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Long-term performance and fouling analysis of full-scale direct nanofiltration (NF) installations treating anoxic groundwater

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A B S T R A C T

Long-term performance and fouling behavior of four full-scale nanofiltration (NF) plants, treating anoxic groundwater at 80% recovery for drinking water production, were characterized and compared with oxic NF and reverse osmosis systems. Plant operating times varied between 6 and 10 years and pretreatment was limited to 10 μm pore size cartridge filtration and antiscalant dosage (2–2.5 mg L^{-1}) only. Membrane performance parameters normalized pressure drop (NPD), normalized specific water permeability (K_w) and salt retention generally were found stable over extended periods of operation (>6 months). Standard acid–base cleanings (once per year or less) were found to be sufficient to maintain satisfying operation during direct NF of the described iron rich (≤8.4 mg L^{-1}) anoxic groundwaters.

Extensive autopsies of eight NF membrane elements, which had been in service since the plant startup (6–10 years), were performed to characterize and quantify the material accumulated in the membrane elements. Investigations using scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDS), total organic carbon (TOC) and adenosine triphosphate (ATP) measurements revealed a complex mixture of organic, biological and inorganic materials. The fouling layers that developed during half to one year of operation without chemical cleaning were very thin (<2 μm). Most bio(organic) accumulates were found in the lead elements of the installations while inorganic precipitates/deposits (aluminosilicates and iron(II)sulfides) were found in all autopsied membrane elements.

The high solubility of reduced metal ions and the very slow biofilm development under anoxic conditions prevented rapid fouling during direct NF of the studied groundwaters. When compared to oxic NF and RO systems in general (e.g. aerated ground waters or surface waters), the operation and performance of the described anoxic installations (with minimal pretreatment) can be described as very stable.

Abbreviations: Π, osmotic pressure; *A*, surface area; act, actual; ATP, adenosine triphosphate; *c*, concentrate; CIP, cleaning in place; DV, Diepenveen; EC, electrical conductivity; EDS, energy dispersive X-ray spectroscopy; EN, European norm; *f*, feed; FCP, feed channel pressure drop; ISO, international organization for standardization; K_w, normalized specific water permeability; MFI, modified fouling index; NEN, Nederlandse norm (dutch norm); NF, nanofiltration; NOM, natural organic matter; norm, normalized; NPD, normalized pressure drop; *P*, pressure; *p*, permeate; *Q*, volume flow rate; ref, reference; RM, Rodenmors; RO, reverse osmosis; SEM, scanning electron microscopy; SRB, sulfate reducing bacteria; *T*, temperature; TCF, temperature correction factor; TOC, total organic carbon; tot, total; WH, Witharen; WW, Weerseloseweg

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1. Introduction

High pressure driven membrane filtration processes, such as nanofiltration (NF) and reverse osmosis (RO) are capable of generating large amounts of high quality drinking water. The excellent removal capacity of contaminants, decreasing prices for the membranes and enhanced membrane lifetimes led to the wide acceptance and popularity of NF and RO. NF is applied for the treatment of surface waters [1,2], as pre-treatment in desalination [3], during waste water reclamation [4] and for the treatment of oxic groundwater [5].

Vitens Water Supply Company (The Netherlands) uses direct NF, with minimal pretreatment, for drinking water production from anoxic groundwater [6], in 9 of its installations. Four of these installations were investigated in the present study (Table 1) and compared to literature data of oxic NF and RO systems. At present, approximately 55% of the water volume produced by the Dutch drinking water companies originates from deep groundwater layers, corresponding to a total volume of 675 million m^3 $year^{-1}$ in 2010. The composition of the untreated deep groundwater does not meet the Dutch drinking water standards or the standards of Vitens in terms of hardness, color, total organic carbon (TOC), sulfate, ammonium, iron, manganese, barium and/or organic micro pollutants (Table 2). Therefore the groundwater needs to be treated. Conventional groundwater treatment (aeration and filtration) does not remove, e.g., color, hardness and pollutants such as pesticides [7]. NF is used by Vitens when 2 or more of the water standards (that cannot be removed with conventional treatment, aeration and sand filtration) are not met, as the simultaneous removal of these compounds by NF is cheaper compared to the combination of other treatments, such as softening and carbon filtration [6,7]. Similar to other membrane filtration processes and feed water sources, the major concern is reduced membrane

Table 1
Design and operational characteristics of the 4 described installations.

Installation name	Weerseloseweg	Rodenmors	Diepenveen	Witharen
Abbreviation	WW	RM	DV	WH
Years of operation[a]	10	9	8.8	5.7
Cleaning frequency [CIPs $year^{-1}$]	0.7	0.6	0.6	0.7
Production capacity [m^3 day^{-1}]	2880	1785	4608	2880
Membrane area [m²]	2412	1809	4221	2412
Number of stages	2	2	2	3
Recovery (%)	80	80	78	80
Well depth [m]	80–150	23–45	33–35	60–90
Antiscalant dosage [mg L^{-1}]	2–2.5	2–2.5	2–2.5	2–2.5

[a] At day of autopsies for fouling analysis.

