

SOIL AQUIFER TREATMENT: A SUSTAINABLE PROCESS FOR ORGANICS REMOVAL?¹

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This paper examines the efficacy of soil aquifer treatment (SAT) as a method to reliably attenuate the concentration of natural organic matter (NOM) in Colorado River water. NOM is of concern because it serves as an organic precursor for production of halogenated disinfection byproducts (DBPs) during drinking water treatment. This paper reviews previous research on SAT with wastewater effluent, provides an overview of results from riverbank filtration studies in Europe and in the United States, and finally, provides some thoughts regarding the application of SAT as part of a treatment process for Colorado River water.

Results from a multi-year field and laboratory-based investigation performed by researchers involved with the project “An Investigation of Soil-Aquifer Treatment for Sustainable Water Reuse” indicate that SAT is a sustainable process for significantly reducing the concentration of dissolved organic carbon (DOC) in wastewater effluent. Intensive monitoring at field sites in Tucson and Mesa, Arizona, demonstrated DOC removal efficiencies ranging to 90% or more during SAT. Moreover, removal efficiencies have not declined since these facilities began operation in 1990. Based on field and laboratory evidence, biodegradation is considered to be the most important removal mechanism for dissolved effluent organic matter during SAT; thus, the process is sustainable.

Riverbank filtration (RBF) has been successfully used for more than 100 years in several European countries. Interest in RBF is increasing in the United States. Several municipalities in the United States use RBF to reduce the concentration of DBP precursors in their source waters and interest in using RBF continues to increase in this country.

Based upon the combination of information available on SAT of wastewater effluent and RBF of surface water, it appears likely that NOM removal rates during SAT of Colorado River water are sustainable. Recommendations for additional research to resolve information gaps are discussed.

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Introduction

The City of Tucson has embarked upon a large-scale program of groundwater recharge to utilize their allotment of Central Arizona Project (CAP) water (approximately 140,000 ac-ft per year). The Central Avra Valley Storage and Recovery Project (CAVSARP) consists of 11 recharge basins covering about 330 acres with 25 recovery wells on site. The facility can recharge up to 60,000 acre-feet (ac-ft) of CAP water per year into the unconfined aquifer in Avra Valley. The City is currently working towards the goal of adding a second recharge facility (Southern Avra Valley Storage and Recovery Project, SAVSARP) to provide an additional 30,000 to 45,000 ac-ft per year of recharge capacity for CAP water.

During percolation, there are significant improvements in the quality of CAP water with respect to removal of natural organic matter (NOM). CAP water has a relatively low content of NOM for a surface water, with an average dissolved organic carbon (DOC) concentration of about 3 mg/L (Figure 1) (GeoSystems Analysis, 1998). The figure shows a histogram of Colorado River water (CRW) total organic carbon (TOC) data collected from a number of sites from 1991 to 1997. CRW has generally lower TOC concentrations than surface waters in other regions of the United States. However, the concentration of TOC is of concern because it serves as organic precursor material for the formation of disinfection byproducts, including trihalomethanes (THMs) and haloacetic acids (HAAs) during subsequent drinking water treatment.

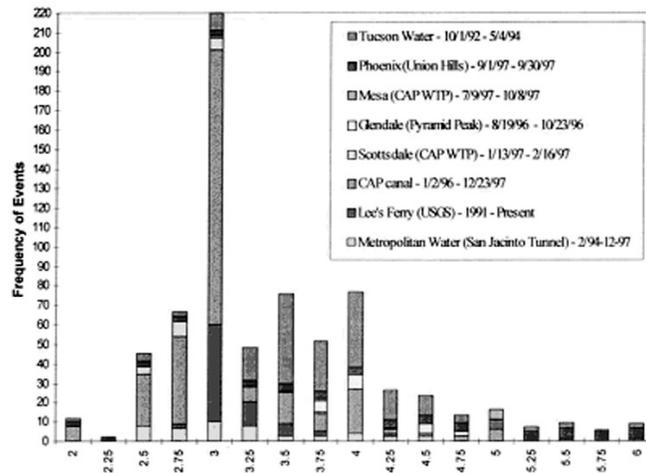


Figure 1. Total organic carbon concentration in Colorado River water from various sites, 1991-1997 (from GSA, 1998).

