Water quality changes during Aquifer Storage and Recovery (ASR): results from pilot Herten (Netherlands), and their implications for modeling

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Abstract

ASR (Aquifer Storage and Recovery) is a water management technique to store surplus water in an aquifer, for use during future needs, using dual purpose wells (that both inject and recollect). The quality changes of drinking water during a recent ASR test in a deep anoxic, sandy aquifer in the Southern Netherlands were dictated by (redox) reactions with organic material, pyrite and manganous siderite. These reactions decreased during successive ASR-cycles by leaching, coating with iron(hydr)oxides and increasing pH.

Tests with adding O_2 and $NaNO_3$ to the injection water, to speed up aquifer inactivation, failed probably by not adding also a pH buffer. An anoxic zone developed around the ASR bore hole wall during storage, by decay of microbes grown during injection.

Iron and manganese, which dissolved more remote from the ASR well, immobilized in the most aerobic zone during recovery. This is explained by sorption to iron(hydr)oxides that precipitated during previous injection and storage phases. As a result, with increasing ASR cycles the recovery efficiency (dictated by iron removal) improved.

The water quality changes have been modelled with an ASR version of the Easy-Leacher model, incorporating the crucial changes in the ASR-proximal zone and the effects of sorption to iron(hydr)oxides.

Keywords

Aquifer storage and recovery, ASR, hydrochemistry, pyrite oxidation, siderite dissolution, modeling.

INTRODUCTION

Aquifer Storage and Recovery (ASR) is a water management technique, in which water is stored in an aquifer during periods of water excess, and recovered by the same (injection) well from the groundwater reservoir during periods of water shortage (Pyne, 1995). ASR may also yield financial benefits by reducing the peak factor in water production (and water treatment) and by raising the security of water delivery.

ASR is an increasingly popular technique with a rapidly expanding number of operational sites in the world. In the Netherlands, however, notwithstanding large-scale application of artificial recharge since 1955, no ASR-system is currently working. This failure relates to the following: (a) decreased water demands in the past years do not urge for increasing subsurface storage of drinking water in the ‘wet’ Netherlands; and (b) it is feared that a quality deterioration of (drinking) water during subsurface storage, especially regarding Fe and Mn, will necessitate a costly post-treatment of the water recovered.

Recent bench marks of drinking water prices in the Netherlands push water supply companies to lower their costs. ASR, if operating without quality deterioration, is thus becoming an interesting option also in the Netherlands, mainly to reduce water treatment costs by peak shaving.

In 1999 Water Supply Company Limburg (WML) therefore started with an ASR-experiment in Herten (Limburg), to test the technical feasibility of ASR, not only for that site but also for a larger scale application in the province of Limburg (notably the Ruhr Valley graben). The main questions to be answered were: (1) are there hydrological constraints associated with ASR in this region? and (2) are the water quality changes in the target aquifer, of drinking water from production plant Herten, such that they really necessitate a second treatment? This paper is based on a report by Wakker et al. (2003).

MATERIAL AND METHODS

The ASR test well and 2 observation wells are shown in cross section in Fig. 1. All available well screens (10) have been sampled on a bidaily to biweekly basis for inorganic and microbiological chemical analysis, using conventional purging and analytical procedures. In total 10 aquifer cores of 1 m length were taken from the whole target aquifer (159–170 m below land surface) and were analysed on their geochemical reactivity (Buijs and Van der Grift, 2001). The infiltration and pumping rate was on average 45 m^3/h.

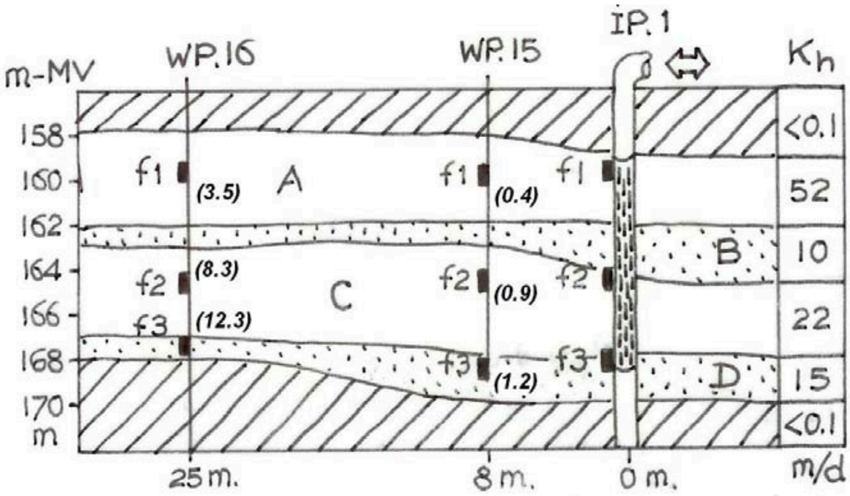


Figure 1. Position of the ASR-well IP1 and the 2 observation wells WP15 and WP16 in cross section, with the hydrogeological structure, coding of aquifer layers (A-D), position of well screens (f1-f3= observation well screens), and mean travel time from/to IP.1 (within brackets, in days). K_h = horizontal hydraulic conductivity.

