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Benefits and hurdles of using brackish groundwater as a drinking water source in the Netherlands

Pieter J. Stuyfzand · Klaasjan J. Raat

Abstract The production of fresh drinking water from brackish groundwater by reverse osmosis (BWRO) is becoming more attractive, even in temperate climates. For successful application of BWRO, the following approach is advocated: (1) select brackish source groundwater with a large volume and a composition that will yield a concentrate (waste water) with low mineral saturation; (2) maintain the feed water salinity at a constant level by pumping several wells with different salinities; (3) keep the permeate-to-concentrate ratio low, to avoid supersaturation in the concentrate; (4) keep the system anoxic (to avoid oxidation reactions) and pressurized (to prevent formation of gas bubbles); and (5) select a confined aquifer for deep well injection where groundwater quality is inferior to the membrane concentrate. This approach is being tested at two BWRO pilot plants in the Netherlands. Research issues are the pumping of a stable brackish source water, the reverse osmosis system performance, membrane fouling, quality changes in the target aquifer as a result of concentrate disposal, and clogging of the injection well. First evaluations of the membrane concentrate indicate that it is crucial to understand the kinetics of mineral precipitation on the membranes, in the injection wells, and in the target aquifer.

Keywords Water supply · Salt-water/fresh-water relations · Reverse osmosis · Waste water disposal · The Netherlands

Introduction

In (semi)arid countries, reverse osmosis of brackish groundwater (BWRO) is increasingly applied to prepare drinking, industrial or irrigation water from brackish groundwater (chloride 300–10,000 mg/l) on a cost-effective basis. Production plants have already been in operation for several decades in many parts of the world, especially in the Middle East (Ahmed et al. 2001; Al-Zubari 2003; Afonso et al. 2004; Jaber and Ahmed 2004) and the USA (Mickley 2006). BWRO is currently also increasingly popular in and around the Mediterranean and Caribbean Seas, and in the Far East and Pacific, including Australia. BWRO and seawater reverse osmosis (SWRO) systems, especially when operated with renewable energy, are considered as two of the most promising solutions to the world water crisis.

Advantages of BWRO over SWRO include a lower membrane-fouling hazard (by biomass, suspended fines, and SO_4 precipitates), a lower energy demand, less need for extensive post-treatment (especially regarding boron removal), availability of source water also far inland, and lower costs. Disadvantages may consist of: (i) relatively high concentrations of Fe, Mn, SiO_2 , NH_4 , PO_4 , and HCO_3 in the source water, which increase the risk of membrane fouling by precipitates of sulphides, silicates, phosphates, and carbonates; (ii) limited availability of brackish groundwater of constant quality, because it is often connate (without actual recharge) and hydrochemically heterogeneous (stratified); and (iii) more problems with disposal of the membrane concentrate (waste water) due to higher nutrient concentrations and remoteness from the sea.

Countries in temperate climates with less severe water scarcity problems are today seriously considering BWRO as a "source-treatment" option to prepare drinking water. The drivers consist of environmental problems like the pollution and salinization of aquifers, drawdown of water tables in phreatic aquifers, effects of climate change like reduced base flows that render surface waters less fit for drinking water production (due to contaminant levels and temperatures exceeding limits for water intake), and increasing costs to produce drinking water from heavily polluted, fresh groundwater.

Even in the Netherlands, a country with abundant surface and groundwater resources (Fig. 1), BWRO is

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Fig. 1 Depth to the 1,000 mg Cl/L interface (in m b.s.l.) with indication of five candidate sites for a BWRO pilot: Deventer, Noordoostpolder, Noord-Bergum, Zevenbergen, and Polder Groot Mijdrecht (modified after Stuyfzand and Stuurman 2008). Also shown: the approximate landward limit of the brackish to salt Holocene transgression waters (*Hol*), relatively shallow occurrences of marine deposits of late Tertiary to early Pleistocene age (*LT*), and relatively shallow Permian rock salt diapirs (*PSD*). Cross section AB in Fig. 3. 1–11 = coastal fresh water lenses down to 50–125 m b.s.l.

currently under serious consideration (Kooiman et al. 2004; Nederlof and Hoogendoorn 2005; Oosterhof and Nederlof 2006), for the following reasons:

- Brackish groundwater is an excellent feed water for reverse osmosis (RO) installations by virtue of its very low content of suspended fines (leading to a very low membrane fouling) and lack of both organic pollutants and pathogenic micro-organisms;
- Salinized or salinizing well fields, with a separate BWRO unit for brackish wells, may continue to abstract the allowed quatum or even abstract more than before;
- A reduced hydraulic impact on the upper aquifer system as compared to fresh groundwater pumping; and

- Taxes are imposed on fresh groundwater exploitation (approximately 0.28 €/m³ in the Netherlands), but not (yet) on brackish and saline groundwater.

In addition, the feasibility of using BWRO to produce drinking water from intercepted saline seepage water in reclaimed land areas (polders) is currently being investigated, in order to reduce the salt and nutrient load of the polder's surface water system. As a matter of fact, BWRO is one of the options to improve surface water quality of two deep (land surface >2 m below sea level (bsl)) polders (Olsthoorn 2008; Vink et al. 2007).

An alternative to BWRO is to apply the "fresh-keeper" approach (Grakist et al. 2002; Kooiman et al. 2004; Fig. 2). In this approach, the upconing of brackish water

Fig. 2 Schematic of (from left to right) a fresh well salinizing by upconing, the fresh-keeper without reverse osmosis (RO), the fresh-keeper with RO, and brackish water with RO (BWRO). The chlorinity of the RO-concentrate is approximately 6,000 mg/l when the quantity of permeate equals the quantity of concentrate

to a fresh water well is prevented by installing an additional deeper or upgradient well that intercepts the (upconing) brackish water. The original single abstraction well is thus kept "fresh". The fresh-keeper approach may be applied without or with RO (Fig. 2). In the first case the brackish or salt groundwater needs to be disposed of. In the latter case, and for standard BWRO systems, the membrane concentrate will constitute the waste water. Its disposal is more problematic than that of brackish to saline groundwater, for several reasons: (i) it may contain antiscalants, (ii) several constituents of the feed water (notably Fe, NH_4 , PO_4 , As, Ba, Cr, F and Ni) can be concentrated above the natural background or even above maximum contaminant levels, and (iii) the regulator has a bias towards lower salinity levels and as a consequence it is difficult to get approval for injection of water higher in salinity than the ambient water.

