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D22.3 Assessment reversed osmosis membrane clogging by varying redox conditions of feedwater

Part 1: Site characterization and ASR performance

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SUMMARY

Coastal areas are generally densely populated and marked by high freshwater demands. Due to the proximity of the sea these areas have to deal with saline water intrusions and salinization of groundwater. Therefore the availability of freshwater cannot always be guaranteed in these regions. Use of local eco-systems by aquifer storage and recovery (ASR) of temporary freshwater surpluses and reverse osmosis (RO) of brackish-saline groundwater are potential solutions for freshwater supply in coastal areas. Both techniques have their drawbacks. ASR in coastal aquifers is marked by freshwater losses by buoyancy effects in the saline groundwater, while RO is accompanied by a saline waste water stream. In DESSIN we aim to demonstrate that a sustainable and reliable freshwater supply can be achieved by combining both techniques in one system (ASRRO). In this report we discuss the potential increase in freshwater recovery by deploying multiple partially penetrating wells (MPPW), a Freshkeeper, and a combination of ASR and RO (ASRRO).

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List of Acronyms and Abbreviations

<i>ASR</i>	<i>Aquifer storage and recovery</i>
<i>ATES</i>	<i>Aquifer thermal energy storage</i>
<i>EC</i>	<i>Electrical conductivity</i>
<i>ICP-OES</i>	<i>Inductively-Coupled Plasma – Optic Emission Spectrometry</i>
<i>RO</i>	<i>Reverse osmosis</i>
<i>RE</i>	<i>Recovery Efficiency</i>
<i>K</i>	<i>Hydraulic conductivity</i>
<i>m-ASL</i>	<i>meters above sea level</i>
<i>m-BSL</i>	<i>meters below sea level</i>
<i>MPPW</i>	<i>Multiple partially penetrating wells</i>
<i>MW</i>	<i>Monitoring well</i>
<i>n</i>	<i>Porosity</i>
<i>VANI</i>	<i>Vertical anisotropy ratio</i>

Executive summary

Coastal areas are generally densely populated and marked by high freshwater demands. Due to the proximity of the sea these areas have to deal with saline water intrusions and salinization of groundwater. Therefore the availability of freshwater cannot always be guaranteed in these regions. Use of local eco-systems by aquifer storage and recovery (ASR) of temporary freshwater surpluses and reverse osmosis (RO) of brackish-saline groundwater are potential solutions for freshwater supply in coastal areas. Both techniques have their drawbacks. ASR in coastal aquifers is marked by freshwater losses by buoyancy effects in the saline groundwater, while RO is accompanied by a saline waste water stream. In DESSIN we aim to demonstrate that a sustainable and reliable freshwater supply can be achieved by combining both techniques in one system (ASRRO). In this report we discuss the potential increase in freshwater recovery by deploying multiple partially penetrating wells (MPPW), a Freshkeeper, and a combination of ASR and RO (ASRRO).

At the Westland field site, the freshwater surplus of 270,000 m³ of greenhouse roof is injected deep into a target aquifer (23-37 m below sea level; 3700 – 4700 mg Cl/l) through two dedicated, recently developed MPPWs. After storage, the water is recovered in the growing season in spring and summer. Unmixed freshwater is recovered at the aquifer top for direct use as high-quality irrigation water. During recovery, the deep wells of the system is used as 'Freshkeepers' for interception of brackish-saline groundwater. This water is directly re-injected in a deeper aquifer (2014) or desalinated by RO for use as irrigation water (ASRRO).

In the first 1,5 years of operation (December 2012 – July 2014), approximately 20% of the injected roofwater was recovered practically unmixed and could be used directly for crop breeding as high-quality irrigation water. Based on the hydrochemical monitoring and groundwater transport modelling, it was found that a deeper borehole of a close by aquifer thermal energy storage (ATES) well caused leakage of deeper saltwater, contaminating the water recovered by the ASR system. The installed Freshkeeper proved to be indispensable to attain the recovery efficiencies (REs) achieved. Despite the leakage of deeper saltwater, the dedicated Westland ASR system proves to be effective to abstract different water qualities separately and attain a significantly better ASR-performance than a conventional system would achieve.

When the performance of the Westland ASR-system was modelled for a case in which the (presumably exceptional) leakage via the ATES borehole was absent, a maximum freshwater recovery was realized by the shallowest wells of the MPPWs, while the deeper wells were gradually intercepting more-and-more brackish-saline groundwater to prevent salinization. The recovery of practically unmixed freshwater for direct use was increased from around 30% (simple well and MPPW) to 50% (Freshkeeper applied) in Cycle 4. Desalination of the intercepted brackish water would lift the RE to 60%.

The modelling of less suitable conditions by assuming a thicker target aquifer indicated that especially in the first cycles, the RE of practically unmixed water will be lower. Cycle-after-cycle, REs will

improve, however, yet requiring abstraction of more brackish/saline groundwater. Likewise for conventional ASR, the highest REs can be attained while storing large volumes in aquifers with relatively low salinities, limited thickness, and relatively low hydraulic conductivities. Under less suitable conditions, more desalination is required to recover the injected freshwater.

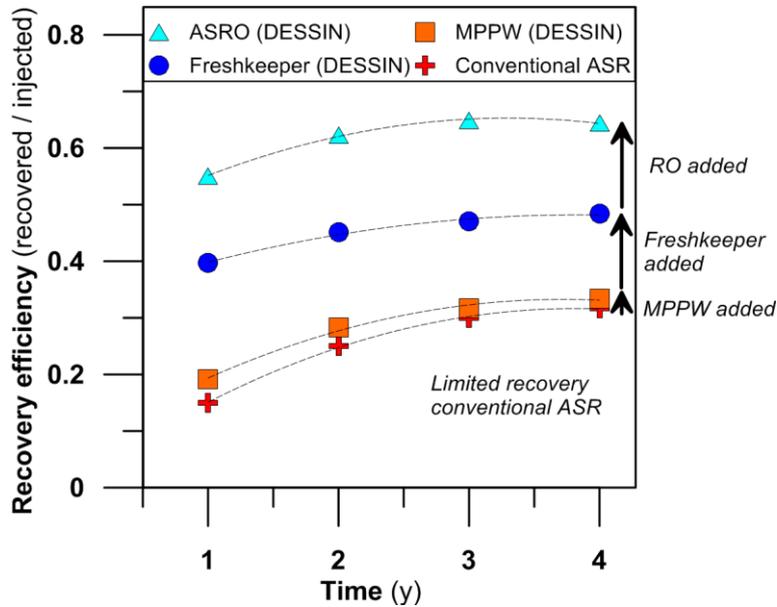


Figure 1: Most relevant results of the SEAWAT modeling study: an increase in recovery efficiencies at the Westland target aquifer thanks to the introduction of the MPPW, the Freshkeeper, and ASRRO.

1.1 Westland: horticultural capital of The Netherlands

The Westland area in The Netherlands (Figure 2) is the Dutch largest intensive greenhouse horticultural area, justifying its second name ‘the glass city’. Glasshouses cover about 2,500 ha of this 10,000 ha large municipality (population: 104,000 inhabitants). For this reason, the horticultural sector including related companies/suppliers are a very relevant contribution to the local and even national economy.

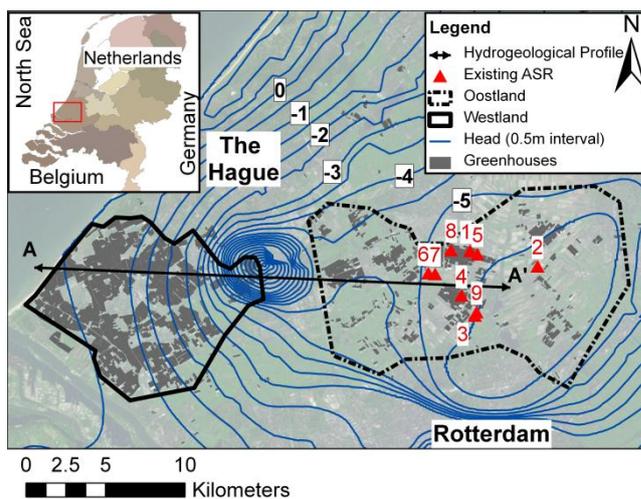


Figure 2: Location of the Westland and neighbouring Oostland greenhouse area.

1.2 The need for additional freshwater in summer

The salinity requirements of the irrigation water in this area (generally measured using electrical conductivity, EC) are exceptionally strict. Drinking water (with sodium concentrations of approximately 50 – 100 mg Na/l) is already too saline for many of the crops (merely tomatoes, cucumbers, peppers) and flowers cultivated. Low salinities allow greenhouse owners to reuse drained water from artificial substrates multiple times, without reaching critical sodium concentrations. Fresh irrigation water supply is realized primarily by storing low-EC rainwater from greenhouse roofs in basins or tanks, complemented by the use of surface water in periods of low salinity and by desalination of brackish groundwater (Stuyfzand and Raat, 2010).