The ASR-experiment in Herten lasted from October 2000 till February 2003. It consisted of 4 ASR cycles (Fig. 2), of which 2 cycles were carried out with additional dosage of oxidant in order to inactivate reactive aquifer phases (notably minerals containing iron and manganese) with the purpose to recover drinking water that would not require any post-treatment. The ASR-cycles were not in normal order (being injection, storage, recovery etc.), for various testing purposes. Results of the test have been simulated and extrapolated to future ASR cycles on a production plant, by using the expert model EL-ASR of Kiwa Water Research, a new variant of Easy-Leacher (Stuyfzand, 1998).

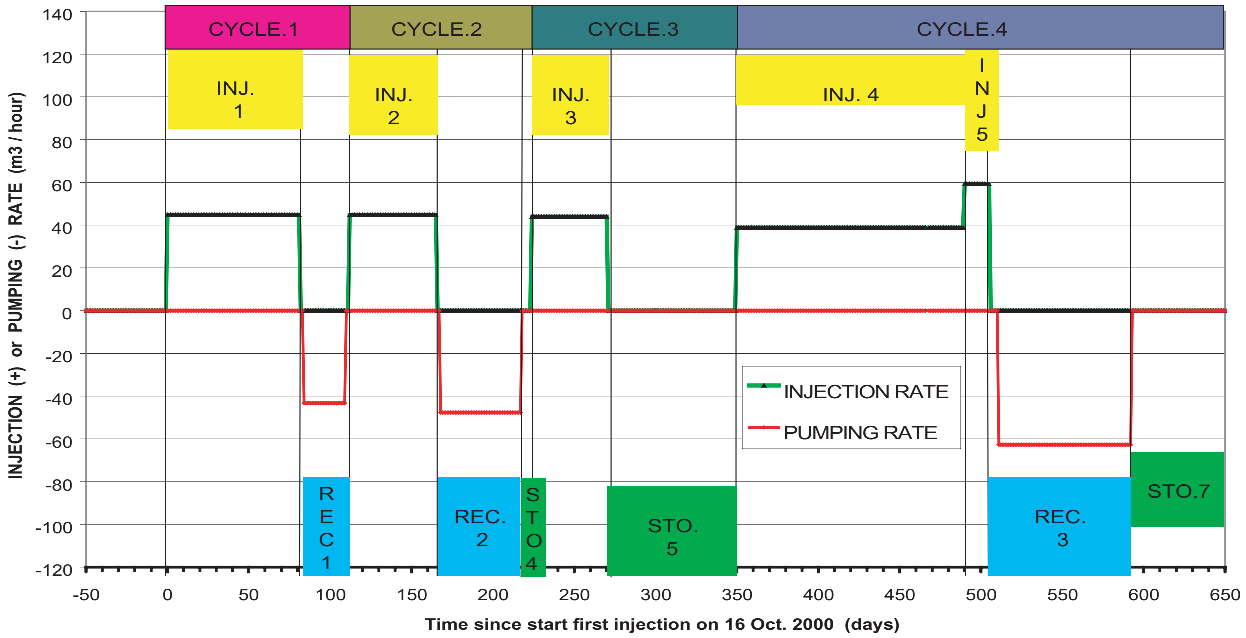


Figure 2. Variation in injection and pumping rate during the discerned periods and ASR cycles. Oxygen was added during injection period 3 (34 mg/L; dissolved thanks to >17 bar water pressure at depth of injection screen), and $NaNO_3$ during injection period 5 (14.6 mg NO_3/L). Storage periods 1-3 too short for marking.

THE TARGET AQUIFER

The target aquifer is composed of fluvial sands of Pliocene age. The grain size distribution, geochemical analyses and reactivity experiments (in an O_2 and CO_2 controled reaction chamber) revealed the presence of 4 aquifer layers (Fig. 1). All layers are deep anoxic, practically without $CaCO_3$, and contain small but significant amounts of reactive BOM (bulk organic material; 0.02–8%) and pyrite (210–1,050 mg/kg d.w.). Only layer D contains a significant amount of reactive manganous siderite (< 0.25%).

