

# Drinking water treatment and chemical well clogging by iron(II) oxidation and hydrous ferric oxide (HFO) precipitation

C. G. E. M. van Beek, C. H. M. Hofman-Caris and G. J. Zweere

## ABSTRACT

Removal of iron(II) from groundwater by aeration and rapid sand filtration (RSF) with the accompanying production of drinking water sludge in the preparation of drinking water from groundwater, and chemical well clogging by accumulation of hydrous ferric oxide (HFO) precipitates and biomass after mixing of oxygen containing and of iron(II) containing groundwater, are identical processes. Iron(II) may precipitate from (ground)water by homogeneous, heterogeneous and/or biological oxidation, where the contribution of these processes, and thus the characteristics of the corresponding HFO precipitates, is a function of pH and process-conditions. Under current conditions in drinking water treatment, homogeneous oxidation dominates above  $\text{pH} \approx 7.75$ , and heterogeneous and biological oxidation below this value. In chemical well clogging, this transition occurs at  $\text{pH} \approx 7.0$ . This information is relevant for the optimization of removal of iron(II) from groundwater by aeration and RSF and the corresponding quality of the produced drinking water sludge, and for the operation of wells clogging by accumulation of HFO precipitates and biomass.

**Key words** | aeration, chemical well clogging by accumulation of hydrous ferric oxide (HFO), drinking water production from groundwater, homogeneous, heterogeneous and biological oxidation of iron(II), rapid sand filtration (RSF)

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## HIGHLIGHTS

- Removal of iron(II) by aeration and rapid sand filtration (RSF) in drinking water treatment and accumulation of hydrous ferric oxides (HFOs) and biomass in chemical well clogging are identical processes.
- Consequently, information and experiences gained in the one field may be compared and applied in the other field and vice versa.
- This information is helpful in optimization of the drinking water treatment process and of the quality of the produced HFO-sludge.

## ABBREVIATIONS

AMD	Acid mine drainage	FeOB	Iron(II) oxidizing bacteria
Fet	Total (divalent and trivalent) (dissolved and solid) iron concentration	HFO	Hydrous ferric oxide
Fe(II)	Total divalent iron concentration	RSF	Rapid sand filtration
FeAC	Iron(II) adsorption capacity	SIR	Subsoil iron removal

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## INTRODUCTION

In the Netherlands (Western Europe) drinking water distributed by water utilities may contain not more than 0.2 mg/l total iron. This standard has been decreased by the water utilities to 0.05 mg/l, and even 0.02 mg/l. About two-thirds of the drinking water in the Netherlands is prepared from groundwater, abstracted at somewhat more than 200 well-fields from alluvial sediments at depths between 25 to 150 m. As the abstracted groundwater may contain up to circa 25 mg/l iron(II), removal of iron(II) is a relevant item. Usually, iron(II) is removed by aeration, producing hydrous ferric oxide (HFO) precipitates, which are removed by rapid sand filtration (RSF). The same applies to manganese(II). After removal of iron and manganese, this water is distributed as drinking water. About 10% of the plants need additional treatment for removal of DOC/color, and many more for pH/hardness correction. For extensive information see Gimbel *et al.* (2004) and Sommerfeld (1999).

In the Netherlands, about a third of the number of well fields, abstracted groundwater consists of a mixture of iron(II) as well as oxygen containing water (Supplementary material, Figure A1). In such waters HFOs will precipitate, resulting in clogging of these wells and raw water lines. For extensive information see Houben & Treskatis (2012), Smith & Comeskey (2010) and Tyrrel & Howsam (1994).

Removal of iron(II) by aeration and RSF, and chemical clogging of wells by accumulation of HFOs and biomass are identical chemical/biological processes occurring under very different conditions. Here both processes will be compared mutually and with physical-chemical and microbiological information. The purpose of this review is to arrive at a clear understanding of the relevant processes, elucidate, and if possible remedy eventual problems, and apply experiences from the preparation of drinking water on the occurrence of chemical well clogging and vice versa.

## OXIDATION OF IRON(II) BY OXYGEN

Dissolved iron(II) may be oxidized by oxygen by three processes, i.e. homogeneous, heterogeneous and biological

oxidation (Supplementary material, section C), producing HFO precipitates:

$$\frac{d}{dt}[\text{Fe(II)}] = \frac{d}{dt}[\text{Fe(II)}]_{\text{homo}} + \frac{d}{dt}[\text{Fe(II)}]_{\text{hetero}} + \frac{d}{dt}[\text{Fe(II)}]_{\text{biol}} \quad (1)$$

This combination of processes occurs everywhere where dissolved iron(II) and oxygen mix: in nature (groundwater-seeps), in preparation of drinking water from groundwater by aeration and RSF, in wells clogging by abstraction of a mixture of oxygen containing and of iron(II)-containing water, etc. The contributions of the various iron(II) oxidizing processes in Equation (1) depends on the circumstances.