A mismatch in precipitation and water demand creates a large winter freshwater surplus, which is discharged to sea, as only a small part can be stored in basins or tanks. Surface water is generally unsuitable as a source of irrigation water during summer droughts, as they are fed by brackish seepage water (de Louw et al., 2010) and contain too high concentrations of sodium. Fresh surface water can be brought in from major rivers, but the inlets suffer increasingly from salinization caused by seawater intrusion during summer droughts, which is exacerbated by sea level rise (Barends et

al., 1995; Kooi, 2000; Kwadijk et al., 2010; Oude Essink et al., 2010; Post, 2003; Schothorst, 1977). Wintertime precipitation is expected to increase, whereas summer droughts may become more intense and prolonged, (Intergovernmental Panel on Climate Change (IPCC), 2007; Royal Netherlands Meteorological Institute, 2014). Freshwater availability for irrigation during summer will likely be reduced due to the changing temporal precipitation distribution in combination with a predicted rise in temperature. Up to now, desalination by reverse osmosis is the only proven technology to ensure additional freshwater supply. Major disadvantages of this technique are the high energy consumption, the required maintenance, and especially the disposal of leftover concentrate in deeper aquifers. Discharge of this concentrate to sewage systems or surface waters is not allowed and its disposal in deeper aquifers can conflict with the goals set in the EU Water Framework Directive.

1.3 Aquifer storage and recovery (ASR) of freshwater in a brackish/saline aquifer as a sustainable but yet too vulnerable source

A more sustainable use of the precipitation surplus collected by greenhouse roofs will improve freshwater availability in the area. ASR is a cost-effective, readily applicable technique to store large water volumes, without the need for large surface areas. In the study area, ASR has been applied on a small scale since the 1980s in the upper, relatively shallow aquifer (10 - 50 m below surface level (m-BSL)), which is the thinnest and least saline aquifer found in the area. The performance of ASR (i.e., the percentage of freshwater that can be recovered upon storage) using this target aquifer, even though it is the least saline aquifer available, is limited especially in the Westland area (Zuurbier et al., 2013). The main cause for the reduced performance are the buoyancy effects induced by the difference in density of the native groundwater (high density), and the injected freshwater (low density), which leads to early salinization at the bottom of the ASR well (Figure 3).

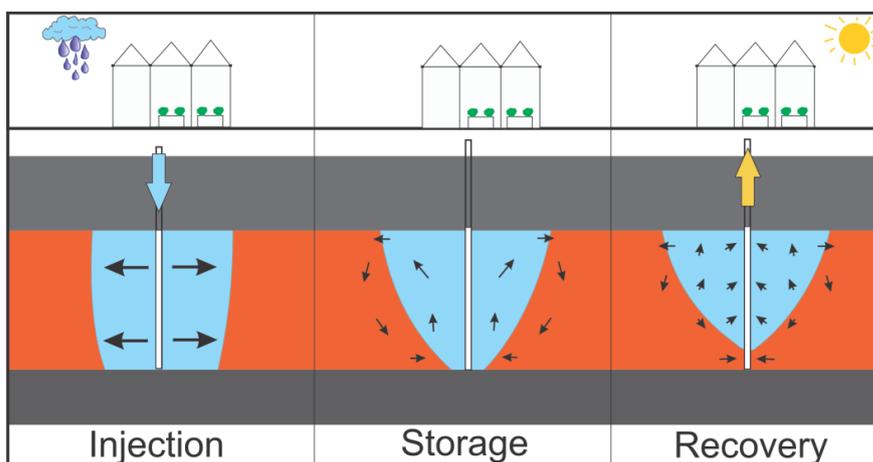


Figure 3: Freshwater loss during ASR in brackish and saltwater aquifers due to buoyancy effects.

1.4 Aquifer storage and recovery combined with reverse osmosis (ASRRO) to provide a robust and sustainable freshwater source

An innovative ASR solution, combined with a Freshkeeper and RO (ASRRO), is proposed to maximize the recovery of injected freshwater surpluses. Multiple partially penetrating wells (MPPW) allow for deep injection and shallow abstraction, postponing the salinization during recovery to attain higher recovery efficiencies. By simultaneously abstracting upper fresh and lower brackish groundwater, salinization of the fresh water well is prevented even longer (Figure 4). The abstracted brackish water is used as additional and reliable freshwater source after desalination. The hybrid ASR/desalination (ASRRO: aquifer storage and recovery and reverse osmosis) system thus combines the best of two techniques and it contributes to optimal durable use of ‘free’ natural sources as rainwater and soil, saving expensive aboveground space and mitigating salinization. The potential is high in coastal areas facing water shortages for drinking water, agricultural, and industrial applications and/or salinization.

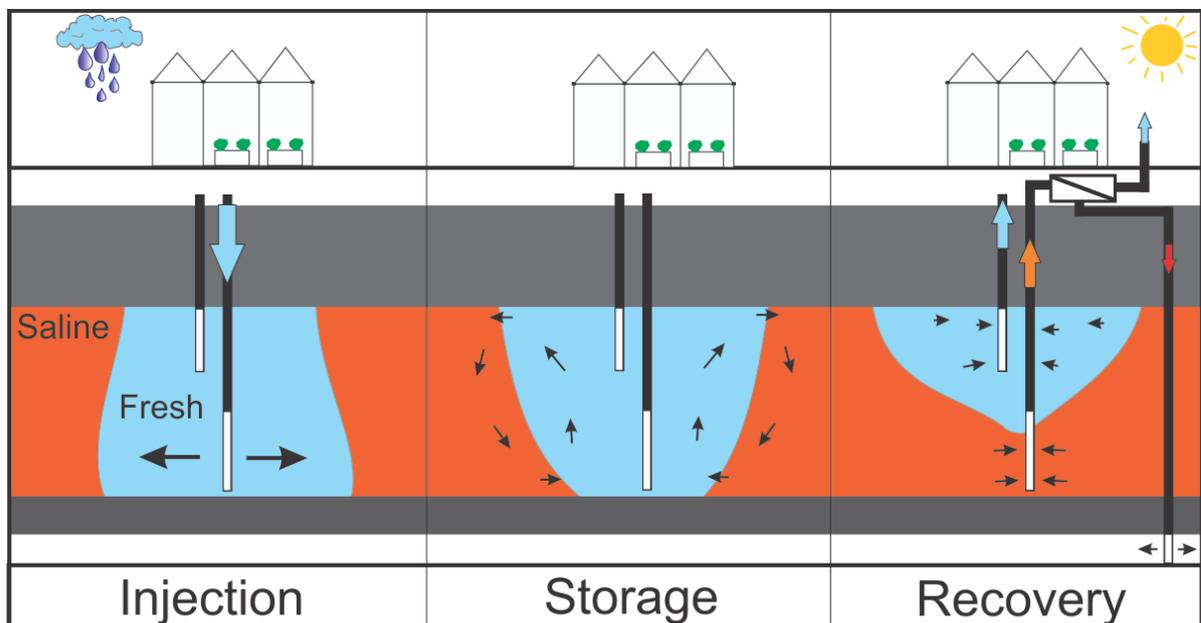


Figure 4: The introduction of the MPPW for deep injection and shallow recovery in combination with a Freshkeeper and optional RO-treatment for a maximal recovery of freshwater (ASRRO).

1.5 Research aims during first application of ASRRO

The task descriptions and accompanying research aims reported in this report are listed in Table 1.

Table 1: Tasks within WP22.2

<u>Task</u>	<u>Task description</u>	<u>Research aim</u>	<u>Time</u>
22.1	Quantification of the freshwater recovery increase by an innovative well design: In this task the freshwater recovery increase by Multiple Partially Penetrating Wells (MPPW), injection/recovery schemes, and the use of the Freshkeeper at the base of the freshwater bubble is quantified.	To assess the optimal well configuration and potential increase in freshwater recovery in the Westland case and in differing hydrogeological settings.	M1-12 (Part I)
22.2	Assessment of membrane clogging by varying redox conditions of the feedwater. Reversed Osmosis (RO) membrane clogging due to varying redox conditions of the feedwater from Freshkeeper is quantified and potential in-situ (e.g., subsurface iron removal) and ex-situ (e.g., pre-treatment of membrane feedwater) techniques to prevent membrane clogging are evaluated.	To quantify and cope with potential negative effects on the RO-feedwater quality induced by introduction of oxic rainwater in the anoxic, saline target aquifer.	M1-24 (Part II)

2 Research approach and methods

2.1 General approach/methodology

In order to complete the defined tasks, a multiphase approach with specific methodologies was set up. These approaches and methodologies are listed in Table 2 and visualized in Figure 5.

Table 2: The approaches and methodologies applied to complete the defined tasks.

