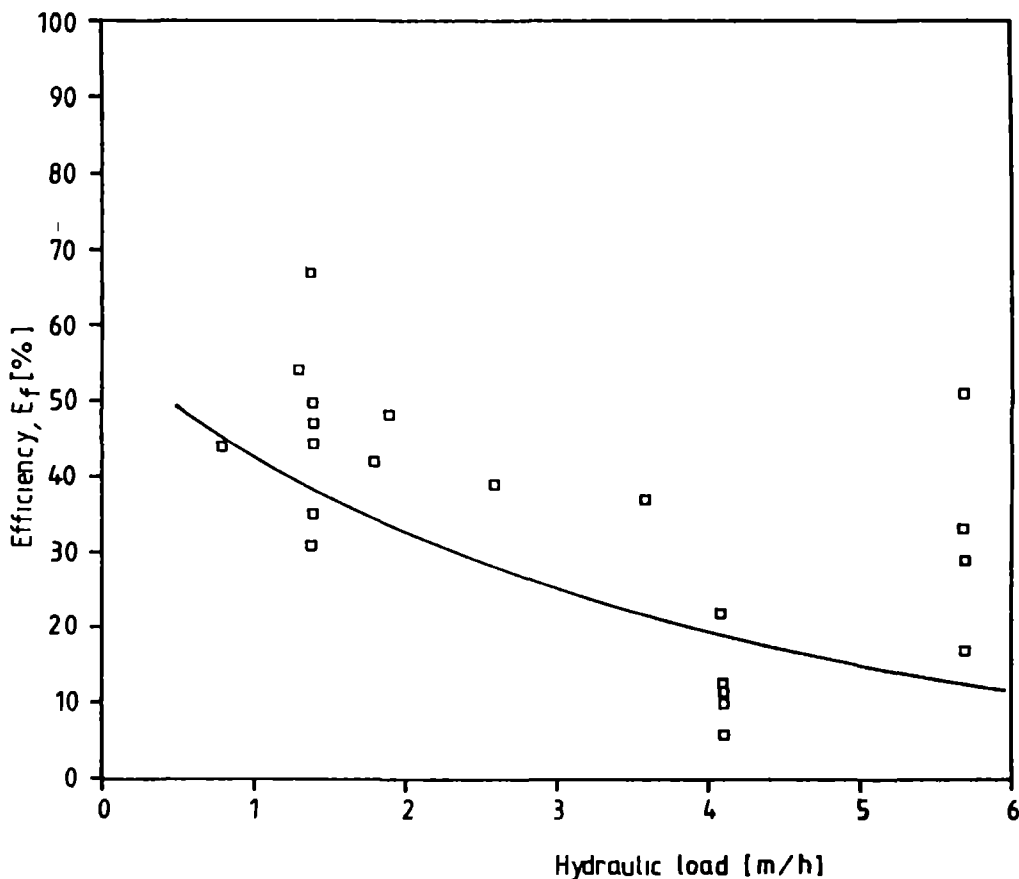


Figure 6.2 Iron removal efficiency of the dry filter as a function of hydraulic load from Haukipudas pilot plant

From figure 6.2 iron removal efficiency varies even under the same hydraulic load. However, fit in curve with correlation coefficient of 0,53 was developed with the equation

$$E_f = 56e^{-0,26q} \quad (6.2)$$

where E_f = iron removal efficiency (%)
 q = hydraulic load (m/h)

From equation (6.2) the theoretical maximum efficiency that can be reached is 50 %, however, efficiencies up to 67 % were reached in certain particular cases. The main reasons for low efficiency of this plant could be

- the pilot plant was too small
- the slots on the PVC pipe column were very small and sufficient air could not enter into the filter
- the raw water pumps were automatically controlled and hence continuous supply of raw water to the pilot plant was not possible
- the water sprayed by the shower head was not distributed equally over the top of the filter surface. In other words the effective surface area was smaller than actual surface area of the filter

The reasons above were more of operational problems. But it could also be possible that the characteristics of the raw water might affect the iron removal efficiency of the pilot plant. This has to be investigated in relation to the development of the micro-organisms which are responsible for iron removal by dry filter.

Raahe

The dry filter plant in Raahe was a production plant serving as a pretreatment unit for iron removal before the slow sand filter. Since this plant has been in operation for more than one year, it was already in a matured stage during the investigation period. Analysis of iron concentration in the effluent water both after dry and slow sand filters were made. The results are presented in appendix 2 and average values are shown in figure 6.3.

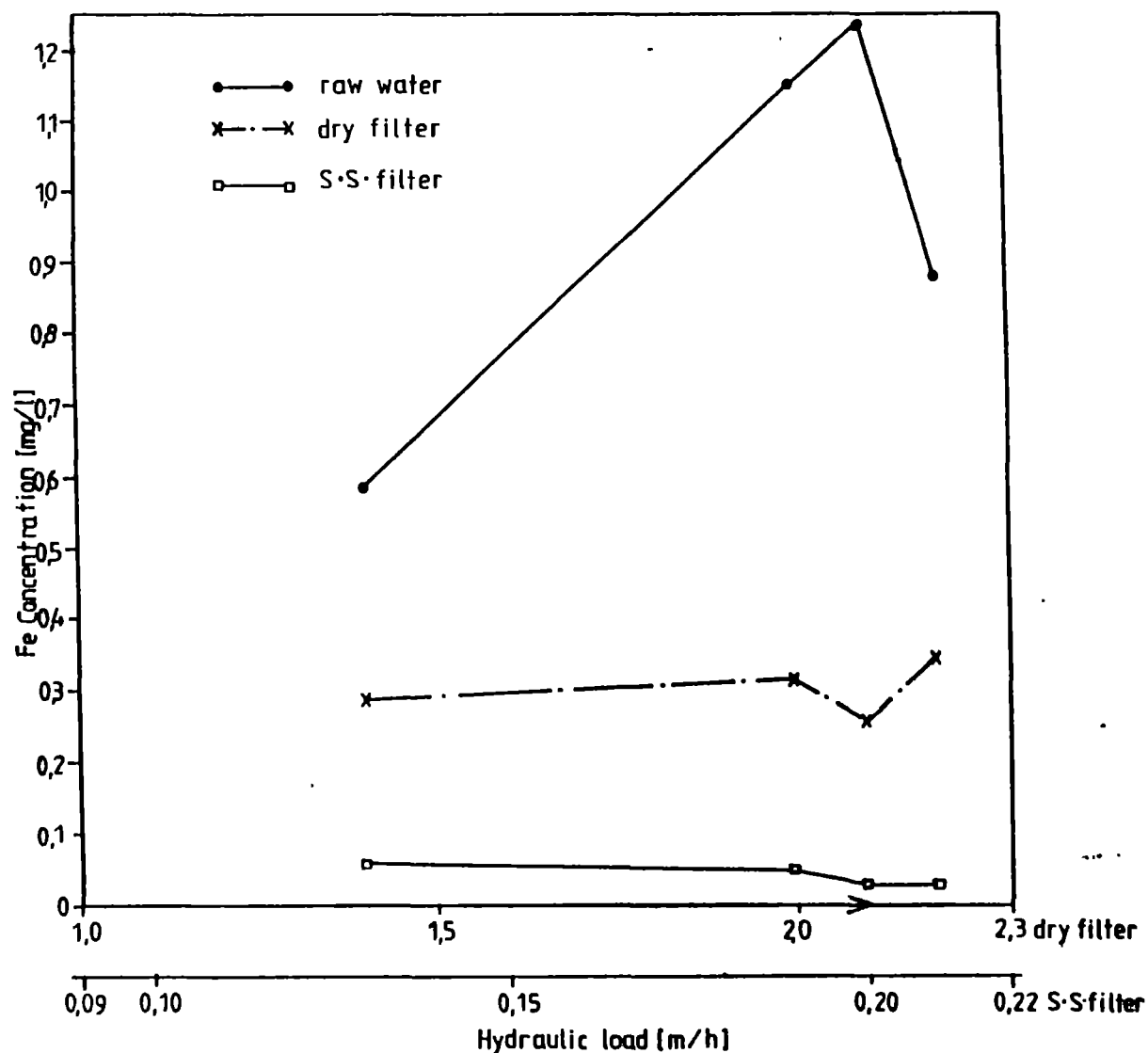


Figure 6.3 Average iron concentration in raw and treated water after dry and slow sand filter of Raahe plant as a function of hydraulic load

Figure 6.3 shows that the average iron removal by the dry filter seems constant over the range of 1,4 m/h to 2,0 m/h hydraulic loading. From the same figure, iron removal by the dry filter is hardly affected even if iron concentration in the raw water increases. This means that the filter has extra capacity for certain additional iron loading. The maximum iron load applied per volume of the filter per day was 34 gram Fe/m³ day with the total raw water flow of 400 m³/day. The effluent iron concentration was 0,26 mg/l with hydraulic load of 2,1 m/h. Iron concentration of less than 0,3 mg/l in the effluent was reached with the hydraulic loads of less than 1,7 m/h and iron load of less than 27 gram Fe/m³ day.

Iron removal efficiency of this plant tends to decrease with the decrease in hydraulic load which is not true. This is because of the few points available to draw the figures. However, the removal efficiency was still bigger at higher iron loading as well as hydraulic loading. This also indicates that the filter has extra capacity for additional iron loading. Figure 6.4 shows the iron removal efficiency of the dry and the slow sand filter at different hydraulic load.

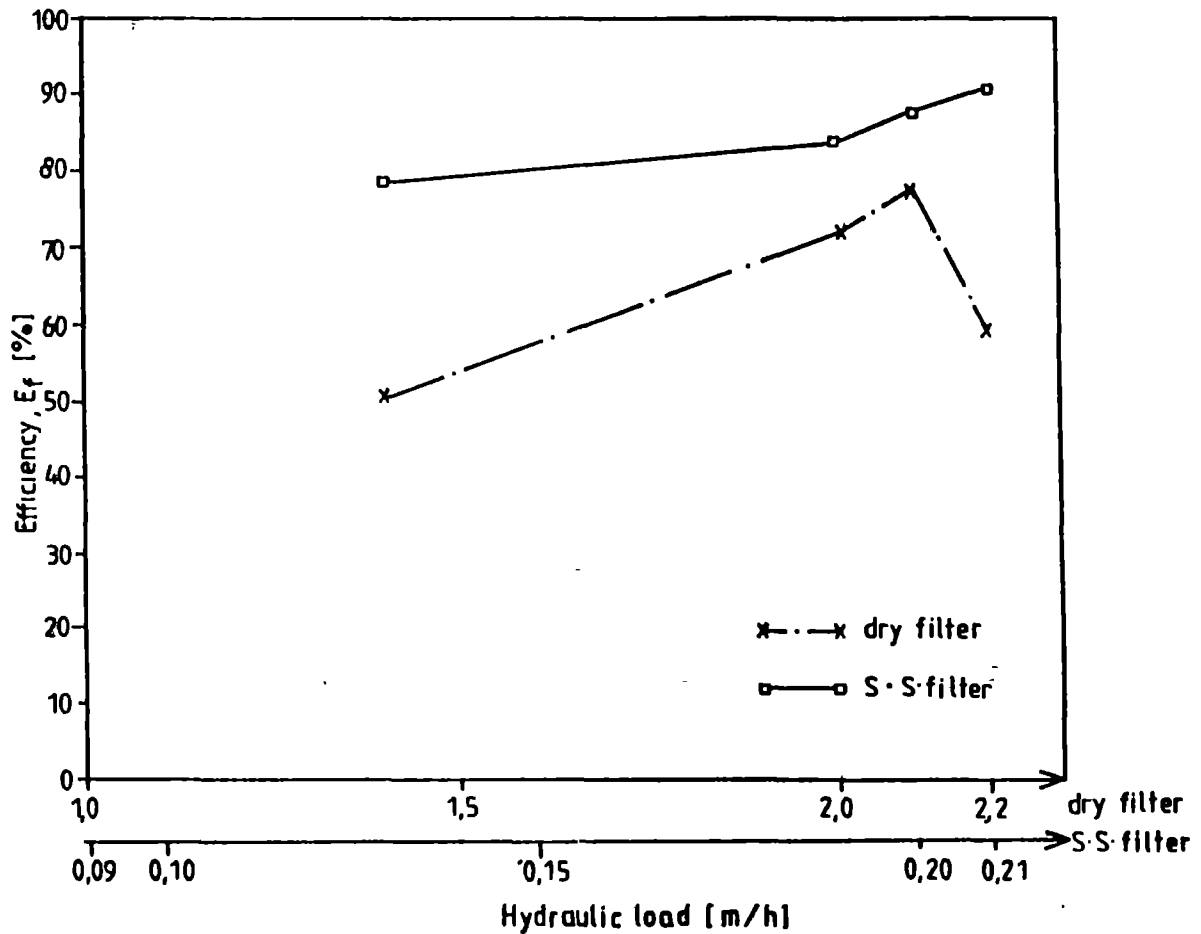


Figure 6.4 Iron removal efficiency of the dry filter and the slow sand filter of Raahé plant at different hydraulic load.

