

Chapter 2

Stormwater Reuse via Aquifer Storage and Recovery: Risk Assessment for Sandy Aquifers

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Abstract In the light of increasing demand and diminishing supplies due to climate change and related alterations in rainfall pattern, a sustainable urban water management for Melbourne and other cities will need to include water recycling and reuse of reclaimed water and stormwater. One key issue in water reuse is the need for storage between times of collection and demand. Aquifer storage and recovery (ASR), which is defined as well injection of source water into a suitable aquifer for later recovery, would be a valuable option as it has limited space requirements, restricts loss from evaporation and limits the potential for contamination. However, stormwater commonly contains elevated levels of heavy metals, of which Zn and Cu are the most frequent and mobile. Stormwater and treated wastewater also contain suspended solids, organic carbon, oxygen and nutrients, which influence the behaviour of injected metals and induce geochemical changes in the aquifer. While ASR has been practised in limestone aquifers in South Australia, field data for sandy aquifers, which are more prevalent around Melbourne, are very limited. After giving an overview about current ASR practices, the general aim of the study was the risk assessment of injected metals derived from stormwater during ASR in sandy aquifers. As no field sites fulfilling these criteria are in operation in Melbourne or anywhere else in Australia, this study was limited largely to literature review and laboratory experiments.

Keywords Aquifer storage and recovery • Stormwater • Heavy metals • Risk assessment • Groundwater protection • Urban water management • Water reuse

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Introduction

Water is vital to the survival and growth of all life and all economic and environmental processes, but the world's supplies of freshwater are under increasing threat from overexploitation, pollution and climate change. Among other countries, Australia has experienced a severe drought over the last 10 years and in Melbourne water supply reservoirs are at an all-time low with the lowest storage levels on record of below 30% at the end of June 2009 (Fig. 1).

A climate change study for Melbourne predicts higher temperatures and reduced rainfall for the future, which would increase the demand and decrease the supply (Howe et al. 2005). A sustainable water management is therefore vital for Melbourne and other cities around the world facing a similar challenge. The sustainable water management will also be an essential part of rural Australia, where the drought has resulted, for example, in a decline of more than 95% in rice production in the past 10 years (Barclay 2010).

Decision-makers have largely opted for desalination as the main solution to the problem. The high energy needs and the problems associated with disposal of highly saline brine make it an environmentally challenging option (Lattemann and Höpner 2008). On the other hand, urban areas produce stormwater and treated effluent streams, in volumes matching the demand (Mitchell et al. 2002) and dispose of them into urban waterways and adjacent coastal areas leading to degradation of these ecosystems (e.g. Walsh et al. 2005). It is therefore of twofold benefit to recycle stormwater and reclaimed water and this has been identified as an important strategy in maintaining a sustainable water supply for Melbourne.

Benefits and Constraints of ASR

The progress in this direction is often hindered, not by technical or economical reasons, but due to perceived health concerns by the public and decision-makers (Dillon et al. 2009; Po et al. 2003; Toze 2006) and by institutional structures that are not suitable for non-centralized systems. Water treatment is capable of rendering wastewater to a quality superior than that of potable water (e.g. Cheremisinoff 2002) and hence reuse as drinking water is possible. There is an even greater potential to use water treated to lower standards, hence using less energy, for non-potable uses, which constitutes more than 70% of urban water demand (Mitchell et al. 2002).

The benefits of ASR are manifold. Public confidence in recycled water is increased when the water has been put back into the natural system before reuse (Dillon et al. 2009; Po et al. 2003) as ASR increases water quality, e.g. through the removal of pathogens and nutrients (Toze 2006). ASR could provide capacity for inter-seasonal or inter-year storage, especially in aquifers of higher salinity that are of limited beneficial use (Pyne 1995; Maliva et al. 2006). Compared to surface storage, ASR has limited loss of water due to

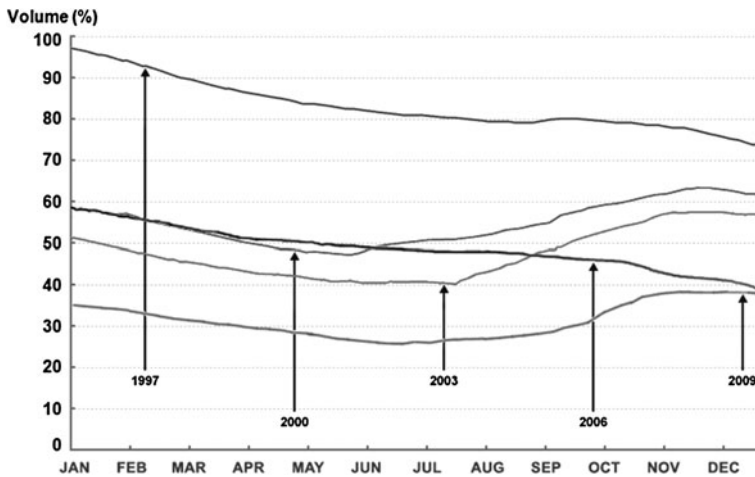


Fig. 1 Melbourne's total water system storage (10 reservoirs combined, total volume 1810.5 GL) development from 1997 to 2009, adapted from (Melbourne Water 2010)

evaporation and is also less affected by contamination or algal blooms and does not provide mosquito breeding grounds (Pyne 1995; Dillon et al. 2006). Space requirements are also limited, which is especially valuable in urban areas for projects larger than household scale (Hatt et al. 2006). And additionally, the costs of artificial groundwater recharge are less than investments necessary for large traditional dams (BGR 2008) or desalination (Dillon et al. 2009).

Despite these benefits, uptake of ASR in Melbourne has been limited by a number of restraints. Insufficient legislation and guidelines for water recycling including water ownership issues (Hatt et al. 2006), lack of experience and knowledge in the water industry and relevant authorities (Dillon et al. 2005; Hatt et al. 2006) and lack of detailed hydrogeological properties (Dudding et al. 2006) have restricted the application of ASR. In addition, more research on well-designed and monitored field projects with long-term performance data, as well as increased hydrogeological knowledge on contaminant attenuation processes and contaminant mobility (Dillon et al. 2005; Maliva et al. 2006) are needed to increase public acceptance (Toze 2006; Dillon et al. 2009).