The native groundwater composition (Table 1) can be characterized as pH-neutral, calcareous (slightly calcite undersaturated), deep anoxic (however without methane), oligohaline-fresh, high in iron, unpolluted, and of the $Ca(HCO_3)_2$– type.

CLOGGING OF THE ASR-WELL

The ASR-well in Herten did not clog (Fig. 3). Especially during the additional oxygen dosage there was a temporary increase in resistance at the well screen (0.2–0.8 m; probably due to gas bubbles), but this disappeared again during the next storage phase. During storage and recovery the ASR-well regenerated spontaneously. The excellent infiltrability of Herten drinking water was to be expected, because it satisfies all general quality guidelines of infiltration water for ASR application to sandy aquifers (see for instance Perez-Paricio and Carrera, 1998; Hijnen et al., 1998).

WATER QUALITY CHANGES IN THE TARGET AQUIFER

Mean changes

The test results demonstrate that the quality of the injected drinking water changed indeed (Table 1). There were (small) increases for sulphate, iron, manganese, arsenic, barium, cobalt, nickel, zinc and colony counts 22°C, and

Table 1. Mean water quality during the 3 ASR cycles in the period 16 October 2000 till 11 June 2002. IP.1-f0 = ASR injection/pumping well screen.

SAMPLE POINT	t50 or daynr. day	pH field	O2-field	Cl	SO4	HCO3	NO3	Ca	Fe	Mn	NH4	As	Co
			mg/L									g/L	
INJECTION (mean input, for each monitoring well samples with comparable number of pore flushes)													
INPUT	0	7.47	12.0	22.5	26.0	220	1.1	73.0	0.01	0.01		0.5	0.5
WP 15-f1	0.4	7.31	6.7	21.0	28.8	213	1.0	71.3	0.01	0.03	0.03	2.5	6.0
WP 15-f2	0.8	7.38	8.1	21.3	31.8	224	1.2	74.0	0.01	0.01	0.03	2.8	8.5
WP 15-f3	1.2	7.22	5.2	18.3	26.3	210	10.0	70.0	0.07	0.02	0.03	1.5	0.5
WP 16-f1	3.5	7.11	0.5	21.0	36.0	201	0.3	70.0	1.27	0.04	0.03	2.8	0.5
WP 16-f2	8.3	7.11	2.5	21.3	34.4	209	0.4	70.9	0.46	0.07	0.03	1.8	5.0
WP 16-f3	12.1	7.05	1.0	21.0	28.0	205	1.7	64.0	2.30	0.11	0.03	1.5	0.5
STORAGE (mean all periods)													
IP.1-f1	58	7.11	0.6	21.8	40.7	215	0.5	77.0	0.64	0.07	0.07	7.0	4.1
IP.1-f2	98	7.10	0.8	21.5	45.0	220	0.4	84.5	0.94	0.17	0.10	3.0	14.0
IP.1-f3	58	7.02	0.8	21.5	40.1	226	0.4	84.1	0.11	0.14	0.04	1.9	1.3
WP 15-f1	59	7.09	0.9	20.8	38.0	208	0.4	71.4	0.05	0.06	0.03	2.9	6.1
WP 15-f2	29	7.00	0.5	22.5	39.8	216	0.4	74.1	0.01	0.09	0.03	3.4	26.3
WP 15-f3	59	6.88	0.3	21.7	39.7	225	0.3	76.0	1.03	0.40	0.03	1.9	7.2
WP 16-f1	58	7.00	0.5	21.9	44.4	200	0.3	71.2	0.81	0.08	0.03	4.8	5.8
WP 16-f2	29	6.82	0.8	21.5	41.2	206	0.3	70.0	1.55	0.20	0.03	4.8	8.3
WP 16-f3	58	6.91	0.3	22.1	39.4	205	0.3	67.0	4.21	0.22	0.07	3.1	0.6
RECOVERY (mean all periods)													
IP.1-f0	20	7.16	0.8	20.7	33.0				0.01	0.02			
IP.1-f1	18	7.18	1.4	20.9	37.5	208	1.1	70.7	0.02	0.01	0.03		
IP.1-f2	21	7.18	1.2	20.8	31.4	211	0.8	69.4	0.02	0.02	0.03		
IP.1-f3	18	7.11	1.9	21.4	31.5	221	0.5	71.2	0.01	0.02	0.03		
WP 15-f1	18	7.18	1.1	20.7	35.1	211	1.4	70.6	0.02	0.03	0.03		
WP 15-f2	17	7.24	2.0	20.9	31.3	212	1.9	69.1	0.01	0.02	0.03		
WP 15-f3	18	7.07	0.6	20.9	29.9	213	0.5	68.6	0.57	0.10	0.03		
WP 16-f1	22	7.16	0.4	19.7	30.9	204	0.6	69.0	1.04	0.05	0.03		
WP 16-f2	17	7.14	0.5	19.6	25.5	210	0.3	67.3	2.25	0.10	0.04		
WP 16-f3	22	7.02	0.4	19.7	27.1	202	0.3	64.0	3.36	0.16	0.06		
NATIVE GROUNDWATER													
IP.1-f0	0	7.00	<0.5	7	<5	300	<0.5	72.0	5.60	0.20	0.14	12.0	4.0

