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# Iron and manganese removal: Recent advances in modelling treatment efficiency by rapid sand filtration



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D. Vries <sup>a, \*</sup>, C. Bertelkamp <sup>a</sup>, F. Schoonenberg Kegel <sup>b</sup>, B. Hofs <sup>c</sup>, J. Dusseldorp <sup>d</sup>, J.H. Bruins <sup>e</sup>, W. de Vet <sup>f</sup>, B. van den Akker <sup>a</sup>

<sup>a</sup> KWR Watercycle Research Institute, P.O. Box 1072, 3430 BB Nieuwegein, The Netherlands

<sup>b</sup> Vitens N.V., P.O. Box 1205, 8001 BE Zwolle, The Netherlands

<sup>c</sup> Evides Waterbedrijf, P.O. Box 4472, 3006 AL Rotterdam, The Netherlands

<sup>d</sup> Oasen Drinking Water N.V., P.O. Box 122, 2800 AC Gouda, The Netherlands

<sup>e</sup> Waterlaboratorium Noord, Rijksstraatweg 85, 9756 AD Glimmen, The Netherlands

<sup>f</sup> Water Supply Company Limburg (WML), Limburglaan 25, 6229 GA Maastricht, The Netherlands

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## ABSTRACT

A model has been developed that takes into account the main characteristics of (submerged) rapid filtration: the water quality parameters of the influent water, notably pH, iron(II) and manganese(II) concentrations, homogeneous oxidation in the supernatant layer, surface sorption and heterogeneous oxidation kinetics in the filter, and filter media adsorption characteristics. Simplifying assumptions are made to enable validation in practice, while maintaining the main mechanisms involved in iron(II) and manganese(II) removal. Adsorption isotherm data collected from different Dutch treatment sites show that Fe(II)/Mn(II) adsorption may vary substantially between them, but generally increases with higher pH. The model is sensitive to (experimentally) determined adsorption parameters and the heterogeneous oxidation rate. Model results coincide with experimental values when the heterogeneous rate constants are calibrated.

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## 1. Introduction

The main (60%) source for drinking water production in The Netherlands is groundwater. Abstracted groundwater is generally anaerobic and thus requires aeration followed by rapid sand filters (RSFs) to remove iron(II) and manganese (II). Iron and manganese need to be removed from the water, since their presence in water can result in: (1) the sedimentation of iron/manganese precipitates in distribution pipelines which can cause clogging potentially leading to increased energy loss, (2) iron/manganese precipitates in the distribution pipelines can re-suspend during temporary higher discharges leading to discoloured water at the customer tap, (3) metallic taste and odour and (4) staining of laundry and household fixtures (Carlson et al., 1997; Buamah et al., 2009a; Jez-Walkowiak et al., 2015).

According to the Directive 98/83/EC and the Dutch Directive

\* Corresponding author. E-mail address: dirk.vries@kwrwater.nl (D. Vries). (Drinkwaterbesluit) the maximum concentration levels in drinking water should not exceed 0.20 mg/L for iron and 0.050 mg/L for manganese (Council, 1998; Drinkwaterbesluit, 2011). For instance, Vitens, the largest drinking water company of the Netherlands, aims at keeping iron and manganese levels lower than 0.05 and 0.01 mg/L respectively. A common treatment process applied in The Netherlands to remove iron and manganese from groundwater is aeration followed by rapid sand filtration. Iron and manganese can be removed by three processes occurring in the sand filter: homogenous oxidation, heterogeneous oxidation and biological oxidation.

Homogeneous oxidation is the oxidation of Fe(II)/Mn(II) to Fe(III)/Mn(II)/Mn(IV) ions and the subsequent hydrolysis which results in flocs that precipitate on and between the sand grains (Sung and Morgan, 1980; Tamura et al., 1980; Beek et al., 2012). Homogeneous oxidation is pre-dominant in the zone where an oxidant is present and no adsorbent material, like (coated) sand grains, is present. In water treatment practice, homogeneous oxidation occurs pre-dominantly in the supernatant layer of the pre-aerated submerged RSF, but also in the water phase of the filter

bed. In contrast to homogeneous oxidation of iron, homogeneous oxidation of manganese in an RSF is negligible (Diem and Stumm, 1984). Heterogeneous oxidation describes the process in which Fe(II)/Mn(II) sorbs to a surface area (e.g. hydrous ferric oxides) where it can subsequently be oxidised (Tamura et al., 1980; Park and Dempsey, 2005; Beek et al., 2012), resulting in the formation of new sorption sites. This autocatalytic process (Sung and Morgan, 1980) takes place mainly at the surface of the filter medium (Sharma, 2001) of submerged sand filters and trickling filters (a trickling filter is continuously emptied via a free outflow and has no supernatant layer present). The third mechanism of iron and manganese removal in sand filters is the biological oxidation of these compounds by iron and manganese oxidising bacteria (IOB and MOB). A number of IOB have been reported in literature to be involved in the biological oxidation of iron, such as: Leptothrix ochracea, Gallionella ferruginea, Toxothrix trichogenes, Thiobacillus ferrooxidans and Crenothrix (Michalakos et al., 1997; Kirby et al., 1999; Sharma et al., 2005; Rentz et al., 2007). Similarly, several bacterial strains involved in the oxidation of manganese are found on filter grains and in the water phase and have been reported, like Pseudomonas sp., Streptomyces sp., and Leptothrix sp. (Bruins, 2016). Monod expressions to model growth rates of bacteria is the most common assumption to deal with biological activity (Liu et al., 2001; Aa et al., 2002), but as far as the authors are aware, there are few studies that have focused on mathematically describing removal of iron and manganese by biologically driven processes.