Stormwater Versus Treated Effluent

Compared to treated effluent, stormwater has the advantage of lower concentrations in nutrients, organic matter and total dissolved solids (Mitchell et al. 2002) and is perceived as more acceptable for reuse by the public (Po et al. 2003). On the other hand, stormwater is generated in single events of different magnitudes and is often seasonally confined. Stormwater quality and quantity thus is rather variable

and depends on a range of parameters, e.g. land use, rainfall intensity, frequency and volume (e.g. Wong 2006). Even within a catchment, pollutant concentration can change by more than one magnitude (Sansalone et al. 1995) during the hydrograph of a single event [e.g. first flush effect (Sansalone and Buchberger 1997; Barbosa and Hvitved-Jacobsen 1999)], from one event to the next and over the seasons [e.g. seasonal first flush (Lee et al. 2004)]. A vital part of stormwater reuse is therefore the collection and storage of the variable sources for later access.

Treated effluent, on the other hand, is a relatively constant and continuous source, but storage for summer months would commonly be needed nevertheless. It contains higher concentration of nutrients, salinity and organic matter. Its use for agricultural purposes, which commonly comprise about 70% of the total water demand, is therefore of twofold benefit, as the addition of fertilizers can be reduced due to the nutrients contained in the water. Another advantage of using reclaimed wastewater instead of stormwater is its lower impact on stream flows. Firstly, treated effluent would not be discarded into streams, deteriorating its quality and, secondly, stormwater flows would still be available as environmental flows. The reuse of reclaimed wastewater with and without aquifer recharge has been successfully implemented in many countries around the world and is being promoted in rural areas (e.g. Abu-Madi et al. 2008).

ASR Technique: Theory and Case Studies

The artificial recharge of groundwater can occur via infiltration or injection. The infiltration via infiltration basins, seepage trenches or vadose zone wells requires the availability or construction of an adequate permeable soil and unconfined aquifer (Bouwer 2002). Injection of water into the aquifer can involve one well for injection and recovery and is defined as ASR, while the use of one well for injection and another well for recovery is defined as aquifer storage, transport and recovery (ASTR). It requires a suitable aquifer and better infrastructure, but injection rates are usually higher than infiltration rates (Bouwer 2002).

In general, managed underground storage involves the following elements (Fig. 2):

1. Collection or harvesting: Water sources can include potable water, surface water, groundwater, treated effluent, and stormwater. This study will be mainly concerned with urban stormwater runoff.
2. Pretreatment: Pretreatment options can include standard stormwater pollution control design practices such as litter and sediment traps, wetlands and ponds, advanced treatment and disinfection (Victoria Stormwater Committee 1999; Ellis 2000; Hatt et al. 2006).
3. Injection: Suitable aquifers can be of different types: confined or unconfined, from unconsolidated sands and gravels to limestones and fractured sedimentary or volcanic rocks.

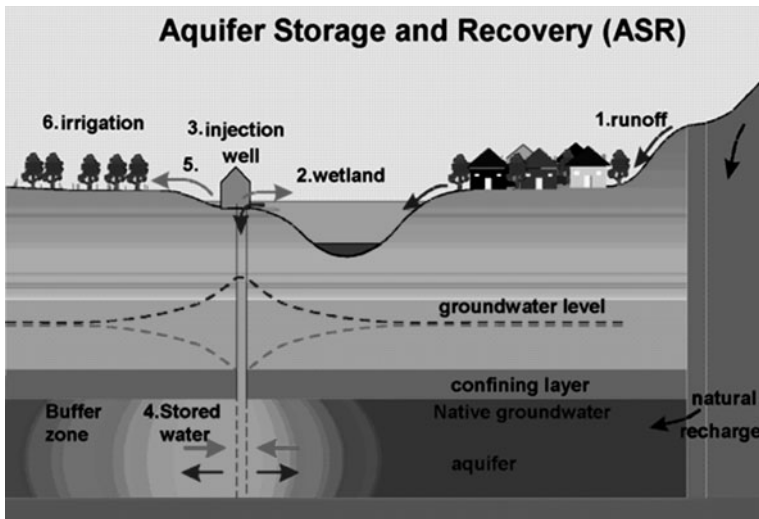


Fig. 2 Schematic aquifer storage and recovery system, modified after (CSIRO Land and Water graphics [2010](#))

4. Storage: A buffer zone separates the native groundwater from the injected water.
5. Recovery: Recovery is typically achieved in a number of recovery stages depending on demand.
6. End use: Recovered water can be used for drinking water, irrigation, firefighting, environmental flows, toilet flushing, industrial cooling and other purposes, and might be subjected to post treatment.

Site Selection Considerations

The site selection for an ASR scheme will depend on a range of factors. Firstly, a suitable water source and a local demand together with the availability of some space for stormwater collection and pretreatment are needed. Secondly, administrative, economic and operational constraints have to be met and finally, a suitable aquifer must be available (Pyne [1995](#); Dillon and Molloy [2006](#)).

The suitability of the aquifer is mainly defined by its storage capacity and recovery efficiency (Pyne [1995](#); AGT et al. [2002](#)). Both fractured and unconsolidated rock systems could be used for ASR systems. While storage capacity in fractured rock can be quite high, its recovery efficiency is often lower due to the usually not well-known heterogeneous characteristics of the fractures. Dual porosity limestone aquifers have been favoured in many regions due to their potential for increasing the storage volume by calcite dissolution and water quality

improvements even with low recovery efficiency in karstic regions (Gerges et al. 2002). Unconsolidated porous aquifer systems have potentially the highest storage capacity. Predictions about the hydraulic performance of the system are much more reliable than for fractured rocks. In general, the aquifer should be sufficiently transmissive to avoid excessive build-up of groundwater mounds, preferably homogeneous and of sufficient extent for the scale of the ASR scheme.

Another consideration is the groundwater quality. It is commonly advisable to have a lower-quality groundwater, so beneficial use will not be impaired by the injection of non-potable water. Very high salinity levels, on the other hand, will increase the buffer zone of an ASR scheme substantially or will limit the recovery efficiency. Contaminated aquifers need to be avoided, as this would compromise the quality of the recovered water.