Analysis of solid iron escaping from the dry filter was also analysed and solid iron concentration was found to vary from 0,1 - 0,2 mg/l. This will affect the removal efficiency of the slow sand filter, although it was more than 80 % and iron concentration in the effluent was very much below 0,3 mg/l.

The lowest total iron concentration result obtained in the effluent water after the dry filter was 0,11 mg/l at hydraulic load of 1,4 m/h.

Siikajoki

The results of the analysis of iron in raw and treated water obtained during the study of the pilot plant is presented in appendix 3 and plotted in figure 6.5.

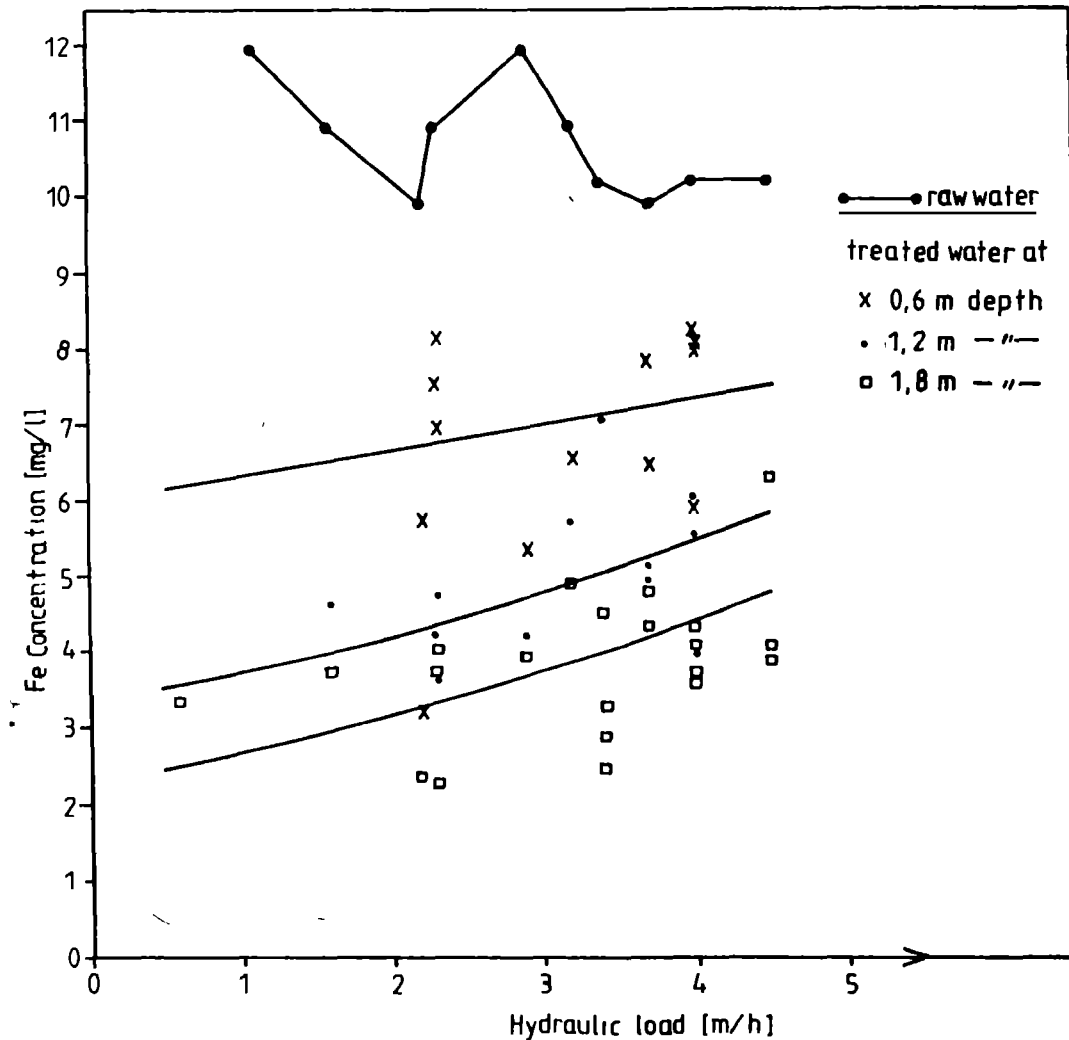


Figure 6.5 Iron concentration in raw and treated water at different depths of Siikajoki pilot plant as a function of hydraulic load.

Figure 6.5 shows that the iron concentration in the effluent decreases as the hydraulic load decreases. This trend can be clearly seen at different depths in the filter. The equations developed at depths of 0,60 m, 1,20 m and 1,80 m are

$$Fe = 6,01 + 0,34q \quad \text{at } 0,60 \text{ m} \quad (6.3)$$

$$Fe = 3,38e^{0,12q} \quad \text{at } 1,20 \text{ m} \quad (6.4)$$

$$Fe = 2,33e^{0,16q} \quad \text{at } 1,80 \text{ m} \quad (6.5)$$

where Fe = effluent iron concentration (mg/l)
 q = hydraulic load (mg/l)

The correlation coefficients are 0,29, 0,52 and 0,53 in sequences of the equations.

Iron removal at the depth of 0,60 m exhibited linear relationship to the hydraulic load while it was exponential at the other depths. The linear relationship may not be true as shown in equation (6.3). Because the relationship is exponential as in equations (6.4) and (6.5) and also can be seen in other equations later on.

The iron concentration in the effluent was more than the standard limit of 0,3 mg/l. Although the standard limit has not been reached iron concentration of 2,5 mg/l in the effluent was reached at hydraulic load of 3,4 m/h. The iron load and the hydraulic load affect the efficiency of the plant. Figure 6.6 shows iron removal efficiency of the plant.

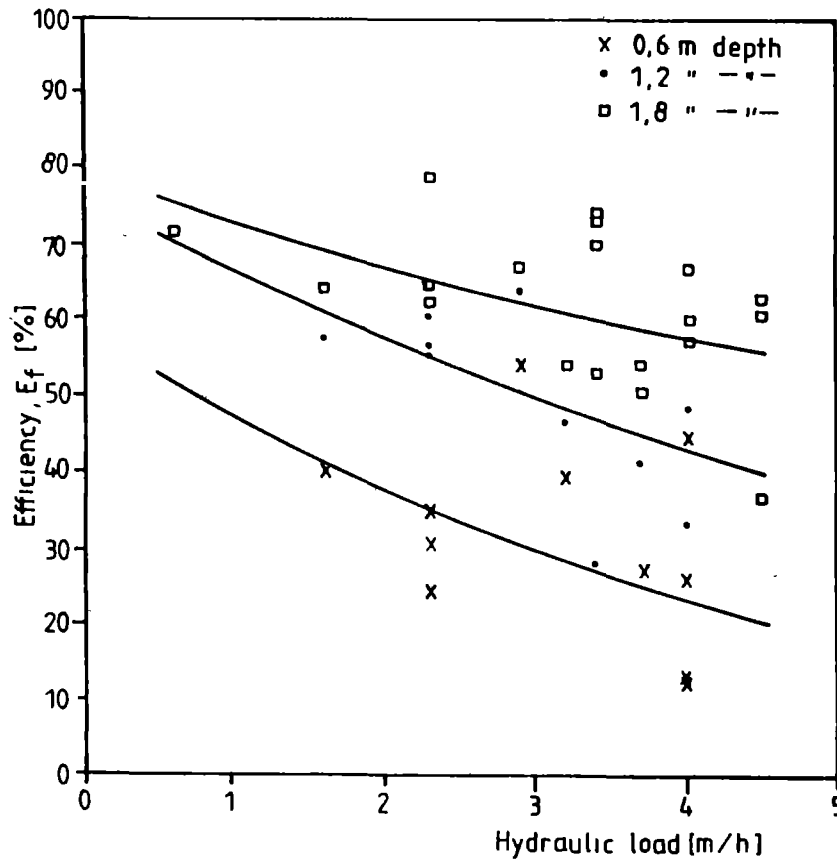


Figure 6.6 Iron removal efficiency of Siikajoki pilot plant as a function of hydraulic load.

Figure 6.6 shows that the iron removal efficiency increases as the hydraulic load decreases. The relationship between the efficiency of the plant at different depths and hydraulic load was found to be exponential. The equations of the fit in curves at different depths were

$$E_f = 60e^{-0,22q} \quad \text{at } 0,60 \text{ m} \quad (6.6)$$

$$E_f = 76e^{-0,14q} \quad \text{at } 1,20 \text{ m} \quad (6.7)$$

$$E_f = 78e^{-0,08q} \quad \text{at } 1,80 \text{ m} \quad (6.8)$$

where E_f = removal efficiency in percent
 q = hydraulic load in m/h

The theoretical maximum efficiency of this plant that could be obtained from equation (6.8) was less than 78 %. However, removal efficiency of 79 % at the hydraulic load of 2,3 m/h was obtained during the study. Removal efficiency of 74 % was also reached at the hydraulic load of 3,4 m/h. These results indicate that the removal efficiency of the dry filter does not only depend on the hydraulic load. It is also dependent on the iron concentration the raw water which was higher during test at the hydraulic loads of below 3 m/h.

The iron removal takes place all along the filter depth as the water trickles through the dry filter.

Figure 6.7 shows the effect of filter depth on iron removal under different hydraulic loading.

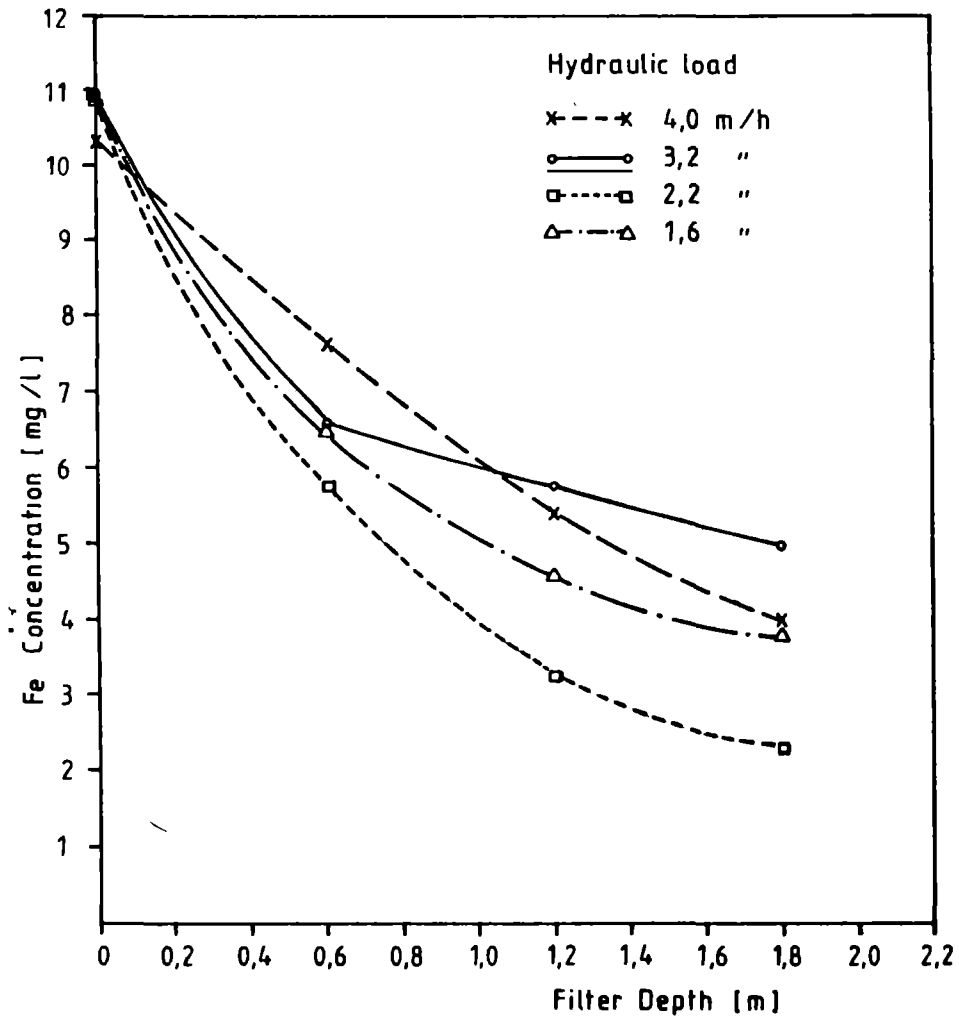


Figure 6.7 Influence of filter depth and hydraulic loading intensity on iron removal of Siikajoki pilot plant.

