

The Relationship between Source and Recovered Water Quality during Storage in a Sherwood Sandstone Aquifer

M. Nandha¹, B. Holden², B. Jefferson¹, P. Jeffrey¹ and K. Le Corre¹

m.pindoria@cranfield.ac.uk, bHolden@anglianwater.co.uk, b.jefferson@cranfield.ac.uk, p.j.jeffrey@cranfield.ac.uk,
k.s.lecorrepidou@cranfield.ac.uk

¹Cranfield Water Science Institute, Cranfield University, College Road, Cranfield, MK43 0AL

²Anglian Water, Thorpewood House, Thorpe Road, PE3 6WT

Abstract

Understanding the potential changes in water quality during storage in an aquifer is crucial if storage schemes are to be designed appropriately. In this study, column experiments are used to determine the changes in water quality when treated water from the River Trent in the UK is stored in a Sherwood sandstone aquifer. We also examine how variation in recharge water quality is reflected in recovered water quality. Water taken from four different points in the treatment train was introduced into four columns, one for each sampling point, which were sealed and left for 15 days. In all four columns, the abstracted water showed a decrease in nitrate and phosphorus concentrations with a corresponding increase in organic carbon content which implied the occurrence bio-denitrification. The concentration of sulphate in the abstracted water also reduced which was attributed to the reduction of sulphate to bisulphide which in turn promotes the precipitation of metals. As expected, the concentrations of metals and major ions increased in all the columns, except for Mo, Co and As which increased in all but one column where a slight reduction was seen. The increase in concentration was attributed to a combination of precipitation and the reduction of minerals in the formation, and the slight decrease in Mo, Co and As in one column to a combination of lower microbial activity and slight adsorption onto the material. The abstracted water met drinking water standards for all measured parameters except for Mn, the levels of which were above the prescribed concentration in all the columns, due to a significant increase in concentration during storage.

Keywords: Aquifer storage, Water quality; Soil columns; Sandstone aquifer

Introduction

Interactions between water used for aquifer recharge, the aquifer material, and native groundwater can potentially affect the quality of the water ultimately abstracted for reuse and can therefore constitute an operational risk to Aquifer Storage and Recovery (ASR) schemes. This risk is site specific and its magnitude is difficult to quantify without detailed environmental, hydrogeological and geochemical characterisation studies combined with pilot-scale investigations. The water used for recharge typically interacts with both the native groundwater and the aquifer rock through a combination of physical, chemical and biological processes that are also linked to subsurface conditions. For instance, it is subject to alterations in redox states, pH, nutrient supplies and temperature, leading to changes in the quality of the abstracted water (Dillon et al., 2006). During the storage phase, it may also react with the adjacent geology. For example, large differences in the pH or redox conditions between recharged water and native groundwater can result in the mobilisation of trace elements such as iron (Pyne, 2005). The method of recharge also influences the type and magnitude of these changes. Recharge can be via infiltration (Soil Aquifer Treatment

(SAT), river bank infiltration, infiltration basin, dune filtration) and/or injection through a well (includes ASR and Aquifer Storage Transfer and Recovery (ASTR)).

Understanding the envelope of potential changes in water quality during storage in the aquifer is therefore crucial if schemes are to be designed appropriately and risks managed effectively. To date, significant research has been undertaken to understand the risks associated with aquifer recharge schemes using infiltration techniques such as SAT (Drewes et al. 2001; Maeng et al. 2012; Rauch-Williams et al. 2010; Fox and Shah 2006; Rauch et al. 2006) and River Bank Infiltration (Horner et al. 2006), however studies of ASR at a lab scale are scarce. Patterson et al. (2012) studied the fate of N-Nitrosodimethylamine (NDMA) in a sandstone aquifer using column experiments, while Johnson et al. (1999) used columns to study the geochemical reactions that may alter water quality during vadose zone injection into an unconsolidated sand and gravel aquifer. The study further evaluates the influence of different source water qualities (micro-filtered Colorado River water, micro-filtered effluent and reverse osmosis treated effluent) on these reactions. Patterson et al. (2010) used column experiments to measure the changes in concentrations of trace organic and inorganic compounds, when reverse osmosis treated recycled water was recharged into a sandstone aquifer. For the purpose of this research, the release of metals and major ions during storage were identified as a key concern for an ASR scheme in the Sherwood Sandstone aquifer. Patterson et al. (2010) and Johnson et al. (1999) suggested that organic carbon, nitrate, sulphate and ammonia can all influence the release of metals and major ions, therefore the literature was further queried to identify any studies of the potential changes in concentration of these parameters in a Sherwood Sandstone aquifer during storage.