Task	Task description	Approach	Methodology
22.1 (Part I)	Quantification of the freshwater recovery increase by an innovative well design	<ol style="list-style-type: none"> 1. Field testing ASR-cycle 2012/2013: use of MPPW only; 2. Field testing ASR-cycle 2013/2014: addition of the Freshkeeper (no RO); 3. Modelling the performance of a conventional (fully-penetrating) ASR-well instead of a MPPW; 4. Modelling and evaluation of the MPPW-benefits in various hydrogeological settings. 	<ol style="list-style-type: none"> 1. Recording of injected/recovered volumes and EC; 2. Lab analysis on (ground)water samples; 3. SEAWAT groundwater transport modelling; 4. SEAWAT groundwater transport modelling.
22.2 (Part II)	Assessment of membrane clogging by varying redox conditions of the feedwater. Reversed Osmosis (RO) membrane clogging due to varying redox conditions of the feedwater from Freshkeeper is quantified and potential in-situ (e.g., subsurface iron removal) and ex-situ (e.g., pre-treatment of membrane feedwater) techniques to prevent membrane clogging are evaluated. (Bruine de Bruin, KWR, M1-24)	Field testing of the Freshkeeper including desalination of saltwater recovered by the Freshkeeper to increase freshwater production	M1-24

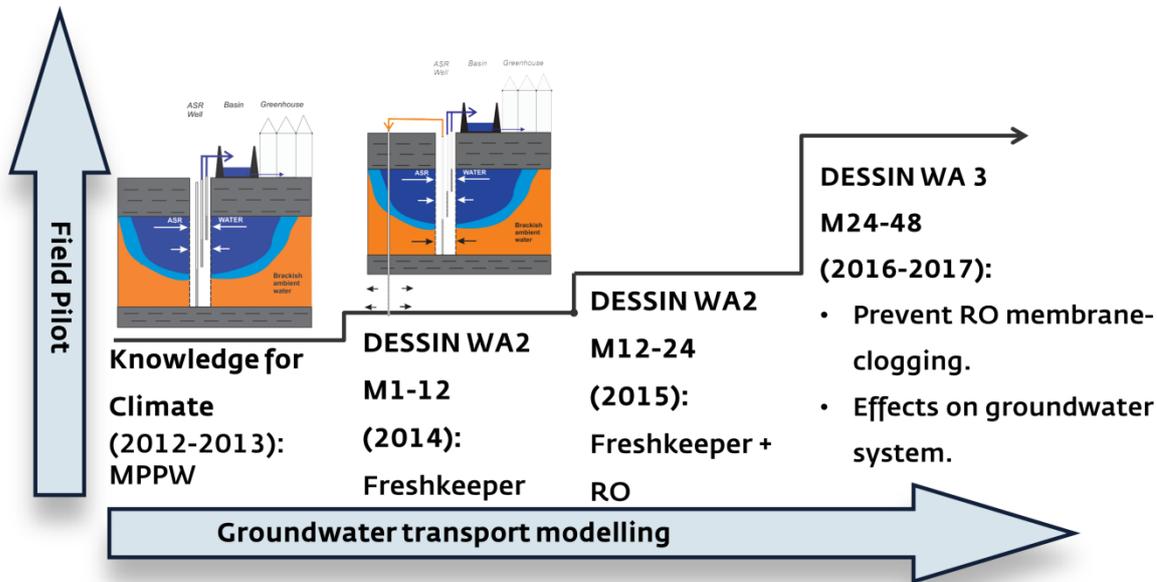


Figure 5: Visualization of the approach and methodologies applied and the Westland ASRRO study

2.2 Westland ASRRO field pilot

A major part of the ASRRO study takes place at the world's first ASRRO-system ever built. This pilot system was initially funded by Knowledge for Climate national research program to test the performance of ASR using MPPWs in coastal (brackish, saline) aquifers. Within the DESSIN project, the ASR-system was stepwise converted to an ASRRO-system. The system is realized at a cluster of tomato growers with a total greenhouse area of 270.000 m²).

2.2.1 Set-up Westland ASR system and hydrogeological setting

The Westland ASRRO system is installed to inject the rainwater surplus of 270,000 m² of greenhouse roof in a local shallow aquifer (23 to 37 m-below sea level (m-BSL); surface level = 0.5 m-above sea level (m-ASL)) for recovery in times of demand. For this purpose, two multiple partially penetrating wells (MPPWs) were installed, Figure 6), so that water could be injected preferably at the aquifer base, and recovered at the aquifer's top in order to increase the recovery efficiency (Zuurbier et al., 2014). All ASR wells (AW1 and AW2, installed in 2012) and the nearby aquifer thermal energy storage (ATES) well (K3-a (abandoned) and K-3b (active), Figure 7) were installed using reverse-circulation rotary drilling, while the monitoring wells (MW1-5, Figure 6) were installed using bailer drillings. Bentonite clay was applied to seal the ASR wells (type: Micolite300) and ATES well K3 (Micolite000 and Micolite300). The ASR wells used a 3.2 m high standpipe to provide injection pressure, whereas the ATES well used a pump to meet the designed injection rate of 75 m³/h. Optionally, water abstracted by the ASR-system can be injected in Aquifer 2 via a disposal well, which is installed approximately 250 m downstream from the ASR-site in Aquifer 2.

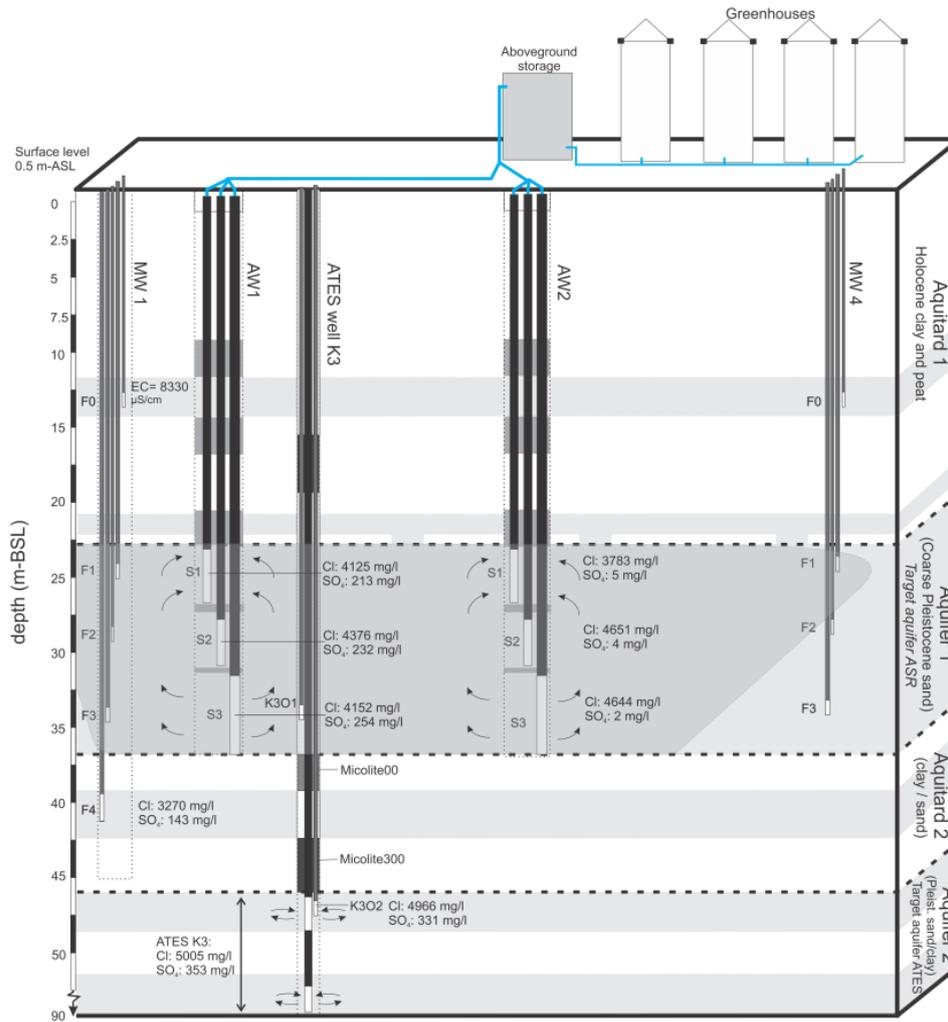


Figure 6: Cross-section of the Westland ASR-pilot, including the ambient groundwater quality observed preceding the ASR operation.

