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# Water quality and treatment of river bank filtrate

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#### Abstract

In drinking water production, river bank filtration has the advantages of dampening peak concentrations of many dissolved components, substantially removing many micropollutants and removing, virtually completely, the pathogens and suspended solids.

<sup>5</sup> The production aquifer is not only fed by the river bank infiltrate but also by water percolating through covering layers. In the polder areas, these top layers consist of peat and deposits from river sediments and sea intrusions.

This paper discusses the origin and fate of macro pollutants in river bank filtrate, based on extensive full-scale measurements in well fields and treatment systems of

- the Drinking Water Company Oasen in the Netherlands. First, it clarifies and illustrates redox reactions and the mixing of river bank filtrate and polder water as the dominant processes determining the raw water quality for drinking water production. Next, full-scale results are elaborated on to evaluate trickling filtration as an efficient and proven one-step process to remove methane, iron, ammonium and manganese. The
- interaction of methane and manganese removal with nitrification in these systems is further analyzed. Methane is mostly stripped during trickling filtration and its removal hardly interferes with nitrification. Under specific conditions, microbial manganese removal may play a dominant role.
- 20 Abbreviations and notations:
  - PW = Polder water
  - RBF = River bank filtration
  - WTP = Water treatment plant

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#### 1 Introduction

Although the role of surface water as a source for drinking water is gradually increasing, in the Netherlands still over 60% is produced from groundwater (VEWIN, 2008). Groundwater generally has the advantage of a good hygienic and consistent quality

<sup>5</sup> compared to surface water. Groundwater abstraction may, however, be restricted especially in areas with desiccation or sea water intrusion. In these cases natural or artificial recharge of the groundwater with surface water may provide a solution, like with dune infiltration and river bank filtration (RBF). Depending on the hydrological situation, the river may infiltrate or drain the surrounding land. In all cases, abstraction of river bank filtrate enhances the infiltration of river water compared to the natural situation.

Although water from a properly designed RBF plant has the general advantages of groundwater, there are some drawbacks. First of all, persistent micropollutants present in the river water will eventually reach the groundwater wells, although high concentra-

- tions are effectively reduced by adsorption, biological breakdown and residence time variation (Sontheimer, 1991). These remaining substances will have to be removed by specific techniques in the treatment plant. In the Netherlands, all treatment plants for river bank filtrate include advanced oxidation, adsorption and/or membrane filtration steps, which will not be discussed in this paper.
- Infiltration implies aquifer passage, resulting in water quality changes. In most cases, the raw groundwater consists of a mix of river bank filtrate and locally infiltrated polder water (PW). This article describes the origin of the concentrations of methane, ammonium, iron, manganese, phosphate and sulfate in the raw water at the Oasen Drinking Water Company in the Netherlands (Oasen). Differences in concentration between in-
- <sup>25</sup> dividual wells can be explained by the ratio of PW compared to river bank filtrate and by redox processes during transport. Changes in river water composition over the last 50 years also play a role.

Dutch standards for sulfate are so high that removal is not necessary, but results are

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presented in the hydrology section of this article because it is a good indicator for the changes in oxidation reduction potential (ORP) during aquifer passage. The hardness of the water greatly increases during RBF. Calcium and bicarbonate are not removed by conventional filtration techniques but are reduced efficiently by supplemental tech-<sup>5</sup> niques such as pellet softening (van Dijk and Wilms, 1991). That, however, will not be discussed in this article.

In the section about treatment, this article focuses on the applied trickling filtration, also known as dry (bio)filtration, as an effective combined treatment step for methane, iron, ammonium and manganese. As part of the joint Oasen-TUDelft PhD-project "Ni-trification in trickling filters for drinking water production", this article further elaborates on possible interactions between the removal processes in a filter. This article focuses further on methane and manganese removal mechanisms and their possible adverse

#### 2 Bank filtration in Oasen Polders, The Netherlands

#### **2.1** Oasen groundwater quality

effects on nitrification.

The Water Treatment Plant (WTP) Reijerwaard uses river bank filtrate as a source for drinking water production. The groundwater is abstracted with vertical wells in a semi-confined aquifer of unconsolidated sandy sediments along the river Nieuwe Maas, which is a branch of the river Rhine. A 10 m thick Holocene layer of clay and peat sediment lies over the aquifer. The area is a polder below river water level, with a dense network of ditches.