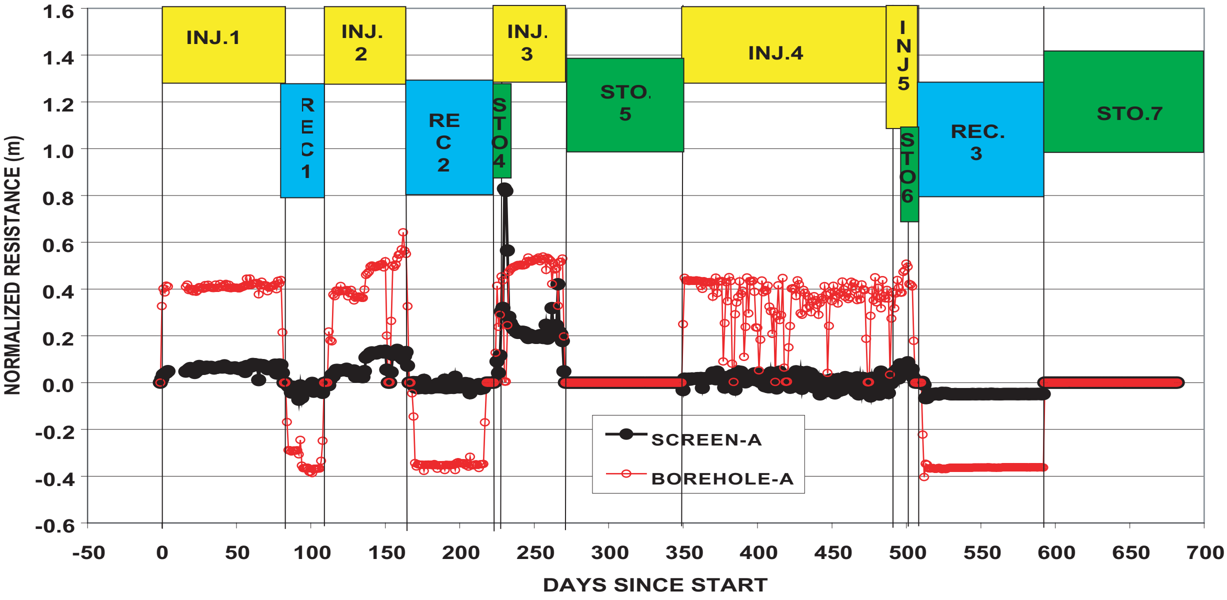


Figure 3. Time plot for the normalized resistance by the ASR well screen (Screen) and the borehole wall (Borehole) in aquifer layer A. The curves do not show a structural head build-up during injection (INJ) and backpumping (REC), thus showing that the ASR well did not clog. Both storage (STO) and recovery contribute to the reduction of the hydraulic resistance formed during injection.

decreases for oxygen, nitrate, silicate, pH, hydrogencarbonate and the calcite saturation index. Ammonia and methane did not show significant changes (< 0.05 mg/L), nor did organic micropollutants (all below detection) and indicators of faecal contamination (not found).

The observed changes are mainly caused by the oxidation of BOM and pyrite (FeS_2), and the oxidation or dissolution of a manganous siderite ($Fe_{0.90-0.95}Mn_{0.05-0.1}CO_3$). In the upper aquifer layer (A) pyrite oxidation is the maior reaction (largest increases of sulphate, arsenic, cobalt, nickel and zinc), and in the lowest layer (D) the weathering of siderite is dominating (iron, manganese and TIC increasing most).

The results in Table 1 reveal that the mean quality changes are stronger during storage than during recovery. This is curious, but can be explained as follows. First, the longer storage phases 4 and 7 were preceded by recovery periods, and thus reflect water that remained in the aquifer for a longer time than the water previously recovered. And second, the other long storage phase 5 followed upon the exceptional injection phase 3 with additional O_2 inputs (34 instead of 9.5 mg/L), and was not followed by a recovery phase.

