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D33.1: Valorisation and demonstration of an ASRRO application in a field application

Westland demo site 2014 - 2017

KWR Watercycle Research Institute, July 2017

The research leading to these results has received funding from the European Union Seventh Framework Programme (FP7/2007-2013) under grant agreement no. 619039

Title of the report

D33.1:VALORISATION AND DEMONSTRATION OF AN ASRRO APPLICATION IN A FIELD APPLICATION
Westland demo site 2014-2017

SUMMARY

ASRRO is a combination of aquifer storage and recovery and reverse osmosis. The results at the demonstration site Westland (2013-2017) indicate that ASRRO is technically viable and beneficial. The biggest operational threat during ASRRO in a sand aquifer is clogging of RO-membranes and potentially also of the saline water re-injection well(s). This is caused by mobilization of clay particles (during freshening) and formation of Fe-colloids (by infiltration of oxic water in an area with adsorbed Fe around the ASRRO wells), both in the infiltration stage. Abstraction of brackish water in deeper sections of the aquifer and regular flushing of the RO-membranes are viable methods to overcome these operational threats.

The impact of widespread use of ASRRO on the regional Westland groundwater system was limited based on regional groundwater modelling. It was shown that ASRRO decreased the chloride concentration with respect to the autonomous scenario and with respect to the current use of brackish water reverse osmosis (BWRO). ASRRO was successful in mitigating the local negative impact (saltwater plume formation) caused by the deep disposal of membrane concentrate during BWRO and significantly reduced the potential saltwater intrusion that was found in the BWRO case. Albeit more expensive, the use of ASRRO is considered competitive with the current BWRO.

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List of Acronyms and Abbreviations

ASR	*Aquifer storage and recovery*
RO	*Reverse osmosis*
ASRRO	*ASR and RO combined in one integrated system*
RE	*Recovery Efficiency*
K	*Hydraulic conductivity*
n	*Porosity*
ASRRO	*Aquifer Storage and recovery and Reverse Osmosis*
BWRO	*Brackish Water Reverse Osmosis*
MFI	*Modified Fouling Index (also: membrane fouling index*
SDI_{15}	*Silt Density Index*
m-ASL	*meters above sea level*
m-BSL	*meters below sea level*
MPPW	*Multiple partially penetrating wells*
MW	*Monitoring well*
ATES	*Aquifer thermal energy storage*
EC	*Electrical conductivity*
ICP-OES	*Inductively-Coupled Plasma – Optic Emission Spectrometry*
ΔP	*Pressure difference (between feed side and reject side of RO-feed channel*
RO-recovery	*Part of the water feeding an RO-treatment that is transformed to freshwater*

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Executive summary

The Westland ASRRO (aquifer storage and recovery in combination with reverse osmosis) demo site is situated in the western coastal zone of the Netherlands, which is marked by presence of brackish to saline groundwater almost up to surface levels. At surface level, greenhouses dominate the landscape, which require rainwater quality (low salinities) for irrigation. Rainwater from greenhouse roofs is therefore used as the main irrigation water source. Challenging is the storage of temporary rainwater surpluses for use in subsequent droughts. As a consequence, supplementary high-quality water is now produced by using brackish water reverse osmosis (BWRO) in combination with concentrate disposal in deep aquifer, resulting in a net freshwater mining of the groundwater system.

Aquifer storage and recovery (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 sea level (m-BSL)), which is the thinnest and least saline aquifer found in the area. Even though it is the least saline aquifer available, the performance of ASR (i.e., the percentage of freshwater that can be recovered upon storage) in this target aquifer is limited, especially in the Westland area. The main causes for the reduced performance are the buoyancy effects induced by the difference in density between the native groundwater (high density), and the injected freshwater (low density), which leads to early salinization at the bottom of the ASR well.

An innovative ASR solution, combined with a Freshkeeper and RO, 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. The abstracted brackish water is used as additional and reliable freshwater source after desalination. The hybrid aquifer storage and recovery and reverse osmosis (ASRRO) system thus combines the best of two techniques and it contributes to optimal durable use of ‘free’ natural sources as (rain)water 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

The objectives at the Westland ASRRO demo site defined at the start of the DESSIN project were:

- To quantify freshwater recovery by an ASR well design.
- To demonstrate the added value of an ASR/RO system on freshwater recovery.
- To demonstrate the effect of enhanced subsurface iron removal on membrane clogging.
- To demonstrate the impact of freshwater supply from brackish aquifers on regional groundwater quality and Water Framework Directive goals.
- To evaluate innovative solutions to enhance freshwater supply from brackish aquifers.

The Westland ASR system is installed to inject the rainwater surplus of 270,000 m^2 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)) with negligible lateral displacement for recovery in times of demand. For this purpose, two multiple partially penetrating wells (MPPW) were installed, such that water can be injected preferably at the aquifer base, and recovered at the aquifer top in order to increase the recovery. Rainwater can be pre-treated and injected with a total rate of 40 m^3/h, and recovered with a total maximum rate of 50 m^3/h.

The results at the demo site indicate that ASRRO is technically viable and beneficial. Freshwater surpluses up to 70 000 m^3/ 6 months could be treated, stored, and partially recovered for direct use (22.5% of the stored water). Additional freshwater could be produced by abstracting the mixed freshwater and saline water and subsequently treating this with RO. This created a high-quality freshwater stream and a waste stream with a quality similar to the native groundwater in a deeper more saline aquifer. Infiltration of rainwater from greenhouse roofs upon sand filtration could be done virtually within legal standards, except for Zn (zinc).

The biggest operational threat (besides the common operational threat using normal brackish water RO) during ASRRO in a sand aquifer (as present at the Westland site) is clogging of RO-membranes and potentially also the saline water re-injection well(s). This can be caused by mobilization of clay particles (during freshening) and formation of Fe-colloids (by infiltration of oxic water in an area with adsorbed Fe around the ASRRO wells), both in the infiltration stage. Abstraction of brackish water in deeper sections of the aquifer and regular flushing of the RO-membranes are viable methods to overcome these operational threats.

The impact of widespread use of ASRRO on the regional Westland groundwater system was considered limited based on regional groundwater modelling. It was shown that ASRRO decreased the chloride concentration with respect to the autonomous scenario and with respect to the current use of brackish water reverse osmosis (BWRO). ASRRO was successfull in mitigating the local negative impact (saltwater plume formation) caused by the deep disposal of membrane concentrate during BWRO and significantly reduced the potential saltwater intrusion that was found in the BWRO case.

Based on this case study, an overall positive to neutral impact of ASRRO on a coastal groundwater system is presumed, which is an improvement with respect to the use of BWRO in the same setting. ASRRO thus provides means to more sustainable use of coastal groundwater systems. However, several operational (e.g. infiltrated and recovered volumes) and hydrogeological (e.g., aquifers, aquitards, drainage levels, nearby abstractions) controlling factors will affect the overall and their cumulative impact on any groundwater system and should be considered before ASRRO implementation elsewhere.

Albeit more expensive, the use of ASRRO is considered competitive with the current BWRO. The cost price per m^3 is 0.06 eur/m^3 higher (0.70 versus 0.64/m^3) as a result of higher CAPEX. Both alternatives are economically more interesting than aboveground storage (in basins).

1 Introduction

1.1 The Westland ASRRO demo site

In 2013, a one year aquifer storage and recovery (ASR) pilot was initiated in the Westland area in The Netherlands. In the DESSIN project, the pilot was prolonged and extended with a Freshkeeper and RO-system in order to create an ASRRO system and enable a robust and sustainable freshwater supply. Between 2014 en 2017, the performance and the impact of the ASRRO system was studied in detail. The findings are presented in this report.

The Westland ASRRO site is situated in the western coastal zone of the Netherlands, which is marked by presence of brackish to saline groundwater almost up to surface levels (Figure 1). At surface level, greenhouses dominate the landscape, which require rainwater quality (low salinities) for irrigation. Rainwater from greenhouse roofs is therefore used as the main irrigation water source. Challenging is the storage of temporary rainwater surpluses for later droughts. As a consequence, supplementary high-quality water is now produced using brackish water reverse osmosis (BWRO) in combination with concentrate disposal in deep aquifer, resulting in a net freshwater mining in the groundwater system.

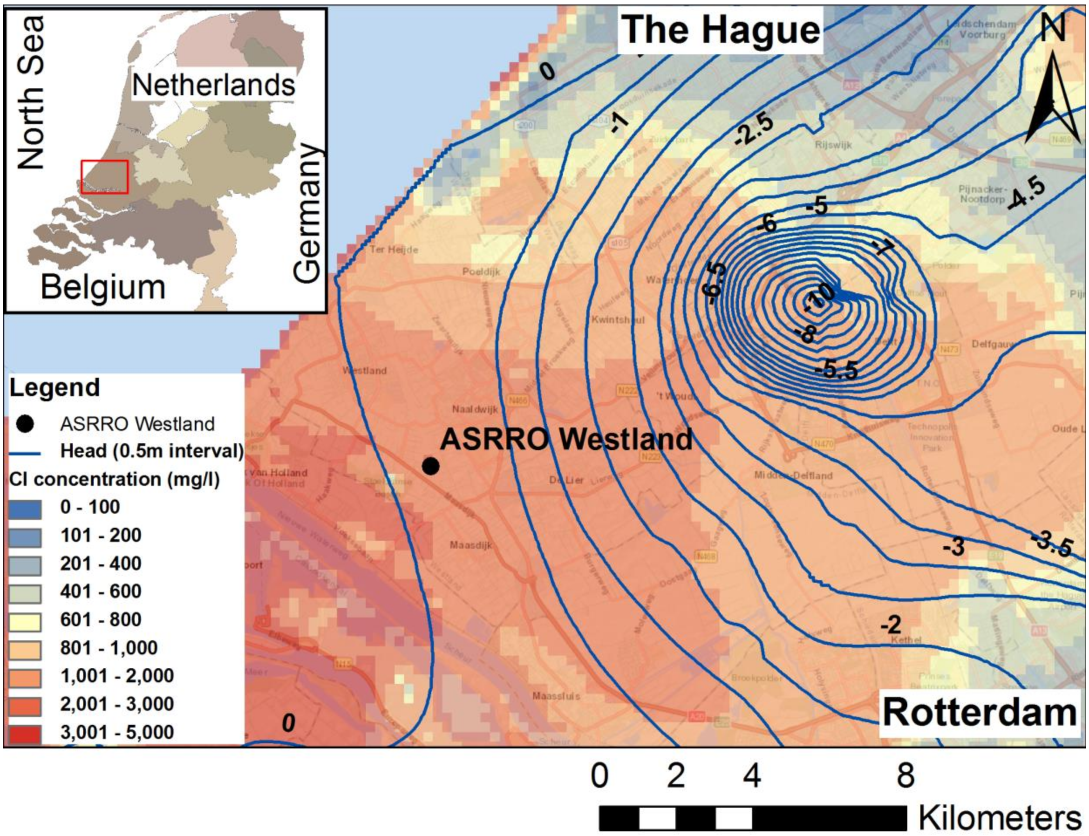


Figure 1: Location of the Westland ASRRO site.

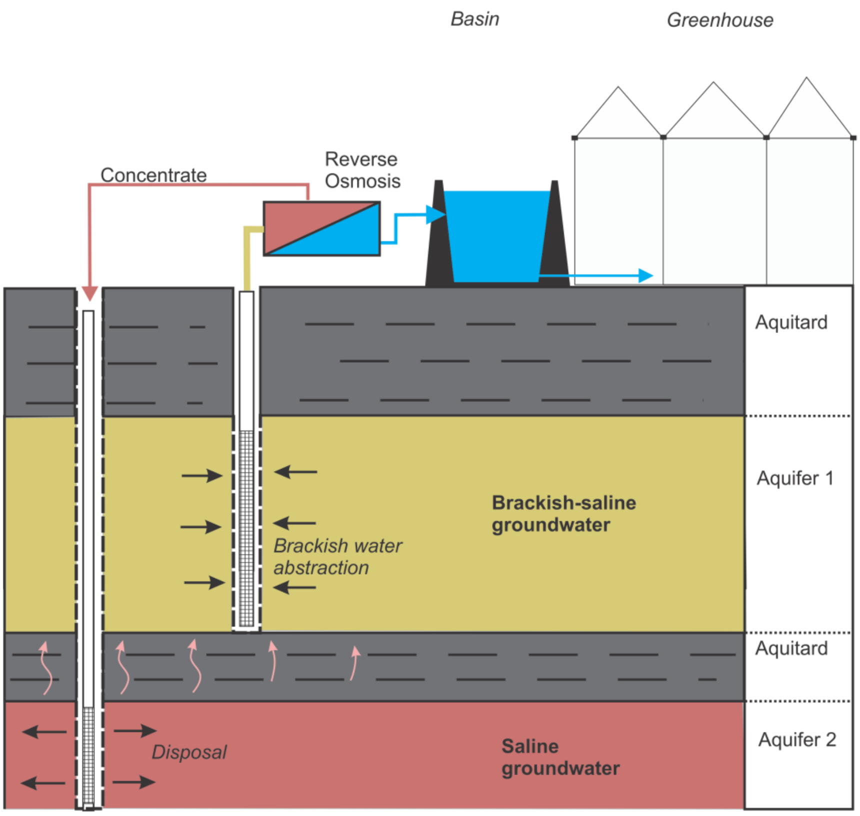


Figure 2: Use of brackish water reverse osmosis (BWRO) accompanied by concentrate disposal in the area.

1.2 Aquifer storage and recovery (ASR) as a sustainable but yet too vulnerable freshwater source via ecosystem services

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 sea 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 causes 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).

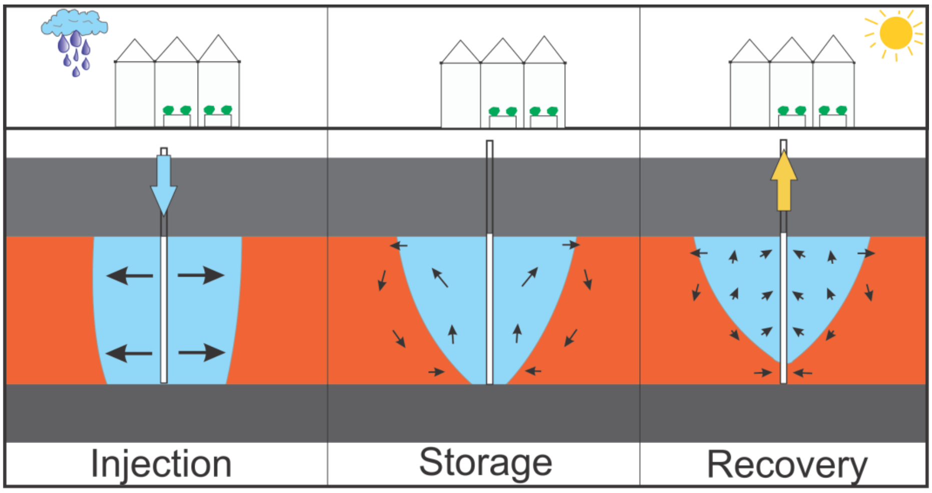


Figure 3: **Freshwater loss during ASR in brackish and saline aquifers due to buoyancy effects.**

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

An innovative ASR solution, combined with a Freshkeeper and RO, 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. The abstracted brackish water is used as additional and reliable freshwater source after desalination. The hybrid aquifer storage and recovery and reverse osmosis (ASRRO) system thus combines the best of two techniques and it contributes to optimal durable use of 'free' natural sources as (rain)water 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.

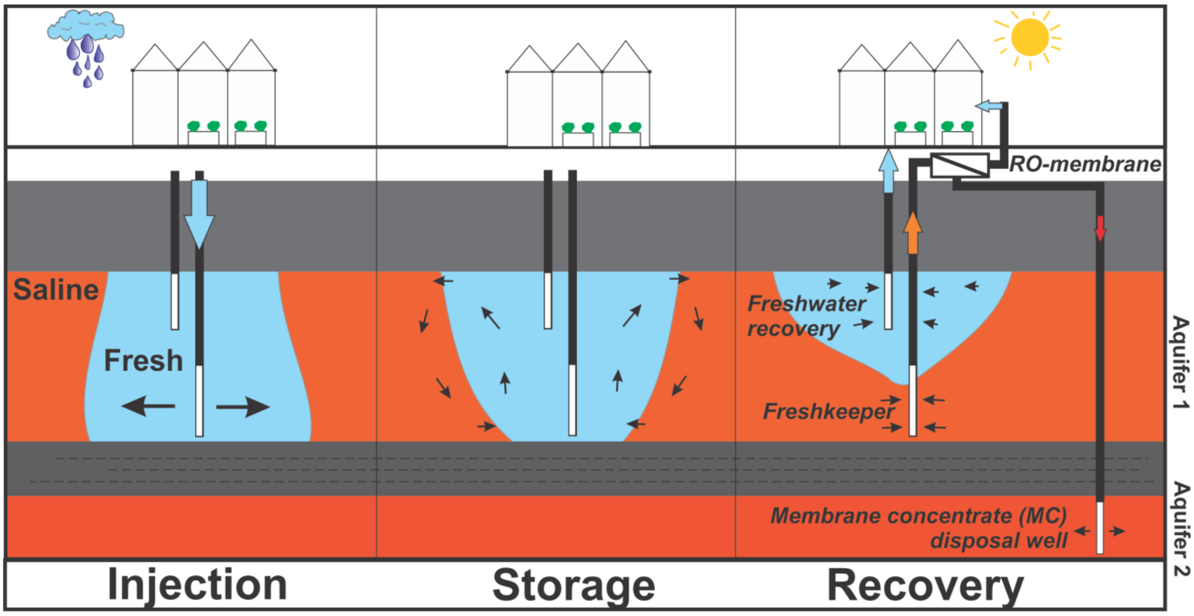


Figure 4: **Operation of an ASRRO system, as realized at the Westland demo site**

1.4 Objectives and approach

The objectives at the Westland ASRRO demo site defined at the start of the DESSIN project were:

- To quantify freshwater recovery by an ASR well design.
- To demonstrate the added value of an ASR/RO system on freshwater recovery.
- To demonstrate the effect of enhanced subsurface iron removal on membrane clogging.
- To demonstrate the impact of freshwater supply from brackish aquifers on regional groundwater quality and Water Framework Directive goals.
- To evaluate innovative solutions to enhance freshwater supply from brackish aquifers.

The tasks planned in order to meet these objectives are listed in Table 1. Since it was shown in D22.3 that Fe was not a source of membrane clogging, T33.3 was not relevant anymore. Clogging was mainly caused by particles in the feedwater, which cannot be mitigated by subsurface iron removal.

Table 1: Task description and approach set at the Westland ASRRO demo site and reported in this deliverable.

Task	Task description	Approach
T33.1	Quantification of the freshwater recovery by an innovative well design	Operation of the full scale field demonstration site; different ASR cycles will be run to quantify maximum freshwater recovery.
T33.2	Demonstration of the added value of an advanced ASRRO system	Freshkeeper and RO installation and operation, monitoring of operation, optimization
T33.3	Demonstration of the effect of enhanced subsurface iron removal on membrane clogging	Building on results of Task 22.2.2
T33.4	Demonstration of the impact of the Westland ASR/RO pilot on the regional groundwater quality	Monitoring of the water quality development of the brackish water target aquifer. Evaluation of the effect of the innovative ASR/RO system on regional water quality.

1.5 Relation with WA2 and D22.3

While the work in WA2 (reported in D22.3) was on research and development of (elements of) ASRRO in 2014 and 2015, the work in WA3 focusses on demonstration of the ASRRO functioning at the Westland demo site between 2014 and 2017. Additionally, the impact of the (wide-spread) use of ASRRO on a groundwater system was assessed.

The methodology for monitoring the ASRRO performance at the Westland ASR site was extensively discussed in D22.3 and is not repeated in D33.1.

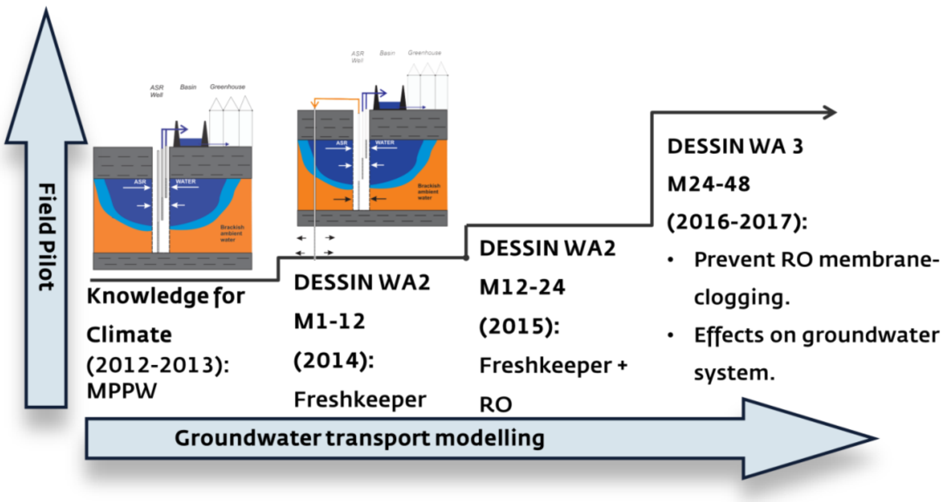


Figure 5: Visualisation of the approach and methods applied in the Westland ASRRO study

2 Set-up of the Westland ASRRO pilot

2.1 Set-up of the Westland rainwater infiltration and recovery (ASR) system

The Westland ASR system is installed to inject the rainwater surplus of 270,000 m^2 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)) with negligible lateral displacement (Zuurbier et al., 2013) for recovery in times of demand. For this purpose, two multiple partially penetrating wells (MPPW) were installed (Figure 7), such that water can be injected preferably at the aquifer base, and recovered at the aquifer top in order to increase the recovery (Zuurbier et al., 2014). Rainwater can be pre-treated and injected with a total rate of 40 m^3/h, and recovered with a total rate of 50 m^3/h.

For more information on the set-up and the hydrological short-circuiting that occurred during the pilot, the reader is referred to D22.3 and ANNEX B: Scientific analysis short-circuiting during ASRRO Westland.

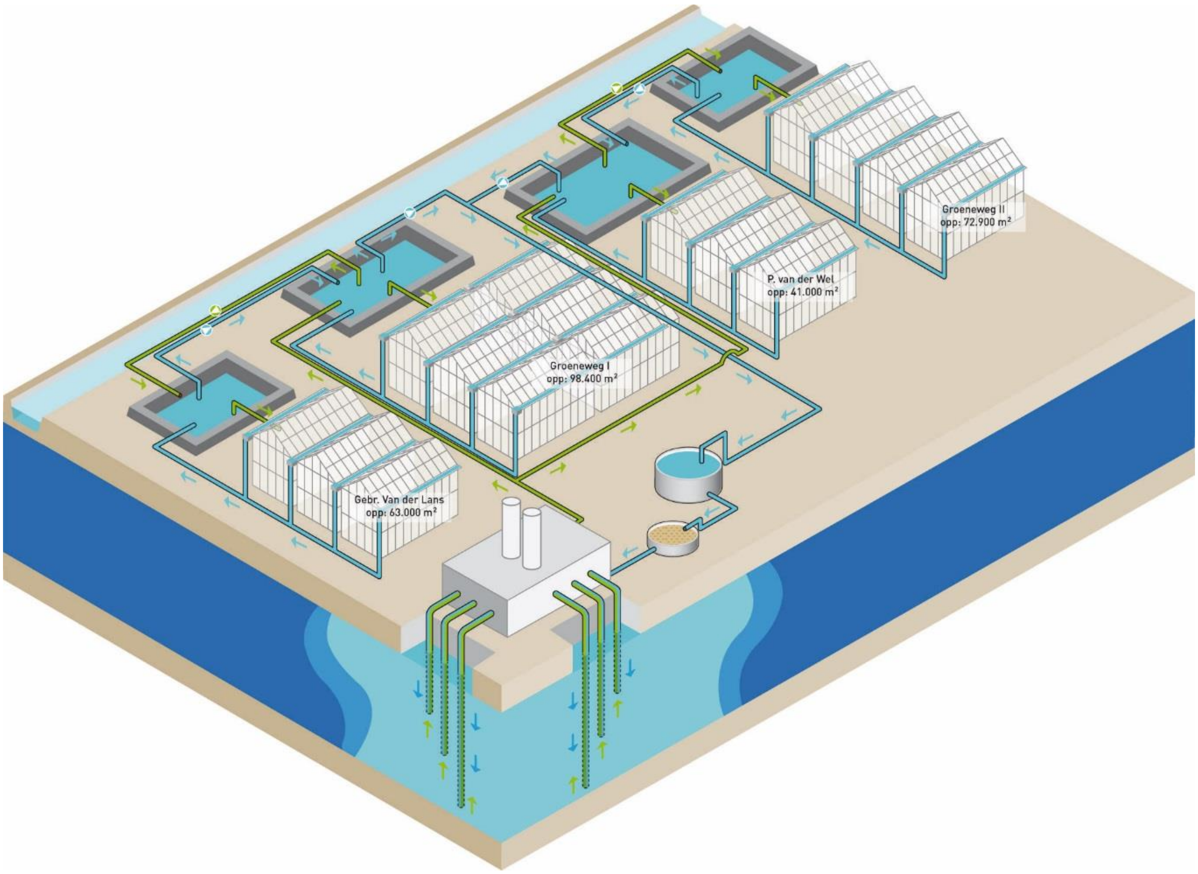


Figure 6: **Overview of the Westland-ASR system**

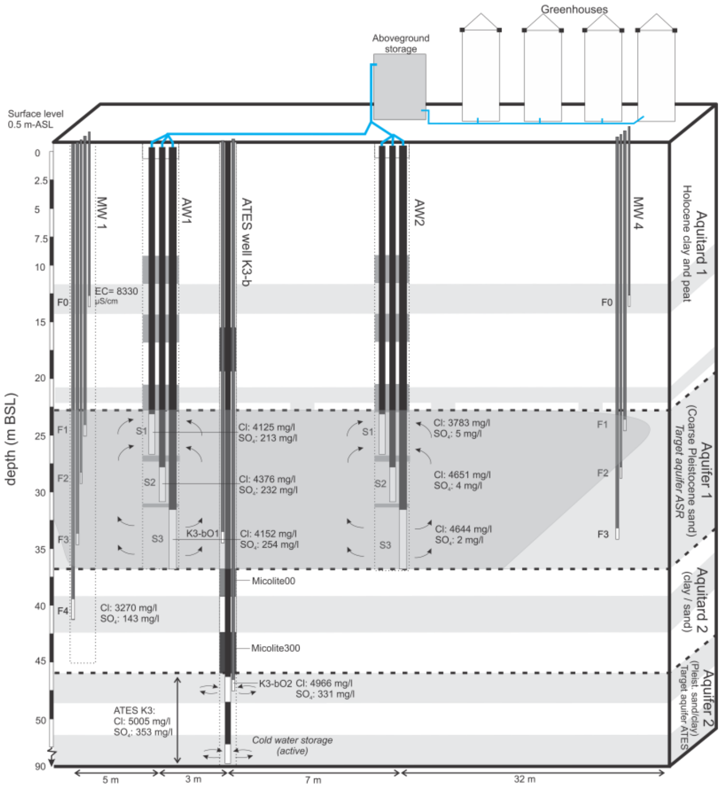


Figure 7: Cross-section of the Westland ASR site to schematize the geology, ASR wells, aquifer thermal energy storage (ATES) well, and the typical hydrochemical composition of the native groundwater. Horizontal distances not to scale.

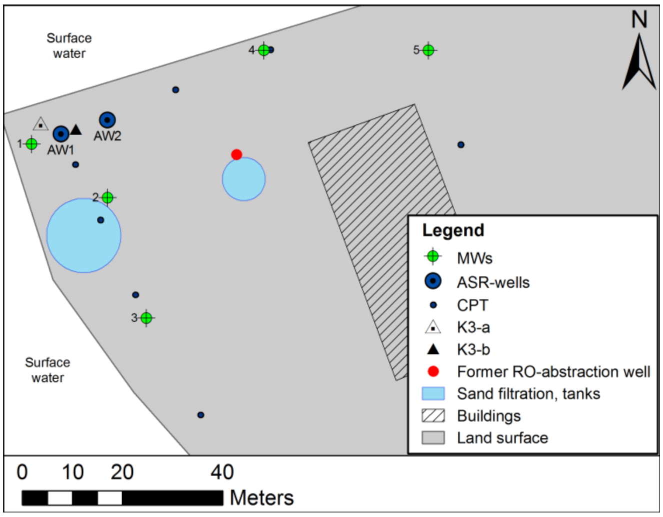


Figure 8: Locations of ASR (AW), ATES, and monitoring wells (MW). The former RO abstraction well is used during the DESSIN pilot to abstract mixed rainwater / groundwater at the fringe of the freshwater bubble and use this for desalination with the 'BWRO' plant.

2.2 Supplementary treatment using RO to achieve ASRRO

When recovery of unmixed water becomes unattainable due to admixing of brackish groundwater with the injected rainwater, treatment via reverse osmosis is applied to maintain the production of fresh irrigation water. Two wells are used to feed two separate RO-facilities.

One is the original brackish water RO-plant present at the site (coded 'BWRO'), which was formerly used to abstract brackish groundwater for RO (without rainwater admixed). This BWRO-system has been active since 2006, and forms the original supplementary freshwater supply of the local greenhouse. Since the start of the ASRRO pilot, the BWRO-well abstracts water from the whole aquifer thickness at the fringe of the injected freshwater body. The BWRO-plant is therefore fed by a mixture of water qualities present at this fringe.

The wells of the ASR-system were connected to a new RO-plant, realized in the DESSIN project to test the desalination of mixed injected water / brackish groundwater from below the freshwater bubble. This will simultaneously enable longer shallow recovery of unmixed injected water for direct use (Freshkeeper). This system is coded 'ASRRO' and the treatment part is coded ASRRO-plant.

The main difference between the feed water of ASRRO and BWRO consists of the location of abstraction. The water for the BWRO-plant is abstracted via a long, fully penetrating well screen at approximately 20 m from AW2 (Figure 8). This well is in the unmixed freshwater bubble at the end of the winter, but that deeper segments of the well completely salinizes as recovery proceeds. The abstracted water will therefore be a mixture of unmixed rainwater, mixed rainwater/groundwater, and unmixed brackish groundwater. This BWRO-plant was designed to be fed by 40 m^3/h of brackish groundwater to produce 20 m^3/h (480 m^3/d) of freshwater, which should result in an equal stream of concentrate at an RO-recovery of 50%.

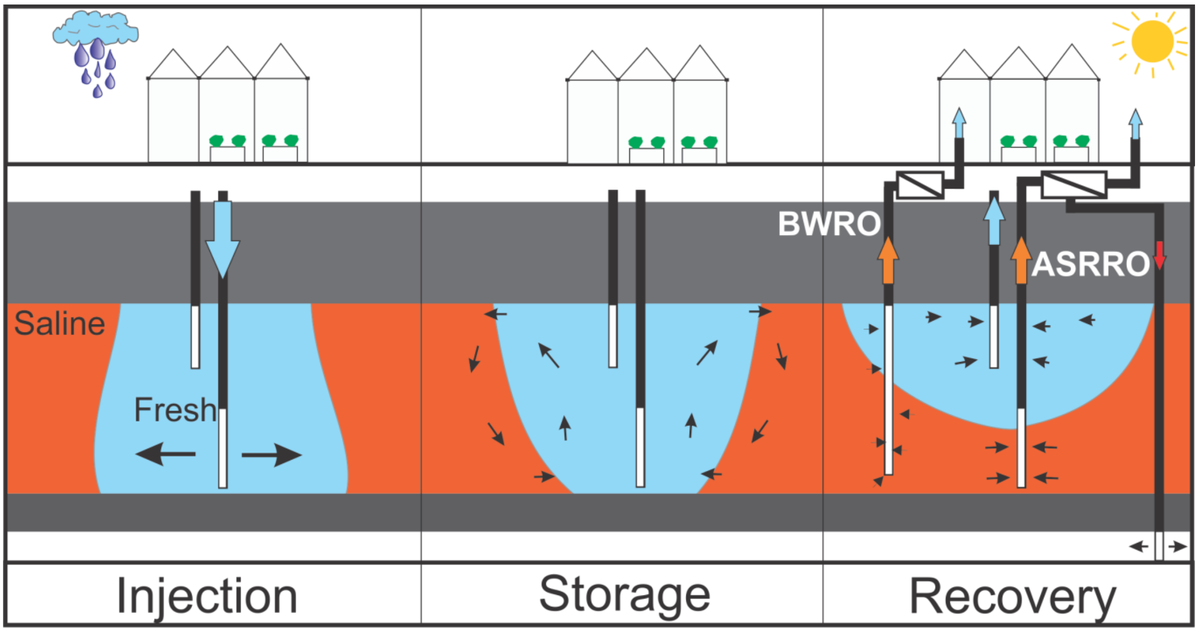


Figure 9: Abstraction of mixed rainwater / brackish groundwater via BWRO and ASRRO

2.3 Detailed hydrogeological characterization based on local drillings

The target aquifer for ASR (Aquifer 1) was found to be 14 m thick and consists of coarse fluvial sands (average grain size: 400 µm) with a hydraulic conductivity (K) derived from head responses at the monitoring wells upon pumping of 30 – 100 m/d (ANNEX B). Aquifer 2 (target aquifer for ATES) has a thickness of more than 40 m, but is separated in two parts at the ATES well K3-b by a 20 m thick layer clayey sand and clay. A blind section was installed in this interval, and the borehole was backfilled with coarse gravel in this section. The K-value of the fine sands in Aquifer 2 derived from a pumping test at approximately 500 m from the ASR-wells is 10 to 12 m/d and is in line with the estimated K-value from grain size distribution (Mos Grondmechanica, 2006). The effective screen length of K3-b in this aquifer is only 8 and 5 m.

