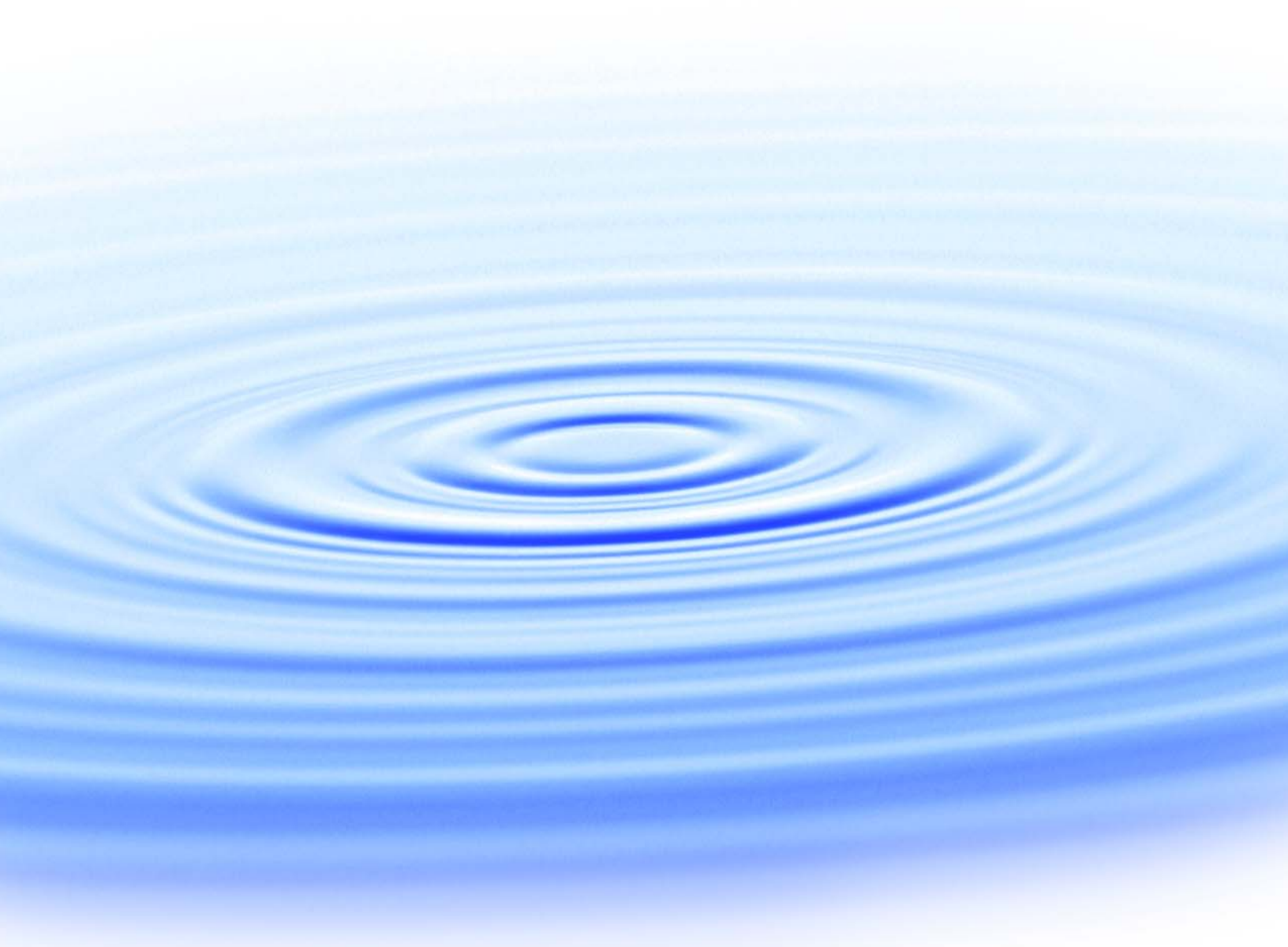
WATEREUSE

Enhancing the Soil Aquifer Treatment Process for Potable Reuse



WateReuse Research Foundation

Enhancing the Soil Aquifer Treatment Process for Potable Reuse

About the WateReuse Research Foundation

The WateReuse Research Foundation builds support for the reclamation, recycling, reuse, and desalination of water through research and education. The Foundation's research advances the science of water reuse and supports communities across the United States and abroad in their efforts to create new sources of high quality water for various uses through reclamation, recycling, reuse, and desalination while protecting public health and the environment.

The Foundation sponsors research on all aspects of water reuse, including emerging chemical contaminants, microbiological agents, treatment technologies, reduction of energy requirements, concentrate management and desalination, public perception and acceptance, economics, and marketing. The Foundation's research informs the public of the safety of reclaimed water and provides water professionals with the tools and knowledge to meet their commitment of providing a reliable, safe product for its intended use.

The Foundation's funding partners include the supporters of the California Direct Potable Reuse Initiative, Water Services Association of Australia, Pentair Foundation, and Bureau of Reclamation. Funding is also provided by the Foundation's members, water and wastewater agencies, and other interested organizations.

Enhancing the Soil Aquifer Treatment Process for Potable Reuse

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Acronyms

3-D	three-dimensional
AOP	advanced oxidation process
BDCM	bromodichloro methane
BDOC	biodegradable dissolved organic carbon
BHA	butylated hydroxyanisole
CEC	contaminant of emerging concern
cfu	colony-forming unit
DBAA	dibromoacetic acid
DBCM	dibromochloro methane
DBP	disinfection byproduct
DCAA	dichloroacetic acid
DDW	Division of Drinking Water (formerly California Department of Public Health)
DEET	*N,N*-diethyl-meta-toluamide
DO	dissolved oxygen
DOC	dissolved organic carbon
DOM	dissolved organic matter
EC	electro-conductivity
EEM	excitation-emission matrix
EfOM	effluent organic matter
EPA	Environmental Protection Agency
GAC	granular activated carbon
gpm	gallons per minute
HAA	haloacetic acid
HRT	hydraulic retention time
LACSD	Los Angeles County Sanitation Districts
MBAA	monobromoacetic acid
MCAA	monochloroacetic acid
ND	non-detect
NDBA	nitrosodi-*n*-butylamine
NDEA	nitrosodiethylamine
NDMA	*N*-nitrosodimethylamine
NDPA	nitrosodi-*n*-propylamine
NMEA	nitrosodimethylamine
NOM	natural organic matter
NPYR	nitrosopyrollidine
NWRI	National Water Research Institute
PFAA	perfluoroalkyl acid
PFBS	perfluorobutane sulfonic acid

PFHxA	perfluorohexanoic acid
PFOA	perfluorooctanoic acid
PFPnA	perfluoropentanoic acid
pfu	plaque-forming unit
PLC	programmable logic controller
PVC	polyvinyl chloride
QA/QC	quality assurance/quality control
qPCR	quantitative polymerase chain reaction
RO	reverse osmosis
rpm	revolutions per minute
SAT	soil aquifer treatment
SBS	sodium bisulfite
SFSG	Santa Fe Spreading Grounds
SJCWWRP	San Jose Creek West Water Reclamation Plant
SUVA	specific ultraviolet absorbance
TBM	bromoform
TCAA	trichloroacetic acid
TCM	chloroform
TCPP	*tris* (chloroisopropyl) phosphate
TDCPP	chlorinated organophosphate
TCEP	*tris* (2-carboxyethyl) phosphine
TOC	total organic carbon
TOrC	trace organic chemicals
TTHM	total trihalomethane
TTHMFP	TTHM formation potential
USGS	United States Geological Survey
UVA	ultraviolet absorbance
UV/H_2O_2	ultraviolet light/hydrogen peroxide
UVT	ultraviolet transmittance
WRRF	WateReuse Research Foundation
WW	wastewater

Foreword

The WateReuse Research Foundation, a nonprofit corporation, sponsors research that advances the science of water reclamation, recycling, reuse, and desalination. The Foundation funds projects that meet the water reuse and desalination research needs of water and wastewater agencies and the public. The goal of the Foundation's research is to ensure that water reuse and desalination projects provide sustainable sources of high-quality water, protect public health, and improve the environment.

An Operating Plan guides the Foundation's research program. Under the plan, a research agenda of high-priority topics is maintained. The agenda is developed in cooperation with the water reuse and desalination communities including water professionals, academics, and Foundation subscribers. The Foundation's research focuses on a broad range of water reuse and desalination research topics including:

- Defining and addressing emerging contaminants, including chemicals and pathogens
- Determining effective and efficient treatment technologies to create 'fit for purpose' water
- Understanding public perceptions and increasing acceptance of water reuse
- Enhancing management practices related to direct and indirect potable reuse
- Managing concentrate resulting from desalination and potable reuse operations
- Demonstrating the feasibility and safety of direct potable reuse

The Operating Plan outlines the role of the Foundation's Research Advisory Committee (RAC), Project Advisory Committees (PACs), and Foundation staff. The RAC sets priorities, recommends projects for funding, and provides advice and recommendations on the Foundation's research agenda and other related efforts. PACs are convened for each project to provide technical review and oversight. The Foundation's RAC and PACs consist of experts in their fields and provide the Foundation with an independent review, which ensures the credibility of the Foundation's research results. The Foundation's Project Managers facilitate the efforts of the RAC and PACs and provide overall management of projects.

Soil aquifer treatment (SAT) is a long-established potable reuse strategy, whereas ozonation is less frequently applied for potable water reuse. This project evaluated the benefits of substituting ozonation for chlorination prior to SAT as a disinfection strategy. Independent of the disinfection strategy, SAT is an effective natural process for attenuating total organic carbon (TOC), the excitation-emission matrix (EEM) fingerprint characteristic of wastewater effluent organic matter (EfOM), disinfection byproducts (DBPs), contaminants of emerging concern (CECs), and microbes (MS-2 virus, coliform bacteria, and *Cryptosporidium* protozoa). Ozonation prior to SAT enhanced SAT performance as seen through improved ultraviolet transmittance (UVT) and wastewater fingerprint and the attenuation of TOC to levels that surpassed those observed with chlorination prior to SAT.

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Executive Summary

Soil aquifer treatment (SAT) is a long-established potable reuse strategy, whereas ozonation is less frequently applied for potable water reuse. This project evaluated the benefits of substituting ozonation for chlorination prior to SAT as a disinfection strategy. The combination of ozonation and SAT has the potential to afford disinfection of a broader variety of pathogens, greater removal of bulk organic matter [e.g., total organic carbon (TOC), as measured by ultraviolet absorbance (UVA) and emissions-excitation matrices (EEMs)], and the removal of a wider variety of contaminants of emerging concern (CECs) while producing water with similar characteristics to those of natural raw waters.

For this study, two Soil Column Systems were tested. One was fed tertiary effluent disinfected with chlorine (Cl_2 Soil Column System) and the other was fed ozonated secondary effluent (O_3 Soil Column System). Each Soil Column System consisted of two soil columns in series. The first column represented the application area of a groundwater recharge basin and the unsaturated zone beneath it. The second column was designed to simulate the behavior in the saturated zone beneath the water table. The total estimated travel time for each Soil Column System was 30 days. The removal of a number of contaminants was examined (e.g., bulk organic matter, CECs, and disinfection byproducts [DBPs] present in the recycled water, as well as spiked MS-2 virus, coliform bacteria, and *Cryptosporidium* protozoa).

Within the context of a 30-day subsurface retention time, bulk organic matter was substantially attenuated through SAT. On average, the Cl_2 Soil Column System reduced TOC from 5.9 to 3.3 mg/L and the O_3 Soil Column System from 5.5 to 2.1 mg/L. On average, the Cl_2 Soil Column System increased ultraviolet transmittance (UVT) from 77 to 86% and the O_3 Soil Column System from 87 to 92%. The ozone disinfection step alone significantly increased UVT from 78 to 87%.

Within the Cl_2 Soil Column System, total trihalomethanes (TTHMs) and haloacetic acids (HAAs) were attenuated to non-detect (ND) levels during all sampling events (reduced on average from 25 to <0.5 ng/L and from 27 to <1.0 ng/L, respectively). Within the O_3 Soil Column System, bromate was reduced to ND levels during all sampling events (reduced on average from 2.5 to <0.5 ng/L). In addition, both systems consistently degraded NDMA to concentrations below the detection level (Cl_2: 448 to < 2 ng/L and O_3: 384 to < 2 ng/L).

Of the 42 CECs tested, 38 CECs were present in the secondary wastewater. Atorvastatin, bisphenol A, caffeine, and diazepam were not detected. Chlorine disinfection did not further reduce any CECs completely. The Cl_2 Soil Column System resulted in the complete removal of 15 CECs (acetaminophen, atenolol, azithromycin, erythromycin, fluoxetine, galaxolide, ibuprofen, metoprolol, naproxen, octylphenol diethoxylate, propranolol, tonalide, triclocarban, triclosan, and trimethoprim). Ozone disinfection removed 18 CECs completely (acetaminophen, atenolol, azithromycin, carbamazepine, diclofenac, erythromycin, fluoxetine, furosemide, gemfibrozil, ibuprofen, naproxen, octylphenol monoethoxylate, octylphenol diethoxylate, propranolol, tonalide, triclocarban, triclosan, and trimethoprim). The O_3 Soil Column System resulted in the complete removal of 3 additional CECs (galaxolide, iopromide, and metoprolol). The efficacy of the O_3 Soil Column System is

driven by the ozone disinfection step. Ozonation reduced some CECs, namely carbamazepine, dilantin, fipronil, meprobamate, and primidone, which would otherwise be recalcitrant through the biological processes in SAT within the 30-day subsurface retention time investigated. Sucralose and TCEP were recalcitrant through both SAT systems and could serve as intrinsic tracers to track the recycled water. For the secondary wastewater tested, sucralose is especially interesting because it is consistently present at high concentrations (around 30,000 ng/L) and has a sufficiently low detection limit (40 ng/L).

Under conditions of saturated flow and a 30-day retention time, spiked microbes were reduced through SAT to below detection levels. These results suggest removals greater than 8.1 log for MS-2 virus, 7.8 log for coliform bacteria, and 9.5 log for *Cryptosporidium* oocysts.

Overall, these findings, along with others from this study, indicate that SAT is an excellent, effective, natural treatment option for the attenuation of TOC, UVA, wastewater fingerprint, DBPs, CECs, MS-2 virus, coliform bacteria, and *Cryptosporidium* protozoa. The use of ozonation as a disinfection alternative prior to SAT, as compared with chlorination, proved to be universally beneficial, with improved removal of all CECs tested, a cleaner water fingerprint via EEM, and enhanced removal of TOC.

Chapter 1

Introduction

1.1 Background

Recycled water use is expanding throughout the world as communities augment their existing water supplies with this drought-proof alternative. One of the most economic and valuable means of utilizing recycled water is through potable reuse projects. Potable reuse projects supplement the potable water supply with highly treated recycled water. California is a leader in potable reuse projects with large projects implementing reverse osmosis-based (RO-based) full advanced treatment, which is an extremely effective treatment train consisting of microfiltration (MF) or ultrafiltration (UF), RO, and an advanced oxidation process (AOP) such as ultraviolet light/hydrogen peroxide (UV/H_2O_2). However, other examples of successful large-scale potable reuse projects are harnessing the power of natural attenuation processes by discharging recycled water (filtered and disinfected) into spreading basins as a means of managed aquifer recharge. The recycled water in the spreading basin then undergoes a natural process called soil aquifer treatment (SAT), as depicted in Figure 1.1 (Drewes and Jekel, 1996; Drewes and Fox, 1999).

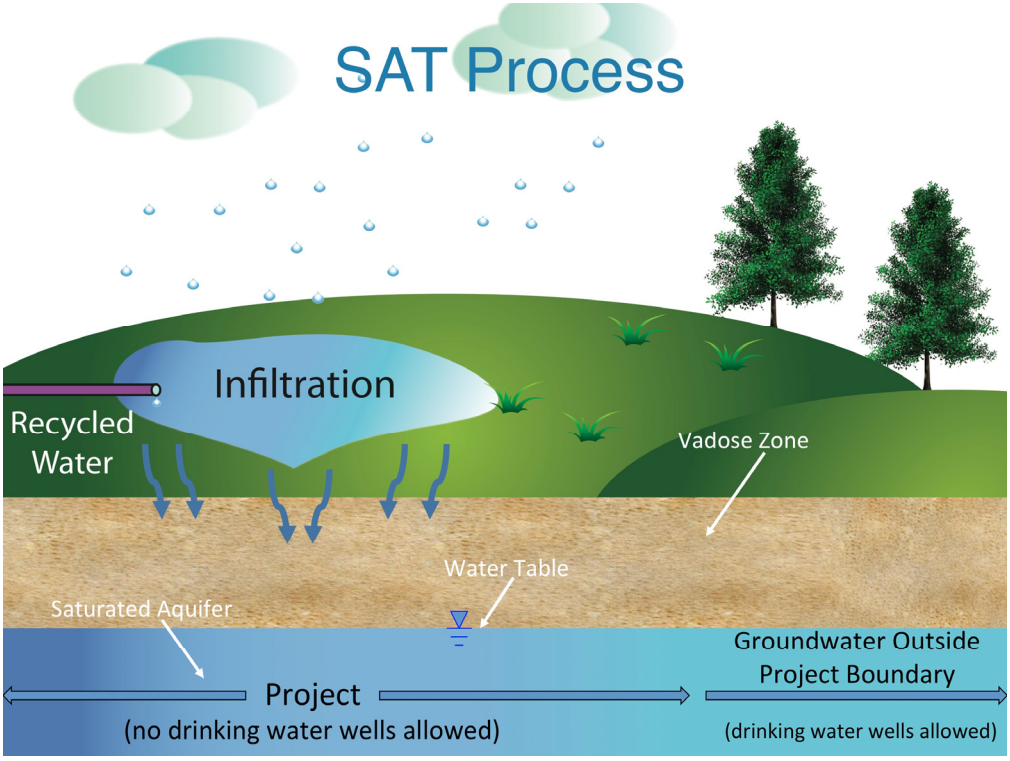

Figure 1.1. The SAT process.

SAT is a long-established potable reuse strategy. For example, SAT has been practiced successfully at the Montebello Forebay in Southern California for more than 5 decades. Montebello Forebay spreading basins are shown in Figure 1.2.

Figure 1.2. Montebello Forebay Rio Hondo Coastal Spreading Grounds.

SAT naturally provides an additional barrier for pathogens and for manmade organic matter and contaminants of emerging concern (CECs) without the additional energy consumption and brine waste stream that come with RO-based treatment trains, thus making it an attractive process for inland communities. The simplicity and effectiveness of SAT has allowed this process to become the most economical potable reuse option when a suitable aquifer is available. In general, SAT removes organic matter and CECs primarily through biodegradation as water percolates through the soil, with filtration, adsorption, and redox reactions providing additional treatment. Although recent studies have shown that a vast majority of CECs are removed by SAT, and extensive public health studies have shown it to be safe (Sloss et al., 1999; Robeck, 1987), SAT has encountered resistance in the past from the public, who often perceive SAT as insufficiently advanced technology compared with the technology required for potable reuse. The San Fernando and San Gabriel Valley areas of California both experienced failures of projects in the 1990s because of resistance by the public. Ozonation is an advanced technology with features that make it an attractive companion to SAT in potable reuse.

1.2 Literature Review

This section consists of the literature review and outlines the current state of knowledge in relation to SAT performance and the potential benefits of ozonation prior to SAT. The literature review is divided into the following subsections:

- attenuation of bulk organic matter through SAT;
- attenuation of CECs through SAT;
- attenuation of microorganisms through SAT;
- potential enhancement of organic carbon attenuation with ozonation prior to SAT; and
- laboratory-scale simulations of SAT.

1.2.1 Attenuation of Bulk Organic Matter through SAT

Recent studies conducted by the WateReuse Research Foundation (WRRF; WRRF-10-10 by Hogg et al., 2013; WRRF-05-04 by Drewes et al., 2011) and other funding entities provide insights into specific mechanisms of contaminant removal and conditions that influence removal. Engineering factors that affect SAT performance include pretreatment, site characteristics that impact travel time, and operating conditions. SAT is ideal for semiarid areas with a groundwater basin composed of permeable, free-draining soil (Fox et al., 2006).

Several field- and laboratory-scale studies have demonstrated SAT to be a robust process. Data from several studies support the fact that dissolved organic carbon (DOC) removal is minimally impacted by aquifer material, infiltration rates, vadose zone depth, and recycled water quality (tertiary verses secondary) but significantly impacted by travel time (Drewes et al., 2011; Laws et al., 2011; Rauch-Williams et al., 2010; Amy and Drewes, 2007; Drewes et al., 2006; Fox et al., 2006; Mansell and Drewes, 2004; Rauch and Drewes, 2003; Drewes et al., 2001; Drewes and Fox, 2000; Westerhoff and Pinney, 2000; Drewes and Fox, 1999; Drewes and Jekel, 1998; Quanrud et al., 1996; Quanrud et al., 1996b). Several studies supporting these findings are summarized in Appendix A.