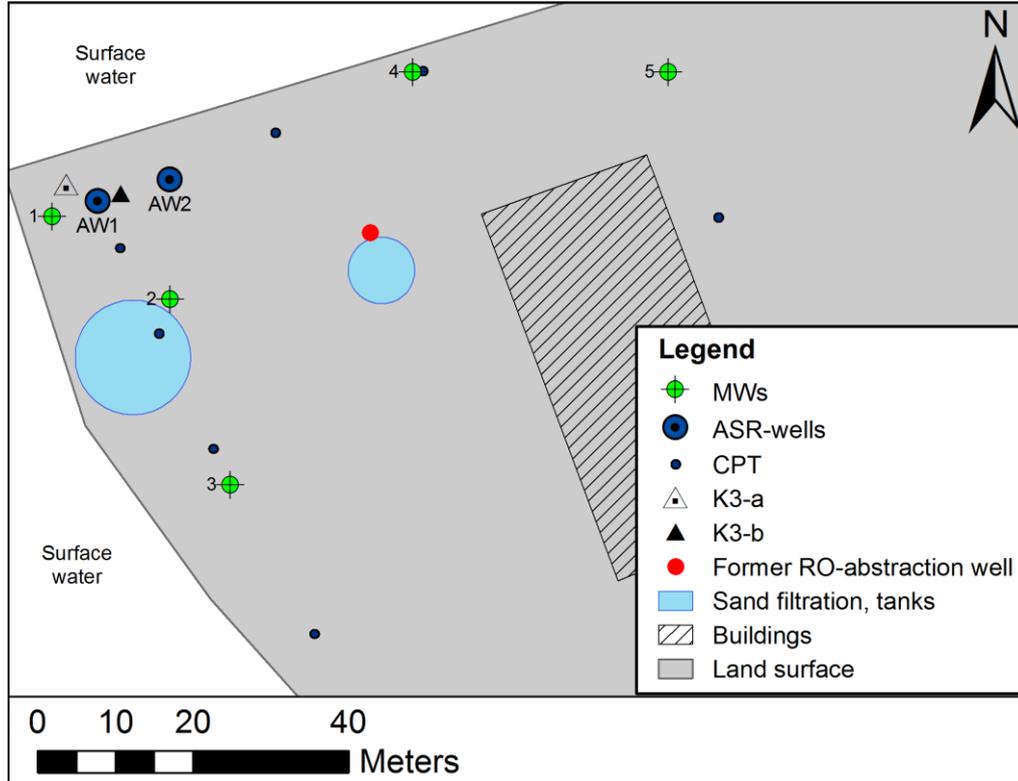


Figure 7: Topview of the Westland ASR-pilot.

The target aquifer for ASR (Aquifer 1) is 14 m thick and consists of coarse fluvial sands (average grain size: 400 μm) with a hydraulic conductivity (K) of 30-100 m/d, which was derived from the head response in MW1 and MW2 upon pumping. The groundwater is typically brackish, with Cl concentrations ranging 3,793 to 4,651 mg/l in Aquifer 1 and approximately 5,000 mg/l in Aquifer 2. A fine sand layer in Aquitard 2 contains remnant fresher water (Cl = 3,270 mg/l). SO_4 was a useful tracer to separate the brackish water in Aquifer 1 and 2, as it is typically virtually absent in Aquifer 1 (presumably younger groundwater, infiltrated when the Holocene cover was already thick, which caused SO_4 -reduction), whereas it is high in Aquifer 2: 300 to 400 mg/l SO_4 (older, infiltrated through a thinner clay cover which limited SO_4 -reduction, see Stuyfzand (1993) for more details). The lateral displacement of the groundwater based on regional hydraulic heads is limited to only a few m per year (Zuurbier et al., 2013).

The target water quality during recovery is again rainwater (low salinity, $\text{Na} < 0.5 \text{ mmol/l}$), which means that the water should be recovered by the ASR-system practically unmixed.

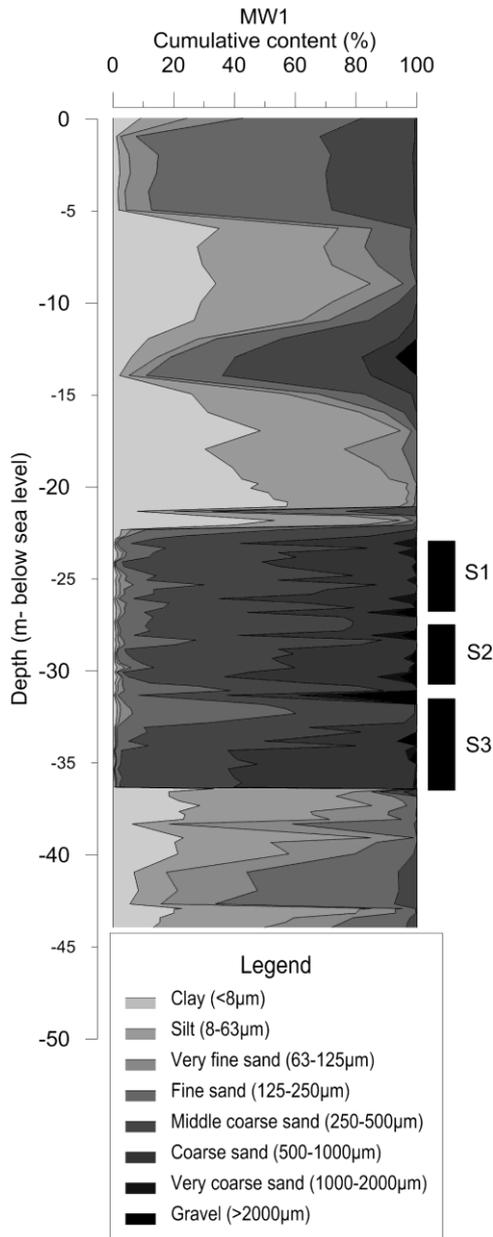


Figure 8: Cumulative grain size contents observed at MW1 (at 5 m from ASR well 1). S1-S3 mark the depth intervals of the ASR well screens.

2.3 Monitoring during Westland ASR cycle testing

All ASR and monitoring well screens were sampled prior to ASR operation (November and December, 2012). MW1 and 2 were sampled with a high frequency during the first breakthrough of the injection water at MW1 (December 2012, January 2013), while all wells were sampled on a monthly basis until March 2014. Three times the volume of the well casing was removed prior to sampling. The injection water was sampled regularly during injection phases. All samples were analyzed in the field in a flow-through cell for EC (GMH 3410, Greisinger, Germany), pH and temperature (Hanna 9126, Hanna

Instruments, USA), and dissolved oxygen (Odeon Optod, Neotek-Ponsel, France). Samples for alkalinity determination within one day after sampling on the Titrilab 840 (Radiometer Analytical, France) were stored in a 250 ml container. Samples for further hydrochemical analysis were passed over a 0.45 µm cellulose acetate membrane (Whatman FP-30, UK) in the field and stored in two 10-ml plastic vials, of which one was acidified with 100 µl 65% HNO₃ (Suprapur, Merck International) for analysis of cations (Na, K, Ca, Mg, Mn, Fe, S, Si, P, and trace elements) using ICP-OES (Varian 730-ES ICP OES, Agilent Technologies, U.S.A.). The second 10 ml vial was used for analysis of F, Cl, NO₂, Br, NO₃, PO₄, and SO₄ using the Dionex DX-120 IC (Thermo Fischer Scientific Inc., USA), and NH₄ using the LabMedics Aquakem 250 (Stockport, UK). All samples were cooled to 4 °C and stored dark immediately after sampling.

CTD-divers (supplied by Schlumberger Water Services, Delft, The Netherlands) were used for electronic recording of conductivity, temperature, and pressure in the target aquifer at MW1 and MW2. Calibrated, electronic water meters were coupled to the programmable logic controller (PLC) of the ASR-system to record its operation per well screen.

2.4 Westland ASR groundwater transport model (Set up)

SEAWAT Version 4 (Langevin et al., 2007) was used to simulate the ASR operation. A half-domain was modelled in 3-D to reduce computer runtimes. Equal constant heads were imposed at two edges of the aquifers, the top of the model (controlled by drainage systems) and at the base of the model (controlled by deeper aquifers). No-flow boundaries were given to the other two side faces of the model. Initial Cl-concentrations were based on the results of the reference groundwater sampling at MW1. SO₄-concentrations in Aquifer 1 were based on MW2, since these concentrations were more representative for the field site. For Aquifer 2, the concentrations found at ATES well K3 (bulk) and the observation well K3O1 were used (see Figure 6). The density of the groundwater was based on the Cl-concentration using:

$$\rho_w = 1000 + 0.00134 \times Cl(mg/l)$$

A longitudinal dispersivity of 0.1 m was derived from the freshwater breakthrough at MW1 and was applied to the whole model domain. Constant heads were based on the local drainage level (top model layer) the (higher) observed heads in the aquifer (which indicated seepage), and the regional hydraulic gradient.

The ASR operation was modelled with a properly sealed and an unsealed ATES borehole, since unreliable sealing of the ATES borehole was expected. In the latter case, a conductivity of 1000 m/d (a realistic K as apart from filter sand around the well screen, the borehole was backfilled with gravel with a grain size of 2-5mm) was given to the cells (1.0 x 1.0m) in Aquifer 1, Aquitard 2, and Aquifer 2 at the location of this pumping well to force borehole leakage.