It is said that iron and manganese removal in aeration and rapid sand filtration have been operated by drinking water treatment companies based mostly on experience and rules of thumb (Mouchet, 1992; Sommerfeld, 1999). Despite the wide-spread application of RSF for groundwater treatment in Western Europe, US and Canada, specific insights in the fundamental mechanisms (homogeneous, heterogeneous and biological oxidation) underlying the removal of iron and manganese in this process step as a function of varying process conditions and influent water composition is still missing. Modelling these mechanisms can provide a mean to investigate assumptions regarding mechanisms, and compare model simulation results with measurements from groundwater treatment practices. Moreover, process optimization and advanced control of filter operation can be supported (validated) by model simulations. For example, high supernatant levels, intensive aeration and high pH will result in a relatively high rate of homogeneous formation of iron (hydroxy-)oxides, eventually increasing the filter bed resistance. To prevent clogging of the filter, more frequent backwashing will be needed and this will result in increased use of backwash water. Heterogeneous oxidation leads to minimal head loss, which indicates that heterogeneous oxidation is generally preferred over homogenous oxidation, especially where the heterogeneous oxidation rate becomes favourable when compared to the homogeneous or biological oxidation rate -e.g. in the case of anoxic groundwater with a slightly alkaline pH (Sharma, 2001) and medium to high iron concentrations (Beek et al., 2012). Due to continuous deposition of iron(III) (hydroxy-)oxides by heterogeneous oxidation, the size of the coated filter grains will increase in time. The increase of filter grain size leads to a decrease of the specific surface area and an increase of the filter bed, which is also experienced in practice (Beek et al., 2012). However, it is believed that microbiology can play a key role in the oxidation of iron (II), and it seems that depending on the exact circumstances, one of the three mechanisms is dominant (Beek et al., 2015).

Depending on the type of filtration used (trickling filters or submerged filters) and the contact time distribution with oxygen, the ratio of homogeneous versus heterogeneous oxidation can be different. Besides the concentration of manganese and iron, the type of filtration is largely dependent on the concentration of ammonium and/or methane (Vet et al., 2011). Another important aspect of removal efficiency is that many water quality parameters (e.g. pH, alkalinity, temperature, organic matter,  $Cu^{2+}$ ,  $Co^{2+}$ ,  $H_2PO_4^-$ ) can affect iron and manganese removal (Stumm and Lee, 1961; Graveland, 1971; Theis and Singer, 1974; Sharma, 2001). Since the water quality can differ from source to source, it is difficult to predict iron and manganese removal efficiency. Likewise, the number of processes involved are characterised by different timescales (including distribution time affecting the amount of oxidation) and can influence one another.

A model describing iron and manganese removal mechanisms could potentially be used as an assessment tool for finding operational conditions or even be part of RSF process monitoring and control with the aim to maintain a desired level of filtrate quality while keeping the operational costs at a minimum. Most studies, e.g. (Ives, 1970; Schwager and Boller, 1997; Jegatheesan and Vigneswaran, 2005), with respect to iron and manganese removal in RSF treatment focus on physical and hydraulic aspects rather than changes of water quality due to physico-chemical and biological reactions. Çakmakci et al. (2010) developed an artificial intelligence model for iron removal in sand filtration, based on a laboratory-scale experiment. However, such a model is difficult to interpret since it does not explicitly relate to (known) chemical, physico-chemical and biological reactions and mechanisms. A model that incorporates both hydraulic resistance, particle removal and chemical removal reactions, i.e. oxidation and adsorption, is described by (Teunissen, 2007). However, in this model changes in water quality due to chemical speciation (with e.g. carbonate ions) is not incorporated and adsorption isotherm values have not been determined from field data. In addition, equilibria reactions with iron and manganese hydroxides were not mass-balanced. The idea to use adsorption isotherms in heterogeneous oxidation has been adopted here.

To overcome the aforementioned limitations, the main objective of this study is to develop a knowledge based model for iron/ manganese removal in RSFs to test and validate the assumed removal mechanisms by oxidation. Specifically, it is aimed to assess the following aspects: (1) the effect of pH and the height of the supernatant influent water layer on iron/manganese removal, (2) to apply Freundlich adsorption isotherm parameter values from batch-experiments for iron and manganese adsorption and compare these with the values reported in literature, (3) to validate model simulation results with field data, calibrate and assess critical model parameters. Note that the inclusion of particle removal and hydraulic resistance, as has been done in the work of Teunissen and co-workers (Teunissen, 2007) is not yet included in the modelling work. Pursuing these aims, the model needs to have enough degrees of freedom to cover different operational conditions but at the same time, not be over-parameterised in order to keep model validation feasible and keep the variance of model parameters at an acceptable level. Furthermore, modelling focus is set on submerged rapid sand filtration (i.e. there is supernatant layer on top of the filter medium) and treatment of groundwater with small amounts of methane and organic material. Because of lack of sufficient experimental data, biological iron and manganese oxidation is excluded from this modelling study.

#### 2. Materials and methods

Assumptions have been made to model iron, manganese and nitrification. Material and methods relating to modelling assumptions are described in more detail in the section 'Model description'. For validation purposes, data is collected from literature and additional samples are taken from the influent water, filter media, and water collected at different bed heights at five Dutch water Download English Version:

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