SAT performance is optimized when operating with favorable infiltration rates and redox conditions. An effective way to sustain infiltration rates and reintroduce oxygen is to use a cyclic surface application regime. The cyclic program can be optimized to effectively restore infiltration rates, aid with pest control, and re-aerate the vadose zone. Typically, the cycle consists of flooding, draining, and drying, as depicted in Figure 1.3.

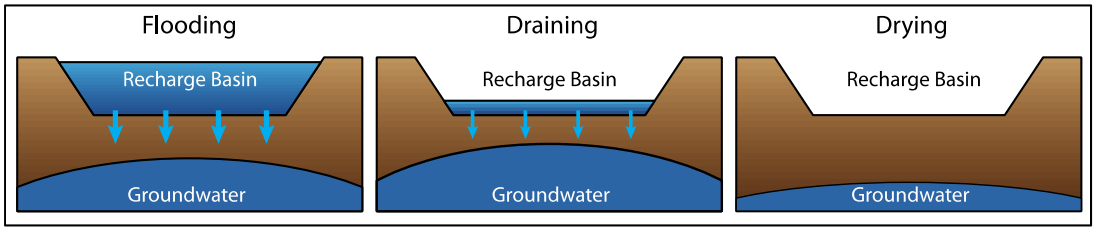

Figure 1.3. Cyclic operation for maintenance of infiltration rates.

Flooding entails filling the basins continuously for 3 to 7 days. Draining entails allowing the water to percolate through the soil column without applying additional water for up to 7 days.

Drying entails a period of 4 to 21 days during which the basin is allowed to dry. A variety of wetting and drying cycles have been tested at a recharge site in Tucson, Arizona (typically operated with 2 days of wetting and 4 days of drying). Research showed that increasing the length of the drying time allowed oxygen to penetrate to greater depths (Fox et al., 2006), which, in turn, promoted oxic conditions for more rapid degradation of organic matter.

Full-, field- and laboratory-scale studies have shown significant removal of bulk organic material [as measured by parameters such as total organic carbon (TOC) and DOC] by SAT within relatively short vadose zone travel times—on the order of days (e.g., Drewes et al., 2006; Drewes and Jekel, 1998; Amy et al., 1996; Wilson et al., 1995). These parameters are indicative of the presence of disinfection byproduct (DBP) precursors and are particularly important to maintain at low levels for prevention of further DBP formation. In addition, SAT has been shown to significantly remove DBPs present in disinfected wastewater effluent, namely total trihalomethanes (TTHMs) and haloacetic acids (HAAs; Amy et al., 1996; Wilson et al., 1995).

The State of the Science Study published by the WRRF (WRRF-11-02 by Trussell et al., 2013) suggested that water for potable use should be free of wastewater properties evident to the informed consumer. To accomplish this, two criteria were proposed for the recycled water:

- it should be free of dissolved organic matter (DOM) of wastewater origin; and
- trace organic chemicals (TOrC) and CECs should be reduced to acceptable levels.

Also suggested in the State of the Science Study was that the "DOM of wastewater origin" criterion could be met either by meeting the current Division of Drinking Water (DDW, formally known as California Department of Public Health) Groundwater Recharge Regulations requirement that TOC of wastewater origin be less than 0.5 mg/L or by transforming the effluent organic matter (EfOM) into a DOM that is more like natural organic matter (NOM). It was suggested that meeting this criterion might be based on a 90% reduction in excitation-emission matrix (EEM) total fluorescence.

EEM fluorescence spectroscopy is one tool that can be used to develop a "fingerprint" of a water sample (Drewes et al., 2011; Laws et al., 2011). With this tool, a range of electromagnetic radiation (typically in the 290–530 nm wavelength range) is projected onto a water sample through a series of incremental pulses, and the resulting fluorescence from each pulse is measured (typically in the 240–450 nm wavelength range). Each pulse, and subsequent fluorescence, makes up the data points on a three-dimensional (3-D) plot. On this plot, the y-axis represents the excitation wavelength, the x-axis represents the corresponding emission wavelength, and the corresponding emission intensity is represented by contours and colors. This type of imaging is referred to as EEM fingerprinting throughout this report. The different regions of an EEM fingerprint represent different types of organics in a given water sample. An example is provided in Figure 1.4 for a secondary wastewater sample, and characteristic regions are highlighted.

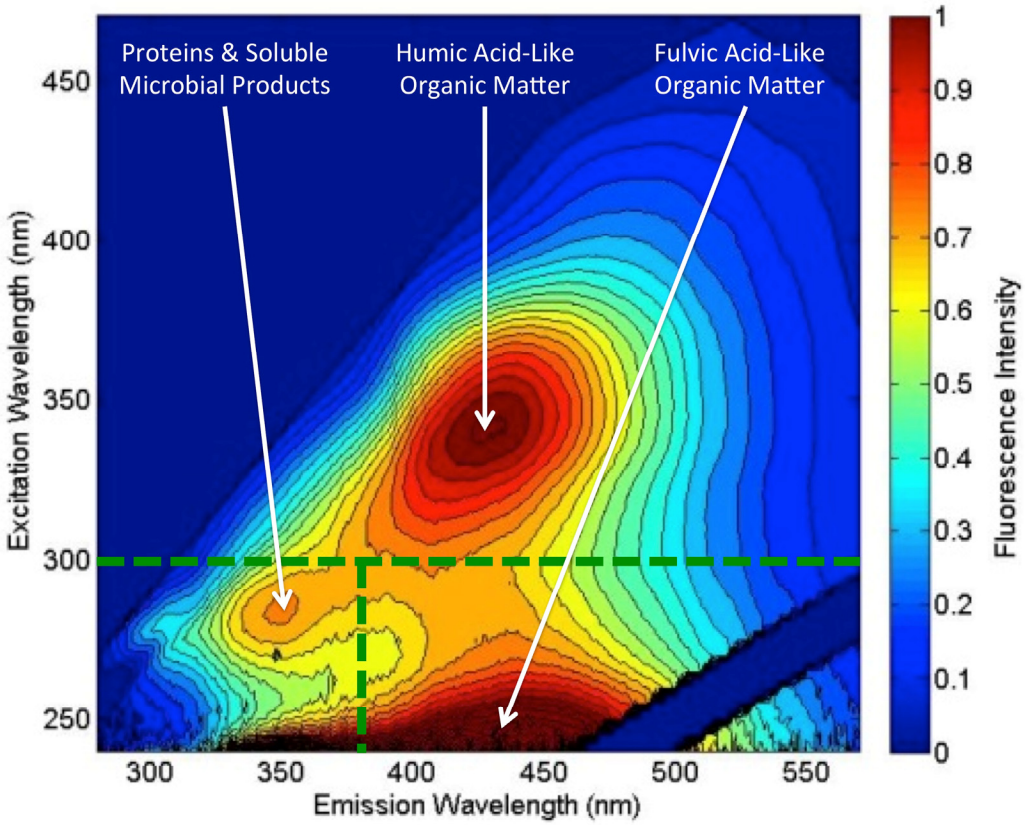

Figure 1.4. An example EEM fingerprint of secondary wastewater with characteristic regions indicated.

Wastewater effluents have been shown to have characteristically high fluorescence intensity (typically shown as red) at the 260 to 290 nm excitation and 320 to 370 emission wavelength ranges (as shown in the lower left region of the EEM fingerprint plot in Figure 1.4). This region of the EEM plot generally has low fluorescence intensity (shown as dark blue) for surface waters not impacted by wastewater effluent (Seong-Nam et al., 2008). EEM fingerprints of recycled water begin to have characteristics similar to those of natural water sources within months after initial surface application (Drewes et al., 2011; Lin et al., 2011). A study conducted at the United States Geological Survey (USGS) Research Basin within the Montebello Forebay Spreading Grounds system examined the effects of subsurface retention time on surface applied tertiary wastewater. Infiltrated water samples were extracted downstream at locations depicted in Figure 1.5. EEM fingerprints were developed for all samples, and the results are show in Figure 1.6. The upper left plot represents the initial condition of the water, and the plots to the right and downward represent subsequent water samples.

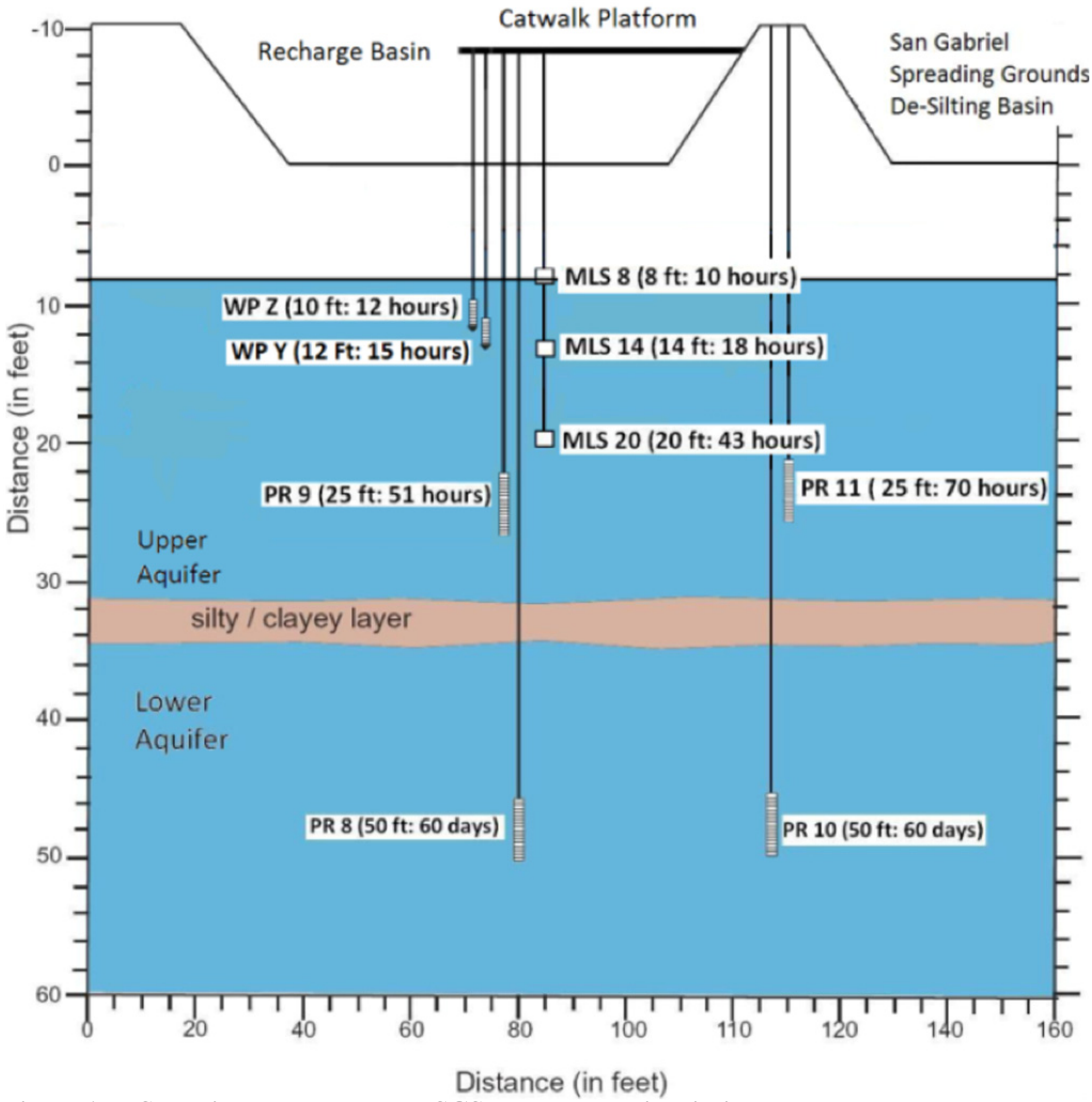

Figure 1.5. Sampling scheme at the USGS Research Basin within the Montebello Forebay Spreading Grounds system—examining the effects of subsurface retention time on surface-applied tertiary wastewater.

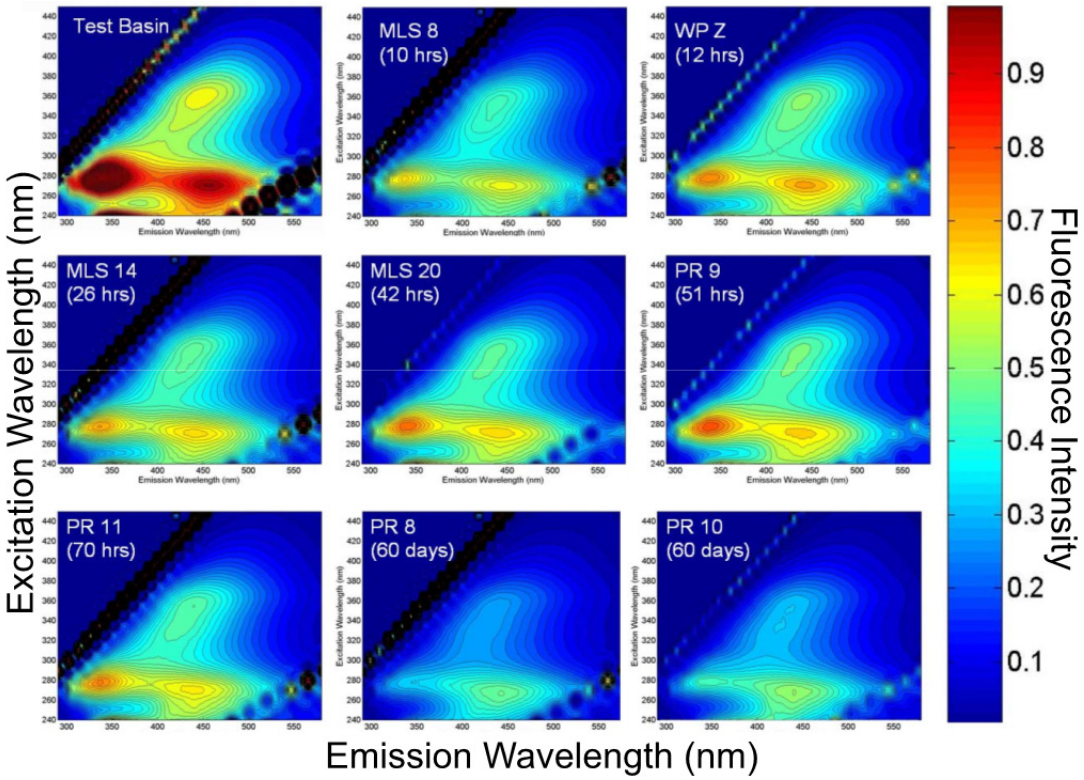

Figure 1.6. EEM fingerprints of tertiary treated wastewater and extracted infiltrated tertiary treated wastewater samples after various travel times (noted in parentheses) at Montebello Forebay Spreading Grounds.

Source: Drewes et al., 2011

1.2.2 Attenuation of CECs through SAT

In the public forum, concern is often expressed over the presence of CECs (e.g., pharmaceuticals, personal care products, and pesticides) existing in drinking water supplies. Wastewater effluents inherently have higher concentrations of these anthropogenic chemicals compared with conventional drinking water sources; therefore, the ability of SAT to remove these constituents is of particular interest in potable reuse applications. Studies have shown that SAT removes a number of the CECs typically found in wastewater effluents (e.g., Drewes et al., 2011). Table 1.1 summarizes the findings from Drewes et al. (2011) on SAT performance for a variety of CECs at several spreading basins. Performance is broken down into three columns based on removal rates. CECs occurring in table cells spanning multiple columns exhibited more variable removal rates compared with those in single-column cells. Note that data for some CECs came exclusively from single locations. The data used to generate Table 1.1 are provided in Appendix B.

Table 1.1. Typical CEC Removal by the SAT Process for Potable Reuse

Excellent Removal (>90%)	Fair Removal (90 to 50%)	Poor Removal (50 to <25%)
Atenolol, Atorvastin, Butylated hydroxyanisole (BHA), Caffeine, Dioctyl phthalate, Enalapril, Fluoxetine, Galaxolide, Nonylphenol, Norfluoxetine, Salicylic acid, Simvastatin hydroxy acid, Trimethoprim		Carbamazepine, Primidone, Chlorinated organophosphate (TDCPP)
Benzophenone, Ibuprofen, *N,N*-Diethyl-meta-toluamide (DEET), Ethylenediaminetetraacetic acid (EDTA), Iopromide, Meprobamate, Sulfamethoxazole		
Diclofenac, Naproxen, Gemfibrozil, Octylphenol, Tonalide, Triclosan		
	Dilantin (Phenytoin), tris (2-carboxyethyl) phosphine (TCEP), tris (chloroisopropyl) phosphate (TCPP)	

Source: Compiled using data from Drewes et al. (2011) for travel times up to 2 weeks

As evident from Table 1.1, several CECs have the potential to be reliably removed through SAT, including atenolol, butylated hydroxyanisole (BHA), caffeine, diclofenac, fluoxetine, gemfibrozil, ibuprofen, naproxen, triclosan, and trimethoprim. Three CECs, namely carbamazepine, primidone, and chlorinated organophosphate (TDCPP), are not substantially attenuated via SAT. In addition, there are several CECs that show variable removal, spanning from <25% to >90%, such as benzophenone, ibuprofen, *N,N*-diethyl-meta-toluamide (DEET), and ethylenediaminetetraacetic acid (EDTA). Recent work at the USGS Research Basin at the Montebello Forebay Spreading Grounds demonstrated that most of the removal by SAT of DOC, biodegradable dissolved organic carbon (BDOC), caffeine, gemfibrozil, and atenolol occurs in the first 10 ft of the vadose zone (Laws et al., 2011).

Despite the high removal efficiencies of SAT for bulk organic material, DBPs, DBP precursors, and pathogens, some CECs are resistant to SAT biodegradation. In addition, data has shown that CECs well removed in one basin may not be well removed in another, as is the case for benzophenone, ibuprofen, and iopromide. Several factors may contribute to poor removal of certain CECs, including chemical form (e.g., biodegradability, hydrophobicity, functional groups, and molecular weight) and variability among and within soil aquifer environments (e.g., soil properties, oxidation reduction potential, travel time, groundwater velocity, hydrodynamic dispersion, and temperature; Drewes et al., 2011). Issues related to the analytical method employed (e.g., matrix interference, susceptibility to contamination during sample collection, and analysis) are another possible source of data variability. In

practice, however, removal of these recalcitrant CECs would require one or several additional treatment processes either before or after SAT. SAT with ozone pretreatment provides a particularly attractive option that allows for the benefits of SAT without the disadvantages of RO-based treatment trains (i.e., higher capital cost and power consumption and the need to dispose of 15% or more of the flow as a brine waste).

1.2.3 Attenuation of Microorganisms through SAT

SAT provides an additional treatment barrier for pathogen removal, particularly for viruses but also for *Cryptosporidium*, the oocysts of which are not easily inactivated by typical disinfection methods. The two mechanisms for removal of *Cryptosporidium* during SAT are physical straining (i.e., filtration) and die-off (i.e., oocyst inactivation over time). Removal of *Cryptosporidium* in groundwater soil systems is fairly well accepted; the Environmental Protection Agency (EPA) allows riverbank filtration to achieve 0.5 log or 1.0 log removal credits to drinking water treatment for aquifers that meet certain physical standards (e.g., unconsolidated sandy aquifers that are setback from the river 25 or 50 ft). One laboratory study using columns with a 1-meter soil depth found *Cryptosporidium* removals of between 3.8 and 4 log (Mays et al., 1998). Field studies of actual *Cryptosporidium* removal are rare, as the actual concentration of oocysts is typically low even in raw surface waters (Trussell et al., 2012). Instead, log removals in the field are typically demonstrated using surrogates, such as microspheres and bacterial spores. For example, Havelaar et al. (1995) found 3.1 and 3.6 log removals of bacterial spores in two riverbank filtration systems for drinking water with 30 and 25 m travel distances, respectively. Schijven et al. (1998) found 1.9 log removal of bacterial spores with just 2 m of travel distance. Santamaria et al. (2011) used laboratory soil columns and spike studies to evaluate the relationship between *Cryptosporidium* removal and travel time in sandy soil and found that log removals increased with travel time, as summarized in Table 1.2.

Table 1.2. *Cryptosporidium* Removal at Three Travel Times

Lab versus Field Scale	Travel Time	*Cryptosporidium* Removal
Lab	10 hours	3 log
Lab	19 hours	4 log
Field	2.4 days	5 log

Embedded within travel time are a number of variables that impact removal, such as aquifer type and infiltration rate. Whereas log removal does increase with travel time, the relationship is not log-linear, leading Santamaria et al. (2011) to hypothesize that variability in the physical properties among the oocysts would lead to potentially greater mobility than predicted by the conventional filtration theory for colloids.