In temperate climates and in countries with severe environmental laws, many of the disposal options mentioned by Mickley (2006), like evaporation ponds and discharge to surface water, are deemed to fail or to be rejected by regulators. In the Netherlands, deep well injection into a more saline, confined aquifer is considered to be the best solution to the waste water problem (Fig. 2).

Prior to the application of deep well injection, there are various questions to be addressed, like: (a) is the quality of the injected water acceptable when compared to the ambient groundwater in the target aquifer? (b) which reactions occur in the mixing zone between the native groundwater and injected concentrate, and what are their consequences? (c) which reactions continue to operate between the injected bubble and the aquifer matrix, and what are their consequences? (d) how can well clogging be prevented when injecting a supersaturated solution? (e) will clay minerals mobilize with a risk of mechanical aquifer plugging? and (f)

how can one prevent short-circuiting between the pumped aquifer and the injected aquifer, as well as undesired fresh or salt water intrusions into both aquifers?

In this contribution, some preliminary answers and ways to solve or circumvent the above problems are explored. The deep well injection system was scheduled to be tested in practice at two pilot plants in the Netherlands in the summer of 2009. Some details of the experimental setup of both pilot plants are presented.

Selection of a suitable source water

The sustainable use as drinking, industrial, or irrigation water puts several constraints on the chemical composition of brackish to saline groundwaters as feed water for BWRO:

- (1) the salinity must be favorable for optimum membrane purification at reduced costs: preferably chloride $<10,000$ mg/l or total dissolved solids (TDS) $<20,000$ mg/l. BWRO systems normally operate between 1,000 and 7,000 mg Cl/L;
- (2) the concentration of ions less soluble than Na, K, and Cl should be low enough to prevent scaling of membranes or injection wells with, for instance, silicate ($\text{SiO}_2 \cdot n\text{H}_2\text{O}$, $\text{MgSi}_2(\text{OH})_6$), sulphate (BaSO_4 , $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), carbonate (CaCO_3 , $\text{CaMg}(\text{CO}_3)_2$, FeCO_3), or phosphate ($\text{Ca}_5(\text{PO}_4)_3\text{OH}$; $\text{NH}_4\text{MgPO}_4 \cdot 6\text{H}_2\text{O}$) minerals. The use of antiscalants should be avoided, in order to increase the chance of getting a license for deep well injection of the waste water;
- (3) the concentration of ions in the permeate should not exceed the maximum permissible concentrations for drinking water. Simple post-treatment like aeration is adequate to get rid of gases like CH_4 , H_2S and CO_2 . But

uncharged dissolved species with a small size, like boron (H_3BO_3) may pose serious problems. Boron is very hard to remove; it can attain rather high concentrations in brackish groundwater, may pass membranes and may then exceed the World Health Organization's (WHO) and the European Union's (EU) drinking water standards of 500 and 1,000 $\mu\text{g/l}$, respectively;

- (4) groundwater is preferred that, after membrane concentration, remains below the levels in the target aquifer for parameters like salinity, nutrients and heavy metals, because its deep well injection is then more likely to be permitted by regulators; and
- (5) the abstraction of brackish feed water should not result in salinization or freshening of the aquifer, well clogging or corrosion of well and transport mains.

The reverse osmosis process

Unpolluted brackish groundwater can easily be treated to drinking water quality by RO, if followed by aeration to get rid of gases like CH_4 , H_2S , and CO_2 , and by rapid sand filtration to eliminate the remaining Fe, Mn, and NH_4 . The permeate does not need any separate conditioning, if mixed with sufficient fresh or slightly brackish groundwater before their combined conventional treatment (aeration and rapid sand filtration). Otherwise a further post-treatment of the permeate is needed to stabilize pH at a higher level while meeting hardness requirements. When high boron concentrations in the feed water cannot be avoided, specific high boron removal RO membrane elements or ion exchangers should be applied (Taniguchia et al. 2004; Glueckstern and Priel 2007).

Antiscalants like polyphosphonates are frequently applied to prevent clogging of the membranes and also of the deep well injection facilities. HCl can be applied to reduce supersaturation of, for instance, calcite. It is to be preferred, however, not to apply them, because their use lowers the chance of getting a deep well disposal permit in the upper 500 m, at least in the Netherlands. In that case the permeate-to-concentrate ratio should be kept low (ca.1.0) in order to keep mineral supersaturation levels in the concentrate low.

All unpolluted, brackish groundwaters in the Netherlands are composed of (deep) anoxic palaeowaters containing significant amounts of Fe, Mn, and NH_4 (Tables 1, 2). By keeping the feed water and the membrane concentrate anoxic, the oxidation and precipitation of Fe(III) and Mn (IV) flocs can be completely prevented (Nederlof and Hoogendoorn 2005), thus facilitating the direct injection of the membrane concentrate without pretreatment.

Selection of a suitable disposal aquifer

A sustainable, deep well injection can be realized when the hydrogeological and hydrogeochemical properties of the target aquifer meet the following criteria:

- (1) hydraulic conductivity and storativity should be high enough to store a large quantity of waste water for a long time;

Table 1 Overview of the chemical composition of various brackish and saline groundwater types, coastal North Sea water and Atlantic Ocean water (modified after Stuyfzand and Stuurman 2008)