The formation of disinfection byproducts (DBPs) during the disinfection of surface waters is of concern due to potential adverse health effects. In order to minimize public exposure to DBPs, the USEPA has implemented stages of regulation (i.e., D/DBP Rule) to reduce the amount of DBPs allowable in public water systems, and which

requires that large public water systems collect data on organic carbon content and DBPs in water they serve to their customers (i.e., the Information Collection Rule, ICR). Currently, the DBP constituents of concern relevant to CAP water are total THMs, haloacetic acids (HAA5), and bromate. Additional types of DBPs and specific DBPs may be regulated in the promulgation of the Stage 2 rule. Within the Tucson Active Management Area, the City of Tucson has established a goal of delivering CAP water with total THM concentrations less than the proposed Stage 2 MCL (i.e., 20 µg/l) to their customers.

The character of NOM in CAP water has not been studied extensively, although a large data set of TOC concentrations exists and some recent studies have provided some new details. Data from CAP water treatment plants and DBP formation tests indicate that current or future USEPA regulations of DBPs in drinking water may not be met using conventional treatment and disinfection methods.

Because of the relatively short period of water quality monitoring at CAVSARP, uncertainty remains regarding the sustainability of NOM removal during recharge of CAP water. The focus of this paper is to examine the question of whether NOM removal during SAT of CAP water is a sustainable process. This paper provides a review of previous research on SAT of wastewater effluent, an overview of results from riverbank filtration studies in Europe and in the United States, some thoughts regarding the expected efficacy of SAT as a sustainable treatment process for NOM removal in CAP water, and concludes with recommendations for additional research to resolve information gaps.

Additional Background

The dissolved organic matter in natural waters can be divided into humic/fulvic (hydrophobic) and hydrophilic fractions. The humic/fulvic fraction tends to be more aromatic and higher molecular weight and comprises the more halogen-reactive material, i.e. forms more DBPs after chlorination (Croue *et al.*, 1999; Singer, 1999). The NOM in CAP water contains a relatively smaller humic/fulvic fraction than most other U.S. surface waters, implying that DBP formation potential is lower in CRW than in other U.S. waters.

Owen *et al.* (1995a,b) studied six different surface waters representing the following: southwestern waters with low color and high alkalinity (i.e., the Colorado and Salt Rivers), waters with moderate color and low to moderate alkalinity (i.e., State Project, Ohio and Laurel Rivers), water with high color and moderate alkalinity (i.e., Harwood Mill Reservoir), and southeastern groundwater with high color and high alkalinity (i.e., Biscayne Aquifer). The authors concluded the following about CRW:

- The humic fraction of organics in CRW is lower than that of other U.S. surface waters.
- The humic and non-humic fractions of organics in CRW water have comparable reactivities with chlorine in the formation of DBPs.
- The fraction of NOM with low molecular weight (i.e., less than 1,000 g/mole) in CRW is approximately half the total DOC.

- The fraction of NOM with low molecular weight in CRW is more reactive in forming THMs than other surface waters studied.
- The percentage of brominated THM is greater for non-humic fractions with lower molecular weight than that of bulk CRW.
- The ratio of THMs to HAAs is lower for CRW than for other surface waters.

This study indicated that although CRW has a relatively low content of humic substances, non-humic matter could be a significant DBP precursor source. In addition, the creation of HAAs in CRW may be relatively greater than in other waters. Nonetheless, total THM formation from CRW in this study was notably less than in the other waters studied. Furthermore, the non-humic fraction may be more susceptible to biodegradation, and hence less likely to be transported into groundwater. Overall, the authors concluded that southwestern waters behaved differently than other surface waters found in the U.S. (Owen *et al.*, 1995a,b). These differences were attributed to generally lower organic carbon concentrations, lower humic substance fractions, and higher alkalinity.