From figure 6.7 it can be seen that at shallow depths iron removal per unit depth was higher than it was at greater depths. This exhibits similar trends as substrate removal by trickling filter for waste water treatment (Rich 1980).

The results presented in figures 6.5 to 6.7 were obtained from the analysis made over three months time. Analysis of iron removal by the same dry filter with the same sizes of filter material was made every four hours over three days time by varying the hydraulic load. This was done in order to know the immediate respond of the dry filter to the change in hydraulic load. This enables to know how quickly the filter stabilizes

and reaches its maximum efficiency. Figure 6.8 shows the iron concentration in raw and treated water under different hydraulic loads as analysed every four hours. The results of the analysis are presented in appendix 4.

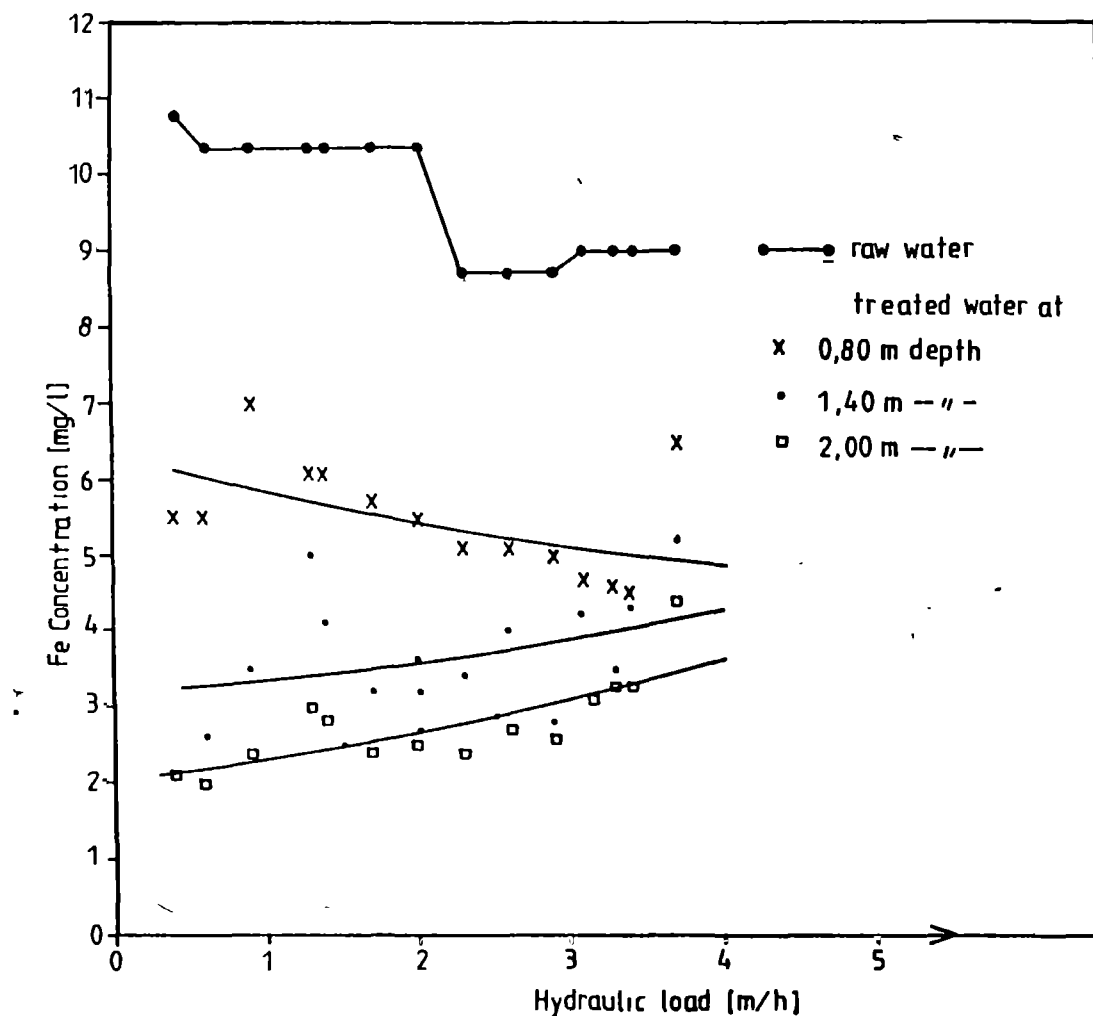


Figure 6.8 Iron concentration in raw and treated water as a function of hydraulic load as analysed every 4 hours from Siikajoki pilot plant.

Iron concentration in the treated water decreases as the hydraulic load decreases. The relationship between iron concentration in the effluent and hydraulic load at different depths was explained by the equations:

$$\text{Fe} = 6,2e^{-0,06q} \quad \text{at } 0,80 \text{ m} \quad (6.9)$$

$$\text{Fe} = 3.1e^{0,08q} \quad \text{at } 1,40 \text{ m} \quad (6.10)$$

$$Fe = 1,99e^{0,15q} \quad \text{at } 2,00 \text{ m} \quad (6.11)$$

where Fe and q are the same as in equation (6.3) to (6.5)

Correlation coefficients are 0,51, 0,62 and 0,80 in sequence of the equations

Figure 6.8 also shows that the iron concentration in raw water was higher during the smaller hydraulic loading at which better iron removal was obtained. The lowest iron concentration reached during the analysis was 2,0 mg/l at the hydraulic load of 0,6 m/h. Also iron concentration of 2,6 mg/l in treated water was obtained at 2,9 m/h hydraulic loading.

The iron removal efficiency of the plant decreases as the hydraulic load increases. Figure 6.9 shows the removal efficiency of the Siikajoki pilot plant under different hydraulic loads as analysed every four hours.

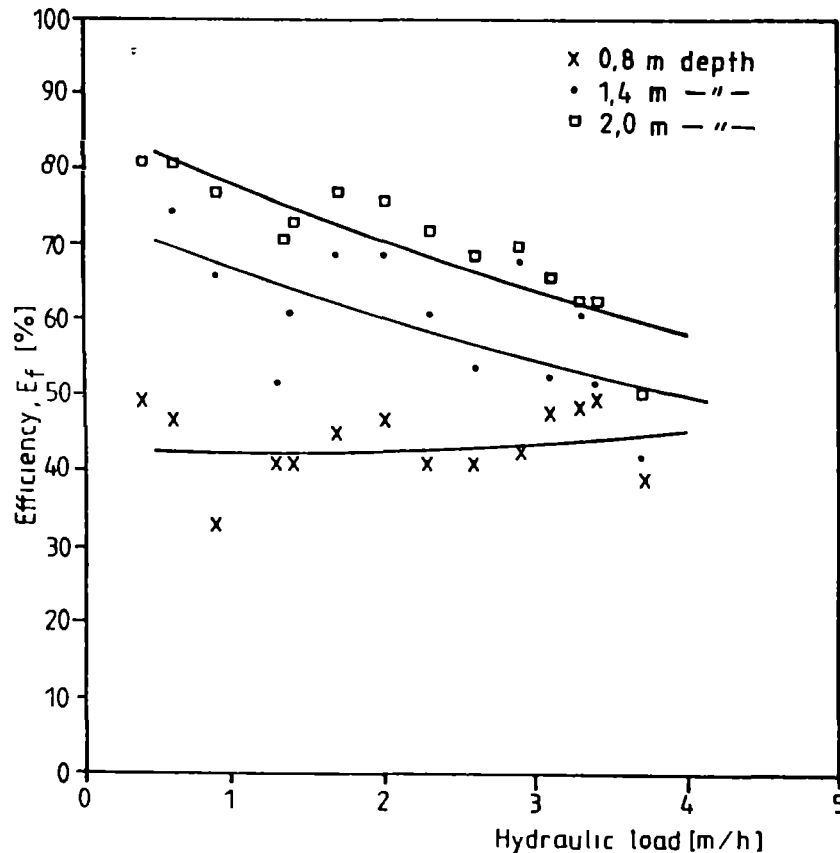


Figure 6.9 Iron removal efficiency of the Siikajoki dry filter plant under different hydraulic loads

The relationship between the removal efficiency and hydraulic load as developed from the fit in curves at different depths of the filter are

$$E_f = 0,42e^{0,02q} \quad \text{at } 0,80 \text{ m} \quad (6.12)$$

$$E_f = 74e^{-0,10q} \quad \text{at } 1,40 \text{ m} \quad (6.13)$$

$$E_f = 86e^{-0,10q} \quad \text{at } 2,00 \text{ m} \quad (6.14)$$

where E_f and q are the same as in equations (6.6) to (6.8).

The correlation coefficients are 0,17, 0,62 and 0,86 in sequence of the equations.

The influence of filter depth on the iron removal was also analysed from the results of the study made every four hours over three days. During this analysis the depth of the dry filter of Siikajoki pilot plant was increased by 20 cm. This was done by filling more crushed stones of the same sizes on the upper part of the filter. Figures 6.10 and 6.11 show the influence of filter depth on iron removal and efficiency under different hydraulic loadings.

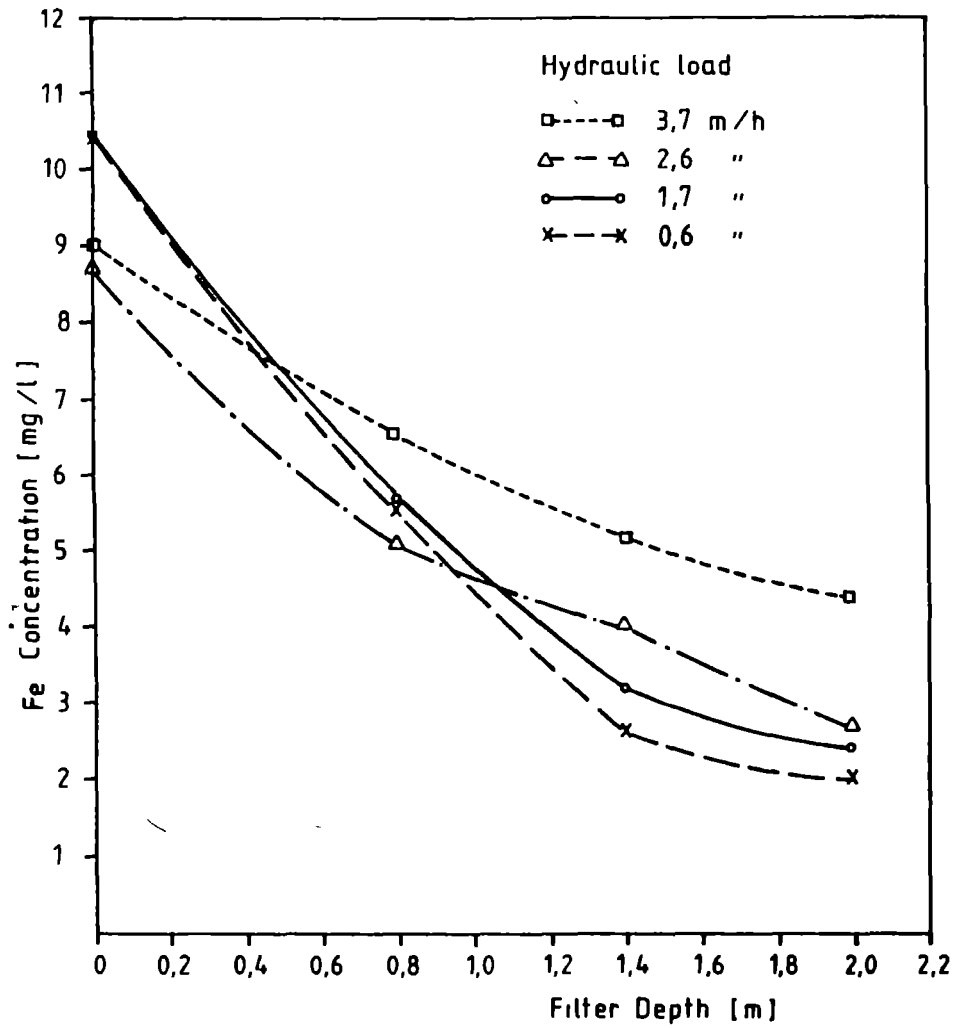


Figure 6.10 Influence of filter depth on iron removal under different hydraulic loading as analysed every 4 hours.