One problem not only experienced with infiltration but also associated with injection ASR is clogging. This reduction in porosity mainly around the injection well leads to a decrease in injection rate and an increase in hydraulic head. Clogging is a process that will usually occur, but in most cases can be managed with pretreatment and redevelopment (Pyne 1995; Brown et al. 2006). The pretreatment can include filtration/coagulation of suspended particle and micro-organisms, addition of pH-elevating substances to reduce iron precipitation, and disinfection to avoid microbial growth (Pyne 1995). Nevertheless, periodical redevelopment by purging the well will be undertaken in regular intervals at all sites. The disposal of these backflushed waters that are usually of low quality have to be accounted for in the planning stage of ASR schemes. Limestone aquifers are likely to compensate for loss in porosity by dissolution of carbonic matrix. Addition of acidic agents has been used to enhance this process and decrease the effect of clogging (Gerges et al. 2002).

Water Quality Considerations

A number of issues such as the uncertainty regarding the possible impacts of recharge water on the beneficial use of the native groundwater resources, the potential for increased discharge into streams and wetlands as well as the quality of the recovered water have to be considered.

A better understanding of processes such as the fate of contaminants present in the recharge water and the interactions taking place in the disturbed groundwater system will also help with guideline development. These are of crucial importance for protecting environmental values and beneficial uses of the groundwater as required by statutory authorities such as the Environmental Protection Agency (EPA Victoria 2003).

Depending on the source water and the end use, different aspects of water quality will be of importance. A good compilation of water quality improvements that can be achieved during ASR has been presented by Dillon et al. (2005). When using lower quality water as recharge, the removal of pathogens, natural organic

matter (NOM) and nutrients and associated redox reactions are of more importance. For all schemes, biogeochemical interactions with the aquifer matrix such as ion-exchange and dissolution/precipitation will have an impact on the recovered water quality, however.

Microbial Pathogens

Pathogens comprise a diverse group of microorganisms and have been found in high concentrations in wet weather urban runoff originating from animal faeces and sewer overflows (Wong 2006). The fraction of organisms associated with settleable particles varies by type of microbe, and the partitioning behaviour of each organism generally changes between dry weather and storm conditions (Characklis et al. 2005). Once injected, their inactivation in the subsurface can be achieved by adsorption, filtration, oxidation, degradation by native groundwater microorganisms and die-off (Pitt et al. 1999; Foppen and Schijven 2006). The main factors favouring long-term survival of pathogens in the ground are small size (reduced straining especially for viruses), soluble organics (reduced adsorption), low oxygen levels (reduced oxidation), low temperature, high pH and low salinity (prolonged life span) and the native microbiota (John and Rose 2005). Monitored ASR sites achieved complete removal of pathogens at storage times longer than 90 days (Dillon et al. 2005). It is therefore not necessary to disinfect water before injection. This will also avoid the formation of carcinogenic disinfection by-products.

Natural Organic Matter

Natural organic matter (NOM), usually measured as total organic carbon (TOC), consists of both particulate (POC) and dissolved (DOC) organic carbon and comprises a complex mixture of low to high molecular weight organic molecules. The bulk of NOM found in stormwater are fulvic and humic acids. The degradation rate of NOM varies widely. In general, smaller molecules with carboxylic and phenolic groups are favourable for biodegradation while large aromatic molecules are more stable (e.g. Howard 2000). Fulvic acids are soluble over the wide range of pH, while humic acids are insoluble at lower pH. Humic substances are most elongated and flexible at high pH, low salinity and low NOM concentration (e.g. stormwater), while they form spheres at higher salinity (>0.05 M), in the presence of trivalent cations and at high NOM concentrations (e.g. Ghosh and Schnitzer 1980). This behaviour plays an important role for cation and metal binding and pH buffering. Depending on the properties of the organic molecule, pH and surface properties of the clay or oxide, NOM can adsorb to clay minerals and oxides via a range of mechanisms (Stevenson 1982; Jardine et al. 1989). The usually positive

surface charge of the underlying clay or oxide thus changes to negative surface charge and thereby influences the cation exchange capacity, sorption of other organic molecules and colloidal stability of the particles (e.g. Stevenson 1982; Kretzschmar and Sticher 1997). A complete reversal of surface charge and complete coating of matrix with NOM can be achieved with as little as 0.5 mg-C/L TOC (Day et al. 1994). Mono-layer coverage of organic matter may impede further sorption of NOM (Jardine et al. 1989; Day et al. 1994), but competitive exchange of more hydrophobic macromolecules with less hydrophobic molecules has been observed (Gu et al. 1996a, b), resulting in higher mobility of smaller organic molecules.

Redox Reactions

NOM in the injectant is the major driving force for microbial growth. Its oxidation acts as an energy source for microorganisms and controls the redox status especially near the injection well. If the electron acceptor is an aqueous species such as O₂, protons are consumed, while reactions involving reduced minerals (e.g. sulphides, ferrous oxides) can release protons (Appelo and Postma 1999). In the natural subsurface environment, these processes will be partly taking place simultaneously due to heterogeneities of the matrix, distribution of reactants and microorganisms, meaning that there is usually no equilibrium between all redox couples in the groundwater. Typically, groundwaters are low in both particulate and dissolved organic carbon (<0.5 mg-C/L). The recharge with organic, oxygen and nutrient-enriched injectant, therefore, is a major disturbance to the system (Bahr et al. 2002). Around the well (<10 m) where particulates are entrained, a so called “treatment” or “proximal” zone with highly elevated microbial activity develops. Here dissolved oxygen is eliminated in a few days, followed by nitrate reduction (Greskowiak et al. 2005; Vanderzalm et al. 2006) triggering further geochemical reactions such as degradation of organic contaminants, mineral dissolution/precipitation, sorption and pH changes that influence the quality of the recovered water (Dillon et al. 2005).

Ion Exchange

The aquifer matrix provides a number of cation exchange sites on clay minerals, oxides and organic matter (Appelo and Postma 1999). The injection of low salinity, mostly Ca-dominated recharge water, into brackish often Na–Mg-dominated groundwater, firstly decreases the salinity, which might cause swelling of clays and a decrease in permeability (e.g. Konikow et al. 2001). Secondly, it will lead to simultaneous homo- and heterovalent ternary cation exchange. The selectivity sequence would usually favour Ca, which would replace Mg, which in turn

replaces Na, leading to a high loading of Ca on the exchanger and increased concentrations of Na and Mg in the solution (Appelo and Postma 1999). This usually helps to restore the permeability. During the establishment of an ASR scheme, these ion exchanges will be moved outwards into the buffer zone and not play a major role for later ASR cycles (Bahr et al. 2002).

