RECLAIMED WATER AQUIFER STORAGE AND RECOVERY: POTENTIAL CHANGES IN WATER QUALITY

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ABSTRACT

Aquifer storage recovery (ASR) operations recharge and recover water through the same well or wells, creating a “bubble” of stored water that typically mixes minimally with ambient groundwater once the storage zone has been developed. Because ASR recharge water does not flow through a vadose zone and is not drawn through an aquifer to a distant point of withdrawal, changes in water quality that occur in surficial aquifer recharge may not apply in ASR operations. Nevertheless, significant water quality changes have been observed during ASR operation, which is of special interest when reclaimed water is stored and recovered. A two-year study was conducted to observe water quality changes in reclaimed water ASR operations at four test sites over differing recharge and recovery periods. The 97 constituents measured at each site included conventional wastewater analytes as well as other constituents of concern, e.g., arsenic, estrone, and carbamazepine. Storage periods ranged from two weeks to eleven months. Where possible, samples were collected from both the ASR well and nearby monitoring wells to assess lateral variations in water quality. Total coliform and heterotrophic plate counts, combined with a reduction in total organic carbon (TOC), nutrients, and dissolved oxygen (DO), suggested microbial activity near the ASR well at most sites. Total trihalomethanes (TTHMs) and haloacetic acids (HAAs) were found to decrease in concentration at three of the four study sites. Pathogens were rarely detected. Microcontaminants exhibited high variability, largely attributed to assumed recharge water variability. Of the trace organics measured, only atrazine showed consisted reduction at all sites detected.

KEYWORDS

Aquifer Storage and Recovery (ASR), reclaimed water, natural treatment systems, pharmaceuticals, microcontaminants

INTRODUCTION

Aquifer storage and recovery (ASR) is a type of groundwater recharge in which water is injected into an aquifer for storage and extracted at a later time from the same well. Storage of reclaimed water through ASR represents a powerful tool for water conservation. A significant problem challenging more widespread application of reclaimed water ASR, however, is the uncertainty regarding the impact of recharged water on native groundwater resources. This concern, and the concern for the quality of the reclaimed water after it is stored, drive the need for an understanding of the fate of contaminants present in the recharge water, from common wastewater compounds to trace organics not addressed in primary or secondary drinking water standards.
In regulated reclaimed water applications, public health is protected by a series of barriers, such as source water control, advanced wastewater treatment processes, and cross-connection control. By adding time and travel through the subsurface, ASR potentially offers another step in the series of barriers that protect public health in the use of reclaimed water. This project aimed to define organic and other wastewater constituents deemed most significant for reclaimed water ASR and measure the concentrations of those contaminants at various stages of recharge, storage, and recovery of reclaimed water. The data was then analyzed with respect to likely attenuation mechanisms and rates to characterize water quality changes through ASR.

**Analytes**

The constituents monitored in this study were selected based on their regulatory relevance, frequency of occurrence in the environment, expected mobility in aquifers, usefulness in aquifer characterization, significance to irrigation water quality, and their degree of public and/or scientific concern.

The analytical plan relied upon local laboratories for analysis of conventional contaminants and the Southern Nevada Water Authority (SNWA) for analysis of all trace organics, including pesticides, disinfection byproducts, and pharmaceuticals and personal care products. Trace organics were specifically selected to help characterize removal mechanisms. Compounds of varying physical characteristics, namely molecular weight and affinity for partitioning to solids, were chosen with the aim of analyzing potential attenuation trends relative to these characteristics.

The final constituent list consisted of 97 compounds grouped into the following categories: basic water quality parameters (e.g., pH, dissolved oxygen), nutrients, metals, disinfection byproducts, radionuclides, pathogens, and microcontaminants (trace organics).

**Test Sites**

This study was designed to investigate the variables of aquifer characteristics, storage time, travel distance, recharge water quality, and operational history. The four test sites chosen for study were ASR wells at Chandler, Arizona; Englewood, Florida; Manatee, Florida; and Bolivar, South Australia. A sampling plan was developed for each of the four testing sites to capture unique environmental and operational conditions. The plans designated the time and location of sampling with respect to actual recharge, storage, or recovery practice. Sampling events were integrated with planned operating and monitoring activities where possible. Table 1 summarizes the key physical and operational parameters of interest at the four sites. Table 2 summarizes the sampling plan.
Table 1. Summary of Site Characteristics

<table>
<thead>
<tr>
<th>Attribute</th>
<th>Chandler, Arizona</th>
<th>Englewood, Florida</th>
<th>Manatee, Florida</th>
<th>Bolivar, South Australia</th>
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<tbody>
<tr>
<td>Mineralogy</td>
<td>alluvium</td>
<td>carbonate</td>
<td>carbonate</td>
<td>calcite, quartz</td>
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<tr>
<td>Salinity, TDS (mg/L)</td>
<td>1,000</td>
<td>20,000</td>
<td>2,000</td>
<td>2,100</td>
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<tr>
<td>Temp. groundwater (°C)</td>
<td>n/a</td>
<td>n/a</td>
<td>26</td>
<td>22-26</td>
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<tr>
<td>Temp. recharge water(°C)</td>
<td>24</td>
<td>26-28</td>
<td>24</td>
<td>7 - 18</td>
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<td>Pre-treatment before ASR</td>
<td>ext. aeration</td>
<td>aeration</td>
<td>aeration</td>
<td>activated sludge</td>
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<tr>
<td></td>
<td>NdeN tertiary filters</td>
<td>filtration chlorination</td>
<td>traveling bridge filter chlorination</td>
<td></td>
</tr>
<tr>
<td></td>
<td>UV</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>TOC in injectant (mg/L)</td>
<td>7.4</td>
<td>9.6</td>
<td>10.6</td>
<td>12.6</td>
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<td>Residence time in aquifer</td>
<td>0.5 to 5 mos.</td>
<td>0.5 to 2 mos.</td>
<td>0.5 mos.</td>
<td>0-11 mos.</td>
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<tr>
<td>Prior Use of ASR site</td>
<td>2004 to present</td>
<td>2001 to present</td>
<td>None</td>
<td>1999</td>
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<td>Target Storage Volume (MG)</td>
<td>300</td>
<td>700</td>
<td>10</td>
<td>40</td>
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<tr>
<td>Travel time to obs wells</td>
<td>14 days</td>
<td>no obs well</td>
<td>no obs well</td>
<td>1-2d; 90-120d</td>
</tr>
</tbody>
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Notes:
- n/a - Not available.
Table 2. Sampling Plan