#### 2.1.1 Hydrology

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The abstracted groundwater consists of a mixture of two anoxic water types with different origins (Fig. 1), compositions and redox states:



- River bank filtrate. Rhine water infiltrating through the river bottom and flowing horizontally through Pleistocene sand deposits to the wells. Subsurface residence times range from 3 to 50 years. This water type accounts for approximately 70% of the abstracted water.
- PW. Water abstracted from shallow groundwater and ditches through the clay and peat layer. Residence times range from a year to several decades. This water type accounts for approximately 30% of the total.

As shown in Fig. 1, the hydrologic setting of RBF well fields is such that the wells near the river abstract mainly river bank filtrate, while the share of PW increases with distance from the river. This increase is caused by the longer flow paths for the filtrate – as a function of distance, but even more as a function of time – combined with the increasing drawdown in hydraulic head by the abstraction, causing a larger aquifer recharge with PW. The mixing of river bank filtrate and PW occurs mainly within the wells; in an aquifer, mixing is generally limited.

#### 15 2.1.2 Redox processes

Redox processes are the crucial initiator of changes in composition for both the river bank filtrate and the PW. In general, it can be stated that when organic matter is sufficiently available, it is oxidized by a sequence of redox processes (from Stuyfzand, 1988):

- 20 aerobic respiration
  - denitrification
  - reduction of manganese and iron
  - reduction of sulfate
  - methanogenesis



Methanogenesis will generally only occur after all sulfates have been reduced, so methane will only be present simultaneously with sulfate as a result of mixing (Stuyfzand, 1994).

Measured concentrations in individual production wells are presented in Fig. 2 for the Reijerwaard well field, together with their distance to the river. The concentrations show signs of a lowering redox level with increasing distance from the river, which is a logical consequence of the increasing share of PW with deep redox conditions.

#### 2.1.3 Redox parameters sulfate and methane

In the river water, the annual averages of sulfate concentration have varied between 50 and 80 mg L<sup>-1</sup> over the last 30 years. The river bank filtrate reaches the redox level of sulfate reduction while passing the deposits on the river bottom and remnants of the Holocene layers that the river intersects, resulting in sulfate concentrations of one half that of the river water. The PW passes the Holocene clay and peat layer and reaches an even lower redox level, with methanogenesis. Since methane and sulfate do not occur simultaneously, the methane and sulfate concentrations were used as indicators for estimating the water fractions derived from both origins. PW is identified by the presence of methane and river bank filtrate by the presence of sulfate. Since the ratio of PW to river bank filtrate determines the methane and sulfate concentrations of

individual wells, the concentrations show a negative correlation (Fig. 3). Corresponding
to the hydrology, wells near the river abstract mostly river bank filtrate, so with low methane and relatively high sulfate concentrations (e.g. wells RK-P12, 17, 34 and 35). In the same way, wells with high methane concentrations and low sulfate concentrations are located in the back of the well field and abstract a larger share of PW. The sulfate concentrations are, however, even lower than might be expected based on the share of river bank filtrate because of the long residence times for it to reach the back of the well field. The old Rhine water had lower sulfate concentrations and, during transport, sufficient time was available for sulfate to be reduced by the small amounts



the aquifer needs long travel times; reduction is limited when travel times are less than 10 to 20 years. This is shown by the wells in the center of the well field (RK-P07 to 09) where sulfate concentrations remain at the level that is expected from the mixing ratio of sulfate-free PW to river bank filtrate and show no indication of a decrease caused by reduction processes during the aquifer transport.

### 2.1.4 Origin of the macro components iron, ammonium, phosphate and manganese

#### Origin of iron and ammonium

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For ammonium, an obvious source is the oxidation of nitrogenous (N-containing) organic matter in the confining clay and peat layer, in the river bottom, and to a lesser 10 extent in the aquifer itself. In this process, iron and manganese oxides act as oxidizing agents and dissolve. In river bank filtrate, the iron concentrations are limited because of precipitation as iron sulfides with HS<sup>-</sup> resulting from subsequent sulfate reduction (Stuyfzand, 1985). This iron sulfide precipitation leaves only very low hydrogen sulfide concentrations in the groundwater. Although mostly below the detection limit, the 15 highest measured concentration of hydrogen sulfides in raw groundwater of another, yet comparable Oasen WTP was  $0.03 \text{ mg L}^{-1}$ . In the PW, all sulfate has been reduced - even methanogenesis occurs - and iron is introduced from the dissolution of minerals like siderite (FeCO<sub>3</sub>) and vivianite (Fe<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub>·8(H<sub>2</sub>O)) that are common in areas with peat. A further increase in iron concentrations in the aquifer can occur due to the 20 reductive dissolution of iron oxides. The positive relationship between the redox indicator, methane, and iron is illustrated in Fig. 3. This figure also shows a strong positive correlation between iron and ammonium, indicating that a relevant contributing process may be the release of adsorbed or organically bound ammonium during the reductive

<sup>25</sup> dissolution of iron oxides.