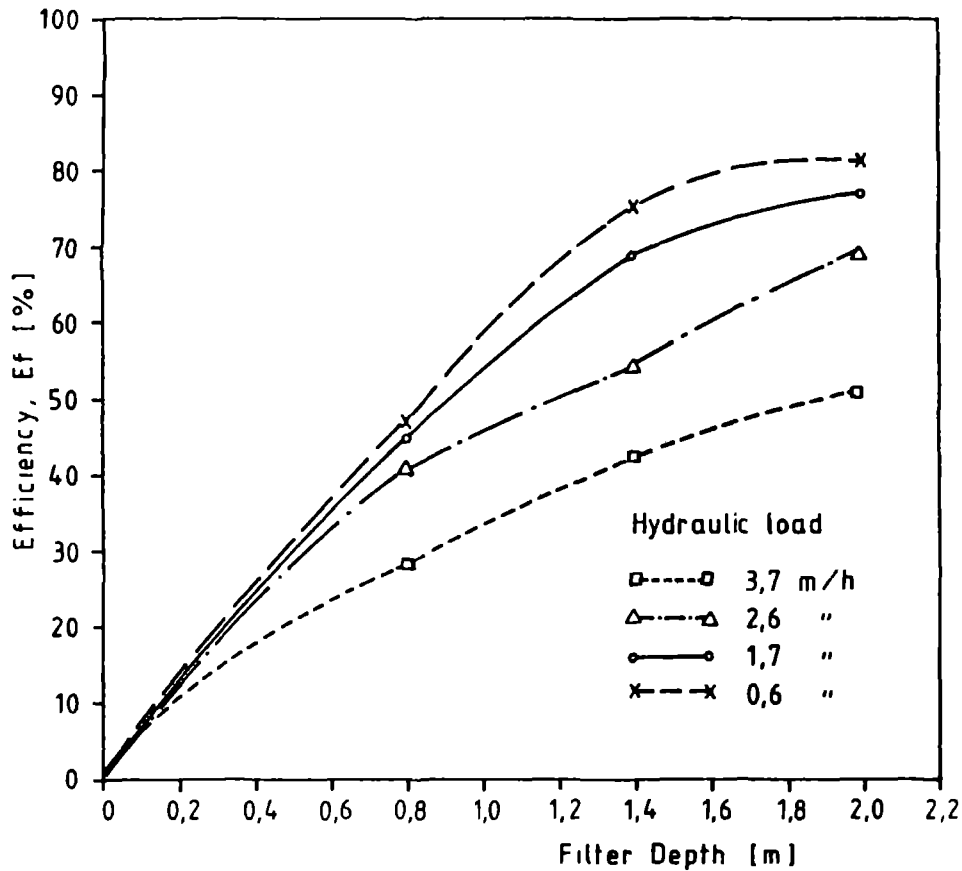


Figure 6.11 Iron removal efficiency at different depth of the filter under different hydraulic loading.

The maximum iron removal efficiency obtained from this analysis was 81 % at the hydraulic loads of below 0,6 m/h. No significant effect was found due to the increment of the depth of the filter by 20 cm. However, iron removal efficiency was increased from 76 % at the hydraulic load of 2,2 m/h to more than 76 % at the hydraulic loads below 2,0 m/h. For instance removal efficiency of 72 % was increased to 81 % under hydraulic load of 0,6 m/h after increasing the filter depth.

Analysis of solid iron escaping through the filter was made both before and after the increment of the filter depth. From this analysis 0,6 mg/l to 1,4 mf/l of iron precipitates were passing through the filter as solid form. Some of these iron solids could be retained if separate settling basin was provided.

6.2 Development and efficiency of iron bacteria

The development of iron bacteria started to be effective after two weeks of operation in Siikajoki and after three weeks of operation in Haukipudas pilot plants. The development of these bacteria was accompanied by the reduction in iron concentration of the effluent water after the dry filter. From iron sludge samples taken for the analysis it was found that the percentage of organic material presented in a unit weight of sample were 31 % and 22 % from Raahe dry filter and dry filter of Siikajoki pilot plant respectively. The difference in the result could probably mean that the number of the organisms will increase after long time operation of the dry filter which was the case in Raahe.

Samples of iron sludge were taken at different depths of the dry filters from Haukipudas, Raahe and Siikajoki plants for the analysis of iron bacteria. The results from the analysis of these samples proved the existence of different iron bacteria all over the filter depth.

Haukipudas

Samples taken from the top surface of the filter indicated the presence of Ochrobium type of dominating species. At this part any filamentous iron bacteria were not present. Metallogenium cell groups covered with a thick iron and manganese precipitate were also present. From samples taken at the middle depth of the filter dominating species of Ochrobium type of bacteria were found. Indication of Arthrobacter types of bacteria with yellowish iron precipitation and few Gallionella filaments were existing. No indication of about manganese precipitation was identified. In the samples taken from the bottom of the filter Ochrobium and Arthrobacter types of species were dominating. No filamentous iron bacteria were present and there was no indication about manganese precipitation.

These bacteria were developed in the raw water characteristics of pH varying from 6,4 - 7,3, redox potential of 110-230 mV and oxygen concentration of less than 0,4 mg/l. Unfortunately, the raw water was not analysed for organic matter, however, the presence of yellowish iron precipitation indicates the existence of organic matter.

Raahe

This plant was in operation more than one year and the sample taken from the top of this filter contained Ochrobium and Arthrobacter types of bacteria as the dominating species. Few Gallionella filaments and manganese precipitates were also present. Samples from the middle depth of the filter contained dominating species of Gallionella. About half of the Gallionella filaments were covered with a thick iron precipitate. Metallogenium cell groups covered with a thick iron and manganese precipitate were found. Some Ochrobium and Siderocapsa cells were also present. This sample indicates an active iron and manganese precipitation. In the samples taken from the bottom of the filter Gallionella was the dominating species. Other bacteria present were Metallogenium, Siderocapsa and Ochrobium. A part of Gallionella without iron precipitate and others covered with a thick iron precipitate existed. Few filaments of Leptothrix trichogenes bacteria were observed.

These bacteria developed in the filter treating raw water of pH 6,1 - 7,3, redox potential of 200-300 mV and oxygen concentration of 2-6 mg/l.

Siikajoki

From the pilot plant of Siikajoki iron sludge samples were taken both when sizes of 40-50 mm and 10-20 mm filter gravel were used.

Samples taken from the top of the filter, during the use of 40-50 mm gravel sizes, contained dominating species of filamentous Gallionella bacteria covered with a thick iron precipitate. Metallogenium cell groups covered with thick iron and manganese precipitation were also observed. This sample indicates an active iron and manganese precipitation. Samples taken at the middle depth of the filter showed Metallogenium and Arthrobacter types as the dominating species. Few Gallionella filaments covered with a thick iron precipitate were present. This sample indicates an active iron precipitation but no manganese activity. Similarly iron sludge sample taken from the bottom of the filter contained dominating species of Ochrobium, Metallogenium and Arthrobacter types. Few Gallionella filaments covered with a thick iron precipitate were present. This sample also indicates active iron precipitation but there is no indication about manganese activity. These samples were taken after three months of operation and the development of some species of iron and manganese bacteria was not complete.

From samples taken after the sizes of the filter media was changed to 10-20 mm the presence of certain iron bacteria was indicated although their type as well as concentration was smaller. Samples taken from the top and middle depth of the filter contained dominating species of Ochrobium and few filaments of Gallionella. Sample from the bottom of the filter showed no other types of iron bacteria except the dominating species of Ochrobium. These samples were taken after 2 weeks of operation. From these results it could be clearly understood that the filter was not yet matured.

In both cases these bacteria were developed in the raw water of pH 6,3 - 7,4, redox potential of 140-250 mV and oxygen concentration of less than 2,7 mg/l.

The iron removal efficiency of the dry filters is more or less the efficiency of the iron oxidizing bacteria. It also depends on the amount of iron bacteria present in the filter and the size of filter material used on which these bacteria develop.

This study was conducted during the autumn and beginning of the winter seasons. Because of the cold weather of these seasons, it was supposed that the efficiency of the plants might be affected. However, there was no effect on the iron removal efficiency as indicated by the results obtained from the summer time analysis from Raahe dry filter. The results are presented in appendix 5.

6.3 Raw and treated water quality analysis

During the study period 24 samples from Haukipudas, 27 samples from Raahe and 23 samples from Siikajoki plants were taken for analysis. In addition to iron, other water quality parameters were also analysed and the average results are shown in table 6.1.

Table 6.1 Average results of the analysis of raw and treated water quality parameters.

No	measured parameters	unit	Haukipudas		Raahe			Siikajoki	
			raw water	treated water dry filter	raw water	treated water dry filter	slow sand filter	raw water	treated water dry filter
1	pH	-	6,9	7,0	6,6	7,1	7,2	6,7	6,9
2	Tem	°C	4,5	5,5	5,5	4,5	4,5	3,5	4,5
3	O ₂	mg/l	0,0	8,0	4,4	9,6	9,3	0,0	9,2
4	NH ₃	mg/l	0,23	0,23	0,02	0,01	0,01	0,16	0,19
5	Hardness	mg/l	52	55	37	36	35	36	34
6	Mn	mg/l	0,56	0,56	0,14	0,07	0,02	0,31	0,30
7	Fe(II)	mg/l	17,8	10,0	0,52	0,14	0,03	10,3	3,0
8	Fe total	mg/l	17,8	12,0	1,0	0,31	0,05	10,4	4,3
9	CO ₂	mg/l	39,2	18,9	19,9	3,1	2,7	30,4	6,5
10	Alkalin.	meq/l	1,07	0,77	0,51	0,50	0,49	0,61	0,37
11	E _h	mVolt	+142	+163	+250	+253	+277	+174	+224
12	Conduct.	ms/m	20,3	18,1	8,0	8,7	8,7	14,6	13,2
13	Turbidity		0,19	6,5	1,5	0,45	0,36	1,5	10,4
14	Colour	pt/l	0,08	0,26	0,02	0,02	0,02	0,05	0,12
15	Ca	mg/l	12,0	12,0	6,0	5,9	6,1	7,5	7,9
16	Zn	mg/l	0,05	0,05	0,05	0,05	0,05	0,05	0,05

Quality parameters from 1-11 were average values analysed regularly during the study period and parameters from 12-16 were analysed once. Also bacteriological quality was analysed but there was no indication of coliforms or other hygiene indicators.

Table 6.1 also shows that the turbidity and colour of the treated water after the dry filter increased in the case of Haukipudas and Siikajoki plants. This was due to the presence of iron precipitates and the washed-down of iron sludge from the filter. After the water passed through the filter and reached the bottom of the filter it was collected in a basin which was also supposed to serve as a sedimentation basin.

However, the water passing through the filter drops into the basin from a height disturbing and even probably breaking the iron precipitates present in the collection basin. This leads to the requirement of separate sedimentation basin where the iron precipitates present in the filtered water and the washed-down iron sludge can settle without any disturbance. This condition can be seen from the result of turbidity and colour removal by Raahé dry filter where the filtered water is collected and lead by a conduit to a separate sedimentation basin.

It is also shown in table 6.1 that some of the iron was still in ferrous form although there was sufficient oxygen supply in treated water. It is possible that the ferro iron was present in combination with complex organic compounds which the micro-organisms might not be able to oxidize. Analysis of raw water from Siikajoki showed 17 mg/l and 4 mg/l of CODCr and TOC respectively. At Haukipudas and Siikajoki Fe(II) is high if compared to total iron. The rise in pH and E_h after the filtration process confirms to the conditions of most iron removal processes, e.g. the Vyredox method. The pH was raised during the process of filtration and therefore addition of chemical for pH adjustment may not be required.

6.4 Analysis of selected water quality parameters

6.4.1 Oxygenation and decarbonation

The raw waters of Haukipudas and Siikajoki are anoxic and contained high carbondioxide. Oxygen starts to enter the water and carbondioxide escapes as the water trickles through the filter during filtration. Average results are shown in figure 6.12.