<table>
<thead>
<tr>
<th></th>
<th>Chandler</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
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<th></th>
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<tbody>
<tr>
<td></td>
<td>5/3/05¹</td>
<td>7/6/05</td>
<td>7/22/05</td>
<td>8/17/05</td>
<td>5/18/05</td>
<td>5/25/05</td>
<td>5/31/05</td>
<td>12/1/04</td>
<td>12/14/04</td>
<td>12/28/04</td>
<td>5/6/04</td>
<td>9/28/04</td>
<td>5/12/05</td>
<td>7/21/05</td>
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<tr>
<td>Event</td>
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<td>recovery</td>
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<td>recovery</td>
<td>recovery</td>
<td>background</td>
<td>recharge</td>
<td>recovery</td>
<td>recharge</td>
<td>storage</td>
<td>recovery</td>
<td>recovery</td>
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<tr>
<td>Wells sampled</td>
<td>ASR, obs. well²</td>
<td>ASR</td>
<td>ASR, obs. well</td>
<td>ASR</td>
<td>ASR</td>
<td>ASR</td>
<td>obs. well</td>
<td>ASR</td>
<td>ASR</td>
<td>ASR</td>
<td>ASR, 4m, 50m obs.well</td>
<td>ASR, 4m, 50m obs.well</td>
<td>ASR, 4m, 50m obs.well</td>
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</tr>
<tr>
<td>Flow in or out (mgd)</td>
<td>1.3</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>0.562</td>
<td>0.438</td>
<td>0.430</td>
<td>0</td>
<td>2</td>
<td>2</td>
<td>0.256</td>
<td>0</td>
<td>0.454</td>
<td>0.342</td>
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<tr>
<td>Approx. storage volume (MG)</td>
<td>310</td>
<td>330</td>
<td>313</td>
<td>269</td>
<td>660</td>
<td>656</td>
<td>649</td>
<td>0</td>
<td>5</td>
<td>5</td>
<td>35.7</td>
<td>40.8</td>
<td>32.1</td>
<td>8.35</td>
</tr>
</tbody>
</table>

Notes:
¹All dates are presented in Month/Date/Year format.
²Observation well.
Conceptual Model
A generic conceptual model of a single aquifer storage recovery (ASR) well and monitor well system was utilized to aid with analysis of the data. This conceptual model, shown in Figure 1, is based upon typical characteristics of ASR operations worldwide (Pyne, 2005).

Figure 1. Conceptual Model of Aquifer Storage and Recovery

Proximal Zone
Immediately surrounding the borehole or screened section of the ASR well is a “proximal” zone, typically within a few feet to a few tens of feet surrounding the well. This is a zone of relatively high geochemical and microbial reactivity, driven primarily by the generally high oxidation-reduction potential (ORP), possibly high nitrate content, and relatively high dissolved organic carbon (DOC) content of the recharge water. Nutrients such as ammonia and phosphate and small but important traces of ferric hydroxide may also be present in the recharge water due to pretreatment processes. Water quality gradients in this zone are potentially high, with ORP typically dropping from positive several hundred millivolts to negative several hundred millivolts during injection (Vanderzalm et al., in press). pH values also may vary, typically lower near the well and increasing away from the well. Velocities decline as the recharge water moves away from the well into the aquifer, providing some opportunity for settling out of any entrained particulates and sorption of colloids (Skjemstad et al., 2002; Greskiowak et al., 2005). Microbial metabolism products, corrosion and precipitation products accumulate in this zone. Desorption, adsorption and other processes also occur. A geochemical “rolling front” may occur in this zone, tending to mobilize metals during each successive cycle and move them away from the well a short distance during recharge, where they adsorb again. Microbial processes contribute to the mobilization and attenuation of metals and other constituents.
The number of pore volume flushes experienced in the proximal zone during a single long ASR recharge period will be at a maximum next to the well, declining with the square of the distance from the well. Some rapid geochemical reactions reach equilibrium within a few pore volume flushes while others may take hundreds of pore volume flushes to reach equilibrium. It is perhaps helpful to look upon this proximal zone as a kind of “sponge” with a high capacity to cause water quality changes due to a variety of reaction processes, whether during ASR recharge, storage or recovery.

During extended storage periods, a die-off of microbial biomass occurs due to lack of new dissolved organic, carbon and nutrients. This may result in negative redox potential at the ASR well (Vanderzalm et al., in press). Some of this organic material is then backflushed from the ASR well at the beginning of recovery.