In homogeneous oxidation, iron(II) as well as oxygen are present in solution, producing HFO-flocs. The accompanying rate-equation reads (Stumm & Morgan 1996):

$$\frac{d}{dt}[\text{Fe(II)}] = -k_{\text{homo}} \frac{[\text{Fe(II)}][\text{O}_2]}{(\text{H}^+)^2} \quad (2)$$

with  $k_{\text{homo}}$  rate-constant,  $[\text{Fe(II)}]$  and  $[\text{O}_2]$  concentrations of iron(II) and oxygen, and  $t$  time. Assuming constant concentration of oxygen and constant pH, Equation (3) is obtained:

$$[\text{Fe(II)}] = [\text{Fe(II)}]_0 \exp\left(-k_{\text{homo}} \frac{[\text{O}_2]}{(\text{H}^+)^2} t\right) \quad (3)$$

with  $[\text{Fe(II)}]_0$  and  $[\text{Fe(II)}]$  as initial and actual concentration of iron(II). Under favorable conditions (high pH, oxygen saturation) this is a fast process (Supplementary material, section C) producing large HFO flocs; under unfavorable conditions small flocs, which may pass the sand-filter and enter the drinking-water distribution-system, and after back washing hardly will settle down in the sludge basin.

Heterogeneous oxidation of iron(II) by oxygen is a two stage process: first adsorption of iron(II) by HFO surfaces, and subsequently oxidation of adsorbed iron(II) by oxygen. The accompanying rate-equation, somewhat modified after Tamura *et al.* (1976), may be represented as:

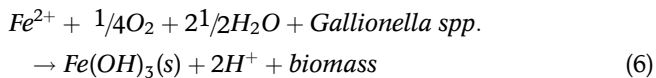
$$\frac{d}{dt}[\text{Fe(II)}] = -k_{\text{hetero}} \frac{[\text{Fe(II)}][\text{O}_2]}{(\text{H}^+)} \text{FeAC} \quad (4)$$

with FeAC as the iron(II) adsorption capacity of HFO, which is a function of pH and HFO mineralogy (Schwertmann & Fechter 1982). Solution of (4) at constant oxygen concentration, pH and adsorption-capacity yields:

$$[Fe(II)] = [Fe(II)]_0 \exp\left(-k_{hetero} \frac{[O_2]FeAC}{(H^+)} t\right) \quad (5)$$

This relation has been experimentally confirmed by, among others Demir & Tufekci (2002), Tufekci *et al.* (2000) and Dietz & Dempsey (2017): the higher the pH and the more HFO-sludge present (the greater the surface area), the faster the oxidation. Subsoil iron removal (SIR) is based on this process (van Beek 1985).

In the oxidation of iron(II), (a small amount of) energy is produced, which may be used by neutrophilic iron(II) oxidizing bacteria (FeOB):



*Gallionella* spp. completely depend on this process for their energy supply, while others, like *Leptothrix* spp., in the (near) absence of iron(II) or oxygen may switch to another chemical process/energy source (Kappler *et al.* 2015). Until recently, it was assumed that *Gallionella* spp. especially occur in acidic micro-aerophilic environments, but recently it has been demonstrated that they play an important role in the removal of iron(II) under neutral oxygen-rich conditions (Hanert 2006; de Vet *et al.* 2011; Eggerichs *et al.* 2014; Wullings 2016).

*Gallionella* spp. excrete the produced HFOs, covered by 'extracellular polymeric substances' (EPS), in twisted stalks (spirils), characteristic for *Gallionella*, as amorphous HFO, Ferrihydrite, Lepidocrocite or Akaganeite (Chan *et al.* 2011). These biologically produced HFO precipitates serve as a substrate for further heterogeneous oxidation of iron(II) (Chan *et al.* 2011; Melton *et al.* 2014; Wang *et al.* 2014), in some way preventing encapsulation of FeOB by these precipitates. Bruins (2016) demonstrated the same phenomenon for manganese removal by oxidation and RSF: initially biological oxidation, after some time followed by heterogeneous oxidation.

## DISTRIBUTION OF IRON(II) OXIDATION PROCESSES AS FUNCTION OF PH

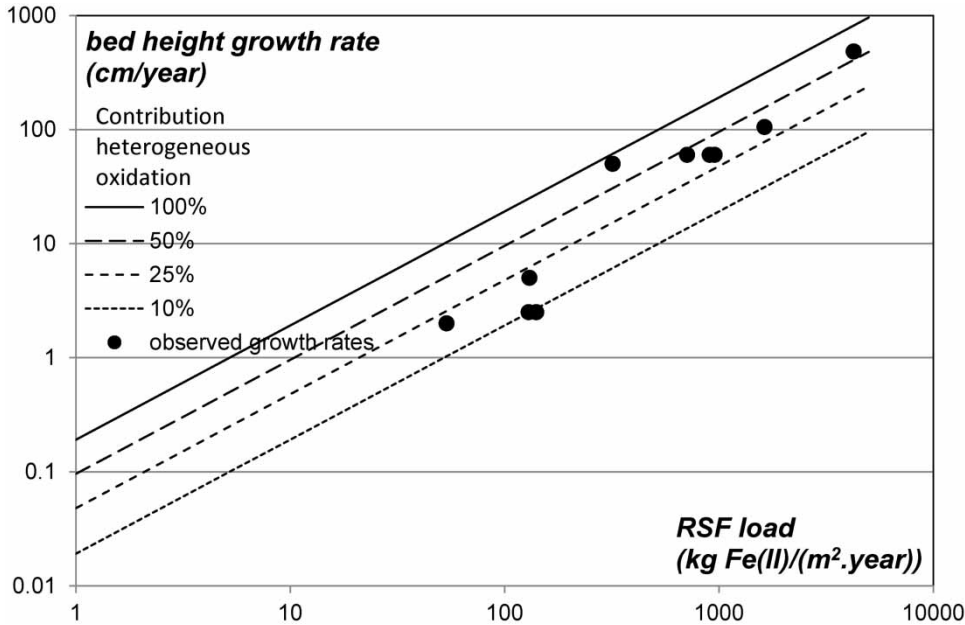
A rough distribution of the various iron(II) oxidizing processes as a function of pH may be obtained in water treatment, as processes here are designed for ease of study. The contribution of homogeneous oxidation may be estimated by calculation (van Beek *et al.* 2016), of heterogeneous oxidation from bed-height growth, and of homogeneous and heterogeneous oxidation from back-washing. The complement is ascribed to biological oxidation. However, de Vet *et al.* (2011) was able to arrive at correct global estimates between two consecutive back-washings (van Beek *et al.* 2016).