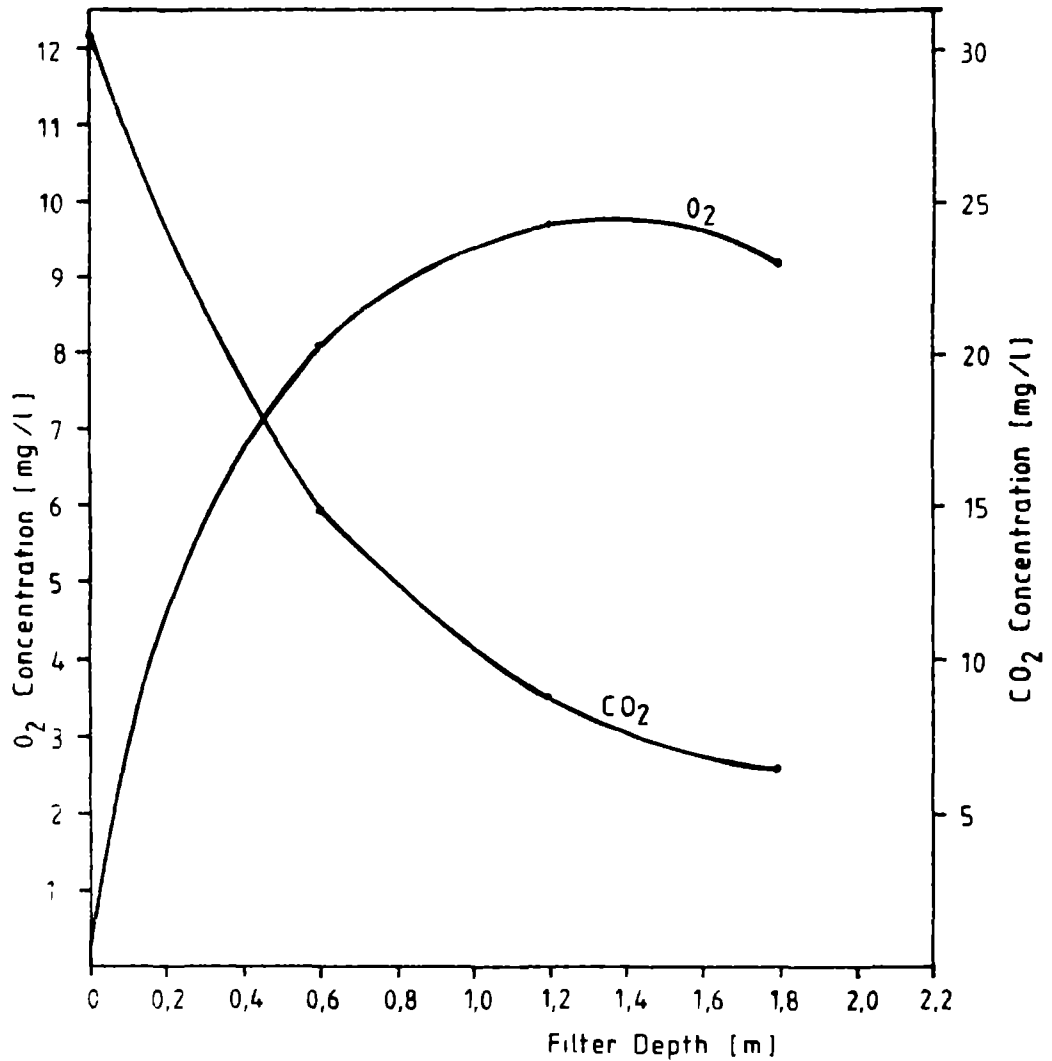


Figure 6.12 Influence of dry filter depth on oxygenation of the water and carbondioxide removal from Siikajoki pilot plant (gravel sizes of 40-50 mm).

Figure 6.12 shows that oxygen supply and carbondioxide removal was mostly completed on the upper portion of the dry filter. The oxygenation process was dependent on the depth of the filter, available slots or holes on the filter body and the water distribution methods on the surface of the filter. From figure 6.12 it can be seen that dissolved oxygen seemed to decrease beyond 1,20 m of filter depth.

This was perhaps because the oxygen was consumed by certain biological process. This process could be the process by which some of the sloughed off bacteria are washed down through the filter and might be degraded together with some other organic matters in the lower part of the dry filter.

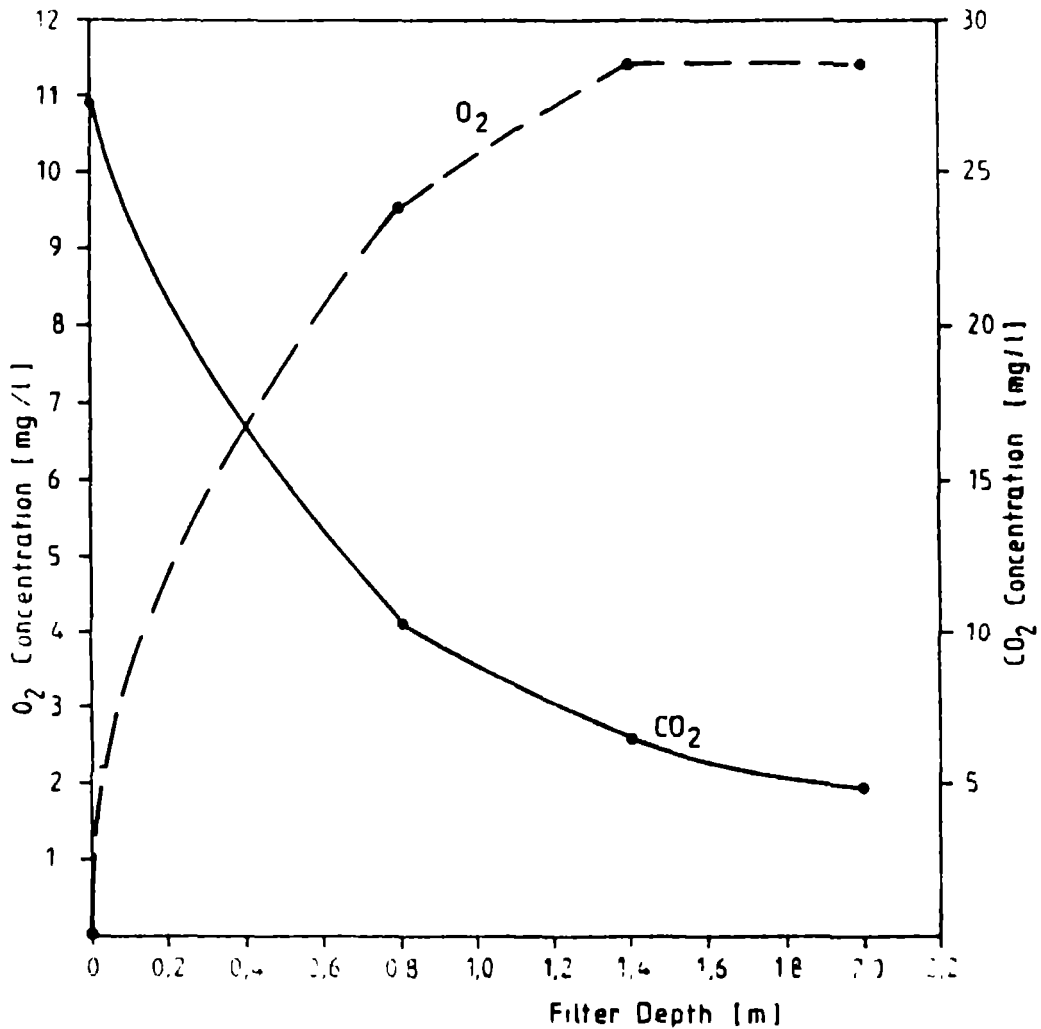


Figure 6.13 Influence of filter depth on oxygenation and carbondioxide removal as analysed every 4 hours from Siikajoki pilot plant (gravel sizes of 40-50 mm).

Oxygen concentration at different depth of the filter especially at the lower depths showed the tendency of being consumed both in figure 6.12 and 6.13. These results were obtained after fairly long time operation of the filter. However, results obtained from the analysis of every 4 hours over three days, with the dry filter filled by 10-20 mm filter material and functioning for three weeks, showed different phenomenon as presented in figure 6.14.

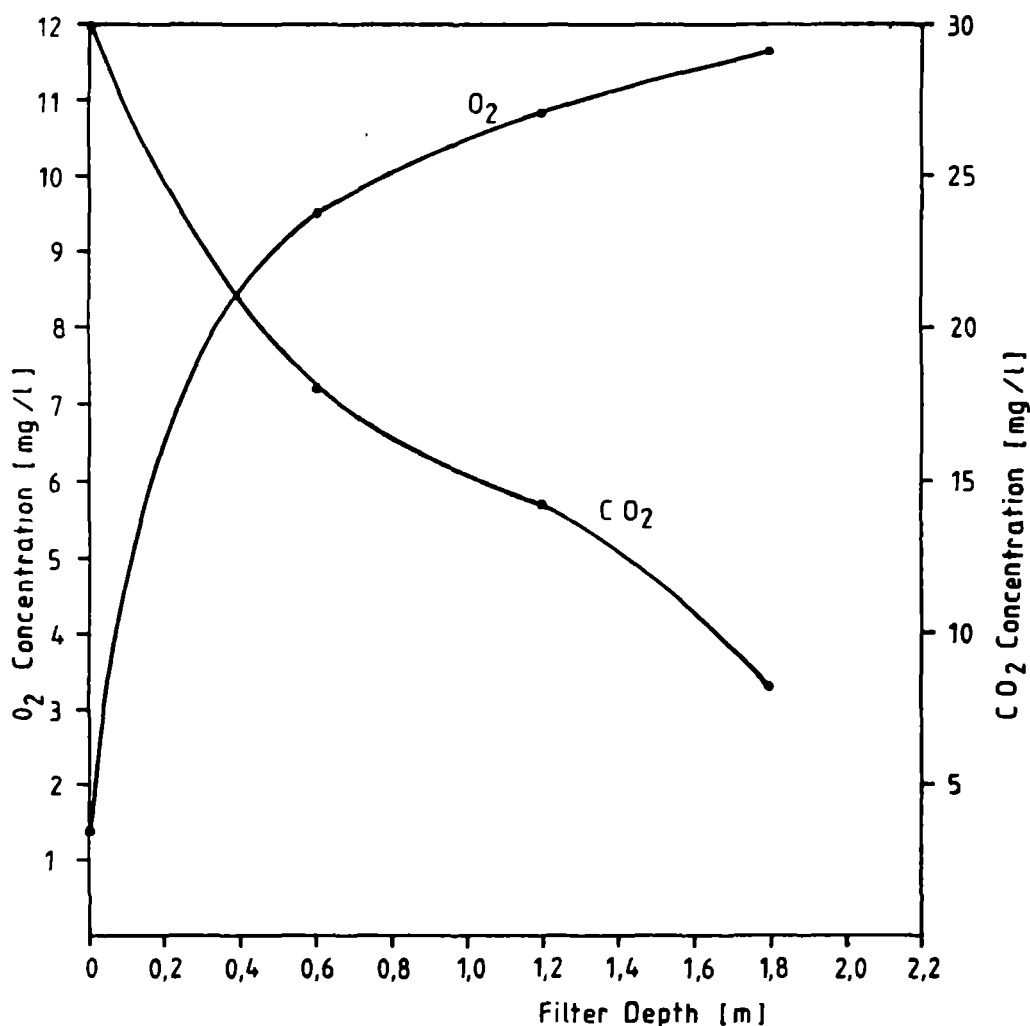


Figure 6.14 Effect of filter depth on oxygenation and carbondioxide removal as analysed every 4 hours from Siikajoki pilot plant (gravel sizes of 10-20 mm).

From figure 6.14 it can be seen that oxygen concentration was increasing with respect to the filter depth. Since, the plant was in operation only for three weeks time when these results were taken, it could be possible that sloughing off of aged or

dead micro-organisms and biological degradation were not yet started, which might be the reason for oxygen reduction at lower depth of the seasoned filter.

The supply of oxygen and removal of carbondioxide with respect to filter depth was not investigated in Haukipudas and Raahe dry filters as it was done in Siikajoki. Nevertheless, the oxygen and carbondioxide concentration in the raw and treated water were analysed during the study period. The results can be seen from table 6.1. From the results obtained the carbondioxide removal in Haukipudas dry filter was not satisfactory. The reason could be due to unefficient water distribution by the spary nozzle and insufficient provision of ventilation slots on the filter body of the PVC column pipe.

In the three dry filters investigated, the carbondioxide concentration in raw water was different and the removal efficiencies of the three plants were also different from each other. The average removal efficiency in Haukipudas was 50 % while it was 84 % in Raahe. The carbondioxide removal efficiency of the dry filter in Siikajoki is shown in figure 6.15.