Table 3: Hydrogeological properties of the Geological Layers in the Westland SEAWAT model

Geological Layer	Model layers	Base (m-BSL)	K_h (m/d)	VANI (K_h/K_v)	S_s (m^{-1})	n (-)	Initial C (mg/l Cl)	Initial C (mg/l SO_4)
Aquitard 1	6	22.3	0.2 - 1	100	10^{-4}	0.2	2000-3000	4
Aquifer 1	12	33.7	35	1	10^{-7}	0.3	4000-4800	4
	3	36.4	100					
Aquitard 2 (clay-sand)	8	47.5	0.05-10	1-10	10^{-4}	0.2-0.3	3200	160
Aquifer 2	6	96	12	1	10^{-6}	0.3	4100-7900	331-375

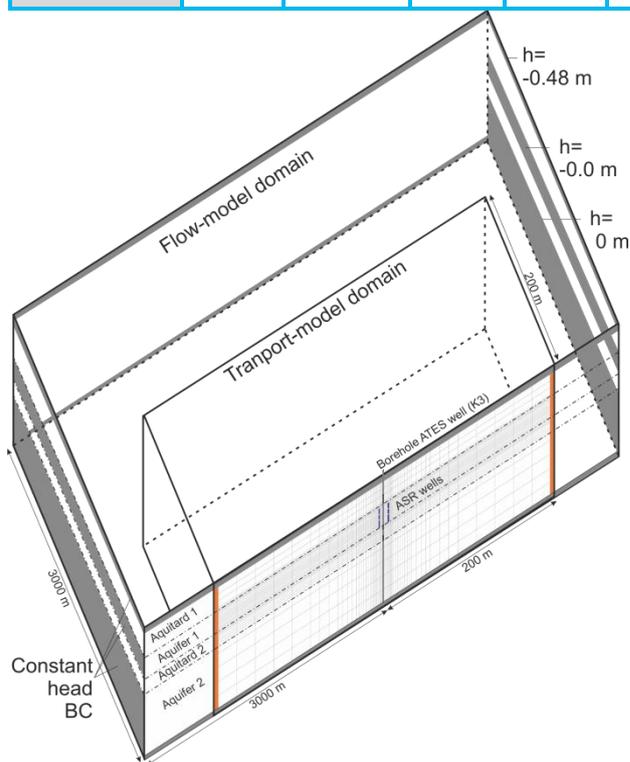


Figure 9: Set-up of the Westland ASR groundwater transport model (half-domain).

3.1 Cycle 1 (2012/2013): testing of the MPPW-ASR system, identification of ATES borehole leakage

Cycle 1 contained the injection of around 13,175 m³ of freshwater in December/January 2012/2013, followed by a first attempt to recover unmixed injection water. Despite the improved design of the ASR system with the MPPW, a rapid and severe salinization of even shallow recovery wells was found already during the first days of recovery (Figure 10). No more than 8% of the injected water could be recovered with a satisfying quality. Remarkably, the salinization at the ASR well 1 (AW1) preceded salinization of the monitoring wells situated further from the ASR well (MW1, MW2), indicating that this salinization was not caused by buoyancy effects in the target aquifer, which would have led to salinization of monitoring wells away from the ASR-wells first. Additionally, high SO₄-concentrations (up to >50 mg/l) in the recovered water were found, which could not be explained by the a SO₄ increase attained by pyrite oxidation by oxygen observed in the injected water (max. SO₄-enrichment <15 mg/l). The remaining source of contamination based on the hydrochemistry was therefore the deeper saltwater from Aquifer 2.

The SEAWAT-model underlined that tilting of the interfaces by buoyancy effects did not lead to the early salinization observed. Additionally, it is shown in Figure 11 by the model that an increase in SO₄-concentrations was not predicted during this cycle, or even when the recovery period was extended (results not shown). Note that SO₄-production by pyrite (FeS₂) oxidation was neglected in the model, but that SO₄-concentrations >15 mg/l could not be explained by this process, however. When the leaky borehole was incorporated in the model (by assigning K=1000 m/d in a 1 x 1 m column at the location of the current ATES well), it was able to introduce the early recovery of deep (SO₄-rich) saltwater (Figure 11). Other scenarios were found unable to improve the simulation of the observed SO₄-trends. These scenario's included: leakage via other ATES-wells further from the ASR wells (at 5 m; too late arrival of SO₄), a high-K borehole (2000 m/d; too early arrival, contamination flux too high), a low-K borehole (500 m/d; too late arrival, flux too low), a vertical anisotropy (K_h/K_z = 2; too early arrival, too high flux), and neglecting the ATES operation (i.e., not incorporating the deep cold water abstraction in Aquifer 2: SO₄-flux too high).

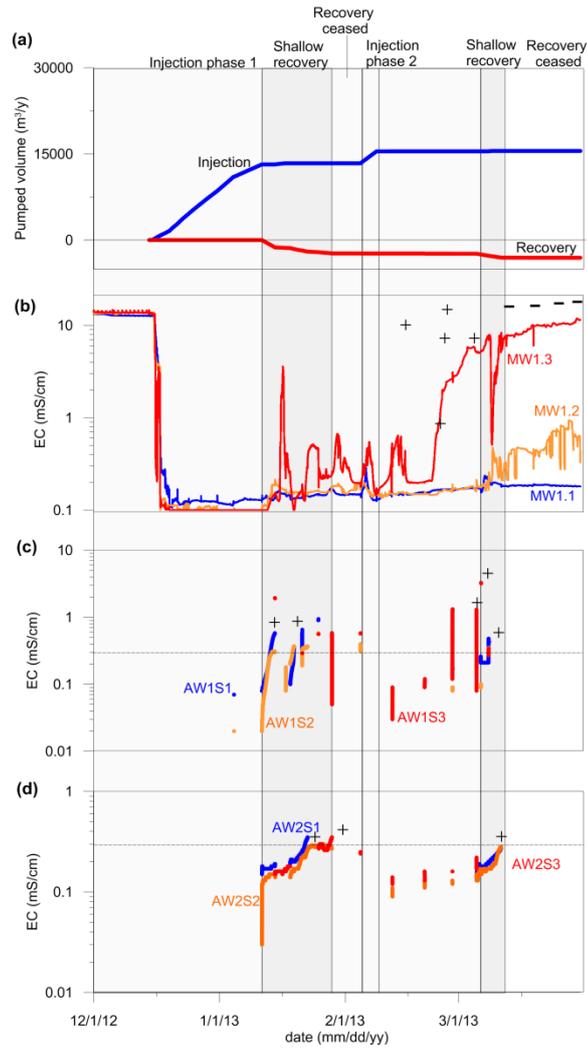


Figure 10: Pumping of the ASR system during cycle 1 in 2012/2013 (a), EC observations at MW1 at 5 m from AW1 (b), and the EC in the recovered water at AW1 (c) and AW2 (d).

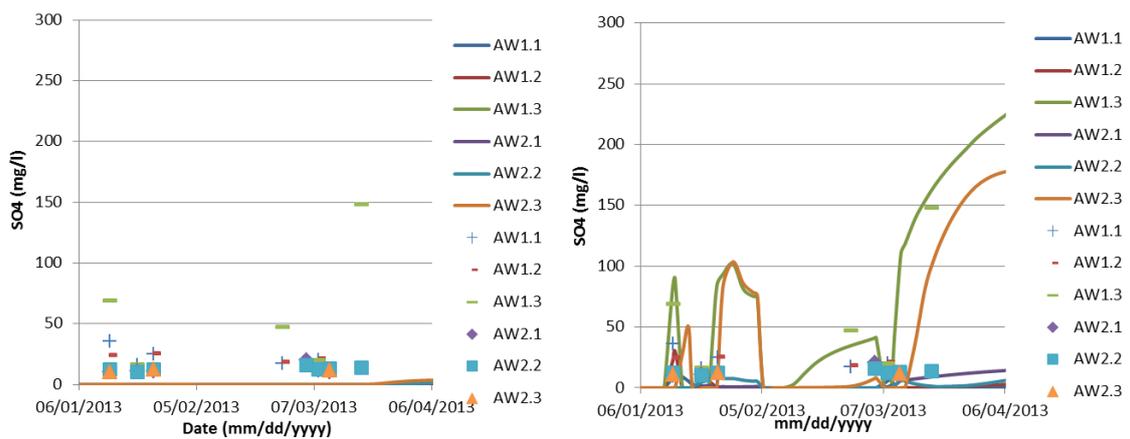


Figure 11: Modelled (solid lines) and observed (data points) SO_4 -concentrations. High concentrations indicate admixing of deeper saltwater. Left = no borehole leakage, right = borehole leakage via a 1x1 m borehole with $K=1000$ m/d).

Based on the hydrochemical observations and model outcomes of Cycle 1 it was concluded that:

- The source of the early salinization is the intrusion of deeper saltwater from Aquifer 2;
- Considering the lithology, thickness, and continuity of Aquitard 2 (confirmed by grain size analyses and cone penetrating tests), leakage via natural pathways through this confining layer was unlikely;
- The nearby ATES boreholes provide more presumable pathways. According to the rate and sequence of salinization, the leakage was situated close to AW1 (ATES well K3: installed in 2006 and replaced in 2008; both boreholes are situated approximately 3 m from AW1 and >7 m from AW2).

3.2 Cycle 2 (2013/2014): improving the ASR operation by addition of the Freshkeeper well

Prior to Cycle 2, the borehole of the abandoned ATES well K3 (replaced in 2008 after signs of short-circuiting and situated approximately 5 m from ASR well AW1) was sealed by injection of Dämmer (Heidelberg Cement, Germany) at the interval from 52 to 36 m-BSL. The current ATES well K3 (situated between ASR well AW1 and AW2) was left unaltered as it was still in operation. Cycle 2 started with the injection of 66,178 m³ of rainwater using both ASR wells between September 2013 and March 2014, which was followed by recovery at AW2 (start: March 5, 2014). Despite sealing of the abandoned K3 borehole, a rapid salinization by SO₄-rich saltwater was again observed (Figure 12) and the recovery was terminated after 26 days (March 21, 2014) after recovering no more than 2,500 m³ (<4%). This time, a monitoring well present in the gravel pack of the ATES K3 well (coded K3O1; a 1m-well screen at 33 m-BSL) was sampled and equipped with a CTD-diver, unraveling high ECs and presence of SO₄-rich saltwater from the deeper aquifer (Figure 12). This presence of intruding deep saltwater was also found at MW1S3 (5m from the ASR wells) as a consequence of re-injecting part of the abstracted freshwater at deeper intervals, which caused lateral displacement of earlier intruded saltwater.