The main mechanisms for attenuating NOM in the subsurface are biodegradation and sorption. Biodegradation is a sustainable attenuation mechanism, whereas sorption has a finite capacity and is non-sustainable. Sorption potential is site-specific, and will vary as a function of CAP water characteristics, the depth to water, distance to recovery wells, and the types of soils and aquifer material.

DBP monitoring and DBP formation potential tests have been performed by the Las Vegas Valley Water District (Miller *et al.*, 1995) and Tucson Water (Ward, 1995) in separate injection studies using treated and chlorinated CRW. Both studies showed reductions in the formation potential of DBPs, total THMs, and HAA5 after aquifer storage of CRW. Based on mechanistic studies, HAA5 biodegradation occurs under aerobic conditions, whereas THM biodegradation is anaerobic; the biodegradable fraction of NOM should also degrade aerobically.

Insight from Research on SAT using Wastewater Effluent

Results from field and laboratory-based investigations performed by researchers involved with the multi-year project “An Investigation of Soil-Aquifer Treatment for Sustainable Water Reuse” support the conclusion that SAT is a sustainable process for significantly reducing the concentration of dissolved organic carbon (DOC) in wastewater effluent.

A consortium of universities (University of Arizona, Arizona State University, University of Colorado, and Stanford University) participated in this five-year research project. Investigations with focus on fate of effluent organic matter (EfOM) were conducted at eight water reuse sites in Arizona, California, and Texas which differ in sources used for drinking water supply, source water quality, applied water treatment prior to reuse, and geohydrological settings. Extensive supporting laboratory investigations were also conducted to help determine the fate of EfOM during SAT. The overall goal of the project was to examine the efficacy of SAT as part of a treatment train leading to eventual indirect potable reuse.

The study investigated how NOM from drinking water and soluble microbial products generated during wastewater treatment influence the organic chemical composition of reclaimed water in water reuse systems. The fate of organic matter during SAT was investigated using biodegradation studies consisting of adapted soil-columns, bench-scale microcosms, and at the full-scale using lysimeter, multi-depth sampler and groundwater monitoring wells.

During field studies of SAT, the majority of DOC in wastewater effluent was removed during percolation through the top 0-10 ft (0-3.0 m) of soil. For sites where aerobic or anoxic conditions existed in the upper vadose zone, the DOC concentrations decreased to 3 mg/L or less and were similar to laboratory studies under controlled conditions. Under anaerobic conditions, the DOC concentrations ranged from 4-5 mg/L and were greater than the DOC concentrations observed under saturated anoxic conditions. Considering the time-scale of SAT systems, which is on the order of months, the removal of DOC was similar for both aerobic and anoxic conditions. The removal and transformation of DOC continues to occur over longer time scales. DOC concentrations continue to decrease at low rates while evidence for continuing biological transformations is provided by a decrease in the specific UV absorbance.

Potential structural changes in EfOM were examined with ultrafiltration tests, size exclusion chromatography, XAD fractionation, carbon-13 nuclear magnetic resonance (¹³C-NMR) spectroscopy, infrared spectroscopy, elemental analysis, and fluorescence. Based on ¹³C-NMR results, the decrease in specific UV absorbance was attributed to a decrease in aromaticity (e.g. preferential reduction of aromatic carbon versus aliphatic carbon). The similarity of drinking water and reclaimed water ¹³C-NMR spectra suggested that structure and composition of organic isolates were dominated by natural organic matter. Disregarding small structural changes in the spectra, the characteristics of organic material did not change significantly during drinking water treatment, household use, and wastewater treatment.