Although not the focus of this study, inactivation also contributes to *Cryptosporidium* attenuation in soil aquifer systems, although generally to a lesser degree than physical removal. A review by Peng et al. (2008) concluded that *Cryptosporidium* inactivation follows a first-order exponential formula: $y_t = y_0 e^{-Kt}$, where y_t is the number of oocysts at time t, y_0 is the number of oocysts at the initial conditions, and K is the inactivation rate coefficient. For soil systems, K was estimated to be 0.011 day^{-1} at 20 °C, corresponding to a time estimate of approximately 210 days for 1 log *Cryptosporidium* inactivation. Inactivation rates are strongly a function of temperature, with warmer temperatures corresponding to increased

inactivation. Soil type (e.g., clay, silt, and sand) also affects inactivation rates, but research is unclear on the exact correlation, and currently it is unknown how concentrations of organic matter, nutrients, and other microorganisms play a role (Peng et al., 2008). Research has demonstrated a lack of knowledge regarding *Cryptosporidium* removal and inactivation in SAT systems for potable reuse. Inactivation of *Cryptosporidium* is generally viewed to be minor compared with physical removal (Nasser et al., 2003), and the associated time scales for the desired inactivation are relatively long (Peng et al., 2008).

WRRF-10-10 (Hogg et al., 2013) explored SAT filtration and disinfection compliance for the disposal of unfiltered secondary effluent into the ground under waste discharge requirements in Fresno and Dinuba, CA. The subsurface retention time was not specified at the two sites. Through field-testing, it was shown that both sites were capable of achieving California Code of Regulations Title 22 tertiary recycled water filtration and disinfection criteria that focus on removal of bacteria and viruses. The Fresno-Clovis Regional Wastewater Reclamation Facility spreading operation achieved more than 7 log removal of MS-2 coliphage and 5.6 log removal of total coliform. The Dinuba Wastewater Reclamation Facility spreading operation achieved approximately 5 log removal of MS-2 coliphage and 4.4 log removal of total coliform. Both sites demonstrated post-SAT turbidity of less than 2 NTU and ultraviolet transmittances (UVTs) greater than 87%. As part of a different study, 7 log attenuation of indigenous coliphage was estimated through 100 ft of subsurface travel in field-scale studies (Fox et al., 2006). Using lab-scale soil columns operated with 7-day wetting and 7-day drying cycles, Quanrud et al. (2003) demonstrated more efficient coliphage removal in finer grained soils (93%) than in coarse sand (76%). A relationship between percolation rate and virus removal was also shown; detention times of 5 and 20 h in a 1 m soil column resulted in removals of 70 and 99%, respectively (Quanrud et al., 2003).

1.2.4 Potential Enhancement of Organic Matter Attenuation with Ozonation Prior to SAT

In addition to disinfection, ozone as a pretreatment technology to SAT offers several benefits for the enhancement of CEC attenuation. Ozone transforms organic matter in wastewater by making it more amenable to biodegradation. This biodegradation supports SAT's primary removal mechanism for CECs, which is thought to be co-metabolism (Fox et al., 2006; Drewes et al., 2001b; Drewes and Jekel, 1998). Ozone alone also removes many CECs through oxidation, both directly and through the formation of hydroxyl radicals (Pocostales et al., 2010; Gerrity et al., 2011; Gerrity and Snyder, 2011; Pisarenko et al., 2012). Other AOPs such as UV/H_2O_2 have similar treatment benefits. However, ozone has a cost advantage, as detailed in WRRF Project 02-009, because it operates both through direct oxidation and through hydroxyl radicals formed by the action of ozone with natural promoters present in wastewater. Hydroxyl radical formation with ozone in the presence of high TOC waters with poor UVT is less expensive than alternative AOPs (Bennoti et al., 2009; Gerrity et al., 2010; Pisarenko et al., 2012; WRRF-02-009). These conclusions were supported by the work of Swiss researchers who studied three lake waters and a wastewater effluent and showed that CEC removal by ozone oxidation is more energy efficient than CEC removal by UV/H_2O_2 (Katsoyiannis et al., 2011). In a recently completed WRRF project, the use of ozone to remove CECs from surface water and wastewater effluent before RO treatment was shown to be more efficient than the use of UV/H_2O_2 because of lower UVT in ozonated waters (WRRF-08-08, Stanford et al., 2013). Along with other results cited earlier, these results suggest that ozonation is a superior pretreatment for SAT. Table 1.3 shows the same data as Table 1.1 with strikethroughs that indicate CEC removal of 70% or greater through ozonation alone at a dose of approximately 0.5 to 1.0 mg O_3/mg DOC.

Table 1.3. Typical CEC Removal by the SAT Process for Potable Reuse and Removal Potential through Ozonation[1]

Excellent Removal (>90%)	Fair Removal (90 to 50%)	Poor Removal (50 to <25%)
~~Atenolol, Atorvastin, BHA, Caffeine,~~ *Dioctyl phthalate*, *Enalapril*, ~~Fluoxetine, Galaxolide, Nonylphenol,~~ Norfluoxetine, *Salicylic acid*, *Simvastatin* hydroxy acid, ~~Trimethoprim~~		~~Carbamazepine, Primidone,~~ TDCPP
~~Benzophenone, Ibuprofen, DEET,~~ EDTA, ~~Iopromide, Meprobamate, Sulfamethoxazole~~		
~~Diclofenac, Naproxen, Gemfibrozil, Octylphenol,~~ Tonalide, ~~Triclosan~~		
	~~Dilantin (Phenytoin),~~ TCEP, TCPP	

Notes: [1]Compiled using data from Trussell Technologies, 2011; Lee et al., 2012; Reungoat et al., 2012; U.S. EPA, 2010; Hollender et al., 2009; Wert et al., 2009; Yue et al., 2009; Nakada et al., 2007; Huber et al., 2005

[2]Text with strikethrough indicates 70% or greater removal potential by ozone; text without strikethrough indicates <70% removal efficacy; italicized text indicates unknown O_3 removal efficacy

Several of the CECs removed by ozone are recalcitrant or exhibit variable removal with SAT alone, indicating that ozone has the potential to supplement CEC removal by SAT. Notably, some compounds that are recalcitrant to SAT removal (e.g., carbamazepine and sulfamethoxazole) show excellent removal by ozone, to levels below their method reporting limits (Lee et al., 2012; Reungoat et al., 2012; Trussell Technologies, 2011; U.S. EPA, 2010; Hollender et al., 2009; Wert et al., 2009; Yue et al., 2009; Huber et al., 2005). Particularly interesting are the results of a recent study by Trussell Technologies (2011), which suggest that sucralose, a compound of well-known resistance to biodegradation, can be significantly reduced through high levels of ozonation. Of the 30 CECs in Table 1.3 for which removal data have been reported for both SAT and ozone, 10 can reliably be reduced by 90% or more using SAT alone, and 11 additional CECs have the potential to be reduced by 70% or more by ozonation alone at approximately 0.5 to 1.0 mg O_3/mg DOC (e.g., carbamazepine, primidone, DEET, dilantin, and sulfamethoxazole).

Historically, bromate formation has been the most significant concern related to the use of ozone in water treatment (Orlandini et al., 1997; Wert et al., 2007; Snyder et al., in press); however, more recent studies have also shown potential for the formation of *N*-nitrosodimethylamine (NDMA) and perfluoroalkyl acids (PFAAs) during the ozonation of wastewater (Hollender et al., 2009; Pisarenko et al., 2012; Snyder et al., in press; Dickenson et al., in press). In the case of NDMA, formation varies in different wastewaters and under

different ozone dose conditions, and the formation of NDMA may necessitate post-mitigation measures (Snyder et al., in press; Dickenson et al., in press). Drewes et al. (2006) and Nalinakumari et al. (2010) showed that SAT could be effective to remove NDMA. In the case of PFAAs, a recently completed study by Dickenson et al. (in press) that surveyed full-scale wastewater facilities that use ozone found that the extent of PFAA formation varied among wastewaters but commonly included perfluoropentanoic acid (PFPnA), perfluorohexanoic acid (PFHxA), perfluorooctanoic acid (PFOA), and perfluorobutane sulfonic acid (PFBS). The most consistently formed PFAA was PFHxA with a maximum increase in concentration up to 11 ng/L at a dose of 1.0 mg O_3/mg TOC. (Further increases in ozone dose did not result in higher formation.) The U.S. EPA provisional short-term health advisories in 2009 for the two predominant PFAAs (PFOA and PFOS) were set at 400 ng/L and 200 ng/L, respectively. The potential for forming perfluorinated substances at these rates by ozone is relatively low.

Although extensive studies have been done on the combination of ozone and granular activated carbon (GAC), relatively few have been done with ozone and inert materials. Past studies of wastewater treatment by ozone followed by biological filtration, which may be viewed as analogous to SAT with preozonation, showed increased removal of CECs during biological filtration (e.g., Reungoat et al., 2012; Hollender et al., 2009). In addition, lab-scale studies of SAT systems (i.e., soil columns) with preozonation, as compared with SAT alone, have shown overall greater removal of DOC because of the increase of organic matter biodegradability (i.e., BDOC fraction). For example, Amy et al. (1996) found that BDOC increased from approximately 54 to 65% with ozone doses of approximately 1.6 mg O_3/mg DOC, as compared non-ozonated secondary effluent. In addition, DBP formation was lower in recirculating soil columns with ozone pretreatment than in those without (Amy et al., 1996).

Research by Drewes and Jekel (1998) investigated how ozone changed the biodegradability of DOC through SAT. Without ozone, DOC was 20% biodegradable (removed within 0–3 days), 14% poorly biodegradable (removed within 3–16 days), and 66% recalcitrant. With a dose of 0.86 mg O_3/mg DOC, DOC was 47% biodegradable, 6% poorly biodegradable, and 47% recalcitrant. This demonstrates that ozone coupled with SAT improves overall removal of DOC by approximately 20% by making DOC more amenable to biodegradation through SAT.

Linlin et al. (2011) studied ozonated (0.6 to 1.0 mg O_3/mg DOC) tertiary water, followed by short-term, unsaturated flow SAT (hydraulic retention time[(HRT] of 14 h) and long-term, saturated flow SAT (HRT of 20 days). The authors reported significant reductions in both DOC and TTHM formation potential (TTHMFP) using this treatment strategy, as summarized in Table 1.4.

Table 1.4. DOC and TTHMFP Reductions in Tertiary Water Treated with Ozone, Unsaturated Flow SAT, and Saturated Flow SAT

Sample Location	DOC, mg/L	TTHMFP, µg/L
Tertiary	6.5	268
Post-ozonation	6.4	224
Post-unsaturated column	2.6	126
Post-saturated column	1.6	105

In addition to removing CECs, ozone effectively eliminates the typical EEM wastewater fingerprint (introduced at the end of Section 1.2.1), changing it to a level comparable to raw drinking water, as shown in Figure 1.7. Linlin et al. (2011) and Drewes et al. (2006) found that the residual organic matter remaining after SAT had characteristics very similar to those of natural water sources, although some EfOM was still distinguishable. Ozone with SAT has the potential to further reduce this residual EfOM by transforming it into NOM, further enhancing the public's perception of SAT.

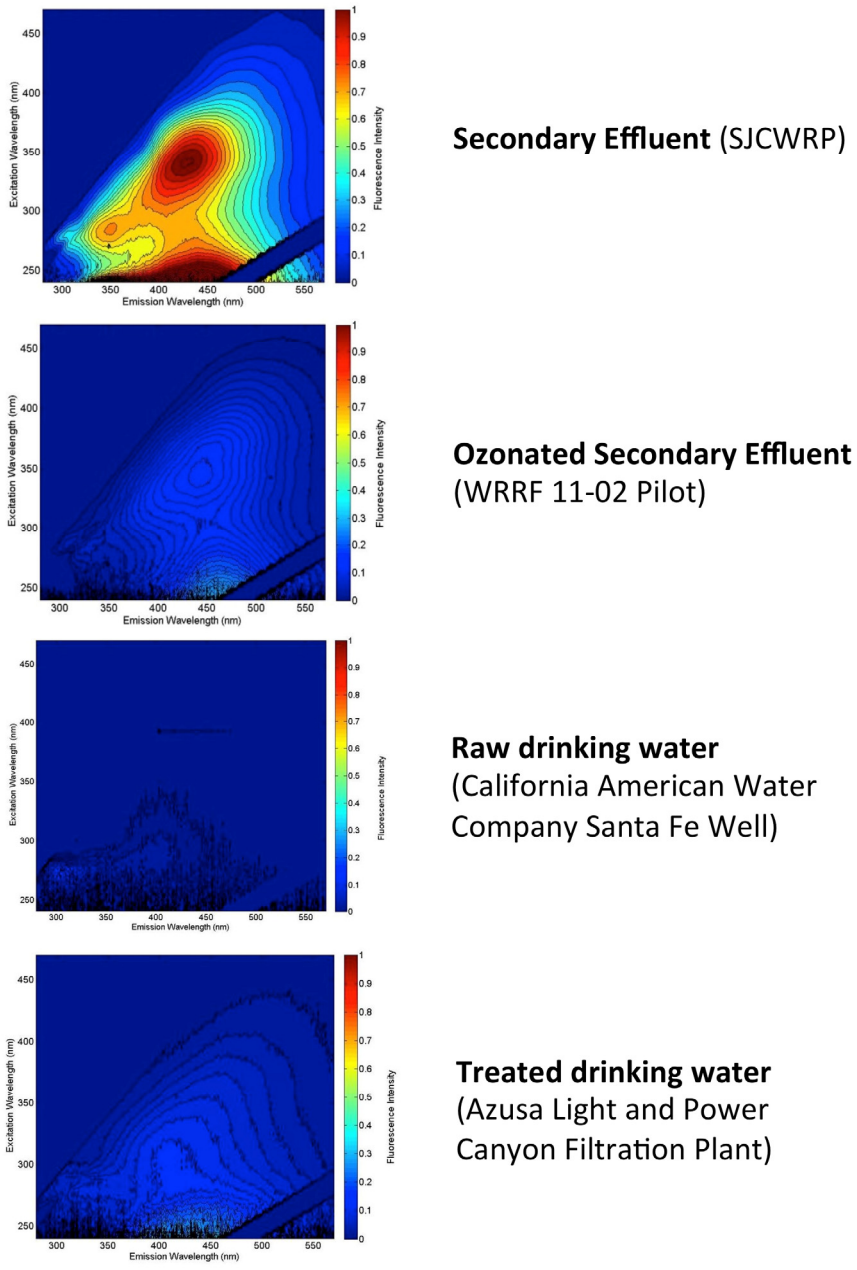

Figure 1.7. Ozone eliminates the wastewater fingerprint and results in a plot that approaches that of raw drinking water.

Note: SJCWWRP = San Jose Creek West Water Reclamation Plant

1.2.5 Laboratory-Scale Simulations of SAT

Various soil column studies have been conducted under controlled laboratory conditions in an effort to better understand the potential of SAT to remove organic matter, nutrients, CECs, and pathogens. Previous studies have varied in terms of column size, soil material, feed water quality, operations, and maturation period. The details of soil column configurations used in the past for groundwater recharge reuse applications with tertiary/secondary wastewater are

provided in Appendix C and summarized as follows (Drewes et al., 2011; Pavelic et al., 2011; Rauch-Williams et al., 2010; Debroux and Drewes, 2007; Fox et al., 2006; Gungor and Unlu, 2005; Cordy et al., 2004; Mansell and Drewes, 2004; Rauch and Drewes, 2004; Quanrud et al., 2003; Drewes et al., 2001; Drewes et al., 2001b; Drewes and Fox, 2000; Westerhoff and Pinney, 2000; Drewes and Fox, 1999; Drewes and Jekel, 1996; Kopchynski et al., 1996; Quanrud et al., 1996; Quanrud et al., 1996b):

- The configuration utilized an upflow regime.
- The configuration utilized column diameters ranging from 8 to 32 cm, with 14 cm being most common.
- The configuration utilized column heights ranging from 1 to 5 m, with 4 m being most common.
- For a column system to resemble field conditions, the system biology must be developed and acclimated to the feed. To achieve a representative biology within the soil column, most studies used an acclimation period of 1 to 4 months for systems operated under saturated flow, continuous feed conditions. Acclimation typically entailed continuous feed of secondary or tertiary effluent with the ultimate goal to achieve steady-state DOC removal through the column or DOC concentration in the column effluent. Studies using intermittent, cyclic feed have shown similar acclimation periods. Two studies, however, supported the need for longer acclimation periods: (1) Drewes et al. (2006) operated parallel anoxic columns that were continuously fed, but one had been acclimated for a duration on the order of months and the other on the order of years. The longer acclimation period resulted in more effective DOC and NDMA reductions. (2) Drewes and Jekel (1998) found that DOC removal improved for the first 13 months of operation. However, this may be attributed to the use of technical sand rather than aquifer material.
- The soil was sourced from recharge sites, from riverbeds, or, less commonly, from sand quarries. Multiple authors reported sieving out material greater than 2 mm.
- Soil columns reported in the literature were operated under a variety of conditions: anoxic versus aerobic, saturated versus unsaturated, and continuous versus cyclic feed.

1.3 Research Needs

Ozonation is an advanced technology that has features that make it an attractive companion to SAT in potable reuse, namely its reputation as a powerful disinfectant, its effectiveness in oxidizing TOrC that are not easily removed by SAT alone, and the way in which it transforms EfOM, reducing its color and making it more amenable to removal by downstream biological processes, such as SAT. Whereas significant work has been done with ozonation in conjunction with GAC, limited studies are available evaluating ozonation as compared with chlorination. Therefore, this study compares SAT performance following these two disinfection alternatives (i.e., Cl_2-SAT and O_3-SAT) on bulk organic carbon, CECs, and DBPs. This project also attempts to document pathogen removal as measured by levels of MS-2 virus, coliform bacteria, and *Cryptosporidium* protozoa.

1.4 Project Objectives

In association with the research needs identified in Section 1.3, this project has the following objectives:

- characterize water samples after each step during Cl_2-SAT and O_3-SAT processes by measuring bulk organic carbon (e.g., UVT and TOC);

- characterize the wastewater "fingerprint" for both disinfection alternatives using ultraviolet absorbance (UVA) and EEM analyses;
- characterize the efficacy of DBP attenuation (e.g., bromate, NDMA, TTHMs, and HAAs) through SAT;
- measure the removal of a suite of CECs, including those known to be recalcitrant to SAT but amenable to ozonation (e.g., carbamazepine, dilantin, meprobamate, primidone, and sulfamethoxazole); and
- determine the efficacy of MS-2 virus, coliform bacteria, and *Cryptosporidium* attenuation through SAT.

Chapter 2

Experimental Approach

2.1 Research Approach

The Soil Column Systems, analytical methods, and experimental plan used to investigate the potential enhancement of SAT with ozonation are described in this chapter. This study consisted of three phases:

- Phase 1: Design and construction of two Soil Column Systems, each system consisting of two columns to compare SAT performance with two different feed sources (chlorinated-disinfected tertiary effluent and ozonated secondary effluent); acclimation of all four columns to establish the microbial population; and tracer testing to evaluate hydraulics;

- Phase 2: Evaluation of DBPs related to each disinfection alternative as well as evaluation of a suite of CECs using ambient concentrations in the source waters being treated; and

- Phase 3: Microbe challenges to explore the attenuation of spiked virus (MS-2 coliphage), bacteria (E. coli), and protozoa (*Cryptosporidium* oocysts).

Each phase and the related analytical methods are detailed in the following sections.

2.2 Phase 1: Design and Construction of Soil Column Systems

The design and operation of the columns for this study were developed based on past published research (as described in Section 1.2.5), as well as on advice from Professor Jörg Drewes, who has more than 2 decades of experience with both field and column studies. The overall process flow diagram and Soil Column Systems are depicted in Figure 2.1. This study included two Soil Column Systems: the Cl_2 Soil Column System and the O_3 Soil Column System. Each system consisted of two columns in series. The first column in each system (Cl_2 Column #1 and O_3 Column #1) represented the application area of a groundwater recharge basin and the unsaturated zone beneath it. Cl_2 Column #1 and O_3 Column #1 were fed their respective source waters from the top of the column (downflow regime). These columns were designed to operate downflow to simulate the vadose zone. A flow control device was used to maintain the target residence time and soil submergence. This device consisted of a tee at the effluent side that was raised and mounted at a level that allowed approximately 1 in. of submergence. To simulate the operations of the spreading basins in the field, the columns were periodically taken offline to drain and dry.

The second column in each series (Cl_2 Column #2 and O_3 Column #2) was designed to simulate the behavior in the saturated zone in the water table beneath the infiltration basin. These columns were designed to operate upflow, following the practice of earlier work by Professor Drewes and others. The filtrate from Cl_2 Column #1 and O_3 Column #1 was collected in a reservoir and pumped into the bottom of Cl_2 Column #2 and O_3 Column #2 (upflow regime), respectively.