Compared to river water, ammonium concentrations in river bank filtrate and PW are very high. In the river Rhine, ammonium was present at yearly averaged concentrations



over  $0.5 \text{ mg L}^{-1} \text{ N}$  before 1990, which has decreased to less than  $0.1 \text{ mg L}^{-1} \text{ N}$  in the last decade. In the river water, the ammonium concentration is generally five times higher in winter than in summer; in river bank filtrate, however, no seasonal variation has been observed due to attenuation.

#### **5** Origin of manganese

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Manganese concentrations are known to rise to a high level during infiltration from the river. At WTP Reijerwaard, and in the nearby Opperduit test location, studied by Stuyfzand (1996), concentrations are found of 1.4 and  $1.5 \text{ mg L}^{-1}$ , in observation wells at several tens of meters after infiltration into the aquifer. Stuyfzand (1985) assumed that manganese mainly originates from the reductive dissolution of manganese oxides and observed that the river bank filtrate is in equilibrium with MnCO<sub>3</sub> precipitate which may be a source, but also a limit, for the manganese concentration.

The high concentration of manganese in the river bank filtrate decreases with travelling time and distance after infiltration. This decrease is visible in the raw water <sup>15</sup> concentrations of the first five wells shown in Fig. 2 and is also found in a row of observation wells perpendicular to the river at Opperduit (Stuyfzand, 1996). At the far half of the well field, away from the river, the manganese concentration in the river bank filtrate has decreased to a low level, and PW contributes predominantly to the concentrations that are found in the wells. Here, the highest concentrations are found in the wells with <sup>20</sup> the highest share of PW.

#### **Origin of Orthophosphate**

Orthophosphate originates from the dissimilation of organic matter and the dissolution of phosphate minerals, with vivianite as the most important one. From Fig. 2 it can be concluded that both the river bank filtrate and PW contain high concentrations. In the river bank filtrate, however, a decrease in the orthophosphate concentration is observed with increasing distance from the river, probably as a consequence of



adsorption on iron (oxy)hydroxides present in the aquifer.

#### 2.1.5 Geochemistry of aquifer sediments

The availability of iron and manganese oxides is, of course, a condition for their reductive dissolution. Samples of the aquifer sediment on a nearby well field (WTP Lekkerk-

- <sup>5</sup> erk) confirm their presence in large quantities. Analysis showed 1000 to 3000 mg kg<sup>-1</sup> by dry weight of amorphous iron (oxy)hydroxides in the sediment (extractable with oxalic acid), whereas a similar amount was found in the form of ferrous iron precipitates as iron sulfides, like pyrite, and in the form of crystalline iron oxides (extractable with aqua regia). For manganese, the samples showed concentrations of 30 to 60 mg kg<sup>-1</sup> of
- dry weight of easily soluble manganese oxides, manganese carbonate (rhodochrosite) and reduced species mainly adsorbed to iron (oxy)hydroxides. A similar amount of manganese was found in more persistent manganese containing minerals. These iron and manganese precipitates might be source or sink for dissolved iron and manganese species.

#### 15 2.2 Oasen groundwater treatment

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The complex mixture of river bank filtrate and PW in the groundwater poses a number of challenges in the treatment process. Before discussing some of these issues based on long-term observations in existing, full-scale filters systems, the next paragraph provides an overview of existing treatment processes for the removal of the relevant macro components.

#### 2.2.1 Removal processes for macro components

The presence of the macro components methane, iron, ammonium and manganese is unwanted in the drinking water for several reasons and, therefore, is severely restricted by drinking water standards. The compounds can be removed from the groundwater by a wide array of physical, chemical and microbiological processes, or combinations



of them. All four compounds can be removed by oxidation with oxygen or a chemical oxidizing agent like ozone, permanganate or peroxide. In all cases, the oxidation can be biologically catalyzed, or indeed sometimes is strictly biological, like for methane, ammonium and manganese under neutral conditions with oxygen as the only oxidizing

agent. In the bacterial processes, the (catabolic) oxidation reaction is used for energy generation and growth of the bacteria. Non-oxidative removal processes range from membrane filtration through flocculation, sedimentation and filtration to adsorption or gas stripping. The membrane filtration techniques of reverse osmosis and nanofiltration, although rather effective for removal of iron and manganese, are generally not applied for the removal of only inorganic macro components. An overview of applicable removal processes is given in Table 1.

