

# Environmental impacts of aquifer thermal energy storage investigated by field and laboratory experiments

Matthijs Bonte, Boris M. Van Breukelen and Pieter J. Stuyfzand

## ABSTRACT

Aquifer thermal energy storage (ATES) uses groundwater to store energy for heating or cooling purposes in the built environment. This paper presents field and laboratory results aiming to elucidate the effects that ATES operation may have on chemical groundwater quality. Field data from an ATES site in the south of the Netherlands show that ATES results in chemical quality perturbations due to homogenisation of the initially present vertical water quality gradient. We tested this hypothesis by numerical modelling of groundwater flow and coupled  $\text{SO}_4$  transport during extraction and injection of groundwater by the ATES system. The modelling results confirm that extracting groundwater from an aquifer with a natural quality stratification, mixing this water in the ATES system, and subsequent injection in the second ATES well can adequately describe the observation data. This mixing effect masks any potential temperature effects in typical low temperature ATES systems ( $<25^\circ\text{C}$ ) which was the reason to complement the field investigations with laboratory experiments focusing on temperature effects. The laboratory experiments indicated that temperature effects until  $25^\circ\text{C}$  are limited; most interestingly was an increase in arsenic concentration. At  $60^\circ\text{C}$ , carbonate precipitation, mobilisation of dissolved oxygen concentration, K and Li, and desorption of trace metals like As can occur.

**Key words** | aquifer thermal energy storage, environmental impacts, ground source heat pumps, groundwater quality

**Matthijs Bonte** (corresponding author)

**Pieter J. Stuyfzand**

KWR Watercycle Research Institute,  
Groningehaven 7,  
Postbus 1072,  
3430 BB Nieuwegein,  
The Netherlands  
E-mail: [matthijs.bonte@kwrwater.nl](mailto:matthijs.bonte@kwrwater.nl)

**Boris M. Van Breukelen**

**Pieter J. Stuyfzand**

Critical Zone Hydrology Group,  
Department of Earth Sciences,  
VU University Amsterdam,  
De Boelelaan 1085,  
1081 HV,  
Amsterdam,  
The Netherlands

## INTRODUCTION

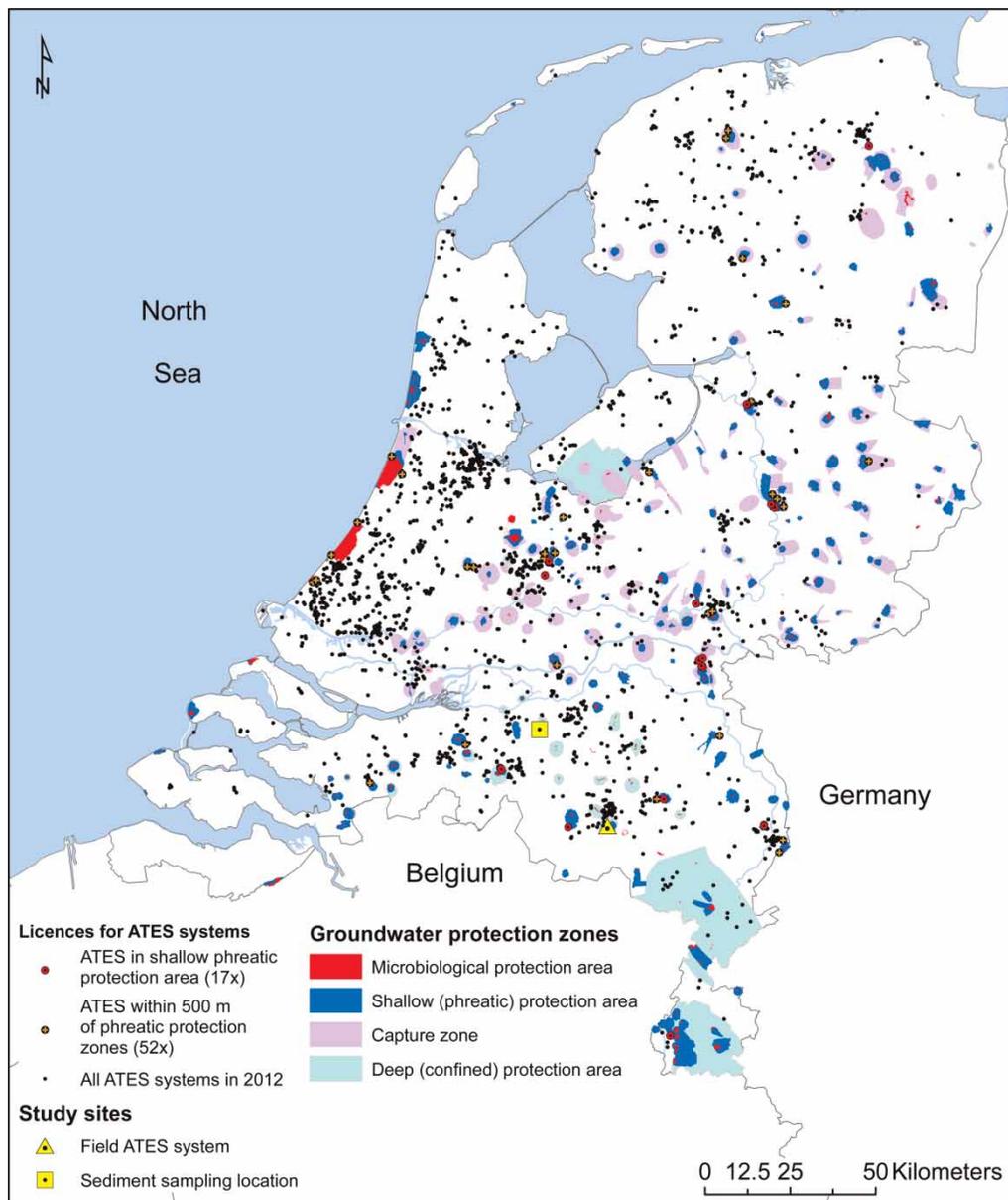
Aquifer thermal energy storage (ATES) uses groundwater for heating and/or cooling purposes in buildings, horticulture and road de-icing. ATES systems operate very similar to the better known closed ground source heat pumps (GSHP). ATES systems generally aim at storing heat derived from buildings in an aquifer and recover this in the subsequent season. In contrast, GSHP systems generally rely on the temperature around the borehole being ambient again in the next season, the same borehole is thus used in both winter and summer. The recovery of aquifer temperature relies on a proper design where the thermal capacity of the borehole is based on thermal parameters of the aquifer and the groundwater flow velocity in the aquifer. ATES systems, however, aim to recover the stored thermal energy of the previous season, which