In order to enable recovery of freshwater, K3O1 was equipped with a small pump (1.4 m³/h) to intercept intruding saltwater, while also the deeper wells of the ASR-system (AW1S3 and AW2S3) were transformed to scavenger wells (“Freshkeepers”), abstracting the intruding saltwater and injecting this in the deep disposal well in Aquifer 2 at 250 m distance from the ASR-site. This way, the ASR-system could again attain an acceptable water quality (practically unmixed rainwater) at AW2S1 and AW1S2 (from April 15 onwards). As a consequence, the deeper segments of the target aquifer (S3 levels, Figure 12bcd) first freshened, followed by again salinization as recovery proceeded. After recovery of in total 12,324 m³ of practically unmixed rainwater (18.6% of the injected water), the recovery had to be ceased due to the slightly increased salinity. During this last salinization, the water at the deeper (S3-)levels of the target aquifer at AW1, MW1, and MW2 was free of SO₄, indicating ‘normal’ salinization by saltwater from Aquifer 1, caused by buoyancy effects. High SO₄ - concentrations were only found close to the currently operating K3 ATES well (at AW1 and K3O1).

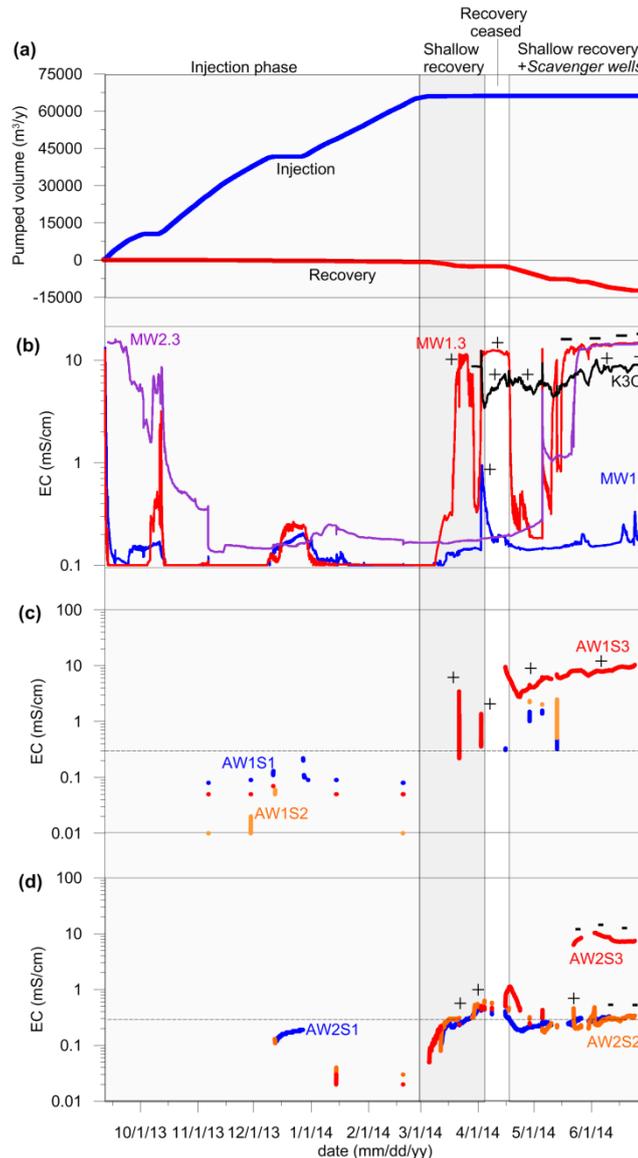


Figure 12: Pumping of the ASR system during Cycle 2 in 2013/2014 (a), EC observations at MW1 at 5 m from AW1 (b), and the EC in the recovered water at AW1 (c) and AW2 (d). AW2.1 and AW2.3 were used for freshwater recovery (12,324 m³). Presence of increased SO₄-concentrations (deep saltwater from Aquifer 2) are marked by '+', while its absence is marked by '-' (indicating shallow saltwater from Aquifer 1).

The modelling results of cycle 2 underline the continuing leakage via the borehole of ATEs well K3. The SEAWAT model was again able to simulate the water quality trends regarding SO₄ and Cl (Figure 13 and Figure 14). Remaining deviations in observed concentration were attributed to uncertainties in the model input, mainly aquifer heterogeneity and potentially disturbing abstractions and injections in the surroundings, mainly for ATEs and brackish water reverse osmosis, the latter abstracting in Aquifer 1 and injecting in Aquifer 2.

In Cycle 2 it was also demonstrated that salinization during recovery was independent of the injected freshwater volume: salinization occurred after recovery of a similar volume as in Cycle 1. Modelling of Cycle 2 revealed that this is caused by the fact that injected freshwater will not reach deep into the deeper saline aquifers since the hydraulic head in the freshwater in the leaky ATEs borehole during injection is more or less equal to the freshwater head in the deeper saltwater aquifer. In other words: it is hard to push freshwater through the ATEs borehole into the deeper aquifer. The freshwater that may reach the deeper aquifer is rapidly displaced laterally as a result of buoyancy effects (Figure 15). A significant head difference ($\Delta h(\text{fresh}) = 0.3 \text{ m}$ to 0.65 m) can be observed during recovery, however, and in combination with the high permeability of the ATEs borehole, this results in a significant intrusion of deeper (SO_4 -rich) saltwater. Even during storage phases, a freshwater head difference ($\Delta h(\text{fresh}) = 0.15 \text{ m}$) was observed as a consequence of the replacement of saltwater by freshwater, causing intrusion (yet with a lower rate) of deep saltwater. Seepage induced by this pressure difference is of course hampered by the low permeability of the aquitard in a 'normal situation'. A continuous aquitard is therefore indispensable for the success of ASR in such a setting, where intrusion of deeper saltwater is not permitted.

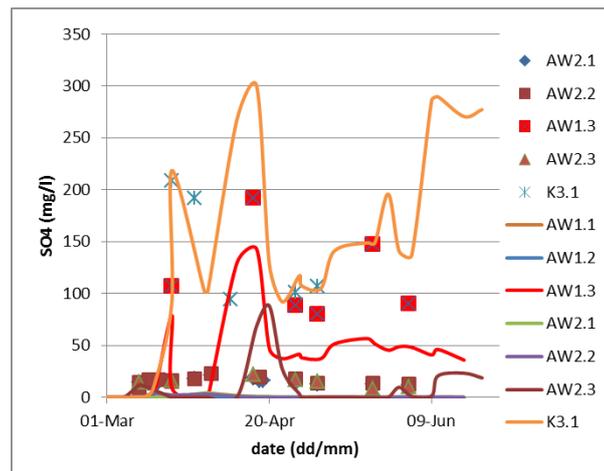


Figure 13: Modelled and observed SO_4 -concentrations at the most relevant well screens.

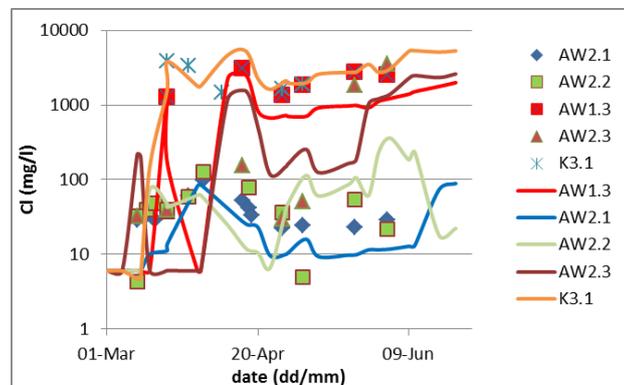


Figure 14: Modelled and observed Cl -concentrations at the most relevant well screens.