#### 2.2.2 Oasen groundwater treatment systems

In groundwater treatment in the Netherlands, only oxygen from the air and no chemical agents are used for oxidation purposes. The central process for treatment of river bank filtrate/PW groundwater is filtration over a granular medium, in most cases single layer silica sand, but other bed configurations (dual layer) and materials (e.g. anthracite, expanded clay pellets) are increasingly used. Because of the oxygen demand of the macro component removal processes, the anoxic groundwater requires aeration before or during filtration. In some cases a simple spraying directly on top of the filter bed may suffice, but in most cases there is a specialized gas exchange process step

- before filtration. Such a gas exchange step will also strip oversaturated gases like methane and carbon dioxide from the groundwater. Removal of the latter increases the pH of the water, which in turn may stimulate the oxidation of iron, ammonium and manganese, the rates of which are proportionate to OH<sup>-</sup> concentrations in circumneu-
- tral circumstances. The scheme of typical Dutch groundwater treatment is shown in Fig. 4.

Due to the high oxygen consumption of ammonia-oxidation (4.6 mg  $O_2$  per mg  $NH_4$ , see Table 1), trickling filtration (also called dry biofiltration) is applied in cases of high



ammonium content in the groundwater, like at several Oasen WTPs. As an alternative, submerged (also called wet) filtration with the injection of pure oxygen might be considered in these situations.

Of the four groundwater components considered, only ammonium and manganese removal pose regular problems for Oasen. The nitrification problems have previously been described by de Vet et al. (2009b). Iron removal in itself is never problematic for Oasen but some of its possible interactions with nitrification were discussed in that earlier paper. The role of microbial iron oxidation, although possibly of importance in the Oasen trickling filters, will not be discussed here. For methane and manganese, different removal mechanisms and their interactions with nitrification are discussed in the next two sections.

#### 2.2.3 Methane removal and its effect on nitrification

Methane removal can be achieved physically by stripping, or biologically by methaneoxidizing bacteria. Biological degradation is characterized by a relatively high biomass

- <sup>15</sup> over substrate yield (19 to 70% of the substrate carbon is incorporated into cell material; Leak and Dalton, 1986), and may lead to the clogging of filter material by production of extracellular polymeric substances (Streese and Stegmann, 2003). Therefore, because biological methane oxidation may interfere with other filtration processes, it is generally not chosen.
- <sup>20</sup> Several systems for intensive gas transfer, such as cascades, tower and plate aerators and high pressure spraying, are effectively applied in full-scale plants for this purpose. A vacuum stripper is applied in situations were aeration is unwanted, as is the case in front of trickling filtration, to avoid clogging the distribution spraying with oxidation products. The stripping efficiency for dissolved gases is determined by gas
- <sup>25</sup> properties, especially the water/air distribution (or Henry) coefficient, and system characteristics for the equilibrium state (determined by the air-to-water ratio, RQ), and kinetics (described by the transfer coefficient). The distribution coefficients in Table 2 show methane's lower affinity for water compared to that of carbon dioxide, implying





a better removal of the former under the same system characteristics. Oxygen and methane are comparable in this respect.

The difference in removal efficiencies for methane and carbon dioxide is illustrated in Table 3 for the full-scale gas transfer systems at two Oasen WTPs. The vacuum stripper had distinctly lower gas removal efficiency for carbon dioxide than the cascade did. For methane, the difference was less pronounced, and both systems achieved over 90% efficiency.

Methane will be completely removed by trickling filtration. Although methane is not measured above the detection limit in the trickling filter effluent, no direct assessment can be made of the role of physical and biological methane removal. However, from the removal efficiency for carbon dioxide, it can be concluded that the air stripping of the trickling filters performed much better than the vacuum stripper at Oasen WTP Lekkerkerk (Fig. 5). The trickling filter contained coarse filter sand (1.7–2.5 mm), was concurrently ventilated with an RQ of 10 and had an average filtration rate of 2.2 m h<sup>-1</sup>.