Several field investigations have been conducted to investigate changes in metals and major ion concentration during storage (Vanderzalm et al. 2010; Vanderzalm et al. 2009; Overacre et al. 2006; Dillon et al. 2005a; Dillon et al. 2005b), and there is literature addressing water quality changes during storage in a sandstone aquifer at the lab scale (Patterson et al. 2010; Patterson et al. 2012). However, no literature was found with respect to changes in nutrients and inorganic compounds when treated surface water is recharged into a consolidated Sherwood Sandstone aquifer, and the influence of recharge water quality on the recovered water quality. It is important to note that each sandstone aquifer varies in its mineralogical composition and will influence recovered water quality in a different manner. Johnson et al. (1999) also demonstrated that differing qualities of recharge water will also influence the recovered water quality. This study therefore aims to improve understanding in this area by (i) determining the potential changes in water quality during storage in a Sherwood sandstone aquifer that may impact recovered water quality, and (ii) exploring the influence of different recharge water qualities on recovered water quality.

Methods and Materials

Newton on Trent, Lincolnshire, UK has been identified as a potential ASR site through a staged water resource and hydrogeological study. Water from the River Trent will be treated in Hall water treatment works (WTW) using the treatment train shown in Figure 1 below, following which it will be recharged into the confined Sherwood Sandstone aquifer. Potential changes in water quality during storage in this aquifer that could impact the quality of abstracted water were investigated using soil columns simulating field conditions. For this purpose, a core sample of the Sherwood Sandstone located approximately 11 miles west of this site was crushed and sieved to <2mm then packed into four PVC columns, one for each sampling point, with a diameter of 9 cm and length of 40 cm. Before sealing the columns, stainless steel mesh was placed in the endcaps to support the

aquifer material and to help spread the inflow laterally through the columns. The columns were then flushed with fresh native groundwater for 10 days in an upward flow to remove all free oxygen, allow stabilisation and inoculation of the columns, and allow microorganisms to acclimatise. The columns were then purged with nitrogen gas to create anaerobic conditions representative of natural aquifer conditions before the source water was introduced. In order to assess the impact of differing recharge water sources on abstracted water quality, water was sampled at different points of the treatment process as illustrated in Figure 1.

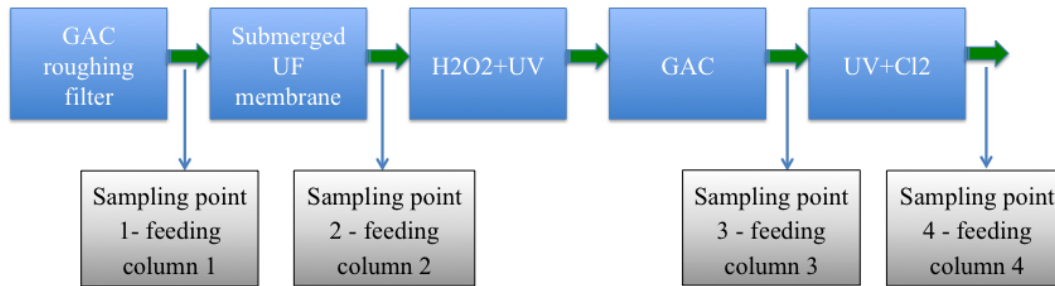


Figure 1. Hall WTW treatment train and sampling points used as source water for the columns

Water from the four sampling points (S1, S2, S3 and S3) were introduced into columns 1, 2, 3 and 4 respectively and left for 15 days to allow any microbiological and geochemical reactions to occur before the water was extracted. There was little information in the literature with regards to residence times in batch column experiments thus a decision was made to use a 15 day retention time. The quality of the abstracted water was then compared to the source water to determine changes that had occurred during storage. The parameters chosen for the evaluation of water quality changes during storage are shown in Table 1.