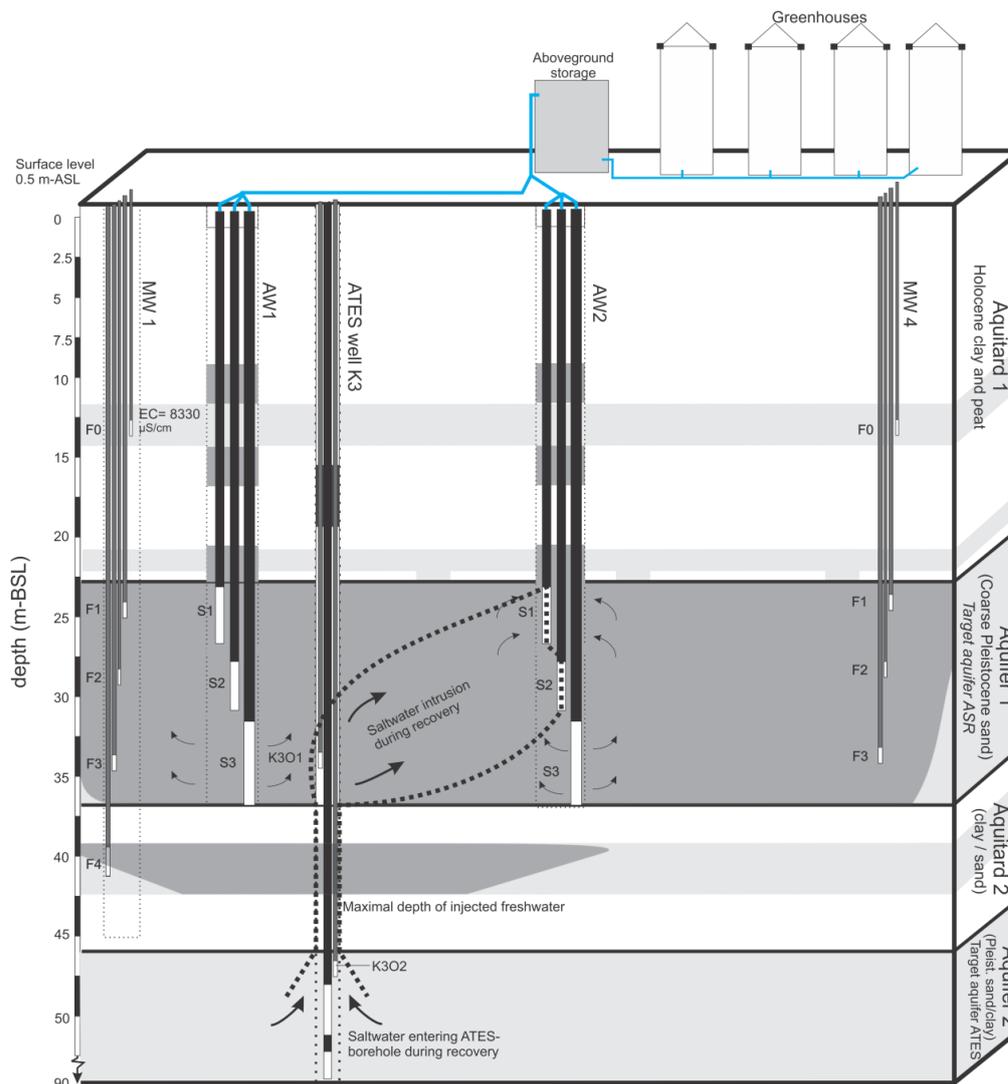


Figure 15: Deep saltwater intrusion via a borehole during recovery of injected freshwater at the Westland ASR site.

3.3 Simulation of ASR-cycles in a undisturbed subsurface or after sealing of the leaking ATEs-borehole

The collected information on the aquifer characteristics in the SEAWAT groundwater model can be used to analyze the performance of the MPPW-ASR system for a ‘normal field site’: i.e. without leakage from deeper aquifers via a perturbations, or after sealing of the perturbation. The latter was attempted on February 3 and 4, 2015 via injection of Dämmer (Heidelberg Cement, Germany). The SEAWAT model was used to simulate three ASR-cycles with the following representative characteristics for the Westland site (Zuurbier et al., 2012):

Table 4: Set-up of the modelled, representative ASR-cycle for the Westland subsurface without short-circuiting of deeper saltwater.

Stage	Duration	Pump discharge
Infiltration	120 days	60,000 / 120 = 500 m ³ /d
Storage	30 days	0 m ³ /d
Recovery	120 days	-60,000 / 120 = -500 m ³ /d
Idle	65 days	0 m ³ /d

During the 120 days of recovery it was aimed to recover as much of freshwater (marked by Cl <50 mg/l) as possible. Three strategies were applied with equal abstraction rates for both AW1 and AW2.

Table 5: Modelled recovery efficiencies at the Westland ASR site using different pumping strategies. The relative discharge per MPPW-screen is given for each particular screen.

Strategy	Distribution pumping rate	Efficiency results	Intercepted brackish water (via deep (S3-)wells)
Conventional ASR-well	In: 100% via one fully penetrating well Out: 100% via one fully penetrating well	Year 1: 15% Year 2: 25% Year 3: 30% Year 4: 32%	
Deep injection, shallow recovery	In: 10/20/70% (year 1) In : 0/20/80% (year 2-3) Abstract: 60/40/0% (year 1-3)	Year 1: 19% Year 2: 29% Year 3: 32% Year 4: 33%	
+ 'freshkeeper'	In: 10/20/70% (Year 1) In : 0/20/80% (Year 2) Abstract: Decreasing from 60/40/0% to 60/0/0% (Year 1-3) Intercept Freshkeeper: increasing from 100 to 500 m ³ /d	Year 1: 40% (55%) Year 2: 46% (62%) Year 3: 47% (65%) Year 4: 48% (64%)	Year 1: 18,500 m ³ Year 2: 20,500 m ³ Year 3: 21,500 m ³ Year 4: 19,300 m ³

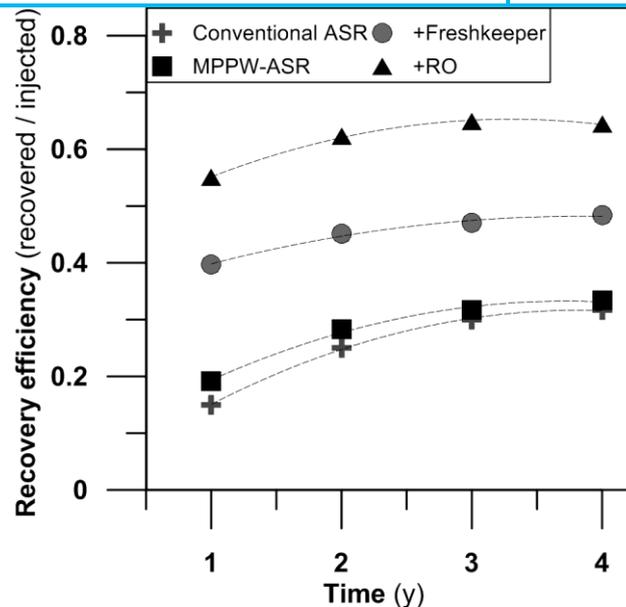


Figure 16: Recovery efficiencies at the Westland ASR site without the borehole leakage resulting from the SEAWAT groundwater transport model for a conventional ASR well (one well screen, fully penetrating), multiple partially penetrating wells without a ‘Freshkeeper’ (scenario MPPW-ASR), for a MPPW in combination with a ‘Freshkeeper’ (scenario +Freshkeeper), and for a scenario in which RO is applied on the intercepted brackish water to produce additional freshwater (50% of the flux).

Recovery with conventional ASR wells will be limited to around 30% of the injected freshwater. The use of a MPPW for deep injection and shallow recovery has a limited positive effect due to the limited thickness of the aquifer: one-third of the injected water is recovered. The extension of the length of the recovery period with the MPPW is limited since the saltwater can rapidly migrate from the base of the aquifer to the shallower recovery wells of the MPPW-system (‘upconing’).

The introduction of the Freshkeeper to protect the shallow recovery wells by interception of this deeper saltwater significantly extended the recovery period, enabling recovery of already 55% in the first year for direct use. Ultimately, this will yield recovery of approximately 50% of virtually unmixed injected freshwater in Cycle 4. In the ASRRO-concept, the intercepted brackish water is desalinated via the process of reverse osmosis, resulting in the production of freshwater (50%, supplied to end user) and concentrate (50%, reinjected in the deeper Aquifer 2).

In this case, more than 60% of freshwater was produced with respect to the injected freshwater. With such recovery efficiencies, the ASRRO can provide more than sufficient freshwater to most of the horticulturists in the Westland area (Zuurbier et al., 2013) and achieve generally accepted levels (Maliva and Missimer, 2010; Pyne, 2005). Additional freshwater can however still be produced by extending the freshwater production via RO, preferentially with water from the freshest wells of the MPPW (generally the shallowest wells) to limit the required salt removal by RO.

3.4 Performance of ASR with Freshkeeper under different hydrogeological settings

The current modelling of the conventional ASR, MPPW, Freshkeeper, and ASRRO set-ups was performed particularly for the Westland pilot site. Relevant is the translation to hydrogeologically differing target aquifers. The most relevant hydrogeological characteristics for the Westland target aquifer and generic characteristics for ASR target aquifers are given in Table 6.

Table 6: Hydrogeological characteristics of the Westland pilot target aquifer.