Since soluble microbial products are added to refractory DOC in reclaimed water, but do not cause significant differences in the NMR-spectra, the chemical characteristics of effluent-derived and naturally derived organic matter overlap extensively. These findings are consistent with chlorine reactivity of effluent organic matter, which is generally comparable to NOM in the formation of DBPs such as THMs and HAAs. When normalized to DOC, the formation potential of SAT product water was not distinguishable from the trihalomethane formation of drinking water sources. Bromide incorporation into the DBP species was, on average, twice as high for THMs as HAAs.

Sweetwater Recharge Facilities, Tucson, Arizona

As part of this project, a five-year study was conducted at the Sweetwater Recharge Facilities (SRF) in Tucson, Arizona, to assess the sustainability of surface spreading operations for organics attenuation during field-scale soil aquifer treatment (SAT) of municipal wastewater.

Removals of DOC were robust during SAT at the Sweetwater site, averaging >90 percent during percolation through the local 37-m vadose zone, with the majority of removal occurring within the first 1.5 m of soil (Figure 2a). The hydrophilic (most polar)

fraction of DOC was preferentially removed during SAT; removals were attributed primarily to biodegradation. Reductions in THM-FP averaged 91 percent across the vadose zone profile. The reactivity (specific THM-FP) of post-SAT organic residuals with chlorine decreased slightly from pre-SAT levels (60 vs. 72 μg THM per mg DOC, respectively). Variations in the duration of wetting/drying periods did not significantly impact organic removal efficiencies.

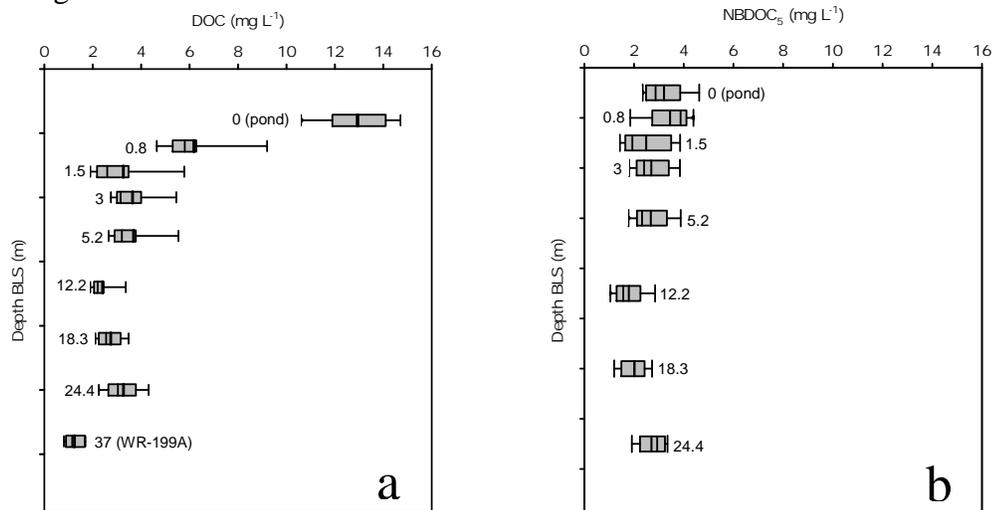


Figure 2. Boxplots of DOC concentration data (a) and NBDOC₅ data (b) as function of depth for samples obtained from suction lysimeters under a recharge basin at the Sweetwater Recharge Facilities during the time period 1996-2001. Soil depth (m) is given next to each box (from Quanrud *et al.*, 2003).

In the original field-scale study at the Sweetwater site, Wilson *et al.* (1995) reported a reduction of 90 percent for DOC during percolation through the 37-m vadose zone over the first 3-4 years of full-scale operations (i.e. 1990-1993). Equivalent removals occurred during the period 1996-2001, indicating that DOC removal efficiency had not diminished over the ten-year operation of the facility.