Sample code	Depth m+MSL	EC uS/cm	Temp °C	pH	Cl mg/l	SO_4	HCO_3	NO_3	PO_4	Na	K	Ca	Mg	NH_4	Fe	Mn	SiO_2	DOC	O_2
Saline surface waters																			
North Sea @	-0.1	39,000	10.5	8.1	16,800	2,355	160	3.0	0.51	9,350	346	350	1,122	0.40	0.10	0.05	0.9	2.4	9.4
Atl. Ocean	-0.1	45,150	10.0	8.1	19,805	2,775	145	0.3	0.06	11,020	408	422	1,322	0.00	0.00	0.01	4.4		10.0
Saline groundwater, type: (sub)recent North Sea																			
24H.259	-104.6	40,700	11.5	7.2	16,300	2,294	253	<0.5	2.78	9,208	330	357	1,080	6.1	6.40	0.46	10.5	4.9	0.1
19A.328	-116.6	40,600	11.6	7.0	15,896	2,220	187	0.0	1.01	8,700	192	476	1,114	8.9	21.43	1.24	14.8	5.8	0.0
Brackish - saline groundwater, type: Holocene transgression, open/gully																			
25C.183	-70.2	16,380	12.3	7.2	6,016	603	424	0.0	0.16	2,625	118	792	277	8.1	9.66	0.28	10.4		0.0
7C.101	-61.5	23,927	11.0	6.6	8,781	892	525	0.0	0.48	4,320	23	905	292	7.2	0.40	1.47			0.0
Brackish - saline groundwater, type: Holocene transgression, lagoonal																			
19A.182	-30.0	7,450	10.5	7.7	1,765	41	2,458	<0.1	37.00	1,395	84	81	215	123.0	5.40	0.53	65.0		0.0
19B.109	-16.0	38,900	11.0	7.1	14,262	780	3,752	<0.1	62.70	8,317	256	343	1,131	164.0	1.20	0.56	57.6	71.1	0.1
Brackish - saline groundwater, type: late Tertiary - early Pleistocene																			
19A.259	-323.4	29,670	16.0	7.4	11,000	840	461	<0.1	6.20	6,000	100	310	630	75.3	4.10	0.52	13.9	3.5	0.1
44F.146	-370.0	23,700	12.8	7.1	8,960	18	311	0.0	0.02	5,221	174	193	378		2.40	1.38	13.2		0.0
31E.176	-277.0	9,834	14.0	7.5	2,925	0	309	0.0	1.10	1,959	24	26	68	43.7	0.60	0.05	18.1		0.0
Brackish - hypersaline groundwater, type: halite leachate																			
52E.114	-524.0	29,400	21.0	6.6	12,939	230	514	<0.5	0.56	7,200	185	610	290	22.6	3.30	0.41	28.3	70	0.0
8D.034	-582.0	103,000	22.0	6.5	73,867	1,740	163	<0.5	0.02	43,000	290	2,200	870	36.1	9.90	1.20	20.3		0.0
61F.296	-420.0	15,400		7.1	5,810	354	921	0.0	0.02	3,910	117	181	95	6.6	1.60	0.20			0.0

@ = in the 1980s

Table 2 Saturation indices (SI_M) for selected minerals, of various brackish and saline groundwater types, coastal North Sea water and Atlantic Ocean water. Selection of minerals, based on the following criteria (1) natural occurrence of the minerals, (2) relevance of minerals for membrane fouling, and (3) reliability of the underlying data. Potentially problematic SI 's in *italics*

Sample code	Ba ug/l	Br	F	I	Li	Sr	δ^2H ‰	3H TU	$\delta^{13}C$ ‰	^{14}C pmc	$\delta^{18}O$ ‰	SI_M cal- cite	dolo- mite	side- rite	rhodo- chrosite	gyp- sum	ba- rite	vivia- nite	OH- apatite
Saline surface waters																			
North Sea @+AJ146	40	56000	1200	55	160	7100		25.0			-1.4	0.48	1.66	-2.02	-1.04	-0.65	0.38	-8.44	1.69
Atl. Ocean	30	67300	1300	60	170	8100	0.0		1.0	95.0	0.0	0.49	1.66	-3.44	-2.13	-0.54	0.28	-14.45	-0.91
Saline groundwater, type: (sub)recent North Sea																			
24H.259	120	58000	240	73	175	7000		-1.5			-1.5	-0.16	0.37	0.24	-0.71	-0.65	0.83	-0.03	0.45
19A.328	178	58571			108	5570						-0.36	-0.15	0.45	-0.60	-0.54	0.99	0.23	-1.17
Brackish - saline groundwater, type: Holocene transgression, open/gully																			
25C.183												0.56	0.86	0.79	-0.50	-0.59		-1.35	-0.61
7C.101								1.7	-13.8	57.1	-5.2	0.09	-0.13	-1.08	-0.33	-0.45		-6.03	-1.93
Brackish - saline groundwater, type: Holocene transgression, lagoonal																			
19A.182	17	9400	420	2590	52	2100		-1.0	12.0		-6.4	0.94	2.43	1.47	0.76	-2.5	-1.05	3.08	4.58
19B.109	120	63000	290	3550	155	7000		0.0				0.88	2.46	0.32	0.23	-1.14	0.44	-0.48	3.9
Brackish - saline groundwater, type: late Tertiary - early Pleistocene																			
19A.259	180	55000	100	2	97	6500		0.0				0.38	1.34	0.57	-0.05	-1.02	0.67	0.69	2.97
44F.146	1255	35133			271	5239				<5.0		-0.30	-0.11	-0.05	-0.08	-2.76	0.08	-5.16	-6.78
31E.176						730		-2.6	-10.4	4.9	-7.2	-0.53	-0.43	-0.17	-0.86			-1.97	-2.34
Brackish - hypersaline groundwater, type: halite leachate																			
52E.114		20200					-44.5	-0.7		1.0	-6.5	-0.10	-0.36	-0.25	-0.94	-1.25			
8D.034	600	113000	50	3700	4600	147000	-25.6	-0.5		9.1	-3.2	-0.23	-0.61	-0.74	-1.62	-0.16	1.06	-3.54	-0.82
61F.296	160	8600		203	6160	14230	-56.2	0	-4.78	3.0	-8.2	0.09	0.04	0.11	-0.54	-1.36	0.66		

@ in the 1980s

Pmc Percent modern carbon

- (2) the aquifer must be as confined as possible, so that the water injected remains contained in a limited space well below land surface;
- (3) to obtain the necessary permits, the native groundwater should preferably have a higher salinity and higher concentration of critical compounds (like nutrients, heavy metals) than the injectate; and
- (4) the tendency of clay minerals in the aquifer to mobilize by deflocculation should be low, because it could provoke mechanical clogging of the aquifer when the particles strand in the pore necks. This tendency is low when the sodium adsorption ratio (SAR) of the injectate and of the native groundwater is low, their salinity is high, the clay mineral content is very low, and the dominant type of clay minerals is favorable (kaolinite > illite > smectite). Further details are given by Scheuerman and Bergersen (1990).