**Storage Zone**

Once the recharge water leaves the proximal zone and enters the main portion of the storage zone further away from the well, geochemical and hydraulic gradients are reduced, the number of pore volume flushes during each ASR cycle declines and water quality changes are less pronounced. During the initial ASR operating cycle, water quality changes are at a maximum. With successive cycles at approximately the same storage and recovery volumes, the storage zone around the well gradually becomes purged of ambient groundwater. Geochemical and microbial reactions tend to be less significant. Typically, after about three to six cycles at the same storage volume, the storage zone achieves close to a geochemical and microbial equilibrium. This process is accelerated if a portion of the stored water is left in the well during each of the first few cycles.

**Buffer Zone**

At the edge of the storage zone is a buffer zone surrounding the well, typically at a radial distance of several hundred feet. However, in layered media, layers with lower hydraulic conductivity may not necessarily be flushed by injectant and residual native groundwater diffuses slowly into the flushed zone during the storage and recovery phases (e.g., Pavelic et al., 2006). The buffer zone separates the stored water from the surrounding water in the aquifer. Depending upon the differences in water quality between the stored water and the ambient groundwater, water quality changes in the buffer zone may be substantial. Ideally, water in the buffer zone is not recovered during the withdrawal stages.

If the buffer zone is formed in one step at the beginning of ASR operations, instead of cumulatively over a period of several operating cycles, geochemical and microbial quasi-equilibrium may be achieved during the initial operating cycle. Otherwise, equilibrium will be achieved over several cycles at approximately the same volume, typically requiring several years of operations. The sum of the volume in the buffer zone and the volume to be recovered seasonally is called the Target Storage Volume (TSV). The TSV is typically measured in terms of million gallons (MG) per million gallons per day (mgd) of recovery capacity, which converts to a unit of “days.” Experience to date with ASR wells in brackish aquifers of Florida suggests typical TSV values of 50–350 days. TSV values in excess of 70 days have been associated with attenuation of arsenic, i.e., lower arsenic concentrations in the recovered water than in the recharge water.
Mechanisms of Water Quality Change

During ASR recovery, the first flush of water recovered may contain particulates and biosolids, as discussed above. The subsequent water, after a few operating cycles at approximately the same volume, or after formation of the buffer zone, is usually relatively uniform in quality. Contact time between the stored water and the aquifer matrix is typically weeks to months for ASR wells compared to hours to days in SAT systems, some bank filtration wells, and wastewater treatment plants. Perhaps more importantly, most, but not all, ASR wells are in deep, confined anoxic aquifers under reducing to highly reducing conditions. A few ASR wells are in deep water table aquifers with low level oxidizing conditions. Flow in ASR wells is, by definition, in two directions, away from the well during recharge and back toward the well during recovery.

Due to the long flow path through the aquifer for water recovered toward the end of an extended recovery period, any opportunity for desorption may tend to lead to steadily increasing concentrations of desorbed compounds in the recovered water along the flow path toward the well, reaching a peak concentration and then decreasing as desorption proceeds to completion. Conversely, microbial processes may augment, inhibit or override desorption processes. The science has yet to be developed regarding the complex interplay of microbial and geochemical processes occurring during ASR storage.

RESULTS

The data sets from the four sampling sites were analyzed with this conceptual model in mind. General constituents are addressed first, followed by microcontaminants. The following site features were considered throughout the analysis:

- Chandler, AZ underwent a salt tracer study at the beginning of sampling. Salt was injected into the ASR well to measure its travel time to the nearby monitoring well. While conductivity at the monitoring well never increased, a spike in conductivity, TDS, and chloride was observed in the final sample of recovered water from the ASR well. This indicated that the salt traveled in a different direction from the monitoring well but was later recovered after sufficient pumping.

- Manatee, FL was sampled during its first ever injection phase. Prior to injection, a sample was taken of native groundwater. Because there would be known mixing of injected water and groundwater for at least the first few cycles of recharge, concentrations in recovered water were interpreted with respect to a mixing ratio established by TDS and boron concentrations.

- Bolivar, Australia, had previously injected secondary wastewater in its aquifer. Thus, the recharge water sample collected at the start of the study period does not reflect the residual water characteristics, which included higher organic and nitrogen concentrations. Also, the water recovered in the final stages of this study appeared to contain a substantial fraction of groundwater. Using TDS as a tracer, the percentage of recharge water in samples taken on this last date from the ASR well, the 4m monitoring well, and the 50m monitoring well were calculated to be 60%, 40%, and 80%, respectively.
General Parameters

Chandler

Bicarbonate in the recharge water increased from 132 to 204 milligrams per liter (mg/L) during movement of the water from the ASR well to the observation well. It also increased from 132 to 164 mg/L during two months of storage at the ASR well. No significant change in bicarbonate occurred during six weeks of recovery at the ASR well. Calcium also increased at the ASR well during the recovery period, from 71 to 110 mg/L. It appears that some carbonate may be present in the storage zone mineralogy.

Chloride was detected in the range 410–464 mg/L at the ASR and observation well except for the last sample during recovery (936 mg/L). This peak may have been a result of the salt slug that was added three months earlier that appeared to have migrated outside of the monitoring zone until sufficient recovery efforts drew it back in.