### Bed-height growth

As a result of heterogeneous oxidation of iron(II), the grains of the sand filter will grow, increasing the height of the filter bed (van Beek *et al.* 2016). As homogeneous and biological oxidation will not or negligibly contribute to bed-height-growth, the contribution of heterogeneous iron(II) oxidation may be estimated from this increase as the product of bed-height-growth, and density and iron content of this iron-precipitate. Due to mutual friction of the grains during back-washing, a part of the HFO precipitates will be removed. Comparison of this calculated value with the iron load of the sand-filter, yields a (relative) minimum contribution of heterogeneous oxidation. Disturbance of the bed-height-growth rate by leaching of filter sand during back washing, or supply of sand by sand-delivering wells is not considered here.

Figure 1 shows the calculated bed-height-growth-rate as a function of bed load for various relative contributions of heterogeneous iron(II) oxidation (van Beek *et al.* 2016). The observed values for the bed-height-growth-rate will be lower than the maximum calculated values: next to heterogeneous oxidation also homogeneous and biological oxidation of iron(II) may occur, and during back washing abrasion of the 'coated' grains results in the release of iron-sludge, which will be carried off.

Figure 1 shows high relative contributions of heterogeneous oxidation (25 to 90%) at high loads, and low



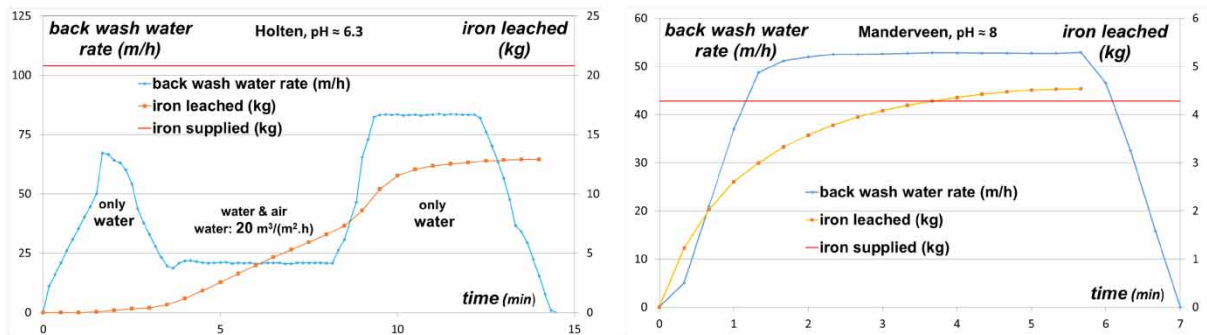
**Figure 1** | Calculated bed-height-growth-rate, in cm/year, as function of bed load, in kg Fe(II)/(m<sup>2</sup>·year), for various relative contributions by heterogeneous oxidation. A bed-height-growth-rate of 10 cm/year and a bed-load of 100 kg Fe(II)/(m<sup>2</sup>·year) compare with a contribution by heterogeneous oxidation of (minimal) 50%. Applied values: volume density of the accretion: 1.22 kg dry matter/l, and iron content of the accretion: 0.428 kg Fe/kg dry matter. Data provided by several drinking water utilities.

relative contributions (10 to 25%) at low loads: high loads are treated in open basins, which are easy to clean, and low loads in closed vessels, which are hard to clean.

### Back-washing

Information about the contributions of the various oxidation processes may also be obtained from back-washing of sand-filters. Figure 2 shows the results of such experiments of well-field ‘Holten’ (pH after filtration ca. 6.3), and of well-field ‘Manderveen’ (pH after filtration ca. 8).

By back washing, all HFOs, precipitated during the preceding filtration period by homogeneous oxidation (in the supernatant water and accumulated upon the top of the sand filter), will be removed with minimal effort, see well-field Manderveen. At well-field Holten, with heterogeneous and biological oxidation, not all HFOs, accumulated during the preceding filtration period, could be removed, even not with more extensive backwashing procedures (involving higher flow rates, longer back washing times or a combination of water and air). The remaining part is considered the minimum contribution by heterogeneous oxidation.



**Figure 2** | Amount of iron (kg) removed by back-washing, compared with the amount accumulated during the preceding filtration period. Left: well-field Holten (iron concentration in abstracted groundwater: 3–8 mg/l, pH effluent 6.2–6.3). Right: well-field Manderveen (iron: 0.7–0.9 mg/l, pH effluent 7.9–8.0), both operated by water utility Vitens.