The groundwater is typically saline, with observed Cl concentrations ranging 3,793 to 4,651 mg/l in Aquifer 1 and approximately 5,000 mg/l in Aquifer 2 (see also Figure 7). This means that with the accepted Cl-concentrations during recovery, only around 1% of admixed ambient groundwater is allowed. A sand layer in Aquitard 2 contains remnant fresher water (Cl = 3,270 mg/l). SO_4 is a useful tracer to distinguish the saltwater from Aquifer 1 and 2: it is virtually absent in Aquifer 1 (presumably younger groundwater, infiltrated when the Holocene cover was already thick), whereas it is high in Aquifer 2 (older water, infiltrated through a thinner clay cover which limited SO_4-reduction, see Stuyfzand (1993) for more details): 300 to 400 mg/l SO_4.

2.4 Borehole leakage near ASRRO well AW1

A very particular and undesirable phenomenon is observed at around 3 m from ASRRO well AW1, where leakage of deep, saline groundwater is occurring via an interconnection with a deeper aquifer. This interconnection was caused by an earlier perforation (borehole) for the sake of the installation of a deeper well for aquifer thermal energy storage (ATES). Apparently, this borehole was not properly sealed, or a too high injection pressure on the ATES well was used. Despite an attempt to seal this borehole on February 3, 2015, the leakage persisted. More information on this leakage can be found in *ANNEX B: Scientific analysis short-circuiting during ASRRO Westland.*

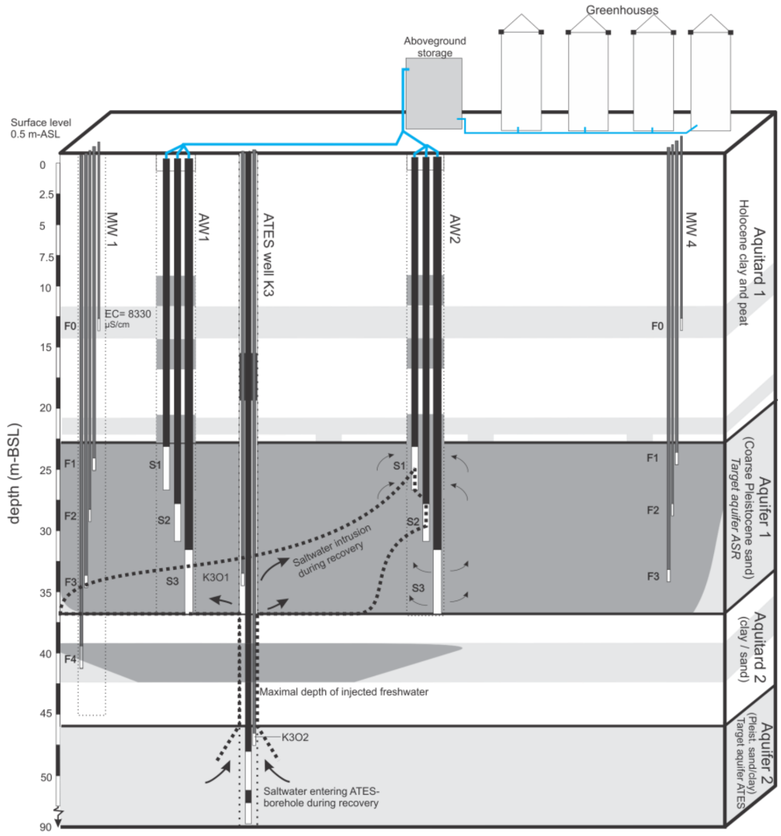


Figure 10: **Leakage of deep, saline groundwater during recovery, hampering optimal recovery of freshwater from the Westland target aquifer.**

2.5 Groundwater transport model

Groundwater transport modelling was executed to validate the added value of the MPPW set-up under the local conditions. In the later stage of the research, the groundwater transport model was used to test potential pathways for deeper groundwater to enter the target aquifer and explore the characteristics of a potential conduit via scenario modelling. Correction for groundwater densities in the flow modelling was vital, due to significant contrast between the aquifer's groundwater and the injected rainwater. In order to incorporate variable density flow and the transport multiple species, SEAWAT Version 4 (Langevin et al., 2007) was used with PMWIN 8 (Chiang, 2012) to simulate the ASRRO operation.

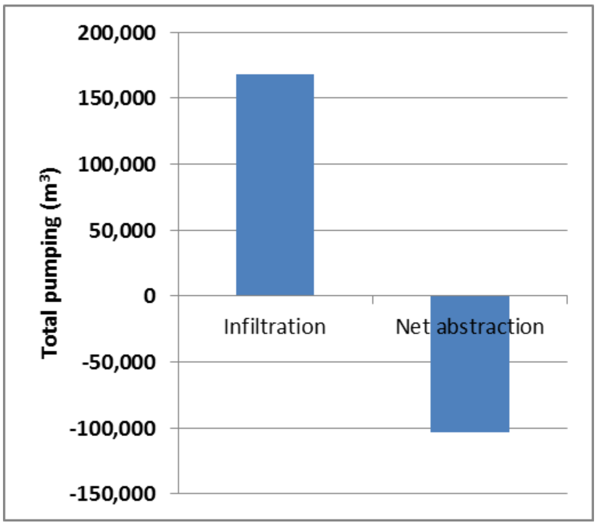
For more information on the groundwater model, the reader is referred to *ANNEX B: Scientific analysis short-circuiting during ASRRO Westland.*

3 Freshwater production by the Westland ASRRO system

3.1 Evaluation of freshwater infiltration and recovery

To get an overview of the performance of the Westland ASRRO water supply system, only the three consecutive and complete cycles of 2014, 2015, and 2016 are analyzed to obtain the water balance. In 2013, the system was first put in operation halfway the wet season, missing a large part of the infiltration stage. At the time of writing this report, the data of the dry season (recovery) of 2017 was incomplete. These cycles were therefore omitted when analyzing the ASRRO water balance.

In 2014 – 2016, in total 168,000 m^3 of rainwater surplus was infiltrated and 102,000 m^3 (64%) of freshwater was produced (Figure 11), either by direct recovery (ASR) or desalination (RO).



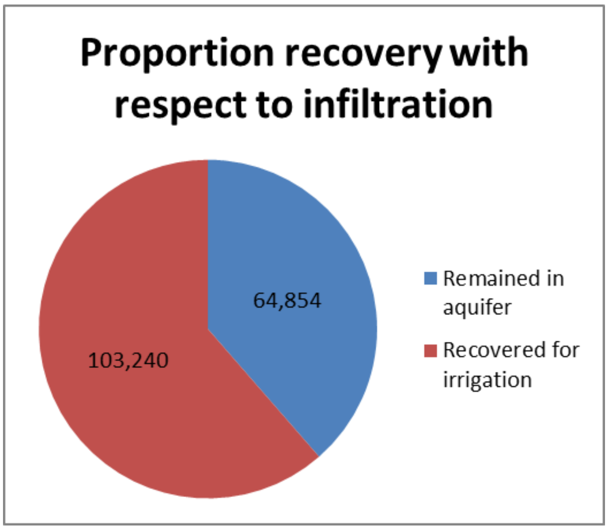


Figure 11: Total freshwater infiltration in and abstraction from the Westland target aquifer at the Westland demo site (left, 'net abstraction' = the production of freshwater from the aquifer by direct recovery of rainwater or *after* RO-treatement) and the resulting water balance (right). Based on three complete cycles (2014 – 2016).

3.2 Direct recovery of freshwater

The volumes of infiltrated and recovered unmixed rainwater with the Westland ASRRO system are presented in Table 2 and Figure 12. This recovered rainwater could be used directly and without post-treatment for irrigation in the tomato greenhouse.

Only Cycle 2013 was incomplete, starting only after completion of the installation half-way December 2012. During that Cycle (before the DESSIN project), the Freshkeeper not installed yet. Especially 2017 was marked by fairly dry autumn and winter, firmly reducing the water available for infiltration and subsequent recovery. In total, 22.5% was recovered unmixed (23.1% in the years with a Freshkeeper).

Table 2: **Infiltration, recovery, and recovery efficiency of unmixed rainwater at the Westland ASRRO demo site. This was directly used for irrigation.**

Cycle	Yearly infiltration (m^3)	Yearly recovery (m^3)	Recovery efficiency (%)
2013*	18,313	3,082	16.8%
2014	70,710	13,320	18.8%
2015	37,166	9,625	25.7%
2016	64,846	15,855	24.5%
2017**	27,968	7,482	26.8%
Total	**219,003**	**49,306**	**22.5%**

**Started half-way December 2012. No Freshkeeper added to the MPPW-ASR system*

*** Cycle until April 26, 2017: maximal direct recovery was attained.*

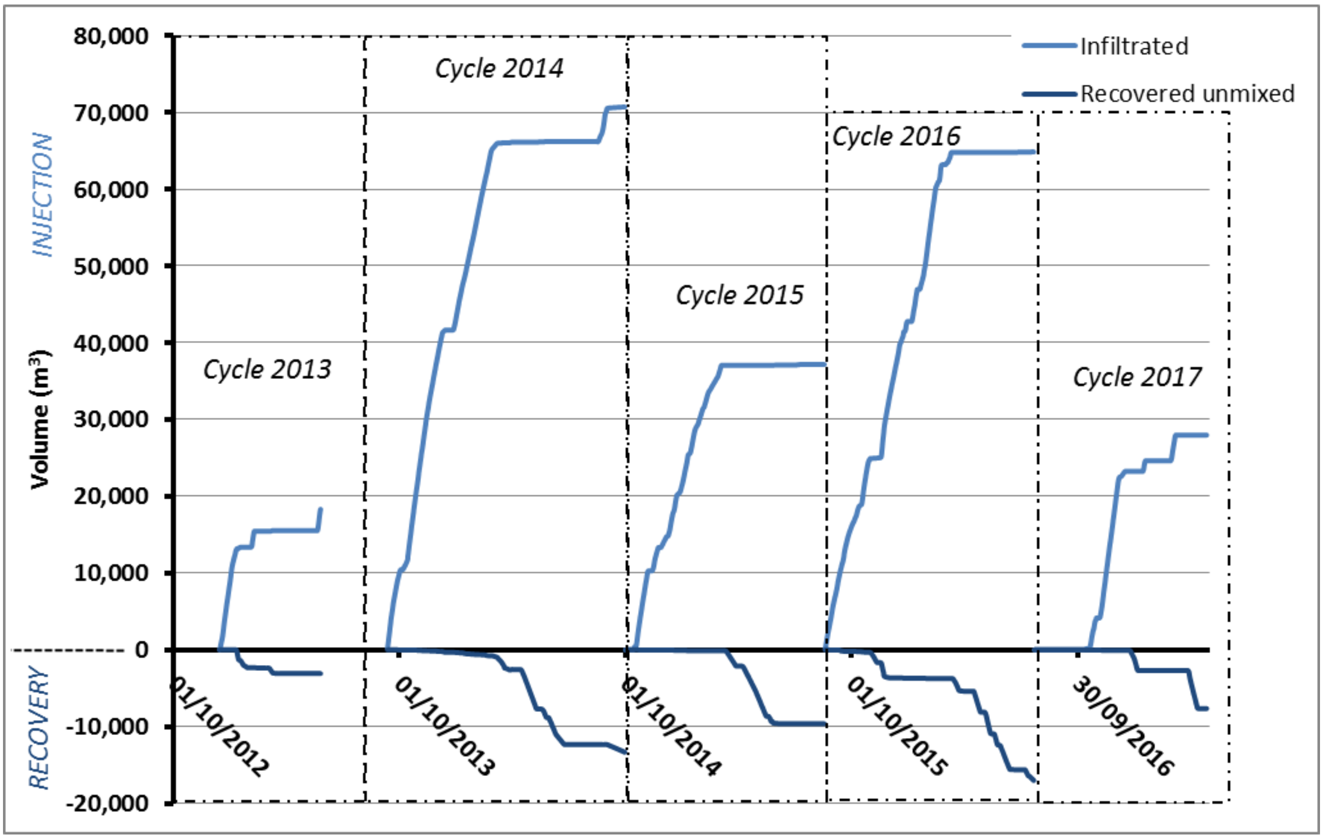


Figure 12: Infiltrated and recovered volumes per cycle versus time (data on a daily basis)

3.3 Production of freshwater upon RO-treatment

Recovered water at the ASRRO found unsuitable for irrigation due to its high salinity due to mixing with native brackish groundwater, was treated with the BWRO and the ASRRO plant in order to keep producing fresh irrigation water.

Table 3: Abstraction of brackish groundwater as feedwater for the ASRRO and BWRO plant and transformation to freshwater and concentrate upon RO-treatment

Cycle	Abstracted brackish / Feedwater ASRRO	Produced freshwater ASRRO	Re-injected / Concentrate ASRRO	RO-Reco-very	Feedwater BWRO	Produced freshwater BWRO	Concentrate BWRO	RO-Reco-very
2014*	10,226	0	10,226	0%	33,480	13,392	20,088	40%
2015	15,661	6,841	8,820	44%	61,771	19,415	42,356	31%
2016	28,192	11,547	16,645	41%	28,196	12,095	16,664	43%
Total	**54,079**	**18,388**	**35,691**	**34%**	**123,447**	**44,902**	**79,108**	**36%**

**ASRRO plant not in operation yet, brackish groundwater from Freshkeeper directly re-injected*

3.3.1 Production via Freshkeeper wells with the ASRRO-Plant

The ASRRO plant was used as a supplementary freshwater source since May 2015 to treat the water abstracted by the Freshkeeper wells (brackish water from the deepest ASRRO wells) and later the shallower ASRRO wells (AW2.1 and AW2.2). The freshwater production and concentrate disposal of ASRRO are shown in Figure 13.

The ASRRO-plant was operated with a constant frequency of the feed pump, allowing changes in RO-recovery upon changes in the feed water quality or membrane condition. The RO-recovery resulting in the ASRRO-plant (percentage of the water transformed to high-quality freshwater) is shown in Figure 14. During the production of freshwater with the ASRRO plant using feedwater from the deeper wells (AW1.3, AW2.2, AW2.3), the RO-recovery of the ASRRO-plants initially remained stable at around 44%. However, during the second stage, especially when also AW2.1 (mostly fresh rainwater, slightly mixed with brackish groundwater) was used to feed the plant, the RO-recovery slowly decreased to 36% in 2017, despite an initial increase in RO-recovery due to the lower salinity. There was no membrane cleaning performed during the entire experiment.

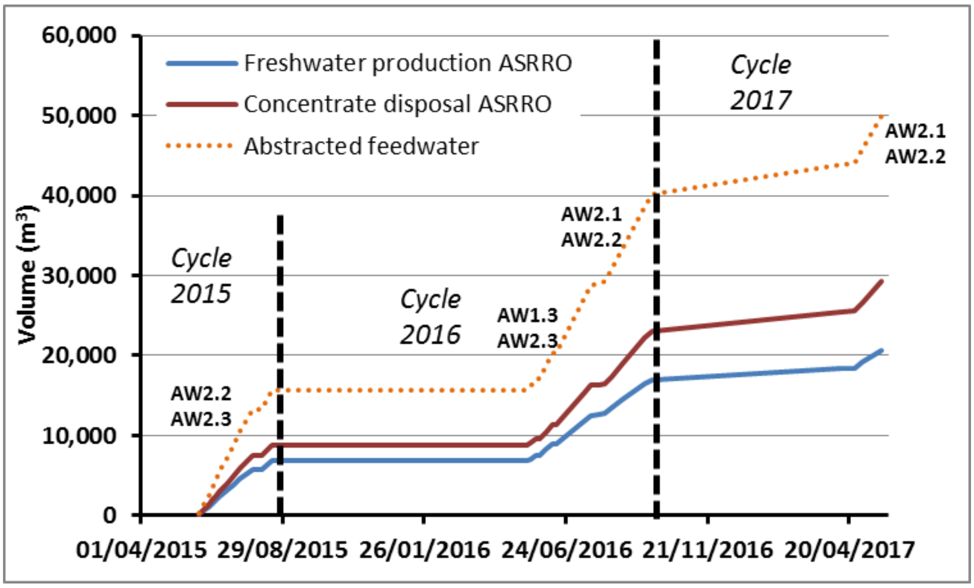


Figure 13: Freshwater production and concentrate disposal by the ASRRO-plant. The ASRRO wells feeding the ASRRO-plant during the various stages are indicated.

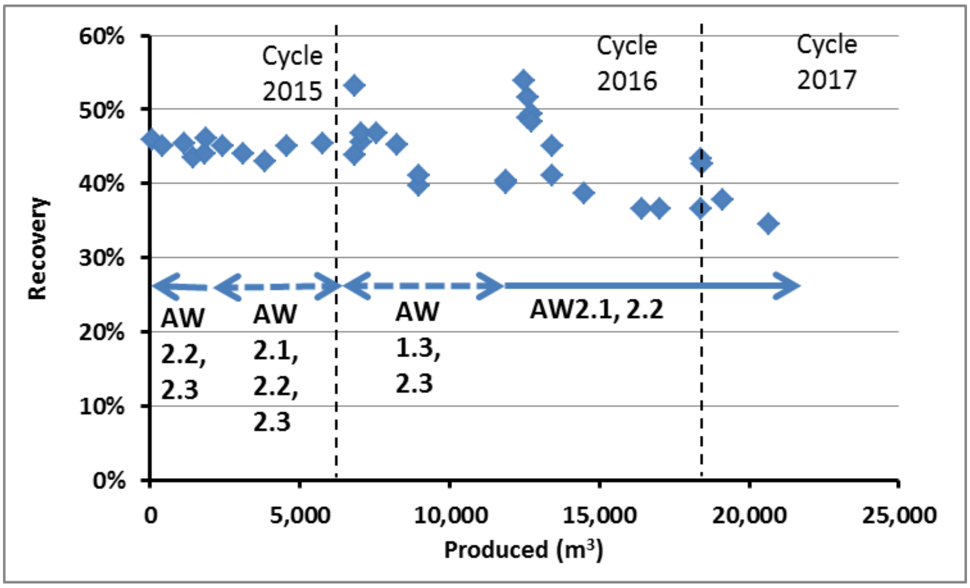


Figure 14: RO-recovery (percentage of abstracted water transformed to freshwater) of the ASRRO-plant

3.3.2 Production via the BWRO well

The BWRO-plant was operated with a constant frequency of the feed pump, allowing changes in RO-recovery upon changes in the feed water quality or membrane condition. The BWRO-plant was used when abstraction of unmixed rainwater and production of freshwater with the ASRRO-plant were insufficient: in 2014 to supply extra irrigation water during August, during June and July 2015, and during August and September 2016. The freshwater production and concentrate disposal of BWRO are shown in Figure 15.

The recovery of the BWRO membranes rapidly decreased from almost 50% to <30% in 2013 and 2015, indicating severe membrane clogging. Membrane cleaning was therefore performed twice with Genesol703 (supplied by GeneSys) in order to restore the production capacity of the BWRO-plant. More information on this process can be found in DESSIN deliverable D22.3 (Zuurbier et al., 2016).

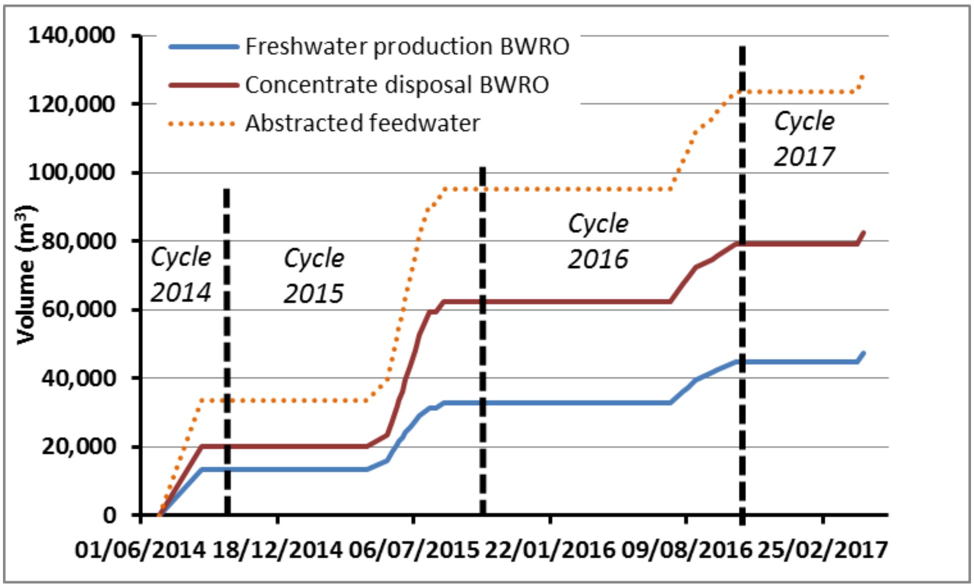


Figure 15: Freshwater production and concentrate disposal by the BWRO-plant.

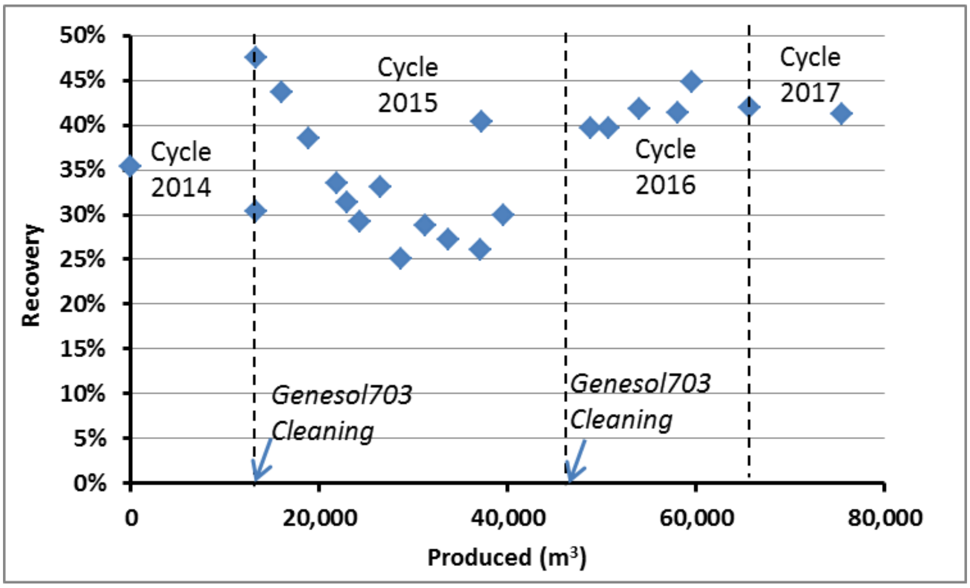


Figure 16: Recovery (percentage of abstracted water transformed to freshwater) of the BWRO-plant

4 Performance of the RO-plants

4.1 ASRRO-plant

The performance of the ASRRO-plant for treating abstracted brackish water from the ASRRO can be evaluated based on the recovery (Figure 14) and the feed pressure and the pressure on the reject side of the membranes (delta-pressure), which are shown in Figure 17.

The recovery shows the main decrease during:

- Salinization of the ASRRO-wells and therefore salinization of the feedwater. This can be explained by the increased osmotic pressure and may not be an indication of clogging. This often occurs at the start of the season, when feed water is still largely rainwater and RO-recovery can shortly be >50%
- Long-term decrease from 44% to 36% in Cycle 2016 and 2017, after a stable recovery in Cycle 2015.

At the same time, the feed pressure (Figure 17) and the feed pressure – recovery ratio (Figure 18) show an increase in 2016 and 2017, especially when water from AW2.1 is part of the feedwater in the final stage, despite the lower EC of the feedwater. This is an indication for a decreasing RO-performance. These results suggest that for desalination during ASRRO, the deeply anoxic and more brackish water can be preferred, despite the higher salinity. This is caused by the presence of clay and Fe-colloids in the shallower infiltrated rainwater (see DESSIN deliverable D22.3).

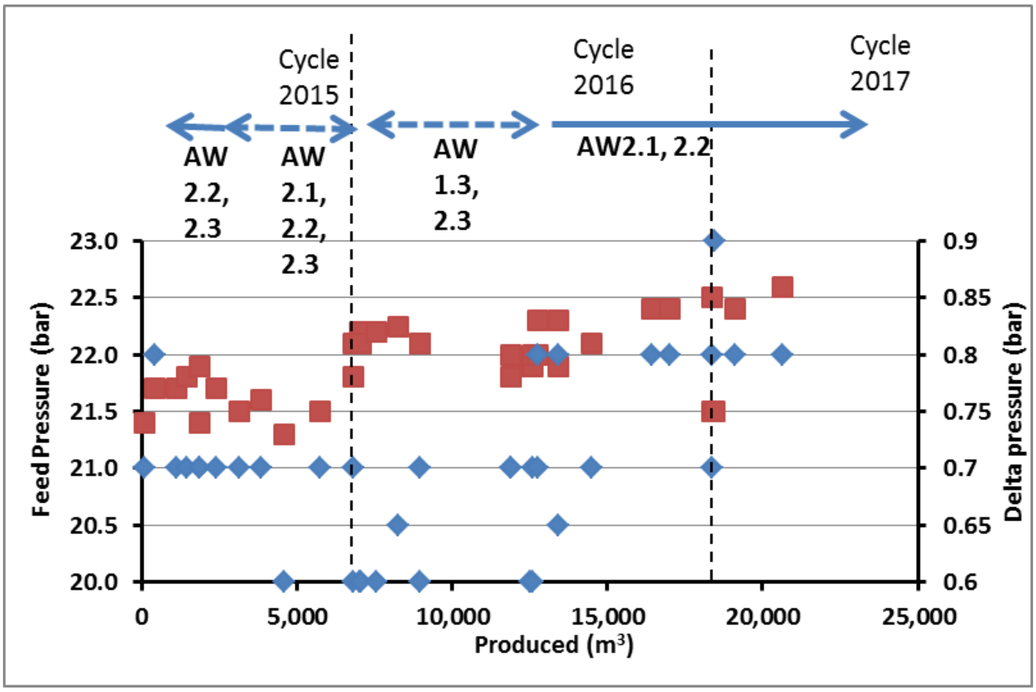


Figure 17: Feed pressure (red) and delta-pressure (blue) between feed and reject side for the ASRRO-plant.

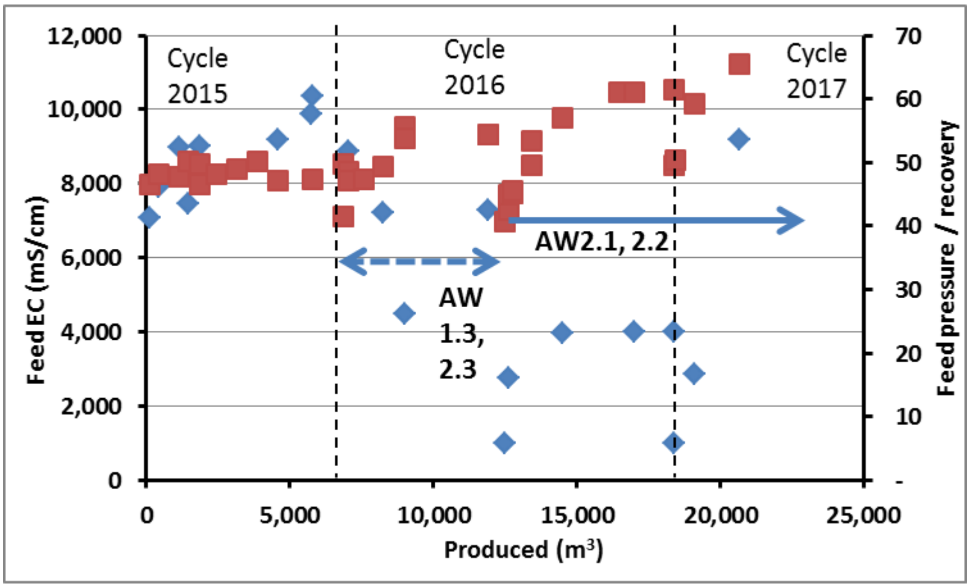


Figure 18: **The feed pressure – recovery ratio (red) and the observed ECs (blue) of the feedwater during sampling at the ASRRO plant.**

4.2 BWRO-plant

The performance the BWRO plant is marked by:

- Rapidly increasing feed pressures and rapidly decreasing RO-recovery during the 2015 cycle (Figure 16, Figure 20), marking a rapid and severe reduction of the membrane performance. This was presumably caused by accumulation of clay particles and some iron oxides (see DESSIN D22.3);
- A relatively stable production in 2016 and 2017, based on the feed pressure, delta-pressure, and the recovery.

From 2016 onwards, the recovery stabilized between 40 and 45%. In this period, an automated flush was implemented at the BWRO system in order to remove accumulated particles. With this flushing added, the BWRO was operated as follows (Figure 19):

1. A 5 minutes flushing stage at the start-up (operating submersible pump only, all water pumped to the concentrate injection well upon filtration by a 1 micron cartridge filter). During this stage, particles should get washed of the membranes;
2. Start of the RO feed pump and normal operation for 6 hours;
3. The feed pump and submersible pump were switched off, and the membranes were flushed with fresh permeate from a 500 L permeate storage tank;
4. The operation was paused for 30 minutes, leaving the membranes in the permeate with its very low. In this stage, it was aimed to mobilize ('disperse') clay particles accumulated on the membrane surface.

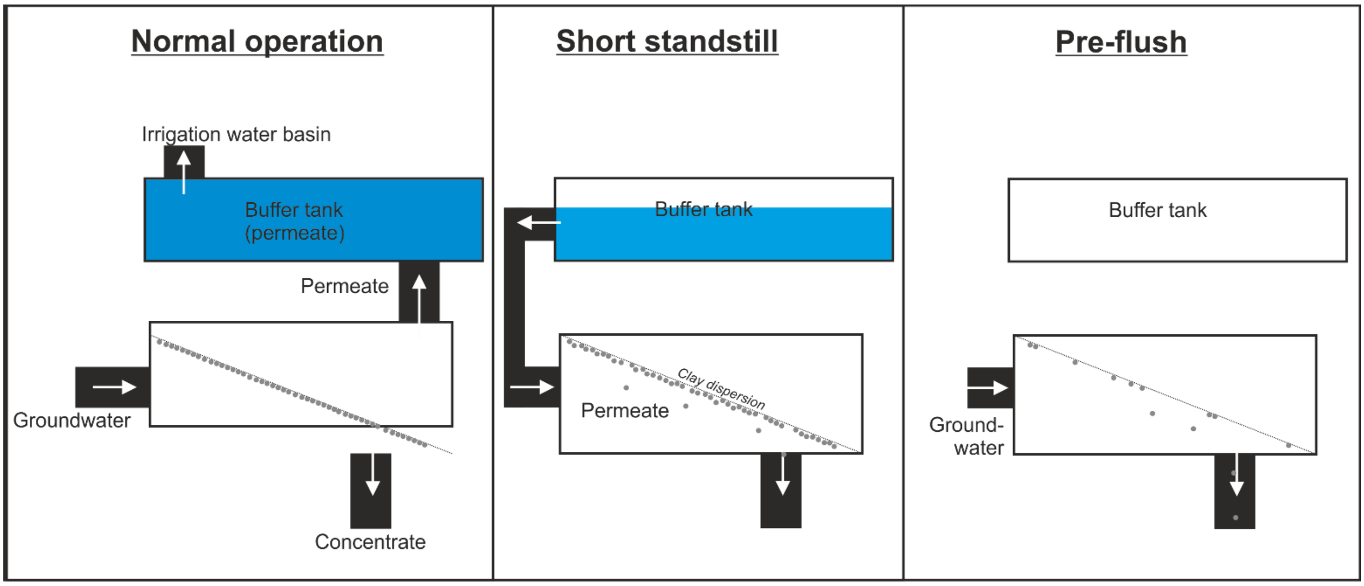


Figure 19: Flushing of the BWRO membranes as frequently applied in 2016 to clean the membrane.

According to the results of 2016 and 2017, this modified operation significantly improved the long-term performance of the BWRO membranes, since a severe reduction of the RO recovery and a severe increase in feed pressure were not observed.

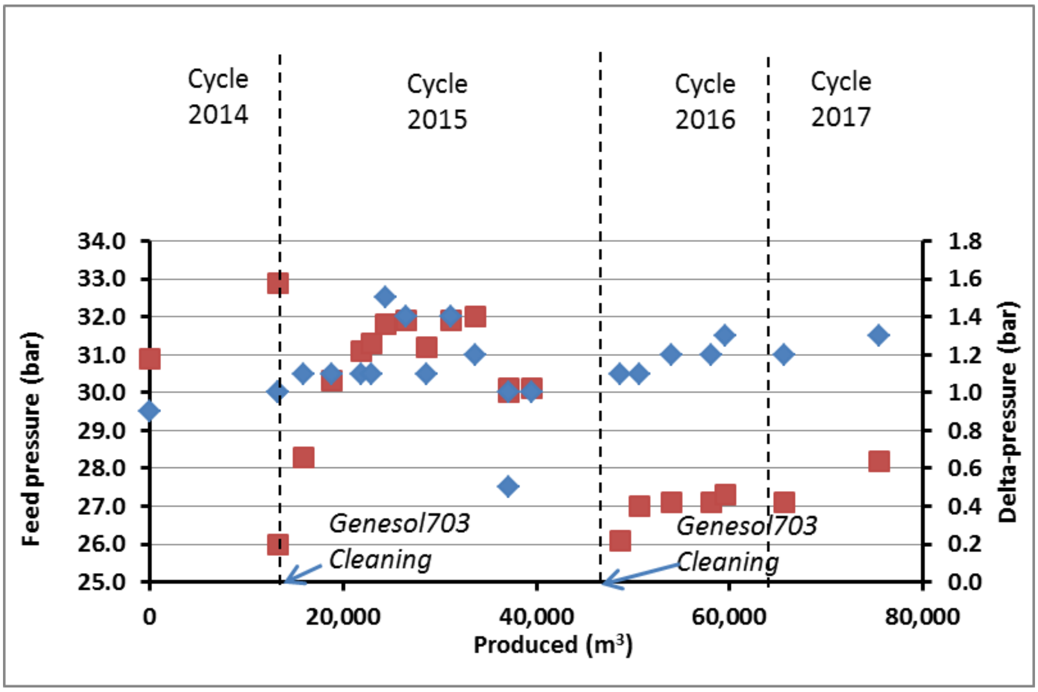


Figure 20: Feed pressure (red) and delta-pressure (blue) between feed and reject side for the BWRO-plant.