Dissolved oxygen (DO) at the ASR well during recharge (8.3 mg/L) did not decline significantly during either two months of storage or movement through the aquifer to the observation well. This is unexpected, suggesting minimal microbial or geochemical activity, or perhaps more likely a faulty measurement. Field measurements of dissolved oxygen were made with an Orion portable meter. It is not known whether a flow cell was used, or whether the equipment had been calibrated.

pH values changed moderately, buffered by the high alkalinity. Of some interest is that the lowest pH (7.2) occurred at the beginning of recovery from the ASR well, consistent with microbial and/or geochemical activity around the well.

Oxidation-reduction potential (ORP) remained positive in all samples, which was unexpected. During the 2-month storage period, ORP declined from 150 to 63 millivolts (mv), but increased to 165 and 151 mv in the subsequent samples from the ASR well. Observation well samples also had unexpectedly high ORP values (122 and 170 mv). This is consistent with the relatively high dissolved oxygen measurements. Subsequently it was determined that ORP measurements were made in the lab using Method SM 2580, not in the field, so the values are undoubtedly inaccurate.

The sodium concentration at the beginning of recovery from the ASR well was very high (550 mg/L), declining during recovery to 210 mg/L after six weeks. Considering the sodium chloride tracer addition at the observation well, and the elevated chloride concentration, a higher sodium concentration might have been expected at the end of recovery at the ASR well. Possibly cation exchange occurred, exchanging sodium for calcium.

Englewood

Uncertainty exists regarding recharge water quality for comparison with the second (5/31/05) recovery sample. That water was stored during mid-March, slightly more than two months previous and may therefore not be adequately represented by the recharge sample collected on 5/18/05. Accordingly, greater weight is placed upon the reliability of
conclusions based upon water quality comparison of the 5/18/05 and 5/25/05 samples, representing a two-week actual storage period between the time when this water was recharged and when it was recovered.

Bicarbonate increased slightly during two weeks of storage, from 158 to 180 mg/L, but did not increase further after 2 months of storage. During the same time period calcium concentration increased from 44 to 58 mg/L and then to 77 mg/L, suggesting some dissolution of limestone.

TOC of the reclaimed water declined substantially. It was initially 9.58 mg/L, declining to 7.89 mg/L in 2 weeks and to 2.58 after 2 months. The decline of TOC and nutrients, combined with the strong indications of microbial activity, suggest that the oxidation-reduction potential should have been reduced, along with the dissolved oxygen concentration. While the dissolved oxygen measurements were taken in the field, the ORP analyses were conducted in the laboratory. Given exposure of the sample to oxygen during travel, the ORP results are invalid.

Manatee
Potential natural tracers for this site included chloride, TDS and boron. However chloride in the aquifer is 514 mg/L while in the recharge water background sample it was 401 mg/L, not much difference on which to base conclusions regarding mixing. Boron concentration was measured at 0.06 mg/L in the groundwater and 0.27 mg/L in the recharge water. No supplemental data was available from the County regarding typical boron concentrations in the reclaimed water.

Considering TDS and boron as reasonable tracers it appears that the recovery sample (12/28/04) represented a blend of between 64 and 76% ambient groundwater with 24 to 36% reclaimed water. Changes in water quality varying significantly from this blend ratio would be indicative of subsurface microbial or geochemical reactions. For those constituents measured at non-detect levels, the blend ratio was calculated using a zero. This introduces a small amount of bias into the resulting conclusions.

Significant departures from predicted concentrations in recovered water based upon blend ratio were that pH and conductivity were higher than expected and DO and TOC were lower than expected given mixing alone.

Bolivar
Chloride was considered the best natural tracer for this site, to distinguish changes in water quality due to blending with groundwater from those due to geochemical and microbial reactions. Departures of greater than about 20% from water quality values predicted through blending were assumed to be significant.

In this suite of samples taken after 11 months storage and three months of recovery, chloride concentrations of 611, 715, and 502 mg/L were detected at the ASR, 4m, and 50m well, respectively. These indicated an increased percentage of native groundwater in the final recovered water samples.
Temperature was 17.6ºC in the recharge water during winter months, however ambient groundwater temperature was 25.9 ºC. At the end of the five month storage period, temperatures at the ASR well and the 4m observation well were 15.1 and 15.8ºC, respectively, while water at the 50 m observation well was 20.6ºC. These values increased to 17, 21, and 20 after 11 months storage and three weeks pumping and 23, 21, and 21 after 11 months storage and three months pumping. A relatively substantial annual temperature variation occurs in the ASR storage zone at this site.

Dissolved oxygen (DO) was 11.9 mg/L in the recharge water and 0.8 mg/L in the ambient groundwater. At the ASR well after 5 months the DO content was 2.7 mg/L, which may perhaps reflect entry of oxygen into the sample. At the 4m and 50 m observation wells DO levels were 0.4 and 0.1 mg/L, respectively. After 11 months storage, DO levels were consistently low, with a minimum of 0.01 and a maximum of 0.67 mg/L detected.

Redox potential was 389 +/- 32mv in the recharge water. Ambient groundwater redox potential averaged 29 +/- 56mv. At the ASR well after 5 months of storage the redox potential was 112mv, which is consistent with the unexpectedly high DO concentration in the well. At the 4m observation well the redox potential had declined from +389mv to -32mv. After 11 months of storage and three weeks of recovery, redox potential values were 9, -36 and 21mv at the ASR well, 4m and 50m observation wells, respectively. After 11 months of storage and three months of recovery, redox potential values were -41, -62 and 9mv, all significantly lower than the previous sample set and well below ambient groundwater values. This indicates continuing microbial and geochemical activity close to the well during extended ASR recovery.