Figure 3 shows the contribution of the oxidation processes distinguished in the removal of iron(II) by aeration and RSF (augmented after van Beek et al. 2016) as a function of pH. In Figure 3, the contribution by homogeneous oxidation has been calculated from the residence time in the supernatant water, or estimated from back-wash experiments (Manderveen). At none of the well fields a complete removal was estimated, which may be explained by a too low value of the reaction-constant, or the removal of the last traces of iron(II) in the top of the sand-filter. Due to the small surface area (the sides and the bottom of the water-volume on top of the sand-filter, versus the grains of the sand-filter) resulting in a few attachment possibilities, and the low concentration of iron(II), the contribution of biological oxidation is considered negligible.

The contribution of heterogeneous oxidation of iron(II) has been estimated from the bed-height-growth and from back-wash experiments. Both ways of estimation are by definition too low, as back-washing will remove an unknown part of the HFO precipitates. The remaining part of

iron(II) has been removed by biological oxidation. Apparently, *Gallionella* spp. is able to survive backwashing (in the sand-filter, in the air-sprayers?).

Notice in Figure 3 the distinction between homogeneous oxidation of iron(II) by oxygen at pH > ca. 7.75 and heterogeneous and biological oxidation at pH < ca. 7.75.

The presence of water on top of the sand-filter serves two different goals: a thick layer of water at 'high' pH serving as a reaction volume for homogeneous oxidation, and a thin layer at 'low' pH facilitating a uniform distribution of water over the filter-surface.

The contributions of heterogeneous and of biological oxidation may be manipulated: the lower the filter velocity, the greater the contribution of biological oxidation (van Beek et al. 2016); the lower the pH, the lower the adsorption capacity of HFOs for iron(II) and the lower the contribution of heterogeneous oxidation; the less frequent and the less rough back washing, the less the loss of FeOB and the greater the contribution of biological oxidation, etc. In

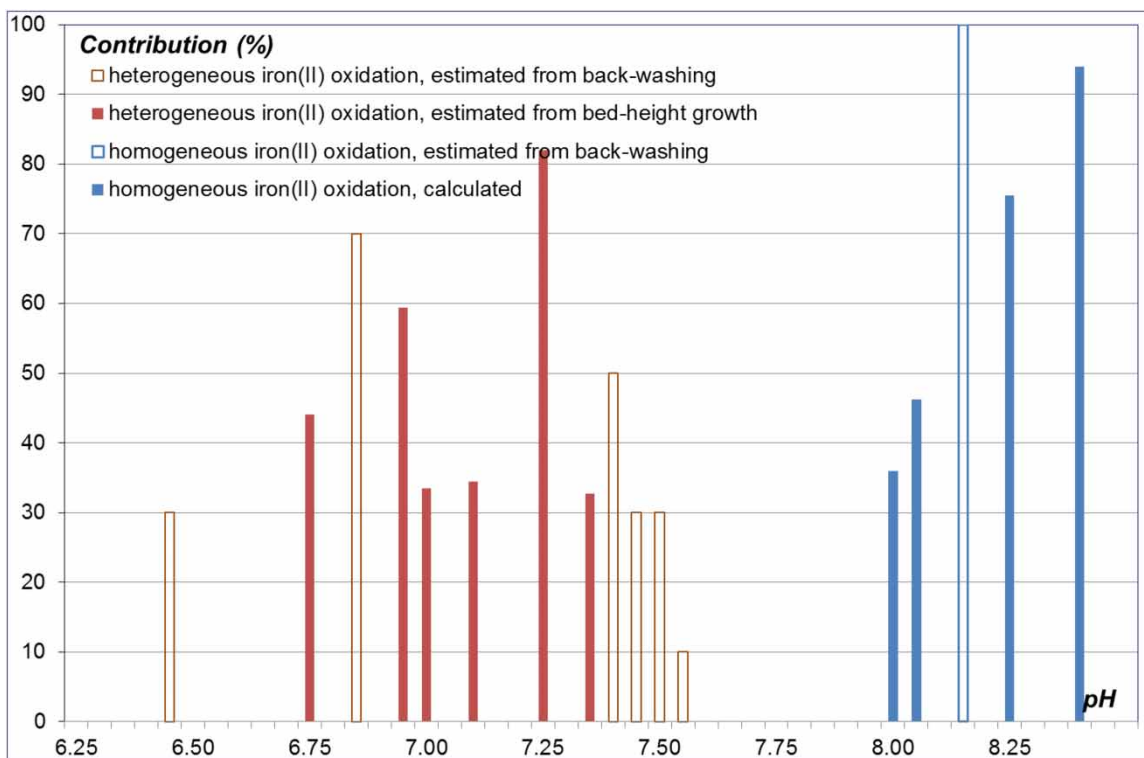


Figure 3 | Distribution of iron(II) oxidation processes in the removal of iron(II) by aeration and RSF as function of pH (extended after van Beek et al. 2016). The values for heterogeneous oxidation are by definition too low; the phenomenon 'bed height growth' is relevant.