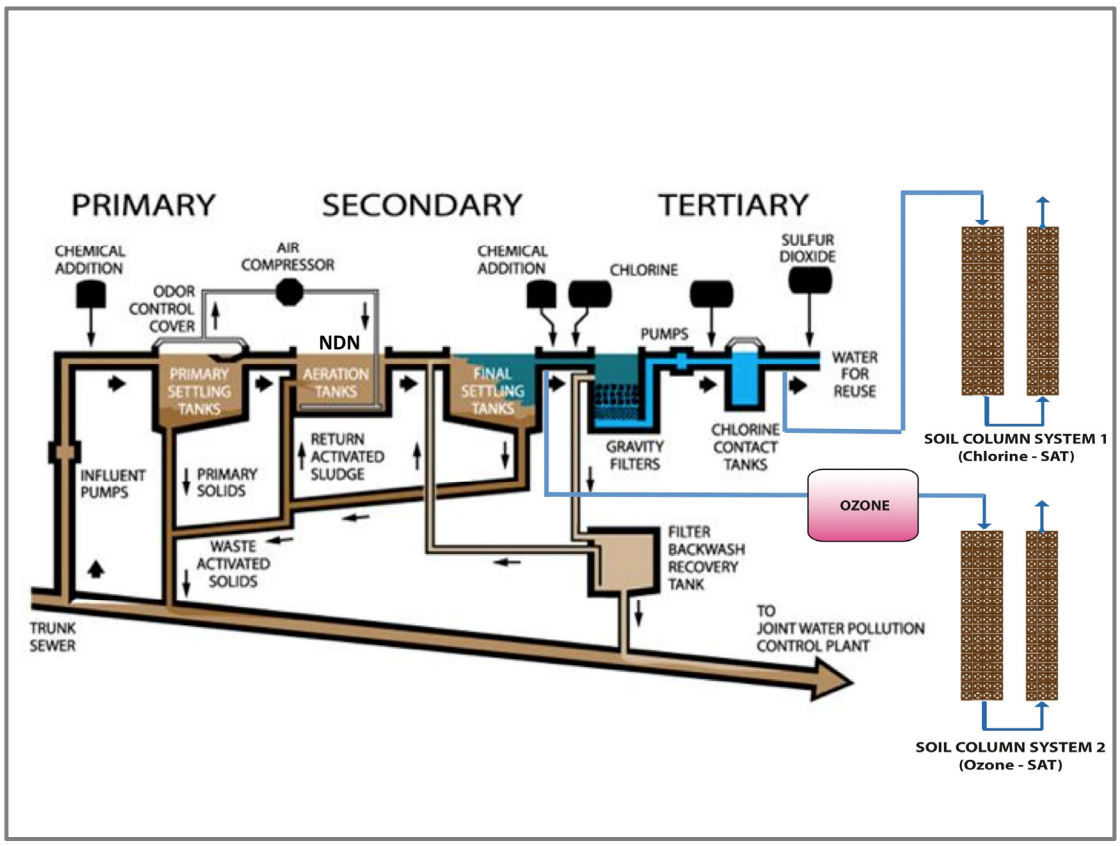

Figure 2.1. Process flow diagram showing the source waters and Soil Column Systems for this project.

2.2.1 Soil Column System Configuration

Cl_2 Column #1 and O_3 Column #1 were 12 ft tall and constructed of 8 in. (outer diameter: 8.6 in.; inner diameter: 7.6 in.) clear polyvinyl chloride (PVC) pipe. Cl_2 Column #2 and O_3 Column #2 were also 12 ft tall clear PVC pipe but of a smaller diameter, 6 in. (outer diameter: 6.6 in.; inner diameter: 6.0 in.; see Figure 2.2).

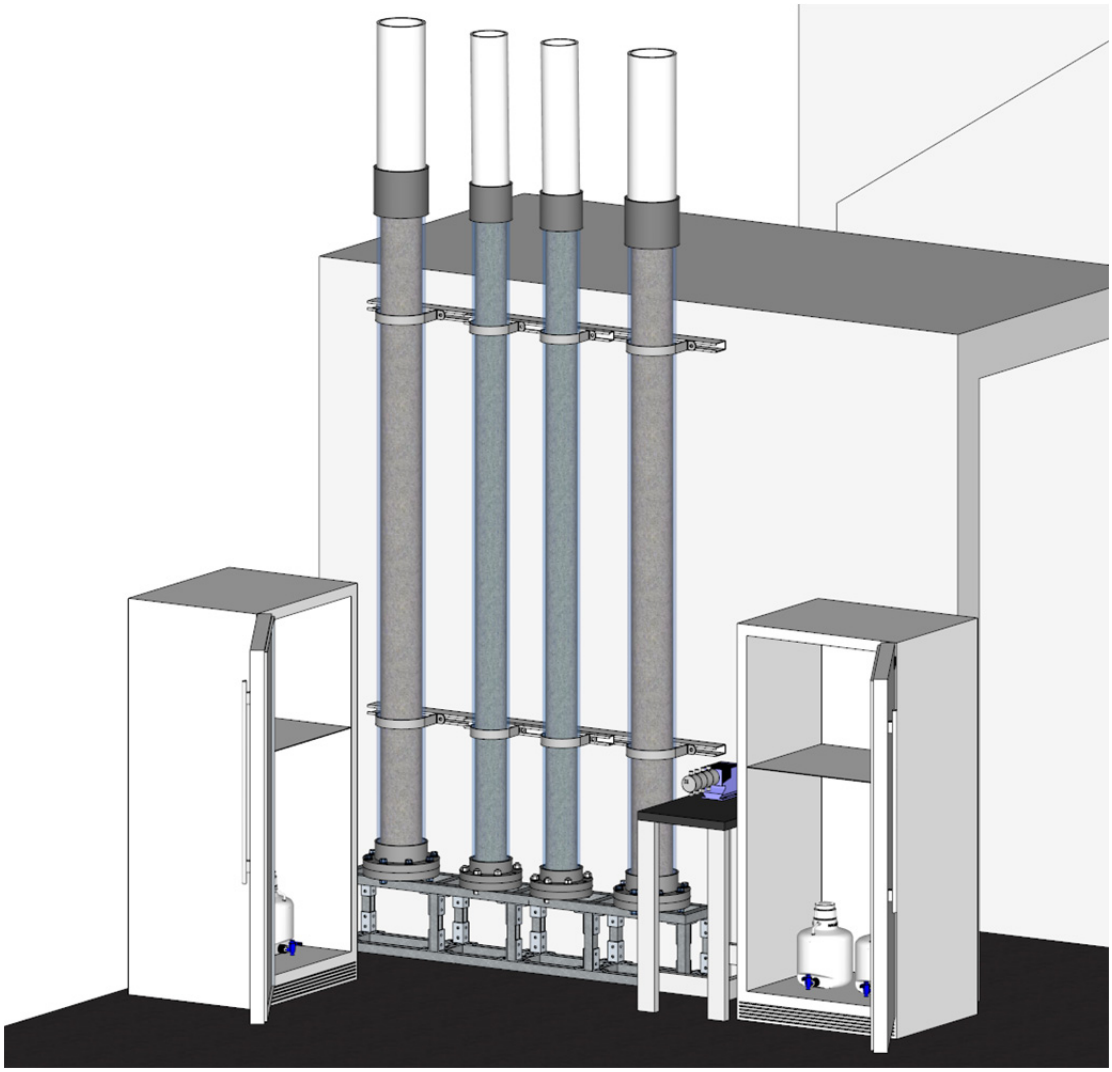

Figure 2.2. Schematic of lab-scale soil columns.

Each column included an integral media support cap and under drain to prevent soil from blocking the openings at the bottom of the column. During testing the clear columns were covered with an opaque polyethylene barrier to prevent light penetration.

2.2.2 Aquifer Material Source and Characterization

Representative soil samples were collected near the Santa Fe Spreading Grounds (SFSG) in the San Gabriel Valley Groundwater Basin (San Gabriel Basin) in Los Angeles County, CA. Stetson Engineers (Covina, CA), who have a broad range of experience in soil science in the San Gabriel Basin, assisted with identifying representative soil, which was collected from a quarry in the same aquifer as the proposed spreading grounds. A 2 mm sieve was used to remove large materials from the soil, which was later used to load the columns. Aquifer material of less than 2 mm was used to have homogeneous soil column fill material, minimize short-circuiting and flow distortions caused by larger soil material, minimize adverse and unrepresentative effects of earthworms and debris, use grain sizes that contribute to removal in the SAT process, and maximize plug flow conditions. This better simulated the subsurface SAT process, independent of soil type. The characteristic parameter that translated

specifically to the SFSG was travel time, which was defined through tracer tests (described in Section 3.3). Viewing SAT performance as a function of travel time allows for a normalized comparison between soil aquifer systems with varying characteristics.

In-house sieve analyses were performed to characterize the particle-size distribution of the aquifer material used in the Soil Column Systems [ASTM D6913-04 (2009): The Standard Test Methods for Particle-Size Distribution (Gradation) of Soils Using Sieve Analysis]. The porosity was determined as follows: (1) a graduated cylinder was filled halfway with water, and this volume (V_{water}) was recorded; (2) sand was then poured into a dry, empty graduated cylinder, and this volume (V_{sand}) was recorded; (3) this sand sample was poured into the water-filled graduated cylinder, making sure no air bubbles formed, and the new volume ($V_{sand+water}$) was recorded; and (4) porosity was calculated using Equation 2.1.

$$Porosity = \frac{V_{void}}{V_{total}} = \frac{V_{sand}+V_{water}-V_{sand+water}}{V_{sand}} \quad (2.1)$$

Using a contracted lab, the sieved soil was characterized by its fractions of organic content, clay content, and metal content using the organic carbon content, loss-on-ignition (~ 500 °C) method by Nelson and Sommers (1996); the soil texture (including clay size fraction); the 6 h hydrometer method by Gee and Or (2002); and the EPA SW 3050 digestion for soil material followed by EPA 6010B for iron and manganese. Saturated hydraulic conductivity was also measured using the constant-head, steady-state, saturated hydraulic conductivity method described by Klute and Dirksen (1986). Evaluation of organic, clay, and metal content has been included to parallel WRRF-10-05, which is studying the effect of these soil characteristics on organic matter and pathogen removal (Drewes et al., *in progress*).

2.2.3 Loading Soil into the Columns

The soil columns were partially filled with water before they were loaded with soil. The soil was dampened with water before being added to the columns one handful at a time. As the columns were being filled, the suspension was constantly mixed, and after settling the soil was compacted by tapping the outside of the column with a rubber mallet near the soil surface. This process reduced stratification and resulted in a more homogeneous composition of soil throughout the column; it also minimized entrapped air pockets. All columns were filled with 10 ft of aquifer material (described in Section 2.2.2), leaving 2 ft of freeboard.

2.2.4 Soil Column Feed Water Sources

2.2.4.1 Tertiary Effluent

The Cl_2 Soil Column System was fed dechlorinated effluent from San Jose Creek West Water Reclamation Plant (SJCWWRP). SJCWWRP uses primary settling tanks, an activated sludge process for nitrification and denitrification, secondary settling tanks, sand/coal media filters, and sequential chlorine disinfection to treat up to 37.5 million gallons of municipal wastewater per day. Sequential chlorination entails the addition of free chlorine to nitrified secondary effluent to achieve breakpoint chlorination and a measurable free chlorine residual, followed by addition of aqueous ammonia to form chloramines and comply with Title 22 disinfection requirements. Title 22 specifies the following State of California Water Recycling Criteria water quality objectives that must be attained for recycled water to be considered tertiary disinfected recycled water that can be used for various reuse applications:

- oxidized (meaning “organic matter has been stabilized, is nonputrescible, and contains dissolved oxygen”);
- filtered (either through granular media or a membrane); and
- disinfected.

The influent to the Cl_2 Soil Column System was this Title 22 effluent from SJCWWRP that was dechlorinated with sodium bisulfite (SBS) prior to feeding Cl_2 Column #1.

2.2.4.2 Ozonated Secondary Effluent

The O_3 Soil Column System was fed secondary effluent from SJCWWRP that underwent ozonation provided by a pilot-scale treatment train being used for WRRF-11-02. That treatment train used an ozone pilot system from WEDECO (Charlotte, NC). This pilot system included an oxygen generator, an ozone generator, side stream injection, contactors, an ozone destruct system, and the respective instrumentation and controls. The ozone generator had a capacity of 3.2 lbs/day for a maximum applied ozone dose of 13 mg/L at the expected water flowrate of 20 gallons per minute (gpm). A series of four contactors after the side stream injector provided 35 gal of residence volume, or nearly 2 min of HRT at 20 gpm. During WRRF-11-02 piloting, the ozone dose was optimized to achieve 6 log virus, 1 log *Cryptosporidium,* and 4 log bacteria inactivation. The ozone dose expected was approximately 5 to 9 mg/L, which corresponded to an ozone dose to TOC ratio of 1.0. The influent to the O_3 Soil Column System was this ozonated secondary effluent from the WRRF-11-02 pilot and was free of ozone residual.

2.2.5 Operations

The system was housed in a room at a temperature of approximately 20 °C. The system was operated with slight differences during each phase, the details of which may be found in Sections 2.2.6, 2.3, and 2.4. A Reglo Independent Channel Control Peristaltic Pump (IDEX Health and Science ISM4408) was used to feed all columns. All reservoirs containing the feed water and effluents were refrigerated at 4 °C to minimize biological growth, and opaque tubing was used to minimize biological growth from ambient light.

2.2.6 Startup for Bio-Acclimation

The first four months of operations were dedicated to allowing a representative biology to grow until steady-state TOC was reached at the outlet of each column. Independent of the designated operations, all columns were operated under saturated conditions during acclimation. Title 22 recycled water from SJCWWRP served as the influent for Cl_2 Column #1, and the column's effluent served as the influent to Cl_2 Column #2. Ozonated secondary effluent from the WRRF-11-02 pilot setup at SJCWWRP served as the influent for O_3 Column #1, and the column's effluent served as the influent to O_3 Column #2. The flowrates that were utilized varied within this phase and are discussed in Chapter 3.

During the acclimation period, weekly water quality samples were collected to monitor the performance of the soil columns. Samples were analyzed for electro-conductivity (EC), ammonia, nitrate, nitrite, pH, TOC, DOC, and UVT-254.

2.2.7 Tracer Tests

Tracer tests based on EC were performed during Phase 1 to characterize the residence time within each column as well as to define the time frame for the collection of seeded microbes during the microbe challenge tests.

Potassium bromide (KBr) solution at a concentration of 10 g/L was used as the tracer. Instrumentation used for the tracer tests included inline EC probes, EC transmitters, a programmable logic controller (PLC) for continuous data logging, and an air temperature logger to compensate for temperature in EC. On injection of the tracer solution, column effluent EC measurements were made at 5-min intervals until levels fell to baseline measurements after the peak concentration was detected.

Although actual residence times of the columns were 5 days and 25 days for vadose and saturated columns, respectively, shorter duration tracer tests were completed and scaled up to the range of design operation. Benefits of using shorter duration tracer tests include (a) less impact on the project schedule and the ability to perform repeated tracer tests, (b) minimized effects of gravity on the denser tracer solution, and (c) minimized effects of dispersion of the tracer. After completion of the tracer tests, the systems were given 3 weeks to recover from any bacterial static effects of the tracer that would result in depressed SAT performance. Results of the tracer tests are provided in Chapter 3.

2.3 Phase 2: Bulk Organic Carbon, CEC, and DBP Evaluation

Soil column operations were configured to represent typical full-scale surface spreading practices. Cl_2 Column #1 and O_3 Column #1 were switched to unsaturated or vadose flow to resemble current stormwater and imported water recharge practices by the Los Angeles County Department of Public Works at the SFSG. To resemble the vadose zone, a 4-week cycle with two distinct operational parameters was used. The first part of the cycle had a saturated, continuous flow regime (downflow) at a flowrate to achieve a residence time of 5 days, lasting for 3 weeks. During the second part of the cycle, the influent flow was stopped and the system was allowed to drain/dry for a week. This cycle was repeated for the duration of this phase. Cl_2 Column #2 and O_3 Column #2 remained saturated to continue representing the region below the water table. These columns were continuously fed vadose effluents using an upflow regime with a 25-day residence time.

Column feeds were not spiked during this phase. The inventory of CECs monitored during this phase was based on

- findings from the CEC monitoring program of the Los Angeles County Sanitation Districts (LACSD) over a period of 5 years; CECs that were consistently present (median based on at least two sampling events and concentration greater than the detection limit) in SJCWWRP were included for assessment during soil column piloting;
- literature on potential SAT indicators, specifically Laws et al. (2011), Drewes et al. (2011), and Trussell Technologies (2011);
- analytical methods available through LACSD Laboratories; and
- consultation with DDW and RWQCB staff.

The suite of CECs listed in Table 2.1 was selected to characterize removal and identify potential SAT performance indicators.

Table 2.1. CECs Monitored During Soil Column Testing

Monitored CECs	Type
4-Nonylphenol (tech mix)	Surfactant/Pesticide
4-tert-octylphenol	Surfactant
Acesulfame-K	Artificial Sweetener
Acetaminophen	Analgesic
Atenolol	Beta blocker
Atorvastatin (Lipitor)	Statin
Azithromycin	Antibiotic
Bisphenol A	Monomer in plastic production
Caffeine	Stimulant
Carbamazepine	Anticonvulsant
Carisoprodol	Muscle relaxant
DEET	Insect repellent
Diazapam	Anti-anxiety drug (Valium)
Diclofenac	Anti-inflammatory (NSAID)
Erythromycin-H2O	Antibiotic
Fipronil	Insecticide
Fluoxetine (Prozac)	Anti-depressant
Furosemide	Diuretic
Galaxolide	Fragrance
Gemfibrozil	Lipid regulator
Ibuprofen	Anti-inflammatory (NSAID)
Iohexol	Contrast agent
Iopromide	Contrast agent
Meprobamate	Anti-anxiety
Metoprolol	Beta blocker
Naproxen	Anti-inflammatory (NSAID)
Nonylphenol diethoxylate	Surfactant
Nonylphenol monoethoxylate	Surfactant
Octylphenol diethoxylate	Surfactant
Octylphenol monoethoxylate	Surfactant

Table 2.1. CECs Monitored During Soil Column Testing (*continued*)

Monitored CECs	Type
Phenytoin (Dilantin)	Anti-seizure
Primidone	Anticonvulsant
Propranolol	Beta blocker
Sucralose	Artificial Sweetener
Sulfamethoxazole	Antibiotic
TCEP	Flame Retardant
TCPP	Flame Retardant
TDCPP	Flame Retardant
Tonalide	Fragrance
Triclocarban	Antibacterial agent
Triclosan	Antibacterial agent
Trimethoprim	Antibiotic

Table 2.2. DBPs Monitored During Soil Column Testing

Monitored DBPs
Bromate
HAA: dibromoacetic acid (DBAA)
HAA: dichloroacetic acid (DCAA)
HAA: monobromoacetic acid (MBAA)
HAA: monochloroacetic acid (MCAA)
HAA: trichloroacetic acid (TCAA)
Nitrosamines: nitrosodimethylamine (NDMA)
Nitrosamines: nitrosomethylethylamine (NMEA)
Nitrosamines: nitrosodiethylamine (NDEA)
Nitrosamines: nitrosodi-*n*-propylamine (NDPA)
Nitrosamines: nitrosodi-*n*-butylamine (NDBA)
Nitrosamines: nitrosopiperidine (NPIP)
Nitrosamines: nitrosopyrollidine (NPYR)
THM: chloroform (TCM)
THM: bromodichloro methane (BDCM)
THM: dibromochloro methane (DBCM)
THM: bromoform (TBM)

DBPs related to each disinfection alternative were also evaluated (Table 2.2). Total trihalomethanes (TTHMs) and individual trihalomethanes (TCM, BDCM, DBCM, and TBM), total HAAs and individual HAAs (DBAA, DCAA, MBAA, MCAA, and TCAA), and individual nitrosamines (NDMA, NMEA, NDEA, NDPA, NDBA, NPIP, and NPYR) were analyzed from the Soil Column System that was treating chlorinated tertiary effluent. Bromate and individual nitrosamines were analyzed from the Soil Column System that was treating ozonated secondary effluent.

In addition to CECs and DBPs, routine water quality parameters were monitored, including ammonia, BDOC, DOC, EC, EEM fingerprint, nitrate, nitrite, orthophosphate, pH, TOC, turbidity, and UVT-254. A summary of the sampling locations and frequencies for Phase 2 are provided in Table 2.3.