assumes that there is little groundwater flow. To achieve this, ATES systems generally require two (or more) separate groundwater wells, one cold and one warm well. Small systems set in aquifers with sufficient vertical anisotropy can also use one well with two production filters. In winter, groundwater is extracted from the warm well and used to heat the building (in combination with a heat pump), the temperature of the extracted groundwater is reduced and the cooled water is injected in the cold well. In summer, groundwater is extracted from the cold well and used to cool buildings, the temperature of groundwater is increased and injected in the warm well. The use of separate wells with a lower and higher temperature compared to the natural groundwater temperature, can result in higher efficiency of the ATES

system compared to a GSHP system and lower energy use for the delivered thermal power. In the Netherlands, most ATEs systems operate at a relatively low temperature of several degrees Celsius above and below the natural groundwater temperature ( $11 \pm 1^\circ\text{C}$ ). The maximum allowable injection temperature is set in provincial regulations and ranges between  $25$  and  $30^\circ\text{C}$  (Bonte *et al.* 2008). Interest in high temperature ATEs, between  $40$

and  $80^\circ\text{C}$  is growing and pressure is increasing to allow this as well.

Over the last decade, the number of ATEs systems in the Netherlands has grown exponentially: from around 200 in 2000 (Segers *et al.* 2006) to more than 2,000 in 2012 (Figure 1). This exponential growth has led to an increasing number of sites where ATEs systems are planned or built near public supply well fields, e.g. in 2012, 17 ATEs systems



**Figure 1** | Map of the Netherlands showing all licensed ATEs systems (2012, based on provincial licensing data), groundwater protection areas and well capture zones for drinking water wells.

are located in a groundwater protection area and an additional 52 systems are located within 500 m distance from a protection zone. This often leads to questions by regulators regarding the risks of ATEs in aquifers used for public water supply, in the Netherlands and abroad (Bonte *et al.* 2011a).

Most published research on this subject focuses on operational aspects, such as scaling due to mineral precipitation occurring at high temperature systems ( $>60^{\circ}\text{C}$ ), and is based on either laboratory experiments (Griffioen & Appelo 1993) or chemical equilibrium modelling (Palmer & Cherry 1984). The results of these studies highlight, for example, that depending on the kind of aquifer geochemistry, precipitation of iron-carbonates can be a problem for ATEs. Arning *et al.* (2006) used batch experiments and PHREEQC modelling at temperatures of  $-24$ ,  $2$ ,  $8$ ,  $30$  and  $50^{\circ}\text{C}$ , and showed the dissolution of silicate minerals, primarily feldspar and the probable precipitation of calcite. A study relevant for drinking water production was performed by Brons *et al.* (1991) who looked at mobilisation of organic carbon from sediments under increased temperature. Incubation experiments showed that at temperatures above  $45^{\circ}\text{C}$  organic carbon was mobilised from sediment resulting in an increased chemical oxygen demand. Microbiological research pertaining to ATEs provided no evidence for growth of pathogens (Winters 1992) or increasing cell counts (Schippers & Reichling 2006), yet a considerable change in the microbiological community composition may occur (Sowers *et al.* 2006; Brielmann *et al.* 2009, 2011).

This paper describes the outcomes of research on ATEs systems in the Netherlands, which builds on the above mentioned studies and focuses on the risks that ATEs may impose on groundwater quality and drinking water production. We have used field data from an ATEs site located in Eindhoven to investigate these effects on groundwater quality in a real system and simulated the observed  $\text{SO}_4$  changes in the ATEs well. This ATEs system is located near a public water supply well field. Because the field data set provides limited information on the direct temperature effects, laboratory experiments were carried out focusing on the impact of changing temperature ( $5$ ,  $11$ ,  $25$  and  $60^{\circ}\text{C}$ ) on sediment-groundwater interactions. These experiments were carried out with sediment collected in

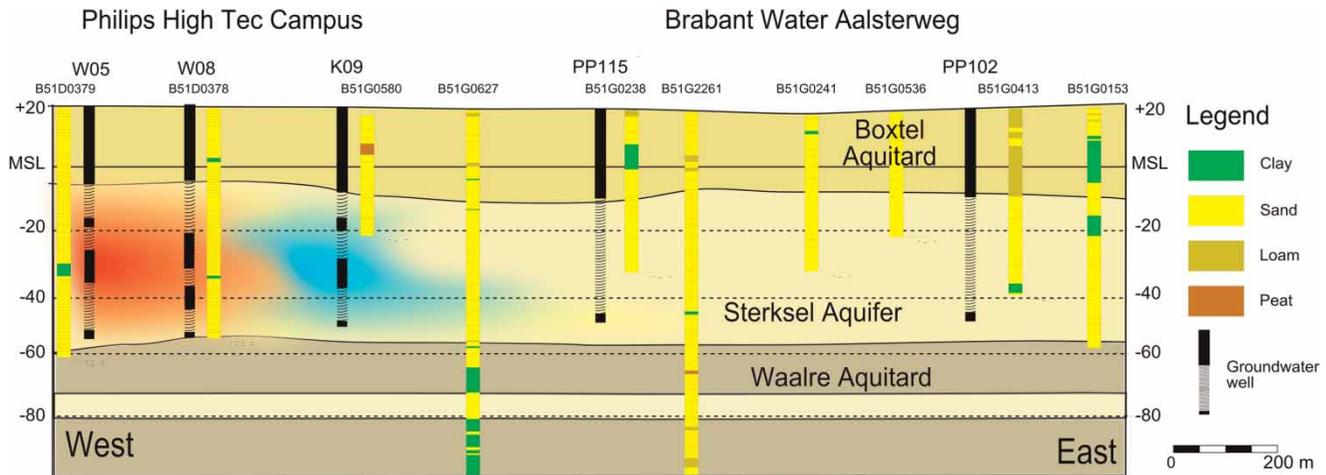
Helvoirt (from the same aquifer as used at the ATEs system in Eindhoven).