- <sup>15</sup> The removal efficiency for carbon dioxide was over 75% for the trickling filter, which is comparable to the removal efficiency of cascade aeration (see Table 3). Before the startup of the acidifying nitrification and manganese removal, the carbon dioxide stripping efficiency in the trickling filter was even higher than 90%. The supposed predominance of physical over biological methane removal was confirmed by two more
- direct methods. First, when balancing the fluxes of methane entering (by water) and leaving (by water and air) the filter, the average and standard deviation for the physical removal efficiency in six trickling filters at several Oasen WTPs was 84%±12%. Differences before and after backwash were found to be within the boundaries of uncertainty. Secondly, the methane-oxidizing activity measured in batch experiments varied only
- <sup>25</sup> slightly between sand samples taken from trickling filters at Oasen WTP Lekkerkerk with and without pretreatment by vacuum stripping.

Figure 6 shows that pretreatment of the groundwater by vacuum stripping had no clear effect on the nitrification in the trickling filters.

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#### 2.2.4 Manganese removal and its effect on nitrification

Without strong oxidizing chemicals, manganese oxidation under circumneutral circumstances may occur chemically (Graveland and Heertjes, 1975) and biologically (Czekalla et al., 1985).

- A comparison of full-scale trickling filters at Oasen WTP Lekkerkerk shows some interesting differences in manganese removal and its occurrence in combination with nitrification problems. At this WTP, groundwater from two separated well fields (Schuwacht and Tiendweg) is also treated with separated double trickling filters. In the Schuwacht well field, subsurface aeration (Appelo et al., 1999) a very mild form of in situ iron removal is applied for the enhancement of nitrification. Figure 7 shows that the enhancement of subsurface aeration worked for manganese removal as it did
- that the enhancement of subsurface aeration worked for manganese removal as it did for nitrification and the interruption of the technique resulted in a similar relapse for both processes.

Incomplete nitrification, however, does not always coincide with incomplete man-<sup>15</sup> ganese removal. Figure 8 shows that, despite incomplete nitrification, manganese removal was not severely reduced in another full-scale trickling filter, which treated normal, non-subsurface aerated groundwater from the Tiendweg well field.

Remarkable differences were also observed in the second trickling filter of the double filter sets during startup with fresh filter sand only in the first trickling filter (Fig. 9). Each

- first filter was directly coupled to its own second filter, making the effluent of the former the influent of the latter. From the startup with new filter material in the first filter, the manganese removal was almost complete in the second filter of the non-subsurface aerated double filter set. In the subsurface aerated double filter set, both the first and second filter required a startup period of over a month for manganese removal.
- <sup>25</sup> The backwash water from a non-subsurface aerated first trickling filter contained more biological formed iron(oxi)hydroxide deposits, resembling those of the *Leptathrix ochracea* (Czekella et al., 1985), iron- and possibly manganese-oxidizing bacteria (Fig. 10).





#### 3 Discussion

#### 3.1 Origin of macro components

When abstracting river bank filtrate, it is nearly inevitable that PW is also attracted and abstracted. The abstraction, therefore, results in a raw water mix of both water <sup>5</sup> types, of which the origins are illustrated in Fig. 11, together with their macro pollutant contributions of interest. The figure shows that PW accounts for the methane and high loads of iron and ammonium, which are relevant for the filtration steps. River bank filtrate and PW contribute an approximately equal load of manganese to the total raw water.

#### 10 3.2 Influencing the ratio of PW to river bank filtrate

River bank filtrate offers a better source for producing drinking water than PW, considering ammonium and iron concentrations. So, with the operation and the design of well fields, it is favorable to maximize the share of river bank filtrate. The percentage of river bank filtrate can be influenced by changing the location of a well field relative to the <sup>15</sup> river or by choosing a location with favorable hydrological conditions. For example, a location close to the river in a polder far below river level will result in a relatively high share (%) of river bank filtrate. But moving well fields is generally not realistic, and other factors will be more important than these aspects of water quality.

For existing well fields, it is a technically challenging idea to separately abstract (and treat) PW and river bank filtrate. Considering the distribution of the PW and river bank filtrate both in the horizontal plane and in depth, as shown in Fig. 11, a technical solution would be to equip each well with two separate screens and pumps. Well screen depth and pump capacity are designed so that the deep screen abstracts only river bank filtrate, while the shallower screen abstracts only PW.