Table 2.3. Sampling Locations and Frequencies for Monitoring of CECs, DBPs, Nitrosamines, Routine Parameters, BDOC, and EEM Fingerprint

Sampling Location	CECs	DBPs	Nitrosamines	Routine (e.g., TOC and UVA)	BDOC	EEM
Secondary	X			X		X
Cl_2 Soil Column System						
Influent	X	X	X	X	X	X
Vadose Effluent	X		X	X		X
Saturated Effluent	X	X	X	X		X
O_3 Soil Column System						
Influent	X	X	X	X	X	X
Vadose Effluent	X		X	X		X
Saturated Effluent	X	X	X	X		X
Total number of events	8	3	7	6	6	3

2.4 Phase 3: Microorganism Evaluation

Cl_2 Column #1 and Cl_2 Column #2 were utilized in the microbiological challenge phase. Both columns were fed continuously with a residence time of 30 days each. Cl_2 Column #1 was used to test coliform removal and Cl_2 Column #2 to test MS-2 virus and *Cryptosporidium* removal. Effluent from Cl_2 Column #1 was analyzed for total coliform. Effluent from Cl_2 Column #2 was analyzed for MS-2 virus and the remaining volume was concentrated and sent to BioVir (a laboratory in Benicia, CA, that specializes in the analysis of microbes) for *Cryptosporidium* enumeration. Each test was carried out three times to determine the attenuation of protozoa, virus, and bacteria through SAT.

The duration of composite effluent sampling was defined using results from the tracer tests, as described in Section 3.3. To bracket the breakthrough period, effluent sample was collected during the period when the majority of the tracer exits the system.

All microbiological spiking specimens were stored at 4 °C to minimize biological growth and die-off. *Cryptosporidium parvum* oocysts were purchased from Bunch Grass Farms, Deary, ID. Male-specific-F^+ (MS-2) coliphages (ATCC #15597-B1) were purchased from BioVir, Benicia, CA. *E. coli* (ATCC #700891) for the total coliform testing was purchased from ATCC, Manassas, VA. All applied water during this phase was quenched to assure that the feed was free of oxidant.

The target pulse microbe spike concentration was selected to demonstrate at least 6-log removal. The target feed dose for each challenge microbe was based on the values provided in Table 2.4: the target spike concentration accounted for the desired maximum removal to demonstrate, and the detection limits and a buffer accounted for matrix effects and dilution.

Table 2.4. Target Microbial Concentrations in the Spike for the Microbe Challenges

Components to Determine Spike Solution Concentration	Bacteria *E. coli* (ATCC #700891)	Virus MS-2 coliphage (ATCC #15597-B1)	Protozoa *Cryptosporidium parvum* oocysts
	Cl_2 Column #1	Cl_2 Column #2	Cl_2 Column #2
Detection limit	0.2 cfu/mL	20 pfu/mL	1 oocyst/L
Detection limit, log	-0.7	1.3	0
Max. removal to demonstrate, log	6	6	6
Dilution buffer[1], log	2	2	2
Target influent concentration	2E+07 cfu/mL	2E+09 pfu/mL	1E+08 oocysts/L

Notes: [1]Additional spike to compensate for dilution that takes place within the column and that is due to the nature of the composite sampling method (i.e., the beginning and ending samples will dilute the peak occurring in the middle samples); this amount was based on results from the tracer tests

pfu = plaque-forming unit; cfu = colony-forming unit

The microbe challenge entailed taking an aliquot of feed water for each column and spiking it with the test organisms. For Cl_2 Column #1 (8 in. diameter with downflow regime), the size of the aliquot was 1.1 L; and for Cl_2 Column #2 (6 in. diameter with upflow regime), it was 0.75 L. For Cl_2 Column #1, after the aliquot was spiked, samples were collected for analysis to measure the influent concentration of total coliform. For Cl_2 Column #2, after the aliquot was spiked, samples were collected for analysis to measure the influent concentration of MS-2 virus and *Cryptosporidium*.

The spiked aliquot was introduced into Cl_2 Column #1 in the following manner:

- the column was drained using a tap at the base of the column until the water level in the column was at the top level of the sand
- spike solution was quiescently pulsed into the top of the column over a period of 45 min
- the column was drained again to bring the liquid level to 1 in. below the sand surface
- unspiked water was introduced over a period of 45 min to bring the standing water level back to a normal operating level of approximately 1 in. above the sand surface; a tee on the effluent tube from the column base was elevated to a level that maintained shallow submergence of the sand with approximately 1 in. of water

The spiked aliquot was introduced into Cl_2 Column #2 in the following manner: The spike solution was pulsed into the system using an upflow regime over a period of 20 min.

After introducing the pulse, normal operations were resumed (i.e., continuous feed of unspiked water, saturated conditions with a 30-day residence time). For *Cryptosporidium*, composite effluent samples were collected for the duration determined during the tracer study to bracket the breakthrough period. For the entire duration of the study, MS-2 virus and total coliform were enumerated in 3- to 4-day composite samples. The volume of each composite sample was noted, thereby allowing the computation of the overall count. All samples were stored at 4 °C and analyzed as described in Sections 2.5.6, 2.5.7, and 2.5.8. Routine water quality monitoring continued during this phase. The attenuation of each challenge microbe was based on the total number of microbes spiked and total number of microbes collected in the effluent composite.

2.5 Experimental Methods

This section describes water quality analyses that were used to evaluate the samples collected during this study.

2.5.1 Routine Water Quality Analyses

The following equipment was used for in-house water quality analyses:

- TOC: GE Sievers 5310 C
- UVA: Hach DR5000 spectrophotometer
- EEM: Horiba Scientific Aqualog spectrofluorometer
- turbidity: Hach 2100AN turbidimeter
- DO, pH, conductivity: Hach HQ40d portable meter
- ammonia, nitrate, nitrite, orthophosphate: Hach DR890 colorimeter

The Horiba Scientific Aqualog spectrofluorometer, Hach DR5000 spectrophotometer, GE Sievers 5310 C TOC analyzer, Hach 2100AN turbidimeter, Hach DR890 colorimeter, and Hach HQ40d portable meter were calibrated and maintained in accordance with manufacturer recommendations. All blanks were from the Thermo Scientific Barnstead Nanopure Ultrapure Water System dispensing 18.2 MΩ-cm and less than 10 parts per billion TOC. Samples were filtered through a 0.45 µm pore size disc filter (25 mm diameter Acrodisc® Syringe Filters with Supor® polyethersulfone membrane by Pall® Life Science) before analysis with the TOC analyzer. Supor polyethersulfone membrane filters have low DOC leaching potential (Karanfil et. al., 2003) for analyses requiring the dissolved fraction of the sample.

The following quality assurance/quality control (QA/QC) procedure was used for microbiological seeding experiments:

- samplers verified the phage, coliform, and oocyst seed stock solution feed rate volumetrically, for at least 1 min before the start of seeding
- samplers wore plastic gloves and changed gloves after each sample was collected
- each sample was individually sealed in a sterile container
- feed and filtered samples were stored in different physical locations

- samples were refrigerated in sealed plastic coolers packed with ice until delivery to the laboratory
- maximum holding times were observed, per standard methods

The following QA/QC procedure was used for CEC and DBP sampling:

- samplers wore plastic gloves and changed gloves after each sample was collected
- each sample was individually sealed in a container provided by LACSD Laboratory
- feed and filtered samples were stored in different physical locations
- samples were refrigerated in sealed plastic coolers packed with ice until delivery to the laboratory; the temperatures of the coolers were recorded upon laboratory receipt
- maximum holding times were observed, per standard methods

To optimize data quality, all samples were taken and preserved in accordance with specified holding times and temperature conditions, and all analyses followed QA/QC procedures related to the associated methods. In addition, detailed instructions and checklists accompanied all methods used.

2.5.2 Fluorescence Excitation-Emissions Matrix (EEM) Spectroscopy

The fluorescence EEM spectroscopy used a PTI fluorometer (Birmingham, NJ) for data acquisition, and data was processed in MatLab (Natick, MA). The EEM images were corrected for the Raman scatter by subtracting the emission of the blank and were corrected for inner-filter effect by following a previously described method (MacDonald et al., 1997). In addition, total fluorescence was measured.

2.5.3 Biodegradable Dissolved Organic Carbon (BDOC)

BDOC is a measure of the DOM of wastewater origin that can be transformed by indigenous microbes. A standardized test for BDOC has yet to be developed; therefore, this project utilized the BDOC method recommended by the National Water Research Institute (NWRI) Independent Advisory Panel (NWRI, 2012). The recommended procedure was developed by Cha et al. (2004) and is specifically designed to assess SAT performance using batch reactors. Although the test is operationally defined by the total travel time of the Soil Column System, the inherent advantages, reproducibility, and simplicity outweigh this disadvantage (NWRI, 2012).

Details for this experimental protocol were extracted from Cha et al. (2004) and Drewes and Dickenson (2007). Washed silica sand served as the medium in the batch reactors. Sand measuring 200 g was washed with saline solution (0.15 M sodium chloride and 1 mM magnesium chloride). A 1 L media bottle was used as the batch reactor. The sample volume was 600 mL. Nine reactors were set up for each specific water sample of interest (i.e., secondary, ozonated secondary, and tertiary from SJCWWRP). Each test was run with triplicate reactors. The soil-attached biology was acclimated using secondary, ozonated secondary, and tertiary water from SJCWWRP for 4 months. The reactors were stored at room temperature in the dark and were manually swirled every other day to maintain aerobic conditions. The solution in each reactor was replaced with fresh feed water every 5 to 10 days during acclimation. The acclimation period continued until the standard deviation of the BDOC was less than 5% for at least four consecutive cycles. Using river soil and tertiary

effluent, Cha et al. (2004) noted an acclimation period of 6 weeks, at which point the DOC removal was roughly 20%.

Once the reactors were acclimated, BDOC sampling was conducted biweekly for a total of six sampling events. During each test, the reactors were manually swirled every other day to maintain oxic, well-mixed conditions. Each test lasted 30 days, during which samples were collected and analyzed for DOC and dissolved oxygen (DO). BDOC is the percentage reduction in DOC between an initial sample (DOC_o) and a final sample (DOC_{30d}). For this study, the days passed since initial DOC measurement were equivalent to the travel time through the Soil Column System (30 days). BDOC was calculated using Equation 2.2.

$$BDOC_{30d} = 100 * \frac{(DOC_o - DOC_{30d})}{DOC_o} \quad (2.2)$$

2.5.4 Contaminants of Emerging Concern (CECs)

LACSD Laboratory analyzed CECs using established protocol (a modified version of EPA 1694 as described by Nelson et al., 2011). Method reporting limits are listed in Table 2.5.

Table 2.5. Method Reporting Limits for CECs Monitored During Soil Column Testing

Monitored CECs	Method Reporting Limit, ng/L
4-Nonylphenol (tech mix)	25
4-tert-octylphenol	5
Acesulfame-K	50
Acetaminophen	10
Atenolol	10
Atorvastatin (Lipitor)	10
Azithromycin	10
Bisphenol A	10
Caffeine	10
Carbamazepine	10
Carisoprodol	10
DEET	10
Diazapam	10

Table 2.5. Method Reporting Limits for CECs Monitored During Soil Column Testing (*continued*)

Monitored CECs	Method Reporting Limit, ng/L
Diclofenac	10
Erythromycin-H2O	10
Fipronil	2
Fluoxetine	10
Furosemide	10
Galaxolide	100
Gemfibrozil	10
Ibuprofen	100
Iohexol	10
Iopromide	10
Meprobamate	10
Metoprolol	10
Naproxen	10
Nonylphenol diethoxylate	25
Nonylphenol monoethoxylate	25
Octylphenol diethoxylate	25
Octylphenol monoethoxylate	25
Phenytoin (Dilantin)	10
Primidone	10
Propranolol	10
Sucralose	40
Sulfamethoxazole	10
TCEP	10
TCPP	20
TDCPP	20
Tonalide	100
Triclocarban	10
Triclosan	10
Trimethoprim	10

2.5.5 Disinfection Byproducts (DBPs)

Bromate and HAA samples were analyzed by Eaton Analytical (Monrovia, CA), and nitrosamines and TTHMs were analyzed by LACSD Laboratory. Methods and method reporting limits are summarized in Table 2.6.

Table 2.6. Methods and Method Reporting Limits for DBPs Monitored During Soil Column Testing

Monitored DBPs	Method	Method Reporting Limit, ng/L
Bromate	EPA Method 326	500
HAAs	EPA Method 552.2	2000
Dibromoacetic acid (DBAA)		
Dichloroacetic acid (DCAA)		
Monobromoacetic acid (MBAA)		
Monochloroacetic acid (MCAA)		
Trichloroacetic acid (TCAA)		
NDMAs	EPA 1625	2
Nitrosodimethylamine (NDMA)		
Nitrosomethylethylamine (NMEA)		
Nitrosodiethylamine (NDEA)		
Nitrosodi-*n*-propylamine (NDPA)		
Nitrosodi-*n*-butylamine (NDBA)		
Nitrosopiperidine (NPIP)		
Nitrosopyrrolidine (NPYR)		
THMs	EPA 624	500
Trichloromethame (TCM)		
Bromodichloro methane (BDCM)		
Dibromochloro methane (DBCM)		
Bromoform (TBM)		

2.5.6 *Cryptosporidium Parvum* Oocysts

Cryptosporidium parvum oocysts were purchased from Bunch Grass Farms (Deary, ID). On receipt the oocysts were irradiated: using a collimated beam, 150 mJ/cm^2 was delivered to the oocyst suspension, which equated to 8 min of exposure. The suspension was then washed with sterile distilled water to remove the storage buffer by centrifuging at 1750 revolutions per minute (rpm) for 10 min. The liquid was decanted, and the pellet was resuspended in 10 mL of autoclaved distilled water. This solution was centrifuged again at 1750 rpm for 10

min. The supernatant was decanted, and the oocyst pellet was re-suspended with 10 mL of distilled water for later use in the pulse spike solution for the three microbe challenges.

BioVir Laboratories (Benicia, CA) analyzed the *Cryptosporidium* samples following EPA Method 1622 for *Cryptosporidium* in water. *Cryptosporidium* removal was determined by comparing the oocyst count between the spiked feed water and the soil column effluent.

2.5.7 Male-Specific F+ (MS-2) Coliphage

Removal of the MS-2 virus was evaluated after SAT. MS-2 stock (>10^{11} infectious units per mL) was purchased from BioVir (Benicia, CA). Additional information about these stocks may be found at the following URL: http://www.biovir.com/phage.html. Viral titers in the stock were enumerated by Trussell Technologies Laboratory using the double-agar-layer method (Standard Method 9224C). The method is used to quantify infectious viruses based on the number of plaques formed on a lawn of an *E. coli* bacterial host strain. Each plaque derives from a single viral plaque-forming unit (pfu) with viral concentrations calculated as pfu per mL of sample volume tested. MS-2 removal was evaluated by using the stock MS-2 solution to create a spiked concentration of ~2 x 10^9 pfu/mL in the spiked feed. Water samples collected from the columns were analyzed without dilution, using 1 mL of sample. If there were any hits, the sample was diluted appropriately. The MS-2 coliphage in the percolating water was enumerated using the same double-agar-layer method.

2.5.8 Total Coliform

Stock of CN-13, a coliform bacterium, washed and suspended in phosphate buffered solution and at a concentration of 10^{10} cfu/mL, was purchased from BioVir (Benicia, CA). Total coliform counts were measured in the spiked feed. Trussell Technologies Laboratory analyzed all samples collected using the membrane filtration procedure (SM 9222b). Effluent samples were analyzed by filtering 100 mL of sample, and additional dilutions were enumerated, if the plaque count was out of range.

Chapter 3

Phase 1: Setup, Acclimation, and Hydraulics

3.1 Construction and Setup

Representative soil was collected from a quarry in the same aquifer as the proposed spreading grounds (washed sand from Cal Blend Soils, Inc., Irwindale, CA). The soil was sieved to remove material larger than 2 mm in diameter (Figure 3.1). Soil characteristics are given in Table 3.1, and the grain size distribution is provided in Figure 3.2. Soil columns were loaded in a manner that ensured compaction, such as by addition of soil into standing water in the column and tapping the side of the column to release air (Figure 3.3). Once the columns were loaded, they were wrapped with sheets of opaque plastic to minimize algae growth (Figure 3.4). The completed columns with flowpaths are shown in Figure 3.5.

Figure 3.1. Sieving of soil for the soil columns.

Table 3.1. Soil Characteristics

Parameter	Results
Soil description	Fine to medium grained sand with silt
D_{10}	0.18 mm
Uniformity coefficient	3.3
Porosity	33%
Total organic content	0.40%
Clay content	1.8%
Metal content: Iron (Fe)	16,800 mg/kg
Metal content: Manganese (Mn)	156 mg/kg
Permeability ($k_{20\ deg\ C}$)	1.13E-02 cm/sec

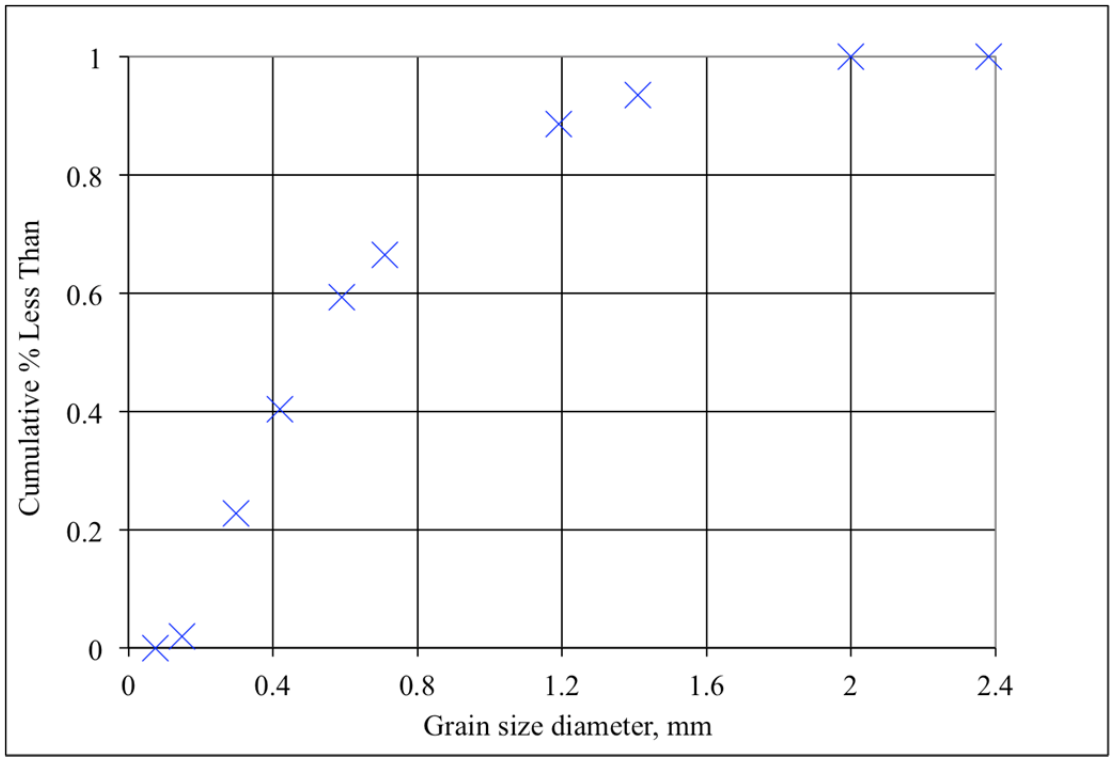

Figure 3.2. Grain size distribution.

Figure 3.3. Soil column assembly: (a) loading the soil columns, which were filled with the source water before soil addition; (b) tapping the columns during loading to help compact the soil; and (c) the columns when soil loading was nearly complete.

Figure 3.4. Soil columns being wrapped with plastic to prevent algae growth.

Figure 3.5. System configuration and flowpaths.

During Phase 1, all columns were operated under saturated conditions. During Phase 2, Cl_2 Column #1 and O_3 Column #1 (larger columns) were operated to resemble the vadose zone, and Cl_2 Column #2 and O_3 Column #2 (smaller columns) were operated to represent the saturated zone. During Phase 3, only the Cl_2 Soil Column System was utilized, and both Cl_2 Columns #1 and #2 were operated under saturated conditions. During Phases 1 and 2, the effluent from the larger columns served as the feed to the smaller columns. During Phase 3, the two columns operated independently. The travel time through each column for the entire duration of this study is graphed in Figure 3.6. During acclimation, the peristaltic pumps delivering water to the columns were not able to maintain a consistent flowrate. Thus, they were replaced with higher performance peristaltic pumps that were better suited to the application. Phase 1 (acclimation) spanned from August 26, 2013, to January 5, 2014, and included tracer testing (November 21 to December 15, 2013) and recovery after tracer testing (December 15, 2013 to January 5, 2014). Phase 2 (CEC, DBP, and bulk organic monitoring) spanned from January 6 to April 11, 2014. Phase 3 (microbe challenges) spanned from April 14 to July 11, 2014.