## METHODS

### Field site

The ATEs system under investigation started operation in 2005 and is located in the south of the city of Eindhoven (the Netherlands). The system is located at about 650 m to the west of a public supply well field (PSWF Aalsterweg, capacity of  $5 \times 10^6 \text{ m}^3 \text{ year}^{-1}$ ) extracting groundwater from the same aquifer as the ATEs system. The ATEs system is partly located within the groundwater protection zone of PSWF Aalsterweg. In this area, activities that may compromise water quality are prohibited, including the establishment of an ATEs system. Here however, authorities allowed the system provided that it would serve as a research site to investigate the effects of ATEs on groundwater quality. More details on the site in Eindhoven are reported in Bonte *et al.* (2011b).

A hydrogeological cross section with both the ATEs wells and the pumping station is presented in Figure 2. The ATEs system is located in sections of the Sterksel aquifer consisting of coarse sand between  $-5$  and  $-55$  m MSL (metres relative to mean sea level). The groundwater in this aquifer is of high quality and very suitable for production of drinking water. However, at the top of the Sterksel aquifer ( $5$ – $25$  m below sea level, BSL), elevated concentrations of  $\text{Cl}$ ,  $\text{SO}_4$  and  $\text{NO}_3$  point to influences from human activities (road salt, atmospheric deposition of sulphur compounds, and N-inputs from fertilisation which, in combination with pyrite oxidation, results in elevated  $\text{SO}_4$  concentrations, respectively). Below this zone of human-influenced groundwater, low  $\text{SO}_4$  concentrations prevail and the groundwater contains  $\text{CH}_4$ . The resulting pattern in groundwater water quality, i.e. the removal of  $\text{NO}_3$  followed by  $\text{SO}_4$  removal and then appearance of  $\text{CH}_4$  is often observed in groundwater systems, testifying to the preferential use of oxidators present in infiltrating rainwater (Appelo & Postma 2005). The ATEs systems consists of 10 'cold' wells (injecting cold water) and 10 'warm' wells concentrated in four well clusters. During an average climatic year, the designed groundwater circulation of



**Figure 2** | Hydrogeological cross section of the ATEs study site in Eindhoven. Aalsterweg is a PSWF of water supply company Brabant Water. PP = pumping well of PSWF; W05, W08 and K09 are part of the ATEs system and simulated in the Modflow-MT3DMS model.

$2.6 \times 10^6 \text{ m}^3$  generates 23,100 MWh of thermal power. The target average injection temperatures are 7 and 16 °C in the cold and hot wells, respectively. From temperature logs taken at monitoring wells not influenced by the ATEs system the natural groundwater temperature in the Sterksel aquifer was determined to range between 10.5 and 12 °C.

In order to assess the effects of the ATEs system on groundwater quality, groundwater samples from ATEs production wells and monitoring wells not influenced by the ATEs system were collected and analysed on a broad range of inorganic, organic, and microbiological parameters.

### Numerical modelling of the field data

The groundwater flow conditions and resulting sulphate concentrations observed in a selection of the ATEs wells were simulated using Modflow (Harbaugh et al. 2000) for groundwater flow coupled with MT3DMS (Zheng 1990) for solute transport. Sulphate was chosen here as a key parameter showing the largest deviations from observed ambient concentrations (further detailed in Results and discussion section).

This modelling framework considers solute transport by advection (based on the flow field calculated by Modflow) and dispersion. The ambient groundwater in the screened aquifer is characterised by a varying chemical composition. A typical redox zonation characterised by the subsequent removal of  $\text{O}_2$ - $\text{NO}_3$ - $\text{SO}_4$  followed by the appearance of

$\text{CH}_4$  is observed (this is described in more detail in Bonte et al. (2011b)). These different water qualities become extracted by the well that fully screens the aquifer, and it is assumed that within the ATEs well, pipework, heat exchanger and pump, the water from different depths is completely mixed and the water quality in the injection well equals the volume-averaged water quality along the extraction well. The coupling between the extraction and injection well and mixing is done by assigning a negative concentration (IC) to the MT3DMS sinks and source packages, according to Zheng (2010):

$$\text{IC} = -\text{NCOL} \times \text{NROW} \times (K - 1) + \text{NCOL} \times (I - 1) + J \quad (1)$$

where NCOL and NROW are the total numbers of columns and rows in the model, respectively, and  $K$ ,  $I$ ,  $J$  are the layer, row, and column indexes, respectively, of the model cell in which the extraction cell is located whose concentration is to be used as the input concentration of the recirculation well. The method allows for using one grid cell as the extraction well coupled with the injection well. In the model the extraction well is set in all layers in the Sterksel aquifer by assigning the extraction cell in the top cell in the Sterksel aquifer and giving underlying cells in the aquifer a very high conductivity ( $10^6 \text{ m d}^{-1}$ ).

The observed  $\text{SO}_4$  variation with depth is used to describe the initial conditions for each layer. Reactive transport was not included in the model (although this is possible in

MT3DMS), because the processes included already provided sufficient detail to describe the main trends in observed sulphate concentrations. This does not mean that reactive processes can be completely ruled out, however anaerobic oxidation of sedimentary organic matter (SOM) by sulphate reduction is a very slow process, and considered to be insignificant within the operation time frame of the ATEs system. This is confirmed by the  $\text{SO}_4/\text{Cl}$  ratio of water sampled from the ATEs system which shows that ATEs water plots on a mixing line between shallow groundwater with high and deep groundwater with low concentrations of  $\text{SO}_4$  and  $\text{Cl}$  (Bonte et al. 2011b). In order to quantitatively check this assumption, the rate for  $\text{SO}_4$  reduction can be estimated using the frequently used Monod kinetics, assuming there is sufficient degradable SOM present (Appelo & Postma 2005):

$$r = r_{\max} \frac{m_{\text{SO}_4}}{k_{\text{SO}_4} + m_{\text{SO}_4}} \quad (2)$$

where  $r_{\max}$  is the maximum rate constant,  $k_{\text{SO}_4}$  is the half saturation constant and  $m_{\text{SO}_4}$  is the aqueous sulphate concentration. Typical values of  $r_{\max} = 9.6 \times 10^{-9} \text{ mg l}^{-1} \text{ s}^{-1}$  and  $k_{\text{SO}_4} = 9.6 \text{ mg l}^{-1}$  based on Parkhurst & Appelo (1999) yield a  $r_{\text{SOM}}$  of  $0.15 \text{ mg SO}_4 \text{ l}^{-1} \text{ year}^{-1}$  for a concentration of  $m_{\text{SO}_4} = 10 \text{ mg l}^{-1}$ , and  $0.25 \text{ mg l}^{-1}$  for  $m_{\text{SO}_4} = 40 \text{ mg l}^{-1}$ . Given the entire simulation period of 3 years, only a small fraction of  $\text{SO}_4$  ( $0.75 \text{ mg l}^{-1}$  for groundwater containing  $40 \text{ mg l}^{-1}$ ) would be removed.