**Nutrients**

**Chandler**

Total nitrogen increased during ASR storage, from 1.7 to 2.4 mg/L, and increased further to 6.4 mg/L during recovery. Nitrate concentrations during this same period increased from 0.9 to 3.9 mg/L, which is opposite what would be expected under reducing conditions. Desorption may have occurred along the flowpath to the ASR well, overriding any decrease in nitrate concentrations resulting from denitrification.

Total phosphorus declined from 4.1 mg/L in the recharge water to 1.5 mg/L after two months of storage, and then declined further to 0.42 mg/L after six weeks of recovery. Ortho-phosphate behaved similarly, declining from 1.9 to 1.47 mg/L during storage and to 0.97 mg/L after six weeks of recovery. Of some interest is that phosphate was never detected at the observation well.

**Englewood**

Nitrogen species showed a consistent reduction during 2 weeks of storage with ammonia dropping from 1.9 to 1.1 mg/L and nitrate from 1.1 to 0.1 mg/L. Phosphorus levels remained high, between 2.4 and 2.8 mg/L.
Manatee
Concentrations of all compounds at the Manatee site were interpreted with respect to mixing ratios determined from boron and TDS as tracers. The data were interpreted with respect to mixing because the Manatee site because sampling occurred during Manatee’s first injection into this aquifer, thus there was no buffer zone preventing direct mixing with native groundwater. Even after accounting for the mixing ratios, nitrite values appeared to have increased while nitrate and phosphorus appeared to have decreased over the two week storage period.

Bolivar
Total nitrogen in the recharge water was 8.2 mg/L. At the ASR well after 5 months of storage, the total nitrogen was 9.41 mg/L while at the 4m observation well it was 1.26 mg/L, a considerable reduction. However, at the 50m observation well it was 7.7 mg/L. After 11 months of storage and 3 weeks of recovery, Total nitrogen values at the ASR well, 4m and 50m observation wells were 0.7, 1.0, and 5.2 mg/L, respectively (mostly ammonia). After 11 months of storage and three months of recovery, total nitrogen values increased from 0.52 mg/L at the ASR well (mostly organic N) to 1.9 mg/L at the 4m well (mostly nitrate). The increases are believed to reflect mixing with residual, nitrogen-rich injectant stored in previous cycles.

Total phosphorus (TP) was 1.58 mg/L in the WRF recharge water sample. The orthophosphate and TP concentrations after 11 months storage and three months of recovery were similar to those after three weeks of pumping, all markedly lower than concentrations after five months of storage. Thus, phosphorus in both total and ortho forms typically decreased with distance from the well and with storage time. This substantial reduction in TP is likely due to sorption, precipitation and microbial activity in the proximal zone.

Metals

Chandler
Changes in metals concentrations were generally unremarkable. A slight increase in barium occurred during ASR storage and recovery, from 0.017 to 0.063 mg/L, possibly due to the presence of barium minerals in the storage zone. Iron concentrations in the observation well were high, corresponding perhaps to the relatively high turbidity values. The observation well is constructed of carbon steel casing, which could have contributed to the iron levels. The WWTP process does not include coagulation, which could otherwise add iron to the reclaimed water.

Englewood
With the exception of an increase in iron from 0.11 to 1.18 mg/L during two weeks of storage, metals concentrations were essentially unchanged. Since the casing material is carbon steel, the increase in iron is not unexpected. A corresponding increase in turbidity, from 0.8 to 1.7 NTU, was also noted, probably representing rust in the recovered water.
Manatee
Iron and barium concentrations were higher in the background sample than in the recharge water at Manatee. These concentrations in the recovered water were consistent with expected mixing ratios, given no evidence for transformation other than mixing. Arsenic showed peculiar results, however, in that it was detected at 0.008 mg/L in the background water, was not detected at an MDL of 0.007 mg/L in the recharge water, and was found at 0.024 mg/L in the recovered water. This could be a spurious data point or reflect mobilization triggered by the injection of recharge water.

Bolivar
Analysis of the metals data indicates mobilization of arsenic close to the ASR well, with concentrations increasing from 2 µg/l in the recharge water to 75 µg/l in the ASR well after 5 months of storage. At the 4m and 50m observation wells the concentrations were 20 and <2 µg/L, respectively. After 11 months of storage, arsenic concentrations were 20, 7 and 1µg/L after three weeks of pumping and 11, 7, and <1 after three months of pumping at the ASR well, 4m and 50m observation wells, respectively.

Ambient groundwater iron content was 1.0 mg/L. The recharge water contained <0.03 mg/L. After 5 months of storage, however, the iron concentration at the ASR well was 6.91 mg/L. This may have been a sample collected after inadequate purging of the well, but the casing is fiberglass and would not have generated any rust. The increased iron concentrations possibly reflect dissolution of ferric hydroxide precipitates under low redox conditions related to microbial activity. A significant spike and eventual reduction in iron concentration occurred at the ASR well.

Radionuclides

Chandler
A small but significant increase occurred in gross alpha radioactivity, from 0.5 milli-Becquerel per liter (mBq/L) in the recharge water to 1.7 mBq/L after two months of storage, increasing to 5.4 mBq/L after six weeks of recovery. The values are well below drinking water action levels (15 mBq/L) but are still of interest, suggesting some baseline radioactivity in area sediments.

Englewood
Neither gross alpha particle activity nor total uranium was detected in any of the Englewood samples.