Following percolation of secondary effluent through about 37 m of unconsolidated sediment at the Sweetwater Recharge Facilities, organic residuals are on the order of 1-2 mg L⁻¹. This value is independent of variation in DOC concentration in the infiltration basins. Reductions of DOC are most dramatic in the uppermost 3 m of sediments, and DOC values are seldom >5mg L⁻¹ below that depth. SAT performance has shown essentially no change over the 10-year life of the facility. The initial performances of newer recharge basins suggest that basin maturation occurs over the first 6-12 months of operation.

Boxplots of nonbiodegradable DOC (NBDOC₅) versus soil depth during 1996-2001 (Figure 2b) show that depth-dependent trends are not strong. Nevertheless, the observed depth-dependent reduction in NBDOC₅ with depth could result from sorptive removal followed by decomposition on a timescale that is much longer than the five-day duration of the BDOC lab test. As implied by the nomenclature, (aerobic) biochemical conversions are expected to be relatively unimportant in contributing to removals of

NBDOC₅ during percolation; anoxic biodegradation may be responsible for decreases in NBDOC₅ at depth. It is also possible that adsorption of residual organics to basin sediments during transport has a modest overall effect on organic composition and bulk concentration. The possible reduction of NBDOC₅ with depth is an intriguing result and suggests percolation can attenuate refractory NOM during recharge of surface waters such as CAP water.

Mesa Northwest Water Reclamation Plant, Mesa, Arizona

Similar studies on SAT were conducted at the MNWRP (Drewes and Fox, 1999a,b; Fox *et al.* 2001). At this site, reclaimed water percolates down to shallow groundwater that migrates laterally to monitoring wells located at travel times estimated at a few days to several years. Concentrations of DOC were reduced to approximately 1 mg/L after 12 to 24 months of SAT with an applied DOC concentration from the NWWRP of 5 to 7 mg/L. THM-FP tests on well waters ranged from 50 to 65 micrograms THMFP/mg DOC.

Efficacy of SAT using Wastewater Effluent: Study Implications

1. The DOC in applied effluents is composed of a combination of easily biodegradable compounds and refractory organic carbon. Refractory organic carbon is assumed to be composed of NOM of drinking water origin, soluble microbial products, and synthetic trace organics.
2. NOM of drinking water origin can persist during effluent pre-treatment and the levels of NOM appear to be independent of effluent pre-treatment. The majority of organic matter added during water use is biodegradable. Therefore, reclaimed water dissolved organic matter is dominated by drinking water DOC and soluble microbial products. These soluble microbial products are similar in character and reactivity to NOM.
3. Based on these results, bulk characterization including XAD fractionation and spectroscopic techniques applied to water samples after long-term SAT could not discriminate organic carbon of wastewater origin from NOM. As a consequence, TOC concentration is an appropriate evaluator of water and wastewater treatment processes, but TOC is less appropriate to represent organics of a wastewater-origin at the point of recovery in water reuse systems.

Riverbank Filtration

While study of EfOM fate during SAT of wastewater effluent is informative, results cannot be directly used to estimate the long-term fate of surface water (i.e. CAP water) NOM during SAT. Research results obtained from studies of riverbank filtration (RBF) provide information useful to predict sustainability of NOM removal during SAT. During RBF, surface water is subjected to passage through aquifer sediments before being collected by extraction wells and subsequently used as sourcewater for drinking water treatment.

The European Experience

Riverbank filtration has been used extensively in Europe to augment drinking water treatment processes for over 100 years. In Switzerland, RBF is used for 80% of all drinking water and in France, RBF is used for 50% of water supply. The same mechanisms acting to improve water quality during SAT are responsible for water quality improvements during RBF. These mechanisms include biological degradation, sorption, ion exchange, and precipitation. The most significant chemical changes are related to microbial activity; most organic matter degradation occurs during early stages of RBF (Tufenkji *et al.*, 2002).

Recent work by Schoenheinz (2003) demonstrated that RBF systems must be actively managed to maintain water quality benefits. The possible release of sorbed hydrophobic organics can occur after stagnation or low flow periods when water is collected directly beneath a river bed with very short flow paths and flow times. Schoenheinz (2003) also noted that use of wet and dry periods to prevent clogging of basins at SAT sites may promote accumulation and release of DOC.