Dissolution/Precipitation

The chemical stability of minerals depends on redox and pH conditions as well as the activity of ions in solution (Appelo and Postma 1999). As stormwater is usually of neutral pH, the change in redox state is the main driver for mineral dissolution and precipitation reactions, which are therefore mainly taking place near the injection well. Minerals of high reactivity and main importance in ASR are carbonates, hydr-/oxides and sulphides. Dissolution of minerals, especially carbonates, is induced by undersaturation with low ion activity in the recharge waters, especially by highly treated injectants (Johnson et al. 1999). The decrease of pH due to release of CO₂ during organic matter degradation also accelerates dissolution of carbonates (Herczeg et al. 2004). In limestone aquifers this relieves the effect of clogging but might lead to collapse of the aquifer matrix structure (Pavelic et al. 2006). The interplay between oxidative/reductive changes induced by ASR and dissolution/precipitation reactions with minerals can be complex. The injection of oxygen into an anoxic aquifer with dissolved Fe and Mn will lead to the precipitation of amorphous hydroxides, while reductive dissolution of Mn- and Fe-oxides starts after oxygen is completely depleted and nitrate is in the progress of reduction (Stumm and Morgan 1996). Both reactions occur in ASR operations close to the injection well, where the injected organic matter induces redox and pH changes (Greskowiak et al. 2005). While the precipitation of minerals will lead to co-precipitation and adsorption of trace metals, the dissolution will in turn mobilise previously incorporated or adsorbed trace metals (Bahr et al. 2002). Pyrite oxidation is of special importance as it releases mobile oxyanions such as arsenate and selenate and mobile cations such as nickel and zinc. It also significantly reduces the pH due to the formation of sulphuric acid in solution, which can mobilise other adsorbed metals. Multiple ASR sites have experienced a decrease in recovered water quality due to geogenic metal release related to pyrite oxidation (Brown et al. 2006). Over a number of ASR cycles, these effects seem to be decreasing and shifting towards the buffer zone of the ASR scheme.

ASR in Use

Research on potable injection and infiltration ASR has been going on for some decades most prominently in the USA (Pyne 1995; Brown et al. 2006), with most emphasis on quantitative aspects such as clogging and recovery efficiency mainly

for long-term and emergency storage. It is becoming more popular in the Middle East together with desalination plants (e.g. Almulla et al. 2005). All schemes in the USA need to comply with the ASCE (2001) standards, which do not allow the injection of poor-quality water (Brown et al. 2006). Therefore the only water quality problem in these schemes is geogenic metal mobilization, mainly concerned with arsenic (Jones and Pichler 2007). Australia appears to be at the forefront of non-potable water recycling and as one of the driest countries in the world has focused on water reuse in urban areas. Adelaide, South Australia, has taken on a leading role in ASR research and has operational, trial and proposed sites, which are more or less well studied. Well-studied examples for stormwater ASR include Andrews Farm (Herczeg et al. 2004; Pavelic et al. 2006) and Parafield (Marks et al. 2005). For treated effluents, the ASR investigations at Bolivar (Dillon et al. 2005; Greskowiak et al. 2005; Vanderzalm et al. 2006) have to be mentioned. As they are all situated in limestone, these schemes cannot directly be compared to potential sites in Melbourne.

Generally, performance results are somewhat limited, as some projects are still relatively new and also because water quality monitoring has only been carried out or documented infrequently. This is partly due to the fact that in Australia no specific guidelines for stormwater recycling are currently in place and the existing (usually treated wastewater recycling) guidelines are not enforceable standards (Hatt et al. 2006).

First Conclusions

From the review so far it can be stated that ASR offers a valuable solution to the storage problem associated with stormwater and treated effluent reuse in urban areas, but implementation has been slow due to administrative hurdles and a lack of political will. In addition, the interactions between microbial, geochemical and hydraulic processes during ASR are complex. They are dependent on injectant, aquifer and groundwater site specifics and have not yet been fully understood. ASR scheme feasibility depends largely on the storage capacity of the aquifer and they are preferably implemented in transmissive sandy or limestone aquifers (Pyne 1995). However, ASR schemes have to take into account the protection of the aquifer and groundwater to reduce the impact on downstream environments and be sustainable in the long term. Limestone aquifers have been used successfully for stormwater ASR in South Australia and provide a high retention capacity for metals due to the alkaline pH. The hydrogeological setting of Melbourne is quite different and potential ASR sites are situated in sandy alluvial aquifers (AGT et al. 2002; Dudding et al. 2006), which have lower pH and might have limited sorption capacity for heavy metals. A clear gap in ASR experience in alluvial sands has been recognized (AGT et al. 2002).

Stormwater ASR in Sandy Aquifers

The risk associated with water recycling to augment drinking water supplies is currently under review and stormwater reuse and managed aquifer recharge are part of the Australian guidelines for water recycling (NWQMS 2007). As elevated levels of metals are ubiquitous in urban stormwater (Makepeace et al. 1995; Wong 2006) and metals cannot be degraded, their behaviour during stormwater ASR was investigated. In contrast to South Australia, where stormwater ASR has been practised for a number of years in limestone, Melbourne's potentially viable aquifers are mainly silicious (Dudding et al. 2006). As no field site was available for this study, laboratory experiments with local aquifer sediments, groundwater and stormwater were undertaken instead to evaluate the risk to the environment posed by injected metals during stormwater ASR. Common empirical methods to investigate metal solid-solution interactions are batch (OECD/OCDE 2000) and column tests (e.g. Schweich and Sardin 1981; Bürgisser et al. 1993) and were hence employed. Currently available hydrogeochemical models (Prommer et al. 2003) were adapted and enlarged to help interpret the empirical results. A detailed description of the experiments goes beyond the scope of this paper and can be found in Wendelborn 2008.

Stormwater Quality

Urban stormwater is a valuable water resource that needs to be integrated into an urban water conservation management to meet future water demands. Nevertheless, urban stormwater contains a range of pollutants that have negative impacts on receiving ecosystems and potentially need to be treated before reuse (Pitt et al. 1999). Stormwater quality and quantity is rather variable, however, and depends on a range of parameters, but commonly occurring priority pollutants have been identified (Eriksson et al. 2007).