The model's hydrogeological setup is based on the geological cross section presented in Figure 2. The Aalsterweg public supply well field comprises of 40 extraction wells located 600 m east from the ATEs system. The wells are aligned in two north-south trending well lines of roughly 1 km length. The effect of this well field on the ATEs system is simulated by a constant head boundary at the east and west boundaries of model domain which sets the hydrological gradient imposed by the well field. Table 1 provides further details on the model discretisation, boundary and initial conditions.

### Column experiments

The column experiments were carried out with two sediment samples (called sediments A and B) collected using Ackerman core sampling. In order to keep sediments anaerobic,  $\text{N}_2$  gas

was bubbled through the drilling water and the Ackerman cores were transported to the laboratory in a chilled cooler filled with  $\text{N}_2$  gas. The sediment cores were taken from two depths in the fluvial Sterksel formation, in a coarse and fine part of the formation. Sediment of cores collected at the same depth were unwrapped in a glovebox under a  $\text{N}_2$  atmosphere, mixed thoroughly to obtain a homogeneous sample and repacked in four Ackerman core samplers with 1 cm increments. After addition of each layer, the sediment was compacted to avoid the formation of preferential flow paths (Rinck-Pfeiffer et al. 2000). The columns were stored at  $4^\circ\text{C}$  to avoid microbiological changes until they were used (Castro et al. 2002). Five subsamples were taken from the bulk sample and analysed for moisture content, organic matter and carbonate content using thermogravimetric analysis (TGA), grain size distribution and clay content using laser particle sizing, total element content using X-ray fluorescence (XRF) and leachable element content by inductively coupled plasma mass spectrometry (ICP-MS) analysis of an aqua regia digest.

The cores were placed in the experimental set-up (Figure 3). The four cores were maintained at temperatures of 5, 11, 25 and  $60^\circ\text{C}$  and flushed with groundwater sampled from a well screen installed at the depth from which the sediment cores were collected. A mixture of  $\text{N}_2/\text{CO}_2$  gas (99%/1%) was bubbled through the holding tank to keep the water anaerobic and at constant pH during the experiment. Dissolved oxygen (DO) was continuously logged in both the holding tank and in-line in the effluent of core 2. DO remained below detection limits during the experiments (holding tank  $< 0.05 \text{ mg l}^{-1}$ , effluent core 2  $< 0.01 \text{ mg l}^{-1}$ ). The sediments were flushed with a flux of  $0.26 \text{ ml min}^{-1}$  resulting in a residence time of around 1 d. Influent and effluent samples were taken in containers pre-filled with  $\text{N}_2$  gas and analysed for major, trace elements and dissolved organic carbon.

## RESULTS

### Water quality field data

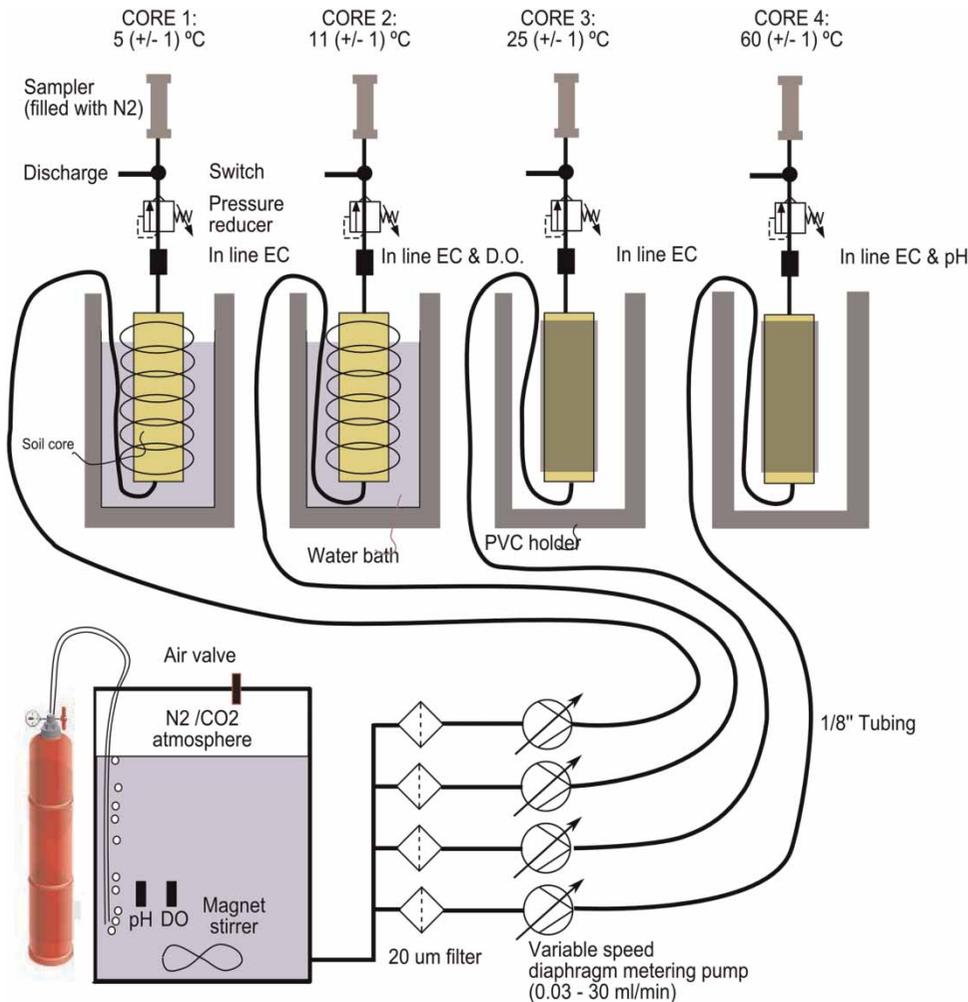
Data for a selection of elements and the effluents' temperature collected from the ATEs field site in Eindhoven are presented in Figure 4. The blue shadings in the figure depict the observed concentration range in ambient groundwater (measured in