Table 1. Parameters measured to determine changes in water quality during storage

Parameter	Reason for selection	Analytical methods
Total organic Carbon (TOC)	<ul style="list-style-type: none"> • Indicates the quantity of organic matter: <ul style="list-style-type: none"> ○ Potential impact on redox conditions (biogeochemical reactions) and metal mobilization. ○ Impact on pH ○ Influences on biological growth 	Shimadzu TOC-V Analyser
Nitrate & sulphate	<ul style="list-style-type: none"> • Oxidants – Dissolved organic carbon can be mineralised (and concentrations reduced) by reacting with injected oxygen, nitrate and sulphate 	Spectroquant cell test kit (Merck Millipore) 1.0 – 50.0 mg/L NO ₃ -N 5 – 250 mg/L SO ₄
Ammonium	<ul style="list-style-type: none"> • Indicator of growth of anaerobic bacteria (nutrient) 	Spectroquant cell test kit (Merck Millipore) 0.20 – 8.00 mg/l NH ₄ -N cell test
Metals, Major ions & P	<ul style="list-style-type: none"> • Release of inorganic compounds during 	Digestion – 1.5ml nitric acid added to 30ml sample and digested in a Mars Xpress

		microwave.
	<ul style="list-style-type: none"> storage. Phosphorus is a nutrient for microbes 	Analysis – Inductively coupled plasma mass spectrometry (ICP-MS) for all but Fe and Na which were measured using Atomic absorption spectroscopy (AAS, Perkin Elmer Analyst 800)
pH	<ul style="list-style-type: none"> Influences microbial activity, Influences biogeochemical reactions Influences dissolution 	pH probe (JENWAY 3540 pH and Conductivity Meter)

Results and Discussion

The quality of the recharged water was altered during storage in all the columns to different extents, as can be seen in Table 2 below.

Table 2. Concentration of Nitrate, Sulphate, TOC, metals and major ions in the different source waters before and after 15 days storage in the columns. *Values below limits of detection. ** Not measured

Parameter	unit	Column 1		Column 2		Column 3		Column 4	
		Before	After	Before	After	Before	After	Before	After
NO ₃	mg/L	8.5	<1	8.4	<1	8.5	1.5	8.4	<1
SO ₄	mg/L	130.5	105.5	142	118.5	137.5	105	141.5	98.5
TOC	mg/L	4.3	4.8	4.4	4.8	0.8	1.8	0.6	3.3
pH		7.9	8.1	7.9	8.1	7.6	8.1	7.6	-**
Mg	mg/L	19.9	24.6	21.7	25.6	19.4	26.3	20.2	25.1
Fe	mg/L	0*	0.2	0*	0.2	0.4	0*	0*	0*
Na	mg/L	50.4	32.9	49.0	36.0	49.6	27.4	51.0	20.6
K	mg/L	10.7	13.3	11.6	14.0	10.2	13.7	10.8	13.9
P	µg/L	471.5	129.2	490.4	57.9	421.4	128.1	914.6	152.8
Mn	µg/L	1.5	773.9	2.4	1554	7.1	894.1	0.7	215.3
Ni	µg/L	7.6	18.2	8.4	13.6	5.4	12.9	4.4	13.6
Co	µg/L	0.4	3.0	0.4	8.5	1.1	0.9	0.2	0.4
Zn	µg/L	66.8	82.7	61.0	90.6	32.7	117.6	32.1	70.4
As	µg/L	2.3	2.3	2.2	3.2	2.0	1.6	2.0	2.5
Mo	µg/L	3.5	3.6	3.4	7.7	4.4	4.3	3.0	5.7
Ba	µg/L	66.7	110.8	72.0	91.7	64.3	107.6	68.0	100.4
U	µg/L	0.9	1.3	0.9	1.5	0.6	1.3	0.6	1.4

In terms of nutrients, no changes in ammonium were noticed as the value was below detection limits in all samples, before and after storage while a net decrease in P concentration was observed across the columns with a 73% reduction in column 1, 88% in column 2, 70% in column 3 and 83% in column 4. P is a nutrient used for microbiological growth therefore an overall reduction in

concentration after storage implies active microbiological growth. The main removal mechanism for nitrates is denitrification, which is the conversion of nitrate and nitrite to nitrogen gas, and is an anaerobic process. Most reduction processes are hydroxide or alkaline producing and therefore pH typically increases (Pyne, 2005), as found in this experiment (Table 2). Dominant electron donors for the denitrification process are organic carbon and reduced sulphur compounds (Dillon et al. 2005b). Dahab, 1993 reported reductions in nitrate of 50 to 100% due to bio-denitrification, however this meant that the recovered water quality would likely have high residual organic material. This is supported by the results found here which show an average reduction of 80% in nitrate concentrations, with a corresponding increase in TOC in the recovered water. Bio-denitrification is therefore the most likely mechanism for the decline of nitrate concentrations during storage.