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#### 3.3 Pretreatment or direct trickling filtration for methane stripping

Full-scale trickling filters at Oasen are versatile removal systems, in optimal conditions capable of removing over 1 g of methane, 10 g of iron, 8 g of ammonium and 1 g of manganese per hour and m<sup>3</sup> of filter bed almost completely. Physical, chemical and biological processes occur simultaneously. Methane is predominantly removed by 5 stripping and the remaining microbial methane-oxidizing activity in Oasen trickling filters was, at most, about 0.7 g methane per hour and m<sup>3</sup> of filter bed only in the upper layer of the filter bed. This is moderate compared to the maximum methane-oxidizing activity measured at a landfill gas treatment filter (63 g methane per hour and m<sup>3</sup> of filter bed; Streese and Stegmann, 2003). In the Oasen trickling filters, ammonia-oxidizing 10 microorganisms may even account for part of the methane-oxidation. Pretreatment by vacuum stripping for methane removal had no effect on the nitrification in the trickling filters. Thus, in well-ventilated trickling filters, biological methane oxidation is limited and does not compete with nitrification. Proper assessment of the physical methane removal capabilities of the trickling filter is recommended to prevent building a costly, but ineffective, extra pretreatment step preceding trickling filtration.

#### 3.4 Manganese removal mechanism and problems

Like in iron oxidation (Sharma et al., 2005), the role of microorganisms in manganese oxidation is being debated. Although no conclusions about the steady state can be
 drawn, manganese-oxidizing bacteria have been shown to play an important role in the startup of the process (Burger et al., 2008). Observations of the full-scale filters at Oasen WTP Lekkerkerk suggest that the role of bacteria in manganese oxidation may be distinctly different under deviating conditions. Thermodynamically, ammonia and nitrite oxidation precede manganese oxidation and the inhibitory effects of incomplete
 nitrification on manganese removal have been reported by Vandenabeele et al. (1995).

With this in mind, we anticipated the results of the subsurface aerated first trickling filter, where incomplete manganese removal accompanied the nitrification problems.





The combination of severely inhibited nitrification without similar manganese removal problems in the non-subsurface aerated filter can not be explained is this manner.

The immediate and almost complete manganese removal in the second filter of the non-subsurface aerated filter set after startup in the first filter is remarkable as well.

- <sup>5</sup> According to Graveland and Heertjes (1975), the autocatalysis of manganese only occurs with unstable manganese oxides like Mn<sub>3</sub>O<sub>4</sub>. These transform over time into more oxidized and stable phases like MnO<sub>2</sub>, losing their catalytic capacity, requiring a constant regeneration of the deposition by freshly formed manganese oxides. Therefore, it should be expected that filter material in the second filters does not contain the unsta-
- ble, catalyzing manganese coating after more than half a year without a supply of manganese in the filter influent. Finally, the microscopic pictures from the backwash water show microbially formed deposits by iron- or manganese-oxidizing bacteria. From this we hypothesize that manganese (and iron) oxidation is predominantly bacteriological in Oasen trickling filters treating normal groundwater. We further hypothesize that sub-
- <sup>15</sup> surface aeration enhances the chemical removal of manganese (and iron), thus limiting the growth of these bacteria. Finally, we hypothesize that the growth of competing ironand manganese-oxidizing bacteria in trickling filters leads to the inhibiting of nitrifying microorganisms.

#### 4 Future research questions

- 20 What processes control manganese concentrations in groundwater, and can they be influenced underground?
  - How can PW and river bank filtrate be abstracted separately? Is separate abstraction of river bank filtrate and PW feasible? And how much mixing is inevitable?
  - What are optimal / fitting treatment schemes for both separated water types?
- Under what conditions does microbial iron and manganese oxidation occur?



- The raw water at the OASEN consists of a mixture of two water types, polder water and river bank filtrate, which have distinct differences in composition, related to their redox levels;
- Polder water has the lowest redox level and accounts for the larger part of the macro pollutants methane, ammonium and iron, while river bank filtrate contributes most to the manganese content of raw water;
- Trickling filtration can be a highly efficient and versatile removal system for macro pollutants in polder river bank filtrate including stripping system of methane;
- Manganese oxidation is probably predominantly microbial in trickling filtration of polder river bank filtrate;
- Coincidence of incomplete manganese removal and nitrification may indicate competition for phosphate or essential trace substrates in biological processes.