Total Suspended Solids

Total suspended solids (TSS) comprise organic and inorganic particulates ($>0.45\ \mu\text{m}$) deriving from natural and anthropogenic sources. TSS concentration in stormwater ranges typically from 50 to 450 mg/L with lower concentrations found in roof runoff and highest concentrations found in urban road runoff. Apart from their clogging potential during infiltration or injection, their importance arises from the attachment of other pollutants (e.g. nutrients, hydrophilic organics, heavy metals) mainly with the finer fraction of particulates (e.g. Dempsey et al. 1993; Mikkelsen et al. 1994; Deletic and Orr 2005). While the coarser particles are likely to be settling out before use in ASR schemes, the finer, more contaminated

proportion is likely to be injected into the aquifer and not to be recovered, as observed at Andrews Farm (Pavelic et al. 2006). Particulate facilitated transport is therefore a potential pathway for contaminants to be distributed in the aquifer and should not be neglected (Dempsey et al. 1993; Liebens 2001) especially as particle size distributions (PSD) analysis of Australian surface runoff showed that higher fractions of smaller particles are generated compared to Europe or the USA with a mean particle size of $\sim 50 \mu\text{m}$ (Wong 2006).

Nutrients and Oxygen

Nitrogen and phosphorus in stormwater derive from fertilizers, faecal matter, plant debris and combustion processes. While they are essential nutrients for plant growth, increased levels may lead to eutrophication and toxic algal blooms. While phosphorus attaches readily to particulates, nitrate is highly soluble and very likely to be injected during ASR. Dissolved oxygen and nitrate are the major electron donors injected into the aquifer stimulating microbial activity, significantly altering the aquatic ecosystem community (Datry et al. 2004).

Organic Carbon

Fulvic acids are the major colloidal and dissolved constituent in natural organic matter. Concentrations typically range from 8 to 35 mg/L. Interactions between DOC, dissolved constituents and solid matter are numerous. DOC stimulates the growth of biofilms, biological production and subterranean ecosystem metabolism and redox reactions (Stumm and Morgan 1996; Baker et al. 2000). Its presence influences the speciation of heavy metals and can increase their solubility through complexation. From the column experiments it is clear that the distinction between mobile and immobile organic carbon is crucial, as mobile organic carbon increases solubility of metals and facilitates their transport, while immobile organic carbon enhances the accumulation of metals in the subsurface. Organic carbon in stormwater consists of a conglomerate of organic molecules with different molecular weights and functional groups. Accordingly, a fraction will be mobile in the subsurface, while other fractions will either be strained, precipitate or sorb to the matrix, becoming immobile. In low ionic strength solution, remobilization of organic carbon can occur by biodegradation, increase in pH or dissolved small organic acids.

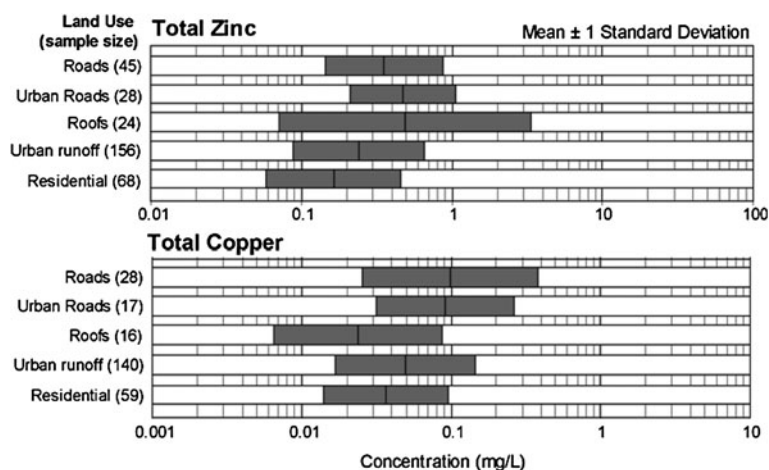


Fig. 3 Typical concentrations of worldwide stormwater samples for Zn and Cu in stormwater (modified after Wong 2006)

Heavy Metals

Heavy metals in stormwater derive from natural sources, as well as from different anthropogenic sources such as hydrocarbon combustion, vehicle exhaust, brake and tyre wear, roof materials, corrosion of metal objects, etc. (Makepeace et al. 1995; Wong 2006). They are often significantly toxic at low concentrations and are therefore considered priority pollutants in stormwater (Eriksson et al. 2007). Their importance stems from the fact that they (in contrast to organic contaminants) cannot be degraded, but are persistent and tend to bioaccumulate.

Metal concentrations in stormwater are highly variable depending on a number of factors such as land use, climate and rain event characteristics. According to their prevalence in stormwater and toxicity Pb, Zn, Cu and Cd (e.g. Barbosa and Hvitved-Jacobsen 1999; Legret and Pagotto 1999; Pitt et al. 1999) are the most important metals for study. Zn and Cu concentrations are highest from urban road runoff and Zn concentrations are increased through runoff from Zn roofs (Fig. 3).

This speciation of metals is vital to assess their potential for injection during stormwater ASR, as larger particulates will be settling out in the detention ponds before injection, while fine particulates and dissolved metals would not (Wong 2006). While Pb has been found to be nearly completely attached to particulates, Zn, Cu and Cd are more associated with dissolved solids and colloidal material (e.g. Sansalone and Buchberger 1997; Legret and Pagotto 1999). As Cd is frequently below detection limit in Melbourne's stormwater and non-settleable fractions of Pb are also low, the main focus of this study is Zn and Cu. Relative to overseas studies catchments in Melbourne showed elevated concentrations of Zn (~ 1.1 mg/L) and comparable concentrations of Cu (~ 50 μ g/L). Compared to

other metals Zn and Cu were predominantly found in the dissolved and fine particulate fraction. Hence their concentrations were still considerably above freshwater ecosystem guideline values after sedimentation. Without further pretreatment noteworthy amounts of Zn and Cu are therefore likely to be injected into the aquifer in dissolved and colloidal form (Wendelborn 2008).

Metal Behaviour During the ASR Cycle

Some of the processes described for water quality considerations are more or less important for stormwater ASR in general and come into play at different stages of the cycle. The main considerations will be highlighted below.