Overall, the increase in Mn concentrations was the most substantial change during storage in the column (Figure 2). On average Mn concentrations increased by three orders of magnitude. A minimum increase from 0.7 to 215.3 $\mu\text{g/L}$ was observed in column 4 which received water that passed through the full treatment train, while column 2 (receiving GAC/UF treated water) released the highest amount of Mn with 1554 $\mu\text{g/L}$ in the recovered water compared to 2.4 $\mu\text{g/L}$ in the source water, that is to say an increase of approximately four orders of magnitude.

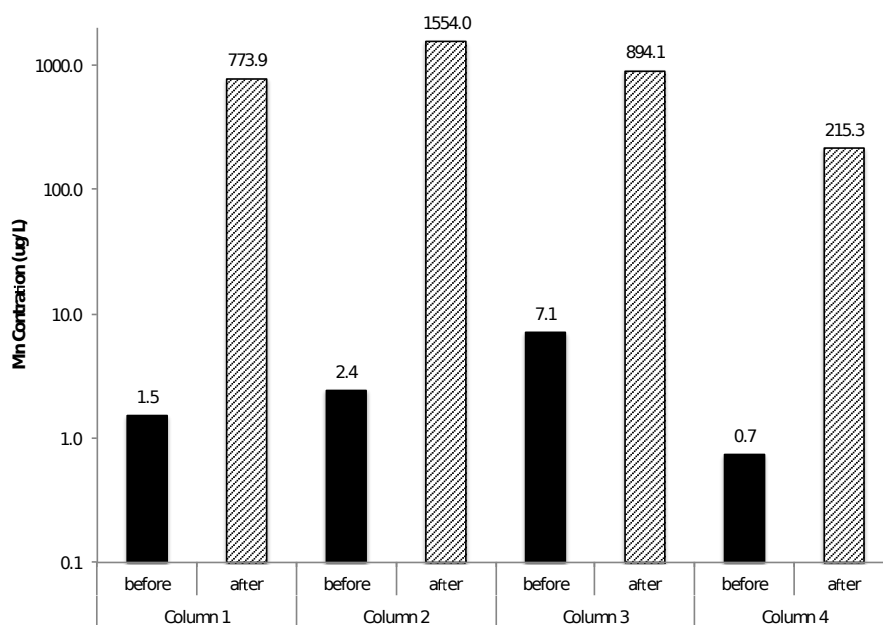


Figure 2: Concentration of Mn in the source (before storage) and abstracted water (after storage)

The concentration of metals, particularly iron and manganese, tend to increase under reducing conditions, which in combination with low bisulphide concentrations can result in high concentrations of iron and manganese in the recovered water (Pyne, 2005). The concentration of sulphate decreased by 19% in column 1, 17% in column 2, 24% in column 3 and 30% in column 4. A decrease in the concentration of sulphate in all columns supports the potential reduction of sulphate to bisulphide, which in turn would promote the precipitation of metals. On its own however such a reaction cannot explain the magnitude of the change observed here. Other mechanisms that could have contributed to this increase in Mn are the reduction and desorption of Mn oxides in the formation (Patterson et al. 2010). These are most likely to be responsible for the

large increase in the concentration in column 2 since the increase does not correspond to a higher reduction of sulphate in the column.

Generally, the concentration of all the metals and major ions increased after storage, which could be explained by the dissolution of metal bearing minerals. However a slight reduction was evident in the concentration of Co and Mo in column 3, from 1.1 µg/L to 0.9 µg/L and 4.4 µg/L to 4.3 µg/L respectively. The concentration of As in the recovered water also varied, with an increase after storage in columns 2 and 4 from 2.2 µg/L to 3.2 µg/L and 2.0 µg/L to 2.5 µg/L respectively, while a slight decrease from 2.0 µg/L to 1.6 µg/L was evident in column 3. The decrease in concentration of Co, Mo and As in column 3 corresponds to a slightly lower removal of nitrate and phosphorus in the same column. This implies a lower microbial activity, since microbial degradation is one of the main removal mechanisms for both nitrates and phosphorus. Lower microbial activity in the column could imply less reduction of minerals which in turn could explain the slight decline in concentration as a result of slight adsorption onto the material without the offsetting increase due to reduction. The concentration of As in column 1 remained the same at 2.3 µg/L. This could be attributed to lower microbial activity in this column, as removal of phosphorus was low in this column, preventing dissolution of metal bearing minerals and a lack of adsorption onto the aquifer material preventing any decrease in concentration.