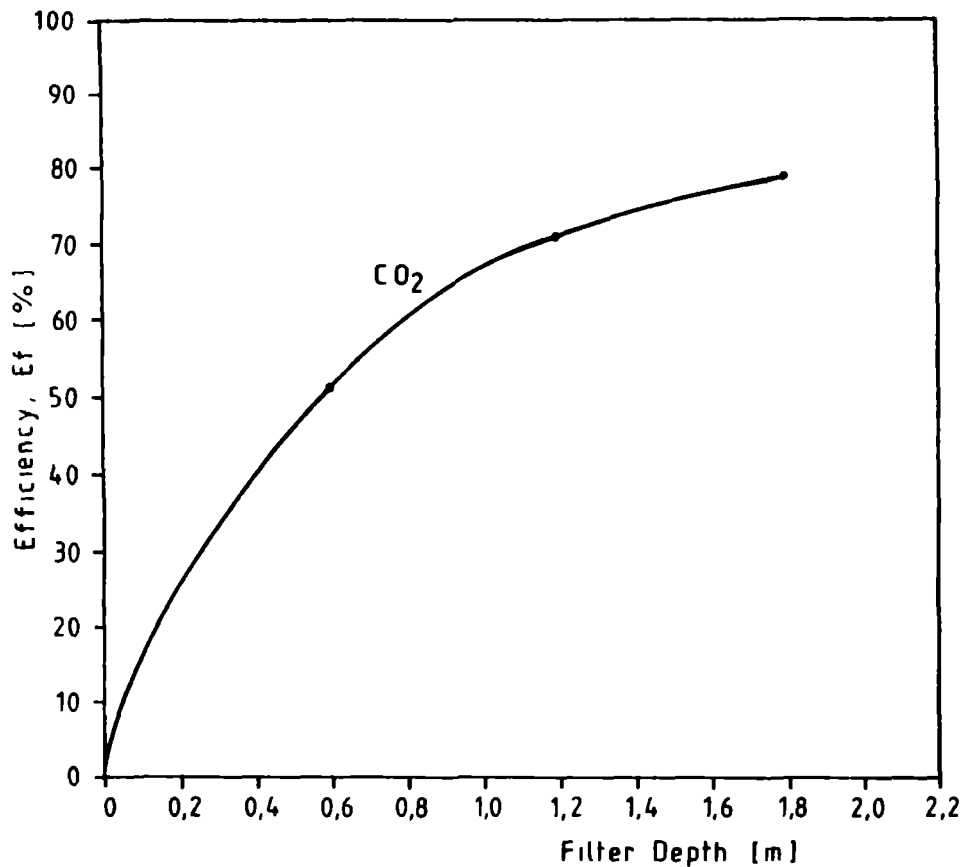


Figure 6.15 Carbon dioxide removal efficiency as a function of filter depth from Siikajoki pilot plant (gravel sizes of 40-50 mm).

In all the cases the concentration of carbondioxide and its removal affected the pH of the water. In most cases the removal of carbondioxide was followed by an increment in pH and to certain extent in reduction of alkalinity of the water.

6.4.2 Other considered parameters

Other water quality parameters relevant with the dry filter and analysed during the study period were manganese, ammonia, ferroiron, hardness, redox potential and temperature.

The removal of manganese and oxidation of ammonia are processes which take place in dry filtration. The accomplishment of these processes requires the maturation of the dry filter and these processes become effective only after long time of operation. Biological manganese removal and ammonia oxidation in Siikajoki and Haukipudas dry filter pilot

plants were not started during this study period. But the removal of both manganese and ammonia oxidation was effective in the dry filter of Raahe which had been in operation for more than one year. In this plant the average manganese concentration in the raw and treated water was 0,14 mg/l and 0,07mg/l respectively. Ammonia concentration in the same water was very small, 0,02 mg/l in the raw water and 0,01 mg/l in treated water.

In the process of filtration the pH and the redox potential of the water increased. This was due to the removal of carbondioxide. However, the results from the analysis of the redox potential can hardly always indicate the effectiveness of the dry filter due to the practical difficulties that exist in its measurement.

6.5 Iron removal efficiency and change in size of filter media

In order to know the effect of the size of filter media on iron removal by dry filter, the filter gravel of 40-50 mm was replaced by gravel of sizes varying from 10-20 mm in the Siikajoki pilot plant. The iron removal of this plant are presented in appendix 6 and plotted in figure 6.16.

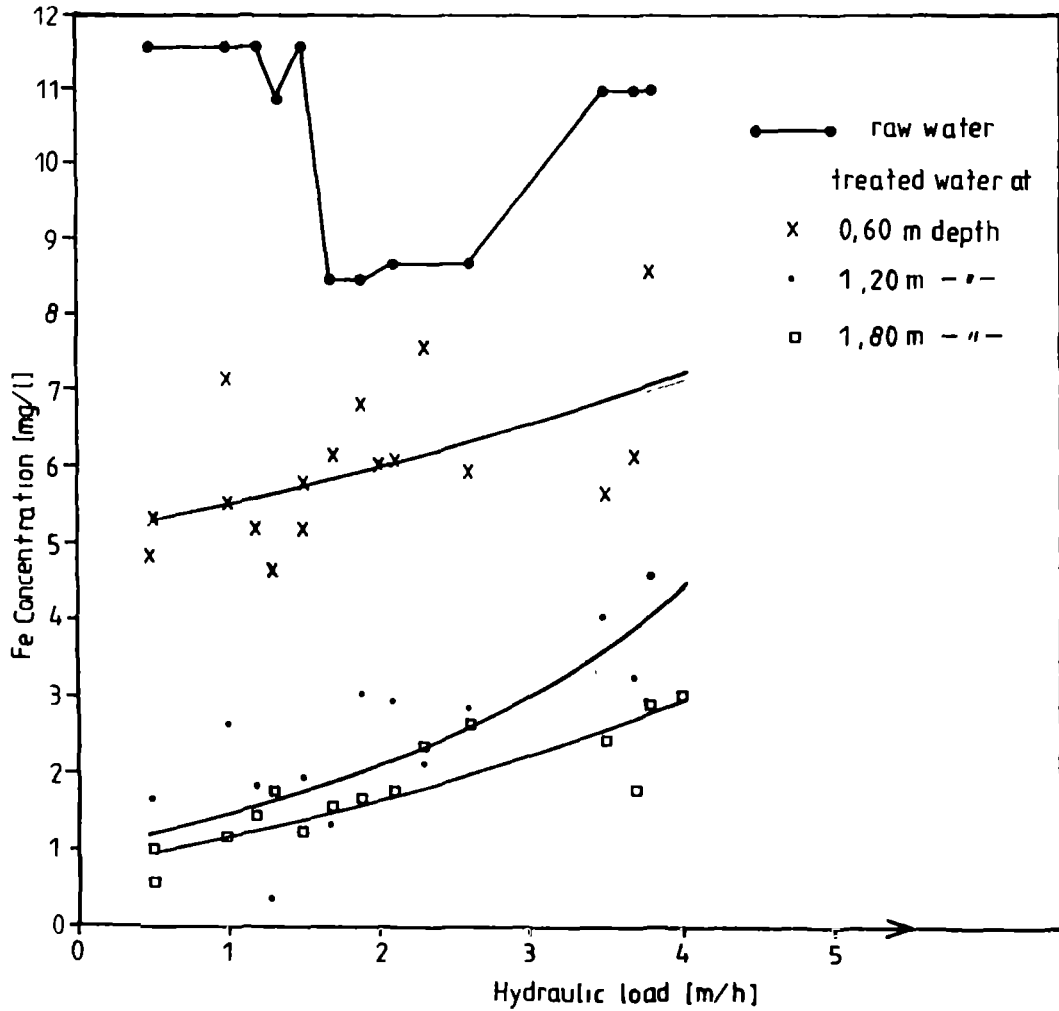


Figure 6.16 Iron removal as a function of hydraulic load as analysed every 4 hours from Siikajoki pilot plant (gravel size of 10-20 mm).

As shown in figure 6.16 iron concentration of 0,6 mg/l in effluent was reached with gravel sizes of 10-20 mm at hydraulic load of 0,5 m/h. From the results obtained mathematical relationship of iron removal and hydraulic load was developed at different depths. The equations describing the relationship are:

$$Fe = 5,1e^{0,08q} \quad \text{at } 0,60 \text{ m} \quad (6.15)$$

$$Fe = 1,07e^{0,35q} \quad \text{at } 1,20 \text{ m} \quad (6.16)$$

$$Fe = 0,9e^{0,31q} \quad \text{at } 1,80 \text{ m} \quad (6.17)$$

where Fe = effluent iron concentration in mg/l
 q = hydraulic load m/h

The correlation coefficients are 0,50, 0,60 and 0,78 in sequence according to the equations.

Treated water samples were taken for analysis at different depths of the filter. Accordingly iron concentration of 0,4 mg/l was obtained at the depth of 1,20 m of the filter while the effluent water contained 1,8 mg/l after the filter depth of 1,80 m. The iron concentration in the effluent was the average of all the effluent waters passing through a particular unit path through the filter. From these results it can be said that there were some areas in the filter where complete iron removal might take place and on the contrary there might be areas where iron removal was very minimal. The reason for such kind of phenomenon was the nonuniform distribution of the water on the top of the filter. During this study it was observed that some parts of the filter were highly loaded while some parts were supplied with small amount of water. However, better iron removal efficiency was obtained with gravel sizes 10-20 mm than when using gravel sizes of 40-50 mm as filter media. Figure 6.17 shows efficiency of the plant.

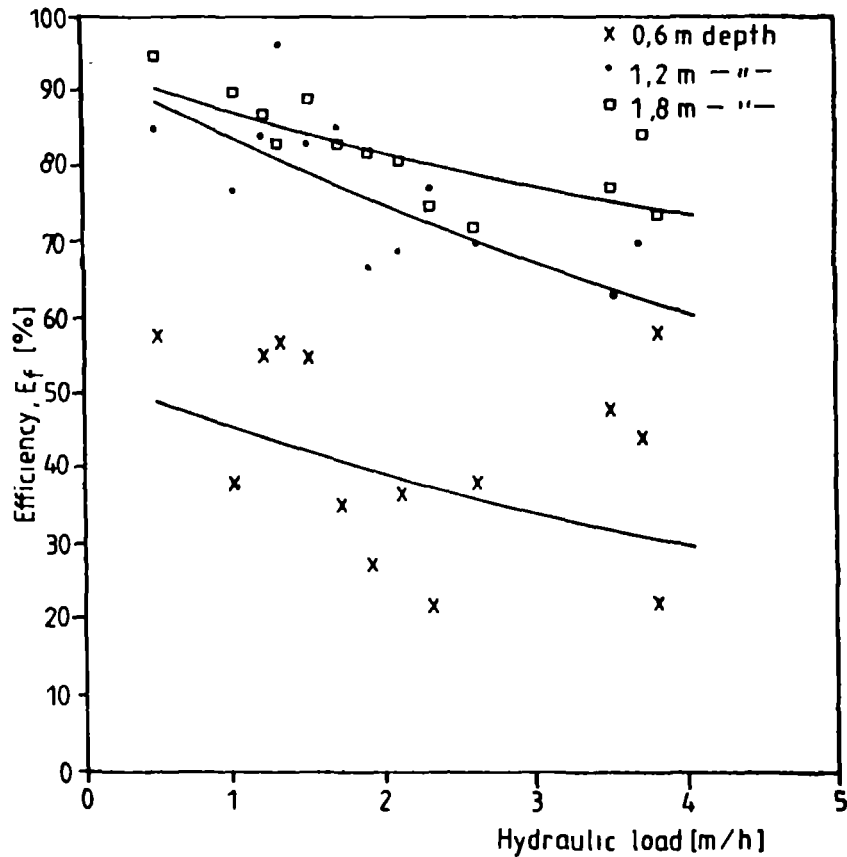


Figure 6.17 Iron removal efficiency at different depths as a function of hydraulic load as analysed every 4 hours from Siikajoki plant (gravel size 10-20 mm).

The equations describing the relationship of the efficiency of the plant to the hydraulic load are:

$$E_f = 53e^{-0,14q} \quad \text{at } 0,60 \text{ m} \quad (6.18)$$

$$E_f = 94e^{-0,11q} \quad \text{at } 1,20 \text{ m} \quad (6.19)$$

$$E_f = 92e^{-0,06q} \quad \text{at } 1,80 \text{ m} \quad (6.20)$$

where E_f and q are the same as defined in equation (6.3) to (6.5)

The correlation coefficients are 0,44, 0,8 and 0,73 in sequence of the equations

For comparison of the effect of different gravel sizes on iron removal by dry filter, the results obtained using gravel sizes of 40-50 mm and 10-20 mm are presented in figure 6.18.