The presence of reactive aquifer constituents like carbonate minerals, soil organic material and pyrite seems less inhibitive, because the membrane concentrate normally is already (super)saturated by itself and anoxic. This prevents strong dissolution reactions taking place, which can leach the aquifer. In extreme cases, dissolution could lead to the collapse of sedimentary structures and local leakage of the injectate from the target aquifer. This combination of processes might happen when injecting waters strongly undersaturated with respect to gypsum or anhydrite and halite, into an aquifer containing these minerals, or when injecting waters that became strongly aggressive towards calcite by HCl dosage, into a limestone aquifer.

In the Netherlands, deep well injection (> 100 m b.s.l.) resides under the Mine Act, which regulates the exploration and exploitation of resources like gas and oil. Clearly, this act was not designed to deal with issues like membrane concentrate disposal. In fact, the results of two small-scale BWRO pilots (see below) will be used by national and regional authorities for the development of future legislation of BWRO systems and concentrate injection. Before permitting the small-scale pilots, the legislator advocated that the injectate should preferably have a better quality than the original groundwater in the target aquifer, which conforms to the Australian draft guidelines for Managed Aquifer Recharge (EPHC–NHMRC–NRMCC 2008). This implied that the injectate should have salinity levels lower than or equal to the ambient groundwater in the target aquifer and that concentrations of heavy metals should not exceed threshold levels of the EU Water Framework Directive. Note that the Australian draft guidelines apply to injected water that is intended to be reused. Injection of BWRO concentrate would be considered waste disposal in Australia, for which specific waste disposal acts apply (Peter Dillon, CSIRO, pers. comm. 2009).

The bias of Dutch regulators towards injecting water lower in salinity stems from current problems with

salinization of aquifers and surface waters in the Netherlands. From a hydro-technological viewpoint, however, it may be advisable to inject higher salinity water, as this water is more likely to migrate downwards and reside in the lower regions of the target aquifer. In fact, upward migration of injected, lower salinity water has been documented at several wastewater injection sites in Florida, USA (Maliva et al. 2007).

There is no map yet of candidate disposal aquifers in the Netherlands, especially not with regard to the upper 500 m. At greater depth, more information exists through explorations for gas and oil. Disposal via abandoned gas or oil wells can be an option as discussed by Nicot and Chowdhury (2005). Disposal at large depth (below ca. 900 m) is also applied at the world's largest inland BWRO plant in El Paso, Texas, USA (Landers 2007). Even though the injectate is supersaturated towards calcite, barite and silica, clogging is not considered an issue there, as the target dolomite aquifer is highly fractured.

Deep well injection

When the above-mentioned conditions are satisfied, deep well injection still may encounter difficulties due to well or aquifer clogging. The main causes are probably chemical precipitation, microbiological fouling, and the formation of gas bubbles. The latter can be prevented by keeping the membrane concentrate pressurized. Microbiological fouling is frequently prevented by chlorination, but that would again reduce the chance of getting a disposal permit in the Netherlands, where chlorination of water received a bad reputation since the banning of drinking water chlorination in the 1980s due to the formation of hazardous by-products. If well clogging occurs, then periodical mechanical well-rehabilitation methods are preferred over chemical methods (see for instance Houben and Treskatis 2007).

The mixing of the injectate with native groundwater normally does not pose serious clogging phenomena, because the water quantity involved is too small, especially in single porosity aquifers. By refraining from antiscalants and acids, mineral phases will hardly or not dissolve, as the solution is already supersaturated by itself. Redox reactions will be minimal as well, because the water injected and the host aquifer are both (deep) anoxic. Water–rock interactions are therefore expected to remain limited to cation exchange and the deposition of some minerals like calcite and apatite, and possibly also dolomite, siderite, vivianite, barite, and amorphous silica.

Suitable BWRO source waters in the Netherlands

An inventory of brackish to hypersaline groundwaters by Stuyfzand and Stuurman (2008) forms a good starting point for the selection of a suitable brackish water source aquifer in the Netherlands. On the basis of environmental tracers, the hydrochemical fingerprint and geological

evolution, they discerned five types of non-anthropogenic, brackish to hypersaline groundwater in the upper 600 m of the Dutch subsurface: (sub)recent, intruding North Sea water along the coast; Holocene transgression water in a high-energy environment like open-marine, tidal gully or estuary; Holocene transgression water in a low-energy environment like a lagoon; relict marine groundwater of late Tertiary to early Pleistocene age; and Permian or Devonian halite leachate. The chemical composition of some examples of these groundwaters is shown in Tables 1, 2.

The spatial distribution of three types of brackish to hypersaline groundwaters in the upper 600 m of the subsoil of the Netherlands is indicated in Fig. 1 (planar view) and Fig. 3 (cross section AB). The Holocene transgression waters dominate in the upper 100–200 m of the western and northern parts of the country constituting the low-lying coastal plain. The top of relict groundwater of late Tertiary to early Pleistocene age is observed at relatively shallow depth (<150 m) in the areas indicated in Fig. 1 (LT₁, LT₂, and LT₃) and as local upconings in several reclaimed lakes with land surface at 4–7 m b.s.l., which need to be heavily drained (Fig. 3). Elsewhere its upper boundary normally resides at depths between 200 and 600 m b.s.l. Permian halite leachate occurs within the upper 600 m on a very local scale in the northeast, only where salt diapirs occasionally rise up to 200–400 m b.s.l. Devonian halite leachate could be important at depths >600 m b.s.l. in the southern parts, and at depths >50 m b.s.l. in the far southeast, the only region in the Netherlands

(South Limburg) where Cretaceous limestone crops out. The extent of (sub)recent, intruding North Sea water is limited to a coastal strip of about 6 km wide (Stuyfzand 1995; Fig. 3).