Pretreatment

In most cases stormwater will not be used for injection straight from the source, but would be collected and pretreated to a degree. Best management practices for urban stormwater (Victoria Stormwater Committee 1999; Hatt et al. 2006) offer a range of primary treatment methods to reduce contaminant loads, the most common being wetlands and detention ponds, which utilize the effect of sedimentation of suspended solids and attached contaminants (e.g. Braskerud 2000; Walker 2001). This helps to prevent clogging of bores and also decreases metal loads attached to settleable solids. However, sedimentation does not affect loads from dissolved metals or metals attached to particles about $<20\text{ }\mu\text{m}$ (Walker and Hurl 2002), and the accumulated metals and organic matter in the sediment have the potential to be remobilized (e.g. Kalbitz and Wennrich 1998; Walker and Hurl 2002). Further improvements to the water quality has been achieved by using constructed wetlands with vegetation, which takes up nutrients and accumulates heavy metals (e.g. Zhao et al. 2007; Read et al. 2008). In return, a release of metals can be expected during decay of plant material, and removal rates in the long term are uncertain. Release of nutrients and dissolved organic matter from wetlands has also been reported and can be increased by wildlife, e.g. birds. Further treatment with filtration through gravel, sand or roughing filters has been shown to decrease particle concentration and associated metals further (e.g. Page et al. 2006; Hatt et al. 2007). Secondary treatment with filter material of different material (e.g. alumina, bark, Bauxsol-coated sand, fly ash, granulated activated carbon, iron oxide-coated sand, zeolite, etc.) have been tested with varying degrees of success (e.g. Sansalone 1999; Genç-Fuhrmann et al. 2007). Conventional secondary treatment (Kurniawan et al. 2006) with flocculation/coagulation, liming or dissolved air flotation are possible and effective, but require the right dosing of chemicals and the handling of sludge waste that are usually not practicable within the ASR settings.

Clearly, the pretreatment step is the most crucial step for metal behaviour as their fate might already be established if no metals are injected into the aquifer. This would be a favourable outcome. In this study metal concentrations dropped about 40–50% for Zn and 75% for Cu from mean stormwater concentrations through settlement (Wendelborn 2008).

Injection

The injection phase is the main disturbance of the groundwater system, as oxic, nutrient and organic matter enriched, low ionic strength water enters a commonly anaerobic, nutrient poor, brackish groundwater. The pH is commonly around neutral for stormwater and free dissolved metals are likely to adsorb to the immobile or mobile groundwater matrix near the injection well. Complexed metals and colloidal metals are more mobile and are likely to get transported until they reach areas of higher salinity (Ryan and Gschwend 1994; Bunn et al. 2002). Due to the increase in flow velocity (McCarty et al. 1993; Ryan and Gschwend 1994) and low salinity of the injected water, colloids near the well could also be mobilized (Johnson et al. 1999; Konikow et al. 2001) carrying attached metals with them. The increase in dissolved organic carbon is able to desorb previously adsorbed metals, if complexation is more favourable. If free Fe^{2+} are present in the groundwater, this would precipitate and scavenge dissolved metals. Other mineral phases are likely to dissolve due to undersaturation or change in Eh–pH stability conditions in the solution and release metals. Metals leached from the aquifer matrix near the well are then redeposited further away from the well, within the radius defined by the storage bubble.

Mixing along the flow path will diminish the difference between the injectant and the native groundwater, but due to heterogeneities in the aquifer, zones of fast preferential flow will develop next to diffusion-dominated areas and local disequilibrium might increase, meaning that the geochemical conditions and hence the dominating process can be quite different depending on the location.

During stormwater injection in the column experiments, Zn showed complete breakthrough for sediments with low sorptive capacity and starting breakthrough for sediment with organic coating. Solubility was slightly increased due to the addition of DOC. The breakthrough curve for the former sediments pointed to a high and a low affinity surface site, while the breakthrough for the latter sediment was much more gradual, indicating a more gradual change in surface site metal affinity exhibited by different organic functional groups (Wendelborn 2008). Cu was completely adsorbed during stormwater injection without added DOC, while solubility was significantly increased with the addition of DOC, with the organically coated sediment again showing the highest retardation of all sediments. The shape of the Cu breakthrough curve suggested transport as organic colloids and complexes and ligand influenced sorption for all three sediments (Wendelborn 2008). Even though clay was $\leq 0.2\%$ for all sediments they showed

measurable release of inorganic and organic colloids (~ 200 nm) when injected with neutral low ionic strength stormwater.

Storage

The injection of organic matter and nutrients will have stimulated microbial activity in the vicinity of the injection well. During storage, organic matter degradation and associated reduction in redox potential will be the main factor for metal behaviour. Metals that were attached to the organic matter might be released and reductive dissolution of minerals might also release metals. If redox potential drops low enough sulphide precipitation of metals might occur. Changes in pH during this stage might also influence the solubility of metals.

In general, disequilibrium will be lessened due to time for rate limited reactions and diffusive processes. After a number of ASR cycles, diffusive processes will have replaced the native groundwater with infiltration water completely.

In the column experiments, particle mobilization from the aquifer was observed during storages related to organic biodegradation and probably mineral dissolution. In conjunction, increased concentrations of Zn and Cu after storages were observed due to colloidal transport and release from matrix induced by changes in solution parameters during storage. Kinetic biodegradation resulted in sharp metal concentrations and pH increases after storage (Wendelborn 2008).

Recovery

The extent of geochemical changes during recovery is much lower than during injection and depends also on the change in volume and the extent of the buffer zone. Free metals in solution will be recovered and especially non-specifically adsorbed metals are likely to be desorbed with an increase in ionic strength. In the well vicinity, particles and all attached metals will also be recovered.

In the column experiments Zn was released continuously due to dilution and competition with major ions and protons, resulting in high recovery efficiency. Cu release was triggered by a decrease in pH. Both metals showed decreased desorption when DOC was added, resulting in very limited release of Cu. The retained fraction of added DOC had significantly increased specific sorption sites and anoxic conditions were less favourable for metal release (Wendelborn 2008). Hydraulic non-equilibrium conditions due to small scale heterogeneities in hydraulic properties and diffusion limited flow were observed. Chemical non-equilibrium was due to rate-limited sorption visible in extensive tailing (Wendelborn 2008).

Overall

Speciation of Cu and Zn depends on a range of different processes and factors. Cu speciation is mostly influenced by the presence of organic matter, in solution it is mainly found as organic complex and sorption is dominated by specific surface complexes, resulting in slow desorption. Zn speciation is mostly influenced by pH, in solution it is mainly found as free ion and sorption is dominated by non-specific surface complexes, resulting in fast desorption (Wendelborn 2008).