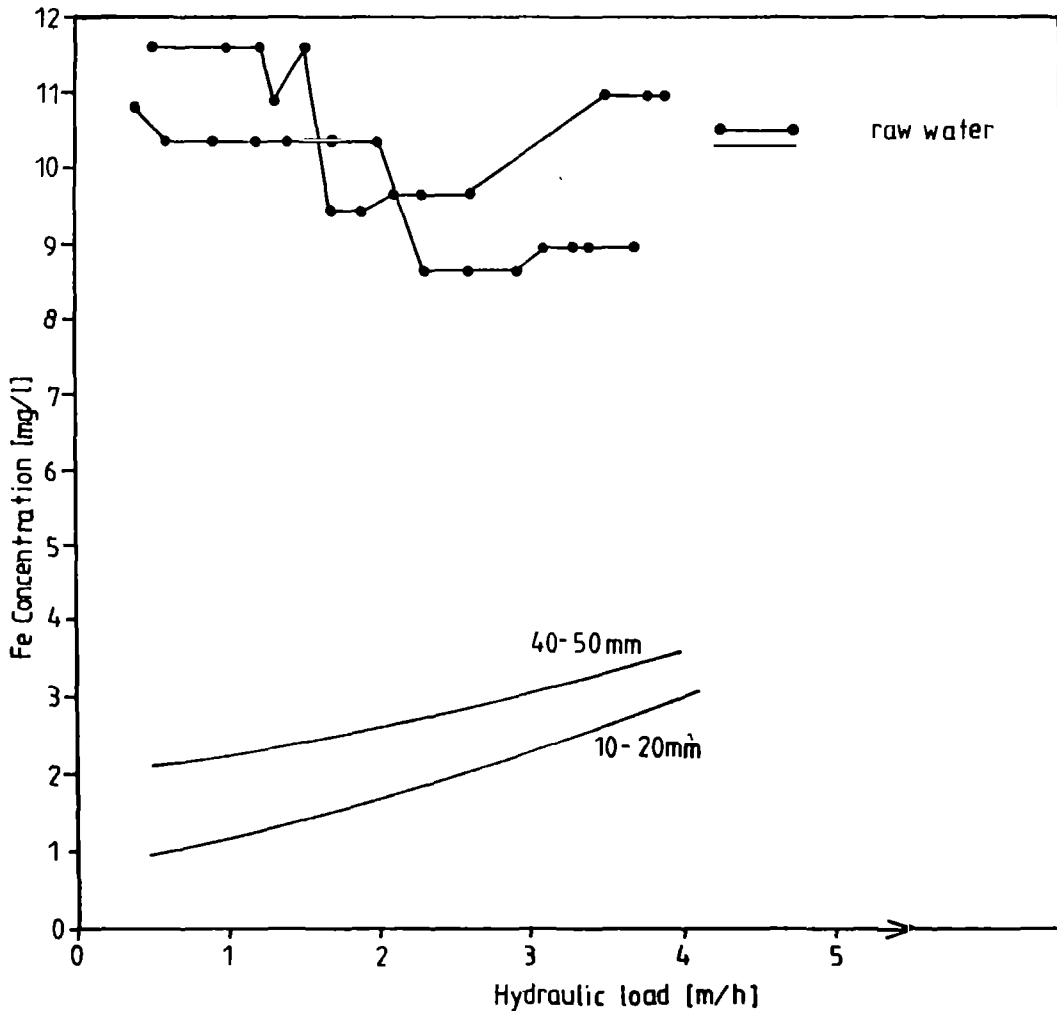


Figure 6.18 Effect of gravel sizes on iron removal under the same hydraulic loading.

From figure 6.18 it can clearly be seen that higher iron removal was achieved by using smaller sizes of filter gravel. This is mostly due to the provision of more surface area per unit volume of filter material for the development of increased number of micro-organisms responsible for iron oxidation. The surface area per unit volume were $110 \text{ m}^2/\text{m}^3$ and $370 \text{ m}^2/\text{m}^3$ for gravel sizes of 40-50 mm and 10-20 mm respectively. This was calculated according to the equation (Huisman 1977 a):

$$S_o = \frac{6}{\phi \sqrt{S_i} \cdot S_j} (1 - P_o) \quad (6.21)$$

where S_o = gross surface area (m^2/m^3)

P_o = porosity of the material (%)

S_i and S_j = the smallest and biggest sizes of filter gravel (m)

ϕ_i = shape factor

= 0,665 for broken gravel (constant)

The void space was measured in the field and approximately found to be 0,45 and 0,42 for gravel sizes of 40-50 mm and 10-20 mm respectively. The change in size of filter material did not affect much the available void volume for the entrance of air and accumulation of iron sludge. Better iron removal efficiency was obtained by using 10-20 mm sizes of gravel than 40-50 mm sizes.

The efficiency of dry filter reaches its maximum capacity when the filter becomes matured after long time of operation. The maturation time under constant hydraulic load was observed in the dry filter of Siikajoki pilot plant. The size of filter gravel used during this observation was 10-20 mm. Figure 6.19 shows the results of the observation.

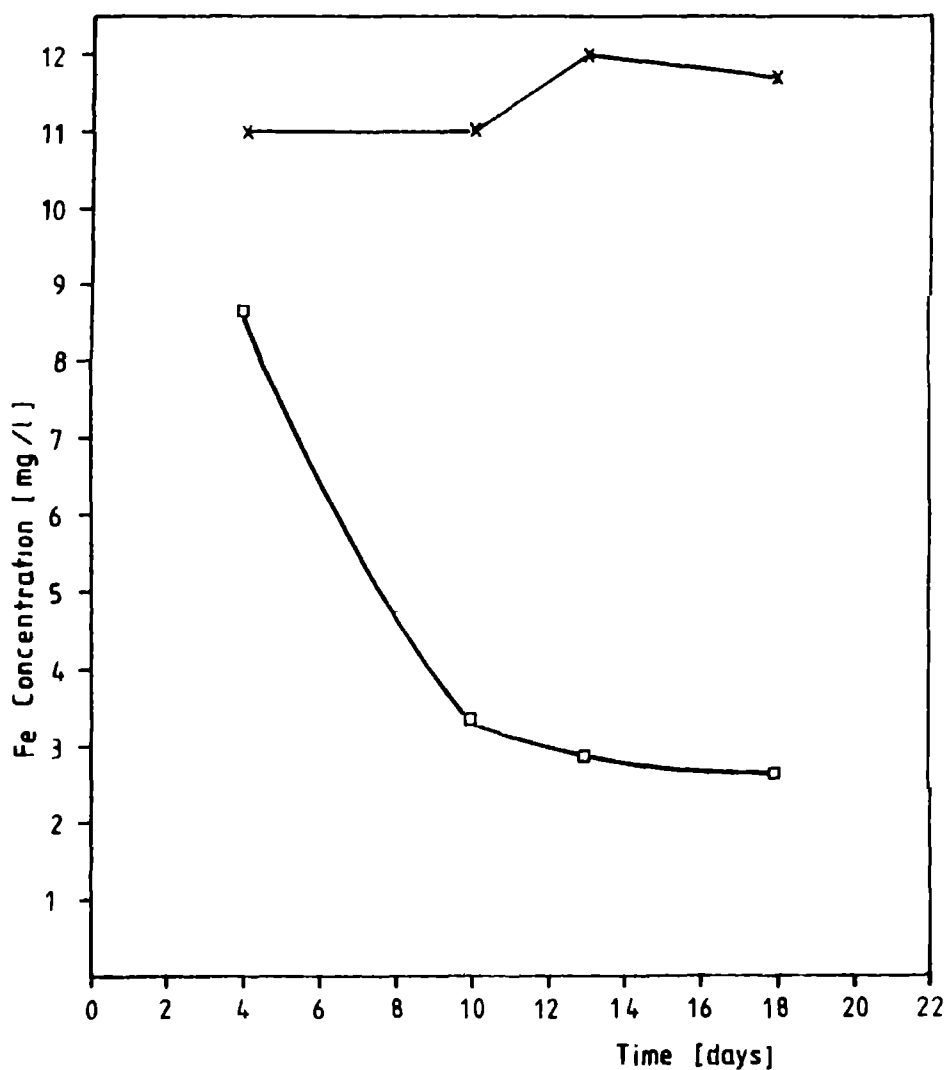


Figure 6.19 Development of dry filter after start up of operation under hydraulic load of 3,7 m/h.

As shown in figure 6.19 the maximum capacity of the plant was not reached. This was because, according to the observation on the 18th day, all the filter materials were not covered by iron biomass. Therefore, there was still available surface to be occupied by iron bacteria and hence the increase in the removal efficiency of the filter was still possible.

7 CONCLUSIONS

Investigation of dry filters for iron removal and water quality improvements was made on Haukipudas and Siikajoki pilot plants and Raahe production plant. According to the results obtained, iron removal by dry filtration is possible provided biological iron oxidation prevailed. Iron removal by dry filter is found to depend on the hydraulic load, depth of filter, iron concentration in the raw water, water distribution on the surface of the filter, size of filter material and ventilation system.

The results obtained from Haukipudas pilot plant were not satisfactory. The reasons for the unsatisfactory results are probably due to uncontinuous raw water supply, insufficient air circulation, inefficient water distribution on the filter surface and some other operational problems. However, iron concentration of 5,3 mg/l in the effluent was obtained at the hydraulic load of 1,4 m/h when the raw water iron concentration was more than 17 mg/l. Therefore, construction of proper and sufficiently bigger pilot plant is very essential to obtain good results.

The results obtained from Raahe dry filter were mostly below the standard limit of 0,3 mg/l at hydraulic loading of less than 1,7 m/h. Effluent iron concentration of less than 0,3 mg/l can be reached at hydraulic loading up to 2 m/h if water distribution of the surface of the filter is made uniformly and air circulation at the upper part of the filter is improved.

In the pilot plant of Siikajoki gravel sizes of 40-50 mm and 10-20 mm were used. The results obtained using 40-50 mm filter sizes gave effluent iron concentration of 2,3 mg/l at hydraulic loading of 2,3 m/h. This result is more than the standard limit, however, iron concentration in the raw water was more than 10 mg/l. The results from Raahe dry filter were mostly below the standard limit. The raw water iron

concentration was small compared to Siikajoki. The use of 40-50 mm sizes of gravel material is not preferred when compared to 10-20 mm gravel sizes since better iron removal can be obtained and construction costs are the same.

Effluent iron concentration of 0,6 mg/l was obtained by using 10-20 mm gravel sizes and filter depth of 1,80 m at hydraulic load of 0,5 m/h. Iron concentration of less than 1,5 mg/l was also obtained at hydraulic loading of less than 2,0 m/h. These results were obtained before the filter was fully matured. Therefore better results can be obtained when the filter becomes fully matured. However, effluent iron concentration of less than 0,3 mg/l may still be obtained by increasing the filter depth to more than 2,0 m. Dry filter of 1,80 m height filled with gravel sizes of 10-20 mm, as it has been tested, can be used as a pretreatment unit e.g. prior to a slow sand filter as a final treatment unit.

The use of dry filter as a pre-treatment unit followed by a slow sand filter as a final treatment unit can be applied provided the iron concentration in the effluent after dry filter is less than 1,0 mg/l. Clogging of the slow sand filter and its cleaning might be a serious problem if higher iron loading is applied.

In both pilot plants iron removal by the dry filters were characterized mostly by an exponential function in relation to hydraulic load. This relationship has to be investigated further to develop a general form of equation that can be applied to any dry filtration condition. When developing such kind of relationship the development of iron oxidizing bacteria, sizes of filter material, filter depth, iron concentration in the raw water and other factors must be taken into account.

From a process point of view, specific surface area and percent void space are the two most important properties of dry filter media. Gravel sizes of 10-20 mm remove more iron

than gravel sizes of 40-50 mm. Smaller gravel sizes provide larger specific surface area which result in greater contact between the iron containing raw water and the micro-organisms. However, the available void volume, which is less for smaller sizes of filter material, will be easily filled by iron sludge and prohibits the entrance of air into the filter and this in its effect might create anaerobic area in the filter body. The oxidized iron then will be reduced to ferro iron and the overall operation of the dry filter will be affected. The use of larger gravel sizes result in increased void space accommodating larger hydraulic loading intensities and better circulation of air through the filter. Air supply to the filter is very essential for the development of micro-organisms, oxygen saturation and removal of carbondioxide from the treated water. Adequate provision of slots, both in size and number is very vital for sufficient entrance of the air into the filter body and escaping of carbon dioxide from the water. Pilot plant study should be made in order to select the necessary sizes of filter material for a particular condition depending on the iron concentration of the raw water. Also, optimum size of filter material that can be used in dry filter should be found. This requires long time investigation of different sizes of filter material under different conditions.