For desalination purposes, it is recommended to use groundwaters with a moderate salinity (1,000–7,000 mg Cl/L) because they require less energy during RO, and with relatively low concentrations of HCO₃, SO₄, Ca, Mg, Fe, Mn, NH₄, PO₄, SiO₂, and Ba, in order to reduce risks on membrane and injection well scaling. This means that (sub)recent, intruding North Sea water along the coast, Holocene transgression water in a low-energy environment like a lagoon, the saltier relict groundwaters of late Tertiary to early Pleistocene age, and the saltier Permian or Devonian halite leachates (Tables 1, 2) should be avoided. Most suitable BWRO source waters are therefore Holocene transgression water in a high-energy environment and the late Tertiary to early Pleistocene groundwaters with relatively low to moderate chloride levels (Cl <7,000 mg/l).

The feasibility of brackish groundwater abstraction depends also on the hydrogeological conditions. The hydraulic conductivity of the pumped aquifer should be sufficiently high and the brackish groundwater stock must be large enough to supply feed water for a long time. The latter could form a serious problem because inland brackish groundwaters are mostly palaeowaters lacking actual recharge. In addition, brackish groundwaters are normally stratified with chlorinity increasing and alkalinity decreasing with depth on a large scale, and with local

Fig. 3 Cross section AB (position indicated in Fig. 1), showing the spatial distribution of groundwater bodies in the Netherlands. Modified after Stuyfzand (1995)

inversions around aquitards (see section *BWRO pilots in the Netherlands*).

Pumping may therefore easily result in temporal and spatial quality variations in the feed water that either adversely affect the RO process or require a sophisticated RO adaptation strategy. A solution to this problem is offered by pumping several wells that vary in salinity (both higher and lower than the desired level) at a rate autocontrolled by on-line electrical conductivity (EC) measurements in each well and their mixture (Fig. 4).

This deliberate mixing of brackish groundwaters of differing salinity and composition could provoke the undesired formation of chemical precipitates, either in the well, transport mains, or on the membranes of the RO installation. Obviously the mixing of oxic with anoxic groundwater should be avoided at all costs, as this is a well-known recipe to create iron flocs, well clogging and scaling (Houben and Treskatis 2007). Brackish groundwaters normally are, however, (deep) anoxic, which excludes this risk. The mixing of waters saturated in carbonate minerals often results in undersaturation (Appelo and Postma 2005), which is even an advantage of mixing in this case. The mixing of waters saturated in sulphate minerals may lead to supersaturation and their precipitation. However, practically all brackish groundwaters in the Netherlands are deeply undersaturated with respect to gypsum, which minimizes this risk as well.

In the planning phase of a BWRO it is advisable to analyze for the various contributing water quality types that may mix in the abstraction system, and then calculate, for instance using PHREEQC-2 (Parkhurst and Appelo 1999), what happens to the mineral equilibria when these waters mix in various ratios.

Fig. 4 Schematic of a well field for BWRO in an aquifer with strong chlorinity stratification, and the solution to yield a stable feed water quality by pumping several wells with different salinity (both higher and lower than the desired level) at a rate autocontrolled by on-line EC measurements in each well and their mixture. Chlorinity of RO-concentrate is approximately 4,000 mg/l in this example

A critical look at mineral saturation indices

The saturation index of water with a particular mineral M (SI_M) is generally defined as :

$$SI_M = \log(IAP/K_S) \quad (1)$$

where : IAP = ion activity product of the mineral–water reaction in the sample [on a mol/kg water basis]; and K_S = the corresponding solubility product in pure water, adjusted to the temperature and pressure of the sample.

Water with a $SI_M=0$ is in equilibrium with the mineral considered; when $SI_M<0$ water is undersaturated and will tend to dissolve the mineral when it is met; and when $SI_M>0$ it is oversaturated and may deposit the mineral. The SI_M , therefore, is an indicator for the risk of mineral precipitation and thus provides insight in the possibility of clogging of the injection wells.

Calculated SIs should be interpreted with care, as is discussed below for the five brackish to hypersaline groundwater types in the Netherlands discerned above. Their chemical composition and SIs with respect to eight common minerals are listed in Tables 1, 2. Most groundwaters are close to equilibrium with calcite due to abundant calcitic and aragonitic shell fragments in most marine formations. Minor deviations (-0.3 to $+0.3$) occur due to errors in pH measurement, and the cases of clear supersaturation (0.5 – 0.9) can be related to high concentrations of dissolved organic matter (DOC) which complex Ca, and of PO_4 and Mg, which inhibit crystallization. The SIs for dolomite ($CaMg(CO_3)_2$) often attained significant supersaturation (1 – 2.5) without evidence of dolomite being actually present in the system. This is explained by a well-known, extremely sluggish crystallization, in combination with dominant sources of high Mg concentrations from both ocean water and cation exchange processes.

Equilibrium and supersaturation with respect to siderite ($FeCO_3$) and rhodochrosite ($MnCO_3$) were also observed but may likewise suffer from inaccuracies in pH measurement, unaccounted complexation of, respectively, Fe^{2+} and Mn^{2+} by DOC, and from kinetic hindrances (Jensen et al. 2002). Nevertheless, the presence of postdepositional manganous siderites in various aquifers in the Netherlands (Huisman 1998) suggests that its dissolution or precipitation is feasible. All waters were aggressive towards gypsum ($CaSO_4 \cdot 2H_2O$), which is explained by its absence as a mineral phase in the aquifers in all cases except where halite dissolved.

Barite ($BaSO_4$) has been demonstrated in geochemical surveys (Huisman 1998) and shows supersaturation (0.3 – 1.1) in most cases. This supersaturation testifies of either unaccounted complexation by organic acids or kinetic hindrances.

