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SUMMARY

Aquifer storage and recovery (ASR) is a water resource management method to balance water supply with demand. During periods of excess water, water is injected and stored in an aquifer for subsequent recovery and use in times of water demand. Wet months and strong rain events constitute the most plausible examples of excess water periods. Periods of water demand usually appear during dry seasons or due to higher total demand such as during tourism season. Storage may, therefore, have a broad duration range depending on whether it aims at covering emergency, seasonal, or long-term demand. Sources of water suitable for injection and use within previous ASR applications include treated drinking water, treated or untreated groundwater, rainwater, high quality reclaimed wastewater, and desalinated seawater.

ASR normally offers financial benefits and water quality improvements. The main financial advantages compared to surface water storage consist of (1) an increased security of water supply and reduced well maintenance, (2) little land occupation, (3) reductions in costs of water storage and evaporation losses, and (4) during water purification, storing the treated water in the subsurface can decrease the peak factor allowing the facility to have a smaller capacity. As opposed to single purpose groundwater abstraction wells, ASR wells offer a decreased well clogging risk due to flow reversals which prevent or delay the accumulation of clogging material along the borehole wall. With respect to water quality improvements, processes such as filtration, sorption, and biodegradation are responsible for the water “recycling” during its passage through the aquifer.

On the other hand, disadvantageous aspects of ASR include water losses due to migration of the stored water through the aquifer and adverse water quality changes that need additional treatment. Interaction between injected water and native groundwater and sediments can result in quality deterioration of the injected source water. Upon injection of the source water, mixing with the native groundwater, mineral dissolution and precipitation processes, cation exchange, and redox reactions may all contribute to the alteration of the injected water quality. Additional complications arise during ASR applications in brackish/saline aquifers due to buoyancy effects between the source water and the heavier native groundwater. These effects result in an earlier chloride breakthrough deteriorating the recovered water quality and reducing the theoretically recoverable fraction due to mixing only.

ASR has been used worldwide for several decades and in various different circumstances. In the Netherlands, ASR has not found wide acceptance yet, mainly due to four, partly outdated, reasons: (1) the fear of well clogging, (2) fear of adverse quality changes due to the relatively reactive subsurface of the Netherlands, (3) lack of confidence in achieving sufficiently high recovery ratios, and (4) lack of motivation due to continuous access to sufficient water. The first three problems can be tackled up to a certain level using various techniques, some of them presented in this thesis. Additionally, the need for underground storage of high quality water has greatly increased due to the increased frequency of flood and drought periods, the expected increase in water-related problems due to climate change and the increasing pressure on both surface area and economic goals.

This thesis is built upon water quality issues identified during a drinking ASR pilot project in the Netherlands. The aquifer is considered of a representative nature for ASR standards. Therefore, studying the issues faced and proposing solutions is considered internationally

relevant for many ASR candidate applications. The hydrogeochemical processes responsible for the deterioration of water quality are identified and quantified in Chapter 2. The injection of oxic source water for ASR purposes triggers oxidation reactions with the most reactive aquifer constituents, namely pyrite, sedimentary organic material, and exchangeable Fe(II), NH_4^+ , and Mn(II). The induced acidity is buffered by conversion of abundant HCO_3^- into CO_2 and by slow dissolution of carbonate minerals such as calcite, siderite, and ankerite. As a consequence, heavy metals such as Fe(II) and Mn(II) are released in the groundwater. During recovery, due to inadequately high pH conditions, adsorptive removal to neoformed ferrihydrite is not efficient and the released heavy metals reach the ASR well. Trace elements (As, Co, Ni) released during pyrite oxidation are also observed in the groundwater at various distances from the ASR well. The water-sediment reactions and the leaching rate of reactive solid phases are quantified following a mass balance approach. The poor, from a drinking perspective, quality of the abstracted water, in combination with expected post-treatment costs, convinced the water supply company in Limburg (WML - The Netherlands) to abandon the idea of ASR application in this type of aquifer.

Chapter 3 describes the reactive transport simulation of one-dimensional flow lines that extend from the ASR well. The Herten aquifer is discretized into 4 geochemically homogeneous sublayers and the spatio-temporal evolution of water quality along the flow lines are successfully simulated by the model. The calibrated and validated model is further used to test pre-treatment techniques of the source water prior to injection. Based on the simulated results, oxygen enrichment of the source water combined with pH-buffering agents (e.g. NaOH) can control the mobilization of heavy metals and increase the recovery efficiency of the ASR plant.

In chapter 4, the reactive transport model is further used to test representative source water types (desalinated seawater, urban storm water) that have been used for ASR purposes in other parts of the world. These scenarios are tested in conjunction with the ASR cycles required to build a buffer zone that allows complete recovery of the stored water body without exceedances of drinking water standards. In order to achieve this, the model is coupled to a Python script that automatically stops recovery and starts the next injection phase when certain specified concentration thresholds are exceeded. The implementation of the buffer zone can be accelerated by enriching the source water with O_2 and/or NaOH in order to suppress the Fe(II), Mn(II) and As problems related to the tested source water types.

Finally, chapter 5 describes the implementation of an experimental column setup, designed to closely simulate ASR cycles in an anoxic environment. The installation is used to test aquifer pretreatment prior to ASR application in order to neutralize the various reductants responsible for the deterioration of the abstracted water quality. Pre-treating the aquifer with a potassium permanganate solution suppresses significantly the pyrite reactivity and generates abundant Mn-oxide precipitates with high sorption capacity. The treatment helps controlling the Mn(II) mobilization and allows for an overwhelmingly improved recovery efficiency.