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#### - Does increased growth of iron- and manganese-oxidizing bacteria result in nutrient limitation for ammonia-oxidizing bacteria?

 Does subsurface aeration inhibit growth of iron- and manganese-oxidizing bacteria?

#### 5 5 Conclusions

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#### Table 1. Removal processes for groundwater macro components.

		Microbial	Physical-chemical	
Component	Reduction-oxidation reaction (with $O_2$ )	Microorganisms (Brock (Ed.), 2006)	Oxidation context	Alternative processes (de Moel et al. (Eds.), 2006)
Methane	$CH_4+2O_2 \rightarrow CO_2+2H_2O$	Methylotrophs like <i>Methylophilus</i> spp., <i>Methylomonas</i> spp., <i>Methylobacter</i> spp. (de Vet et al., 2009a)	-	Stripping
Iron	$Fe^{2+}$ + <sup>1</sup> / <sub>4</sub> O <sub>2</sub> + <sup>1</sup> / <sub>2</sub> H <sub>2</sub> O → FeOOH + 2H <sup>+</sup>	Gallionella ferruginea, Leptothrix ochracea., Sphaerotilus natans, Toxothrix Trichogenes (Czekalla et al., 1985; Emerson and Mover, 1997)	Adsorptive oxidation (Sharma et al., 2001)	(Reverse osmosis, nanofiltra- tion)
			Homogenous oxidation	Flocculation, settling, floc or flocking filtration
Ammonium	2 NH <sub>4</sub> <sup>+</sup> +3 O <sub>2</sub> $\rightarrow$ 2 NO <sub>2</sub> <sup>-</sup> +4 H <sup>+</sup> +2 H <sub>2</sub> O 2 NO <sub>2</sub> <sup>-</sup> +O <sub>2</sub> $\rightarrow$ 2 NO <sub>3</sub> <sup>-</sup>	Nitrosomonas spp., (Nitrosospira spp., Nitrosol- ubus spp.), Ammonia- oxidizing Archaea Nitrospira spp., (Nitrobacter spp.) (de Vet et al., 2009)	(Breakpoint chlorination) (not applied in the Nether- lands)	Stripping of ammonia (high pH), adsorption on zeolites (reverse osmosis)
Manganese	$\begin{array}{ccc} 2 & Mn^{2+} + 1 & O_2 + H_2O & \rightarrow \\ Mn_2O_3 + 2H^+ & \end{array}$	Manganese-oxidizing bac- teria like <i>Leptothrix</i> spp., <i>Metallogenium</i> spp., <i>Hy-</i> <i>phomicrobium</i> spp., <i>Side-</i> <i>rocapsa</i> spp., <i>Siderocystis</i> spp. (Czekalla et al., 1985)	Autocatalytic oxidation	(Reverse osmosis, nanofiltra- tion)
			Homogenous oxidation with strong oxidants like permanganate	Flocculation, settling, floc or flocking filtration

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**Table 2.** Distribution coefficients (in mass/mass) for methane and carbon dioxide (de Moel et al. (Eds.), 2006).

		MW	0°C	10°C	20°C
Methane	$CH_4$	16.0	0.0556	0.0433	0.0335
Oxygen	$O_2$	32.0	0.0493	0.0398	0.0337
Carbon dioxide	$CO_2$	44.0	1.71	1.23	0.942

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**Table 3.** Methane and carbon dioxide in- and outgoing concentrations and stripping efficiency for two Oasen full-scale gas exchange systems.

		WTP Reijer	waard Cascades*	WTP Lekke	erkerk Vacuum stripper**	
	Driving force AVG (MIN)	RQ=24 (5.4)		P=6 kPa		
	Hydraulic load AVG (MAX)	37 (1 CH.	65) m <sup>2</sup> m <sup>2</sup>	50 CH.	(90) m <sup>2</sup> h <sup>-1</sup> m <sup>-</sup>	
IN	$(mg L^{-1})$	7.2±1.7	103±36	0.93±0.31	32.6±4.4	
OUT	$(mg L^{-1})$	$0.28 \pm 0.08$	23.7±3.4	$0.08 \pm 0.03$	25.5±3.6	
Efficiency	%	95.7±1.7	72.3±9.0	90.8±2.6	21.9±5.0	

Efficiency =  $(C_{in} - C_{out})/C_{in}$ ; RQ = air to water ratio v/v;

\* Values for 1995-2004;

\*\* values for Jan–Nov 1998



**Fig. 1.** Layout of the Reijerwaard RBF site (51°52′ N, 4°35′ E, left) and cross-section of the abstraction of river bank filtrate and PW (right).