The situation with respect to phosphate minerals like vivianite ($Fe_3(PO_4)_2 \cdot 8H_2O$) and hydroxi-apatite ($Ca_5(PO_4)_3OH$) is subject to high uncertainty because of a high sensitivity of the calculated SI to analytical inaccuracies (pH, difficulties in distinguishing inorganic from organic PO_4 with current preservation and analytical

techniques) and variations in mineral composition and degree of crystallization. Apatites seem to be rather abundant in marine formations, however, and may be relevant especially in samples with high PO_4 concentrations and without strong redox reactions with organic matter.

Equilibria with sulphur-containing minerals like pyrite could not be calculated by lack of (good) H_2S data. All samples appeared undersaturated with respect to halite, fluorite, and opal. Sample 8D.034 approached halite equilibrium ($\text{SI} \leq -1$), which is consistent with local halite leaching. Samples 19A.182 and 19B.109 approached opal (amorphous silica) equilibrium ($\text{SI} = -0.1$), which is explained by stagnant conditions in clayey marine deposits with high diatom contents. All groundwater samples were supersaturated ($\text{SI} = 0.5-10$) with respect to most silicate minerals (clay minerals, quartz, albite, and K-feldspar). This probably indicates that these minerals play a minor role in dictating the hydrochemistry of brackish to hypersaline waters at low temperatures, owing to kinetics.

It can be concluded that mineral supersaturation of water with respect to, for instance, calcite, dolomite, silicate, phosphate, and iron minerals does not necessarily mean that equilibrium will be attained by mineral deposition. The mineral saturation index, therefore, is a *risk* indicator for precipitation, and subsequent problems with chemical clogging of membranes, injection wells, and host aquifers. When $\text{SI} < 0$, these problems are not to be expected. When $\text{SI} > 0$, the index indicates there is a chance that clogging may occur. Field pilots are needed to further quantify this chance and to see at which saturation levels precipitates start to form and become a nuisance, as a function of contact time and contact surface conditions.

BWRO pilots in the Netherlands

BWRO systems consisting of a brackish groundwater abstraction, treatment with RO and concentrate disposal by deep well injection, have never been operational in The Netherlands. To date, five candidate BWRO pilots have been defined in the Netherlands: by water supply company Vitens, near Deventer, in the Noordoostpolder and close to Noord-Bergum, and by water utility Brabant Water near Zevenbergen, and by Waternet Amsterdam in Polder Groot Mijdrecht (Fig. 1).

The pilots near Deventer (Nederlof and Hoogendoorn 2005) and Polder Groot Mijdrecht (Olsthoorn 2008) have been cancelled, so that they are not further discussed below. Permits for pilots in Noord-Bergum (Vitens) and Zevenbergen (Brabant Water) have recently been issued and both pilots were scheduled to start in summer 2009.

Noordoostpolder

This polder area is part of the former Zuiderzee, a brackish bay of the North Sea into which the Yssel River (a tributary of the Rhine River) discharges. In 1932, this bay was closed off from the North Sea by a dike and automatically filled up with fresh water from the Yssel River. In 1942, the Noordoostpolder was reclaimed from the new Lake Yssel. Permanent drainage of this new land 3 m b.s.l. results in a relatively rapid freshening in the eastern parts that receive deep fresh groundwater from the mainland. This is not the case in the central and western parts where salinities remain higher (Fig. 5). The western parts are expected to slowly freshen due to slow infiltration of fresh water from Lake Yssel, which is hindered by Holocene aquitards and Recent mud layers settling out in the lake. In the central parts of this polder, brackish seepage is expected to continue for decades. This

Fig. 5 Schematic of the aquifer system in the Noordoostpolder, the observed chloride depth profiles, and the position of the projected pumping and injection wells. Lines represent chloride depth profiles observed in various observation wells in the western/central and eastern parts of the Noordoostpolder. Aquifer 2 = fluvial, coarse grained sand of Pleistocene age; Aquifer 3 = marine, fine to medium grained sand of late Tertiary and early Pleistocene age, with clay intercalations

makes the central part of the polder area best fit for BWRO, also because it reduces this brackish and eutrophic seepage, thus contributing to an improvement of surface water quality. This expected improvement, the increasing local population and ecological incentives to reduce the production of fresh drinking water from well fields on the mainland, triggered the idea to start a BWRO pilot. The position of pumping and injection wells is shown in Fig. 5, together with the distribution of aquifers and aquitards and the chlorinity distribution.

Table 3 shows preliminary calculations on the chemical characteristics of the membrane concentrate and permeate, together with the quality of the local source groundwater. It can be observed that increasing the RO efficiency ($= 100 \times \text{permeate volume} / \text{feed volume}$) from 50 to 80% will significantly raise the risk of clogging because of rising levels of supersaturation with respect to many minerals. The less saline, brackish groundwater from shallow depth is to be preferred from a chemical point of view. It yields a somewhat better concentrate quality during RO, and boron concentrations of the permeate would not approach WHO and EU drinking water standards. In the calculations (Table 3) it is assumed that undissociated boric acid (H_3BO_3), which is the dominant

boron species in the feed water, will pass through the membranes completely. This is a worst-case assumption, because some retention is normally observed.

The very high supersaturation levels for siderite, hydroxy-apatite and vivianite, also in the 50% concentrate, are worrisome as these minerals may form and plug the membranes or wells. However, especially vivianite and siderite also show supersaturation in the raw groundwater, indicating that formation of these minerals is kinetically hindered. The concentrate is expected to be supersaturated with respect to pyrite as well, but this is difficult to quantify because of the lack of reliable data on H_2S .