Table 2
Overview of the physical, chemical and biological parameters (average concentration and standard deviation) of the feed waters of the 4 NF installations, as well as the Dutch drinking water standards and the desired drinking water qualities by Vitens. Red color indicates that either Dutch or Vitens standards for drinking water are not met.

Measurement	Units	Dutch Norm*	Vitens Norm**	Diepenveen	Rodenmors	Weerseloseweg	Witharen
$temp_{min,max}$	°C	≤ 25	≤ 25	9.5 - 13.1	10.6 - 13.2	9.5 - 14.2	11.6 - 13.2
pH	pH	7.0 - 9.5	7.8 - 8.3	7.22 ± 0.03	6.99 ± 0.03	7.22 ± 0.05	6.81 ± 0.03
conductivity	mS m^{-1}	≤ 125	≤ 80	63 ± 1	56 ± 4	67 ± 1	53 ± 1
oxygen	mg L^{-1}	≥ 2	≥ 4	< 0.01	< 0.01	< 0.01	< 0.01
colour	mg Pt/Co L^{-1}	≤ 20	≤ 10	≥ 20	≥ 20	≤ 10	≥ 20
total hardness (tH)	mmol (Ca^{2+} + Mg^{2+}) L^{-1}	≥ 1.0	1.0 - 1.2	3.25 ± 0.1	2.74 ± 0.2	3.48 ± 0.1	2.66 ± 0.1
TOC	mg L^{-1}		≤ 3	7.3 ± 0.3	7.9 ± 0.5	3.1 ± 0.3	9.2 ± 0.2
DOC	mg L^{-1}			6.6 ± 0.3	6.8 ± 0.6	2.9 ± 0.2	8.6 ± 0.4
ATP	ng L^{-1}			2.4 ± 1.8	2.3 ± 3.1	1.9 ± 1.4	<1
sulfate	mg SO_4^{2-} L^{-1}	≤ 150	≤ 120	65.3 ± 1.2	9.5 ± 2.7	123.8 ± 4.1	< 2
sulfide	mg S^{2-} L^{-1}			n.d.	< 2	< 2	n.d.
nitrate	mg NO_3^- L^{-1}	≤ 50	≤ 25	< 1	< 1	< 1	< 1
ammonium	mg NH_4^+ L^{-1}	≤ 0.2	≤ 0.05	1.1 ± 0.0	1.5 ± 0.1	0.3 ± 0.0	1.4 ± 0.0
ortho-phosphate	mg PO_4^{3-} L^{-1}			0.5 ± 0.0	0.8 ± 0.4	0.5 ± 0.3	0.6 ± 0.2
hydrogen carbonate	mg HCO_3^- L^{-1}	≥ 60	≥ 90	319 ± 2	329 ± 15	286 ± 3	337 ± 2
$methane_{(headspace)}$	µg L^{-1}			313 ± 8	14857 ± 639	145 ± 20	15143 ± 639
calcium	mg L^{-1}			116 ± 3	99 ± 6	121 ± 4	96 ± 3
chloride	mg L^{-1}	≤ 150	≤ 100	37.3 ± 0.9	38.0 ± 12.0	30.8 ± 1.1	30.4 ± 1.9
sodium	mg L^{-1}	≤ 150	≤ 100	22.1 ± 0.8	21.0 ± 4.8	22.6 ± 0.8	19.4 ± 0.9
magnesium	mg L^{-1}			8.5 ± 0.2	6.8 ± 0.6	11.3 ± 0.6	6.6 ± 0.1
iron	mg L^{-1}	≤ 0.2	≤ 0.05	4.2 ± 0.0	7.1 ± 0.2	2.0 ± 1.2	8.4 ± 0.2
potassium	mg L^{-1}			3.7 ± 0.1	2.2 ± 0.2	3.3 ± 0.1	2.2 ± 0.0
manganese	mg L^{-1}	≤ 0.05	≤ 0.01	0.3 ± 0.0	0.3 ± 0.0	0.2 ± 0.1	0.3 ± 0.0
silicon	mg L^{-1}			6.7 ± 0.2	9.6 ± 0.3	11.6 ± 0.2	10.7 ± 0.3
strontium	µg L^{-1}			409 ± 11	396 ± 30	772 ± 35	400 ± 6
barium	µg L^{-1}	≤ 500	≤ 100	197 ± 6	78 ± 7	101 ± 5	14 ± 0
aluminum	µg L^{-1}	≤ 200	≤ 30	4.1 ± 2.1	2.4 ± 0.3	< 2	2.8 ± 1.0

n.d. = not determined
* = Dutch drinking water standards (Drinkwaterbesluit 2011)
** = Desired drinking water quality (Vitens)