Hydrogeological parameter	Characteristics pilot site Westland	Values pilot site Westland	Typical values Westland area	Typical values for coastal aquifers (generic)
Thickness (H)	Relatively thin aquifer	H = 14 m	H = 10 – 40 m	1 – 100 m

Hydraulic conductivity (K)	Coarse fluvial sands	K = 30 – 100 m/d (average: K = 50 m/d)	average: K = 20 – 50 m/d	K = 1 (fine sands) – 100 m/d (coarse sands), up to 500 m/d (gravel)
Salinity (indicated by Cl-concentration)	Brackish-saline	3700 – 4700 mg Cl/l	1000 – 10,000 mg Cl/l	40 – 20,000 mg Cl/l

It can be derived that the modelled Westland target aquifer is relatively representative for coastal target aquifers. The thickness however, is in the lower range. For this reason, the thickness of the Westland target aquifer was doubled in the SEAWAT model and three cycles were run to assess whether similar recoveries could be achieved. The injection and recovery strategy was exactly similar as for the Westland pilot site. For conventional and MPPW-set-ups it was now found that recovery was severely limited to less than 5% and 21% respectively (Figure 17), which was mainly caused by the fact that with an equal tilting rate (controlled by the unaltered hydraulic conductivity and salinity), brackish water entered the bottom well earlier due to a smaller radius of the injected freshwater bubble. This is in line with previous findings that thicker aquifers will result in lower recovery efficiencies during ASR (Bakker, 2010; Ward et al., 2009).

With the Freshkeeper, the RE in Cycle 3 was already close to the RE found achievable in the Westland aquifer, although a much larger volume of brackish water needed to be intercepted to maintain recovery of practically unmixed water: the Freshkeeper well is switched on earlier. As a consequence, 75 days of recovery of unmixed freshwater were possible in Cycle 3, and 66% of freshwater would be recovered when this was combined with desalination of this brackish water (ASRRO).

Altogether, the degree of tilting (controlled by groundwater salinity and hydraulic conductivity) and the radius of the injected freshwater bubble (controlled by aquifer thickness and injected volume) will control (1) the part of injected water that can be recovered practically unmixed for direct use (especially in the first cycles) and (2) the required interception of brackish water. Low hydraulic conductivities, low salinities, and large volumes in a relatively thin aquifer are optimal for direct recovery and require less or no interception by a Freshkeeper. High hydraulic conductivities, saline aquifers, and smaller volumes in a relatively thick aquifer make the Freshkeeper indispensable to recover a significant part of the injected water.

Table 7: Modelled recovery efficiencies assuming a doubled aquifer thickness using different pumping strategies. The relative discharge per MPPW well screen is given for each particular screen.

Strategy	Distribution pumping rate	Efficiency results	Intercepted brackish water (via deep (S3-)wells)
Conventional ASR-well	In: 100% via one fully penetrating well Out: 100% via one fully penetrating well	Year 1: 2% Year 2: 4% Year 3: 5%	
Deep injection, shallow recovery	In: 10/20/70% (year 1) In : 0/20/80% (year 2-3) Abstract: 60/40/0% (year 1-3)	Year 1: 13% Year 2: 19% Year 3: 21%	

+ 'freshkeeper'	In: 10/20/70% (Year 1) In : 0/20/80% (Year 2) Abstract: Decreasing from 60/40/0% to 60/0/0% (Year 1-3) Intercept Freshkeeper: increasing from 100 to 500 m ³ /d	Year 1: 26% (40%) Year 2: 37% (54%) Year 3: 42% (66%)	Year 1: 16,400 m ³ Year 2: 20,900 m ³ Year 3: 29,500 m ³
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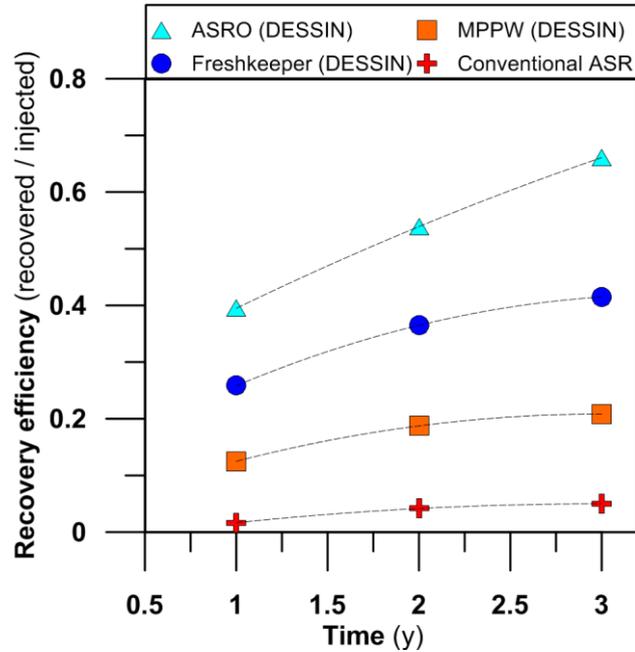


Figure 17: Recovery efficiencies at the Westland ASR site assuming a doubled target aquifer thickness resulting from the SEAWAT groundwater transport model for a conventional ASR well (one well screen, fully penetrating), multiple partially penetrating wells without a 'Freshkeeper' (scenario MPPW), for a MPPW in combination with a 'Freshkeeper' (scenario Freshkeeper), and for a scenario in which RO is applied on the intercepted brackish water to produce additional freshwater (50% of the flux).

4 DISCUSSION AND CONCLUSIONS

4.1 ASR-performance at the Westland field site

In the Dutch horticulture area Westland, a highly-advanced ASR-system was installed in brackish (3700 – 4700 mg Cl/l) coastal aquifer (coarse sands, 14 m thick), where buoyancy effects normally lead to low recovery efficiencies (REs), making the aquifer storage rather inefficient. The Westland ASR-system, however, was equipped with multiple partially penetrating wells (MPPWs) to enlarge the flexibility of injection and recovery. By the enlarged flexibility it was aimed to recover a significant part of the injected fresh rainwater via deep injection, shallow recovery, and interception of deep brackish-saline groundwater to prevent contamination of shallow wells.

In the first 1,5 years of operation (December 2012 – July 2014), approximately 20% of the injected water was recovered practically unmixed. Based on the hydrochemical monitoring and groundwater transport modelling, it was found that a deeper borehole of a close by ATES well (realized before the start of the pilot) caused leakage of deeper saltwater, contaminating the water recovered by the ASR system. The installed Freshkeeper proved to be indispensable to attain the RE achieved. Despite the leakage of deeper saltwater, the Westland ASR-system proves to be effective to abstract different water qualities separately and attain a significantly better ASR-performance than a conventional system would achieve.

4.2 Modelled performance of advanced recovery using a Freshkeeper in combination with RO (ASRRO)

When the performance of the Westland ASR-system was modelled for a case in which the leakage via the ATES borehole was absent or technically sealed, the ultimate performance of the ASR-system at the Westland site could be derived. In this case, a maximum freshwater recovery was realized by the shallowest wells of the MPPWs, while the deeper wells were gradually intercepting more-and-more brackish-saline groundwater to prevent salinization. The recovery of practically unmixed freshwater for direct use was increased from around 30% (simple well and MPPW) to 50% (Freshkeeper applied) in Cycle 4. Desalination of the intercepted brackish water would lift the RE to 60%.

The modelling of less suitable conditions by assuming a thicker target aquifer indicated that especially in the first cycles, the RE of practically unmixed water will be lower. Cycle-after-cycle, REs will improve, however, although this does require abstraction of more brackish/saline groundwater. Likewise for conventional ASR, the highest recoveries of unmixed waters can be attained while storing large volumes in aquifers with relatively low salinities, limited thickness, and relatively low hydraulic conductivities.

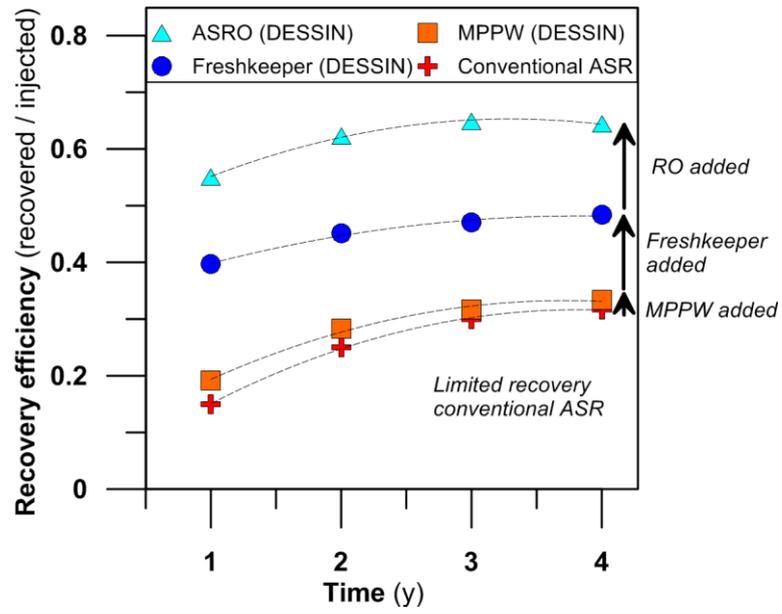


Figure 18: Most relevant results of the SEAWAT modelling study: an increase in recovery efficiencies at the Westland target aquifer thanks to the introduction of the MPPW, the Freshkeeper, and ASRRO.

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