Figure 6 illustrates the calculated changes in water quality in Aquifer 3 (see Fig. 5) upon injection of membrane concentrate, as modeled with PHREEQC-2. The target aquifer is 30 m thick, with a CEC of 10 meq/kg, porosity of 0.3 and a bulk density of 1.8 kg/dm^3 . The native water quality mimicked that of the deeper aquifer in Table 3 (Raw B; 3,290 mg Cl/L). The 50% membrane concentrate of the shallow brackish groundwater (2,038 mg Cl/L; Table 3) was used as injection water, with an injection rate of $75 \text{ m}^3/\text{h}$ and a radial flow pattern. In this example super- and undersaturation

Table 3 Water quality survey of shallow and deeper brackish source water from the Noordoostpolder, and their calculated concentrate and permeate in a hypothetical BWRO-plant with 50 and 80% efficiency ($= 100 \times \text{permeate volume} / \text{feed water volume}$). Concentrate and permeate concentrations were calculated using Troi 1.5.0 (Trisep Corp.). Mineral saturation indices (SIs) were calculated with PHREEQC-2. Potentially problematic concentrations or SIs in *italics*

	Parameter Unit	Raw A	Shallow brackish groundwater, Cl=1,040 mg/l				Raw B	Deeper brackish groundwater, Cl=3290 mg/l			
			Concentrate		Permeate			Concentrate		Permeate	
			50%	80%	50%	80%		50%	80%	50%	80%
pH		6.6	6.84	7.13	5.04	5.2	7.35	7.52	7.36	5.67	5.52
Cl	mg/l	1,040	2038	4949	42	62	3,290	6,657	16,359	69	114
HCO ₃	mg/l	577	1142	2803	11	17	854	1,680	3,797	11	17
SO ₄	mg/l	10	19.9	49.6	0.1	0.1	3.1	6.2	15	0	0.7
PO ₄	mg/l	1.6	3.19	7.93	0.01	0.02	2.2	4.39	11	0.01	0.48
Na	mg/l	480	965	2335	23	34	1,820	3,601	8,840	40	65
K	mg/l	15	29.3	70.7	0.7	1.1	65.8	130.1	319.3	1.5	2.4
Ca	mg/l	260	515	1271	5	7	272	541	1,338	3	6
Mg	mg/l	37	73	181	1	1	168	334	826	2	4
NH ₄	mg/l	20	38.9	93.8	1.1	1.6	13.8	27.3	66.8	0.3	0.5
Fe	mg/l	55	109.6	272.7	0.4	0.6	10	19.9	48	0.1	2.2
Mn	mg/l	1.8	3.59	8.93	0.01	0.02	1	1.99	4.5	0.01	0.22
SiO ₂	mg/l	16	31.6	77.6	0.4	0.6	26.8	53.3	132.2	0.3	0.5
B	μg/l	239	239	239	239	239	757	757	757	757	757
Ba	μg/l	600	1,190	2930	10	20	300	600	1,480	10	10
F	μg/l	100	196	476	4	6	50	101	249	1	2
Sr	μg/l	1,510	2,990	7360	30	50	2,290	4,590	11,320	80	80
Mineral saturation index (SI _M)											
Calcite		-0.12	0.59	1.48			0.68	1.31	1.67		
Aragonite		-0.27	0.44	1.32			0.53	1.15	1.52		
Dolomite		-0.94	0.48	2.26			1.3	2.56	3.33		
Siderite		1.31	1.95	2.72			1.28	1.8	2.38		
Rhodochrosite		0.11	0.74	1.5			0.54	1.06	1.59		
Strontianite		-1.82	-1.11	-0.23			-0.86	-0.23	0.13		
Barite		0.09	0.47	0.94	< 0.0		-0.98	-0.63	0.03	< 0.0	
Hydroxyapatite		-1.61	1.31	4.75			2.03	4.26	5.94		
SiO ₂ amorphous		-0.74	-0.43	-0.02			-0.5	-0.19	0.26		
Al(OH) ₃ amorphous		-1.14	-0.84	-0.61			-1.46	-1.34	0.46		
Vivianite		2.12	3.64	5.31			1.59	2.58	7.28		
Alunite		-0.75	0.04	0.64			-4.66	-4.21	2.84		
Gypsum		-2.37	-1.99	-1.51			-3.11	-2.75	-2.05

Fig. 6 Water quality changes in a target aquifer upon injection of membrane concentrate, as modeled with PHREEQC-2. Breakthrough curves at 20 m distance from injection well (Q_{in} 75 m³/h, aquifer thickness 30 m, radial flow pattern; 1 pore volume = 6.3 days). Mineral precipitation and dissolution were not considered in this simulation. C = concentration at 20m distance from injection well; C_0 = concentration in native groundwater; SI = saturation index for mineral phases

were tolerated for all minerals, i.e., mineral precipitation and dissolution were not allowed for. Clearly, during injection, the water quality simply evolves from the native water type to the injected water type, while the SI values at the start and end of the simulation correspond to those listed in Table 3. The temporary drop in SI for some minerals (e.g., calcite, hydroxyapatite) results from the retardation of cations by ion exchange and, only to a minor extent from the mixing

of two water types by dispersion. For example, the dip in SI in calcite resulted mainly from the retardation of injected Ca by cation exchange and not from mixing.

As mentioned, mineral precipitation and dissolution were not considered in the above example. When accounting for these processes in a model simulation, it is crucial to have an understanding of the kinetics involved. Simple equilibrium modeling would result in an erroneous, rapid dissolution and/or precipitation of

Fig. 7 Schematic of the aquifer system at the Noord-Bergum BWRO pilot, the observed chloride depth profile, and the position of the projected pumping and injection wells. Aquifer 1A = glacial and fluvial, medium fine sand of Pleistocene age; Aquifer 1B = fluvial, coarse grained sand of Pleistocene age; Aquifer 2 = fluvial, coarse grained sand of early Pleistocene age, with clay intercalations

Table 4 Mineral saturation indices (SIs) of the membrane concentrate at Noord-Bergum and Zevenbergen BWRO pilot plants. RO recovery of 50%. SIs were calculated with PHREEQC-2

Mineral saturation index (SI _M)	Noord-Bergum	Zevenbergen
Calcite	1	1
Dolomite	1.2	1.1
Siderite	2	1.2
Quartz	1.2	1
SiO ₂ amorphous	-0.2	-0.3
Hydroxyapatite	3.5	2.9
Vivianite	3.6	0.8
Barite	0	-1.9

large quantities of minerals. While information on precipitation and dissolution kinetics is available for some minerals like calcite (Plummer et al. 1978) and amorphous silica (Rimstidt and Barnes 1980), it is lacking for minerals like vivianite and hydroxyapatite. In addition, there is no precise understanding of SI thresholds for dissolution and precipitation in different water types. This lack of understanding of processes acting upon injection of supersaturated concentrate reveals the urgent need for BWRO pilot studies.