Manatee
Manatee’s background sample contained 4.8 +/- 5.9 mBq/L of gross alpha particle activity and 0.3 +/- 0.4 mg/L total uranium. Neither constituents were detected in the recharge water, and the recovered water was 4.8 +/- 4.4 mBq/L gross alpha and 9.0 +/- 0.4 mg/L total uranium. Given the known dilution effect, these values suggest an
unexpected increase in radionuclides in the recovered water, but the measurements are reported with a high level of uncertainty.

**Bolivar**
Total uranium increased slightly over time at the ASR and 4m observation well, reaching an overall peak of 0.0038 mg/L at the 4m well after 5 months of storage. Uranium was never found above the 0.0005 mg/L detection limit at the 50m observation well. Gross alpha particle activity decreased then increased over the course of storage, with a peak value of 110 mBq/L being found at the ASR well after 11 months of storage and three months of pumping.

**Disinfection Byproducts**

**Chandler**
Total trihalomethanes (TTHMs) declined in the ASR well from 0.17 micrograms per liter (µg/L) to 0.019 µg/L during two months of storage, and declined further to 0.014µg/L during six weeks of recovery. Haloacetic acids (HAA5) declined similarly from 0.096 µg/L to <0.02µg/L during two months of storage, probably all of which occurred during the first few days.

**Englewood**
A significant reduction in TTHMs and HAA5 occurred during 2 weeks of storage, from 8.4 to <0.5 mg/L and from 12 to 1.3 mg/L, respectively. All are very low values. This is a strong indication of microbial activity.

**Manatee**
TTHMs were not detected in the background water, but were found at 12.27 ug/L in the recharge water and at 4.24 ug/L in the recovered water. Given a mixing ratio of up to 3 parts native groundwater to 1 part recharge water, these values do not indicate a reduction in TTHMs beyond dilution over the two week storage period. HAA5 was not measured in the background or recharge water but was found at 0.6 ug/L in the recovered water.

**Bolivar**
TTHM concentrations were 234 µg/l in the recharge water. At the ASR well after 5 months of storage THMs had declined to 18 µg/l while at the 4m and 50m observation wells they had declined to 80 and ≤4 µg/l, respectively. Haloacetic acid (HAA5) concentrations declined from 72 µg/l in the recharge water to <9 µg/l in all observation well samples after 5 months.
Microorganisms

Chandler
Heterotrophic Plate Count (HPC) for the recharge water was 8/mL, increasing to 500/ml at the observation well. However, pathogenic microbiota and protozoa were not detected at the method limits in both wells. Following 2 months of storage, HPC values were essentially unchanged and very low in the ASR well and had declined to 26/ml in the observation well. After 6 weeks of recovery, HPC values in the recovered water had increased to 146/ml.

Englewood
After two weeks of storage, total coliforms were “Too Numerous To Count” and Heterotrophic Plate Count increased from 7 to 850/ml. However fecal coliforms, E. coli, enterococci and coliphage were not detected at the method limits.

Manatee
The only microbial parameter detected in the background sample was Heterotrophic Plate Count of 8.5 counts/mL. The recharge water showed 71/mL and the recovered water 3/mL, a decrease beyond that expected from mixing. The only other microbiological detect at Manatee was 4 cfu/100 mL total coliform in the recovered sample.

Bolivar
Total coliform bacteria declined during the storage period from 110 cfu/100ml in the recharge water to 68 in the ASR well after 5 months of storage, to 15 in the 4m observation well and were not detected in the 50m observation well. Fecal coliform and E. coli bacteria were 6 cfu/100ml in the recharge water but zero in all subsequent samples at all wells. Coliphage, Cryptosporidium and Giardia were analyzed but not detected in all samples.

Microcontaminants

Travel and Laboratory Blanks
When considering microcontaminant data, it is especially important to consider blank contamination. In this study, both travel and laboratory blanks were analyzed. Travel blanks were obtained from reagent water systems at the ASR site by utility personnel, and collected in bottles sent by Southern Nevada Water Authority (SNWA) along with sample bottles. Laboratory blanks were performed by filling a sample bottle with reagent water (Nanopure™) and processing with samples. Both travel and laboratory blanks were handled and processed in the same manner as samples.
The following compounds were dismissed from the dataset due to recurring presence in travel blanks:

- BHT
- DEET
- Estradiol
- NDMA
- Nonylphenol

In several cases, data show that compounds detected in travel blanks were not the result of laboratory contamination, but rather a function of the blank water used at the site or handling of the samples by utility personnel. Where travel blank concentrations were low and only occurred in one sample, the corresponding sample data was not dismissed but was interpreted relative to the travel blank concentration.

**Recharge Water Quality**

Recharge water was sampled at each ASR site. Only one sample was collected at each site due to budgetary restrictions. This is critical, as this grab sample represents one moment in time and does not provide an integrated view of the water entering the ASR well. For the Bolivar data set, reclaimed water variability is probably subdued due to extended storage in oxidation ponds during pretreatment of the wastewater. For the other three sites, less storage is provided at the treatment plant so variability in microcontaminant concentrations is probably more pronounced. Previous data from team members has shown that concentrations of these target compounds can vary diurnally in typical wastewater treatment plants that do not have extended storage to attenuate variability in influent water quality.

**Analytical Approaches**

A significant issue for microcontaminant data analysis is the expected variability in concentrations in the reclaimed water. For this sampling program, only a single sample of the recharge water was analyzed at each site, so it is possible that this sample is not representative. Accordingly, comparison of recharge water quality to recovered water quality involves an inherent degree of uncertainty, evidenced by the frequent occurrence of higher microcontaminant concentrations in recovered water than in recharge water. Conclusions that are more defensible may be obtained by comparing successive recovered water samples in the same well, and comparison of water quality data at adjacent wells obtained at the same time. However, the latter approach fails to address water quality changes occurring during storage periods between the end of recharge and the beginning of recovery.