Development of an anoxic zone around the borehole wall

A remarkable phenomenon was the development of a more anoxic zone of several meters around the ASR well during storage (Fig. 4), probably indicating the die-off of a microbiological community that formed there under

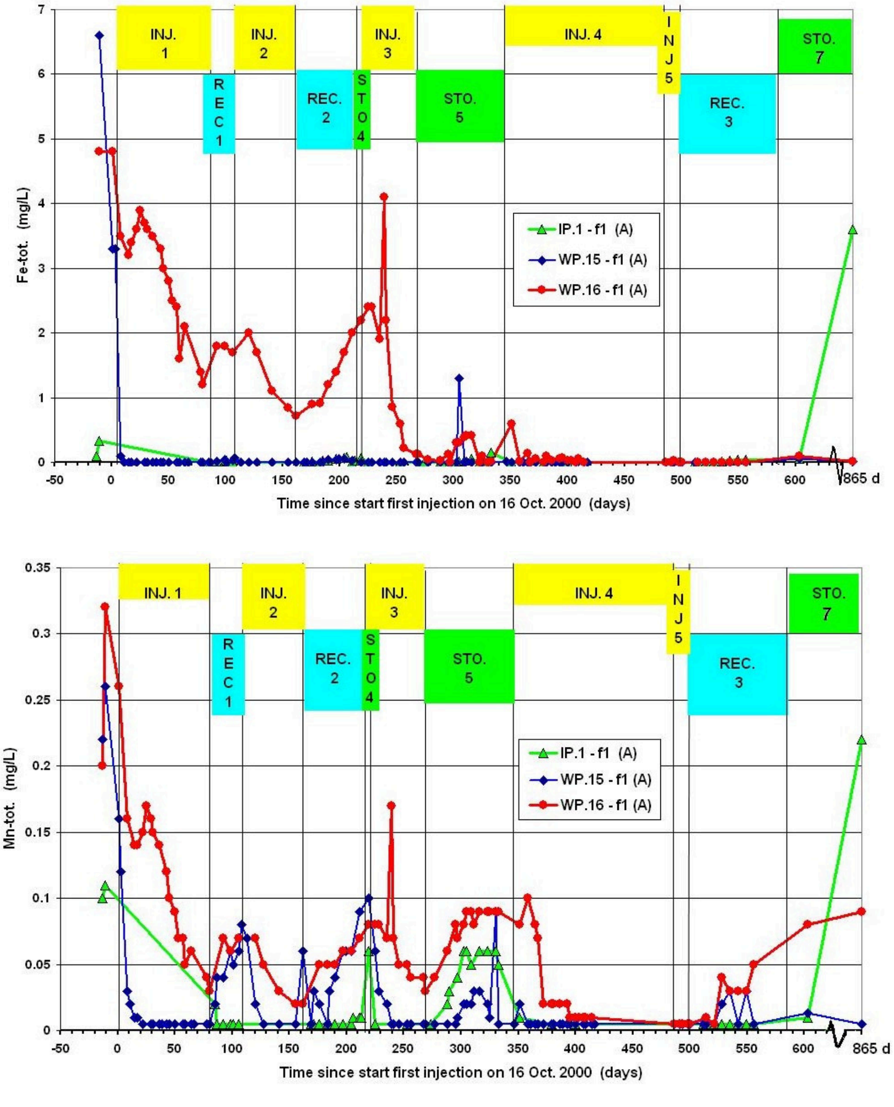


Figure 4. Plot of the iron and manganese concentrations in upper aquifer zone A, in observation screen f1 of the ASR-well and WP.15 (8 m, 0.4–1.2 days) and WP.16 (25 m, 3.5–12 days). Periods with injection (INJ), storage (STO) and recovery (REC) are numbered as in Fig. 2

(sub)oxic conditions during injection (Stuyfzand et al., 2002; Vanderzalm et al., 2002). This is deduced amongst others from the stronger Mn and Fe mobilisation in the observation screens within the ASR-well (amongst others IP.1-f1) during storage phase 5 and especially 7, as compared to observation wells WP.15 and 16 at resp. 8 and 25 m distance.

Subsurface Iron Removal (SIR)

Problems with iron and manganese in the water recovered arose during low O_2–concentrations (<0.5 mg/L) and low pH (<7.1) in the aquifer, especially during the first few ASR cycles. During the recovery phase, after starting up the system, the so-called Subsurface Iron Removal process (Van Beek, 1983; Appelo et al., 1999) strongly reduced the concentrations of iron (Fig. 5) and, to a less extent, those of manganese and ammonia.