**Table 1** | Summary of discretisation, model parameters, and boundary and initial conditions used for Modflow/MT3DMS modelling

| Model aspect   | Model value   |
|--|---|
| <i>Model discretisation</i>  |   |
| Model domain   | 1 × 1 km <sup>2</sup>   |
| Horizontal discretisation  | 40 × 40 m <sup>2</sup> at the boundaries 5 × 5 m <sup>2</sup> at the centre of the model near the ATEs system   |
| Vertical discretisation  | 16 layers, representing Boxel semi-confining layer (4 layers) and Sterksel aquifer (12 layers)  |
| <i>Flow parameters (Modflow)</i>   |   |
| Horizontal conductivity ( $K_h$ ) Based on values of the Dutch geological database (REGIS) accessible via <a href="http://www.dinoloket.nl">www.dinoloket.nl</a> | Upper Boxel (+20 to 0 m-mean sea level, MSL): 20 m d <sup>-1</sup> (medium fine sand) Lower Boxel (0 to -5 m MSL): 0.05 m d <sup>-1</sup> (sandy clay) Sterksel (-5 to -55 m MSL): 30 m d <sup>-1</sup> (medium coarse sand)  |
| Vertical conductivity ( $K_v$ ) Also from REGIS  | Upper Boxel: 10 m d <sup>-1</sup> Lower Boxel: 0.01 m d <sup>-1</sup> Sterksel: 15 m d <sup>-1</sup>  |
| Porosity ( $n$ )   | 0.35  |
| Specific storativity ( $S_s$ )   | 10 <sup>-4</sup> m <sup>-1</sup>  |
| Boundary conditions  | Ambient flow field with hydraulic gradient towards the Brabant Water Aalsterweg PSWF (PP115 and PP102 of this pumping station are presented in Figure 2): simulated by constant head boundaries at east and west boundaries of the model simulating a ambient flow velocity of 55 m per year  |
| ATEs system  | Simulated with injection and extraction wells (based on average monthly extraction rates of 25 m <sup>3</sup> h <sup>-1</sup> )   |
| Initial conditions   | Bases on steady state results   |
| Time discretisation  | Total simulation time: 3 years. Each year comprises: 100 d extraction from warm well, injection in cold well during winter 80 d storage period 100 d extraction from cold well, injection in warm well during winter 80 d storage period  |
| <i>Transport parameters (MT3DMS)</i>   |   |
| Longitudinal dispersivity  | 2.5 m   |
| Horizontal and vertical transverse dispersivity  | 0.25 m  |
| Diffusion  | Ignored   |
| Advection  | Third-order TVD (total-variation-diminishing) scheme (time discretisation based on a Courant number of 0.75)  |
| Boundary conditions  | Solute sinks at extraction wells Solute sources at injection wells, based on average extraction sulphate concentration calculated by MT3DMS by use of Equation (1).<br><br>Fixed concentration at all outer model boundaries (based on observed SO <sub>4</sub> gradient): Boxel aquifer: 40 mg SO <sub>4</sub> l <sup>-1</sup> Upper part of Sterksel aquifer (5–25 m BSL): a linear gradient from 40 to 5 mg SO <sub>4</sub> l <sup>-1</sup> Lower part of Sterksel aquifer (25–55 m BSL): 5 mg SO <sub>4</sub> l <sup>-1</sup> |
| Initial concentration  | Same depth distribution as boundary conditions  |

reference monitoring wells) at the depth of the monitoring screen (the full colour version of this figure is available online at <http://www.iwaponline.com/jwc/toc.htm>). The minimum and maximum temperatures observed at the site

are 7 and 22 °C, respectively. The quality data and temperature data show a cyclic character for many elements. This cyclic character is explained by the relatively high groundwater flow velocity in the aquifer at the site: The ATEs system