Overall, metals are persistent in the environment and mechanisms are reversible, meaning that metals retained in the aquifer could eventually become mobile again with time and change in geochemical conditions. In conclusion, the interactions of metals with solution and solid described in this section are complex and intertwined. At this stage, no single theoretical approach or model is capable of incorporating all processes and factors and empirical approaches are therefore still common. It is therefore critical to undertake site-specific studies and ongoing monitoring to assess key processes and outcomes.

In conclusion, injected amounts of Zn are mobile and will mainly be recovered. Cu can be mobile in the presence of mobile organic carbon, but will mainly accumulate in the aquifer until the sorption capacity is exceeded. The release of metals was triggered by reduction in pH, increase in ionic strength and particle mobilization. Metal concentrations were high after storage phases. Minor sediment constituents, especially organic matter, significantly reduce metal mobility.

Implications and Recommendations

Implications for Stormwater ASR in Sandy Aquifers

While the results obtained during the laboratory experiments are only representative for short-term behaviour, the lessons learned could be translated into long-term behaviour in sandy aquifers. It should be remembered that spiked metal concentrations during the experiments were considerably higher than expected injected stormwater concentrations, i.e. about four times higher for Zn and about 50 times higher for Cu. One also has to consider that other metals would be present in stormwater and especially Al and Fe would strongly be competing for sorption sites.

The prognosis for injected Zn would be that it would be relatively mobile within the aquifer and would be present throughout the injected freshwater zone. It would be partially adsorbed onto surface sites especially during the first number of ASR cycles, but largely be recovered in similar amounts as injected. If we assume a mean injected stormwater concentration of about 500 µg/L and assume that Zn is on average recovered in similar concentrations, then this concentration would be well below the drinking water guideline of 3 mg/L (NHMRC 2004) and

below the irrigation water guideline of 2 mg/L (ANZECC and ARMCANZ 2000) and should be fit for these purposes. In contrast, these concentrations would be drastically exceeding freshwater ecosystem guidelines of 8 µg/L (ANZECC and ARMCANZ 2000) and should not be used for environmental flows.

The prognosis for injected Cu would need to be differentiated into different scenarios.

- Scenario A: Low concentrations of organic carbon

For scenario A, injected Cu would be largely adsorbed to the aquifer matrix slowly filling up sorption sites from the injection well outwards to the edge of the freshwater zone, expelling less strongly sorbed metals such as Zn in its wake. Desorption due to dilution would be highest close to the injection well, where high volumes of recovered water are flowing past. Unless pH values of recovered water are not significantly lower than pH 6, release of Cu would be limited though and Cu would accumulate in the aquifer over the long term. If the surrounding groundwater has lower pH values, the creation of a larger buffer zone and recovery volumes less than injected volumes would drastically reduce the impact of the original groundwater. Nevertheless, a breakthrough of low pH groundwater in zones of higher transmissivity could result in a surge of desorbed Cu in the recovered water, which could be many times higher than injected concentrations depending on the sorption capacity of the aquifer. Hence, special attention should be paid to recovered Cu values in low pH groundwater ASR settings. In these settings, continuous monitoring of pH would be recommendable, as pH would be a good surrogate indicator for elevated metal concentrations.

If we assume a mean injected stormwater concentration of about 15 µg/L and assume that Cu is largely retained in the sediment, Cu concentrations in recovered water would be well below drinking water guidelines of 2 mg/L (NHMRC 2004) and irrigation water values of 0.2 mg/L (ANZECC and ARMCANZ 2000). Use would only be compromised by release of previously accumulated Cu at low pH. As freshwater ecosystem guidelines are set as low as 1.4 µg/L (ANZECC and ARMCANZ 2000), recovered water would most likely be still not fit for environmental flows, especially in the later years of operation of the ASR scheme, when the sorption capacity has been exceeded.

- Scenario B: Elevated amounts of injected organic carbon

Injected organic carbon can be mobile or immobile in the subsurface. The formation of mobile Cu-ligand complexes would increase Cu solubility and transport, spreading Cu faster throughout the freshwater zone than in the previous scenario and would result in higher recovery of Cu. However, significant portions of the previously mobile organic complexes would presumably be deposited with an increase of ionic strength during recovery, and hence increase Cu accumulation throughout the subsurface. The immobile carbon would accumulate close to the injection well and fuel redox reactions. As not all organic carbon will be degraded quickly, retained amounts of organic carbon would significantly increase the sorption capacity of the aquifer allowing for a longer and higher accumulation of

Cu. On the other hand, release of Cu after storage phases would likely be increased due to the biochemical changes and dissolution and degradation of part of the immobile organic carbon during storage. Backflushed solutions and initially recovered volumes could therefore be unsuitable for reuse.

- Scenario C: Elevated amounts of colloids

Relevant concentrations of colloids could result from injected particles, may be due to mobilization of aquifer fines or be produced during mineral dissolution and precipitation. They have the potential to transport adsorbed metals that would usually be classified as immobile. In general, colloids will be transported along preferential flow path, e.g. zones of higher permeability, and will aggregate and be deposited once ionic strength reaches the critical limit at the outer edge of the freshwater zone (Ryan and Gschwend et al. 1994; Bunn et al. 2002). Over a number of ASR cycles, this could lead to a localized accumulation of fines, potentially lowering permeability and leading to a localized higher metal concentration at the fringe of the storage zone. Overall, this would lead to lower recovered metal concentrations.

To estimate the possible accumulated Cu concentrations a rough calculation could be as follows: With an injected Cu concentration of 15 µg/L it would add up to an injected amount of 15 kg of Cu per year and GL and amount to 300 kg/GL in a life span of 20 years. If we further assume a mean porosity of 0.33 an average Cu concentration of 67 mg/kg would result over 20 years. The experiments showed that even sediments consisting to >98 wt% of sand exhibited considerable sorption capacity with more than 200 mg/kg for Cu and hence Cu concentrations could easily be accommodated within the sediment. Comparing the potentially accumulated concentration with the sediment quality guideline target value of 34 mg/kg (ANZECC and ARMICANZ 2000), shows that the accumulation of Cu would be well above the threshold effect level. Similar calculations for an injected Zn concentration of 500 µg/L would result in 2,200 mg/kg Zn compared to a sediment quality guideline value of 150 mg/kg. This would mean that more than 90% of Zn would have to be recovered to stay below the sediment target value, which could be achievable in low sorption capacity sediments.