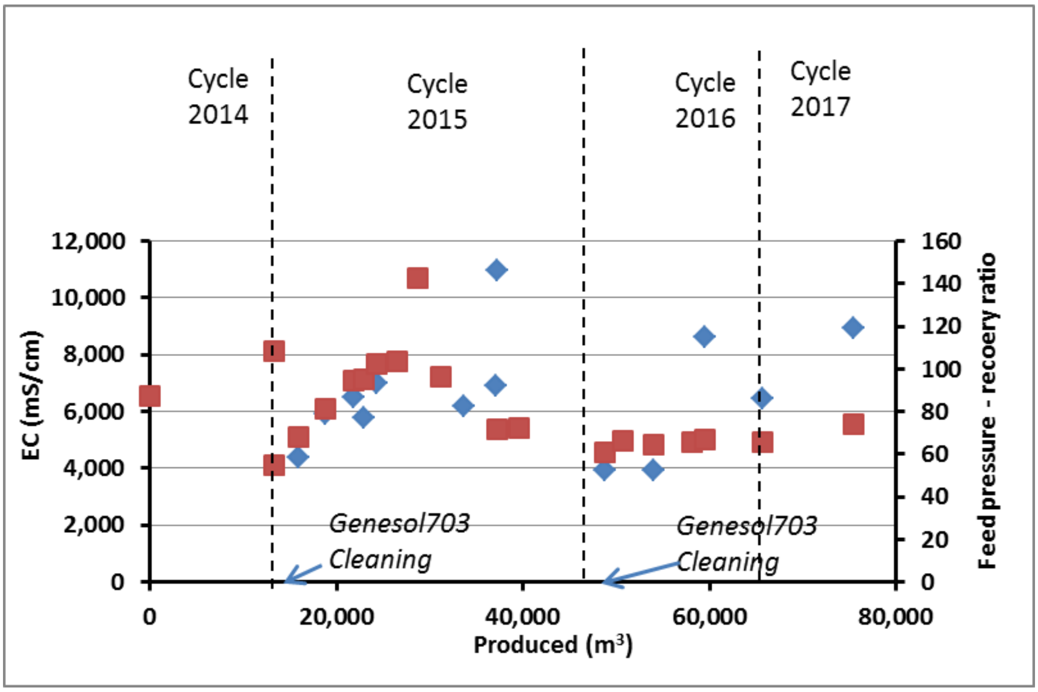


Figure 21: The feed pressure – recovery ratio (red) and the observed ECs (blue) of the feedwater during sampling at the BWRO plant.

5 Water balances at the Westland demo site

5.1 Water balance of the Groeneweg II greenhouse (hosting the ASRRO)

The ASRRO system was installed on the plot of the Groeneweg-II tomato company (10 hectares). This was the primary user of the recovered water upon ASR and RO.

In 2014, half of the supplementary irrigation water demand could be supplied by direct recovery of the stored rainwater (Figure 22, Figure 23). The other half was produced by the BWRO-plant. In 2015, more supplementary irrigation water was required and less rainwater was available for infiltration. Therefore, a larger part had to be produced by BWRO. In 2016, even more water was required due to a late summer drought, but this could be largely supplied by ASR and ASRRO. Almost 40% of the supplementary water demand was supplied by direct recovery of freshwater (Table 4).

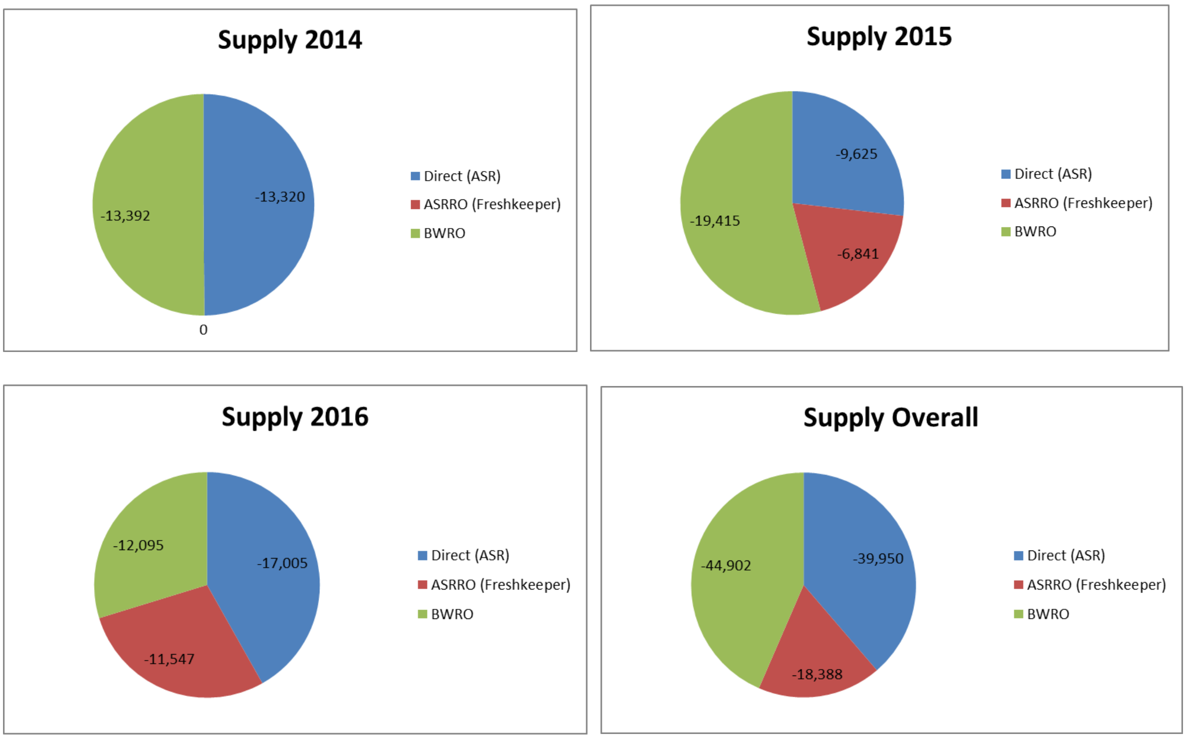


Figure 22: Contribution of the different freshwater sources to the supplementary water supply (2014-2016)

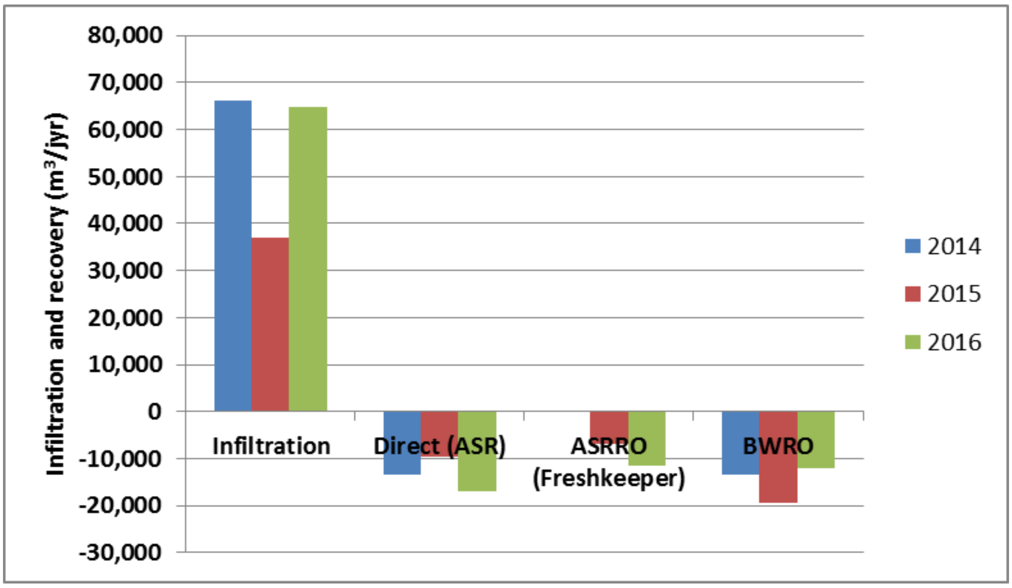


Figure 23: Infiltration, direct recovery (unmixed), production of the ASRRO-plant (brackish water recovered with Freshkeeper), and production of the BWRO plant.

Table 4: Contribution of the different freshwater sources to the supplementary water supply (2014-2016)

Source	2014	2015	2016	Total	Total
Direct (ASR)	-13,320	-9,625	-17,005	-39,950	39%
ASRRO (Freshkeeper)	0	-6,841	-11,547	-18,388	18%
BWRO	-13,392	-19,415	-12,095	-44,902	43%
Total	**-26,712**	**-35,881**	**-40,647**	**-103,240**	**100%**

5.2 All growers connected to the ASRRO scheme

In total, four growers (27 hectares) were connected to the ASRRO scheme to provide rainwater surplus for infiltration, while 5 growers (29 hectares) were supplied by the ASRRO systems and three BWRO systems (GW-II, VdLans, GW-I). Due to their high water demand (tomato's: >1000 mm/yr), a balance between infiltration of rainwater and production of freshwater is not attained at this site (Figure 24). Between 2014 and 2016, about 43% of the water produced from the Westland aquifer was not replenished by artificial infiltration of rainwater via the DESSIN ASRRO plant. However, 57% of the supplementary water demand could be covered by infiltration of the rainwater surplus that would otherwise be drained to sea.

Especially the company Groeneweg-I has a relatively high supplementary water demand (Figure 25), which is due the low capacity of its above ground rainwater tanks (50 mm).

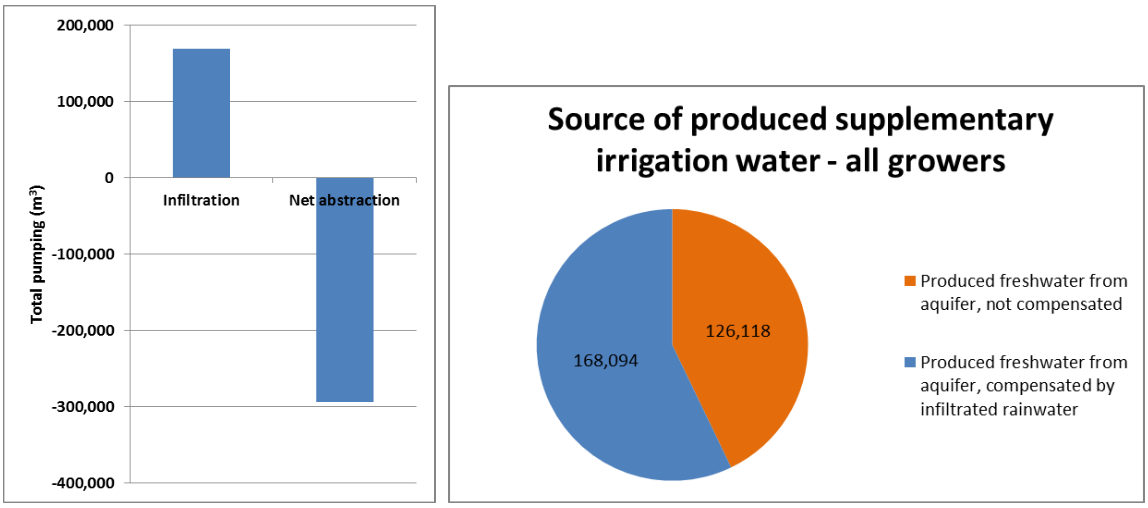


Figure 24: Total infiltration and net abstraction ('production') of freshwater from the target aquifer.

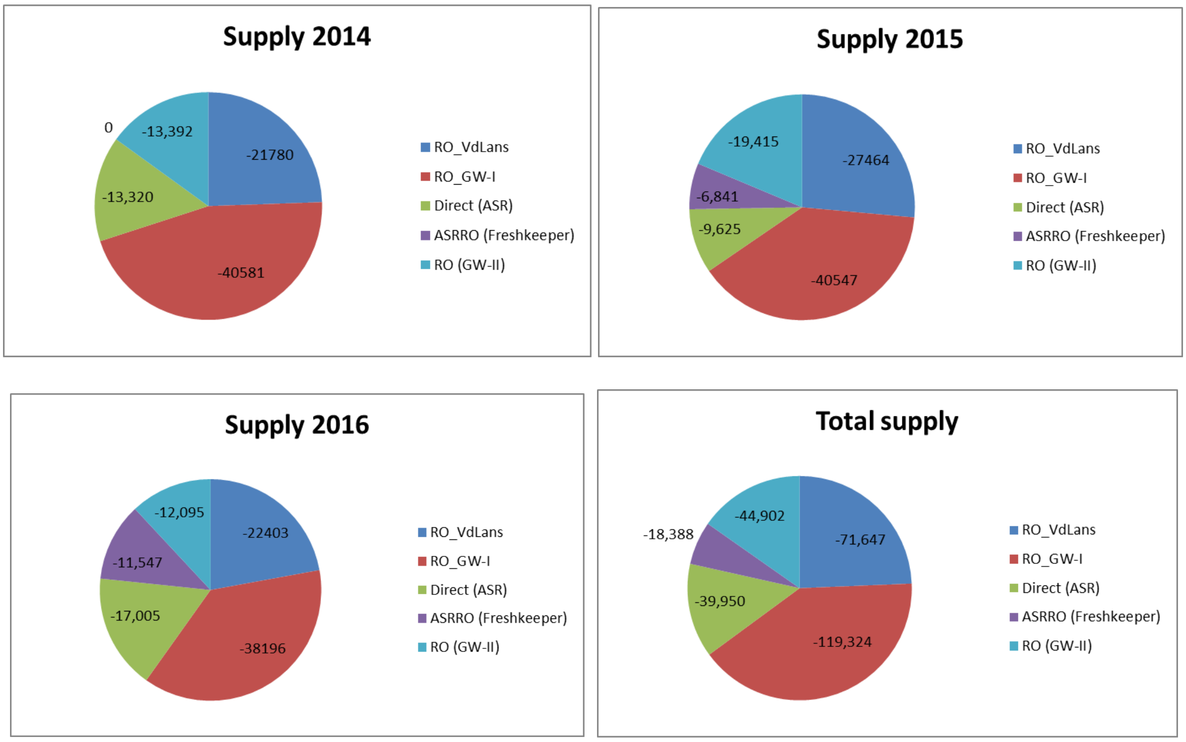


Figure 25: Contribution of the different freshwater sources to the supplementary water supply of all growers providing rainwater surplus to the ASRRO system (2014-2016)

6 Impact of ASRRO on the groundwater system

In order to evaluate the impact of ASRRO on the groundwater system, the local and regional effects of ASRRO were studied.

6.1 Local impact of the Westland ASSRO system at the demo site

6.1.1 Modelling results

Groundwater transport modelling with SEAWAT was performed to simulate the ASRRO operation and to validate the added value of the MPPW-ASRRO set-up at the Westland field site (for more information on the model set-up, see D22.3).

Comparison field data and groundwater modelling

Model results are here compared with real field data to validate its capability to predict future performance. Therefore, measured chloride concentrations at measuring wells MW1 and MW2, and at the ASRRO wells AW1 and AW2 were plotted together with modelled values (ANNEX D). The model is overall accurate in predicting the trends of (measured) increasing and decreasing chloride concentrations. Only the deepest well screens salinize more rapidly than predicted by the model, especially at MW1, MW2 and AW1.

Cross-sections of the chloride distribution

Cross-sections have been included to visualize the dynamics near all wells. The initial chloride distribution (15 December 2012) is given in Figure 26. The chloride distribution in the subsurface after a long period of infiltration (22 January 2015) and abstraction (18 August 2015) are shown in Figure 27 and Figure 28, respectively. The final situation (26 April 2017) is given in Figure 29.

The most relevant observations are:

- A freshwater lens forms and tends to seep through the upper clay layer, getting out of reach for abstraction.
- Infiltrated freshwater is intercepted by the BWRO, at the fringe of the infiltrated freshwater body;
- The conduit to Aquifer 2 between AW1 and AW2 caused short-circuiting of brackish water from this deeper aquifer during periods of abstraction. Only limited freshwater can be pushed downwards during infiltration;
- Overall, membrane concentrate injections in the deeper aquifer by either BWRO or ASRRO led to freshening of the deeper aquifer. This means that their net effect on the deeper aquifer is positive.

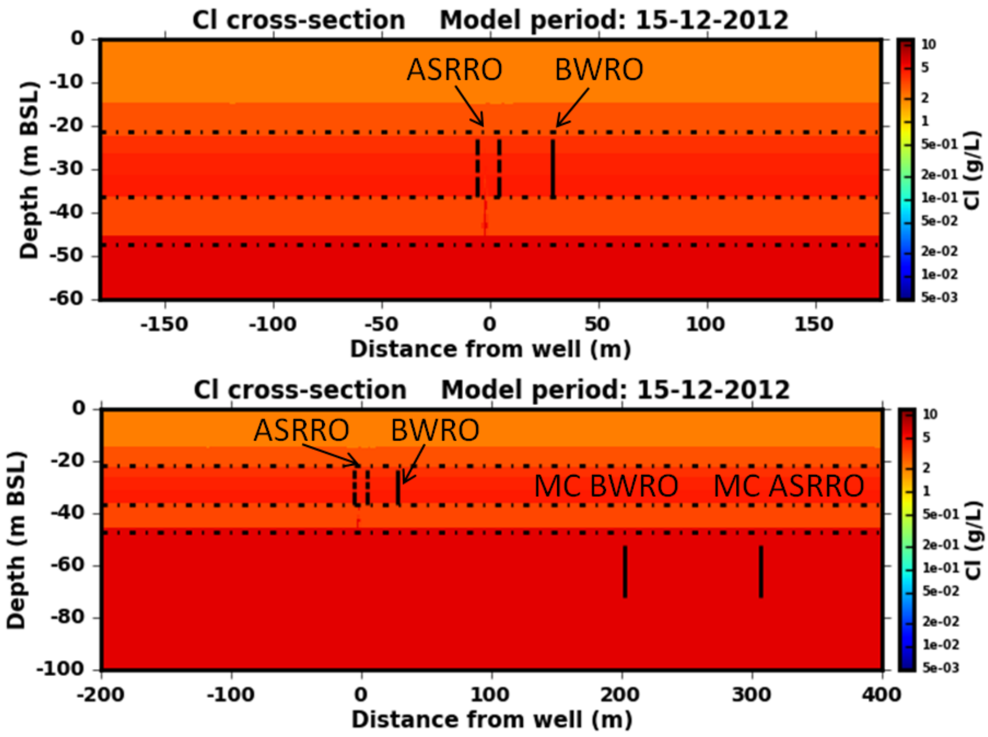


Figure 26: Initial chloride concentrations (15 December 2012) for cross-sections through the ASSRO wells (AW1 and AW2), both near the ASRRO (top) and an overview including membrane concentrate injection wells (bottom).

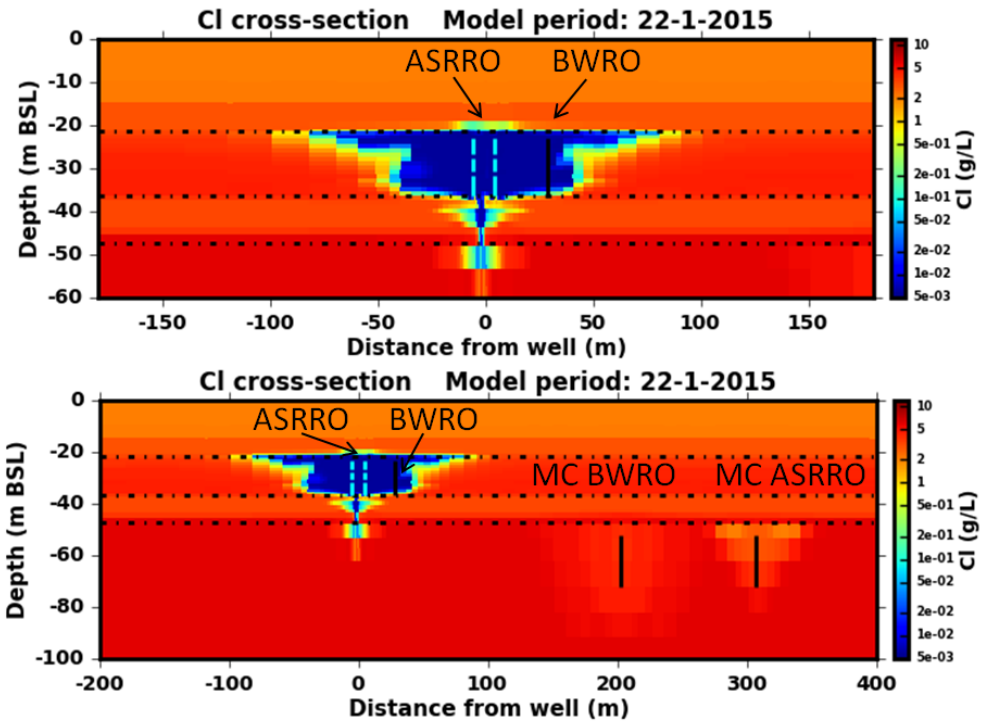


Figure 27: Modelled chloride concentrations after a prolonged period of infiltration (22 January 2015) for cross-sections through the ASSRO wells (AW1 and AW2), both near the ASRRO (top) and an overview including membrane concentrate injection wells (bottom).

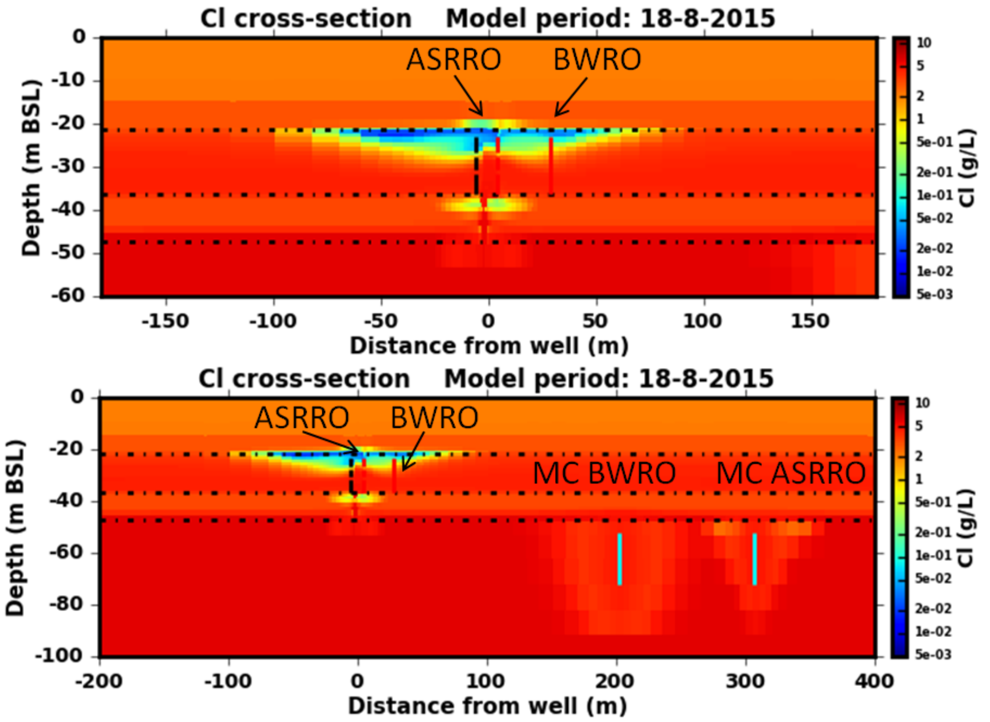


Figure 28: Modelled chloride concentrations after a prolonged period of abstraction (18 August 2015) for cross-sections through the ASRRO wells (AW1 and AW2), both near the ASRRO (top) and an overview including membrane concentrate injection wells (bottom).

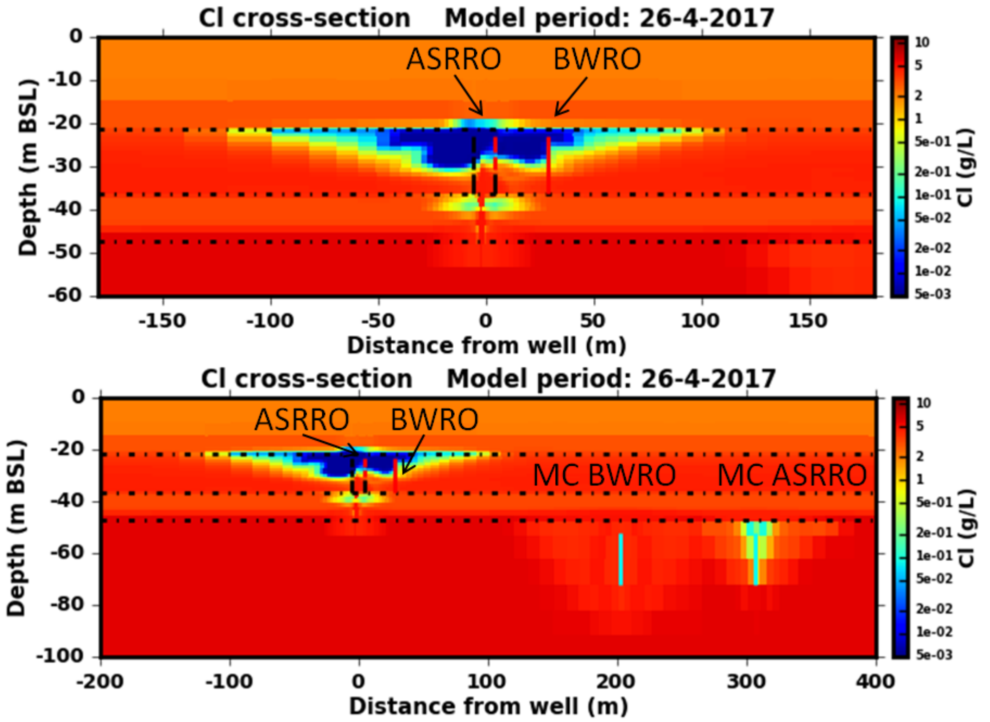


Figure 29: Final modelled chloride concentrations (26 April 2017) for cross-sections through the ASSRO wells (AW1 and AW2), both near the ASRRO (top) and an overview including membrane concentrate injection wells (bottom).

6.1.2 Water quality impacts: infiltrated rainwater

The average water quality of the infiltration water was derived by taking the average of 20 samples taken between 2012 and 2017 (Table 5). The variation of the infiltration water quality can be derived from (Figure 25 to Figure 32). The infiltrated water was typically very fresh and oxic. EC and pH are relatively constant, but the temperature has a seasonal variation. All average concentrations of the infiltrated freshwater remain below the legal limits set by the Water Act of The Netherlands in 2017, except for Zn. Zn often exceeded 100 µg/l, which resulted in an average infiltration concentration above the legal limits. The origin of Zn is the (galvanized) material on the greenhouse roofs. Since concentrations of Zn in the freshwater reaching the surrounding monitoring wells remained <10 µg/l, it was presumed that Zn was adsorbed in the vicinity of the ASR-wells. A better removal of Zn in the pre-treatment facility should, however, be attained.

Table 5: Observed infiltration water quality averaged over 20 measurements between 2012 and 2017, tabulated together with the legal limits set by the Water Act of The Netherlands in 2017. EC-25 Field is the electrical conductivity measured in the field with a reference temperature of 25°C.

Sample code	Average	Legal limits
	2012-2017	Water Act, The Netherlands, 2017
EC-25 Field (µS/cm)	38	-
Temperature (°C)	8.7	-
pH (Field)	7.1	-
DO (mg/L)	9.5	-
Na (mg/L)	5.0	120
K (mg/L)	0.3	-
Ca (mg/L)	1.9	-
Mg (mg/L)	0.7	-
Fe (mg/L)	0.0	-
Mn (mg/L)	0.1	-
NH_4 (mg NH_4/L)	0.19	3.2
Cl (mg/L)	6.5	200
SO_4 (mg SO_4/L)	2.3	150
HCO_3 (mg HCO_3/L)	7.7	-
NO_3 (mg N/L)	3.0	24.8
PO_4-t (mg P/L)	0.1	1.25
As (µg/L)	1.3	10
Zn (µg/L)	171.8	65

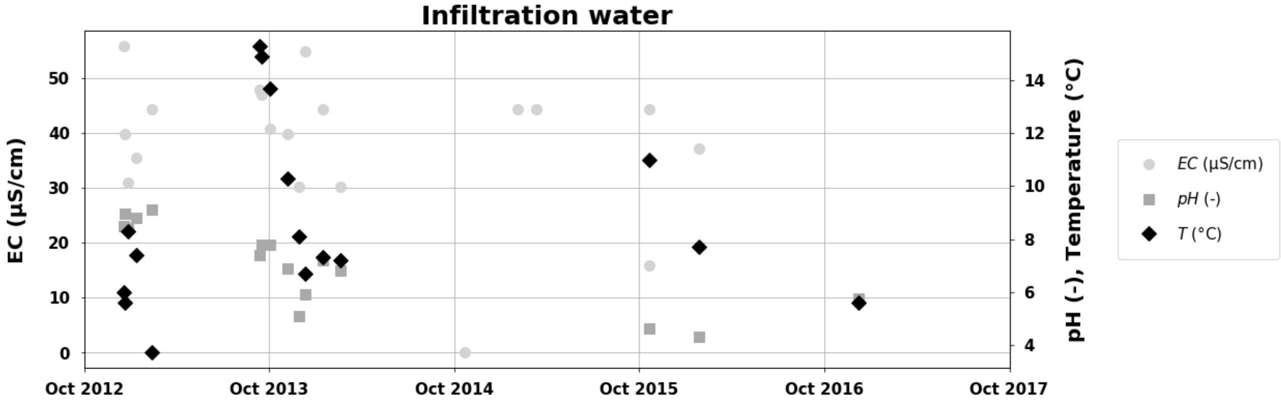


Figure 30: Electrical conductivity (EC in µS/cm), pH (-), and temperature (Temp in °C) of the freshwater used for infiltration at ASRRO Westland.

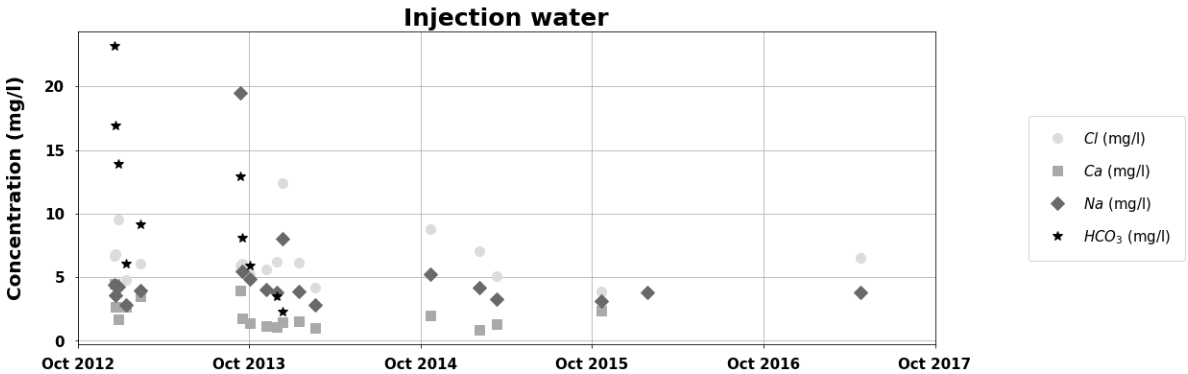


Figure 31: Concentrations of Cl, Ca, Na, and HCO_3 in the freshwater used for infiltration at ASRRO Westland.

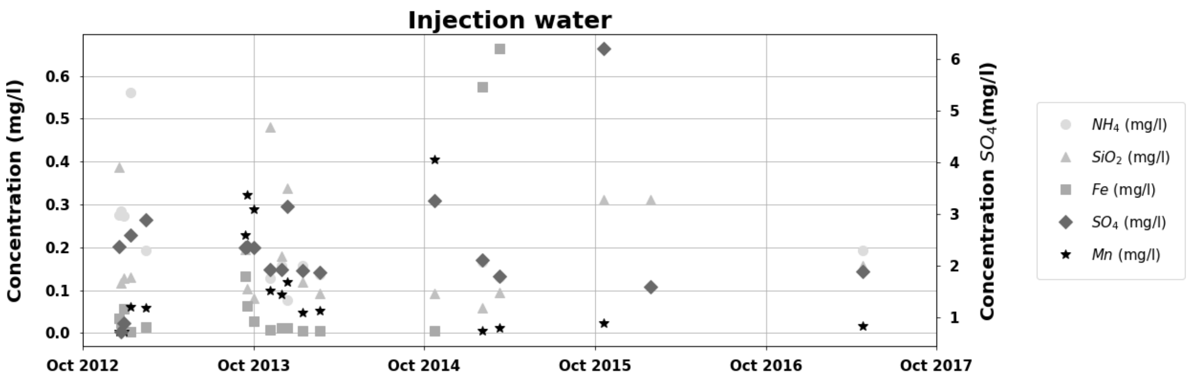


Figure 32: Concentrations of NH_4, SiO_2, Fe, SO_4, and Mn in the freshwater used for infiltration at ASRRO Westland.

The infiltration water was frequently analysed for pesticides and various heavy metals by an independent laboratory (Groen Agro Control) to comply with the obligations set in the permit. These observations underline the exceedance of Zn in the infiltration water, but also indicate that for other metals, no exceedance was observed (Table 6). All pesticides were generally absent or found only in very low concentrations. Only in June 2013, a firm exceedance of the maximum concentration for Pyrimethanil was observed.

Table 6: Observed pesticides and heavy metals in the infiltration water (measurements as required by the permit). Note: red values indicate exceedance of legal limits.