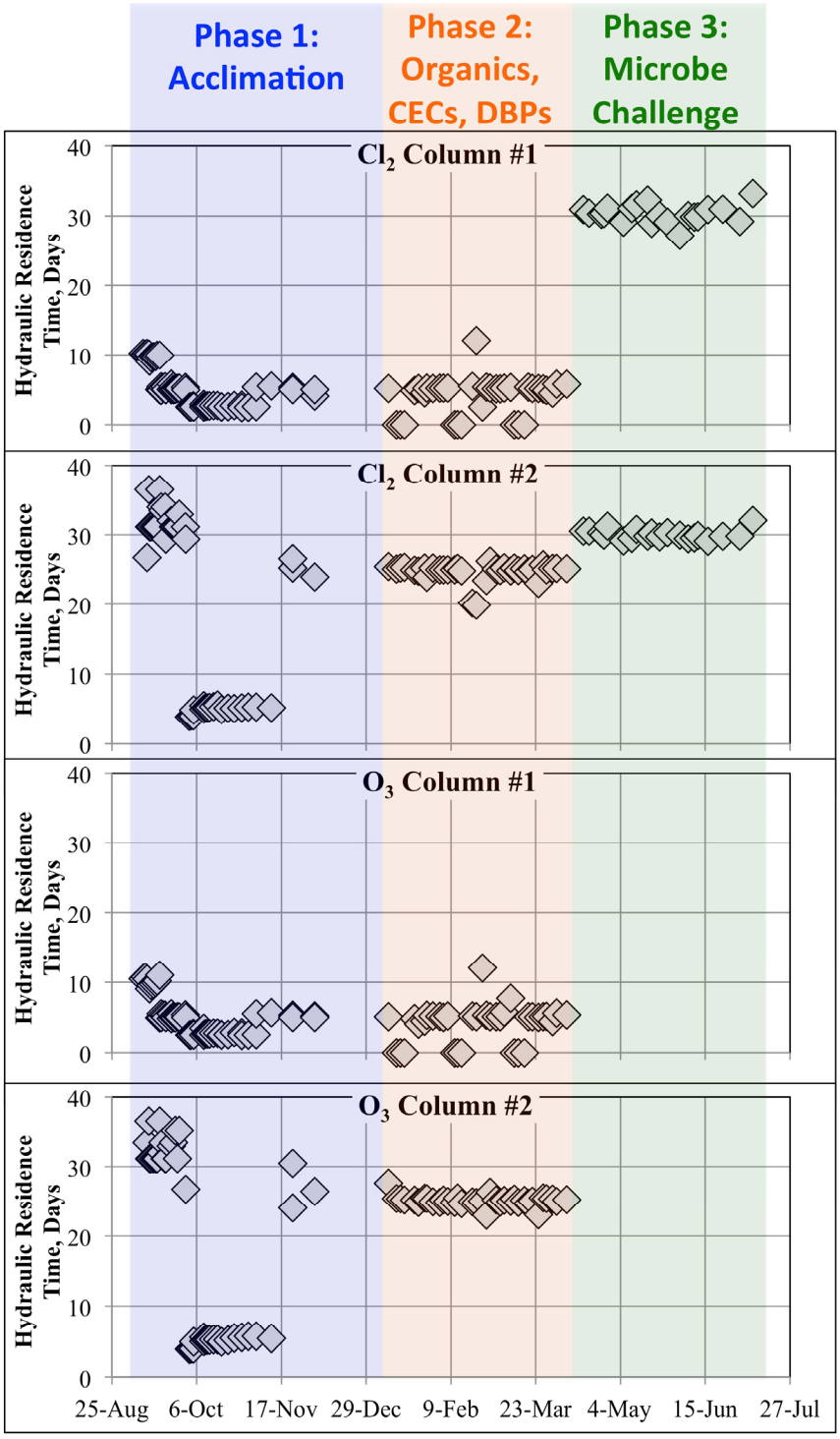

Figure 3.6. System HRT during the entire duration of the project.

3.2 Acclimation

Acclimation consisted of operating the columns under saturated conditions to allow the establishment of a microbial community. The systems were initially operated at residence times equivalent to those planned during the evaluation of CECs (i.e., $C1_2$ Column #1 and O_3 Column #1 operated at a 5-day HRT; $C1_2$ Column #2 and O_3 Column #2 operated at a 25-day HRT). However, it was observed that the TOC in the effluents of all columns was greater than that in the influent. As a result, the flowrate was increased to flush the sand of residual organic carbon (i.e., $C1_2$ Column #1 and O_3 Column #1 operated at a 2.5-day HRT; $C1_2$ Column #2 and O_3 Column #2 operated at a 5-day HRT). In late November 2013, after the TOC had stabilized, the operations were reverted to the original settings (i.e., $C1_2$ Column #1 and O_3 Column #1 operated at a 5-day HRT; $C1_2$ Column #2 and O_3 Column #2 operated at a 25-day HRT). Raw TOC data during the stabilization and acclimation period is shown in Figures 3.7 and 3.8 for the Cl_2 and O_3 Soil Column Systems, respectively.

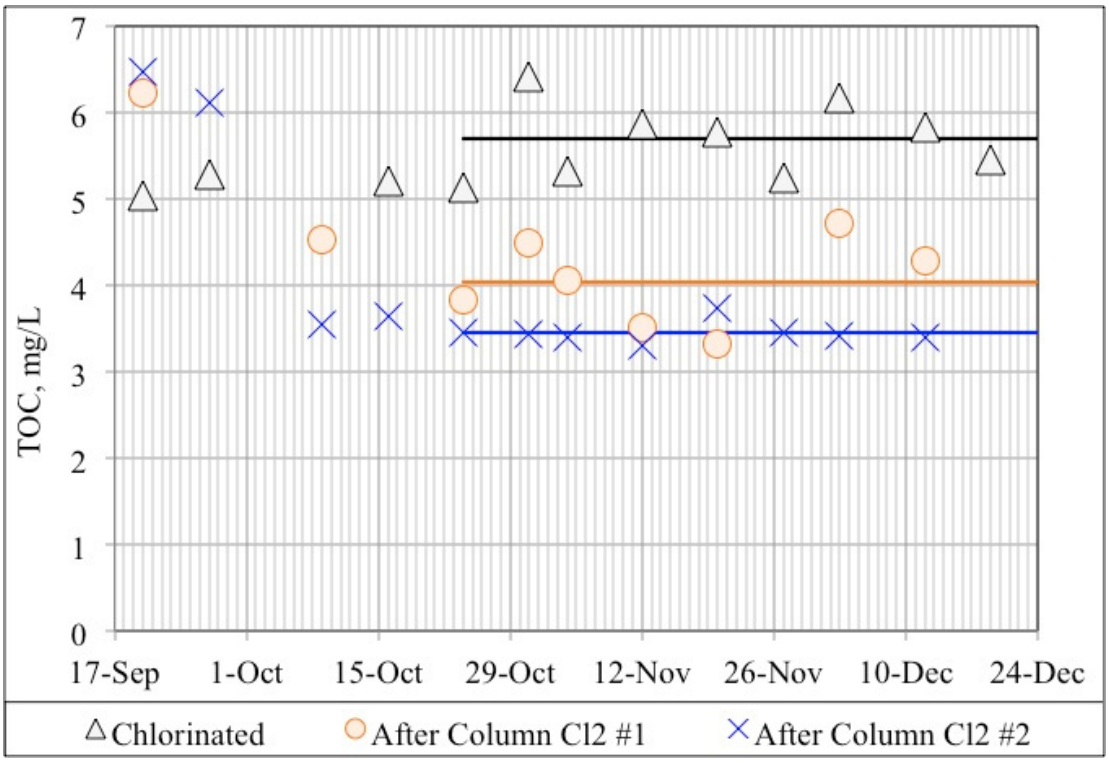

Figure 3.7. Overall Cl_2 Soil Column System performance based on TOC during Phase 1 (acclimation); solid horizontal lines correspond to the average.

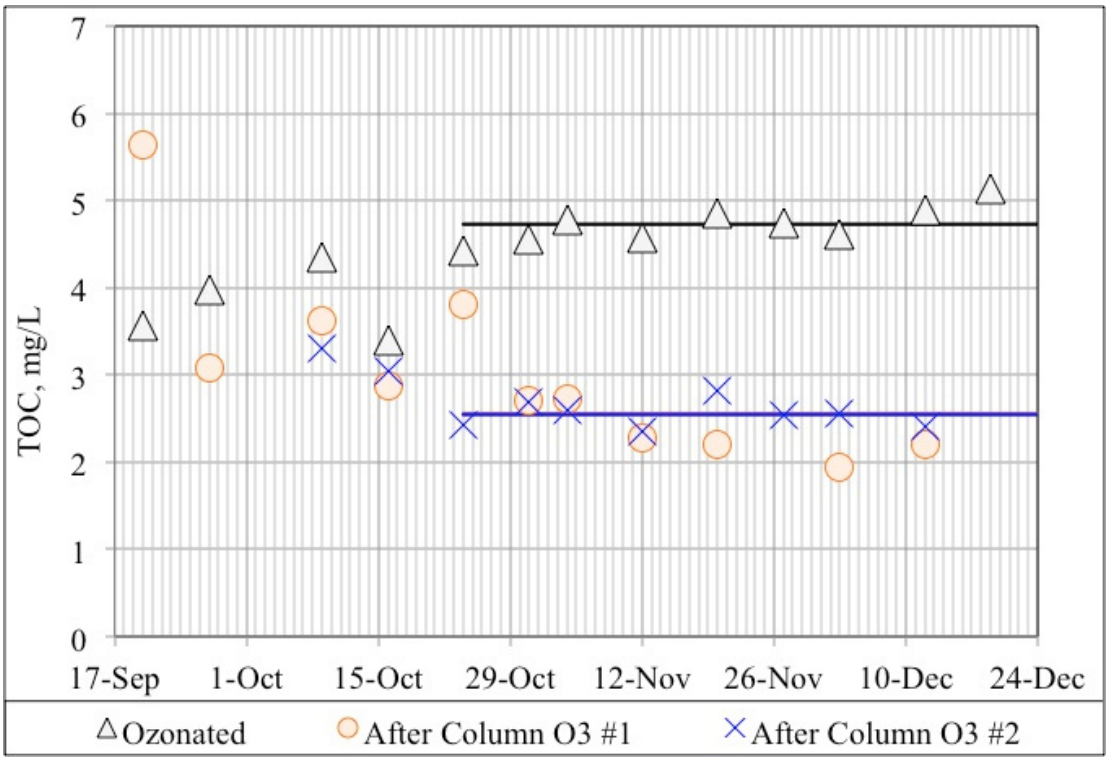

Figure 3.8. Overall O_3 Soil Column System performance based on TOC during Phase 1 (acclimation); solid horizontal lines correspond to the average.

After the unexpected increases in soil column effluent TOC were curbed by flushing out residual organic matter, the TOC stabilized. The Soil Column System being fed chlorinated water attained on average a final TOC effluent concentration of 3.4 mg/L, which equated to 39% removal. The Soil Column System being fed ozonated water attained on average a final TOC effluent concentration of 2.6 mg/L, which equated to 46% removal. TOC data are summarized in Table 3.2.

Table 3.2. Summary of TOC Data During Acclimation

Location	TOC, mg/L Average ± Standard Deviation	Average Difference in TOC, mg/L	Average Percentage TOC Removal
Cl_2 Disinfection Alternative			
Post-Chlorination	5.70 ± 0.44	n/a	n/a
After Cl_2 Column #1	4.00 ± 0.51	1.7	29
After Cl_2 Column #2	3.40 ± 0.13	0.6	14
Overall Cl_2 System	n/a	2.2	39
O_3 Disinfection Alternative			
Post-Ozonation	4.70 ± 0.22	n/a	n/a
After O_3 Column #1	2.60 ± 0.62	2.2	46
After O_3 Column #2	2.50 ± 0.15	0.01	0.3
Overall O_3 System	n/a	2.2	46

During this same period, the UVT was also monitored for the Cl_2 and O_3 Soil Column Systems, respectively shown in Figures 3.9 and 3.10.

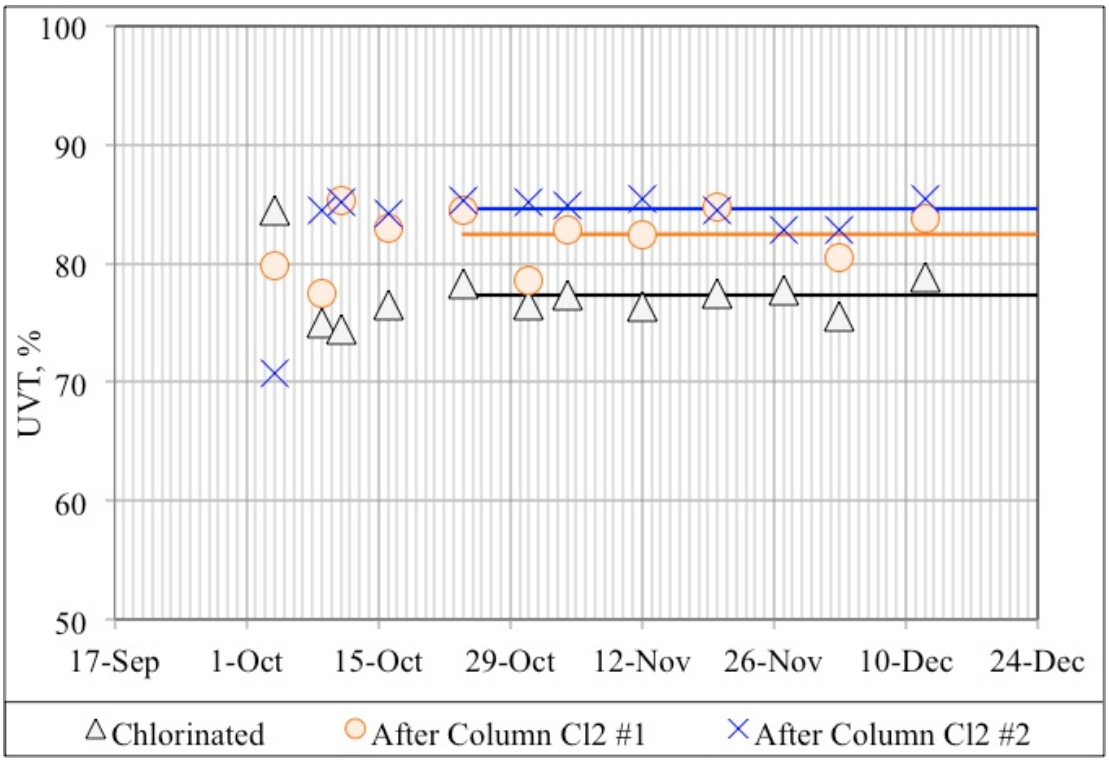

Figure 3.9. Overall Cl_2 Soil Column System performance based on UVT during Phase 1 (acclimation); solid horizontal lines represent averages.

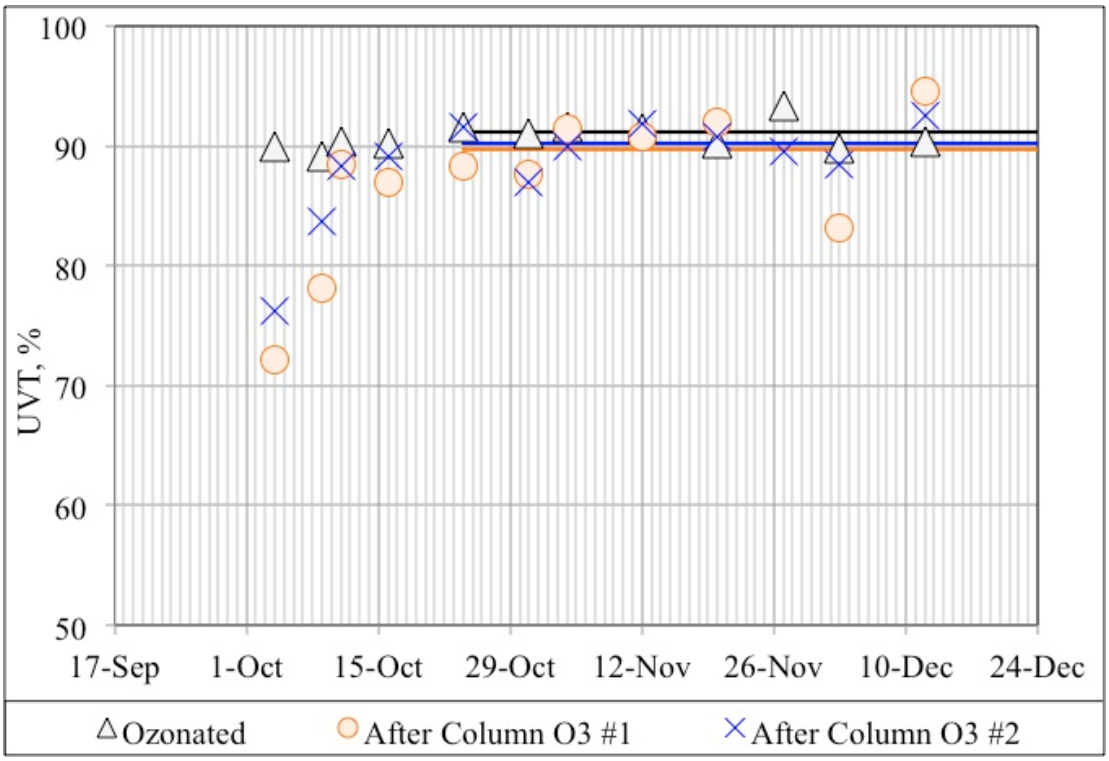

Figure 3.10. Overall O_3 Soil Column System performance based on UVT during Phase 1 (acclimation); solid horizontal lines represent averages.

The average UVT in the final effluent was 85% for the Cl_2 Soil Column System and 90% for the O_3 Soil Column System. UVT and specific ultraviolet absorbance (SUVA), which is UVA/TOC x 100, are summarized in Table 3.3.

Table 3.3. Summary of UVT and SUVA Data During Acclimation

	Parameter	
Location	**UVT, %** **Mean ± Std. Dev.**	**SUVA, L/mg-M** **Mean ± Std. Dev.**
Cl_2 Disinfection Alternative		
Post-Chlorination	77.0 ± 1.1	1.96 ± 0.13
After Cl_2 Column #1	82.0 ± 2.2	2.09 ± 0.22
After Cl_2 Column #2	85.0 ± 1.1	2.11 ± 0.18
O_3 Disinfection Alternative		
Post-Ozonation	91.0 ± 1.1	0.86 ± 0.11
After O_3 Column #1	90.0 ± 3.7	1.95 ± 1.01
After O_3 Column #2	90.0 ± 1.9	1.76 ± 0.30

For the Cl_2 Soil Column System, the average UVT improved with travel time (as seen in Table 3.3: 82% after Column #1 and 85% after Column #2); however, this observation was not apparent for the O_3 Soil Column System.

3.3 Tracer Test Results

Tracer tests were completed 12 weeks into acclimation. Tracer tests using high concentrations of potassium bromide to have measurable differences in electrical conductivity were used to characterize the residence time within each column as well as to define the time frame for collecting seeded microbes during the microbe challenge tests (Phase 3). Pictures of the setup, including the inline EC probes, the EC transmitter, and the PLC are shown in Figure 3.11.

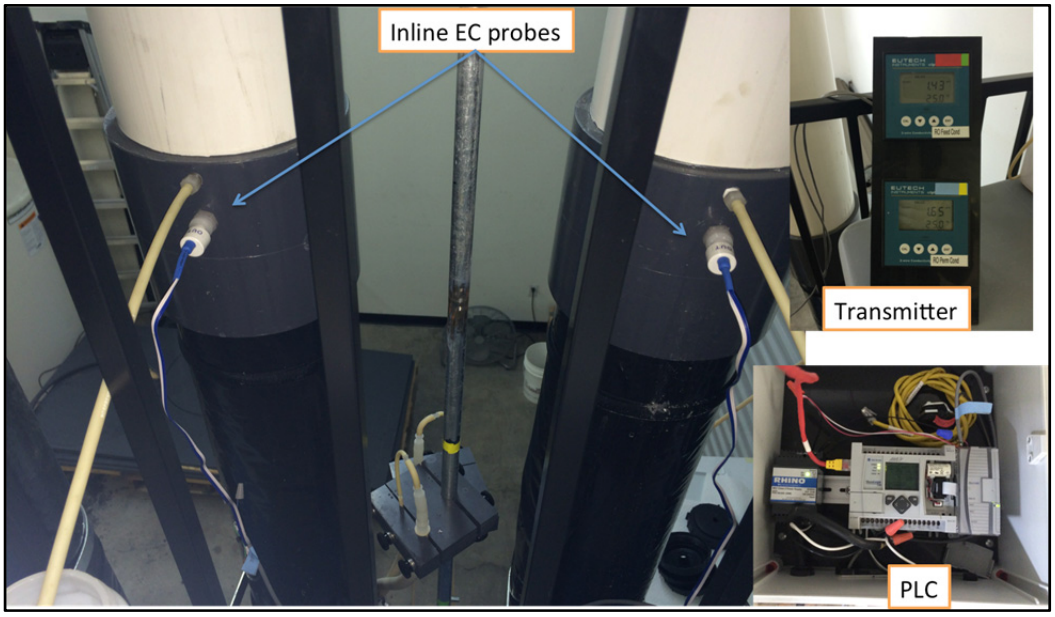

Figure 3.11. Inline EC probes, the EC transmitter, and the PLC for tracer testing.