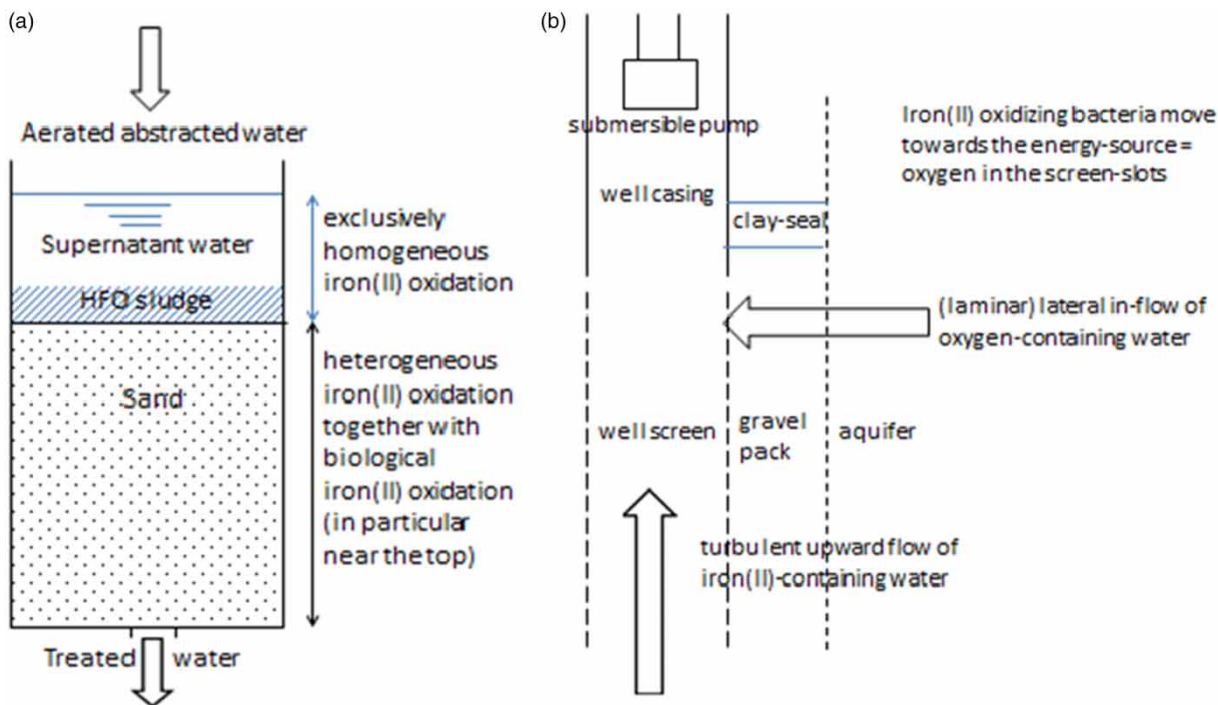
order to prevent agglomeration of filter-sand, (and to prevent the filter bed exceeding the sand-vessel,) regular back washing is necessary. In order to avoid competition with heterogeneous oxidation of iron(II) as much as possible, biological oxidation of iron(II) will predominantly occur on and just below the top of the sand-filter, as there competition with heterogeneous oxidation will be minimal.

### Well clogging by accumulation of HFO precipitates and biomass

Wells, abstracting a mixture of iron(II)- and of oxygen containing groundwater (Supplementary material, sections A and B), will clog by accumulation of HFO precipitates, whether or not together with accumulation of biomass consisting of *Gallionella* spp. Figure 4(a) shows schematically a sand-filter. Due to the unfavorable conditions for biological and heterogeneous oxidation, in the supernatant water almost only homogeneous oxidation of iron(II) will occur.

Figure 4(b) schematically shows the occurrence of chemical well clogging by accumulation of HFO precipitates and biomass. Comparison of Figure 4(a) and 4(b) demonstrates

that conditions in a clogging well are far more comparable with those in the supernatant water of the sand-filter than those in the sand filter itself (small versus large surface area for precipitation of HFOs and for attachment of *Gallionella* spp.). Consequently, in chemical well clogging conditions are much more favorable for homogeneous oxidation than for heterogeneous and biological oxidation (however not optimal, because in chemical well clogging the abstracted groundwater cannot be saturated with respect to oxygen). The HFO flocs, precipitated as a result of homogeneous oxidation, which cannot or hardly attach on surfaces, will accumulate in the (long) raw-water lines, over time resulting in ever smaller effective diameters of these lines, and consequently ever smaller yields, see Figure 5 (van Beek 2018). Burté *et al.* (2019) present a fine example of a heat extraction system: no clogging of the abstraction well, but accumulation of HFO deposits in the waterlines and clogging of the infiltration well. During periods of rest, homogeneous oxidation of iron(II) will continue: small HFO flocs precipitated during these periods may accumulate inside the well, and may, in the presence of a (small) hydraulic gradient over the height of the well-screen, leave the well and accumulate in the



**Figure 4** | (a) (left): Longitudinal section through a sand-filter and (b) (right): longitudinal section through a well suffering chemical well clogging resulting from abstracting a mixture of (deep) iron(II) containing and (shallow) oxygen containing water (not to scale).

screen slots, gravel pack, well bore and further away in the aquifer (Houben & Weihe 2010; van Beek 2018).