Monitoring round	Pesticides	Al	Cd	Cr	Cu	Pb	Ni	Zn
	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l
Legal limits	0.1	-	0.4	15	15	15	15	65
Jan-13	none	13.2	<0.05	8.7	2.9	0.5	1.2	42
Jun-13	Carbendazim: 0.01 Pyrimethanil: 0.60	3.5	<0.05	<0.5	1.7	0.2	1.3	29
Dec-13	Pyrimethanil: 0.06	237	0.19	5.1	2.6	0.28	8	324
Sep-14	none	13.9	<0.05	5.5	3.1	<0.7	2.1	30.5
Mar-15	Fluopyram: 0.024 Spiromesifen: 0.072 Triflumizool: 0.068	68.4	0.09	7.5	3.1	0.75	1.6	172
Sep-15	Fluopyram: 0.010 Triflumizool: 0.034	22	<0.05	1	2.3	<0.07	<0.05	110
Feb-16	Spiromesifen: 0.082 Chloorprofam: 0.01 Indoxacarb: 0.032 Propyzamide: 0.015							
Apr-16	none							
Dec-16	none							
Average		59.7	0.1	5.6	2.6	0.4	2.8	117.9

6.1.3 Water quality impacts: RO feedwater and re-injected concentrate

The average water quality of the ASRRO and BWRO feed water and re-injected concentrate was determined for samples taken between 2015 and 2017 (Table 7).

In general, both feed waters are quite alike, and mainly marked by mixing of infiltrated rainwater and brackish groundwater. Due to redox processes, this water is typically anoxic after aquifer residence. As, Fe, and Mn are slightly higher around the ASR well, presumably by redox reactions around the ASR well (see DESSIN deliverable D22.3) or potentially also by mixing with intruding saltwater from Aquifer 2 (as marked by a higher SO_4 concentration).

Zn concentrations are significantly higher at the ASRRO wells, which probably derives from the infiltration water (which has high Zn concentrations), from which Zn is adsorbed in the vicinity of the ASR wells during infiltration. Desorption during abstraction at those wells can explain the high Zn concentrations. Zn should therefore be a topic for future research in greenhouse ASR(RO) systems.

In general, the re-injected brackish water that is disposed of in Aquifer 2 is less saline than the native groundwater in the receiving Aquifer 2 (around 5000 mg/l Cl). This means that a net 'freshening' occurred in Aquifer 2 at the Westland ASRRO site. For the groundwater systems in the area, which is suffering from salinization, this is a positive effect. Again, the high concentration of Zn in the concentrate of the ASRRO-plant can have a negative impact.

Table 7: **Quality of intercepted brackish groundwater used for reverse osmosis, and of the resulting RO-reject (concentrate), averaged over measurements between 2015 and 2017. EC-25 Field is the electrical conductivity measured in the field with a reference temperature of 25°C.**

Sample code:	BWRO-feed	BWRO-concentrate	ASRRO-feed	ASRRO-concentrate
	2015-2017	2015-2017	2015-2017	2015-2017
#samples	18	11	16	9
EC-25 Field (µS/cm)	6632.3	9129.6	6896	11111
Temperature (°C)	13.1	13.6	12.6	12.9
pH (Field)	7.0	7.1	7.2	7.2
DO (mg/L)	0.0	0.0	0.0	0.0
Na (mg/L)	1073.2	1698.1	1135.9	2075.0
K (mg/L)	37.6	59.2	35.6	65.3
Ca (mg/L)	220.9	358.3	247.2	453.1
Mg (mg/L)	150.6	237.1	160.6	297.9
Fe (mg/L)	5.6	10.9	7.8	18.2
Mn (mg/L)	0.8	1.2	2.0	3.8
NH_4 (mg NH_4/L)	15.6	22.4	12.9	22.8
Cl (mg/L)	2037.2	3305.4	2221.9	3965.9
SO_4 (mg SO_4/L)	11.8	24.0	49.1	96.3
HCO_3 (mg HCO_3/L)	666.1	1000.8	534.3	861.6
NO_3 (mg N/L)	0.2	0.5	0.3	0.6
PO_4-t (mg P/L)	3.8	6.4	1.3	2.2
As (µg/L)	1.0	3.8	5.0	8.9
Zn (µg/L)	5.4	6.2	244.9	474.1

6.2 Regional impact of ASRRO

The regional impact of ASRRO in the Westland region is extensively described in Annex A : "The impact of integrated Aquifer Storage and Recovery and brackish water Reversed Osmosis (ASRRO) on a coastal groundwater system" by Steven Ros and Koen Zuurbier (published in 'Water' 2017, 9, 273).

The impact of widespread use of ASRRO on the regional Westland groundwater system was limited based on regional groundwater modelling. However, it was shown (Figure 33) that ASRRO decreased the average chloride concentration with respect to the autonomous scenario and the current use of brackish water reverse osmosis (BWRO). ASRRO was also successful in mitigating the local negative impact (saltwater plume formation, Figure 34) caused by the deep disposal of membrane concentrate during BWRO and reducing the saltwater intrusion induced by brackish water abstraction in the BWRO case.

Based on this case study, an overall positive to neutral impact of ASRRO on a coastal groundwater system is presumed, which is an improvement with respect to the use of BWRO in the same setting. ASRRO thus provides means to more sustainable use of coastal groundwater systems. However, several operational (e.g. infiltrated and recovered volumes) and hydrogeological (e.g., aquifers, aquitards, drainage levels, nearby abstractions) controlling factors will affect the overall and cumulative impact on any groundwater system and should be considered before ASRRO implementation elsewhere.

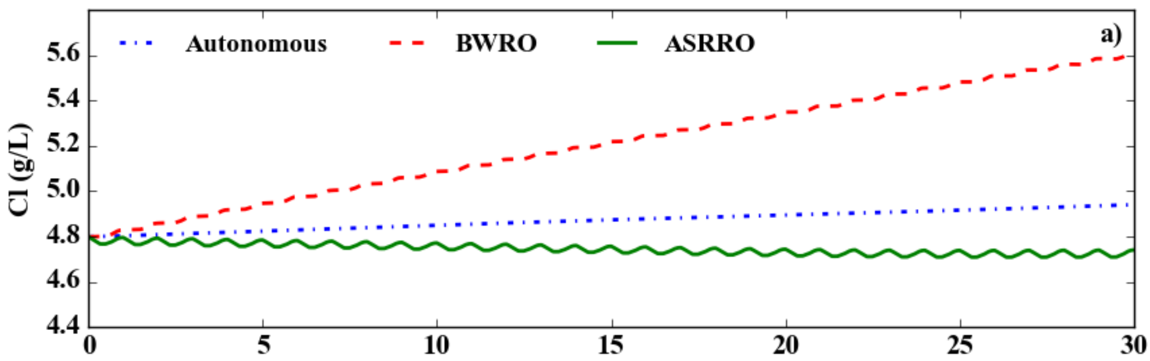


Figure 33: Overage concentrations of chloride in the Westland groundwater system during 30 years based on groundwater modelling assuming an autonomous case (no groundwater use), use of BWRO (current situation), and ASRRO (approach developed in DESSIN).

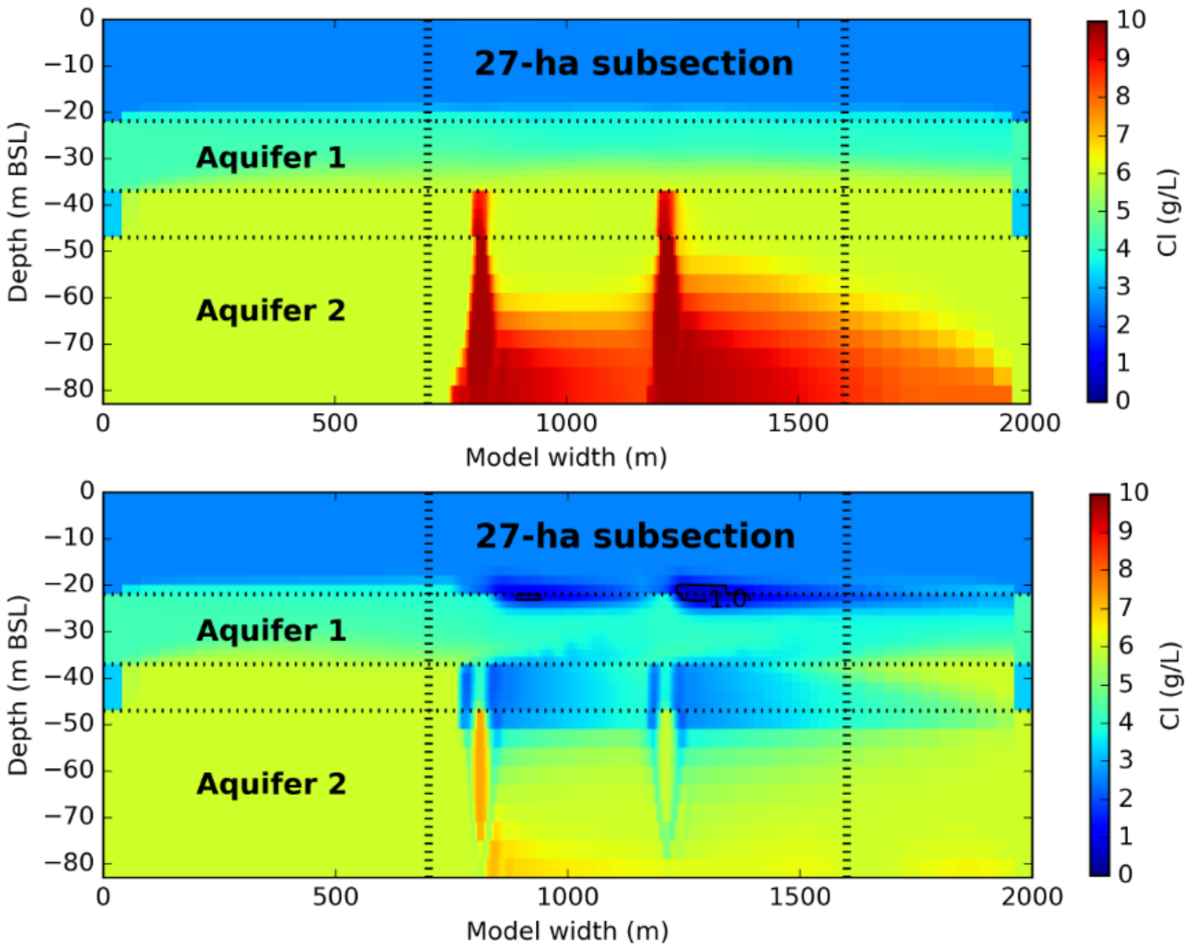


Figure 34: **Cross-section of the average concentrations of chloride in the Westland groundwater system after 30 years with typical use of BWRO (current situation), and ASRRO (approach developed in DESSIN).**

6.3 Relation with (inter)national regulation

Horticulture companies growing crops (e.g. tomatoes) with a high water demand require a secondary freshwater source, complementary to the use of rainwater. Often, horticulture companies (but also industries) in coastal areas use brackish/saline groundwater as secondary irrigation water source. Abstracted groundwater is desalinated by reverse osmosis (RO).The fresh water is used for irrigation in the greenhouses, whereas the residual fraction in which remaining soluble salts accumulate (the concentrate), is injected into the subsurface into a deeper aquifer. With this activity, two activities need legal attention:

1. Abstraction of the brackish water: a net abstraction of groundwater can result in declining groundwater levels (especially in (semi-)arid regions) and saltwater intrusion (in coastal areas, such as the Westland), which is not allowed according to the European Water

Framework Directive (WRD). The (accumulative) effects of the net abstraction(s) therefore need careful consideration.

The replacement of BWRO by ASRRO can result in a shift to a situation with a reduced or absent net abstraction and should therefore be preferred over BWRO to avoid conflicts with the WRD;

2. This disposal of produced concentrate: this can be in conflict with the standards of the WRD. Due to the injection of concentrate, the concentrations of soluble species (such as Cl, Mg, Na, Ca, Cd, Zn) may increase in the receiving aquifer. When this occurs it is in conflict with the'stand still' principles of the WRD. In the Netherlands the injection of concentrate into the subsurface is subject of policy discussions. In the Westland area the injection of brine is banned, but separate permissions can be obtained until at least 2023, only if alternatives are not available.

   ASRRO does not provide a direct alternative for the concentrate injections. However, it can prevent the possible increase of soluble salt concentrations by the infiltration of freshwater. The net result is the soluble salt concentrations in the aquifer do not increase in time. This in accordance with the EU Water Framework Directive.

At this moment the ASSRO concept is worked for the Westland region by the idea of "The Waterbank". The principle of 'The Waterbank' idea is that the salinity increase in the aquifer due to concentrate disposal is mitigated by large-scale and organized infiltration of rainwater and potentially other freshwater surpluses. The use of reverse osmosis can be allowed in such a case, but only will be charged to finance the infiltration facilities.

7 Economic impact of ASRRO

The economic impact of the various water supply alternatives was evaluated using an economical tool set up by KWR (Figure 35). This integrated approach includes all relevant aspects with respect to operational and capital expenditures, including tax shield, discount, and subsidies. Those aspects were included for the Dutch situation. Here, a subsidy (MIA/Vamil) is available for infiltration systems, which lowers the CAPEX of ASRRO. The economic lifespan of the main components of ASRRO is 20 years and is in line with the lifespan of a greenhouse. All re-investments needed to reach this lifespan were taken into account for each alternative. A standard size supply for the Westland area was assumed (Table 8).

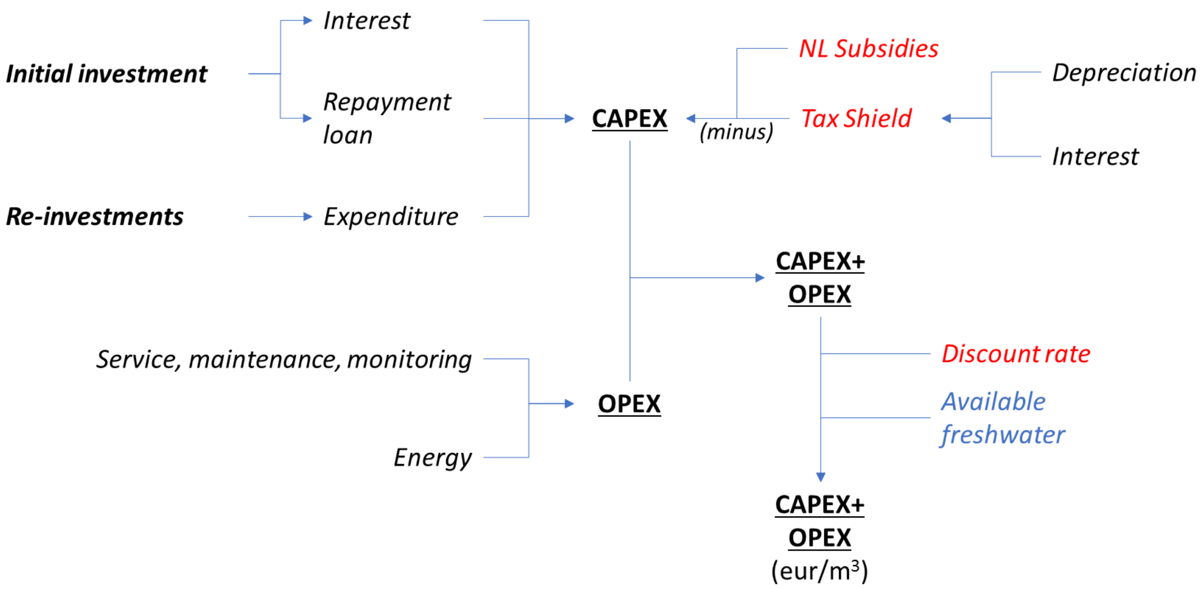


Figure 35: Approach to assess cost price for ASRRO, BWRO, and a basin. Aspects that can be varied are indicated in red.

Table 8: Main operational assumptions for cost price analysis

Assumptions		
Freshwater made available	30 000	m^3/yr
Freshwater infiltrated (during ASRRO)	30 000	m^3/yr
Capacity	25	m^3/h
Unmixed recovery (during ASRRO)	10 000	m^3/yr
Desalinated (during ASRRO)	20 000	m^3/yr
Energy price	0.12	euro/kWh
Economic lifespan	20	yrs

Of the alternatives examined, BWRO is the current and cheapest alternative in the Westland area. Basins storing more of the rainwater surplus are unattractive due to large claim on aboveground land, resulting in a high loss of income. ASRRO is 0.06 eur/m^3 more expensive than conventional BWRO. The difference between both alternatives is reduced by the Dutch subsidy for sustainable investments (MIA/Vamil). The higher price for ASRRO water is caused by the higher initial investment (CAPEX). Lower energy consumption as a result of a reduction of the desalination only partly mitigates this higher CAPEX.

Based on the limited difference in cost price per m^3 and the increased sustainability, ASRRO is considered a competitive source for irrigation water supply.

Table 9: Cost price of produced water with BWRO, ASRRO, and after storage in a basin.

CAPEX + OPEX	BWRO	ASRRO	Basin
	eur/m^3	*eur/m^3*	*eur/m^3*
+ 3% discount rate + tax shield + NL subsidy	0.64	0.70	1.37
+ 3% discount rate + tax shield	0.64	0.73	1.37
+ 3% discount rate	0.68	0.77	1.46
none	0.88	0.98	1.96

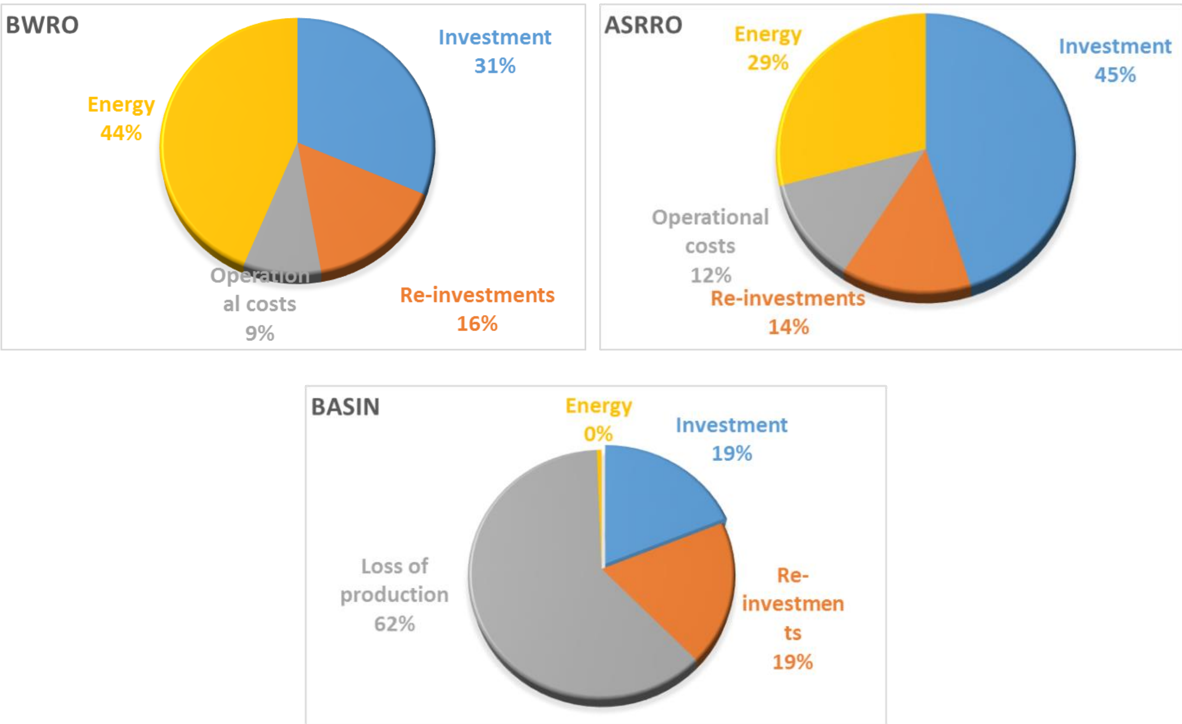


Figure 36: Build-up of the cost-price per alternative

8 Conclusions

From 2014 to 2017, a pilot was conducted at the Westland Demo site in order to integrate ASR, the Freshkeeper, and desalination in one system. The objective was to create a sustainable and robust freshwater supply, using the characteristics of the aquifer as an ecosystem service. This integrated 'ASRRO' system must improve the freshwater production from conventional ASR, while mitigating the negative impact of brackish water reverse osmosis.

The results at the demo site indicate that ASRRO is technically viable and beneficial. Freshwater surpluses up to 70 000 m^3/ 6 months could be treated, stored, and partially recovered for direct use (22.5% of the stored water). Additional freshwater could be produced by abstracting the mixed freshwater and saline water and subsequently treating this with RO. This created a high-quality freshwater stream and a waste stream having a quality comparable to the native groundwater in a deeper aquifer.

The biggest operational threat during ASRRO in a sand aquifer (as present at the Westland site) is clogging of RO-membranes and potentially also the saline water re-injection well(s). This is caused by mobilization of clay particles (during freshening) and formation of Fe-colloids (by infiltration of oxic water in an area with adsorbed Fe around the ASRRO wells). Both processes occur in the infiltration stage. Careful abstraction of the brackish water in deeper zones of the aquifer and regular flushing of the RO membranes are potential mitigation strategies to mitigate membrane clogging.

An overall positive to neutral impact of ASRRO on a coastal groundwater system is presumed, which is an improvement with respect to the use of BWRO in the same setting. ASRRO thus provides means to more sustainable use of coastal groundwater systems. However, several operational (e.g. infiltrated and recovered volumes) and hydrogeological (e.g., aquifers, aquitards, drainage levels, nearby abstractions) controlling factors will affect the overall and cumulative impact on any groundwater system and should be considered before ASRRO implementation elsewhere.

Albeit more expensive, the use of ASRRO is considered competitive with the current BWRO. The cost price per m^3 is 0.06 eur/m^3 higher (0.70 versus 0.64 m^3) as a result of higher CAPEX. Both alternatives are economically more interesting than aboveground storage (in basins).

An overview of the outcomes of the different tasks in T33 is presented in Table 10.

Table 10: Outcomes of the different tasks in T33

Task	Description	Outcome DESSIN
33.1	Quantification of the freshwater recovery by an innovative well design.	Conventional ASR in the typical Westland saline aquifer results in ASR recovery efficiencies <30%. This can be lifted to >50% with the innovative well design and even more by the use of RO.
33.2	Demonstration of the added value of an advanced ASRRO system.	The advanced ASRRO system showed capable of 1) enlarging the recovery of unmixed freshwater upon storage, 2) provided a more robust water supply thanks to the use of RO and 3) can attain a neutral water balance to prevent mining of water from a coastal aquifer
33.3	Demonstration of the effect of enhanced subsurface iron removal on membrane clogging	Clogging of membranes (and potentially: re-injection wells) during ASRRO appears to be driven by mobilization of clay particles and Fe-colloids. This can be mitigated by regular flushing of the RO-membranes with permeate and regular cleaning of the re-injection well
33.4	Demonstration of the impact of the Westland ASR/RO pilot on the regional groundwater quality	The impact of widespread use of ASRRO on the regional Westland groundwater system was limited based on regional groundwater modelling but it was shown that ASRRO decreased the chloride concentration with respect to the autonomous scenario and the use of brackish water reverse osmosis (BWRO). ASRRO was successfull in mitigating the local negative impact (saltwater plume formation) caused by the deep disposal of membrane concentrate during BWRO. Based on this case study, an overall positive to neutral impact of ASRRO on a coastal groundwater system is presumed, which is an improvement with respect to the use of BWRO in the same setting. ASRRO thus provides means to more sustainable use of coastal groundwater systems.

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ANNEX A: Scientific analysis of impact of ASRRO on a groundwater system

water

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Article

The Impact of Integrated Aquifer Storage and Recovery and Brackish Water Reverse Osmosis (ASRRO) on a Coastal Groundwater System

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Abstract: Aquifer storage and recovery (ASR) of local, freshwater surpluses is a potential solution for freshwater supply in coastal areas, as is brackish water reverse osmosis (BWRO) of relatively shallow groundwater in combination with deeper membrane concentrate disposal. A more sustainable and reliable freshwater supply may be achieved by combining both techniques in one ASRRO system using multiple partially penetrating wells (MPPW). The impact of widespread use of ASRRO on a coastal groundwater system was limited based on regional groundwater modelling but it was shown that ASRRO decreased the average chloride concentration with respect to the autonomous scenario and the use of BWRO. ASRRO was successful in mitigating the local negative impact (saltwater plume formation) caused by the deep disposal of membrane concentrate during BWRO. The positive impacts of ASRRO with respect to BWRO were observed in the aquifer targeted for ASR and brackish water abstraction (Aquifer 1), but foremost in the deeper aquifer targeted for membrane concentrate disposal (Aquifer 2). The formation of a horizontal freshwater barrier was found at the top of both aquifers, reducing saline seepage. The disposal of relatively fresh concentrate in Aquifer 2 led to brackish water outflow towards the sea. The net abstraction in Aquifer 1 enforced saltwater intrusion, especially when BWRO was applied. The conclusion of this study is that ASRRO can provide a sustainable alternative for BWRO.

Keywords: aquifer storage and recovery; ASR; ASRRO; BWRO; membrane concentrate disposal; ASR performance; coastal groundwater system; short-circuiting; freshwater barrier

1. Introduction

Coastal areas are often marked by high freshwater demands and a low freshwater availability. Use of aquifer storage and recovery (ASR; [1,2]) of temporary freshwater surpluses and brackish water reverse osmosis (BWRO; [3]) are potential techniques to improve the freshwater availability in coastal areas. ASR is a cost-effective, readily applicable technique to store large water volumes without the occupation of large surface areas. Nevertheless, the performance of ASR, which is marked by the recovery efficiency (RE: the percentage of freshwater that can be recovered upon storage), can be very limited in coastal areas [4]. The main cause for the reduced RE are the buoyancy effects induced by the difference in density of the brackish or saline native groundwater (high density), and the injected freshwater (low density). This leads to early salinization at the bottom of the ASR well [5–7]. On the other hand, BWRO is a proven technology able to continuously desalinate groundwater with a wide range of salinities, while its costs are acceptable for various end users. However, BWRO is accompanied by a saline waste stream (membrane concentrate: 'MC'). Disposal of this MC often occurs into (deeper) aquifers or local surface waters, which can lead to environmental pollution and/or groundwater salinization.

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A more sustainable and reliable freshwater supply can potentially be achieved by combining ASR and RO in one system ('ASRRO', Figure 1). In such a set-up, a balance is obtained between freshwater injection (wet periods) and recovery (dry periods) via a combination of ASR and BWRO-treatment. Doing so, the abstraction of brackish groundwater at the base of the ASR target aquifer for BWRO may improve the direct recovery of freshwater by shallower wells [8] and thus the RE of ASR. The deep well intercepting brackish water is called a 'Freshkeeper' [9]. The integrated approach of ASRRO may provide a much more robust and sustainable freshwater supply than each of the independent techniques, as more freshwater is recoverable for direct use, such that less water requires (energy-consuming) desalination by RO. Additionally, overexploitation of the groundwater may be counteracted by compensating freshwater production with artificial recharge.

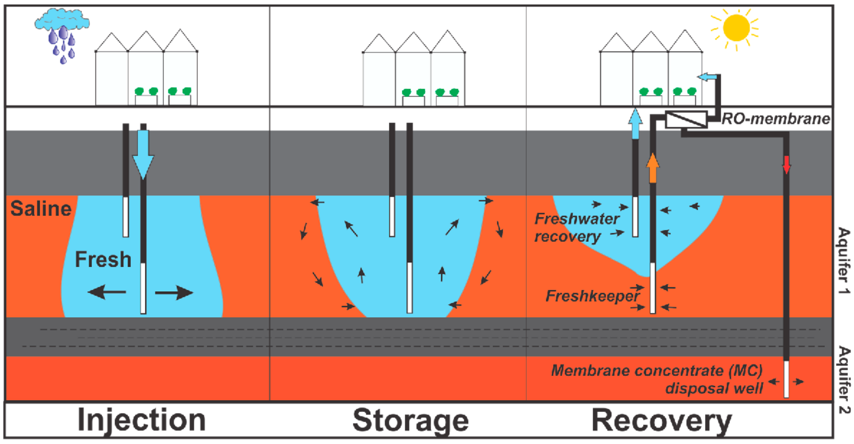

Figure 1. The principle of aquifer storage and recovery and reverse osmosis (ASRRO) for storage and recovery of freshwater in brackish-saline aquifers.

A first ASRRO system was constructed in 2012 and tested as a conventional ASR-system in the first year (2013), while the Freshkeeper well was added in 2014 [10]. Since 2015, the system is operating as a complete ASRRO system. In the first studies, the focus was on the direct recoverability of the injected freshwater [10] and potential clogging of the RO-membrane [11]. However, the impact of ASRRO on the local and regional groundwater system was not evaluated. The aim of this study is therefore to analyse the effects of ASRRO on the groundwater quality in a coastal area, both on a local and a regional scale. The design and operation of ASRRO were derived from a field pilot, situated centrally in the study area [10]. Numerical modelling was performed to assess the water quality development of the Regional groundwater system.

2. Methods

2.1. Study Area

The Westland area in The Netherlands is the country's largest intensive greenhouse horticultural area and situated within 10 km of the North Sea shoreline (Figure 2). It suffers from a significant mismatch between water demand and availability in the horticultural greenhouse sector, presence of brackish and saline groundwater, current use of BWRO, and saltwater intrusion [12]. The horticultural sector in the area requires irrigation water with an extremely low salinity of <0.5 mmol/L [4]. Surface water and drinking water generally fail to meet this water quality limit. Therefore, rainwater is harvested via the greenhouses' roofs and partly stored in aboveground basins. A mismatch in precipitation and water demand, however, results in discharge of a significant part of the available rainwater in wet periods [4]. Therefore, BWRO is required in summers as additional fresh irrigation water supply. Greenhouse owners in the area using BWRO abstract the required brackish groundwater from the upper Aquifer 1 (10–50 m BSL) and dispose of MC in the deeper Aquifer 2 (40–120 m BSL).

Because of the presence of confined, unconsolidated sand aquifers in the shallow subsurface (10–50 m BSL), ASR is a viable option to bridge the periods of rainwater availability and demand [4]. However, although the upper aquifer is the least saline target aquifer for ASR available, the predicted RE of ASR is generally <50% in the Westland area [4]. Therefore, attempts are being made to improve the RE of ASR via independently operated multiple partially penetrating wells in a single borehole (MPPW, [13]).

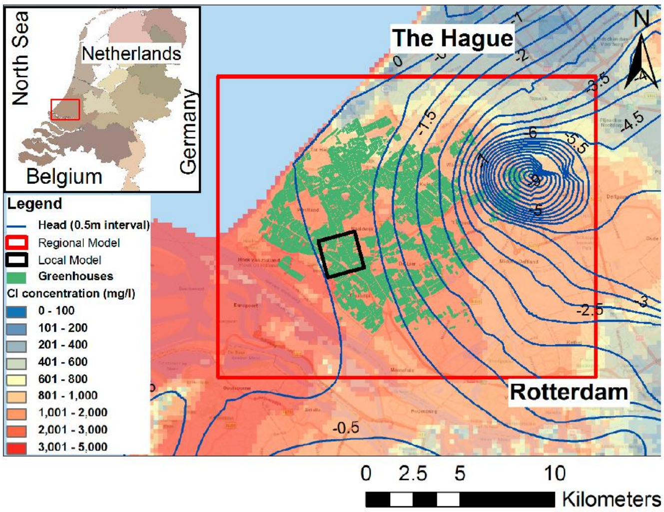

Figure 2. Location of the Westland study area and the regional hydraulic heads, groundwater salinity, location of greenhouse horticulture, and boundaries of the groundwater models.

2.2. Numerical Modelling of ASRRO Application

Since ASRRO is still in a piloting phase and observation of all complex interactions is hard in the field, the impact on the groundwater quality in the area was assessed by numerical modelling. A two-stage approach was applied by first modelling two individual ASRRO systems in a horizontal layer model to study their performance and interaction on a local scale (based on the field pilot: Local Model), followed by modelling of the widespread use of ASRRO in the Westland region (Regional Model). In both cases, SEAWAT Version 4 [14] was used. FloPy [15] was used to generate the models' input and output and to frequently calculate the resulting MC concentration, which provided input to the Source/Sink package in the subsequent stress period.

2.2.1. Modelling Local Impacts (Local Model Based on ASRRO Pilot)

A half-domain (2 × 0.5 km) local model was set up and comprises two ASRRO systems, of which the design and setting was based on the general design, operation, and setting of the first ASRRO field trial [10]. The main model parameters are shown in Table 1 and are based on an extensive ASR pilot in the target sand aquifer in the Westland area, which included calibrated groundwater modelling [10]. The parameterisation was considered representative for the target area. A dispersivity coefficient of 0.1 m was assigned based on the same pilots in the same aquifer [10,13] and a diffusion coefficient of 8.64×10^{-5} m^2/day was assigned based on [10,16]. The grid size varied spatially and was most refined near the wells, i.e., either 5 m (x) × 5 m (y) × 1.5 m (z) at the MPPWs and 5 m × 5 m × 4 m at the membrane concentrate disposal wells (MC disposal well). Western and eastern boundaries were assigned a general head boundary (GHB). The GHB simulated a constant head at a distance of half a cell length (L) from the model boundaries (20 m), using the heads marked in Figure 3 and assuming unchanged horizontal conductivity (K_h). The GHB conductance (C_{GHB}) depended on the boundary cells' width (W) and thickness (D) perpendicular to the regional flow direction and was calculated using

$$C_{GHB} = K_h \cdot \frac{W \cdot D}{0.5 \cdot L} \quad (1)$$

Concentrations at these boundaries were kept constant and were based on [10]. A hydraulic gradient of −0.008 m/m induced background flow as indicated in Figure 3 and was based on the regional heads (Figure 2). Recharge by precipitation was not considered because the target areas for ASRRO are typically covered with greenhouses clusters (capturing rainfall, which is infiltrated with the ASRRO systems) and infiltration through the thick clay cover is limited.