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		Methane (mg/l) RK-P28●23			Sulfate (mg/l) O0.067	Ammonium (mg/l NH4) •28	Iron (mg/l) ●16	Manganese (mg/l) ©0.64	Orthophosphate (mg/l) ●2.5
	1500	- RK-P26 <b>●18</b> RK-P33 <b>◎9.1</b>	Î	more PW (50 %) less RBF (50 %) RBF age > 40 years	- 00.083 00.25	- <b>0</b> 22 <b>0</b> 16	- •14 •10	- ©0.55 ©0.48	- <b>@</b> 2 ©1.6
ver (meters)		PK-D02 <b>0</b> 25	U		00.061	<b>•</b> 29	<b>4</b> 7	00.61	
distance from riv	1000	RK-P05 <b>019</b> - RK-P06 <b>013</b> RK-P07 <b>04.9</b> RK-P36 <b>00.98</b> RK-P09 <b>02</b> RK-P10 <b>0</b> 9			00.17 - 03.4 917 - 30 - 024 - 015	•23 •24 • •21 •16 •11 •10 •13	• 17 • • 14 • • 9.8 • 6.5 • • 5.3 • • 6.3	00.6 - 00.59 - 00.55 - 00.62 - 00.5 - 00.56	•2.3 •2.1 • 02.1 01.6 01.3 01.4 01.5
		RK-P12 <b>9.4</b> RK-P17 <b>2.9</b> RK-P34 <b>00.57</b>	Ĵ	more RBF (90 %) less PW (10 %)	•30 •37 •38	©12 ©8.9 ©6.1	©5.9 ○3 ○1.8	©0.75 ©0.84 ©0.7	<ul><li>○1.4</li><li>●1.8</li><li>●2.1</li></ul>
	500	RK-P35O0.53		RBF age < 10 years	●29	○5.7	01.9	●1.1 	•2.2

Fig. 2. Concentrations in raw water of individual production wells at Oasen WTP Reijerwaard; averages for 2005–2009.



**Fig. 3.** Water qualities of individual wells at the Oasen WTP Reijerwaard; relationship between sulfate and methane; methane and iron; ammonium and iron; averages for 2005–2009.

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Fig. 4. Artist impression of a conventional groundwater treatment system in the Netherlands with submerged sand filters.







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**Fig. 5.** Carbon dioxide concentrations and removal efficiencies for vacuum stripper and trickling filtration at Oasen WTP Lekkerkerk, period January–November 1998; open symbols, concentrations (left Y-axis):  $\Box$  raw water;  $\circ$  effluent vacuum stripper;  $\Delta$  effluent trickling filter; solid symbols, removal efficiencies (right Y-axis):  $\circ$  vacuum stripper;  $\blacktriangle$  trickling filter.



**Fig. 6.** Ammonium concentrations in the effluents of trickling filters at Oasen WTP Lekkerkerk; ▲ PLTOFF08 with pretreatment by vacuum stripping (October 1998 to April 2000), • PLTOFF05 without pretreatment by vacuum stripping (March 1999 to September 2000).

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**Fig. 7.** Ammonium (o, left Y-axis) and manganese (▲, right Y-axis) concentrations in the trickling filter effluent in periods with and without subsurface aeration in the Oasen WTP Lekkerkerk's Schuwacht well field.







**Fig. 8.** Ammonium (∘, left Y-axis) and manganese (▲, right Y-axis) concentrations in the trickling filter effluent treating normal groundwater from the Oasen WTP Lekkerkerk's Tiendweg well field.







**Fig. 9.** Manganese concentrations in the effluent of the first and second filter of double trickling filter sets at the Oasen WTP Lekkerkerk around the startup with fresh filter sand in the first trickling filter;  $\Delta$  and  $\diamondsuit$  effluent of first and second subsurface aerated filter,  $\blacktriangle$  and  $\bullet$  effluent of first and second non-subsurface aerated filter.







**Fig. 10.** Phase-contrast microscopy pictures of backwash water from first trickling filters at Oasen WTP Lekkerkerk; mainly inorganic flocs in backwash water from a subsurface aerated filter (left, 630X), mainly biological deposits from a non-subsurface aerated filter (right, 1000X).

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Fig. 11. Schematic presentation of the origin of discussed parameters.