With decreasing pH, conditions for homogeneous oxidation become less favorable, whereas both heterogeneous and biological oxidation are favored. *Gallionella* spp. will look for the most favorable conditions (energy source): the slots of the well-screen, along which oxygen-containing groundwater enters the well (similar to the accumulation of chemical precipitates and biomass around ‘deep sea vents’). The accumulation of biomass and HFO precipitates, and thus clogging of (the slots of) the well-screen will start where oxygen containing water enters the well (boundary layer oxygen-containing and iron(II)-containing water), and subsequently extend in the downstream as well as slowly in the upstream direction, causing well-screen and well-head clogging, see Figure 5. Over time, the well-screen will become clogged more and more, obstructing the inflow more and more, resulting in an increasingly

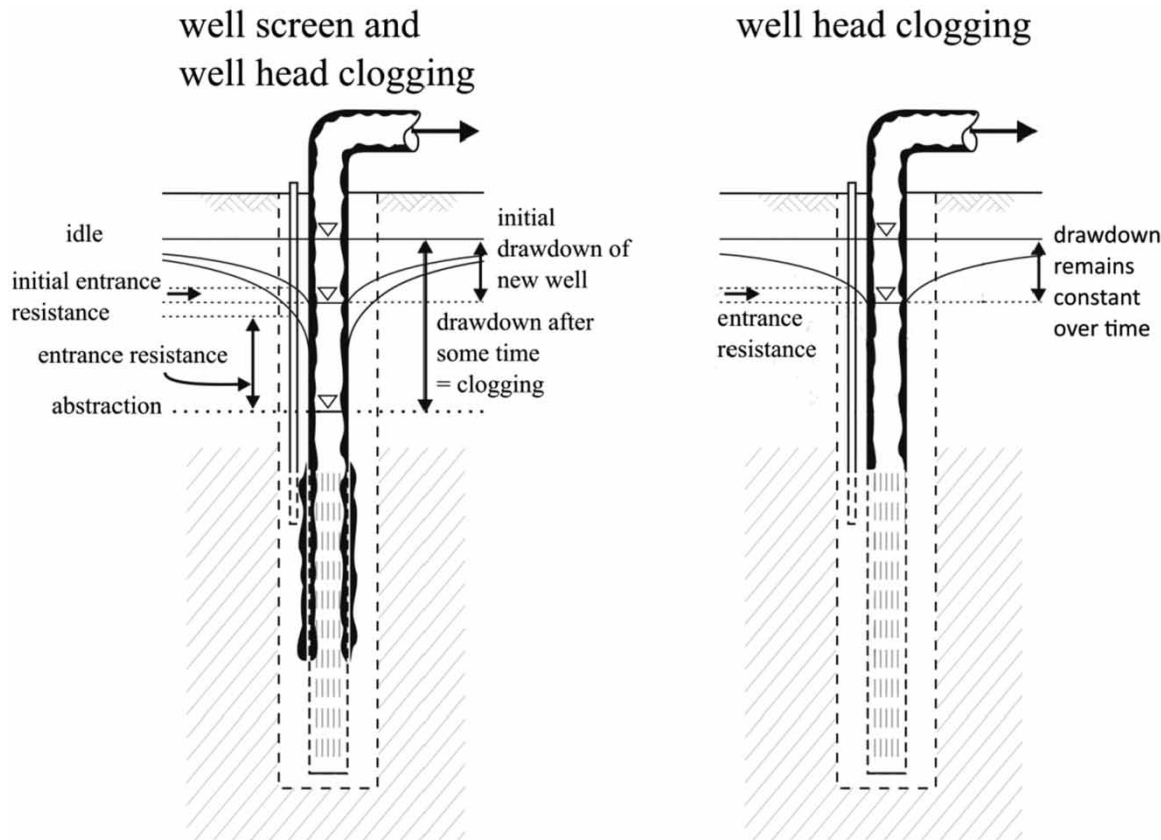
deeper water level inside the well, in the end ceasing production, if not rehabilitated in time.

## DISCUSSION

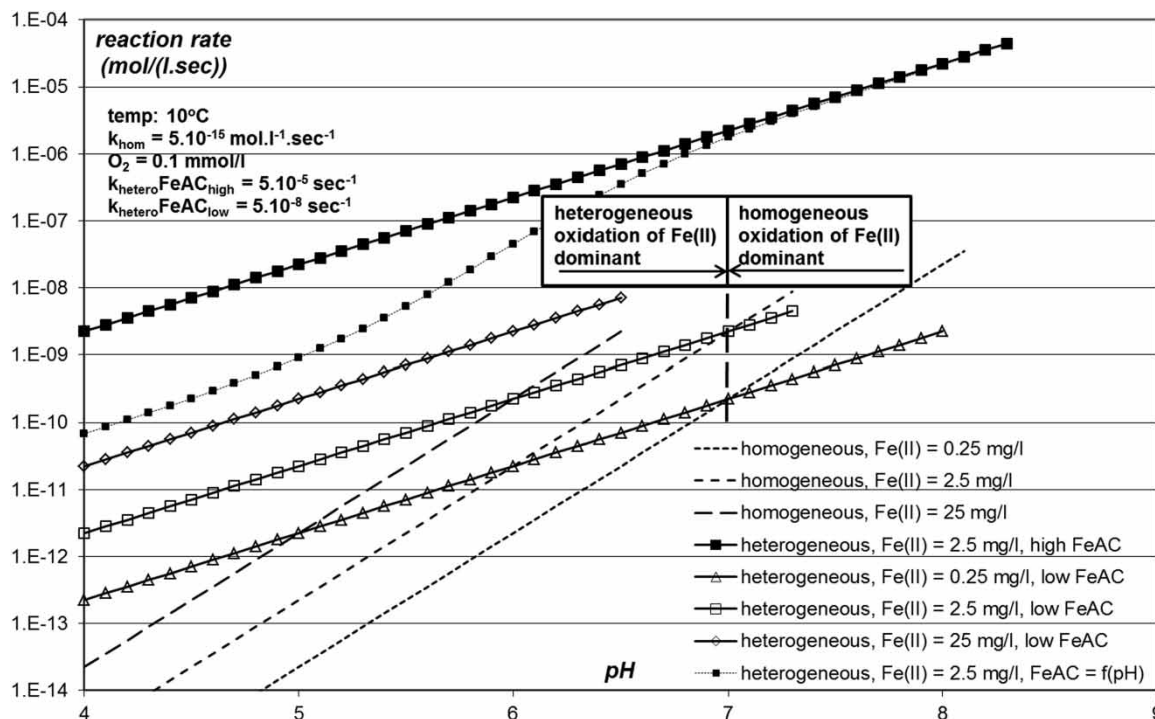
Substitution of Equations (2) and (4) into (1) yields:

$$\begin{aligned} \frac{d}{dt}[Fe(II)] = & -k_{homo} \frac{[Fe(II)][O_2]}{(H^+)^2} \\ & - k_{hetero} \frac{[Fe(II)][O_2]}{(H^+)} FeAC(pH) \\ & + \frac{d}{dt}[Fe(II)]_{biol} \end{aligned} \quad (7)$$

Figure 6 shows the homogeneous oxidation rate of iron(II) by oxygen as a function of pH for three concentrations of iron(II) together with the heterogeneous oxidation rate for various values of the iron(II) adsorption



**Figure 5** | Difference between well-head and well screen clogging (left) and only well-head clogging (right). In well-head clogging the well casing, the submersible pump (not represented here), but in particular the raw-water lines are covered/clogged by HFO precipitates, resulting in an ever decreasing yield; in well-screen clogging the clogging of the screen becomes ever more serious, until the well ceases to deliver water.



**Figure 6** | Reaction rate as a function of pH for homogeneous and for heterogeneous oxidation of iron(II) by oxygen for various concentrations of iron(II) and for various iron(II) adsorption capacities. Values applied:  $k_{\text{hom}}: 5.10^{-15} \text{ mol.l}^{-1}.\text{sec}^{-1}$ , and  $k_{\text{hetero,FeAC}}$ : respectively  $5.10^{-8}$  (chemical well clogging) and  $5.10^{-5} \text{ sec}^{-1}$  (RSF), in such a way that heterogeneous and homogeneous oxidation occur next to each other as function of pH.

capacity (FeAC). Figure 6 demonstrates that for low values of FeAC, as in chemical well clogging, homogeneous and heterogeneous oxidation of iron(II) occur next to each other as function of pH: homogeneous oxidation at 'high' pH (in Figure 6: pH > 7), and heterogeneous oxidation at 'low' pH (in Figure 6: pH < 7). This distribution provides an elegant explanation for the occurrence of both clogging phenomena: well-head clogging by homogeneous oxidation if pH > 7, and well-screen (and also, to a lesser extent, well-head) clogging by heterogeneous and biological oxidation if pH < 7 (van Beek 2018).

Figure 6 also shows the heterogeneous iron(II) oxidation rate for a high value of the FeAC, representative for RSF, and, as the negative charge of HFO's increases with increasing pH, also for a pH-dependent value of the FeAC (Dempsey *et al.* 2012). Under these conditions, over the whole pH-range, heterogeneous oxidation of iron(II) is dominant over homogeneous oxidation.

Because the reaction rates of homogeneous and of heterogeneous oxidation increase with increasing pH,

conditions for *Gallionella* spp. are most favorable at low pH, see Figure 6.

Conditions for biological oxidation are even becoming more unfavorable with increasing pH due to a concomitant decreasing concentration of iron(II) (Supplementary material, section A).

Figure 6 also shows that for lower FeAC values, the dominant iron(II) oxidation process in particular is a function of pH, and to a lesser extent of the concentrations of iron(II) and of oxygen: at high pH the occurrence of homogeneous oxidation of iron(II) is dominant, at neutral pH of heterogeneous oxidation and at low pH of biological oxidation by *Gallionella* spp., moreover favored by a high iron(II) concentration. This distribution is in line with the distribution demonstrated by Dempsey *et al.* (2012) and by Kirby & Kostak (2002): at low pH HFO's still have some negative charge and some heterogeneous oxidation will occur, advantageously utilized in the treatment of acid mine drainage (AMD). Minimum pH values with respect to the removal of iron(II) in the treatment of drinking



water by aeration and RSF are presented by Czekalla (1997): pH = 6.0, and later a value of pH = 5.5 (Czekalla 2008).