An autonomous case without wells was for comparison with ASRRO and BWRO cases. Subsequently, two scenarios were evaluated with transient models: one with the ASRRO systems (infiltration of winter surplus equals combined freshwater production from ASR and RO (+MC disposal) in summer), the other with the conventional BWRO systems (no infiltration in winter) to produce the same volume of freshwater solely from BWRO in summer while reinjecting MC. The average winter precipitation surplus available for ASR is around 200 mm/year or 2000 m³/ha of greenhouse [17]. The impact of ASRRO systems was determined for a predefined 27-ha greenhouse area (Figure 3), which simulates 2 ASRRO systems in a half-domain with areas equal to the area connected to the pilot ASRRO system [10]. Accordingly, 54,000 m³/year of freshwater was injected by MPPWs (coded 'a–d') in Aquifer 1 in winter and abstracted in the next summer after a 59-days' storage period, The operational schemes of the BWRO and ASRRO cases are shown in Table 2. Unmixed freshwater for direct use was recovered first during ASRRO. Upon salinization of the recovery wells, the water was directed to the RO to produce freshwater. The MC was injected into Aquifer 2 via the MC disposal wells. An RO efficiency of 50% (maximal achievable efficiency without anti-scalants) was assumed in both scenarios, resulting in an equal production of freshwater and MC. In total, a net volume of 54,000 m³ of freshwater is produced from Aquifer 1 during both ASRRO and BWRO each year. This is ca. 4.4% and 1.6% of the pore water volume present in Aquifer 1 (1,215,000 m³) and Aquifer 2 (3,402,000 m³) in the 27-ha subsection.

Table 1. Local Model main parameter values.

Geological Layer	Depth	Layers	Porosity, n	K (Horizontal)/K (Vertical)	Storativity, S	Starting Conc
	(m BSL)	(Thickness)	(-)	(m/day)	(-)	Cl (mg/L)
Holocene clay cap	0–22	11 (2 m)	0.2	0.1/0.01	0.001	2550
Aquifer 1	22–37	10 (1.5 m)	0.3	35.0/35.0	1×10^{-6}	4300
Aquitard 1	37–47	5 (2 m)	0.2	0.2/0.02	0.001	3300
Aquifer 2	47–83	9 (4 m)	0.35	30.0/30.0	1×10^{-6}	6000

Table 2. Operational scheme of each of the 4 MPPWs (a–d) in the Local Model.

BWRO	Time	Volume	Rate	ASRRO	Time	Volume	Rate
	(Days)	(m³)	(m³/Day)		(dAys)	(m³)	(m³/Day)
Idle period	182	0	0	Winter injection	123	13,500	109.8
				Storage period	59	0	0
Summer abstraction	153	13,500	88.2	Summer abstraction	153	13,500	88.2
Idle period	30	0	0	Idle period	30	0	0

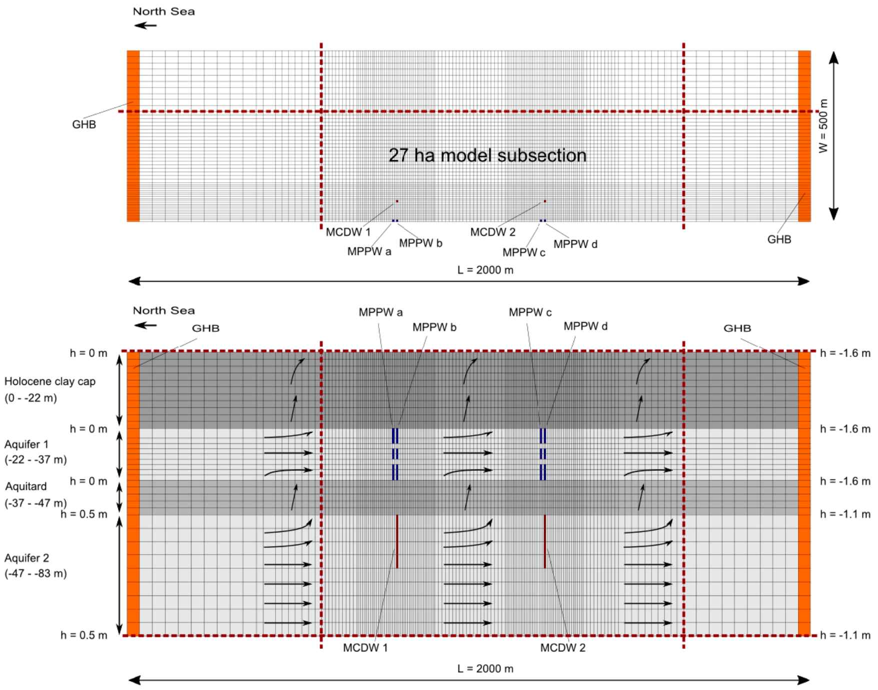

Figure 3. Top: Top view of the Local Model grid. Bottom: Cross-sectional view (West-East) showing the hydraulic heads (h) and the expected flow pattern through the various geological layers for the autonomous situation. The 27-ha subsection (area of the greenhouses connected to the ASRRO system) of the model is located within the marked box. The vertical exaggeration is 10:1.

2.2.2. Modelling of Regional Impact (Regional Model)

Numerous ASRRO systems to supply freshwater in the greenhouse area were modelled to assess the cumulative impacts on hydrological processes further away from the systems (such as saltwater intrusion) and to assess the relative impact of the application of ASRO and BWRO on its current scale with respect to the whole groundwater system. The 3D Regional Model (18 × 16.5 km) covers the Westland groundwater system (Figure 1). The model is based on the PZH (Province of Zuid-Holland) model used and described in earlier studies [10,18]. The main model parameters are shown in Table . Dispersivity and diffusion coefficients are equal to the Local Model. Horizontal cell dimensions are 50 × 50 m. Layer thicknesses are constant in the top 80 m BSL, and vary spatially at greater depths (Figure A1). This results in 1,439,904 active cells. The clay cap confining the upper aquifer (Aquifer 1) is 1 to 3 model layers thick (Figure A2). Aquifer 1 is 3 to 5 model layers thick. The Regional Model's vertical conductivity (Kv) depends on the horizontal conductivity (Kh). The vertical Kh/Kv-anisotropy equals 10 if the Kh is equal to or below 1 m/day, 3 if the Kh is 1 to 10 m/day, and 2 if the Kh is in between 10 and 30 m/day. Grid cells with a higher Kh have been made isotropic. The Kh and Kv distribution of the Phreatic layer, Aquifer 1, and Aquifer 2 are shown as supplementary information (Figure A3 and A4), as well as the Kh of the Clay Cap and Aquitard 1 (Figure A5).

Constant heads were assigned to both the top layer and the vertical boundary planes (Figure A6). The model bottom is a no-flow boundary, since this was considered to be the hydrological base. No constant concentrations were assigned. Starting concentrations and heads decrease landwards, where deep polders are present and groundwater has lower salinities (for more information, Figure A7 and A8).

The BWRO and ASRRO scenarios include 616 ASRRO wells and 616 MC disposal wells in a grid. ASRRO wells are placed in Aquifer 1 at 20–35 m BSL, 500 m apart. MC disposal wells are placed in Aquifer 2 at 50–80 m BSL, 250 m downstream of each accompanying ASRRO or BWRO well. The annual freshwater injection and recovery equals 8885 m^3/year/MPPW, corresponding to a 4.5-ha greenhouse surface per MPPW, which is considered realistic based on comparable existing ASR systems further inland [4]. The durations of injection, storage, and recovery periods are similar to the Local Model. In total, a net volume of 5,473,000 m^3 of freshwater is produced from Aquifer 1 during both ASRRO and BWRO each year. This is ca. 0.4% and 0.1% of the pore water volume present in Aquifer 1 (1.46 km^3) and Aquifer 2 (5.38 km^3), respectively. This means that the ratio of produced versus present groundwater is 11 and 16 times less than in the 27-ha subsection of the Local Model in Aquifer 1 and Aquifer 2, respectively.

Table 3. Regional Model main parameter values. The * indicates that the number of model layers in which the geological layer is present, is spatially variable. The 'var' indicates that the model layer thicknesses vary spatially.

Geological Layer	Depth (m BSL)	Layers (Thickness)	Porosity, n (-)	K (Horizontal) (m/Day)	Storativity, S (-)
Phreatic	0–5	L1 (5 m)	0.3	0.25–75	0.1
Holocene clay cap	5–20 *	L2–4 (5 m)	0.3	<1	0.001
Aquifer 1	10–35 *	L3–7 (5 m)	0.3	9–75	0.001
Aquitard 1	35–40	L8 (5 m)	0.3	0–0.01	0.001
Aquifer 2	40–135	L9–10 (5 m), 11–13 (10 m), 14 (var)	0.3	1–5	0.001
Aquitard 2	100–154	L15 (var)	0.3	0.001–0.002	0.001
Aquifer 3	114–272	L16–17 (var)	0.3	0.1–1	0.001

3. Results

3.1. Local Impacts of ASRRO and BWRO

3.1.1. Relative Concentration Changes and Concentration Profiles

The absolute and relative concentration changes in the 27-ha subsection for ASRRO and BWRO are given in Table 4 and Figure 4 In the autonomous scenario, slow salinization was observed, primarily in the clay layers. Applying ASRRO decreased the subsection's average salinity with 3.7% with respect to the autonomous situation. The formation of an extensive saltwater plume around the MC disposal wells was not observed during ASRRO (Figure 5). During the first part of each summer abstraction, the injected MC was less saline than the ambient groundwater and subsequently moved upwards through Aquitard 1. Near the end of recovery stages, however, the MC was relatively saline and sank to the basal part of Aquifer 2. Unrecoverable injected freshwater moved upwards in Aquifer 1 and was trapped below the Clay cap. Although stratification in groundwater qualities was introduced during ASRRO, the overall effect on the groundwater quality is neutral to positive.

BWRO in combination with local MC disposal by MC disposal wells increased the average salinity by 15.5% with respect to the autonomous situation (Table 4). Saltwater plumes formed and merged around the MC disposal wells in Aquifer 2, obtaining a combined length of 1200 m and width of 300 m in 30 years (Figure 6). During BWRO, Aquifer 1 suffered from upconing of saline groundwater from Aquifer 2. This was marked by an increase in the chloride concentration of the abstracted RO feed water from 4.3 to 5 to 6 g/L and can be also seen in Figure 6 by the higher salinities at the base of Aquifer 1 near the BWROs in the 27 ha subsection. Outside the 27-ha subsection, changes were limited. The overall impact of BWRO on the groundwater system based on the outcomes is negative due to the general salinization introduced.

Table 4. Average concentrations within each geological layer throughout the 27-ha subsection after 30 years' time, for the autonomous situation, ASRRO, and BWRO; the relative concentration increase (in %) of the ASRRO and BWRO cases, and the relative concentration increase (in %) of the ASRRO case compared to BWRO.

Geological Layer	Autonomous Concentration (g/L)	ASRRO Concentration (g/L)	Rel. Conc Change ASRRO (%)	BWRO Concentration (g/L)	Rel. Conc Change BWRO (%)	Rel. Conc Change ASRRO Compared to BWRO (%)
Clay cap	2.67	2.63	−1.5	2.70	+1.2	−2.7
Aquifer 1	3.91	3.97	+1.4	4.71	+20.3	−15.8
Aquitard 1	5.07	4.60	−9.3	6.03	+18.7	−23.6
Aquifer 2	6.00	5.73	−4.5	6.97	+16.2	−17.8
Total	4.90	4.72	−3.7	5.66	+15.5	−16.7

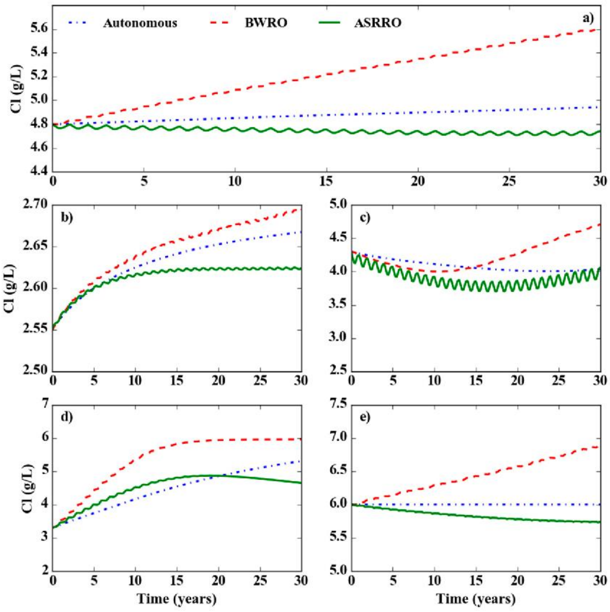

Figure 4. Average chloride concentration (g/L) throughout the 27-ha model subsection for: the total groundwater system (**a**), the Clay cap (**b**), Aquifer 1 (**c**), Aquitard 1 (**d**), and Aquifer 2 (**e**).

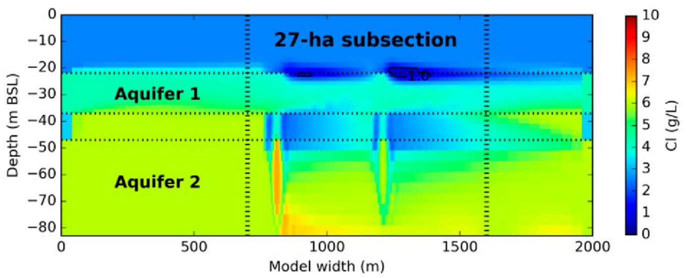

Figure 5. Chloride concentrations along the mirror plane (W- > E) of Local Model where the MC disposal wells are situated (scenario ASRRO; t = 30 year, end of summer). The 27-ha model subsection includes the part shown between the vertical dotted lines (x = 70; x = 160).

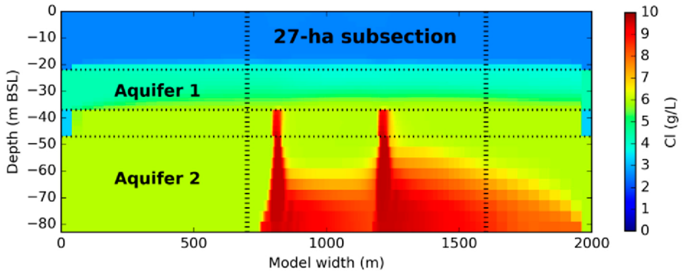

Figure 6. Chloride concentrations along the mirror plane (W- > E) of the Local Model where the MC disposal wells are situated (scenario BWRO; *t* = 30 year, end of summer). The 27-ha model subsection includes the part shown between the vertical dotted lines (*x* = 70; *x* = 160).

3.1.2. Concentration of the Membrane Concentrate

During ASRRO, MC concentrations were initially close to zero and increased to >6 g/L in the final stages of MC injection (Figure 7). On average, the concentration of the disposed water (Cl: 3–4 g/L) was below the initial chloride concentration of Aquifer 2 (6.0 g/L). The water quality of MC reinjected by the downstream MC disposal well 2 (Cl: 3.6 g/L) was slightly better than for MC disposal well 1 (Cl: 3.8 g/L). The injected chloride mass during ASRRO was 47% to 63% lower compared to the BWRO case. In the BWRO case, therefore, disposal of MC by the MC disposal wells (Cl: 7.5–11.6 g/L) significantly exceeded the ambient chloride concentration in Aquifer 2, which was 6 g/L. The average MC disposal concentrations were 8.6 g/L (MC disposal well 1) and 8.7 g/L (MC disposal well 2).

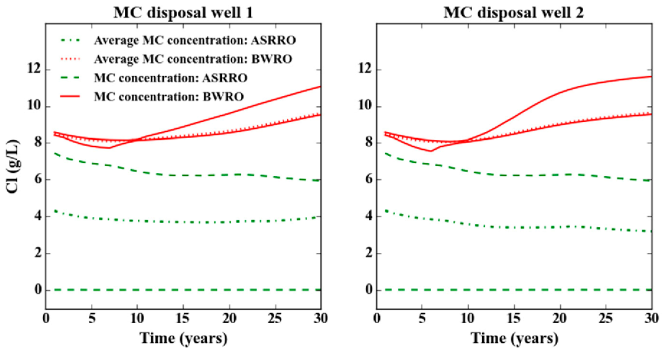

Figure 7. Range of MC concentrations with time of water injected by MC disposal well 1 and MC disposal well 2, for ASRRO and BWRO; and the yearly averaged MC concentration of each MC disposal well for both scenarios.

3.2. Regional Impacts of Wide-Spread Implementation of ASRRO and BWRO

3.2.1. Regional Concentration Changes and Concentration Profiles

Modelling the wide-spread implementation of ASRRO and BWRO in a regional model enabled analyses of the cumulative effects on the groundwater system. The average chloride concentrations of the ASRRO and BWRO scenarios after 30 years are shown in Table 5). Both ASRRO (−0.3%) and BWRO (0.0%) had a minor effect on the average chloride concentration in the whole regional groundwater system, which can be related to the fairly limited freshwater production from the aquifer compared to the modelled domain. ASRRO led to a limited freshening of the Clay cap and

Aquifer 1, but lowered the average chloride concentration within Aquifer 2 by 1.0%. BWRO decreased the average chloride concentration in the Clay cap and Aquifer 1, while increasing the concentrations in Aquitard 1. A concentration increase (+0.2%) was observed in Aquifer 2, which was targeted for MC injection.

Table 3. Average concentrations within each geological layer after 30 years' time for the Westland_auto, Westland_BWRO, and Westland_ASRRO; the relative concentration increase (in %) of the BWRO and ASRRO cases, and the relative concentration increase (in %) of the ASRRO case compared to BWRO.

Geological Layer	Autonomous Concentration	ASRRO Concentration	Rel. Conc Change	BWRO Concentration	Rel. Conc Change	Rel. Conc Change Compared to BWRO
	(g/L)	(g/L)	(%)	(g/L)	(%)	(%)
Phreatic layer	1.06	1.06	0.0	1.06	−0.2	+0.2
Clay cap	1.15	1.13	−2.0	1.10	−4.9	+3.0
Aquifer 1	1.61	1.60	−0.7	1.59	−1.0	+0.3
Aquitard 1	1.95	1.97	+0.9	1.98	+1.6	−0.7
Aquifer 2	4.58	4.53	−1.0	4.58	+0.2	−1.2
Aquitard 2	7.78	7.77	−0.1	7.77	−0.1	+0.0
Aquifer 3	10.49	10.49	+0.0	10.49	+0.0	+0.0
Total	6.77	6.75	−0.3	6.77	+0.0	−0.2

The averaged chloride concentrations after 30 years are given for the grid cells containing the 616 MPPWs or abstraction wells in Aquifer 1 (Figure 8) and MC disposal wells in Aquifer 2 (Figure 9). The average chloride concentration in the autonomous scenario was 1.8 g/L and 5.2 g/L near the MPPWs and MC disposal wells, respectively. ASRRO decreased local chloride concentrations near the wells in Aquifer 1 by 0.7 g/L (−41%) and by 1.0 g/L in Aquifer 2 (−20%) with respect to autonomous scenario, causing a shift to lower salinity classes. In the vicinity of the MC disposal wells of the ASRRO systems, concentrations were in the narrow range of 1.5–4.5 g/L Cl (Figure 9).

In the BWRO scenario, local chloride concentrations in the grid cells of the abstraction wells in Aquifer 1 decreased with 0.3 g/L (−18%) with respect to the autonomous situation and with 0.2 g/L near the MC disposal wells (−3%). The concentrations near the MC disposal wells of BWRO systems were predominantly in the range of 2.5–6 g/L, with a clear peak around 3.0–4.5 g/L Cl, whereas in the autonomous scenario these concentrations were generally in the range of 1.5 to 6.5 g/L Cl (Figure 9).

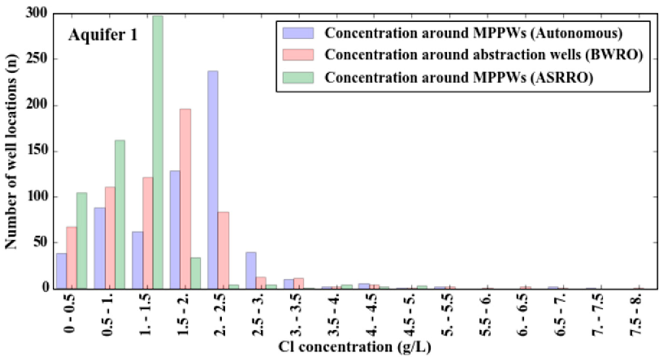

Figure 8. Distribution of local chloride concentrations in the 50 m × 50 m model cells of the MPPWs (Aquifer 1) of each individual BWRO and ASRRO system (average during the final year (year 30)).

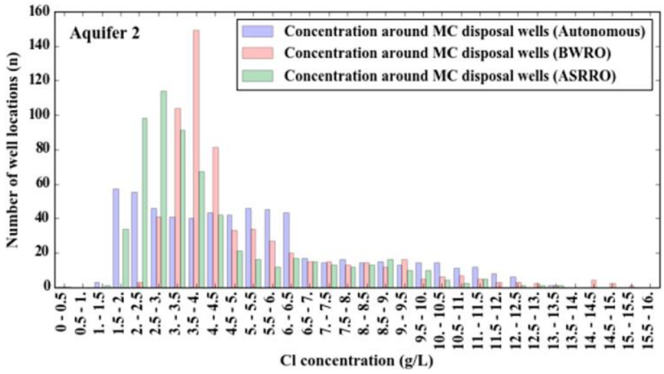

Figure 8. Distribution of local chloride concentrations in the 50 m × 50 m model cells of the MC disposal wells (Aquifer 2) of each individual BWRO and ASRRO system (average during the final year (year 30)).

Chloride concentration changes in Aquifer 1 and 2 at the end of the 30-years simulations for both ASRRO and BWRO with respect to the autonomous situation are shown in Figure . The absolute concentrations are presented as Figure A9. The impact of the BWRO and ASRRO systems is still relatively local after 30 years. However, ASRRO systems significantly reduced the salinization of Aquifer 2 that was locally occurring during BWRO. Differences were less pronounced within Aquifer 1. Here, it was clear that ASRRO led to less saltwater intrusion along the North Sea shore, as indicated by a narrower strip with strong salinization.

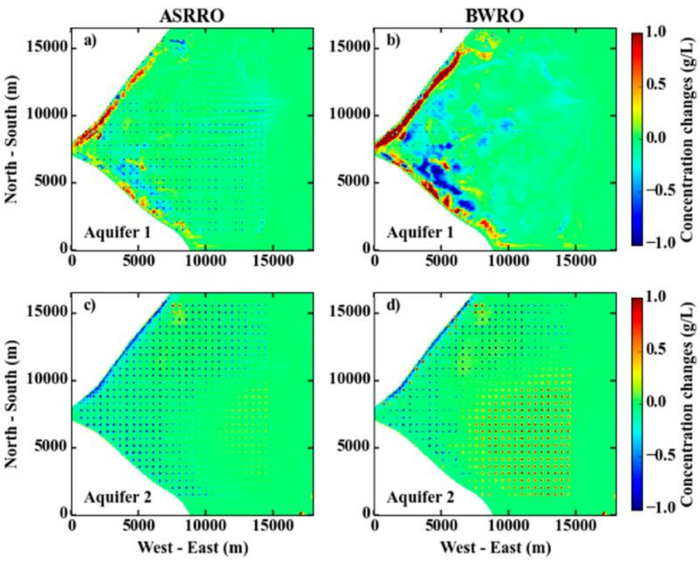

Figure 10. Relative chloride concentration changes (g/L) between ASRRO and Autonomous and between BWRO and Autonomous after 30 years in Aquifer 1 (**a**,**b**) and Aquifer 2 (**c**,**d**).

3.2.2. Concentration of the Membrane Concentrate

A distribution of the MC injection water concentration by the MCDWs is presented in Figure 11. The MC injected in Aquifer 2 had an average concentration of 3.6 g/L (in case of BWRO) and 2.1 g/L (in case of ASRRO).

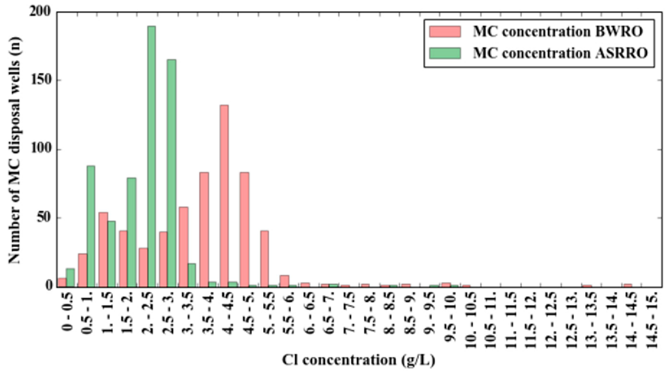

Figure 9. Distribution of the 30-year averaged MC chloride concentration by the 616 MC disposal wells.

4. Discussion

4.1. Impact of ASRRO on the Groundwater System

The timescale and resolution of the impact of ASRRO on the groundwater systems are significantly different on a local scale (vicinity of the system) and a regional scale (a regional groundwater system). Therefore, the impact on both scales is discussed separately.

4.1.1. Local Impacts on Groundwater Quality

Stratification of Freshwater-Saltwater by ASRRO

Besides impacting the average chloride concentration in the various geological layers, the ASRRO systems heavily impacted the distribution of concentrations in the vertical dimension of the aquifers ('stratification'). In Aquifer 1, injected freshwater moved to the top and remained where it formed local freshwater lenses. When such lenses merge, they can form a horizontal barrier for saline seepage, similar to vertical barriers along a coastline [19]. As a consequence, the diffuse saline seepage was replaced by freshwater seepage. The second consequence was that MC injected in Aquifer 2 was relatively fresh in the first stages of disposal. This freshwater moved upwards, replacing ambient brackish groundwater in Aquifer 2 and Aquitard 1, also in the downstream direction. Once this water reached Aquifer 1, it diluted the RO feed water of the downstream ASRRO system (MPPW c and d), which can be considered a positive impact.

Relative Impact of ASRRO

Based on the salt budgets of the Local Model, a positive impact of ASRRO on the groundwater system can be derived (Table 4). Ultimately, the total salt mass present during ASRRO in the vicinity of the wells stabilized, apart from seasonal variations caused by freshwater injection and abstraction, indicating that a local salinity equilibrium can be attained. ASRRO systems in the Local Model decreased the total chloride mass in the arbitrary 27-ha subsection by 3.9% with respect to the autonomous situation and caused long-term freshening within the Clay cap, Aquitard 1, and Aquifer 2. Upstream of and lateral to the ASRROs, short-circuiting increased the rate of salinization of

Aquitard 1 because of the net abstraction in Aquifer 1, which was required to feed the RO system with sufficient brackish water. This is indicated by a long-term increase in chloride mass in Aquifer 1. Only initially there was a positive impact because of the initial release of relatively fresh groundwater from Aquitard 1. The relative impact of ASRRO on Aquifer 1 can therefore be negative with respect to the autonomous situation (no abstractions). The MC did obtain a lower average chloride concentration (Cl: 3.8 g/L (MC disposal well 1) and 3.6 g/L (MC disposal well 2)) within Aquifer 2 (Figure 8) than in the autonomous situation (Cl: 6.0 g/L) and Aquifer 2 is therefore positively affected. This was due to the annual freshwater injection that locally diluted the groundwater in Aquifer 1 in the vicinity of the MPPWs (Figure). The balance between freshwater infiltration and freshwater abstraction is therefore expected to be vital for the total impact of ASRRO on the groundwater system.

Comparing the Impact of BWRO and ASRRO

In the Local Model, ASRRO decreased the total chloride mass in the 27-ha subsection by 16.8% with respect to BWRO and caused relative freshening of the groundwater system (Figure 4). This is regarded a positive impact. In the current practice of BWRO, short-circuiting from Aquifer 2 to Aquifer 1 is more severe than with ASRRO. As a consequence, BWRO intensified the rate of salinization in Aquifer 2 once relatively fresh groundwater from Aquitard 1 was consumed and replaced by relatively saline groundwater from Aquifer 2. As this water reached the MPPWs, the resulting MC became significantly more saline, forming extensive saline water plumes (Cl: 8–12 g/L). This is a negative impact of BWRO and was not observed during ASRRO. No freshwater barrier was formed because of BWRO, but seepage of brackish water towards the surface water system in summer was at least significantly reduced due to the abstraction—and therefore lower heads—in Aquifer 1.

The most noticeable positive impact of ASRRO with respect to BWRO is the absence of a relative salinity increase (and mineral saturation) in the deeper groundwater system. This is however a boundary condition for a long-term use of this groundwater system for freshwater supply, either by ASR (as increasing salinities lower the recovery efficiency [20]) or BWRO (as increasing salinities make BWRO more expensive and energy-consuming [21,22]).

4.1.2. Regional Impacts on Groundwater Quality

Widespread use of ASRRO in the Regional Model decreased the average regional chloride concentration by 0.3%, which can be considered a positive impact. The total impact on the groundwater system is significantly less due to the lower ratio of produced freshwater versus present groundwater (Section 2.2.1). BWRO resulted in higher average chloride concentrations of 0.2% with respect to ASRRO (Table 3). However, because of the net abstraction of groundwater in Aquifer 1 during ASRRO as well, additional saltwater intrusion still occurred along the coastline (Figure 10). This can be considered a negative impact of ASRRO. Yet still, this intrusion is significantly reduced when it is compared to the modelled intrusion caused by the current, wide-spread use of BWRO. In Aquifer 2, on the other hand, the MC disposal resulted in a transformation of saltwater intrusion to an outflow of brackish water towards the sea (Figure 10).

The abstractions for BWRO increased the infiltration of relatively fresh groundwater from the Clay cap and decreased Aquifer 1 concentrations regionally, as occurred in the first 10 years of the BWRO application in the Local Model. The positive effects of increased infiltration even outweighed those resulting from freshwater injection by ASRRO. Infiltration of freshwater from the surface may be exaggerated in this study by the modelling approach, in which constant heads and constant low salinities (freshwater) were assigned to the whole phreatic layer.

In the Regional Model, the MC disposal wells were further away from the brackish water abstraction wells than in the Local Model (250 m instead of 50 m), while each system had a lower capacity (8885 m^3/system/year instead of 27,000 m^3/system/year). This limited the rate of short-circuiting from Aquifer 2 to Aquifer 1 and reduced the local effects. However, once short-circuiting becomes prominent—as in the pilot [10]—salinization of abstraction wells will occur, which can undo this initially positive development (Figure 10). Short-circuiting should therefore be regarded a major obstacle for (long-term) use of BWRO in the study area.

In the Westland regional model, BWRO did not negatively affect chloride concentrations around each individual abstraction and MC disposal well. This depended on the local initial salinity of Aquifer 1 and 2 and the chosen RO recovery of 50%. Moreover, as with ASRRO, the disposal of MC into Aquifer 2 reduced salt water intrusion. This may imply that potentially no negative effects or even freshening may be observed when concentrations in Aquifer 2 are locally double or more than double the concentrations in Aquifer 1 (western and central parts of the Westland: Figure), unless short-circuiting occurs due to a limited separation of both aquifers. In such a case, it is relevant to analyse if this is really a sustainable situation, or that the net abstraction still results in saltwater intrusion in (parts of) the area, and thus, eventually, in salinization of the abstraction well. This risk is also present with ASRRO, albeit smaller due to the winter freshwater injections. Careful planning of wells will be beneficial to the limitation of rapid and severe local effects, although it will not influence the regional effects. Altogether, the regional impact of BWRO seems acceptable, but is strongly dependent on enhanced natural infiltration and sufficient spreading of BWRO systems.

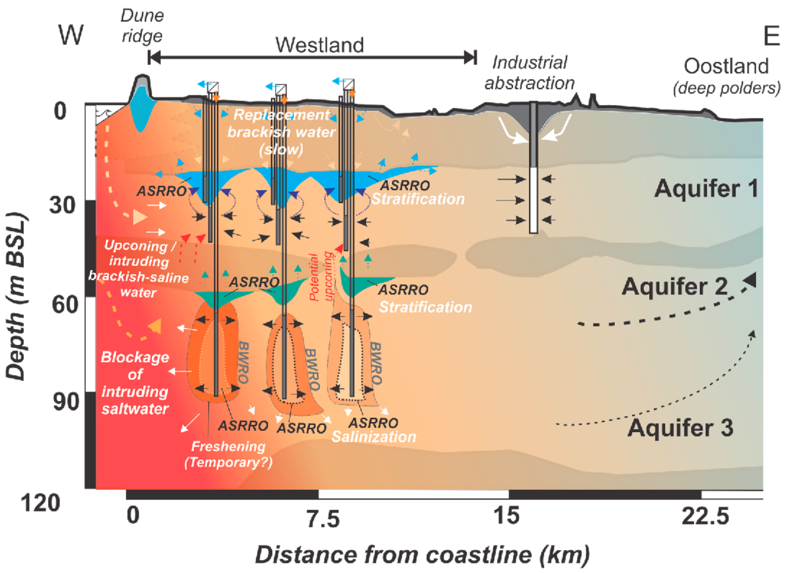

Figure 10. Overview of the integral impacts of ASRRO (and BWRO) on the regional salinity distribution in the Westland study area in a west-east cross-section. Local impact include salinization (BWRO) and stratification (ASRRO), while regionally saltwater intrusion (Aquifer 1) and brackish water outflow are the main phenomena, having the most negative impact in the BWRO scenario.

4.2. Implications of This Study for the Use of the Westland Groundwater System and Coastal Groundwater Systems Elsewhere

In the Westland area, the net abstraction during BWRO results in enhanced infiltration and saltwater intrusion, which was shown by this study. Excessive long-term decreasing groundwater tables are therefore not observed in the area and the predominant effects of water mining are the increasing salinities in the aquifers. Therefore, the current solution for high-quality irrigation water supply in the Westland area (BWRO) is under its hydrogeological conditions an unsustainable water supply solution and leads to slow mining of remnant (relatively) fresh water and saltwater intrusion [23], which is marked as an undesired effect upon exploitation of groundwater systems. Unlike falling groundwater levels, the slow salinization process is difficult to observe and will manifest itself primarily by a slow increase of the salt mass in the groundwater system and of the salinity of the abstracted brackish water. This eventually makes BWRO a less efficient or even infeasible technique.

A switch to ASRRO will prevent or limit the impact on the groundwater body of using the Westland's aquifers as an irrigation water source and may therefore be preferred from a policy point of view, taking into account European Water Framework Directive and especially the European Groundwater Directive [24], as they set specific goals for the condition of groundwater bodies. A boundary condition for success is the balance between (artificial) infiltration of freshwater surpluses and freshwater production during ASRRO. Positive side effects can be a reduction of subsidence and the intentional lowering of water levels in of aboveground rainwater reservoirs by infiltration for enhanced retention during intensive rainfall events.