Preliminary tracer tests were conducted to determine if tracer tests with a reduced residence time could be used to predict hydraulic characteristics at the columns' design flowrates. Figure 3.12 shows breakthrough curves for the three tests. Section 2.2.7 details the purpose for these tests. Normalized curves are presented in Figure 3.13 for the three tests. These responses suggest that if effluent is collected from 80 to120% of the target HRT, then the majority of the spiked material that was added will have passed through the column.

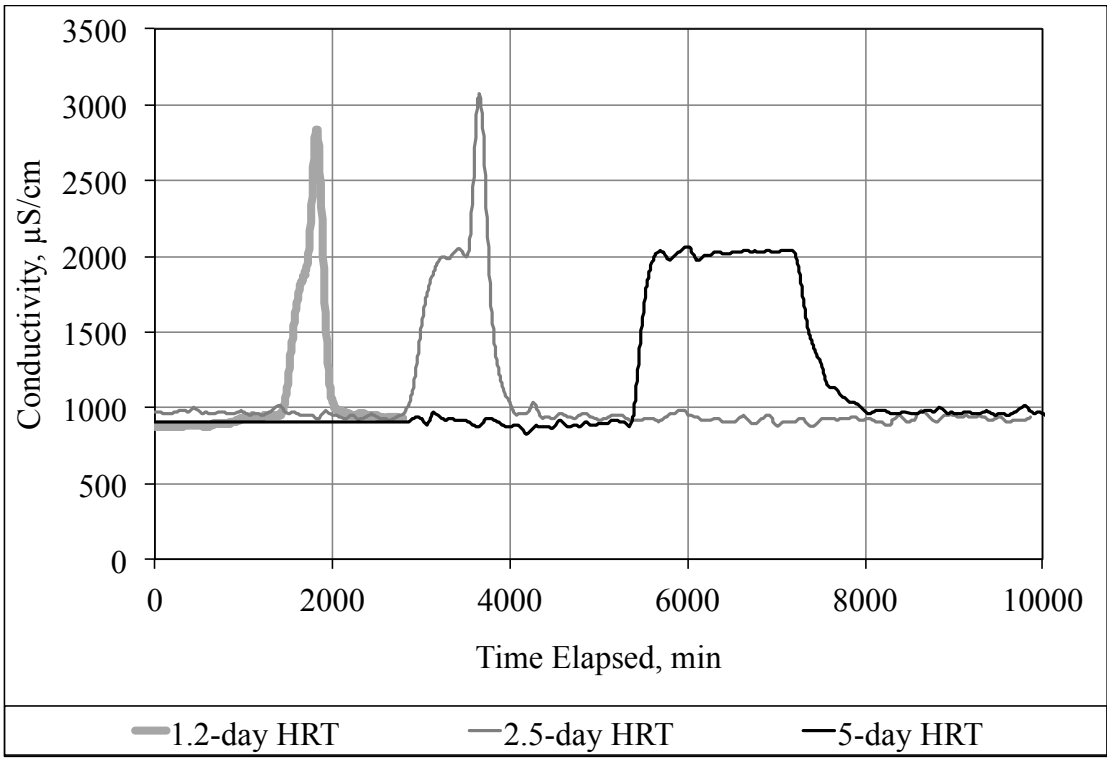

Figure 3.12. Conductivity data for 1, 2.5, and 5 day residence times for preliminary tracer tests in Cl_2 Column #1.

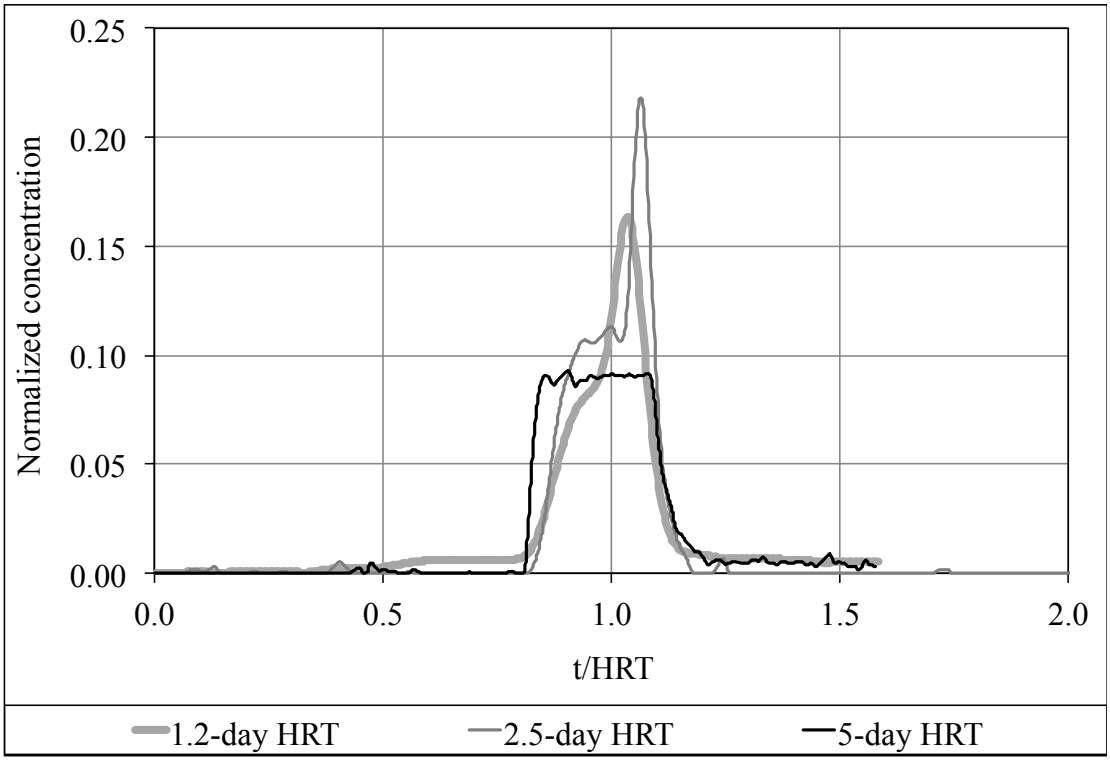

Figure 3.13. Normalized curves for Cl_2 Column #1.

Given this finding, tracer tests were completed on all columns at a residence time of 5 days. After the completion of the tracer tests, the systems were given 3 weeks to recover from any bacterial static effects of the tracer that could have resulted in depressed removals. During this recovery period, Cl_2 Column #1 and O_3 Column #1 were operated at a 5-day HRT, and Cl_2 Column #2 and O_3 Column #2 were operated at a 25-day HRT.

Raw tracer test data and normalized data are provided in Figures 3.14 and 3.15, respectively.

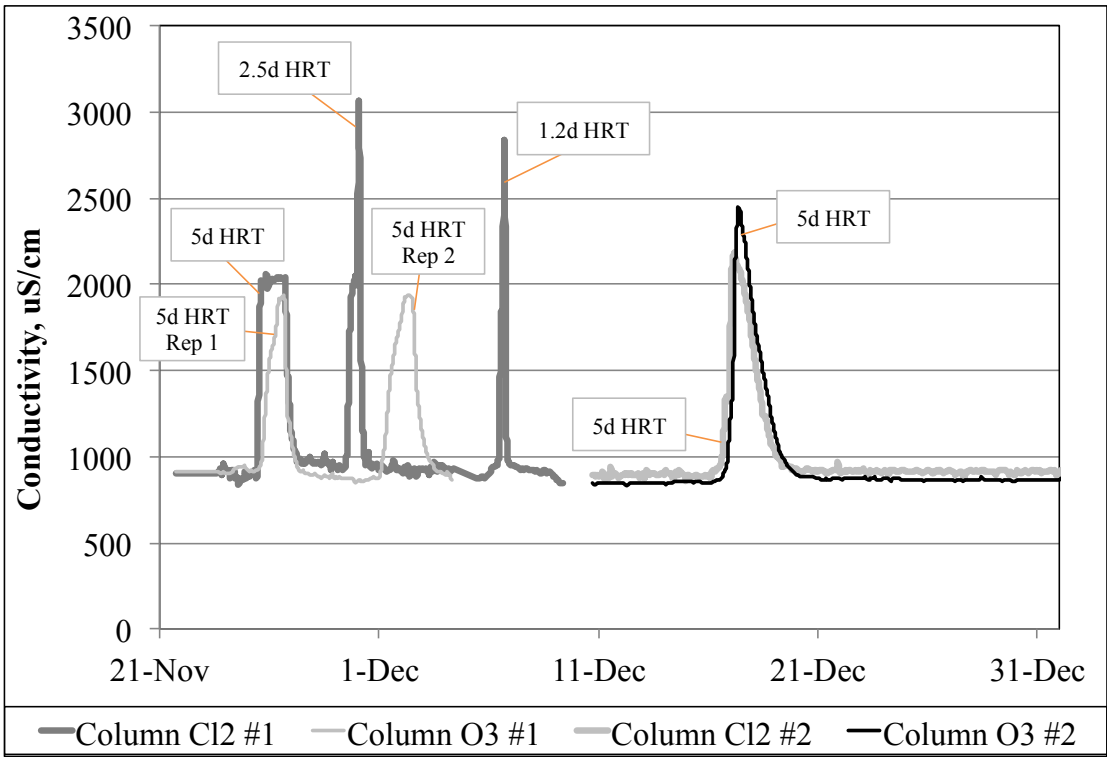

Figure 3.14. Raw conductivity data from tracer tests.

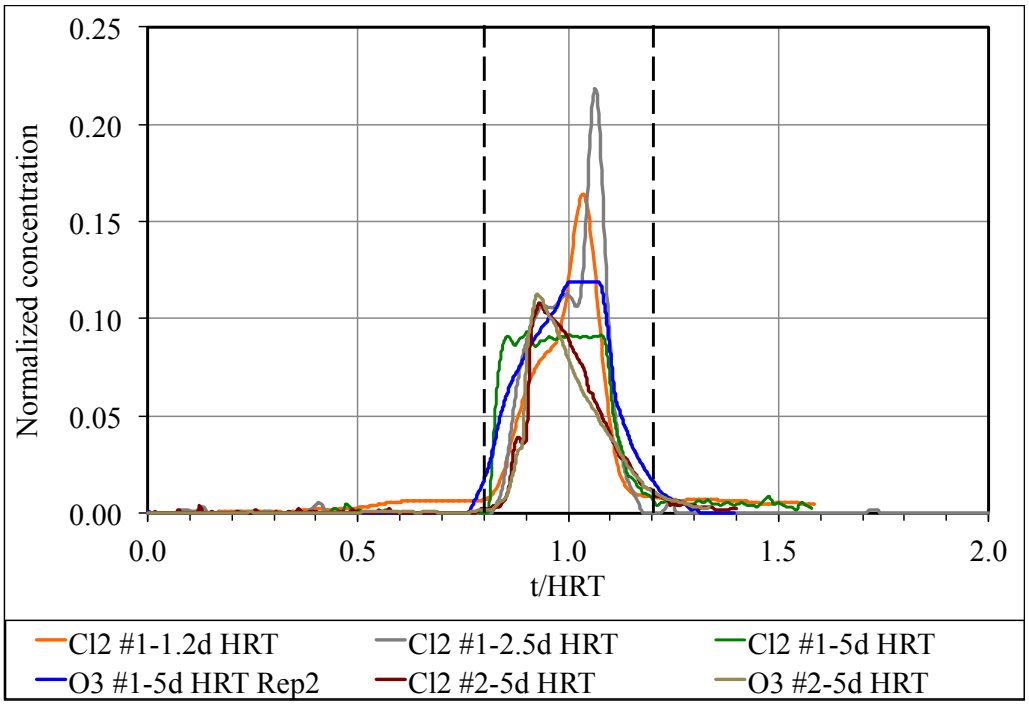

Figure 3.15. Normalized curves for all tracer tests.

The measured HRT, modal time (the time elapsed between tracer addition and the observation of the highest tracer concentration exiting the system), and T_{10} (the time elapsed between tracer addition and the time when 10% of the tracer has exited the system) are summarized in Table 3.4.

Table 3.4. Tracer Test Results for HRT, Modal Time, and T_{10}

	HRT (theoretical)	HRT (measured)	Modal Time	T10
		Days		
Cl_2 Column #1 downflow				
Rep 1	1.2	1.2	1.3	1.0
Rep 2	2.5	2.4	2.5	2.1
Rep 3	4.4	4.6	4.2	3.9
Cl_2 Column #2 upflow				
Rep 1	5.1	7.0	6.5	6.3
O_3 Column #1 downflow				
Rep 1	4.4 (rep 1)	4.6	4.9	4.2
Rep 2	4.9 (rep 2)	4.6	4.8	4.0
O_3 Column #2 upflow				
Rep 1	5.5	7.2	6.7	6.5

For the upflow columns, a systematic bias was observed between the theoretical HRT and the measured HRT. It is unclear why the difference was observed for the upflow columns but not for the downflow columns. Notable design and operational differences were the direction of flow and the diameter (downflow columns were 8 in. in diameter, whereas upflow columns were 6 in. in diameter). Two driving forces may contribute to the differences: (1) expansion of sand caused by the upflow regime, which resulted in greater sand depth and higher porosity and (2) convection from gravity because the salt solution is slightly denser than water and would act in opposition to convective forces from velocity. It was confirmed that the sand depth had not changed since the columns were loaded. If convection that was due to gravity was a factor, the inverse effect should have been observed with the downflow columns (measured HRT < theoretical HRT), but this was not the case. The reason behind this observation remains unexplained. For the purposes of this discussion, the theoretical HRT is used throughout the document.

Chapter 4

Phase 2: Bulk Organic Carbon, DBP, and CEC Attenuation

The following chapter provides results from Phase 2: bulk organic carbon, DBP, and CEC attenuation through SAT. Within the bulk organic carbon discussion, TOC, UVT, SUVA, EEM, BDOC, and the other routine water quality parameters (e.g., nitrogen, phosphate, and pH) are evaluated. This discussion is followed by the DBP results, and lastly, CEC monitoring outcomes are presented.

4.1 Bulk Organic Carbon

4.1.1 TOC and UVT

During Phase 2, Column #1 from each Soil Column System was operated in a fashion that represented the vadose zone. Vadose zone operations were modeled by maintaining a constant water depth at the top of the soil column for 21 days and then allowing the water to percolate through the soil column while ceasing feed water for a draining and drying period of 7 days. This sequence was repeated for the duration of this phase. Samples taken of the soil column feed and effluent were paired. The feed was tracked through the system, and a representative effluent sample was collected based on when the sampled feed was expected to exit the system.

On average, the Soil Column System being fed chlorinated water attained a final TOC effluent concentration of 3.3 mg/L, which equated to 45% removal. The Soil Column System being fed ozonated effluent attained on average a final TOC effluent concentration of 2.1 mg/L, which equated to 62% removal. The average UVT in the final effluent was 86% for the tertiary system and 92% for the ozonated system. Average TOC, UVT, and SUVA data are summarized in Table 4.1.

Raw TOC data collected during this phase for the Cl_2 and O_3 Soil Column Systems are plotted in Figures 4.1 and 4.2, respectively.

Table 4.1. Summary of TOC Data During Phase 2

Location	Parameter				
	TOC, mg/L Mean ± Std. Dev.	TOC Diff., mg/L	TOC Removal, %	UVT, % Mean ± Std. Dev.	SUVA, L/mg-M
Secondary	5.60 ± 0.73	n/a	n/a	78.0 ± 2.2	2.03
Cl_2 Disinfection Alternative					
Post-Chlorination	5.90 ± 0.29	n/a	n/a	77.0 ± 1.9	1.86
After Vadose	3.50 ± 0.33	2.4	40	85.0 ± 3.1	1.96
After Saturated	3.30 ± 0.29	0.3	7	86.0 ± 2.2	2.12
Overall SAT	n/a	2.6	45	n/a	n/a
O_3 Disinfection Alternative					
Post-Ozonation	5.50 ± 0.47	n/a	n/a	87.0 ± 2.0	1.11
After Vadose	2.60 ± 0.30	2.9	53.2	91.0 ± 3.6	1.64
After Saturated	2.10 ± 0.16	0.48	18.8	92.0 ± 2.7	1.74
Overall SAT	n/a	3.4	62	n/a	n/a

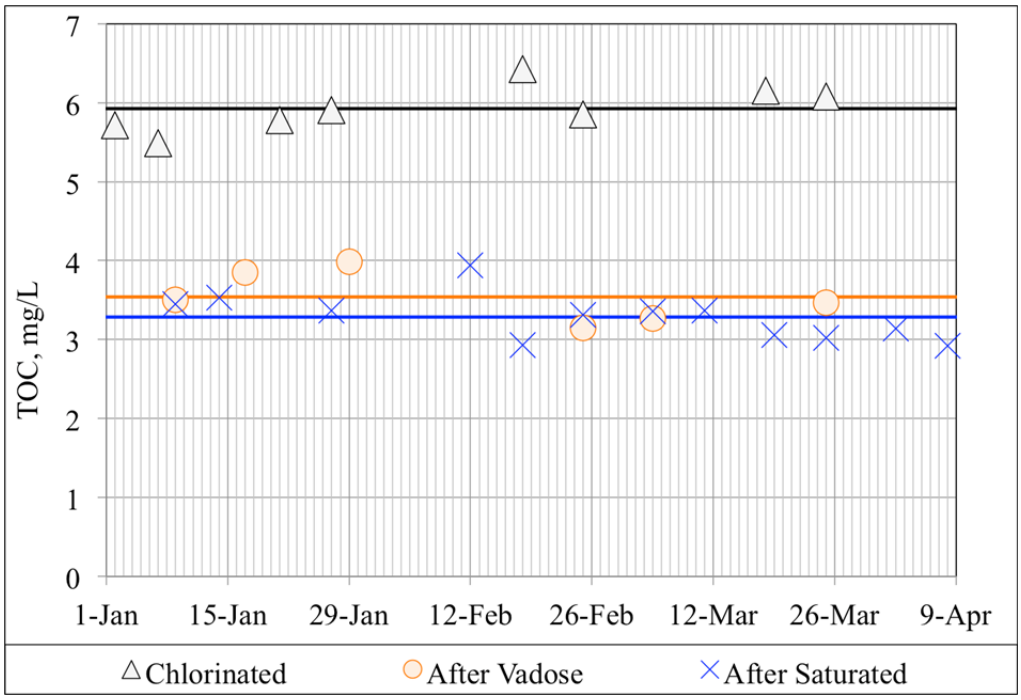

Figure 4.1. Overall Cl_2 Soil Column System performance based on TOC during Phase 2 (bulk organic carbon, DBP, and CEC monitoring); solid horizontal lines represent averages.

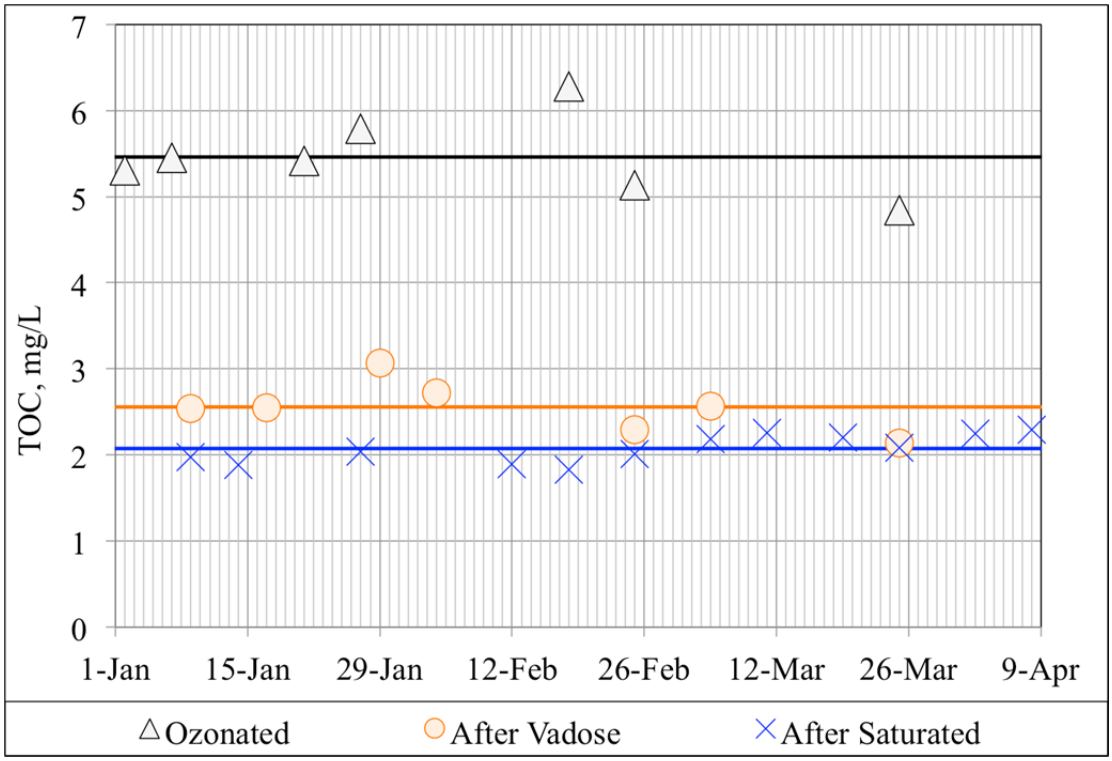

Figure 4.2. Overall O_3 Soil Column System performance based on TOC during Phase 2 (bulk organic carbon, DBP, and CEC monitoring); solid horizontal lines represent averages.