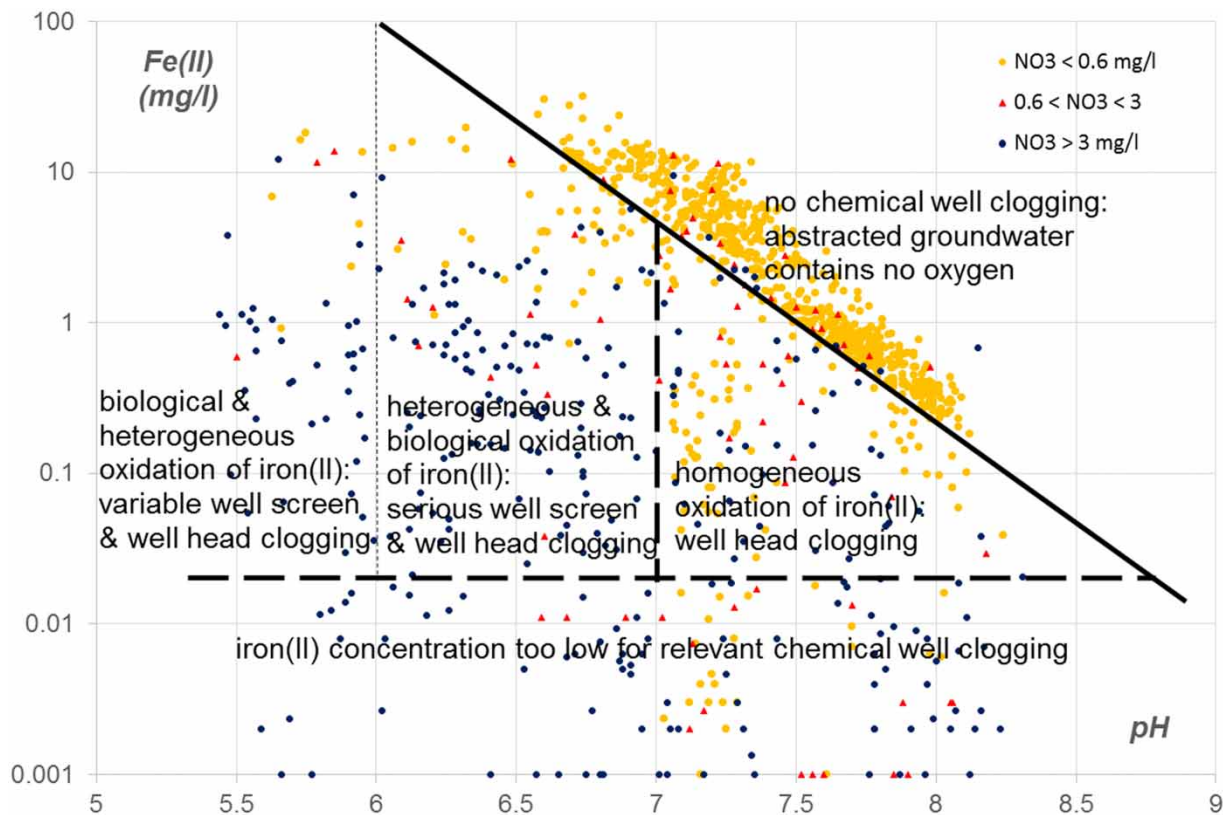
This distribution of iron(II) oxidation processes as a function of pH is in line with the experimentally observed distribution in the removal of iron(II) from groundwater by aeration and RSF, see Figure 3: homogeneous oxidation of iron(II) if pH >7.75, and heterogeneous and biological oxidation of iron(II) if pH <7.75. The same distribution appears present in the occurrence of chemical well clogging, see Figure 7: if pH >7: homogeneous oxidation of iron(II) resulting mainly in well-head clogging (van Beek 2018), and if pH <7: heterogeneous and biological oxidation, resulting mainly in well-screen clogging, where the contributions of heterogeneous and of biological oxidation will cross each other as function of pH. Besides pH and concentrations of oxygen and of iron(II) also other parameters are relevant with respect to this distribution: growth conditions for *Gallionella* spp. (adequate supply of nutrients), well operation (pump capacity,

operation hours/year), well maintenance (well rehabilitation: method and frequency), etc.

If the abstracted groundwater besides iron(II) contains no oxygen, chemical well clogging will not occur; and if the abstracted groundwater contains oxygen but the concentration of iron(II) is below ca. 0.02 mg/l, there will be some, but hardly relevant well clogging.

## CONCLUSIONS

In the removal of iron(II) in the production of drinking water from groundwater by aeration and RSF and in the occurrence of chemical well clogging by accumulation of HFOs and biomass, identical iron(II) oxidation processes are active: homogeneous, heterogeneous and biological oxidation, but under very different conditions. Which oxidation process dominates is determined by chemical (pH, O<sub>2</sub> concentration), physical (specific surface area) and operational



**Figure 7** | Preliminary classification for the occurrence of chemical well clogging by accumulation of HFO precipitates and biomass as function of pH, iron(II) concentration, and presence of nitrate (oxygen, Supplementary material, section A) in groundwater abstracted by 1186 wells operated by water utility Vitens.

(flow velocity, continuous vs. intermittent operation) conditions, and may be, within limits, manipulated. Obviously, there are more options for process-optimization in drinking water treatment than in well clogging: flow direction, method of back-washing, volume of 'supernatant water', etc. Under current conditions the boundary pH between on the one hand homogeneous oxidation, and on the other hand heterogeneous and biological oxidation of iron(II) equals for drinking water treatment  $\text{pH} \approx 7.75$  and for chemical well clogging  $\text{pH} \approx 7$ . The oxidation/precipitation process also governs the characteristics and properties of the HFO precipitates and the contribution of biomass, and consequently of the drinking water sludge and of the clogging material (HFO particle size, HFO floc size, ratio inorganic/organic material).

Many of the phenomena described, as bed-height-growth, differences in back washing of sand-filters, passage of HFO particles, presence of *Gallionella* spp. in sand-filters and in clogging wells, etc., are well known. Making these phenomena quantitative, as a function of pH and also of concentration of oxygen, will result in a better understanding and thus in a more efficient operation.

## ACKNOWLEDGEMENTS

Dirk Vries (KWR Water Research) and Koen Huysman (PIDPA) are acknowledged for commenting on an earlier draft of this paper. Two anonymous reviewers suggested considerable improvements.

## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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