Hurdles for large-scale implementation of ASRRO to mitigate potential impacts on the groundwater body may be the variability of the water demand in the area, however. Horticulturists with a low water demand can suffice with an aboveground rainwater reservoir, whereas horticulturists with a high water demand require more water than available by precipitation. On average, these imbalances can be eliminated in the Westland due to the equal volumes of water surplus and demand over time, but there is currently no incentive for horticulturists with a low water demand to infiltrate their surplus. A water bank [25] may provide a potential governance instrument to overcome this hurdle. A technical hurdle can be the mobilisation of particles upon freshening [26,27], which can lead to clogging of RO membranes during ASRRO [28]. The extent to which this process occurs and potential mitigation strategies are relevant fields of future research.

The Westland coastal groundwater system suffers from many typical water related issues observed in coastal zones worldwide, the most important ones being saltwater intrusion, subsidence, sea level rise, salinization of surface waters, and an increasing water quality and quantity demand, especially during prolonged droughts. The Westland case can therefore be considered a valuable example for improvement of the management of coastal groundwater systems with ASRRO. However, several local operational (e.g., infiltrated and recovered volumes) and hydrogeological (e.g., aquifers, aquitards, drainage levels, nearby abstractions) controlling factors will affect the overall impact and their cumulative impact on any groundwater system. This overall impact should therefore be evaluated before widespread ASRRO implementation in other areas.

5. Conclusions

In this study, the expected impacts of combined aquifer storage and recovery and reverse osmosis (ASRRO) on the water quality of the Westland groundwater system have been assessed through modelling the local effects of ASRRO and effects of widespread ASRRO implementation.

ASRRO reduces the salinities in its vicinity. An initially local, horizontal freshwater barrier forms at the top of the ASRRO target aquifer (Aquifer 1) and the aquifer for MC disposal (Aquifer 2), positively impacting seepage by lowering its salinity. In the deepest interval of Aquifer 2, a plume with slightly increased salinities can form and migrate downstream. However, this plume is significantly smaller compared with brackish water reverse osmosis (BWRO: the current practice, in which no rainwater is injected). In this case, an overall increase in the system's salinization rate was observed. During BWRO, increasingly more saline water will enter Aquifer 2, thereby forming an increasingly large and significantly more saline plume and creating the risk for upconing towards the brackish water abstraction wells in Aquifer 1.

Regionally, both ASRRO and BWRO resulted in an increase in saltwater intrusion in the aquifer targeted for freshwater storage and production (Aquifer 1), while in Aquifer 2 the saltwater intrusion was reduced by the outflow of brackish water upon MC disposal. The saltwater intrusion in Aquifer 1 during ASRRO was limited as a consequence of the freshwater injections. Furthermore, the significantly lower MC concentrations during ASRRO in combination with the brackish water outflow towards the sea improved the overall salinity in Aquifer 2. The same outflow was observed during BWRO, but the high concentrations in the MC deteriorated the groundwater quality in that case, such that a water quality improvement of Aquifer 2 was not attained.

The outcomes of this study highlight the complex interplays when targeting coastal groundwater systems with freshwater supply techniques like ASRRO and BWRO. Based on this case study, an overall positive to neutral impact of ASRRO on a coastal groundwater system is presumed, which is an improvement with respect to the use of BWRO in the same setting. ASRRO thus provides means to sustainably use coastal groundwater systems. However, several operational (e.g., infiltrated and recovered volumes) and hydrogeological (e.g., aquifers, aquitards, drainage levels, nearby abstractions) controlling factors will affect the overall impact and their cumulative impact on any groundwater system and should be considered before ASRRO implementation elsewhere.

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Author Contributions: Steven Eugenius Marijnus Ros and Koen Gerardus Zuurbier conceived and designed the modeling experiments; Steven Eugenius Marijnus Ros performed the modeling; Steven Eugenius Marijnus Ros and Koen Gerardus Zuurbier analyzed the data and wrote the paper.

Conflicts of Interest: The authors declare no conflict of interest. The founding sponsors had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, and in the decision to publish the results.

Appendix A. Model Input Regional Model

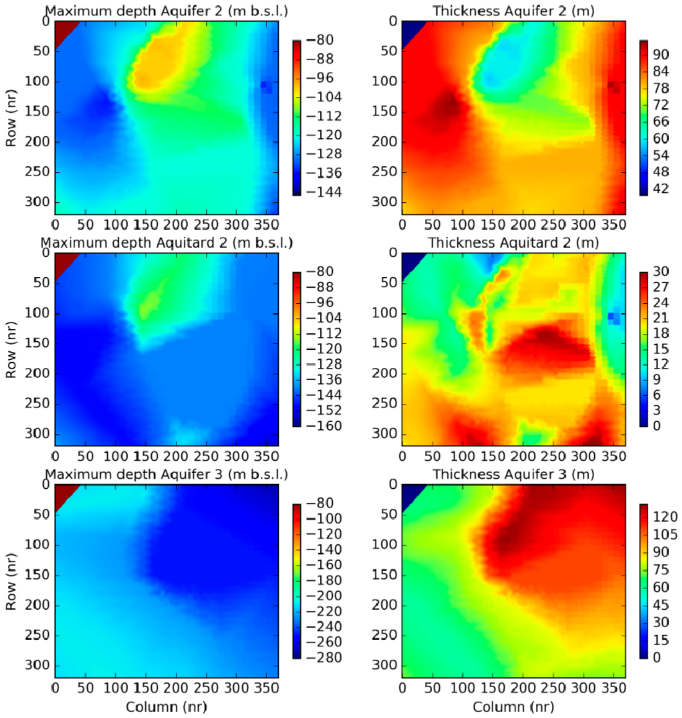

Figure A1. Maximum depth of occurrence (m below surface level) and total thickness (m) of Aquifer 2 (**top**), Aquitard 2 (**middle**), and Aquifer 3 (**bottom**).

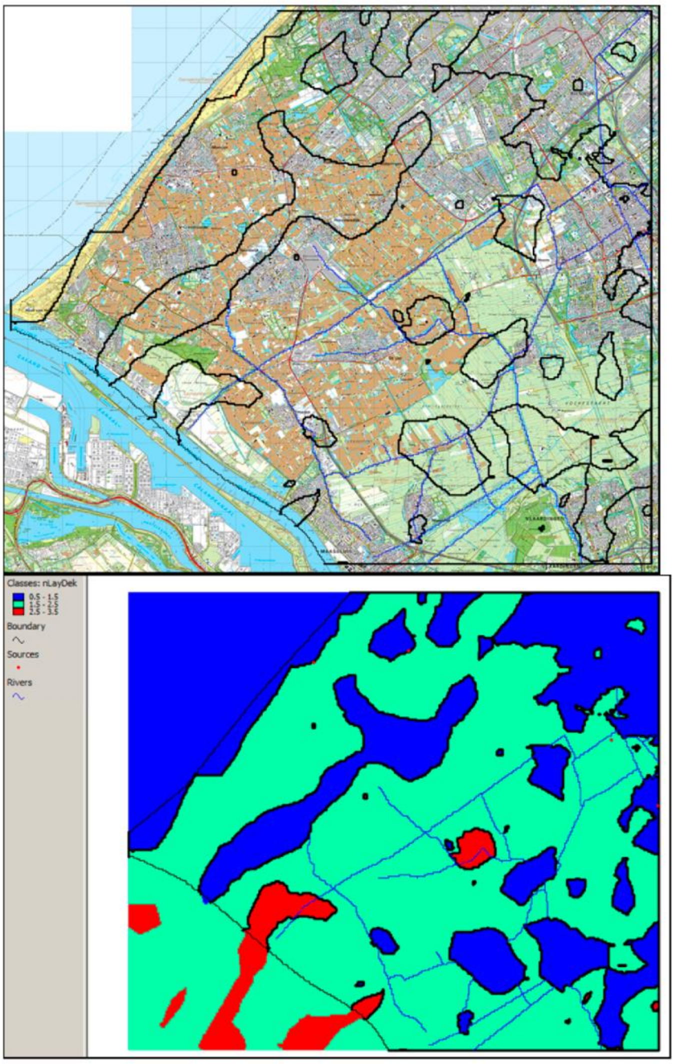

Figure A2. Topographic map (top) showing the outline of the regions wherein the Holocene clay cap is either 1, 2 or 3 model layers thick. The latter is presented in the bottom figure. The clay cap occurs in L2, (L3, L4), and is at most 15 m thick. The clay layer occurrence has been obtained from the PZH-Westland data [18].

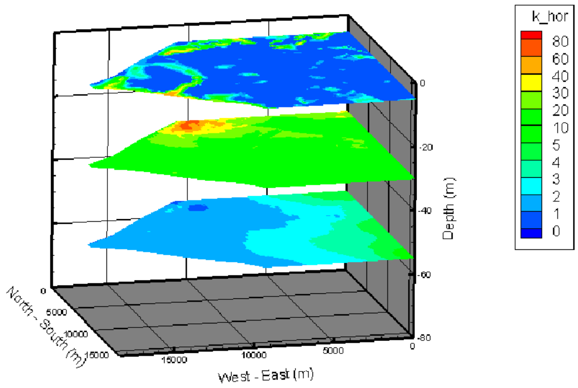

Figure A3. Horizontal conductivities (m/day) in the Regional Model for the Phreatic layer, and Aquifers 1 and 2. The North Sea is located northwest of the model domain.

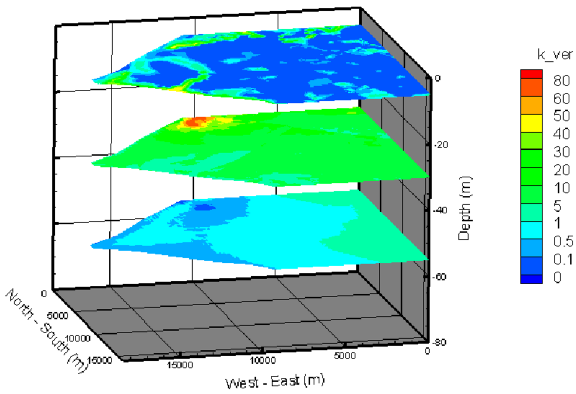

Figure A4. Vertical conductivities (m/day) in the Regional Model for the Phreatic layer, and Aquifers 1 and 2. The North Sea is located northwest of the model domain.

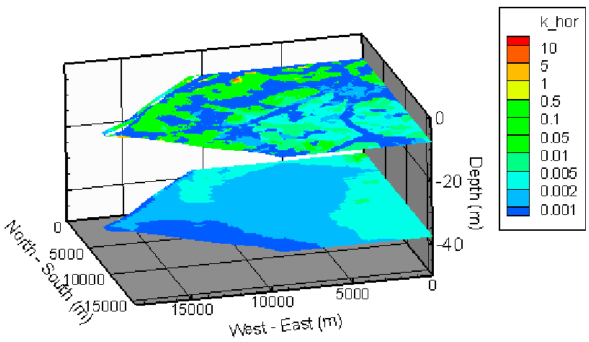

Figure A5. Horizontal conductivities (m/day) in the Regional Model for the Clay cap and Aquitard 1. The North Sea is located northwest of the model domain.

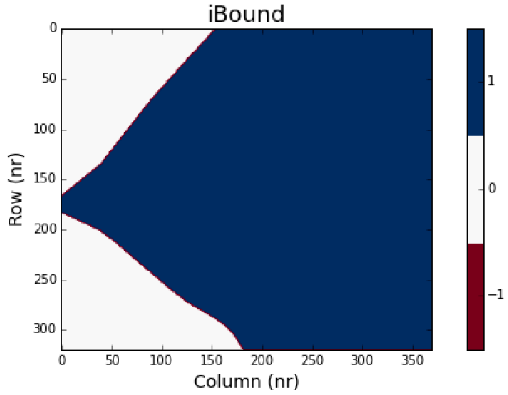

Figure A6. iBounds (−1, 0, 1) used in the Regional Model. The values are set to "−1" along the vertical boundary planes surrounding the active grid cells and in the top layer.

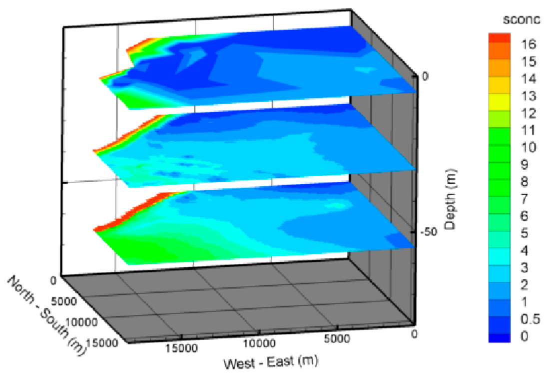

Figure A7. Starting concentrations (g/L) in the Regional Model for the Phreatic layer, and Aquifers 1 and 2. The North Sea is located northwest of the model domain.

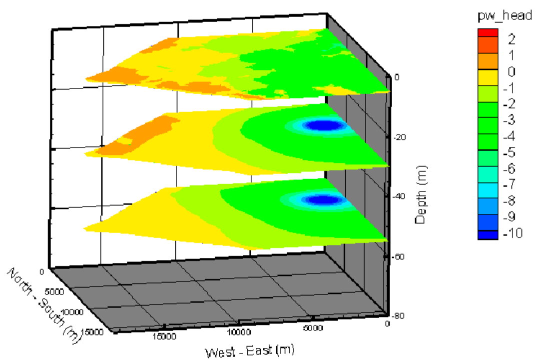

Figure A8. Point water heads (m) in the Regional Model for the Phreatic layer, and Aquifers 1 and 2. The North Sea is located northwest of the model domain.

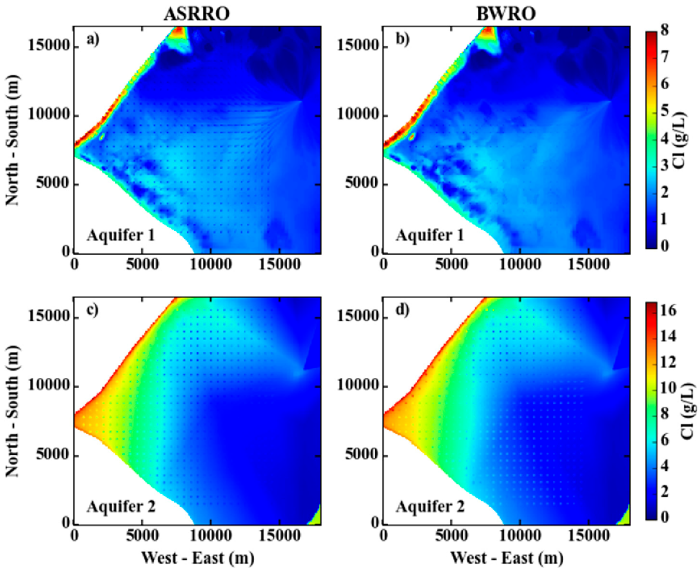

Figure A9. Chloride concentration (g/L) after 30 years of practice of ASRRO and BWRO in Aquifer 1 (**a**,**b**) and Aquifer 2 (**c**,**d**).

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ANNEX B: Scientific analysis short-circuiting during ASRRO Westland

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Consequences and mitigation of saltwater intrusion induced by short-circuiting during aquifer storage and recovery in a coastal subsurface

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Abstract. Coastal aquifers and the deeper subsurface are increasingly exploited. The accompanying perforation of the subsurface for those purposes has increased the risk of short-circuiting of originally separated aquifers. This study shows how this short-circuiting negatively impacts the freshwater recovery efficiency (RE) during aquifer storage and recovery (ASR) in coastal aquifers. ASR was applied in a shallow saltwater aquifer overlying a deeper, confined saltwater aquifer, which was targeted for seasonal aquifer thermal energy storage (ATES). Although both aquifers were considered properly separated (i.e., a continuous clay layer prevented rapid groundwater flow between both aquifers), intrusion of deeper saltwater into the shallower aquifer quickly terminated the freshwater recovery. The presumable pathway was a nearby ATES borehole. This finding was supported by field measurements, hydrochemical analyses, and variable-density solute transport modeling (SEAWAT version 4; Langevin et al., 2007). The potentially rapid short-circuiting during storage and recovery can reduce the RE of ASR to null. When limited mixing with ambient groundwater is allowed, a linear RE decrease by short-circuiting with increasing distance from the ASR well within the radius of the injected ASR bubble was observed. Interception of deep short-circuiting water can mitigate the observed RE decrease, although complete compensation of the RE decrease will generally be unattainable. Brackish water upconing from the underlying aquitard towards the shallow recovery wells of the ASR system with multiple partially penetrating wells (MPPW-ASR) was observed. This "leakage" may lead to a lower recovery efficiency than based on current ASR performance estimations.

1 Introduction

Confined and semi-confined aquifers are increasingly being used for storm water and (Ferguson, 1990), brine disposal (Stuyfzand and Raat, 2010; Tsang et al., 2008) and storage of freshwater (aquifer storage and recovery or ASR; Pyne, 2005), heat (aquifer thermal energy storage or ATES; Bonte et al., 2011a), and CO_2 (Steeneveldt et al., 2006). Additionally, they are perforated for exploitation of deep fossil and geothermal energy and traditionally used for abstraction of drinking and irrigation water. The increased use of the subsurface can lead to interference among aquifer storage systems (e.g., Bakr et al., 2013) or affect the groundwater quality (Bonte et al., 2011b, 2013; Zuurbier et al., 2013b). These consequences form relevant fields of current and future research.

The perforation of aquifers and aquitards accompanying the subsurface activities imposes an additional risk by the potential creation of hydraulic connections ("conduits") between originally separated aquifers or aquifers and surface waters. This risk is plausible, as estimations indicate that about two-thirds of the wells worldwide may be improperly sealed (Morris et al., 2003), although the attention for this potential risk is limited (Chesnaux, 2012). Additionally, many of the new concepts to use the subsurface (e.g., ATES, ASR, brine disposal) require injection via wells, which may cause fractures, even when the annulus is initially properly sealed, by exceedance of the maximum-permissible injection pressure (Hubber and Willis, 1972; Olsthoorn, 1982). The soil fractures are undesirable for most groundwater wells in the

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relatively shallow subsurface, since they create new connections between originally separated aquifers.

The resulting short-circuiting or leakage process has been studied at laboratory (Chesnaux and Chapuis, 2007) and field scale (Jiménez-Martínez et al., 2011; Richard et al., 2014), and for deep geological CO_2 storage (Gasda et al., 2008). Santi et al. (2006) evaluated tools to investigate cross-contamination of aquifers. Chesnaux et al. (2012) used numerical simulations of theoretical cases to demonstrate the consequences of pumping tests and hydrochemistry of hydraulic connections between granular and fractured-rock aquifers, which clearly demonstrated the significant hydrochemical cross-contamination when short-circuiting aquifers have a distinct chemical composition. The impact of short-circuiting on ASR has not been evaluated to date. However, reliably confined aquifers are vital to successfully store energy (Bonte et al., 2011a) and freshwater (Maliva et al., 2016; Maliva and Missimer, 2010; Missimer et al., 2002; Pyne, 2005; Zuurbier et al., 2013a) to bridge periods of surplus and demand, as inter-aquifer leakage may result in a loss of freshwater or undesirable admixing groundwater with a poorer quality, and therefore a reduced ASR performance. Furthermore, although the risks of short-circuiting by perturbation are acknowledged by scientists, it seems that the practical and regulatory communities are less aware (Chesnaux, 2012). This is underlined by the fact that certification for mechanical drilling (applied since the Industrial Revolution) in the Netherlands was not obligatory before 2011 (Stichting Infrastructuur Kwaliteitsborging Bodembeheer, 2013a), while for the subsurface design and operation of ATES systems (> 1500 systems since the 1990s; Bonte et al., 2011a; CBS, 2013), obligatory certification has only been enforced since early 2014 (Stichting Infrastructuur Kwaliteitsborging Bodembeheer, 2013b).

The lack of proper design and regulation of subsurface activities using wells can be partly caused by the lack of clear field examples of how well-intentioned use of the subsurface for sustainability purposes can fail thanks to earlier activities underground. This lack can be caused by the fact that short-circuiting may not be easy to observe (Santi et al., 2006), or because failing or disappointing projects often do not make it to public or scientific reports. Therefore, we present in this study how short-circuiting via a deeper borehole led to failure of freshwater recovery during ASR in a coastal aquifer. The objective of this paper is to demonstrate and characterize the potential consequences of perturbations for coastal ASR systems. Additionally, the use of deep interception of saltwater to improve shallow recovery of freshwater upon ASR was assessed. The Westland ASR site in the coastal area of the Netherlands served as a demonstration and reference case.

Table 1. Depth of the various well screens.

Well screen	Top (m b.s.l.)	Bottom (m b.s.l.)
AW1.1 + AW2.1	23.1	26.6
AW1.2 + AW2.2	27.6	30.6
AW1.3 + AW2.3	31.6	36.4
ATES K3-b	53	61
	80	85

2 Methods

2.1 Setup Westland ASR system and pilot

The Westland ASR system is installed to inject the rainwater surplus of 270 000 m^2 of greenhouse roof in a local shallow aquifer (23 to 37 meters below sea level, m b.s.l.; surface level = 0.5 meter above sea level, m a.s.l.) with negligible lateral displacement (Zuurbier et al., 2013a) for recovery in times of demand. For this purpose, two multiple partially penetrating wells (MPPW) were installed (Fig. 1), such that water can be injected preferably at the aquifer base, and recovered at the aquifer top in order to increase the recovery (Zuurbier et al., 2014). Due to the limited space available at the greenhouse site, the ASR well was installed close to an existing ATES well, injecting (in winters) and abstracting (in summers) cold water of about 5 °C. All ASR (AW1 and AW2, installed in 2012) and ATES (K3-a, installed in 2006 and replaced by K3-b at 3 m from AW1 and 7 m from AW2 in 2008) wells were installed using reverse-circulation rotary drilling, while the monitoring wells (MW1-5, Fig. 2) were installed using bailer drilling. Bentonite clay was applied to seal the ASR well (type: Micolite300) and ATES well K3 (Micolite000 and Micolite300). The depth of the well screens is shown in Table 1. The monitoring wells were installed at 5 m (MW1), 15 m (MW2), 30 m (MW3), 32 m (MW4), and 60 m (MW5).

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 $m^3\,h^{-1}$. The maximum Cl concentration in the recovered water accepted at the site is 50 $mg\,L^{-1}$. The ASR operation was relatively "dynamic" due to the incorporation of the ASR system in the water supply of a greenhouse; injection occurred in times of high levels in the aboveground rainwater reservoirs, whereas recovery occurred when low reservoir levels were observed. This led to the general ASR cycles as presented in Table 2.

2.2 Detailed hydrogeological characterization based on local drillings

The target aquifer for ASR (Aquifer 1) was found to be 14 m thick and consists of coarse fluvial sands (average grain size: 400 µm; see Fig. 3) with a hydraulic conductivity (K) derived

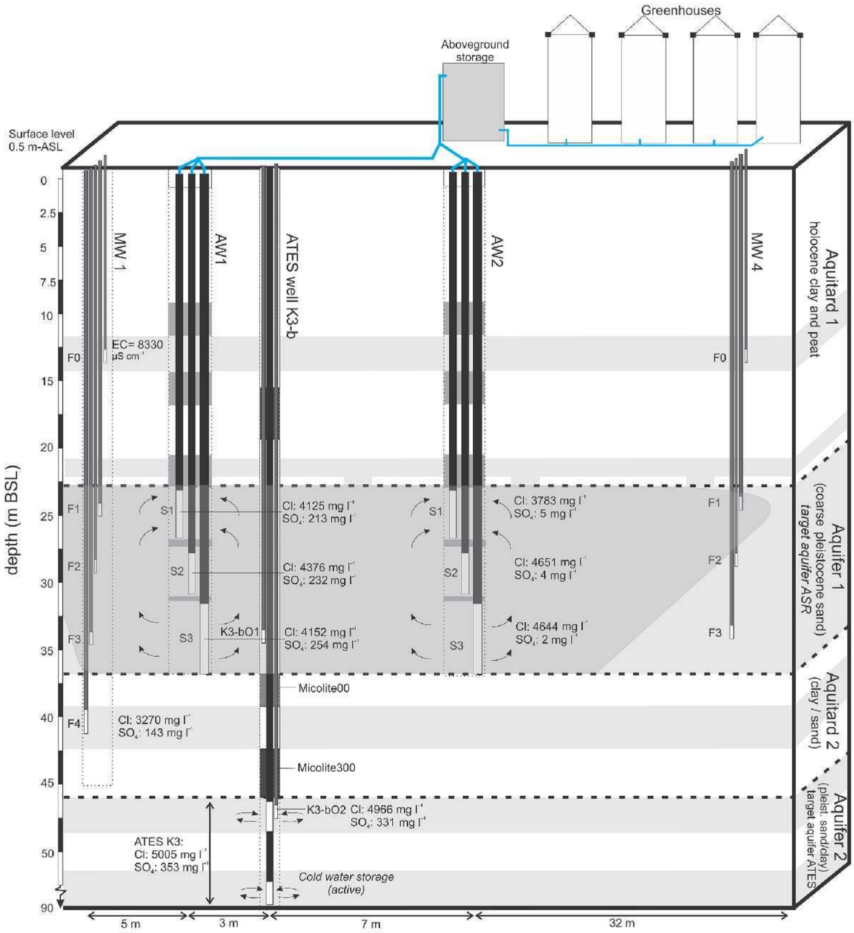

Figure 1. Cross section of the Westland ASR site to schematize the geology, ASR wells, ATES well, and the typical hydrochemical composition of the native groundwater. Horizontal distances not to scale.

Table 2. Summary of the ASR operation.

Stage	Date	Wells
Injection cycle 1.1	12 December (2012)–11 January (2013)	AW1 + AW2
Recovery cycle 1.1	11 January–28 January (2013)	AW1.1 + AW2.1
Injection cycle 1.2	4 February–8 February (2013)	AW1 + AW2
Recovery cycle 1.2	5 March–11 March (2013)	AW2.1 + AW2.2
Injection cycle 2	11 September (2013)–5 March (2014)	AW1 + AW2
Recovery cycle 2	5 March–24 June (2014)	AW2.1 + AW2.2

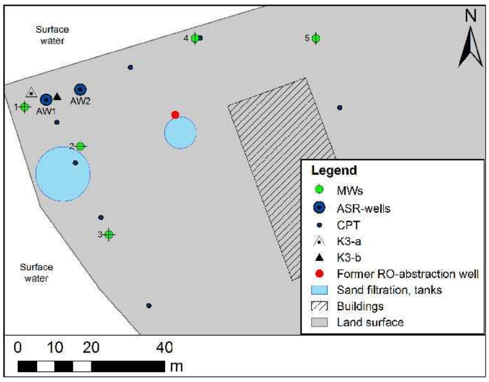

Figure 2. Locations of ASR (AW), ATES, and monitoring wells (MW).

from head responses at the monitoring wells upon pumping of 30–100 m d^{-1}. Aquifer 2 (target aquifer for ATES) has a thickness of more than 40 m, but is separated in two parts at the ATES well K3-b by a 20 m thick layer clayey sand and clay. A blind section was installed in this interval, and the borehole was backfilled with coarse gravel in this section. The K value of the fine sands in Aquifer 2 derived from a pumping test at approximately 500 m from the ASR well is 10 to 12 m d^{-1} and is in line with the estimated K value from grain size distribution (Mos Grondmechanica, 2006). The effective screen length of K3-b in this aquifer is only 8 and 5 m (Table 1).

The groundwater is typically saline, with observed Cl concentrations ranging from 3793 to 4651 mg L^{-1} in Aquifer 1 and approximately 5000 mg L^{-1} in Aquifer 2 (see also Fig. 1). This means that with the accepted Cl concentrations during recovery, only around 1 % of admixed ambient groundwater is allowed. A sand layer in Aquitard 2 contains remnant fresher water (Cl = 3270 mg L^{-1}). SO_4 is a useful tracer to identify the saltwater from Aquifers 1 and 2: it is virtually absent in Aquifer 1 (presumably younger groundwater, infiltrated when the Holocene cover was already thick), whereas it is high in Aquifer 2 (older water, infiltrated through a thinner clay cover which limited SO_4 reduction; see Stuyfzand, 1993, for more details): 300 to 400 mg L^{-1} SO_4.

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 MW2 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 (Table 3). In all, 3 times the volume of the well casing was removed and stable field parameters were attained 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 electrical conductivity (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 1 day after sampling on the Titralab 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 % HNO_3 (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, USA). The second 10 mL vial was used for analysis of F, Cl, NO_2, Br, NO_3, PO_4, and SO_4 using the Dionex DX-120 IC (Thermo Fischer Scientific Inc., USA), and NH_4 using the LabMedics Aquakem 250 (Stockport, UK). All samples were cooled to 4 °C and stored dark immediately after sampling.

Combined electrical conductivity, temperature, and pressure transducers (CTD) divers (Schlumberger Water Services, Delft, the Netherlands) were used for continuous monitoring 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 the operation per well screen.

2.4 Setup Westland ASR groundwater transport model

Groundwater transport modeling was executed to validate the added value of the MPPW setup under the local conditions. In the later stage of the research, the groundwater transport model was used to test potential pathways for deeper groundwater to enter the target aquifer and explore the characteristics of a potential conduit via scenario modeling. Correction for groundwater densities in the flow modeling was vital, due to significant contrast between the aquifer's groundwater and the injected rainwater. In order to incorporate variable density flow and the transport multiple species, SEAWAT version 4 (Langevin et al., 2007) was used with PMWIN 8 (Chiang, 2012) to simulate the ASR operation. A half-domain was modeled to reduce computer runtimes (Fig. 4). Cells of 1 × 1 m were designated to an area of 20 × 20 m around the ASR wells. The cell size increased to 2.5 m × 2.5 m (30 m × 40 m around the well) and was then gradually increased to a maximal cell size of 200 m × 200 m at 500 m from the ASR wells. The pumping rate of each well screen was distributed over the models cells with the well package based on the transmissivity (thickness × hydraulic conductivity) of each cell. The third-order total-variation-diminishing (TVD) scheme (Leonard, 1988) was used to

Table 3. Sampling rounds at the Westland ASR site (2012–2014). "IN" is injection water.

Well(s)	Date
K3-b, K3-bO2	22 August 2012
AW1, AW2	6 November 2012
MW1–MW5	5 December 2012
MW1	14 December 2012
MW1	17 December 2012
MW1, MW2, IN	18 December 2012
MW1, MW2	20 December 2012
MW1, MW2, IN	21 December 2012
MW1, MW2	24 December 2012
MW1, MW2	27 December 2012
MW1, MW2	31 December 2012
MW1, MW2, MW4	4 January 2013
MW1, MW2, IN	11 January 2013
AW1, AW2	14 January 2013
MW1, MW2, MW4	17 January 2013
AW1, AW2	25 January 2013
MW1, MW2, MW4, IN	12 February 2013
AW1, AW2	8, 11, 19 March 2013; 8 April 2013; 11, 14, 17, 21, 28 March 2014; 2, 15, 17, 28 April 2014; 5, 22 May, 2 June 2014
MW1, MW2, MW3, MW4	11 March, 8 April 2013; 17 September 2013; 2 October 2013; 6 November 2013; 11 December 2013 14 January 2014; 19 February 2014; 2 April 2014; 5 May 2014
K3-bO1	21, 28 March 2014; 8, 28 April 2014; 5 May 2014

model advection and maintain the sharp edges of the freshwater bubble by limiting numerical dispersion.

Equal constant heads were imposed at two side boundaries of the aquifers, the top of the model (controlled by drainage) and at the base of the model. No-flow boundaries were given to the other two side boundaries of the model. Initial Cl concentrations were based on the results of the reference groundwater sampling at MW1. SO_4 concentrations in Aquifer 1 were based on MW2, since these concentrations were considered most representative for the field site; this well was close to the ASR wells, but not potentially disturbed by the ATES or ASR wells. For Aquifer 2, the concentrations found at ATES well K3-b (bulk) and the observation well K3-bO.1 were used (see Fig. 1). The density of the groundwater was based on the Cl concentration using

$$\rho_w = 1000 + 0.00134 \times \mathrm{Cl}(\mathrm{mg\,L^{-1}}). \quad (1)$$

Density and viscosity were not corrected for temperature, as all temperatures (background groundwater, injected ASR water, and injected ATES water) were in the range of 8 to 12 °C and should not significantly impact the flow pattern (Ma and Zheng, 2010). 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) and the observed heads in the aquifer. The regional hydraulic gradient was derived from regional groundwater heads (TNO, 1995). Further details are given in Table 4.

The recorded pumping rates of the ASR wells and the ATES K3-b well during two ASR cycles were incorporated in the SEAWAT model. The ASR operation was modeled with a properly sealed and an unsealed ATES borehole. In the latter case, a hydraulic conductivity (K) of $1000\,\mathrm{m\,d^{-1}}$ was given to the cells ($1.0\,\mathrm{m} \times 1.0\,\mathrm{m}$) in Aquifer 1, Aquitard 2, and Aquifer 2 at the location of the ATES pumping well to force a significant borehole leakage. This K was considered realistic since apart from filter sand around the well screen; the borehole was backfilled with gravel with a grain size of 2 to 5 mm. In later scenarios, the ATES well was moved towards the fringe of the ASR well stepwise (10 m further away from AW1 in each scenario), after which Cycle 2 was simulated again. This was to examine the impact of borehole leakages at various distances from the ASR wells.

2.5 The maximal recovery efficiency with and without leakage at the Westland ASR site.

The collected data on the aquifer characteristics in the SEAWAT groundwater model were used to analyze the future performance of the MPPW-ASR system for the current (with leakage) and a "normal field site" (without leakage from deeper aquifers via a perturbation, or after sealing of the perturbation). The SEAWAT model was used to simulate three consecutive ASR cycles with the representative operational characteristics from Table 5 for the Westland site (Zuurbier et al., 2012). Once the recovered Cl concentration exceeded $50\,\mathrm{mg\,L^{-1}}$, the model was stopped, and the length of the stress period with recovery was adjusted, such that no water with Cl > $50\,\mathrm{mg\,L^{-1}}$ was recovered. Subsequently the model was run again after adding another cycle.