Given these issues, two analytical approaches were utilized. The first, “Approach A”, considers all water quality data as being equally representative, even though reasonable concern exists that recharge water quality is quite variable with respect to microcontaminants. Consequently, observations based upon comparisons to recharge water quality have an inherent degree of uncertainty. The second, “Approach B” ignores recharge water quality data and focuses upon comparison of recovered water samples at the same well and also recovery samples collected at the same time from different wells.
By not comparing changes from the start of storage, however, changes occurring over longer time periods may not be detected with the second analytical approach.

Microcontaminant concentrations exceeding five times the Method Detection Limit (MDL) were considered significant to support analysis of removal trends. Concentration differences of less than 20% were not considered significant. Thus, a compound needed to appear at least once at a site at five times the MDL and to increase or decrease greater than 20% (after mixing was taken into account) to be considered indicative of a noteworthy change in concentration.

**Trendable Microcontaminant Data**

Table 3 shows the reduction of 52 originally analyzed microcontaminants to 27 microcontaminants usable for trend analysis by approaches A and B described above. The following sections highlight observations for each site. Interpretations can be found in more detail in the full WRF report by this title (in press).
### Table 3. Reduced Microcontaminant Constituent List

<table>
<thead>
<tr>
<th>Original List</th>
<th>Less Rejected Data¹</th>
<th>Less Non-Detects²</th>
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</table>

**Notes:**

¹ Constituents removed from analysis due to repeat presence in lab and/or travel blanks.
² Constituents not measured above MDL in any sample.
³ Constituents not measured at 5 times the MDL in any sample.
Chandler

Although many target compounds were not detected or were present at very low levels in the recharge and recovered water, several microcontaminants were detectable well above the MDLs. The compounds occurring at the greatest concentration (>5xMDL) include: iopromide, meprobamate, TCEP, carbamazepine, dilantin, erythromycin, ibuprofen, naproxen, triclosan, and gemfibrozil. The occurrence of these microcontaminants in wastewater is well documented (Ternes, et al., 2000; Snyder, et al., 2001; Kolpin, et al., 2002). Team members have found these compounds to occur in wastewater effluents at concentrations similar to those found here (Snyder, et al., 2001; Snyder, et al., 2004; Snyder, et al., 2006; Vanderford, et al., 2003); however, the actual occurrence in recharge (reclaimed) water is highly dependent upon the wastewater treatment processes employed.

Additionally, in some cases, compounds detected in the initial sample were no longer detectable in the final sample (i.e., caffeine). In this case, percent removal is reported based on the MDL, and likely underestimates actual removal. Ibuprofen was well removed during ASR recovery (81%), which can be attributed to biodegradation. The moderate removal of several compounds was most likely due to both biodegradation and adsorption. Reduction may also have contributed, however this would most likely have occurred prior to the beginning of recovery and would therefore not be a significant factor for the second analytical approach.

Englewood

Many constituents increased in concentration between recharge and the first sample, collected after 8 days (1 day of storage and 7 days of recovery). This water had been in storage underground for about two weeks. These included hydrocodone (83–103 ng/L); trimethoprim (1-11 ng/L); acetaminophen (<1–2.3 ng/L); erythromycin-H2O (39–180 ng/L); sulfamethoxazole (24–1410 ng/L); fluoxetine (52–97 ng/L); pentoxifylline (15–20 ng/L); carbamazepine (433–606 ng/L); estrone (7.9–10 ng/L); iopromide (4.9–83 ng/L); ibuprofen (103–126 ng/L); diclofenac (<1.0–62 ng/L); triclosan (<1.0–42 ng/L), and gemfibrozil (<1.0–875 ng/L).

The simplest explanation for the increase in concentration for so many constituents during 2 weeks of ASR storage is short-term variability in recharge water quality. An alternate hypothesis, however, is that during 7 days of recovery there occurred desorption of microcontaminants that had sorbed onto the limestone aquifer matrix and associated microbial biofilms close to the well during the previous extended recharge period from 2001, during which a very large volume of water (net 651 MG) had been recharged. It is hypothesized that during this four-year period extensive biofilm formation would have occurred around the well, probably within a radial distance of a few feet to a few tens of feet. This accumulation of carbon would tend to sorb microcontaminants that are more amenable to carbon adsorption. Microcontaminants would sorb on both the biofilm and the aquifer matrix during recharge, with highest concentrations closest to the well. Desorption would tend to be cumulative along the recovery flow path so that concentrations in the recovered water from the well would initially be high, reaching a peak and then gradually declining as recovery approaches the total volume recharged. For Englewood, this high volume recovery is not likely to ever occur due to practical constraints associated with increasing salinity in the recovered water. Consequently, the potential for recovery of elevated concentrations of microcontaminants, desorbed from the aquifer matrix and from biofilms close to the ASR well would therefore be relatively high. Furthermore, the opportunity for microbial assimilation of these organic compounds under low redox conditions would be minimal during short storage periods.
Compounds at Englewood were generally removed to a lesser extent during the one-week time period between samples as compared to Chandler, even though the differential in subsurface storage time (two weeks to two months) is greater.