The implications of metal and major ion reactions during storage are important in ASR because of their impact on post-treatment requirements. Additionally, the exchange of divalent ions such as Mg²⁺ in the formation with Na⁺ in the recharge water can result in clay dispersion which can cause clogging in the formation (Johnson et al. 1999), hence reducing the storage capacity of the aquifer.

The intended end use of the water to be recharged at the ASR site considered in this study is to provide drinking water. The prescribed concentration or value (PCV) relevant to the parameters measured in this experiment for drinking water in the UK (Drinking Water Inspectorate, 2010), along with the maximum concentration measured after storage are shown in Table 3. It should be noted that not all the parameters measured in this experiment have PCVs.

Table 3: PCV for parameters measured in the experiment along with the maximum concentration measured and the sample point yielding the maximum value. *All values at this sample point were <0.2mg/l

Parameter	Prescribed concentration or Value	Maximum value after storage for 15 days	Sample point
Ammonium	0.5 mg/L	<0.2 mg/L	N/A*
Nitrate	50 mg/L	1.5 mg/L	S3
Sodium	200 mg/L	36.1 mg/L	S2
Sulphate	250 mg/L	118.5 mg/L	S2
Copper	2mg/l	0.008 mg/L	S2
Arsenic	10 µg/L	3.2 µg/L	S2
Iron	200 µg /L	171 µg/L	S2
Manganese	50 µg/L	1554 µg/L	S3
Nickel	20 µg/L	18.2 µg/L	S1

The only parameter exceeding the PCV is manganese and post-treatment to address this would therefore be required. The sample point that yielded the highest concentrations in recovered water for most of the parameters with PCVs was S2. The least treated water, S1 resulted in the highest concentration in only one parameter, while S3 resulted in two and S4 resulted in none. This suggests

that using S2 would present the highest risk with regards to changes in the recharge water quality during storage therefore should be avoided, and S4 would provide the lowest risk option. S4 is also the most treated therefore expensive water thus consideration needs to be given to S1, which could provide the best option as it presents a low risk and is the least expensive water.

Predictions regarding potential concentrations of the measured parameters at field scale, based on the experimental data should be made with caution for the following reasons. Firstly the size of the soil columns used pale in comparison to the extent and heterogeneity of an actual aquifer. Although attempts were made to ensure the columns were representative of aquifer conditions, the aquifer material was obtained from one point in the aquifer and therefore may not truly represent the formation. Secondly, the material used in the columns was crushed. Although using the whole core would have maintained the structure of the rock i.e. the porosity and permeability, which could improve extrapolation of the results to a field scale, the samples were crushed to ensure that the columns would be composed of a controlled homogenous material, as opposed to using different cores each varying in structure and composition. In order to assess the impact of varying recharge water quality on recovered water quality, replication with different waters is necessary. Using crushed material focuses attention on changes in recovered water quality as a result of the different recharge water qualities only, and avoids confounding due to differences in the structure and composition of the core. This is a common approach as evidenced by its use in Patterson et al. (2010), Rinck-Pfeiffer et al. (2013) and Vanderzalm et al. (2013). Using crushed material increases the reactivity of the sediment as surfaces that were not previously in contact with water are exposed. This suggests that not all the changes seen in recovered water during the experiment would correspond to the changes seen in the field, and the magnitude of these changes may be lower in the field. The results of this study however are still very useful in predicting potential changes that may occur since the increased reactivity of the columns provide a “worst case” scenario of changes that may occur.

Conclusions

Despite a relatively short storage time (15 days), the results of this study demonstrated that the quality of recharged water can change in important ways during storage in an aquifer. Significant reductions in the concentration of nitrates were seen in all the source waters, and a reduction in sulphate and phosphorus was also evident. These were attributed to an increase in microbial activity, with a corresponding increase in the TOC due to bio-denitrification. Overall the concentrations of metals and major ions were found to increase and the reduction of mineral oxides were inferred to be the main mechanism responsible for this. There was some evidence of microbiological activity influencing the reduction of minerals as where microbiological activity was limited, the concentration of Co, Mo and As reduced rather than increase.

The only parameter exceeding the drinking water PCV was Mn which implies post-treatment would be required to reduce its concentration. At this stage, it is difficult to establish the influence of the different pre-treatments on the recovered water quality as in order to make any confident predictions on changes during storage and the influence of pre-treatment on the recovered water quality, more data is required. Additional work is currently being undertaken to repeat the tests using varying retention times (from 20 to 60 days) to provide a more robust dataset and exclude any anomalies. This wider range of retention periods will allow for a more reasoned assessment on the influence of pre-treatment and residence time on potential changes that may occur during storage.

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