The American Experience

Interest in RBF has increased significantly in the United States in recent years. A major study conducted by researchers at Johns Hopkins University was performed in 1998-2000 and examined the fate of DBP precursors and selected microorganisms during RBF at three full-scale sites in the Midwestern United States. Findings were published in two articles in the Journal of the American Water Works Association (Weiss *et al.*, 2003a,b). At all three sites, there was significant reduction (35-67%) in DOC concentration during RBF. The formation potentials for THMs and HAAs were reduced by 50-80% at the three sites. Consistent at all sites was a shift in the speciation of DBPs in RBF product waters, with a relative increase in the fraction of brominated species and decrease in the chlorinated species. This was attributed to an increase in the Br⁻/DOC ratio as DOC was removed during RBF without corresponding removal of Br⁻. Of particular interest in this study was the finding that reductions in DBP-FP concentrations at all sites were generally greater than corresponding reductions of DOC concentrations, suggesting preferential removal of the reactive portion of the NOM pool during RBF.

Efficacy of SAT using CAP Water

At least three research studies have been performed in the Tucson Basin examining the fate of NOM during recharge of CAP water. Ward (1995) studied direct injection of treated and disinfected Colorado River water to the Central Well field of the Tucson Basin aquifer. The study documented decreases in DOC, THM, and HAA concentrations after aquifer storage. Attenuation was attributed to biodegradation and dilution during aquifer storage.

Two University of Arizona M.S. thesis projects have examined the fate of DBP precursors in CAP water during recharge operations at surface spreading basin sites in Avra Valley. Stinson (1999) conducted vadose zone and groundwater monitoring at CAVSARP over a two-year period, reporting TOC reductions of 30 and 70% in the vadose zone and at a recovery well, respectively. An upward trend in TOC concentration

at recovery wells was noted at the conclusion of the study. A follow up study by Lutz (2000) found similar TOC removals of 30 and 60% in the vadose zone and at recovery wells at different CAP recharge basins in Avra Valley.

Artiola (2003) reported a consistent 72% reduction of TOC during recharge of CAP water at the CAVSARP facility over a 1.5-year monitoring period in 2001-2002, consistent with monitoring by City of Tucson personnel showing stable rates of TOC removal at around 70% during recharge of CAP water. The majority of removal occurred within the first 90 feet of percolation depth. The fraction of NOM classified as fulvic/humic acid was about 35%, lower than most other surface waters, but in agreement with previous work on Colorado River water.

Conclusions and Recommendations

Based on common experiences derived from studies on EfOM removal during SAT and NOM attenuation studies during RBF, it is expected that NOM removal rates during SAT of CAP water are sustainable. The fraction of NOM classified as fulvic/humic acids is expected to increase after SAT of CAP water, possibly leading to greater *specific* reactivity ($\mu\text{g DBPs per mg DOC}$) in the formation of DBPs upon chlorination. Common experiences also suggest there will be an increase in the fraction of brominated species of DBPs, due to the increase in the ratio of Br^-/TOC as NOM is attenuated during SAT without corresponding reduction in Br^- concentration.

General data gaps include the lack of long-term studies on NOM fate and transport during recharge of CAP water in soils and aquifer material and no direct data on NOM biodegradability in CAP water. As such, future field-based work to resolve remaining information gaps should include continued monitoring at CAVSARP for TOC/DOC removal rates along with testing of recovered waters for disinfection by-product reactivity and speciation (i.e. chlorinated versus brominated species of THMs and HAAs). Additional laboratory-based studies should examine the short-term biodegradability of NOM in CAP water (e.g. using 5-day BDOC tests) along with soil column studies to directly determine the relative importance of removal mechanisms (i.e. biodegradation versus sorption) and the effect of using alternative treatment and disinfectant methods.

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