3 Results

3.1 Cycle 1 (2012–2013): first identification of borehole leakage

The first ASR cycle started in December 2012. The first recovery started halfway January 2013. Despite the abstraction with only the shallow wells of the MPPW, a rapid and severe salinization was found within the first days of recovery, after injecting freshwater for about 1 month (Fig. 5). It was expected that due to mixing and buoyancy effects during ASR, MW2 would salinize first, followed by MW1, and finally the ASR wells (AW1 and AW2) towards the end of the recovery phase, with each time the deepest well screens

Table 4. Hydrogeological properties of the geological layers in the Westland SEAWAT model.

Geological layer	Model layers	Base (m b.s.l.)	K_h (m d^{-1})	K_v (m d^{-1})	S_s (m^{-1})	n (–)	Initial C (mg L^{-1} Cl)	Initial C (mg L^{-1} SO_4)
Aquitard 1	6	22.3	0.2–1	0.002–0.01	10^{-4}	0.2	2000–3000	4
Aquifer 1	12	33.7	35	35	10^{-7}	0.3	4000–4800	4
	3	36.4	100	100				
Aquitard 2 (clay–sand)	8	47.5	0.05–10	0.0005–10	10^{-4}	0.2–0.3	3200	160
Aquifer 2	6	96	12	12	10^{-6}	0.3	4100–7900	331–375

Table 5. Setup of the modeled, representative ASR cycle for the Westland subsurface without short-circuiting of deeper saltwater.

Stage	Duration	Pumping rate
Injection	120 days	60 000/120 = 500 m^3 d^{-1}
Storage	30 days	0 m^3 d^{-1}
Recovery	120 days	−60 000/120 = -500 m^3 d^{-1}
Idle	65 days*	0 m^3 d^{-1}

* Longer when early salinization occurred during recovery.

salinizing first. This salinization would then be caused by the replacement of freshwater by ambient groundwater (very low-SO_4 concentrations) from the same aquifer (Ward et al., 2009). Remarkably, the salinization at AW1 preceded salinization of the monitoring wells situated further from the ASR wells (MW1, MW2). Furthermore, SO_4 concentrations (up to > 50 mg L^{-1}) were found in the recovered water, which could not be explained by the SO_4 concentration attained by pyrite oxidation by oxygen and nitrate present in the injection water (Zuurbier et al., 2016), which would result in SO_4 concentrations of less than 15 mg L^{-1}.

The SEAWAT model underlined that tilting of the freshwater–saltwater interfaces at the fringe of the ASR bubble did not cause the early salinization observed, as this would have led to a much later salinization (Fig. 6) without enrichment of SO_4 (other than be pyrite oxidation), even if the recovery period was extended (results not shown). When the leaky borehole was incorporated in the model (by assigning $K = 1000$ in a 1 m × 1 m column at the location of the current ATES well), it was able to introduce the early recovery of deep (SO_4-rich) water (Fig. 7). Other scenarios that were tested, but unable to improve the simulation of the observed SO_4 trends, were leakage via the former ATES K3-a well further from the ASR wells (arrival of SO_4 too late), a high-K_v borehole (2000 m d^{-1}; arrival too early, flux too high), a low-K borehole (500 m d^{-1}; arrival too late, flux too low), a vertical anisotropy in the aquifers ($K_h/K_z = 2$; arrival too early, flux too high), and omission of the deep cold water abstraction from Aquifer 2 via the ATES well in Aquifer 2 (SO_4; flux too high).

The hydrochemical observations and model outcomes of Cycle 1 indicated that the source of the early salinization was the intrusion of saltwater from Aquifer 2. Considering the lithology, thickness, and continuity of Aquitard 2 (confirmed by grain size analyses and cone penetrating tests on the site), leakage via natural pathways through this separating layer was unlikely. According to the rate and sequence of salinization, the leakage could well be situated at the ATES K3-b well close to AW1.

3.2 Cycle 2 (2013–2014): improving the ASR operation

Cycle 2 started with the injection of 66 178 m^3 of rainwater using both ASR wells between September 2013 and March 2014, which was followed by recovery solely at the downstream AW2 (start: 5 March 2014). A rapid salinization by SO_4-rich saltwater was again observed (Fig. 8) and the recovery was terminated after 26 days (21 March 2014) after recovering no more than 2500 m^3. During this cycle, a monitoring well present in the gravel pack of the ATES K3-b well (coded K3-bO1; a 1 m well screen at 33 m b.s.l., Fig. 1) was also sampled and equipped with a CTD diver and continuously pumped with a rate of 1 m^3 h^{-1}, unraveling high ECs and presence of SO_4-rich saltwater from the deeper aquifer in the center of the injected freshwater body (Fig. 8). This presence of intruding deep saltwater was also found at MW1S3 (5 m from the ASR wells) as a consequence of displacement while re-injecting part of the abstracted freshwater from the shallow AW2S1 wells screen at the deeper AW2S3 well screen and density-driven flow (spreading over the base of the aquifer). The observed Cl concentration (268 mg L^{-1}) on 2 April 2014 at MW1S4 (situated in Aquitard 2 at 5 m from AW1) was significantly lower than at MW1S3 (2528 mg L^{-1}) and K3-bO1 (3341 mg L^{-1}), indicating that salinization of the shallow target aquifer (Aquifer 1) preceded salinization of Aquitard 2.

In order to re-enable recovery of freshwater, the deepest wells of the MPPWs (AW1S3 and AW2S3) were transformed to interception wells or "Freshkeepers" (Stuyfzand and Raat, 2010; Van Ginkel et al., 2014), abstracting the intruding saltwater and injecting this in a deep injection well in Aquifer 2 at of distance 200 m from the ASR site. This way, an acceptable water quality (Cl < 50 mg L^{-1}) could be recovered

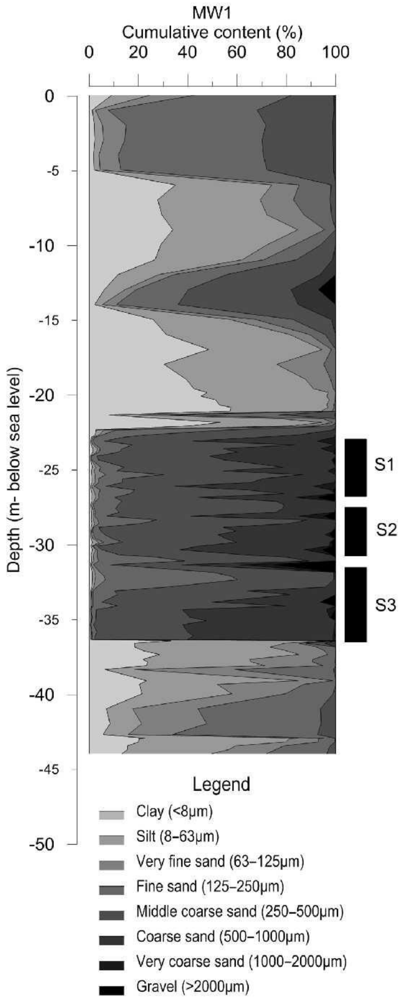

Figure 3. Cumulative grain size contents observed at MW1 (at 5 m from ASR well 1) in this study. S1–S3 mark the depth intervals of the ASR well screens.

at AW2S1 and AW1S2 again (from 15 April onwards). As a consequence, the deeper segments of the target aquifer (S3 levels, Fig. 8b, c, d) first freshened, followed by again salinization as recovery proceeded. Saline water was continuously observed at K3-bO1, indicating that leakage via the K3-b borehole continued. After recovery of in total 12 324 m^3 of practically unmixed rainwater (18.6 % of the injected water),

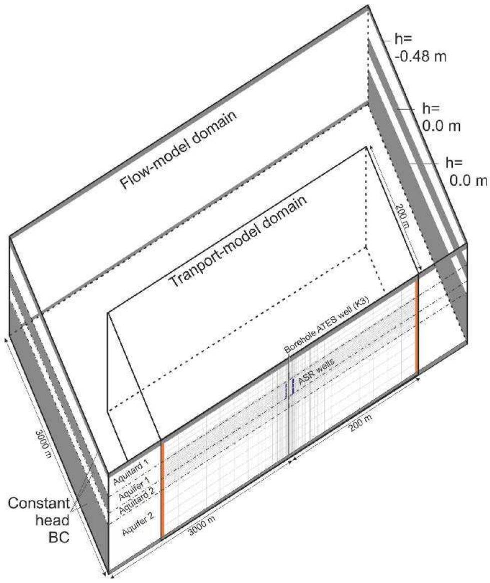

Figure 4. Setup of the Westland ASR groundwater transport model (half-domain).

the recovery had to be ceased due to the increased salinity. During this last salinization, the water at the deep (S3-)levels of the target aquifer at AW1, MW1, and MW2 showed low-SO_4 concentrations, indicating salinization by saltwater from Aquifer 1 instead of deep saltwater from Aquifer 2. High-SO_{4-} concentrations (> 100 mg L^{-1}) were only found close to the K3-b ATES well (the presumable conduit) in this phase (AW1 and K3-bO1).

The SEAWAT model with leakage via the borehole of K3-b was able to reasonably simulate the water quality trends regarding SO_4 and Cl in Cycle 2 (Figs. 9 and 10). Remaining deviations in observed concentrations were contributed to uncertainties in the model input, mainly aquifer heterogeneity, potential stratification of the groundwater quality in Aquifer 2, and disturbing abstractions and injections in the surroundings, mainly by nearby ATES and brackish water reverse osmosis systems, the latter abstracting from Aquifer 1 and injecting in Aquifer 2.

Modeling of Cycle 2 demonstrated that salinization during recovery was independent of the injected freshwater volume. Salinization occurred after recovery with the same rate as in Cycle 1, despite a 4 times larger injection volume. Analysis of the modeled concentration distribution and pressure heads showed that injected freshwater could not reach deep into the deeper saline aquifers since the freshwater head in the leaky ATES borehole during injection was more or less equal to

Table 6. Calculated leakage flux Q_{VGP} via the (unsealed) borehole based on Maas (2011) for different net recovery rates ($Q_{\mathrm{recovery,\,net}}$).

		Storage (no recovery)	Low recovery rate	High recovery rate
$Q_{\mathrm{recovery,\,net}}$	$(\mathrm{m^3\,d^{-1}})$	0	77	371
Δh_{GP}	(m)	0.15	0.30	0.66
Q_{VGP}	$(\mathrm{m^3\,d^{-1}})$	49	99	215
W	$(\mathrm{m^2\,d^{-1}})$	0.0031	0.0031	0.0031
α		4.7	4.7	4.7
r_0	(m)	0.1	0.1	0.1
r_1	(m)	0.4	0.4	0.4
K_{HIN}	$(\mathrm{m\,d^{-1}})$	100	100	100
K_{VIN}	$(\mathrm{m\,d^{-1}})$	100	100	100
K_{VGP}	$(\mathrm{m\,d^{-1}})$	1000	1000	1000

the freshwater head in the deeper saltwater aquifer. In other words, little freshwater was pushed through the conduit into the deeper aquifer. Further on, the freshwater that did reach the deeper aquifer got rapidly displaced laterally as a result of buoyancy effects (Fig. 11).

A significant head difference (Δh (fresh) = 0.3 to 0.65 m) was observed in the model during recovery. In combination with the high permeability of the ATES borehole, this resulted in a significant intrusion of deeper (SO_4-rich) saltwater. Even during storage phases, a freshwater head difference (Δh (fresh) = 0.15 m) was observed as a consequence of replacement of saltwater by freshwater in the target aquifer, causing intrusion of deep saltwater, yet with a lower rate than during recovery.

3.3 Analysis of the leakage flux via the borehole

An analytical solution was presented by Maas (2011) to calculate the vertical leakage via a gravel or sand pack. In this solution, it is presumed that an aquitard was pierced during drilling and the annulus was filled up with sand or gravel without installing a clay seal. The leakage is then calculated as function of the different hydraulic conductivities, pressure difference, and the radius of the borehole and well screen:

$$Q_{\mathrm{VGP}} = \frac{\Delta h_{\mathrm{GP}}}{W}, \tag{2}$$

where Q_{VGP} = vertical leakage via gravel pack $(\mathrm{m^3\,d^{-1}})$, Δh_{GP} = hydraulic head difference between two sections of the gravel pack, one being the inflow and the other the outflow section (meters), and W = leakage resistance $(\mathrm{d\,m^{-2}})$ and is calculated as

$$W \approx \frac{(0.005(\ln(\alpha))^2 - 0.058\ln(\alpha) + 0.19)}{(r_1\sqrt{K_{\mathrm{HIN}}K_{\mathrm{VIN}}})} \tag{3}$$

and a as

$$\alpha = \frac{K_{\mathrm{VGP}}(r_1^2 - r_0^2)}{2K_{\mathrm{VIN}}r_1^2}/, \tag{4}$$

where r_0 = radius of well screen (m), r_1 = radius of borehole (m), K_{VGP} = vertical hydraulic conductivity of gravel pack $(\mathrm{m\,d^{-1}})$, K_{VIN} = vertical hydraulic conductivity of inflow aquifer layer $(\mathrm{m\,d^{-1}})$, and K_{HIN} = horizontal hydraulic conductivity of the inflow aquifer layer $[\mathrm{m\,d^{-1}}]$.

Calculating the leakage flux using the Δh_{GP} from the SEAWAT model underlines that the pressure differences induced by density differences and enhanced during abstraction for freshwater recovery in combination with an unsealed borehole leads to a saltwater intrusion (Q_{VGP}) of around 50 to 200 $\mathrm{m^3\,d^{-1}}$ (Table 6), which is in line with the observed leakage flux in the SEAWAT model.

3.4 The maximal recovery efficiency with and without leakage at the Westland ASR site.

The SEAWAT model was used to evaluate the ASR performance at the Westland field site with three different ASR strategies (Table 7), with and without the saltwater leakage. During the 120 days of recovery it was aimed to recover as much of the freshwater (marked by $\mathrm{Cl} < 50\,\mathrm{mg\,L^{-1}}$) as possible. Equal abstraction rates were maintained for both ASR wells (AW1 and AW2) in the scenarios without leakage, whereas only AW2 was used for recovery in the scenarios with leakage.

Recovery with conventional, fully penetrating ASR wells will be limited to around 30 % of the injected freshwater in a case without the saltwater leakage. For the case with leakage, freshwater recovery will be impeded by the short-circuiting during the storage phase; the wells will produce brackish water already at the start of the recovery phase. 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 can be recovered in a case without leakage. The improvement of recovery efficiency (RE) by introduction of the MPPW is limited in comparison with the conventional ASR well since some saltwater from Aquitard 2 was found to move up to the shallower recovery wells of the MPPW system ("upconing") rapidly after

Table 7. Modeled recovery efficiencies at the Westland ASR site without short-circuiting using different pumping strategies. The relative pumping rate per MPPW well screen is given for each particular screen.

Strategy	Distribution pumping rate	RE (short-circuiting/no short-circuiting)	Intercepted brackish-saline water (via deep (S3-)wells)
Conventional ASR well	In: 100 % via one fully penetrating well Out: 100 % via one fully penetrating well	Year 1: 0/15 % Year 2: 0/25 % Year 3: 0/30 % Year 4: 0/32 %	
Deep injection, shallow recovery (MPPW-ASR)	In: 10/20/70 % (Year 1) In : 0/20/80 % (Year 2–3) Abstract: 60/40/0 % (Year 1–3)	Year 1: 1/19 % Year 2: 1/ 29 % Year 3: 1/32 % Year 4: 1/33 %	
MPPW-ASR + "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 $m^3\,d^{-1}$	Year 1: 29/40 % Year 2: 32/46 % Year 3: 33/47 % Year 4: 33/48 %	Year 1: 32 700/18 500 m^3 Year 2: 33 000/20 500 m^3 Year 3: 31 900/21 500 m^3 Year 4: 31 500/19 300 m^3

the start of recovery. The slight increase in Cl concentrations caused by this process is sufficient to terminate the recovery due to exceedance of the salinity limit. Before the fringe of the freshwater bubble reached the recovery wells, recovery was already terminated. In the case of saltwater leakage, salinization occurred within 2 days, limiting the RE to only 1 %.

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 40 % in the first year for direct use. Ultimately, this will yield a RE of almost 50 % of virtually unmixed ($Cl < 50\,mg\,L^{-1}$) injected freshwater in cycle 4 in a case without leakage. This will require interception of 18 500 m^3 (cycle 4) to 21 500 m^3 of brackish-saline groundwater, such that almost 30 000 m^3 of freshwater can be recovered.

When this ASR operational scheme with the Freshkeeper was applied to the field pilot, where short-circuiting saltwater hampered freshwater recovery, approximately one-third of the injected freshwater could be recovered. The ASR well close to the leaking borehole (AW1) was unable to abstract freshwater in this case. Only AW2 could be used for freshwater recovery, in the end only via the shallowest well (AW2S1). The freshwater loss by short-circuiting cannot be eliminated completely since a large volume of unmixed freshwater is abstracted together with intruding saltwater during the required interception. The RE will therefore remain lower than in an undisturbed geological setting (RE: 48 %). At the same time, the required interception of brackish-saline water will be higher (Table 7), with a total volume of more than 30 000 m^3, while around 20 000 m^3 of freshwater is recovered.

4 Discussion

4.1 Saltwater intrusion during the Westland ASR pilot

In this study, the first focus was on the causes for the significantly lower observed freshwater RE of the system. This RE was initially less than a few percent, whereas recovery of around one-third of the injected water was expected. The hydrochemical analyses clearly indicated that the observed salinization was caused by unexpected intrusion of deeper saltwater, as marked by substantially higher SO_4 concentrations, which could not be caused by arrival of saltwater from the target aquifer or the upper aquitard, or by the SO_4 release upon oxidation of pyrite in the target aquifer. The high-SO_4 concentrations also exclude early salinization by larger buoyancy effects than initially expected, for instance by a higher K or higher ambient salinities in the target aquifer. The high-SO_4 concentrations also excluded rapid lateral drift of injected water, as this would also have led to salinization by saltwater with low-SO_4 concentrations. Additionally, lateral drift would also result in limited REs after addition of the Freshkeeper, which was not the case.

Knowing the source of the salinization, several transport routes can be presumed. First of all, intrusion of deep saltwater may occur when Aquitard 2 has a significantly lower K than derived from grain size analyses, despite the distinct groundwater qualities observed. A more diffuse salinization via Aquitard 2 can then be expected. However, this salinization would be more gradual and better distributed around the wells. It would also mean that Aquitard 2 would quickly freshen during injection and salinize first during recovery. However, the later salinization of Aquitard 2 observed at

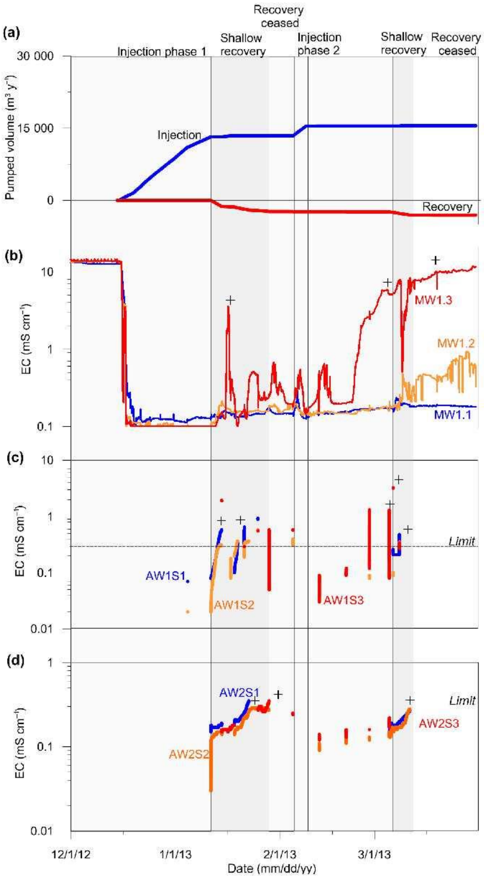

Figure 5. Pumping of the ASR system during cycle 1 (2012/2013), EC observations at MW1 (5 m from AW1), and the EC in the recovered water at AW1 and AW2. MW = monitoring well, AW = ASR well.

MW1S4 with respect Aquifer 1 (observed at MW1S3 and K3-bO1) indicated that Aquitard 2 is bypassed by deeper saltwater during recovery. The presence of (a) conduits therefore provide (a) probable pathways for bypassing saltwater, meaning short-circuiting was occurring between Aquifers 1 and 2. The SEAWAT model underlines that this can indeed explain the early and rapid intrusion by deep saltwater. Since the highest Cl and SO_4 concentrations were found in the borehole of K3-b well (K3-bO1), this borehole provides the most presumable location of (a) conduits. Natural conduits are considered unlikely due to continuity and thickness of Aquitard 2 observed in the surrounding of the ASR wells and the geological genesis (unconsolidated, horizontal lagoonal deposits). The conduits at or around the K3-b borehole may originate from the time of installation (improper sealing) or operation, as recorded operation data of the ATES system reports that incidentally exceeded the maximum injection pressure in the well of 1 bar during maintenance in 2009.

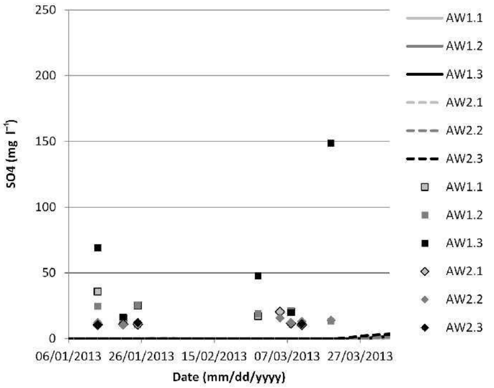

Figure 6. Modeled (solid lines) and observed (data points) SO_4 concentrations without borehole leakage. High concentrations indicate admixing of deeper saltwater. Observed SO_4 concentrations by far exceed the modeled concentrations.

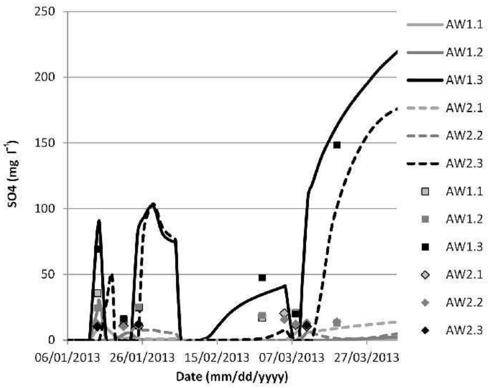

Figure 7. Modeled (solid lines) and observed (data points) SO_4 concentrations. Borehole leakage at the location of the current ATES K3 well via a 1 m × 1 m borehole with $K = 1000\,\mathrm{m\,d^{-1}}$. High concentrations indicate admixing of deeper saltwater. Observed SO_4 concentrations become in line with the modeled concentrations.

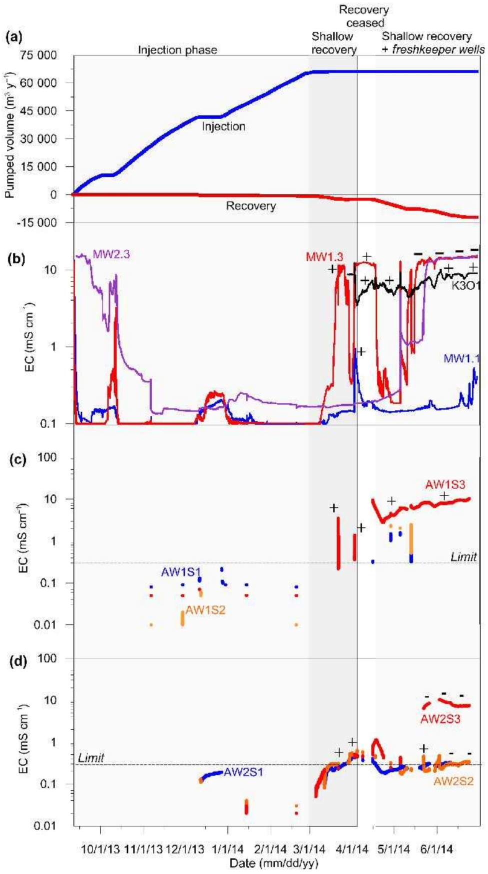

Figure 8. Pumping of the ASR system during cycle 2 (2013/2014), EC observations at MW1 (5 m from AW1), and the EC in the recovered water at AW1 and AW2. AW2.1 and AW2.3 were used for freshwater recovery (12 324 m^3). Presence of increased SO_4 concentrations (deep saltwater) are marked by "+", while its absence is marked by "−" (indicating shallow saltwater).

4.2 The consequences of short-circuiting on ASR in coastal aquifers

The potential effects of short-circuiting induced by deep perturbation on ASR in a shallower coastal aquifer were subsequently explored. In this case of freshwater storage in a confined, saline aquifer, pressure differences induced by the difference in density between injected freshwater, and native groundwater provoked intrusion of native groundwater in the injected freshwater bubbles via the presumed conduit. It is illustrated that a complete failure of the ASR system can occur when the short-circuiting via such a conduit occurs close to the ASR wells and little mixing with ambient saltwater is allowed.

The negative effects of short-circuiting on ASR on coastal aquifers are mainly related to the hydraulics around the conduits. First, freshwater is not easily transported downwards through the conduits into a deeper aquifer, while it is easily pushed back into the shallower aquifer when injection is stopped or paused. Second, the freshwater reaching a deeper aquifer is subjected to buoyancy effects and migrates laterally in the top zone of this deeper aquifer. Finally, during storage and especially during recovery, the pressure differences in combination with a high hydraulic conductivity rapidly induce a strong flux of saltwater from the whole deeper aquifer into the shallower ASR target aquifer, where a relatively low hydraulic head is present. This short-circuiting induced by such a pressure difference is hampered by the low permeability of the aquitard in a "pristine situation". A continuous, undisturbed aquitard is therefore indispensable for the success of ASR in such a setting, as intrusion of deeper saltwater is not desired.

With an increasing distance between the ASR wells and a nearby conduit, the proportion of mixed saltwater in the recovered water decreases while the arrival time increases. When the conduit is situated outside the radius of the injected freshwater body in the target aquifer, a decrease in RE is not expected.

The Westland field example highlights how design, installation, and operational aspects are vital in the more-and-and more exploited subsurface in densely populated areas. First of all, old boreholes are unreliable and their presence should better be avoided when selecting new ASR well sites (Maliva et al., 2016). Second, installation and operation of (especially injection) wells should be regulated by strict protocols to prevent the creation of new pathways for short-circuiting. Finally, it is important to recognize that similar processes may occur in unperturbed coastal karst aquifers, where natural vertical pipes can be present (Bibby, 1981; Missimer et al., 2002).

4.3 Mitigation of short-circuiting on ASR in coastal aquifers

In order to mitigate the short-circuiting and improve the freshwater recovery upon aquifer storage under these unfavorable conditions, several strategies can be recognized. Obviously, sealing of the conduits would be an effective remedy. However, it may not be viable to (1) locate all conduits, for instance when the former wells are decommissioned or when the confining clay layer is fractured upon deeper injection under high pressure, and (2) successfully seal a conduit at a great depth. This is underlined by the fact that only limited reports of successful sealing of deep conduits can be found.

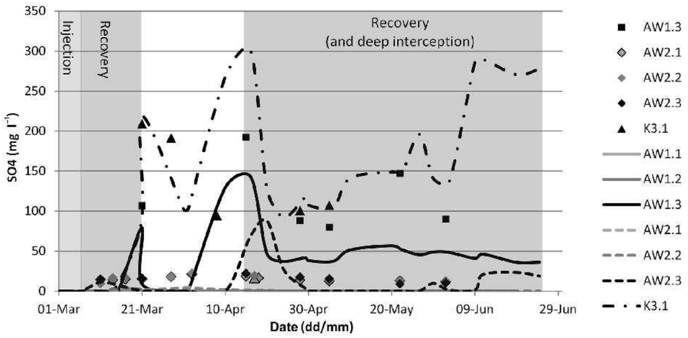

Figure 9. Modeled and observed SO_4 concentrations at the most relevant well screens.

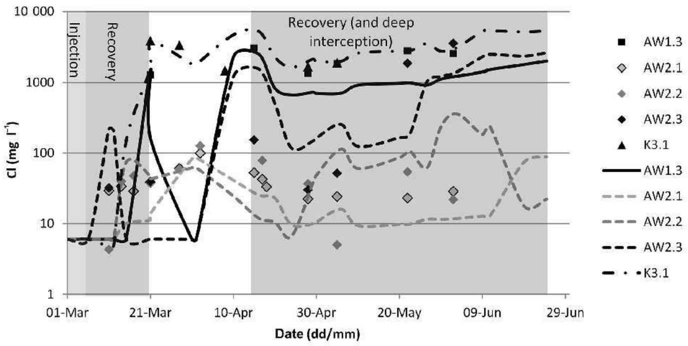

Figure 10. Modeled and observed Cl concentrations at the most relevant well screens.

Apart from sealing, one can also try to deal with these unfavorable conditions. MPPW were installed at the Westland ASR site, for instance, enabled interception of intruding saltwater by using the deeper well screens as "Freshkeepers". After this intervention, about one-third of virtually unmixed injected freshwater becomes recoverable. This way, the RE is brought to a level similar to the level obtained by an MPPW-equipped ASR system without the Freshkeeper interception and without short-circuiting, while the RE would otherwise remain virtually null. It does require interception of a significant volume of brackish-saline groundwater, however, which must be injected elsewhere or disposed of. The addition of a Freshkeeper will therefore inevitably increase the investment costs (additional infrastructure for re-injection/disposal) and operational costs (electricity required for pumping).

A significant part of the unmixed freshwater is blended with saltwater in the Freshkeeper wells, such that the freshwater recovery becomes lower than in the situation in which the Freshkeeper is applied and saltwater intrusion via short-circuiting is absent. At the Westland field site, this is compensated by desalinating the intercepted brackish-saline groundwater, which is a suitable source water for reverse osmosis (RO) thanks to its low salinity. The freshwater (permeate) produced in this process is used for irrigation, while the resulting saltwater (concentrate) is disposed of in Aquifer 2. The resulting RE increase is plotted in Fig. 12. Even when no unmixed freshwater is available, desalination of injected water mixed with groundwater can be continued with this technique to further increase the RE. In comparison with conventional brackish water RO, this leads to a better feed water for RO (lower salinity) while salinization of the groundwater system by a net extraction of freshwater is prevented by balancing the freshwater injection and abstraction from the system.

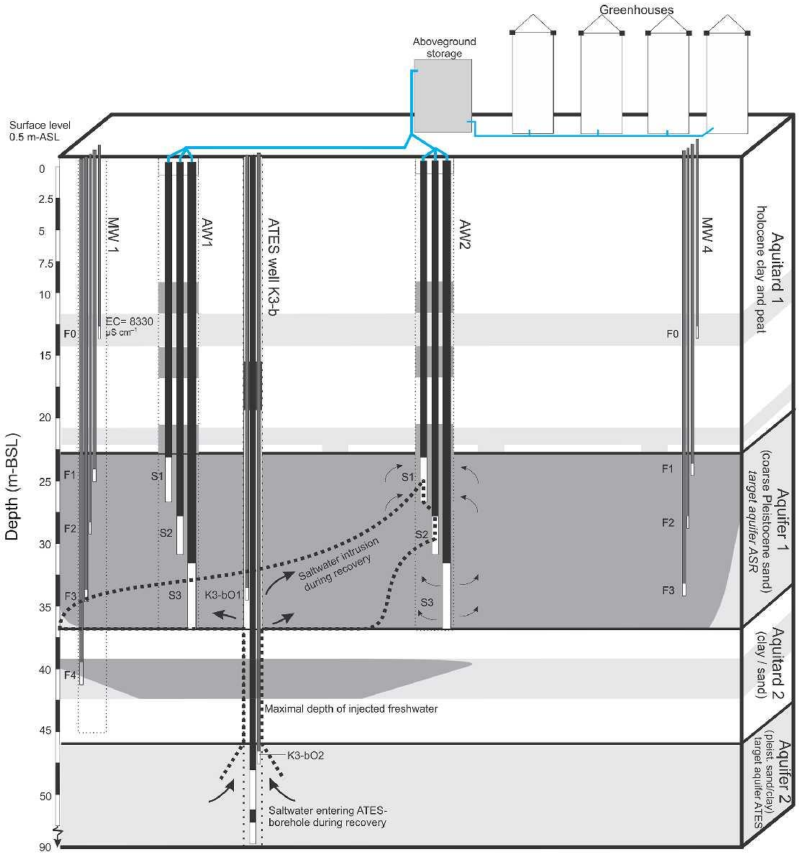

Figure 11. Deep saltwater intrusion via ATES K3-b borehole during shallow recovery of injected freshwater at the Westland ASR site at the start of Cycle 2.

4.4 On the performance of ASR in coastal aquifers without leakage: upconing brackish water from the deeper aquitard

In case of a strict water quality limit and relatively saline groundwater, brackish groundwater upconing from the deeper confining aquitard toward shallow recovery wells is a process to take into account, apart from the buoyancy effects in the target aquifer itself. This was shown by the SEAWAT model runs without short-circuiting, which showed a small increase in Cl concentrations at the ASR wells prior to the full salinization caused by arrival of the fringe of the ASR bubble. The SEAWAT model indicated that the (sandy) clay/peat layer (Aquitard 2) below the target aquifer was the source of upconing brackish-saline groundwater. Although this layer has a low hydraulic conductivity, it is not impermeable and salinization via diffusion can occur in this zone, while brackish pore water can physically be extracted from this aquitard. The transport processes in this deeper aquitard are comparable with the borehole leakage water via conduits in this aquitard: freshwater is not easily pushed downwards during injection, but brackish water is easily attracted during recovery. After the recovery phase this zone salinizes until the next injection phase starts, so a gradual improvement in time is limited. Brackish water may also be attracted from the upper aquitard ("downconing"), but this process is counteracted by the buoyancy effects and did not lead to early termination of the freshwater recovery in the Westland case.