**Manatee**
The background sample from the storage zone at Manatee contained non-detect concentrations of all microcontaminants. That no microcontaminants were found was not unexpected for a confined, artesian limestone aquifer over 500 ft deep. Regarding the recharge to recovery water quality changes, and taking the mixing ratio into consideration, the following comparisons were noted.

Compounds found at concentrations higher than expected were Trimethoprim, Sulfamethoxazole, Iopromide, Naproxen, and Gemfibrozil. Compounds found at concentrations lower than expected were atrazine and chlorate. Compounds observed to be at the same concentrations less dilution were Ibuprofen, Hydrocodone, Meprobamate, Carbamazepine, Dilantin, and Erythromycin.

**Bolivar**
Of the 51 microcontaminants evaluated, 30 were not found in the reclaimed water, as analyzed for the WRF sample, or were found at zero or insignificant (less than 5 x MDL) levels in the recovered water. These included hydrocodone, trimethoprim, sulfamethoxazole, fluoxetine, pentoxifylline, meprobamate, oxybenzone, testosterone, progesterone, triclosan, α-BHC, β-BHC, γ-BHC, diazinon, δ-BHC, aldrin, chlorpyrifos, fluoranthene, dieldrin, BDE#28, BDE#47, BDE#100, BDE#99, BDE#154, BDE#153, diclofenac, perchlorate, bromate, iodate, and chlorate.

The remaining data were interpreted with regard to travel distance and time and showed few consistencies. Acetaminophen increased across the data set according to both data analysis approaches. Atrazine decreased from a recharge concentration of 9.2 ng/L and an average of 4.5 ng/L across all wells at 5 months storage time to an average of 1.8 mg/L across all wells at 11 months storage storage time. Estrone decreased from a recharge concentration of 32 ng/L and an average 5 month concentration of 24 ng/L to being not detectable in any of the samples after 11 months of storage.
DISCUSSION

General
While anoxic and low-redox conditions are reasonably expected to occur in the storage zone at all four reclaimed ASR sites due to the abundance of carbon and nutrients and the aquifer confinement, the field data fails to show this, probably due to sampling error arising from unreliable sampling methods utilized at some sites for these two field measurements. Future efforts should emphasize careful characterization of redox conditions, given that these have been found to have an important influence on degradation of some organics in aquifers (e.g., Ying et al., (2003) for several EDCs and Pavelic et al., (2006) for trihalomethanes).

At most sites, TOC, pH, and nutrient declines appeared to occur close to the well, thus supporting the concept of higher biological activity in the proximal zone. Also, the DBP concentration changes observed at three of the four sites were consistent with disinfection byproduct reductions observed at ASR sites in confined, anoxic aquifers where carbon is available to support microbial activity. HAAs are attenuated rapidly through aerobic microbial activity, typically within a few days, while THMs are attenuated more slowly through anaerobic microbial activity, requiring typically a few weeks of storage for complete attenuation. This has been documented in two American Water Works Association Research Foundation (AWWARF) research investigations (Dillon and Toze, 2005).

Microcontaminants
The sampling and analysis program quantified concentrations of trace organics in reclaimed water recharged into aquifers, in aquifer observation wells, and in recovered water using a minimum number of samples. The number of samples required at any one site to statistically support attenuation rates and condition correlations would be one to two orders of magnitude higher than could be supported by the scope of this project. Atrazine was the only microcontaminant present in three or four recharge waters whose concentrations consistently decreased or increased in recovered water at those sites.

Many microconstituents appeared in higher concentrations in the recovered water than recharge water, suggesting high variability in recharge water. Monitoring well data at sites where these were sampled are also inconsistent with source water and recovered water concentrations again inferring variability of input concentrations is the largest factor affecting measured concentrations in monitoring wells and in recovered water.

Possible causes of temporary increases in microcontaminant concentrations in recovered water have been speculated. These include recovery of colloidal organic material entrapped in the aquifer close to the ASR well, where microcontaminants sorb to these organic colloids and the biofilm in the aquifer near the ASR well during the injection cycle. Another possibility is desorption, i.e., mobilization of microcontaminants from immobile phase to soluble phase as a result of pH or temperature change in water that is returning to the injection well as the result of a radial gradient in the aquifer. It should be noted, however, that strong pH gradients are not expected to persist through the storage phase at any distance from the injection well (Greskiowak et al. 2005). Temperature gradients may be quite persistent in aquifers following ASR (e.g., Pavelic et al., 2006) but it is doubtful that these could be sufficient to cause a enough change in the adsorption isotherm to facilitate measurable desorption.
CONCLUSIONS

The data compilation show that residence times and degradation rates in the aquifers were insufficient to claim biodegradation of trace organics to a degree that eclipses source concentration variability at the four sites. A substantially larger database would be required to establish degradation rates of microcontaminants within the aquifers in field experimental programs.

There is some evidence that certain pollutants degrade in aquifers between recharge and recovery, however, such as TTHMs, HAAs, and atrazine. These species may be contained close to an ASR well and their concentrations may be reduced or eliminated before they have an opportunity to migrate away from the ASR site. For this group, ASR may be considered as part of a treatment train, in addition to providing seasonal and long-term storage.

By observing changes in concentrations of over 90 compounds at four ASR sites with many variables, this study intentionally took a broad assessment of water quality changes in reclaimed water ASR storage. For compounds in which this dataset exhibited more variability than trend, it hopefully provides a baseline for future investigations of narrower focus. The full details of this study can be found in the WateReuse Foundation publication by the same title, currently in press.

ACKNOWLEDGEMENTS

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REFERENCES