**Figure 3** | Column testing set-up.

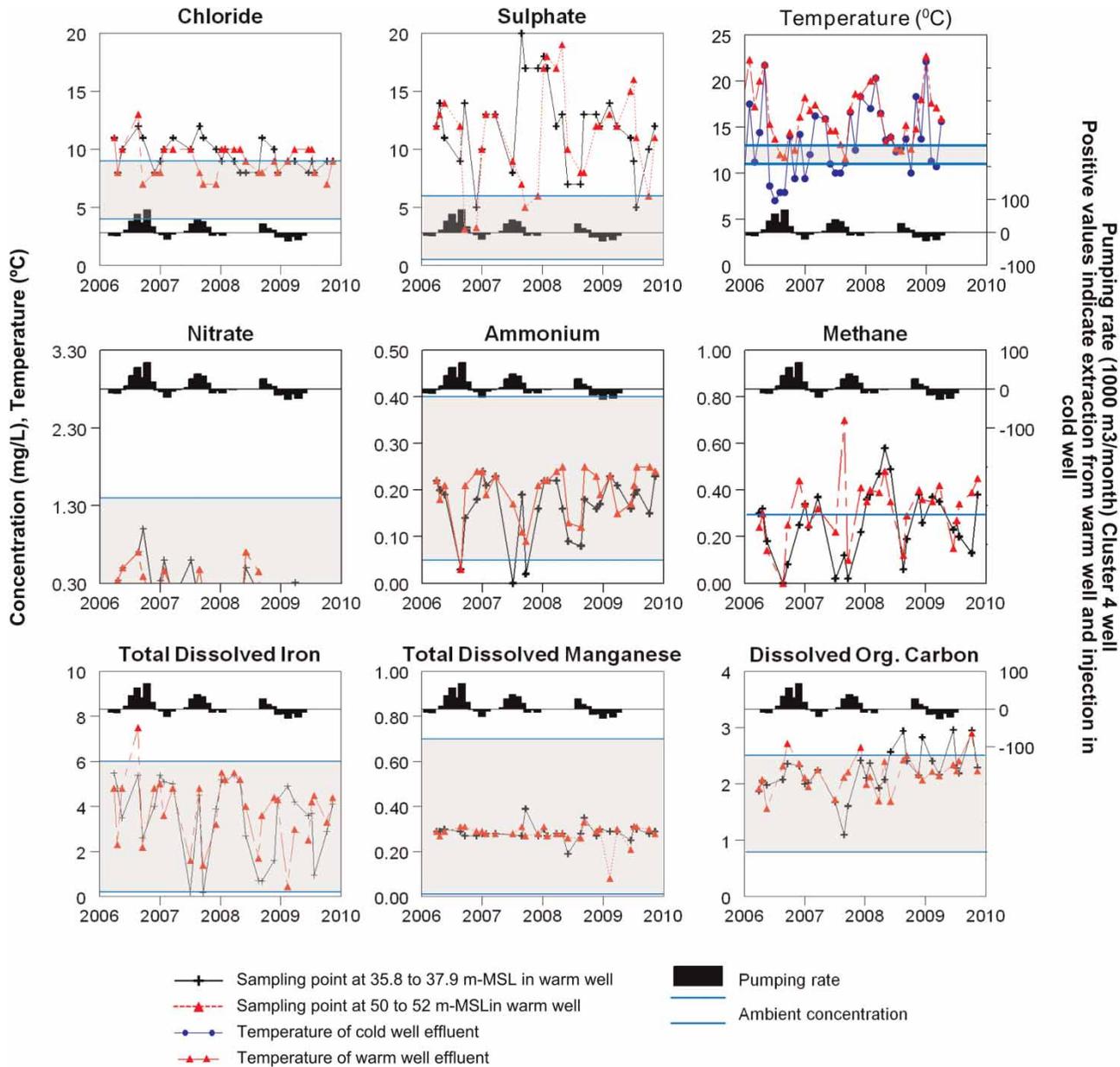
extracts groundwater from the different depths within the aquifer having different qualities, mixes it and injects it back into the aquifer. The injected groundwater mixture has a different quality from the unmixed ambient groundwater: It is a mixture of groundwater originating from the entire depth section of the aquifer which causes the natural vertical concentration gradient to be levelled out.

The ATEs system introduces a fraction of shallow groundwater with a different chemical composition at greater depth, and a fraction of deep groundwater at shallow depth. At the depth of monitoring wells presented in Figure 4, this leads to increased concentrations of  $\text{SO}_4$ , Cl, Na, K and other elements, while concentrations of  $\text{CH}_4$  and Fe decreased. After some time, during the storage phase of ATEs operation, the injected groundwater drifts

away with the natural groundwater flow and groundwater quality returns to ambient conditions.

### Numerical modelling of field data

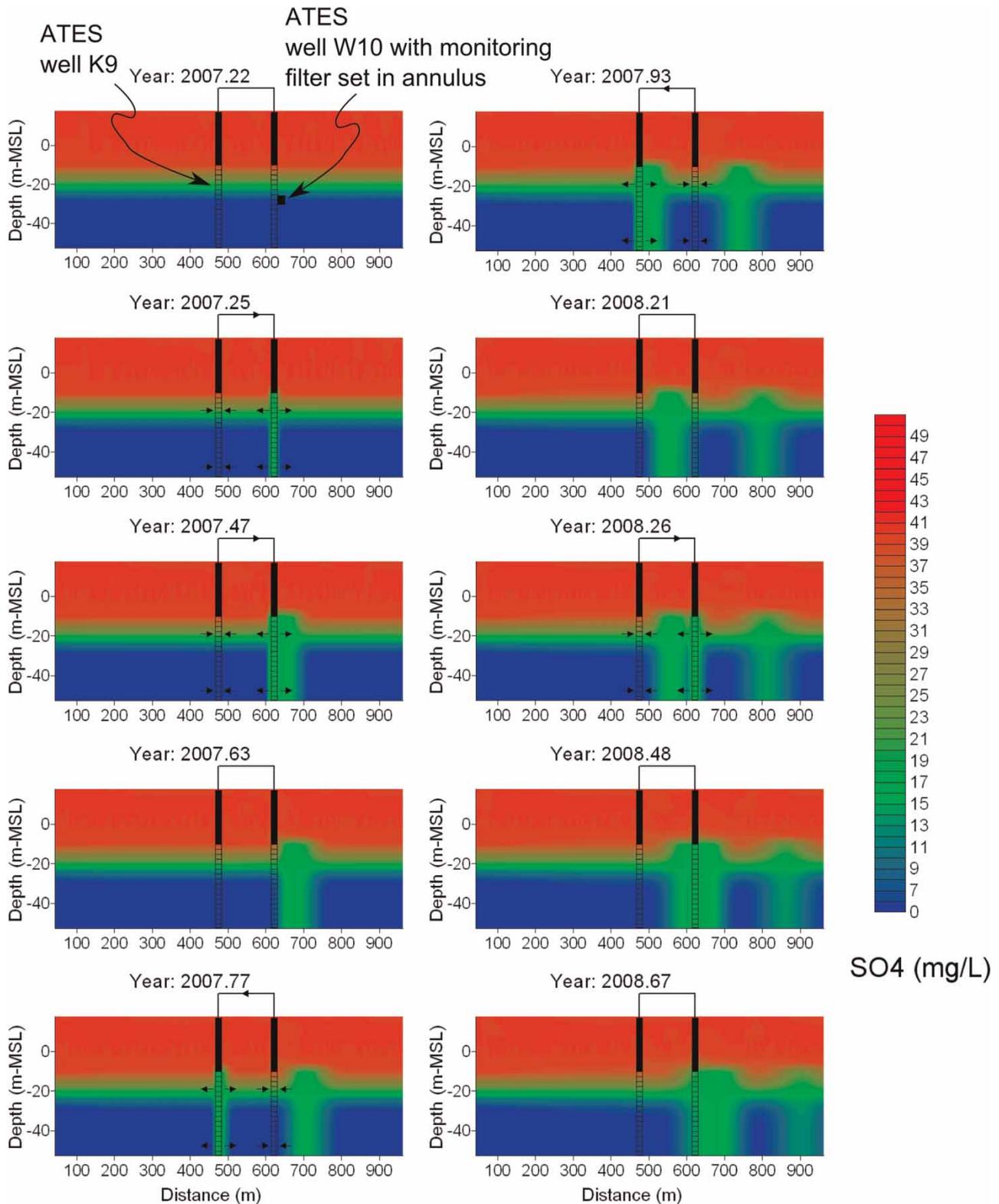
A quantitative analysis of this mixing process is provided by the groundwater flow and solute transport model. Results of the simulations are presented in Figure 5 which contains a number of cross sections over the modelling domain incorporating a cold and warm ATEs well during winter, the storage phase, summer, and another storage phase. Figure 6 presents simulated and observed sulphate concentrations in the observation well screens set in the annulus of one of the ATEs wells (W10 screen between  $-50$  and  $-52$  m MSL).