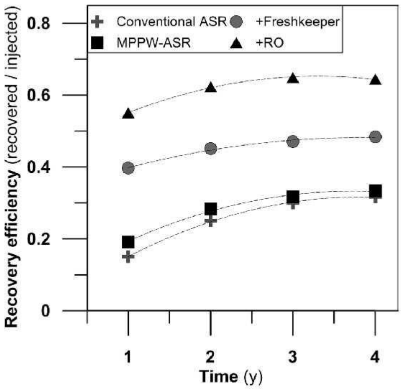

Figure 12. Recovery efficiencies at the Westland ASR site with and without the borehole leakage resulting from the SEAWAT groundwater transport model for a conventional ASR well (one well screen, fully penetrating), deep injection and shallow recovery via multiple partially penetrating wells without 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 abstracted brackish water).

The release of brackish water from the deeper aquitard in coastal aquifers can be relevant when quality limits are strict, the native groundwater is saline, and the native groundwater in the target aquifer is displaced far from the ASR wells. The performance of ASR may then be much worse than is predicted by existing ASR performance estimation methods (e.g., Bakker, 2010; Ward et al., 2009), which assume that impermeable aquitards confine the target aquifer. Even in the first MPPW field test (Zuurbier et al., 2014), this process was not observed, due to a smaller radius of the freshwater bubble, resulting in earlier salinization due to buoyancy effects. The upconing water can optionally be intercepted by a (small, deep) Freshkeeper well screen to extent the recovery of unmixed freshwater, likewise the interception of intruding saltwater at the Westland site.

Finally it should be noted that the ASR system analyzed in this study had very strict water quality limits (practically no mixing allowed) and that a buffer zone (Pyne, 2005) between the injected freshwater and the relatively saline ambient groundwater was not realized before starting the ASR cycles. The boundary conditions for ASR were therefore already unfavorable. Also, the potential improvement after more than three cycles was not explored. The performance of this ASR system should therefore not be considered the typical performance of ASR in a brackish-saline aquifers, which controlled by a complex interplay of geological conditions and operational parameters (Bakker, 2010), well design (Zuurbier et al., 2014, 2015), and the formation of a buffer zone prior to starting the ASR cycles (Pyne, 2005).

5 Conclusions

This study shows how short-circuiting negatively affects the freshwater recovery efficiency (RE) during aquifer storage and recovery (ASR) in coastal aquifers. ASR was applied in a shallow saltwater aquifer (23–37 m b.s.l.) overlying a deeper saltwater aquifer (> 47.5 m b.s.l.) targeted for aquifer thermal energy storage. Intrusion of deeper saltwater was marked by chemical tracers (mainly SO_4) and quickly terminated the freshwater recovery. The most presumable pathway was the borehole of an ATES well at 3 m from the ASR well (forming a conduit) and was identified by field measurements, hydrochemical analyses, and SEAWAT transport modeling. Transport modeling underlined that the potentially rapid short-circuiting during storage and recovery can reduce the RE to null. This is caused by a rapid intrusion of the deep saltwater already during storage periods, and especially during recovery. Transport modeling also showed that when limited mixing with ambient groundwater is allowed, a linear RE decrease by short-circuiting with increasing distances from the ASR well within the radius of the injected ASR bubble is found. Old boreholes should therefore rather be avoided during selection of new ASR sites, or must be situated outside the expected radius.

Field observations and groundwater transport modeling showed that interception of deep short-circuiting water can mitigate the observed RE decrease, although complete compensation of the RE decrease will generally be unattainable since also unmixed freshwater gets intercepted. At the Westland ASR site, the RE can be brought back to around one-third of the injected water, which is comparable to the RE attained with an ASR system without the Freshkeeper in the same, yet undisturbed, setting. With the same Freshkeeper, the setup would be able to abstract around 50 % of the injected water unmixed, if the setting would be undisturbed. This underlines the added value of such a interception well for ASR. Finally, it was found that brackish water upconing from the underlying aquitard towards the shallow recovery wells of the MPPW-ASR system can occur. In case of strict water quality limits, this process may cause an early termination of freshwater recovery, yet it was neglected in many ASR performance estimations to date.

6 Data availability

The data used in this manuscript can be obtained by contacting the authors.

Competing interests. The authors declare that they have no conflict of interest.

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ANNEX C: Hydrochemical observations at ASRRO and BWRO

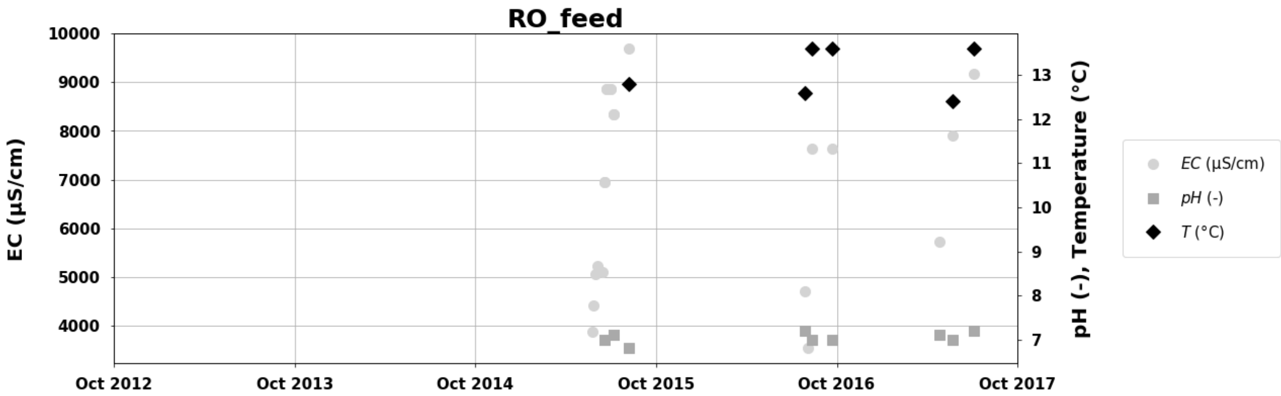

Figure 37: Electrical conductivity (EC in µS/cm), pH (-), and temperature (Temp in °C) of brackish groundwater intercepted to feed the BWRO-system.

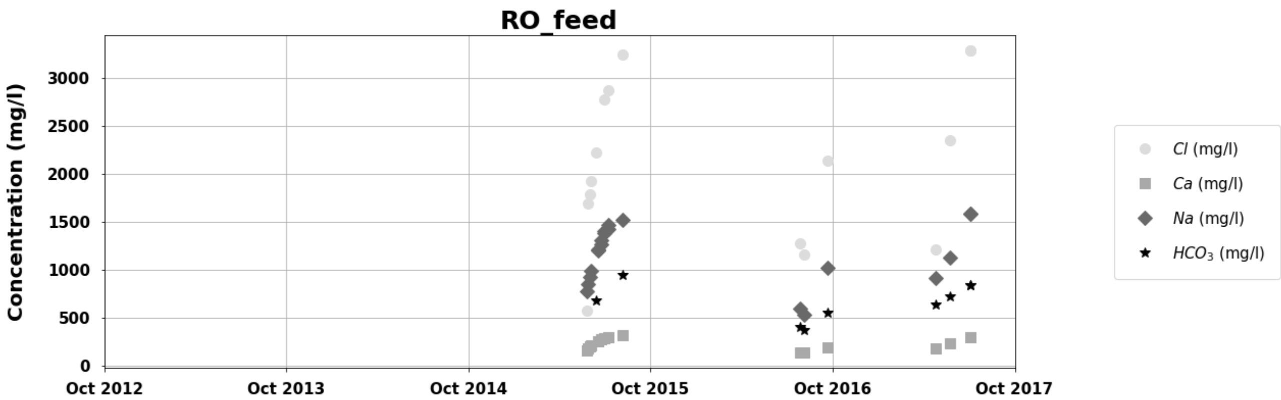

Figure 38: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater intercepted to feed the BWRO-system.

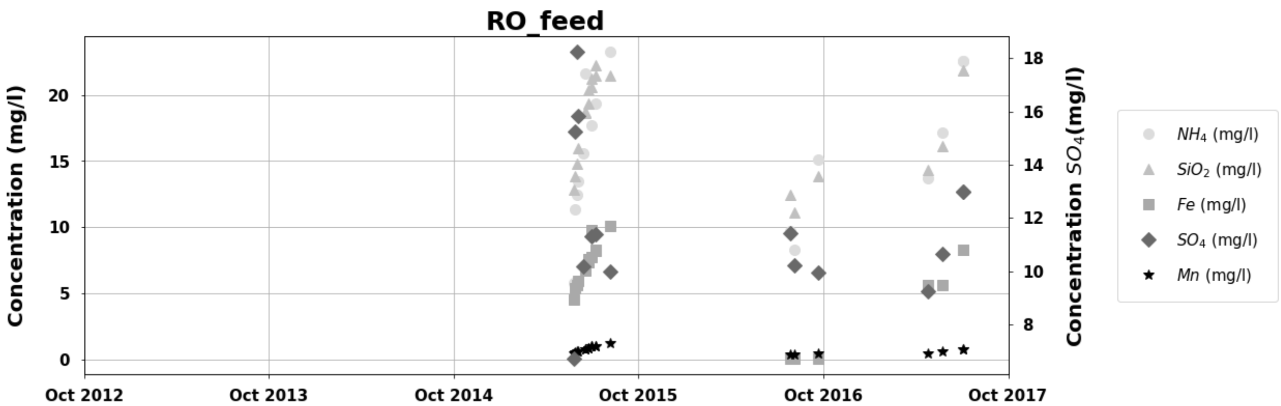

Figure 39: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater intercepted to feed the BWRO-system.

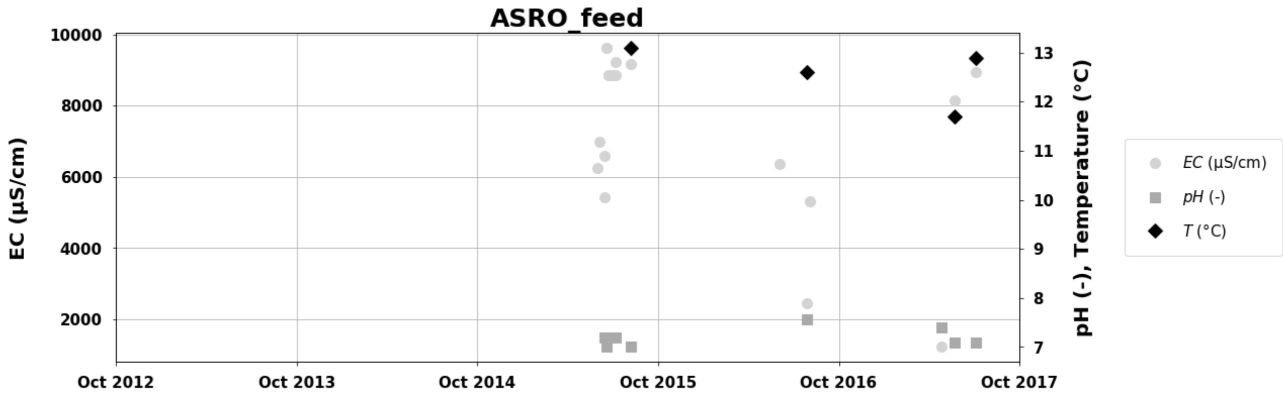

Figure 40: Electrical conductivity (EC in µS/cm), pH (-), and temperature (Temp in °C) of brackish groundwater intercepted to feed the ASRRO-system.

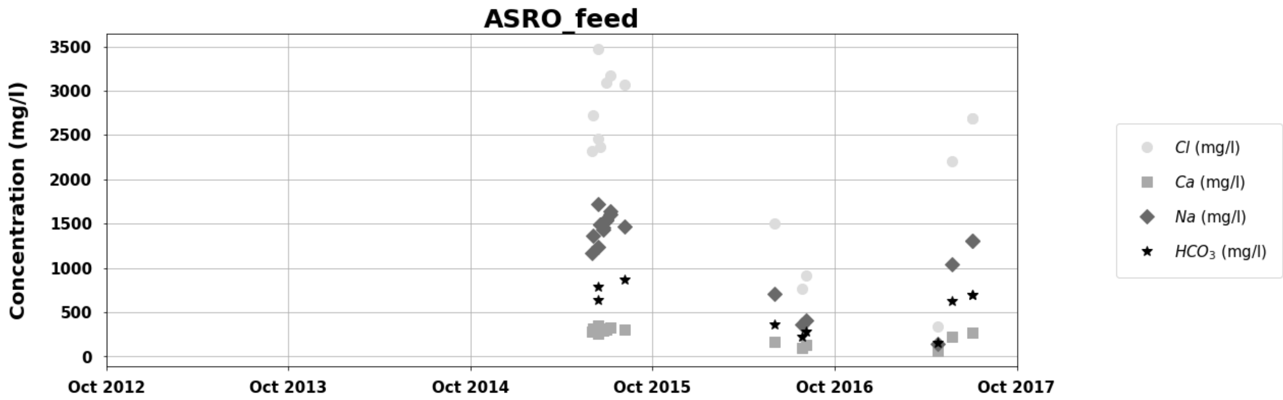

Figure 41: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater intercepted to feed the ASRRO-system.

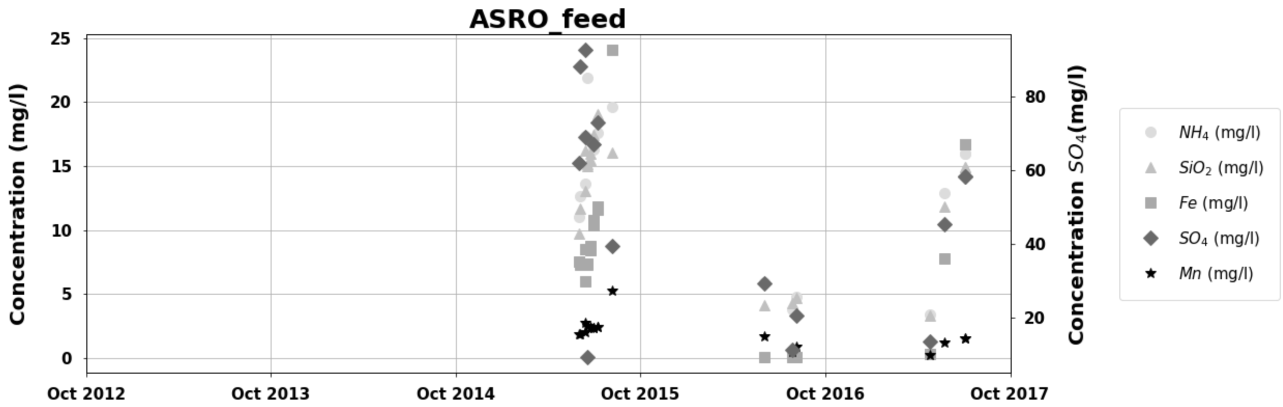

Figure 42: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater intercepted to feed the ASRRO-system.

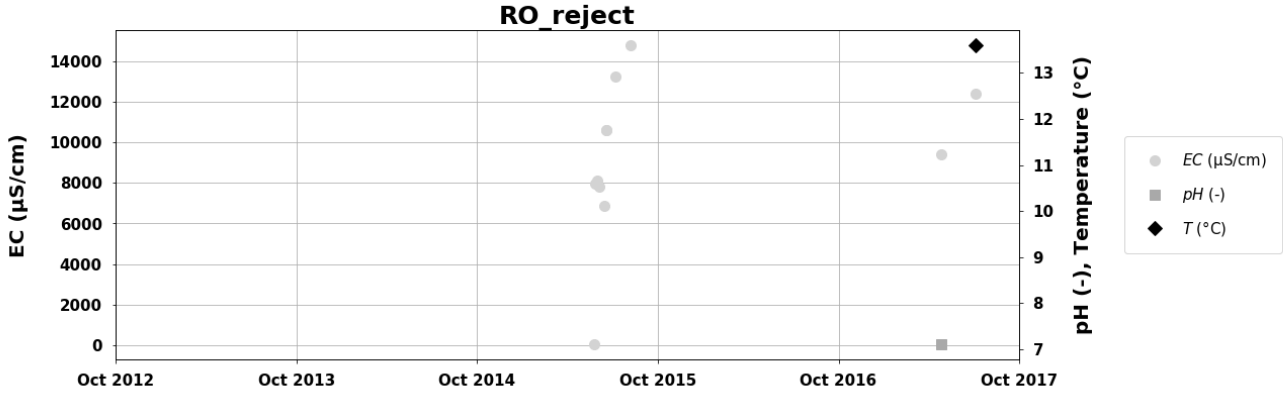

Figure 43: Electrical conductivity (EC in µS/cm), pH (-), and temperature (Temp in °C) of brackish groundwater rejected by the BWRO-system.

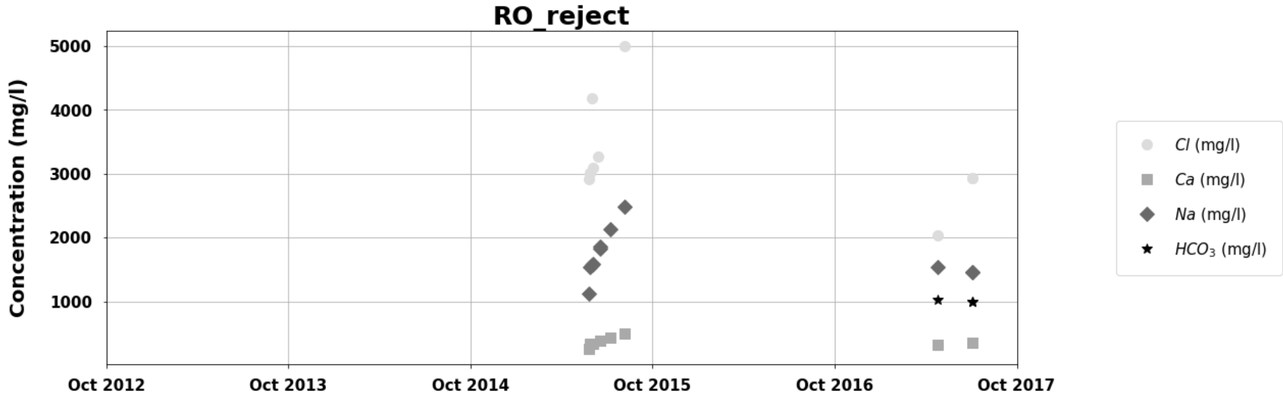

Figure 44: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater rejected by the BWRO-system.

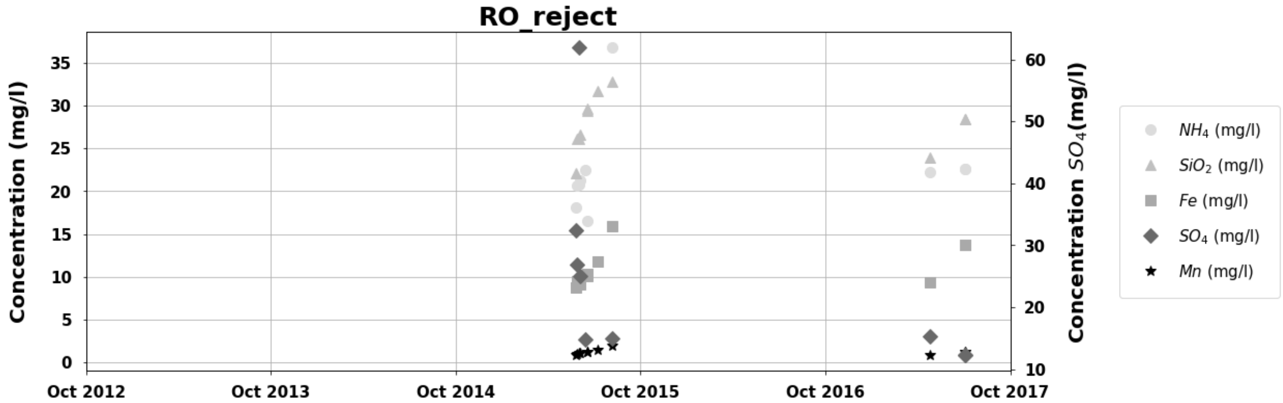

Figure 45: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater rejected by the BWRO-system.

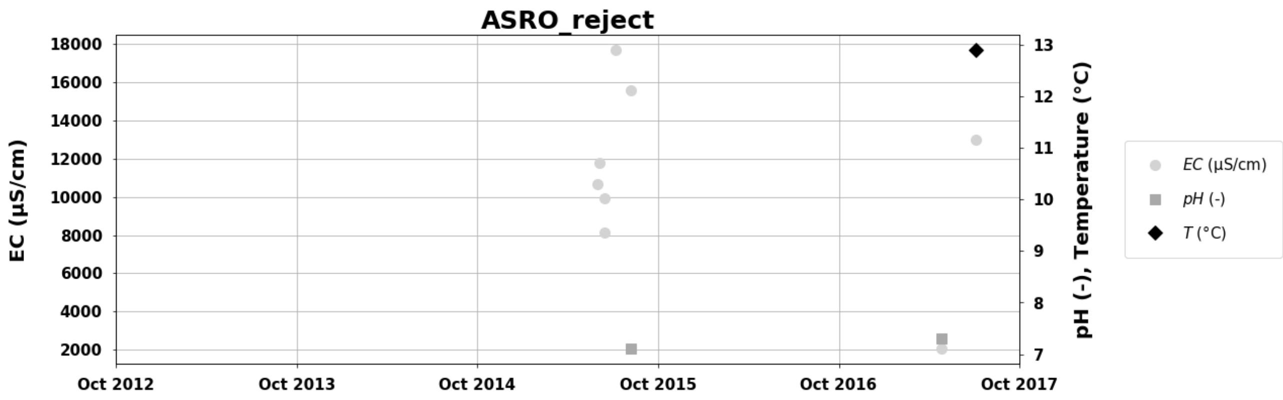

Figure 46: Electrical conductivity (EC in µS/cm), pH (-), and temperature (Temp in °C) of brackish groundwater rejected by the ASRRO-system.

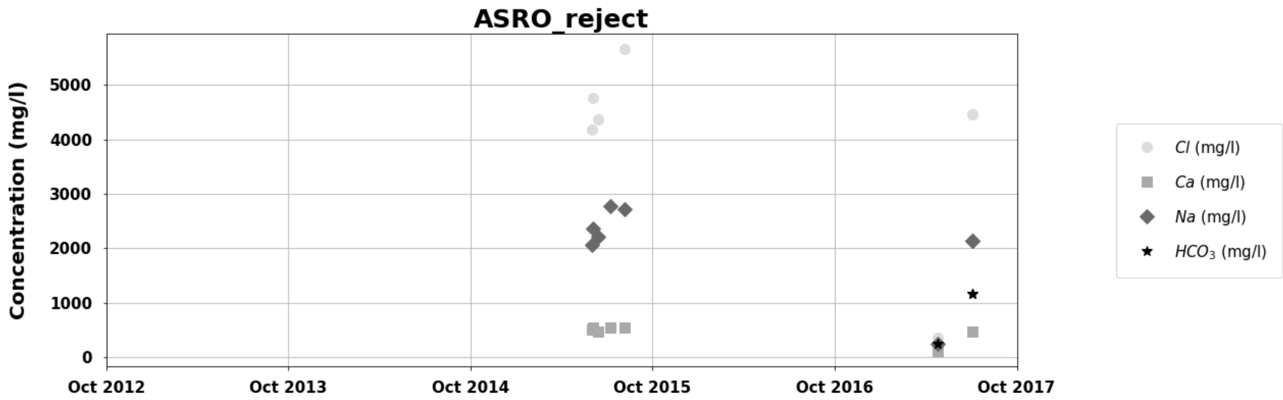

Figure 47: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater rejected by the ASRRO-system.

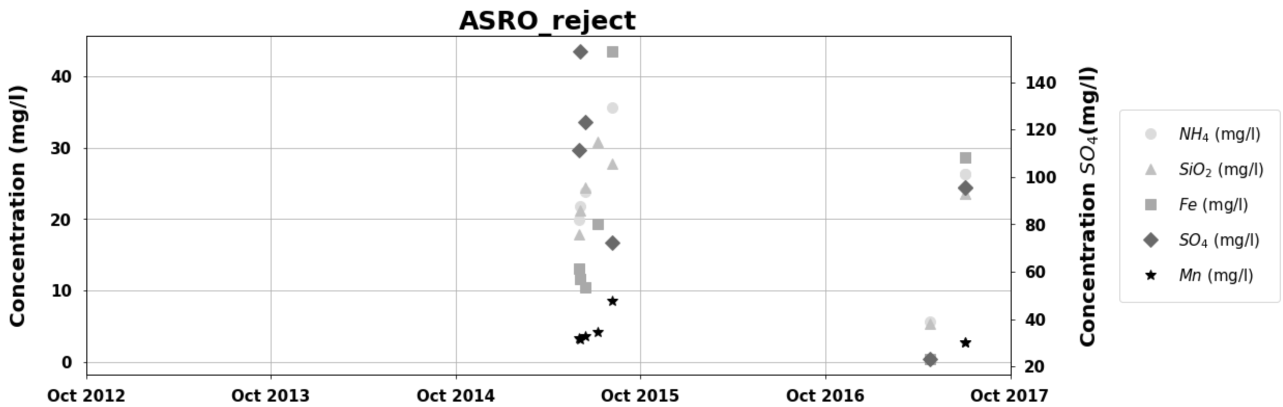

Figure 48: Concentrations of Cl, Ca, Na, and HCO_3 in brackish groundwater rejected by the ASRRO-system.

Annex D: Comparison field and model data

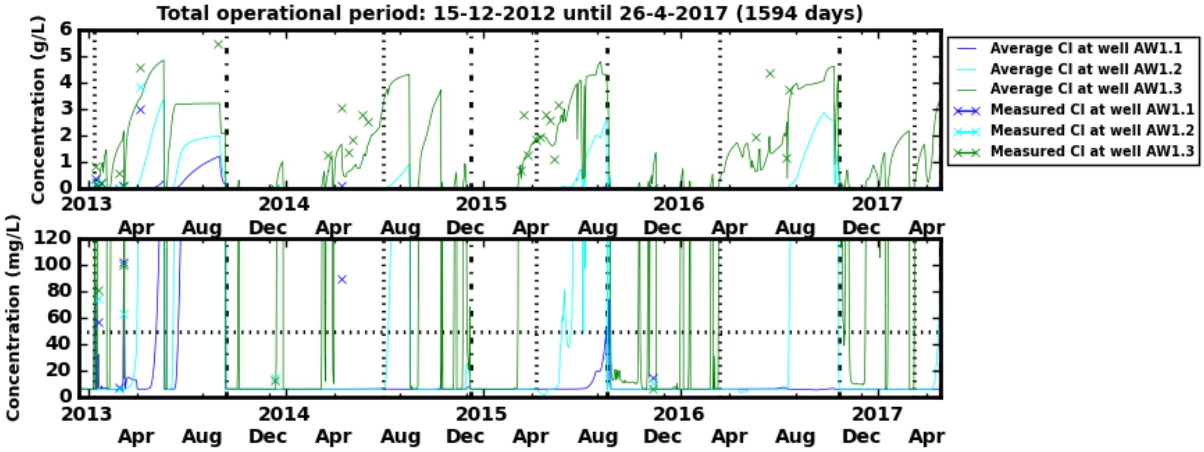

Figure 49: Measured (crosses) and modelled (solid lines) chloride concentration in water in the different well screens of the ASR-well (AW1) at Westland from 15 December 2012 until 26 April 2017. The black dotted and dashed vertical lines indicate the start of the infiltration and recovery period respectively. The horizontal dashed line in the bottom graph represents the maximum allowable concentration limit of chloride upon abstraction.

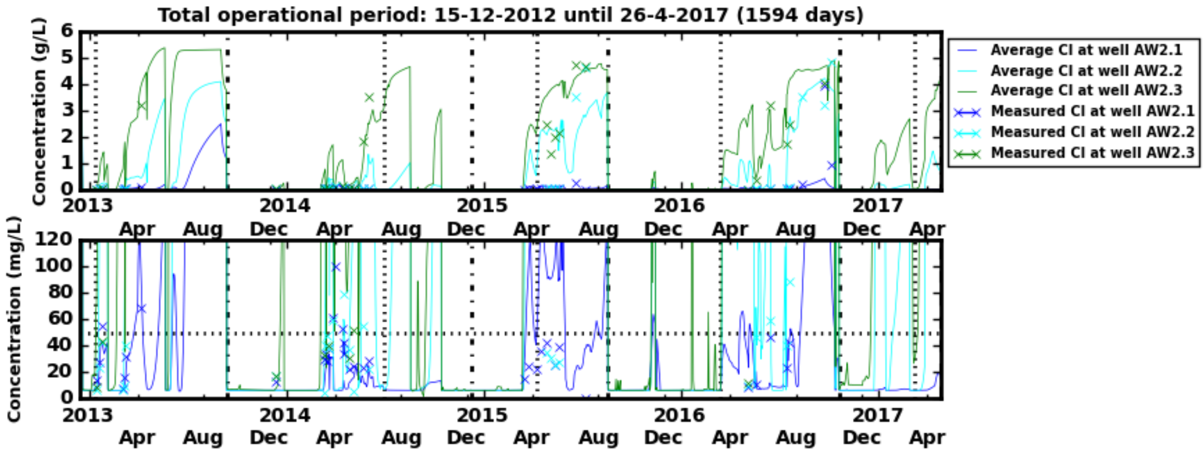

Figure 50: Measured (crosses) and modelled (solid lines) chloride concentration in water in the different well screens of the ASR-well (AW2) at Westland from 15 December 2012 until 26 April 2017. The black dotted and dashed vertical lines indicate the start of the infiltration and recovery period respectively. The horizontal dashed line in the bottom graph represents the maximum allowable concentration limit of chloride upon abstraction.

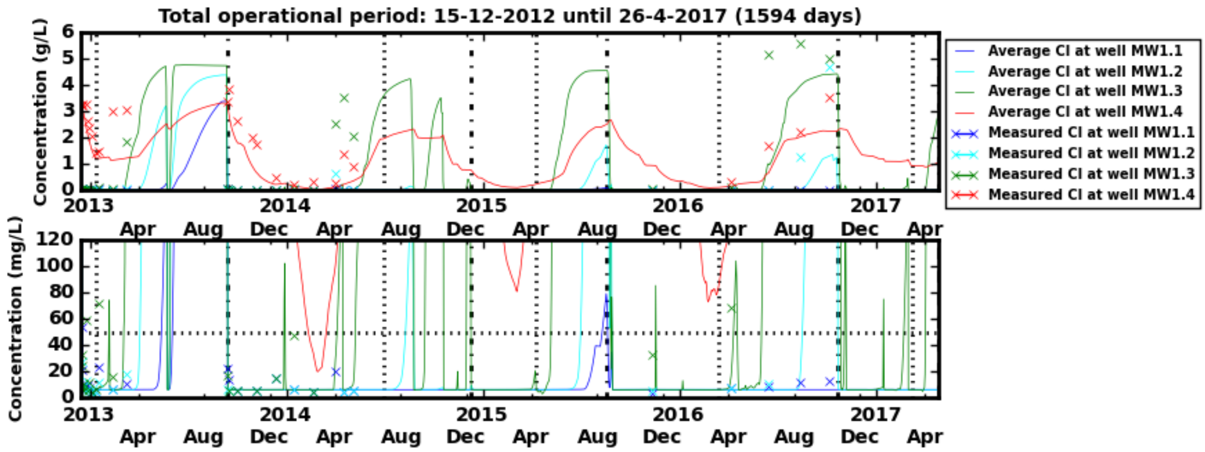

Figure 51: Measured (crosses) and modelled (solid lines) chloride concentration in water in the different well screens of the monitoring well (MW1) at Westland from 15 December 2012 until 26 April 2017. The black dotted and dashed vertical lines indicate the start of the infiltration and recovery period respectively. The horizontal dashed line in the bottom graph represents the maximum allowable concentration limit of chloride upon abstraction.

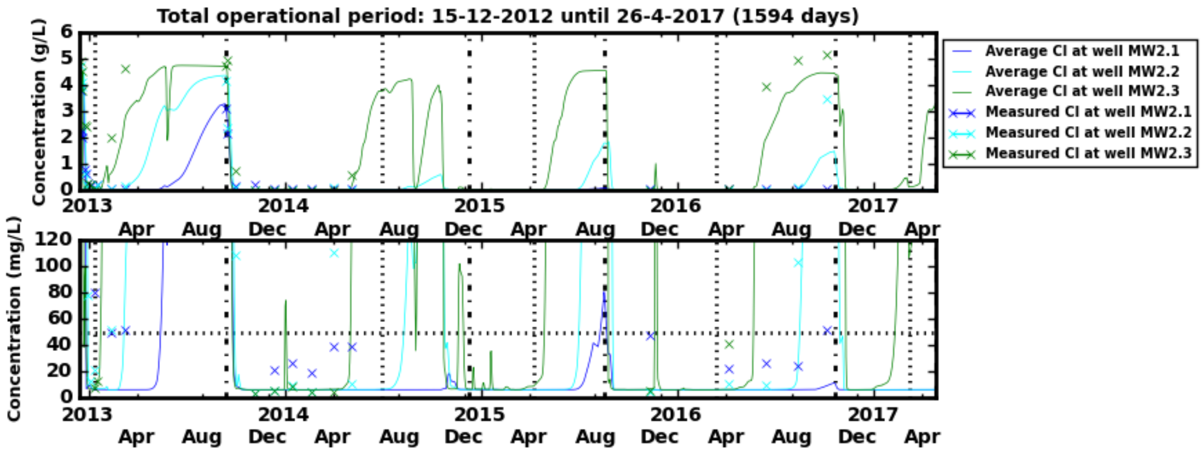

Figure 52: Measured (crosses) and modelled (solid lines) chloride concentration in water in the different well screens of the monitoring well (MW2) at Westland from 15 December 2012 until 26 April 2017. The black dotted and dashed vertical lines indicate the start of the infiltration and recovery period respectively. The horizontal dashed line in the bottom graph represents the maximum allowable concentration limit of chloride upon abstraction.

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