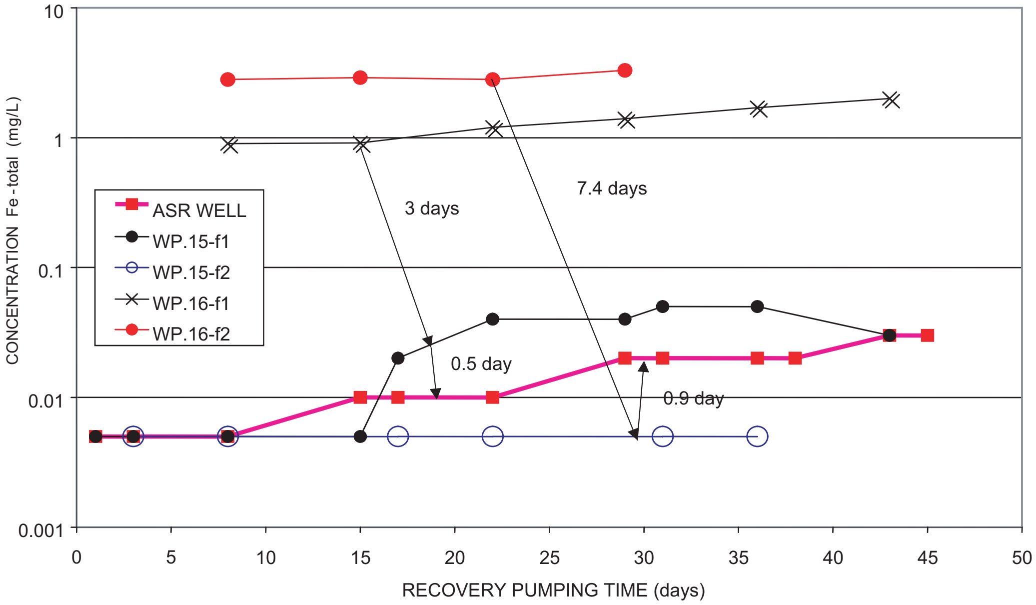
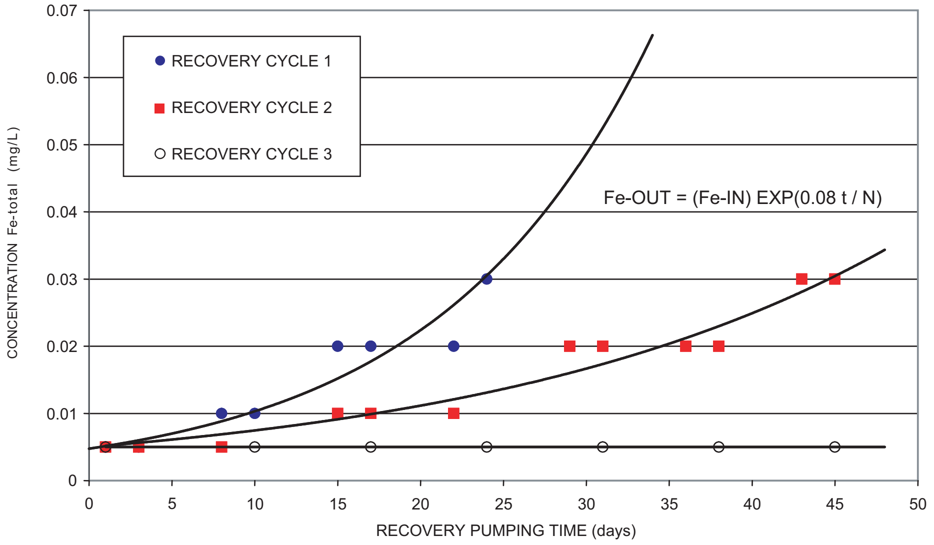


Figure 5. Developments in the iron concentration during recovery period 2 (after 2 days of storage following injection period 2), in the ASR-well (the water recovered), and on 4 sites in the aquifer. The arrows indicate how the iron concentration is changing, within the indicated time (by sorption), during backflow from WP16 (25m) via WP15 (8m) to the ASR well

Extinction of water quality changes

In accordance with experiences elsewhere (Pyne, 1995), also here the reactions of the infiltration water with reactive aquifer components gradually diminished (with increasing cumulative injection volume). Consequently the iron and manganese concentrations decreased (Figs. 4 and 6). This is explained by: (1) leaching of pyrite and siderite; (2) coating of them by iron(hydr)oxides, which reduces their reactivity; and (3) a pH increase in the aquifer thanks to the extinction of the above mentioned oxidation reactions.