**Figure 4** | Observed groundwater quality in two monitoring screens in two ATEs production wells with ambient concentrations depicted by blue shading (the full colour version of this figure is available online at <http://www.iwaponline.com/jwc/toc.htm>). Also shown is the extraction rate of the ATEs wells in black bars; a positive pumping rate means an extraction and a negative pumping rate means that groundwater is injected in the wells, and the temperature of the effluent from the warm and cold ATEs well.

The cross sections of the modelling results (Figure 5) clearly show the extraction, mixing, and injection during ATEs operation which disturbs the natural stratified sulphate distribution with high sulphate concentrations in the Boxtel aquifer and upper Sterksel aquifer, and low concentrations in the lower Sterksel aquifer, generating plumes of ‘injected ATEs water’ with an average concentration of both aquifer layers. The close vicinity of the well field

causes a relatively high groundwater flow velocity. This causes the injected plumes of ATEs water to drift away with the ambient groundwater flow towards the PSWF while somewhat fading out due to dispersion. Also visible in the cross sections is that the drifting plume of injected ATEs water, actually flows from one ATEs well to the adjacent coupled ATEs well, thus causing a second jump in the breakthrough of mixed ATEs water. This is highlighted with



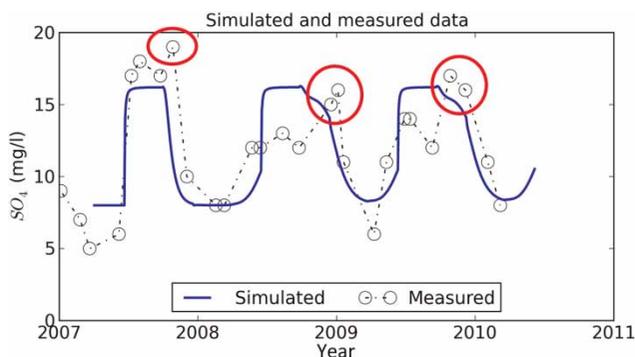
**Figure 5** | Simulated sulphate concentrations (shading) during several periods over the 3 year simulation period. Two ATEs wells (K9 and W10) are shown, top left panel shows location of the monitoring screen set in the annulus of W10. The observed and simulated SO<sub>4</sub> concentrations for this monitoring screen are shown in Figure 6.

the circles in Figure 6, displaying this second breakthrough in the observed data.

The comparison between observed and simulated sulphate concentrations in Figure 6 indicates that the process of extraction, mixing, and injection can adequately describe the observed oscillations in sulphate concentrations. The observed and modelled sulphate concentrations show that the injected, heated and chilled water actually floats away from the ATEs wells before being recovered in the subsequent season. Temperature data in the ATEs warm and cold wells presented in Figure 4 show that the temperature in the ‘warm’ well is increased during summer and decreased during injection in the ‘cold’ well during winter. When extraction starts in the subsequent season, the temperature in the ATEs well has returned to near ambient conditions confirming that the ATEs water has drifted away before it can be recovered. This actually highlights that an ATEs system realised in an area with a high groundwater velocity (which is often the case near a well field) operates like a GSHP, which relies on the groundwater temperature returning to ambient conditions before the next season starts. This is atypical for most areas in the Netherlands where groundwater flow velocities are quite low and indicates that areas near existing well fields are not very suitable for ATEs systems.

### Laboratory experiments

Table 2 shows some characteristics of the two sediment types investigated. The deeper sediment has higher



**Figure 6** | Measured and observed sulphate concentrations in ATEs well W10 with screen between –50 and –52 m MSL over the period 2007–2010. The circles show the second breakthrough of ATEs water from the adjacent ATEs well (see text for further description).