It is clear that ASR schemes would be decommissioned once they are no longer viable, e.g. due to clogging of the aquifer or decreasing volumes of stormwater available for injection. Once the injections have stopped, the previous groundwater flow and original groundwater conditions re-establish themselves over time. In the long term, this would result in desorption of accumulated metals due to dilution effects. Desorption would be accelerated if groundwater was of low pH, which could result in an increased release of metals into groundwater fed surface water ecosystems, which would be potentially problematic. Desorption would be limited if groundwater was highly reducing and metals would be precipitated as sulphides.

Aquifer Selection Recommendations

The choice of the aquifer would be restricted by the available aquifer in the location of source and demand of water, and the storage capacity needed for the ASR scheme. From the point of view of this study, the recommendations for an aquifer selection are divided by the question: (a) Do metal concentrations in the recovered water need to be very low (e.g. use for environmental flows) or (b) would long-term metal accumulation in the aquifer that might later pose a risk to downstream systems be of concern and hence recovery of injected metals would be preferred? In the first case high sorptive capacities are a bonus, while in the latter case low sorptive capacities are needed.

Accordingly, a local aquifer with limited recharge to surface water and adjacent aquifers would be the most suitable situation. In this case the aquifer could be used for water purification and high sorptive capacity would be appreciated to lower metal concentrations of the recovered water to a minimum. The accumulated metals in the aquifer would pose very limited risk to surrounding ecosystems. If, on the other hand, the aquifer is highly connected to other aquifers and discharges into adjacent surface waters, then low sorptive capacity and a high recovery efficiency of injected metals would be more appropriate. Given that irrigation and drinking water guidelines allow for much higher metal concentrations than are tolerated by freshwater organisms, high metal recovery would seem to be the most preferred option. It should be taken into account that the sorptive capacity of the aquifer will generally be increased with the injection of colloids and organic carbon as well.

Analysis of aquifer sediments before the implementation of an ASR scheme should include: fine clay, to assess potential mobilization of colloids from the matrix; organic carbon, which offers high amounts of specific sorption sites; pyrite, which is generally an undesirable mineral due to release of arsenic and other trace metals; and cation exchange capacity and possibly specific sorption capacity to assess the accumulation potential of metals. The evaluation of sediment analysis should take into account that surface coatings consisting of oxides and organic matter do not contribute a great deal of mass but offer a large amount of reactive surface area. Low values of Fe^{2+} in the native groundwater would also be favourable as this would prevent the precipitation of amorphous iron hydroxides, which would also increase the sorption potential.

Monitoring Recommendations

Since the accumulation of metals in the aquifers constitutes a concern, the ASR project should regularly be monitoring metal concentrations of injected and recovered (including backflushed) waters, to be able to calculate the retained amount of metals in the subsurface. Currently no ASR project has undertaken this

mass balance for metals. As no stormwater ASR project in sandy aquifers has been established yet, field data to validate the current laboratory experiments is desirable. Additionally, the monitoring of backflushed waters is of special importance, as these would potentially contain the highest metal concentrations and should include analysis for As. These volumes should not be discharged into streams without the knowledge of metal concentrations and have to be disposed of correctly or treated before discharge. The continuous monitoring of pH would be highly recommended, as pH would be a good surrogate indicator for elevated metal concentrations. Low pH readings could then trigger additional sampling and analysis for metals before reuse.

Pretreatment Recommendations

The remediation of aquifers from heavy metal contamination is a difficult and expensive task and it would therefore be advisable to limit the potential for metal accumulation in the aquifer regardless of the specific situation. Recommendations for pretreatment would therefore include the lowering of injected metal concentrations (including Al and Fe) as well as organic carbon and colloid concentrations, as these increase the retention potential of the aquifer. This kind of pretreatment would also be beneficial in other respects as the potential for biological and mechanical clogging would be reduced.

Pretreatment for metals could be achieved with biofiltration, exchangers or reactive filters, while organic matter and colloids would best be reduced via coagulation and precipitation (Kurniawan et al. 2006). While this kind of pretreatment would be desirable, the benefits have to be weighed against the disadvantages of use of chemicals, disposal of waste sludge and associated carbon emissions. Pretreatment with natural or recycled substances and low carbon imprint should be investigated. For particulates and organic carbon, slow sand filtration or roughing filtration would be possible (Page et al. 2006). Metal removal could be achieved with oxide-coated sands (Sansalone 1999; Genç-Fuhrman et al. 2007), biosorbents such as tree fern (Ho 2003), agricultural waste products such as hulls (Marshall and Champagne 1995) or wetlands (Walker and Hurl 2002).

Conclusions

The final conclusion from this study seems to be that water recycling and reuse is a necessity in the current situation of many cities in the world with dwindling surface and groundwater resources. Stormwater ASR is a valuable tool in fulfilling this task. Additional pretreatment that might be necessary to bring recycled water up to acceptable standards for humans should in reality be undertaken anyway before discarding these waters into waterways.

It must be emphasized that there is no one solution to water shortage and a range of options should be implemented simultaneously. The trend is to decentralize supply according to localized demand and use holistic approaches. The most common form of small-scale stormwater reuse is the collection of roof runoff in rainwater tanks. It has been widely employed in rural areas and is increasingly implemented in urban households for domestic use (Mitchell 2004). Greywater tanks are also a favourable option at household scale. The local reuse also saves the cost of drainage, central treatment and redistribution. For larger catchments, detention ponds, retarding basins and reservoirs are needed (Hatt et al. 2006) requiring increasingly more space, which is a limited and costly asset in the urban environment and poses a major problem. Nevertheless, separation of stormwater and wastewater streams limits the treatment costs and energy needs. All schemes should be monitored for pathogens and heavy metals to limit potential health issues.

Apart from technical considerations (Dillon and Molloy 2006), increased efforts have to be made to educate and involve the population, which is accustomed to a centralized system. The community needs to be made aware of the problems and possible solutions. Information about actual dangers and realistic potentials needs to be given. To increase the acceptance for recycled water, the community should be incorporated in decision-making processes and the administrative and legislative frameworks need to be in place to allow innovative ways of water supply.

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