During recovery, in addition, the efficiency of the SIR process increases (Van Beek, 1983; Appelo et al., 1999), which leads to further declining concentrations of iron, manganese and ammonia.



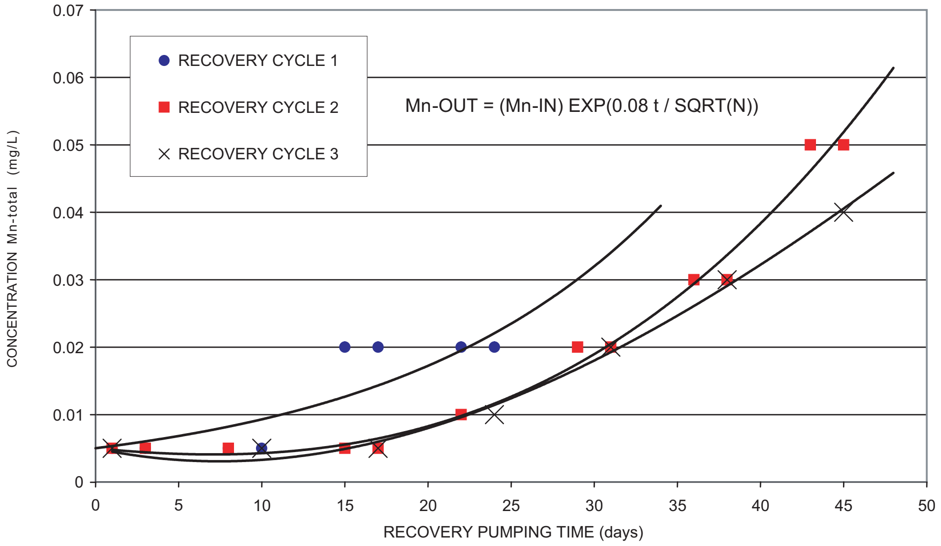


Figure 6. Plot of concentrations of iron (above) and manganese (below) in the backpumped water during recovery cycles N = 1-3 of the Herten ASR test

EFFECTS OF DOSING CHEMICALS

Current experiences

In current ASR practice oxidizing, desinfecting and pH-raising chemicals are dosed to the infiltration water, if needed. The success of ASR in the USA can, as a matter of fact, be partly attributed to the dosage of sodium hydroxide and a desinfectant.

These additives are useful in: (1) preventing exorbitant growth of bacteria; (2) coating the reactive iron minerals with iron(hydr)oxides, (3) keeping the aquifer (sub)oxic for a longer time, and (4) buffering the acidifying action of oxidation reactions. All this may indeed prevent the dissolution of Fe^{2+}, Mn^{2+} and sometimes arsenic, as well as microbiological problems.

Oxygen (tested in Herten)

The positive effects of O_2-dosage (on average 25 mg/L) to nearly saturated infiltration water (9.5 mg/L), during injection period 3, were overshadowed during storage phase 5 by a strong pH decline due to enhanced oxidation processes. This pH drop caused manganese problems during the subsequent storage period. The pH should be prevented to drop below 7-7.2, by adding NaOH.

Nitrate (tested in Herten)

The dosage of nitrate (0.22 mmol $NaNO_3$/L) to the infiltration water (with only 0.02 mmol NO_3/L), during injection period 5, contributed to an accelerated oxidation of BOM and pyrite, with 75% less production of acid (advantageous) and a slower consumption rate (also advantageous because nitrate that way keeps the water (sub)oxic for a longer time than oxygen does). However, from an esthetic point of view the addition of oxygen is to be strongly preferred.

Sodium hydroxide (not tested in Herten)

The acidifying effects, in consequence of adding oxygen, can be easily buffered by also addin NaOH or another pH raising agent.

Desinfectant (not tested in Herten)

In the USA chlorine or hypochlorite is dosed for preventing the development of a dense, microbial community around the borehole wall, because microbes both clog the well and consume O_2 and NO_3. In the Netherlands, these oxidants have been largely abandoned due to the formation of hazardous byproducts. Therefore alternatives are needed, for instance consisting of intermittent (1) backpumping at a high flow rate after juttering with compressed air, or (2) injection of water containing ozone or H_2O_2, followed by the actions mentioned under 1.

MODELING

The ASR testing did not reflect the normal ASR operation in a year, with for instance 150 days of injection, followed by 60 days of storage, 120 days of recovery and 35 days of stand-still. This means that simulations and extrapolations with a transport model were needed, in order to predict the performance of a true ASR plant.