Maturation of dry filter is the development of iron bacteria which is accompanied by iron removal. This requires longer time functioning of the filter and might even taken more than months. The ripening of dry filter with respect to manganese removal and ammonia oxidation takes longer time than for iron removal.

Among the common types of iron bacteria identified during the study were Ochrobium, Arthrobacter, Gallionella and few cells of Siderocapsa and Lepthothrix trichogenes. Metallogenium cell groups covered with iron and manganese precipitates were also identified. These bacteria developed under different iron concentration and characteristics of the raw water. The type of bacteria developed are slightly different at different depths. In addition to their capability to adapt themselves to the prevailing condition, iron bacteria require provision of

favourable environment and media for their growth. The question of the process of biological oxidation of iron is still unanswered and requires full investigation by itself.

Dry filter for iron removal requires very simple construction and does not involve the use of any chemicals. In addition to this, running and maintenance costs are very small. Dry filter reacts very quickly and stabilizes itself to its capacity in short time when loading conditions are altered. This is very beneficial after cleaning of the filter if desired or after maintenance of pumps. The other importance of dry filter is that, regular and complicated method of cleaning is not needed. In principle cleaning of the filter is accomplished during the process of filtration. However, washing the filter by pressure nozzles helps in removing the aged and deposited iron sludge from the filter body. This can be done whenever the need arises, for instance once a year or in two years.

Provision of separate settling tank is essential to catch iron solids escaping through the filter and the washed down iron sludge during the process of filtration. In general dry filter functions as a natural filtration process keeping the natural quality of the water and provides high quality drinking water.

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APPENDICES

APPENDIX 1

Haukipudas - Iron concentration in raw and effluent water and percent removed under different hydraulic load.

Total iron concentration		Area of filter (m ²)	Flow rate (m ³ /h)	Hydraulic load (m/h)	Percent removed %
raw water (mg/l)	treated water (mg/l)				
18,0	15,0	0,126	0,72	5,7	17
17,0	12,0	"	"	"	29
17,0	8,4	"	"	"	51
18,0	12,0	"	"	"	33
17,0	16,0	"	0,51	4,1	6
17,0	16,0	"	"	"	6
17,0	16,0	"	"	"	6
18,0	16,0	"	"	"	11
20,0	18,0	"	"	"	10
17,0	16,0	"	"	"	6
17,0	15,0	"	"	"	12
18,0	14,0	"	"	"	22
19,0	12,0	"	0,45	3,6	37
18,0	11,0	"	0,33	2,6	39
19,0	7,9	"	0,24	1,9	58
17,0	8,2	"	0,21	1,7	52
17,0	11,0	"	0,17	1,4	35
16,0	11,0	"	"	"	31
18,0	9,0	"	"	"	50
16,0	5,3	"	"	"	67
20,6	11,0	"	"	"	47
18,0	10,0	"	"	"	44
18,0	8,2	"	0,16	1,3	54
16,0	8,9	"	0,11	0,8	44

APPENDIX 2

Raahe - Iron concentration in raw water and effluent waters after dry and slow sand filter under different hydraulic load. Influent iron concentration to the slow sand filter is the effluent concentration from dry filter.

Iron concentration and percent removed					Hydraulic load	
Raw water	after dry filter	after slow sand filter	dry filter	slow sand		
Fe (mg/l)	Fe (mg/l)	%removed	Fe (mg/l)	%removed	A = 7,5 m ²	fil.A=80m ²
					(m/h)	(m/h)
1,00	0,35	65	0,02	94	2,2	0,21
1,00	0,32	68	0,02	94	"	"
1,00	0,42	58	0,01	98	"	"
0,80	0,37	54	0,06	84	"	"
0,60	0,28	53	0,02	93	"	"
1,70	0,23	86	0,02	91	2,1	0,20
0,76	0,29	63	0,04	86	"	"
1,30	0,35	73	0,13	63	2,0	0,19
0,77	0,41	47	0,03	93	"	"
2,30	0,44	81	0,09	80	"	"
0,70	0,21	70	0,01	95	"	"
0,69	0,21	70	0,01	95	"	"
0,69	0,22	68	0,02	91	1,4	0,13
0,55	0,23	58	0,13	43	"	"
0,50	0,18	64	0,03	83	"	"
0,43	0,11	74	0,02	82	"	"
0,56	0,36	36	0,10	72	"	"
0,63	0,33	48	0,03	91	"	"
0,58	0,43	26	0,08	81	"	"
0,59	0,27	54	0,07	74	"	"
0,60	0,34	43	0,05	85	"	"
0,50	0,32	36	0,03	91	"	"
0,56	0,20	64	0,06	70	"	"
0,83	0,62	25	0,10	84	"	"
0,62	0,22	65	0,05	77	"	"
0,62	0,33	47	0,03	91	"	"
0,63	0,23	63	0,06	74	"	"

APPENDIX 3

Siikajoki - Iron concentration and percent removed at different depths under different hydraulic load as analysed during the study period (gravel size of 40-50 mm used).

Hydraulic load (m/h)	Iron concentration in raw water (mg/l)	Iron concentration in effluent and % removed at depths of					
		0,6 m		1,2 m		1,8 m	
		(mg/l)	%	(mg/l)	%	(mg/l)	%
4,5	7,4	-	-	-	-	8,6	-
"	8,4	-	-	-	-	8,2	2
"	11,0	-	-	-	-	4,1	63
"	10,0	-	-	-	-	3,9	61
"	10,0	-	-	-	-	6,3	37
4,0	11,0	8,0	27	5,6	49	4,4	60
"	9,7	8,3	14	4,0	59	4,1	58
"	9,3	8,1	13	6,2	34	3,7	60
"	11,0	6,0	45	6,0	45	3,6	67
3,7	9,0	6,5	28	5,2	42	4,4	52
"	11,0	7,9	28	5,0	55	4,9	55
3,4	11,0	-	-	-	-	3,3	70
"	11,0	-	-	-	-	2,9	74
"	9,3	-	-	-	-	2,5	73
"	10,0	-	-	7,1	29	4,6	54
3,2	11,0	6,6	40	5,8	47	5,0	55
2,9	12,0	5,4	55	4,3	64	4,0	67
2,3	11,0	7,6	31	4,8	56	3,8	65
"	11,0	7,0	36	4,7	57	2,3	79
"	11,0	8,2	25	4,3	61	4,1	63
2,2	-	5,8	-	3,3	-	2,4	-
1,6	11,0	6,5	41	4,6	58	3,8	65
0,6	12,0	-	-	-	-	3,4	72

APPENDIX 4

Siikajoki - Iron concentration in effluent at different filter depth under different hydraulic load as analysed every four hours (gravel sizes of 40-50 mm used).

Hydraulic load (mg/l)	Iron concentration in raw water (mg/l)	Iron concentration in effluent and % removed at depths of					
		0,80 m		1,40 m		2,0 m	
		(mg/l)	%	(mg/l)	%	(mg/l)	%
3,7	9,0	6,5	39	5,2	42	4,4	51
3,4	9,0	4,5	50	4,3	52	3,3	63
3,3	9,0	4,6	49	3,5	61	3,3	63
3,1	9,0	4,7	48	4,2	53	3,1	66
2,9	8,7	5,0	43	2,8	68	2,6	70
2,6	8,7	5,1	41	4,0	54	2,7	69
2,3	8,7	5,1	41	3,4	61	2,4	72
2,0	10,4	5,5	47	3,2	69	2,5	76
1,7	10,4	5,7	45	3,2	69	2,4	77
1,4	10,4	6,1	41	4,1	61	2,8	73
1,3	10,4	6,1	41	5,0	52	3,0	71
0,9	10,4	7,0	33	3,5	66	2,4	77
0,6	10,4	5,5	47	2,6	75	2,0	81
0,4	10,8	5,5	49	-	-	2,1	81

APPENDIX 5

Raahe - Iron concentration in raw and effluent water after dry and slow sand filter. Obtained from Raahe water supply company for comparison of summer and winter time functioning of the dry filter. The dates are in the year 1985 and the average flow rate is 500 m³/d.

Date	Raw water		Treated water after dry filter		Treated water after slow sand filter	
	Fe mg/l	pH	Fe mg/l	pH	Fe mg/l	pH
03.01	2,1	-	0,28	-	-	-
22.01	1,0	-	0,35	-	-	-
12.03	0,95	-	0,40	-	-	-
23.04	0,88	6,4	0,56	7,6	0,04	7,4
18.06	0,62	6,3	0,30	7,3	0,02	7,2
02.07	0,71	6,3	0,30	7,3	0,05	7,2
16.07	0,72	6,6	-	7,3	0,03	7,6
30.07	1,0	6,8	0,31	7,3	0,03	7,4
20.08	0,92	-	0,42	-	-	-
03.09	1,80	6,6	0,44	7,3	-	-
17.09	1,10	6,6	0,39	7,4	0,10	7,3
01.10	0,73	6,6	0,55	7,4	0,13	7,5
15.10	0,88	6,7	0,33	(6,7)	0,02	7,5

APPENDIX 6

Siikajoki - Iron concentration in effluent at different filter depth under different hydraulic load as analysed every 4 hours (gravel sizes of 10-20 mm used).

Hydraulic load m/h	Total iron concentration at filter depth of						
	raw	at 0,6 m		at 1,2 m		at 1,8 m	
	water mg/l	Fe total mg/l	% removed	Fe total	% removed	Fe total	% removed
3,8	11,0	8,6	22	4,6	58	2,9	74
3,7	11,0	6,2	44	3,3	70	1,8	84
3,5	11,0	5,7	48	4,1	63	2,5	77
2,6	9,7	6,0	38	2,9	70	2,7	72
2,3	9,7	7,6	22	2,2	77	2,4	75
2,1	9,7	6,1	37	3,0	69	1,8	81
1,9	9,5	6,9	27	3,1	67	1,7	82
1,7	9,5	6,2	35	1,4	85	1,6	83
1,5	11,6	5,2	55	2,0	83	1,3	89
1,3	10,9	4,7	57	0,4	96	1,8	83
1,2	11,6	5,2	55	1,9	84	1,5	87
1,0	11,6	7,2	38	2,7	77	1,2	90
0,5	11,6	4,9	58	1,7	85	0,6	95

APPENDIX 7

Discussion with Mr Tauno Luoto, Raahe water supply technician 10/12/85 at 13,00-14,30 Raahe

The dry filter in Raahe was constructed and put into operation in October 1984. They selected dry filter because the use of chemicals was not liked by themselves and the customers. When they constructed this plant, it was first used as a pilot plant but later on put into a fullscale production plant. During the design there were not as such any design criteria.

After this plant was put into operation they applied sodium hypochlorite once to clean the iron sludge from the filter. At this moment, he stated that, all the iron sludge most probably the iron bacteria were washed down from the filter. Iron removal started in a short time after the application of raw water again. The filter was washed once though they have not changed the stones. As mentioned during the discussion there was no difference in iron removal efficiency during summer and winter seasons.

After the water is filtered by dry filter it is again filtered by a slow sand filter. The slow sand filter was constructed because they have not known that the results of the dry filter will be good.

The top surface area of the slow sand filter was 80 m² and sand depth of 80 cm. The depth of the filter bottom was 52 cm and consists of gravel sizes of 1,5 - 3 mm on top, 4 - 8 mm in the middle and 16 - 32 mm at the bottom with depths of 6 cm and 40 cm respectively. The sand grains in the filter (80 cm thick) consists of $d_{15} = 0,14 - 0,18$ mm and $d_{85} = 0,3 - 0,4$ mm.

Mr Luoto stated that they want to change the top layer of the sand to bigger sizes so that iron can go deeper into the filter and provides longer filter run. At this moment the iron deposits clog the top surface and cleaning is done every 3 weeks.

The water after the slow sand filter was mixed with water coming from other sources and they add Ca(OH)_2 and KMnO_4 to

raise the pH and remove other oxidizable contaminants. The daily water supply to the town of Raahе was 2200 m³/d and one third of this was treated by the dry filter and the slow sand filter in series.

Finally Mr Luoto stated that, if the need for iron removal arises in certain water treatment works his proposal will be to use dry filter. This is because of the easiness in operation of the dry filter and no chemical demand for the process.

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