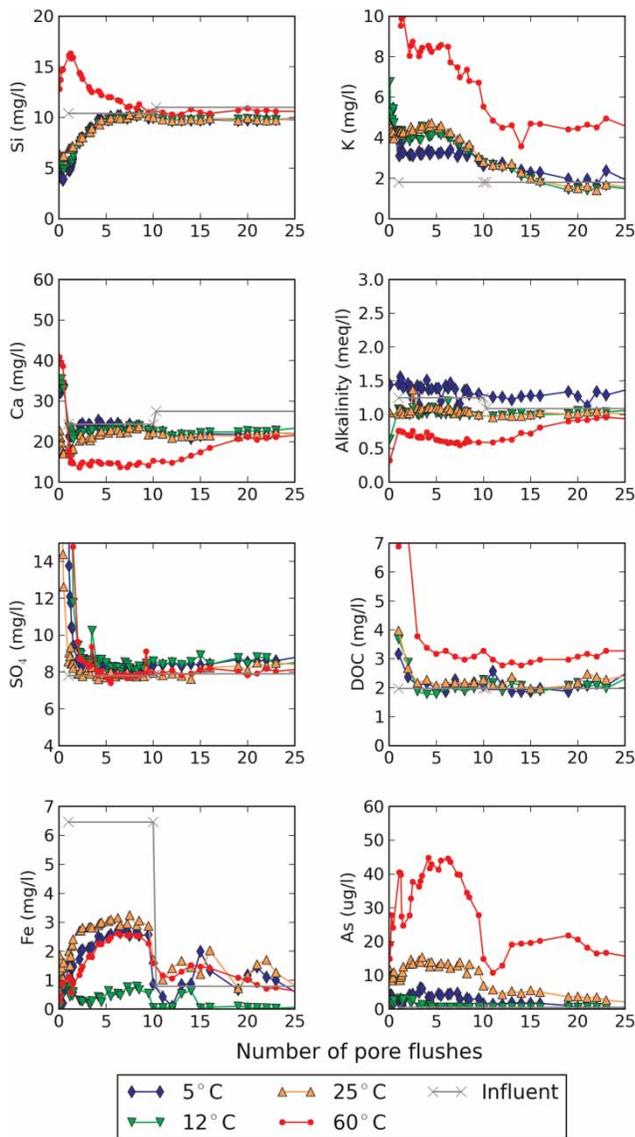
**Table 2** | Sediment characteristics

| Sediment     | Organic carbon (LOI550) (%) | CaCO <sub>3</sub> (%) | Clay% <2 μm | D50 (μm) |
|--------------|-----------------------------|-----------------------|-------------|----------|
| A (42 m MSL) | 0.76                        | 6.01                  | 3.7         | 187      |
| B (33 m MSL) | 0.48                        | 0.53                  | 2.2         | 244      |

carbonate, organic carbon, and clay content and is expected to be more reactive than the coarser shallower sediment.

In Figure 7 a selection of effluent curves from the column experiments with sediment B are shown to illustrate several likely processes. The first two relate to silicate and carbonate mineral phases: silicate minerals become more soluble at higher temperatures, whereas carbonate minerals become less soluble. Si and K in effluent of the 60 °C core during the first five pore flushes (PFs) were almost twice the concentration of effluent obtained from cores 1 to 3 (5–25 °C). The differences may, however, also be explained by de- and adsorption of dissolved Si and K on exchangers and other reactive surfaces, next to the dissolution of K-feldspar (microcline) or K release from mica. The latter process was also held accountable for increased K and Si concentration in batch experiments by Arning *et al.* (2006). Ca and HCO<sub>3</sub> concentrations in effluent of the 60 °C core were relatively low, probably due to precipitation of carbonate minerals such as calcite and dolomite (Mg is not shown but shows a similar pattern as Ca).

The effluent dissolved oxygen concentration (DOC) in both sediments A and B from the two lowest temperature cores was on average equal to the influent concentration, DOC from the 25 °C core was slightly higher (0.1 mg l<sup>-1</sup>) in both sediments, whereas DOC in the effluent from sediment B at 60 °C was increased by 46% from 2.4 to 3.5 mg l<sup>-1</sup>. The DOC increases in effluents of sediment A (having higher organic carbon content; Table 2) is even greater with increasing temperature. These results are consistent with the findings of Brons *et al.* (1991) who showed an increase in DOC concentrations above 45 °C in incubation experiments. The increase of DOC in the 60 °C effluent was accompanied by a slight decrease in SO<sub>4</sub> concentration in sediments A (not shown here) but not in sediment B (Figure 7). This indicates that at higher temperature, the



**Figure 7** | Effluent data of column experiments on sediment B.

mobilised DOC can provide a carbon source for sulphate-reducing bacteria. Up to 25 °C no decrease in  $\text{SO}_4$  was observed indicating that  $\text{SO}_4$  reduction did not occur.

Effluent Fe concentrations show a pattern which is not easily explained: all effluents were depleted in iron relative to the influent. The 11 °C core showed the greatest depletion in Fe and the 25 °C core the least. After seven PFs all cores showed a rapid further depletion. Arsenic was below detection ( $<1 \mu\text{g l}^{-1}$ ) in the influent but was considerably increased above the drinking water limit of  $10 \mu\text{g l}^{-1}$  for both the 25 and 60 °C cores in sediment B. In sediment A,

arsenic was above the drinking water limit only in the 60 °C effluent. In the 11 °C core, arsenic was not significantly above the detection limit, but strangely enough, in the 5 °C core it was.

## DISCUSSION

Overall, the data and modelling of the field site clearly showed that ATEs operation changes the natural redox zoning in the aquifer, which itself may trigger secondary reactions, for example mobilisation of trace metals and organic carbon. This redox mixing combined with the impact of changing temperature was, however, not observed in the ATEs system as the injected water drifted away before it was recovered again. Generally, however, it can be stated that the vulnerability of the public supply well field is increased because shallow water, more influenced by human activity is brought to depth where it can travel through coarser sediment layers and reach the public supply well field more rapidly. In addition to this, the capacity of the aquifer to buffer contaminants (sorption of organic contaminants to organic matter or metals to iron-oxides, or by reducing introduced oxidants) is likely to decrease with depth because of the coarsening of the aquifer downwards. This aspect of heterogeneity (not included in the flow modelling presented here) implies that organic matter and clay content, and related reducing capacity, will reduce with depth.

The results of the laboratory experiments confirm that direct temperature driven impacts of heat storage up to around 25 °C are limited and only at the highest temperature (60 °C) effects resulting from increased temperature are visible. However, a modest increase in arsenic and DOC concentration was also visible at 25 °C. The observed thermal impacts from laboratory experiments showed, in combination with the observed concentration changes at this ATEs site, that the impacts were visible but sufficiently small to keep groundwater suitable for drinking water production. The mixing of shallow human-influenced groundwater with deeper, generally less influenced groundwater, does imply however, that the vulnerability of a public supply well field can be increased by ATEs operation.

## CONCLUSIONS

Field and laboratory data suggest that ATES operation can impact upon groundwater quality in a number of ways. (1) Depending on the construction of the ATES system and local hydrogeology and geochemistry, mixing of groundwater from different depths by ATES can have an important influence on groundwater quality, irrespective of the operation temperature. (2) In ATES systems with operating temperatures up to 25 °C, direct temperature effects are modest but the increased arsenic mobility observed in laboratory experiments requires further attention. This is especially the case in aquifers which under natural conditions have elevated arsenic concentrations. (3) In ATES systems with higher operating temperatures (60 °C), laboratory experiments show that groundwater quality is significantly influenced by the temperature changes with increasing DOC, As, K and Si concentrations. These aspects should be considered when planning or designing an ATES system, especially in aquifers used for water supply.

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