Various models are nowadays available (amongst others PHT3D of Prommer et al., 2003; SWIFT-PHREEQC of Gauss et al., 2000) that could do the job, however, after various extensions to account for: (a) development of an anoxic zone around the borehole wall; (b) the process of subsurface iron removal; and (c) extinction of water quality changes.

A less laborious route was followed by transforming the expert model Easy-Leacher (Stuyfzand, 1998) into a transport code fit for ASR (EL-ASR 2.0). It is beyond the scope of this paper to describe this model, which accounts for the above mentioned problems a-c.

It follows from from Fig. 7 and 8 that, without adding oxygen and NaOH, iron problems (Fe > 0.2 mg/L) disappear after 8 cycles, and that the first few m^3 of water recovered always need to be disposed of.

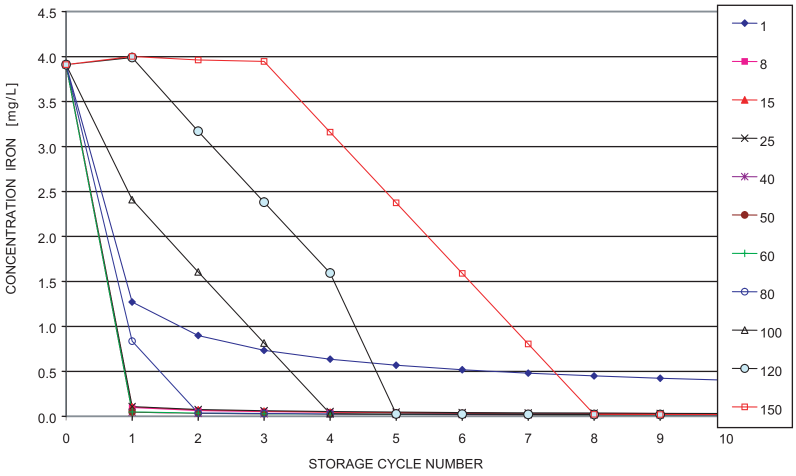


Figure 7. The with EL-ASR 2.0 calculated Fe-concentration at 1-150 m distance from the ASR well in aquifer zone A at the end of each storage period of ASR-cycles 1-10. Each cycle consists of 150 days of injection, 60 days of storage, 120 days of recovery and 35 days of stand-still.

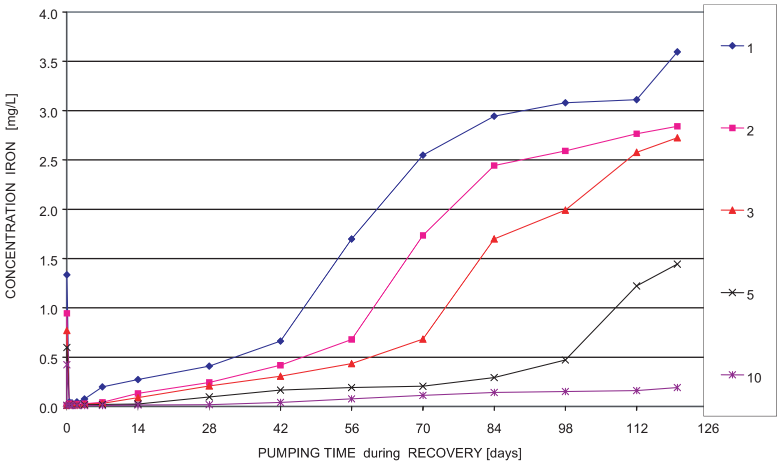


Figure 8. The with EL-ASR 2.0 calculated Fe-concentration in the water recovered by the ASR well during cycles 1-3, 5 and 10. Same cycle program as in Fig. 7.

CONCLUSIONS

ASR on the Herten site proved to be hydrologically feasible, because the water could be injected and recovered with a sufficiently high rate without clogging problems, and there was no evidence of a cumbersome mixing with native groundwater (using Cl as a tracer).

The relatively small quality changes of Herten drinking water in the aquifer studied, pose a problem especially regarding the increases for iron and manganese (drinking water guidelines resp. 0.2 and 0.05 mg/L). The reason is that these changes may require a simple post-treatment. However, the mentioned deviations from the drinking water standards are small, and can be easily cured by (a) dilution with conventionally treated groundwater, or (b) a short aeration followed by rapid sand filtration.

On the other hand, the rather small quality problems can be easily prevented by 2 measures: (i) the addition of oxygen and NaOH during the initial injection periods; and (ii) periodical dosage of a desinfectant like O_3 and/or H_2O_2 and backpumping of it after juttering.

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