Noord-Bergum

Well field Noord-Bergum has a permit for a total abstraction of 20 Mm³/a, but due to salinization abstraction has decreased from 20 Mm³/a in the 1990s to 7 Mm³/a today (2009). The cause of salinization is upconing in combination with lateral flow of brackish groundwater originating from the Holocene transgression close to the northwestern border of the well field (Fig. 1). As was discussed already, this type of Holocene transgression water is in principle suitable as BWRO source water, by

virtue of its low scaling potential and moderate chloride levels. Water company Vitens has plans to add a BWRO unit to the salinized parts of the well field, yielding an additional 3 Mm³/a of drinking water. It is expected that this unit will scavenge the advancing brackish groundwater and thereby safeguard the still operating fresh wells.

A demonstration BWRO project was scheduled to start at Noord-Bergum in the summer of 2009. Together with the pilot at Zevenbergen (see below), this will be the first BWRO system operating in the Netherlands. Technical issues to be addressed, besides the RO operation, are the production of a stable brackish source water, clogging of the injection well and target aquifer, and the quality changes in the target aquifer. In addition, this pilot will constitute the first field application of the fresh-keeper approach. If successful and still competitive with alternative solutions, the demonstration project at Noord-Bergum will be extended to a full-scale plant.

The demonstration project will consist of a fresh water abstraction well (screened at 66–86 m b.s.l.), a brackish abstraction well (133–148 m b.s.l.), an injection well (172–193 m b.s.l.), and four observation wells at 12, 25, 43, and 100 m distance from the injection well. The situation is schematized in Fig. 7. The abstraction wells will both operate at 50 m³/h, rendering an injection rate of approximately 25 m³/h. As a result of simultaneous abstraction and injection, the hydraulic head difference between the source and target aquifer will increase, which could provoke leakage between the two aquifers. Preliminary calculations, however, indicate that this risk is limited, mainly because of the mild abstraction and injection rates and the flow resistance of the 10-m-thick aquitard.

Preliminary calculations with PHREEQC-2 indicate that a RO efficiency of 50% can be reached, while

Fig. 8 Schematic of the aquifer system at the Zevenbergen BWRO pilot, the observed chloride depth profile, and the position of the projected pumping and injection wells. Aquifer 2 = fluvial, coarse grained sands of early Pleistocene age; Aquifer 3 = marine, fine grained sands of late Tertiary and early Pleistocene age, with clay intercalations

maintaining undersaturation for most minerals. Still, potentially problematic SIs (>0.3) were calculated for some minerals, as indicated in Table 4. Precipitation of quartz and dolomite is not expected, as precipitation is generally slow for quartz and because supersaturation of dolomite has often been reported without dolomite being present in the system. Precipitation of calcite, siderite, hydroxy-apatite and vivianite could be an issue and will be addressed in the pilot study. During injection, cation exchange and mixing with the ambient groundwater are expected to have only a minor influence on the SIs.

During the 1-year pilot, clogging of the injection well, hydraulic heads and water quality changes in the source and target aquifers will be monitored closely. A lab experiment using cores from the target aquifer will run parallel to the field experiment, in order to obtain more detailed insight in water quality changes and potential mineral precipitation and dissolution processes.

Zevenbergen

The BWRO pilot at Zevenbergen (Brabant Water) was issued to gain first, hands-on experience with deep well injection of membrane concentrate. When proven successful, the BWRO concept may be applied to well fields that currently suffer from upconing of brackish water (and thus need protection of the remaining fresh wells).

The brackish water source at Zevenbergen originates from the Holocene transgression, and like the Noord-Bergum pilot, a RO efficiency of 50% can be reached with acceptable mineral saturation levels (Table 4). Water will be abstracted from 100–115 meter depth (50 m³/h) and injected in Aquifer 3 between 160 and 185 m BSL (Fig. 8). Observation wells are situated at 25 and 80 m distance from the injection well, and will be used to monitor water quality and hydraulic heads. At present, only limited information is available on the hydrogeology at Zevenbergen, yet it is expected that the 30 m thick aquitard will prevent upward leakage of the water injected.

The focus of this pilot is on the operational issues of membrane fouling and well clogging and on energy consumption of the BWRO plant. The pilot was intended to start in summer 2009 and will run for a total period of 4 years.

Conclusions

BWRO is an interesting "source-treatment" option in temperate humid climates, where water scarcity problems are mild but environmental issues are of big concern. The disposal of the RO waste water in an environmentally acceptable way is the weakness of BWRO. In the Netherlands, deep well injection is the best disposal option and a holistic approach is advocated to reach a high-quality waste water that perfectly fits into the natural chemical environment of the target aquifer. The approach consists of a cautious selection of the brackish source

groundwater and target aquifer for disposal of membrane concentrate, a low RO efficiency of around 50%, keeping the system anoxic and pressurized, and avoiding the use of antiscalants. Preferably, the BWRO source water should: have moderate chloride levels ($\text{Cl}^- < 10,000$ mg/l), for optimum RO purification; be low in HCO_3^- , SO_4 , Ca, Mg, Fe, Mn, NH_4 , PO_4 , SiO_2 , and Ba, to reduce risks on membrane and injection well scaling; not produce permeate that exceeds permissible drinking water levels for constituents like boron; and produce concentrate water with salinity, nutrient and heavy metal levels that do not exceed ambient levels in the target aquifer.

To test this approach, two BWRO pilot studies were scheduled to start in the summer of 2009: at Noord-Bergum, one of the largest well fields in the Netherlands, which suffers from severe salinization problems, and at Zevenbergen. These pilots will provide the first practical experience with the BWRO concept in the Netherlands. The most important technical issues to be addressed are the pumping of a stable brackish source water, the performance of the RO system, the long-term quality changes of the membrane concentrate in the target aquifer and the relation between mineral supersaturation, membrane fouling and clogging of the injection well. For this, it is crucial to develop a better understanding of the kinetics of mineral precipitation on the membranes, in the injection wells and in the target aquifer.

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