Biodegradation of bulk organic material, such as TOC or DOC, is typically observed within the first week of subsurface travel, as shown in Figures 4.1 and 4.2 and as documented by Rauch-Williams et al. (2010), Mansell and Drewes (2004), Drewes and Fox (1999), Drewes and Jekel (1998), and Fox et al. (2006). A detailed summary from the published literature is provided in the second table in Appendix A: Summary of Laboratory- and Field-Scale Studies.

Some of the variation observed in the ozonated water is explained by the fact that during the soil column testing, different pretreatment alternatives prior to ozonation were being examined (as part of WRRF-11-02). The pretreatment step was absent from December 5, 2013, to February 18, 2014, so this was the only period when the ozone columns received ozone-treated secondary effluent. Ultrafiltration served as the pretreatment from September 13 to December 3, 2013, and from February 19 to March 7, 2014; and microfiltration served as the pretreatment from March 10 to May 28, 2014. TOC removal by low-pressure membranes was measured at 10 to 15%. The higher TOC values correlated with the piloting configuration that did not include a pretreatment step. Even with these variations in influent TOC for the O_3 Soil Column System, the final effluent from this Soil Column System was consistent, as shown in Figure 4.2. Average TOC data from Phase 2 is summarized in Figure 4.3.

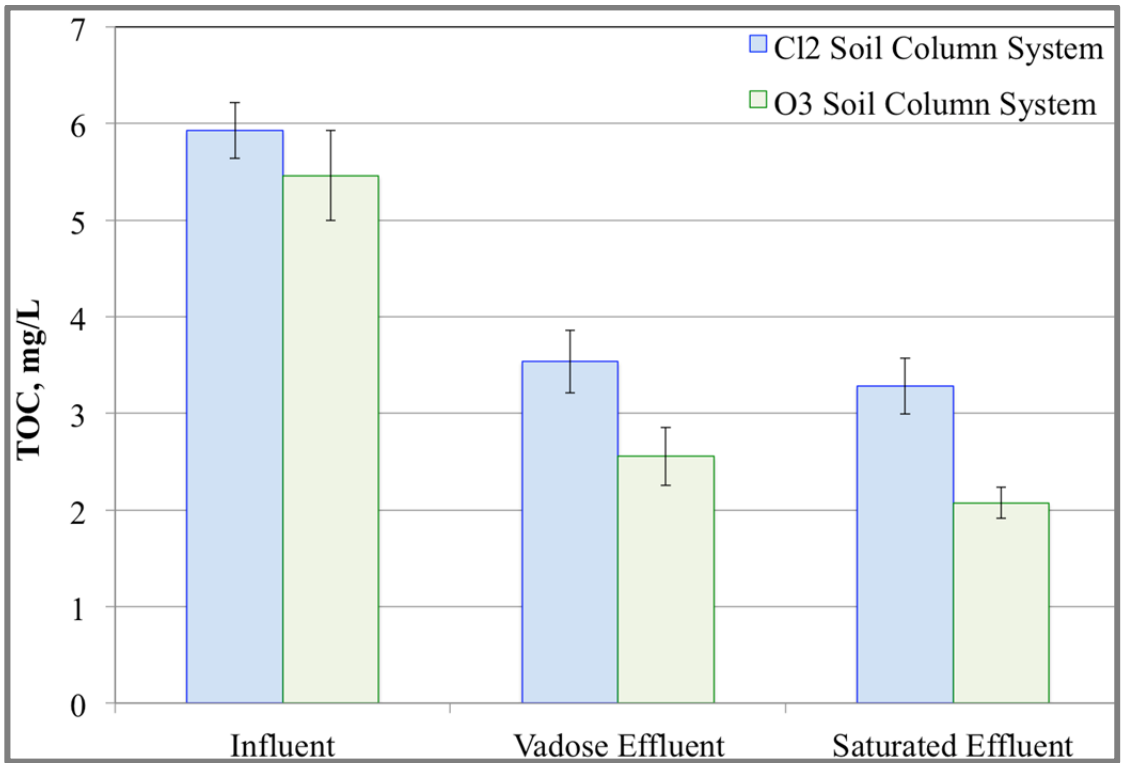

Figure 4.3. Average TOC data from Phase 2 (Oct 24, 2013–Dec 23, 2013).

Note: Error bars represent standard deviation

As previously discussed, multiple studies have also shown that the majority of the TOC is reduced by SAT within relatively short travel times, on the order of days. In addition, TOC is a parameter that is indicative of the presence of DBP precursors, and it is particularly important to maintain bulk organic carbon at low levels to prevent DBP formation during treatment for potable reuse. Ozonation enhances TOC reduction through SAT and aids in reducing DBP formation when the groundwater is later extracted and disinfected for drinking water purposes.

UVT was also monitored during Phase 2. Raw UVT data is shown in Figures 4.4 and 4.5 for the Cl_2 and O_3 Soil Column Systems, respectively.

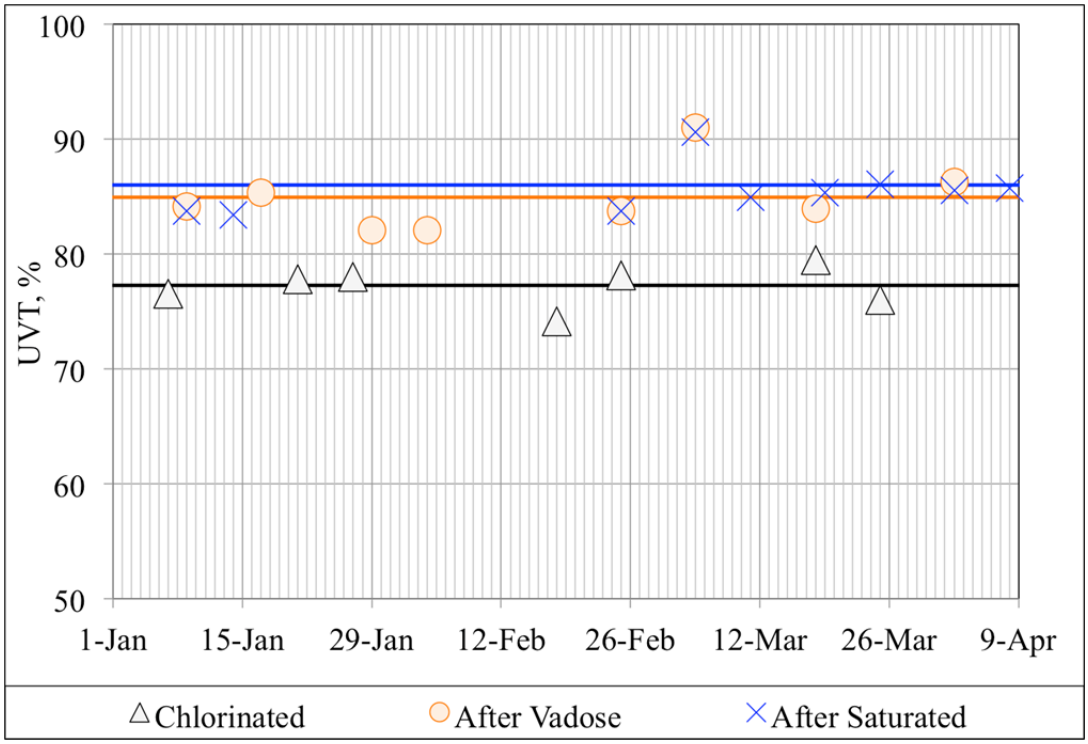

Figure 4.4. Overall Cl_2 Soil Column System performance based on UVT during Phase 2; solid horizontal lines are averages.

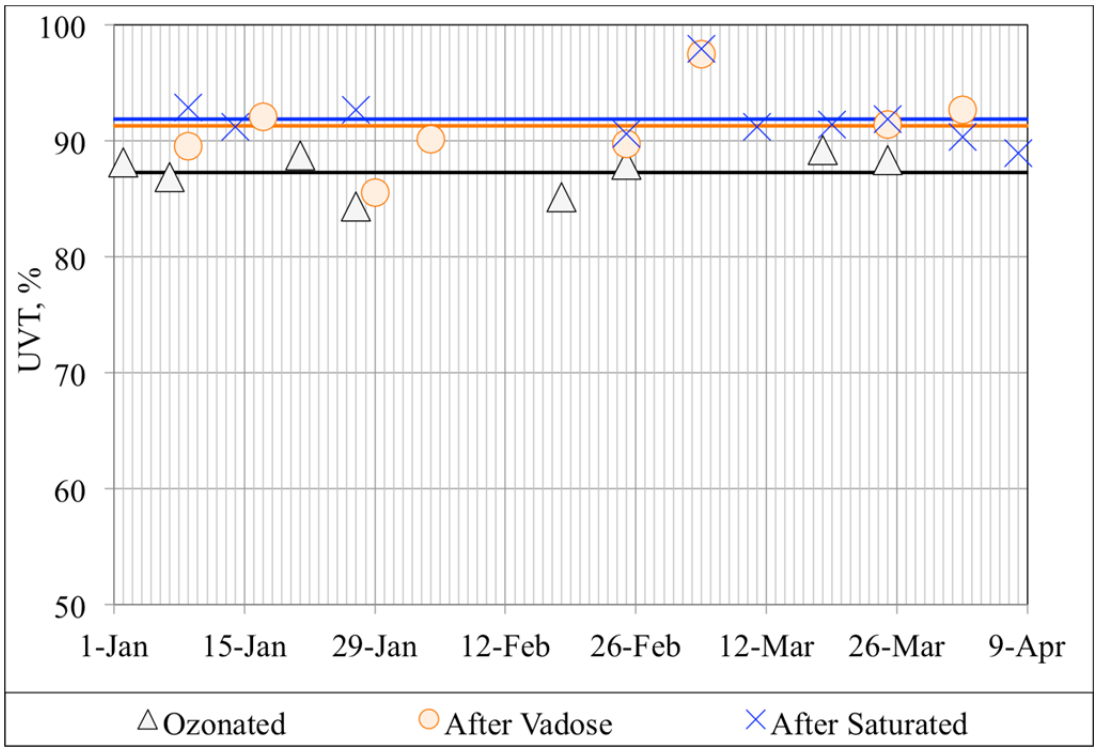

Figure 4.5. Overall O_3 Soil Column System performance based on UVT during Phase 2; solid horizontal lines are averages.

The UVT percentage of the Cl_2 Soil Column System influent was in the high 70s, and that of the O_3 Soil Column System influent was in the high 80s. The majority of UVT improvement occurred within the vadose columns. On average, UVT increased from 77% to 85% and 87% to 91% for the Cl_2 and O_3 vadose columns, respectively. Minor improvements were observed through the saturated columns. On average, UVT increased from 85% to 86% and 91% to 92% for the Cl_2 and O_3 saturated columns, respectively. The influent of the O_3 Soil Column System, ozonated secondary effluent, had a higher initial UVT than the final effluent from the Cl_2 Soil Column System. Overall, the O_3 Soil Column System, as compared with the Cl_2 Soil Column System, was able to achieve a higher quality final effluent with regard to UVT.

Within the 30-day travel time tested in this study, the TOC and UVT of the effluent from the O_3 Soil Column System were better than the TOC and UVT of the effluent from the Cl_2 Soil Column System. The improved removal is likely due to the organics in ozonated water being more amenable to biodegradation through the soil columns, thus resulting in faster kinetics.

There is potential for further removal with an extended subsurface travel time for the Cl_2 Soil Column System. An existing surface application project in California sponsored by IEUA used a combination of lysimeters and monitoring wells to monitor TOC degradation at the full scale during project startup. During a portion of the startup, IEUA basins were operated with only recycled water, and lysimeters were used to extract the infiltrated recycled water from the vadose zone at various depths. The resulting TOC of the water extracted using the deepest lysimeter ranged from 1.0 to 2.1 mg/L. For example, after approximately 3 months of subsurface travel, both Victoria and San Sevaine Basins demonstrated 78% TOC removal (Campbell et al., 2011).

4.1.2 EEM: Wastewater Fingerprint

An introduction to and a background on EEM fingerprinting are provided at the end of Section 1.2.1. EEM data from this study was used to formulate a 3-D plot, which was then used to analyze the composition of the organic matter.

EEM results from this study are provided in Figure 4.6. Given that wastewater exhibits inherent variability over time, both the EEM fingerprints from individual grab samples and the average of those grab samples are provided for comparison purposes. The y-axis is the excitation wavelength, the x-axis is the corresponding emissions wavelength, and the emission intensity is represented by contours and colors, with dark blue being low concentrations and red being high concentrations. As evident in the far right column (the average of the grab samples) in Figure 4.6, disinfection significantly transforms the organic matter. The characteristic EfOM in wastewater (shown as a high fluorescence intensity at the 260–290 nm excitation and 320–370 emission wavelength ranges) is substantially diminished, especially after ozonation.

With ozonation, this peak diminishes such that the fingerprint of the organic matter begins to resemble that of natural water (Seong-Nam et al., 2008). Ozonation cleaves and oxidizes organic carbon of wastewater origin, eliminating the fluorescent signal and characteristic wastewater fingerprint. Also evident, and supported by other studies, is the fact that all peaks within the Cl_2 Soil Column System gradually diminish and look less like wastewater as the water spends more time in the subsurface. The vadose zone alone significantly reduces the wastewater fingerprint after only 5 days of subsurface travel. In the case of the ozone system, bioactivity did not further attenuate fluorescence but rather caused humics to re-appear, possibly byproducts of metabolic processes but not of wastewater origin (Seong-Nam et al.,

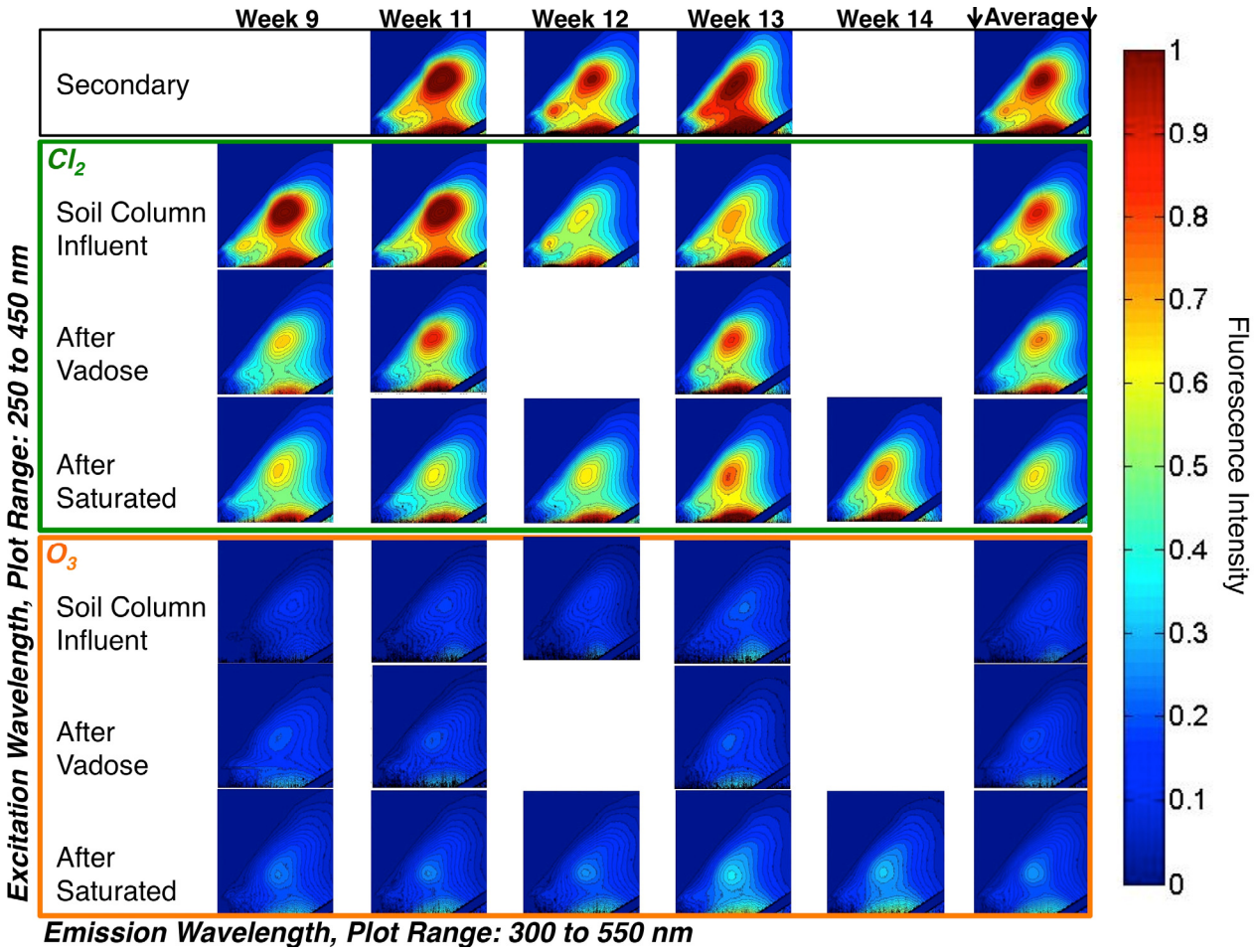

Figure 4.6. EEM data through the Soil Column Systems; y-axis is excitation wavelength, ranging from 250 to 450 nm; x-axis is emissions wavelength, ranging from 300 to 550 nm.

4.1.3 BDOC

The BDOC reactors (Figure 4.7) achieved acclimation within two months. Acclimation was defined as the point when four consecutive samples had a standard deviation of 5% or less. Acclimation data are provided in Table 4.2.

Figure 4.7. BDOC reactor with acclimated sand and water sample of interest.

Table 4.2. BDOC during BDOC Reactor Acclimation

		BDOC (equal to percentage DOC reduction)		
Date	**Duration, Days**	**Secondary**	**Chlorinated**	**Ozonated**
15-Sep	5	4	7	14
20-Sep	5	16	15	16
25-Sep	5	-6	-2	13
2-Oct	7	18	4	13
9-Oct	7	17	7	22
18-Oct	7	16	15	24
25-Oct	7	15	14	35
31-Oct	7	16	17	42
4-Nov	7	19	17	41
12-Nov	8	16	22	35
20-Nov	10	22	20	44
Statistics for samples from Oct 25 to Nov 12	Average, %	**16**	**18**	**38**
	Standard deviation, %	**2**	**3**	**4**

To coordinate timing with CEC monitoring, BDOC testing began 3 months after reactor acclimation. Thirty-day BDOC data are summarized in Table 4.3, and the reported average is calculated from the initial and final DOCs, provided in Table 4.4.

Table 4.3. Thirty Day BDOC Data Collected During Phase 2

Date	BDOC		
	Secondary	Chlorinated	Ozonated
27-Jan	41%	26%	56%
4-Feb	49%	44%	62%
18-Feb	44%	39%	56%
4-Mar	45%	41%	54%
17-Mar	41%	40%	56%
27-Mar	37%	39%	60%
Average	43%	38%	58%
Standard Deviation	4%	6%	3%

Table 4.4. Average DOCs Used to Compute Average 30-Day BDOC

	Initial DOC, mg/L	30-day DOC, mg/L	BDOC, %
	Average ± Standard Deviation	Average ± Standard Deviation	
Secondary	5.40 ± 0.26	3.10 ± 0.20	43
Chlorine column influent	6.00 ± 0.25	3.70 ± 0.31	38
Ozone column influent	5.40 ± 0.42	2.30 ± 0.19	58

As expected, BDOC is higher for the ozonated water because ozonation cleaves the organic carbon, making it more bio-accessible. BDOC is comparable to the DOC removal observed through the soil columns, as shown in Figure 4.8.

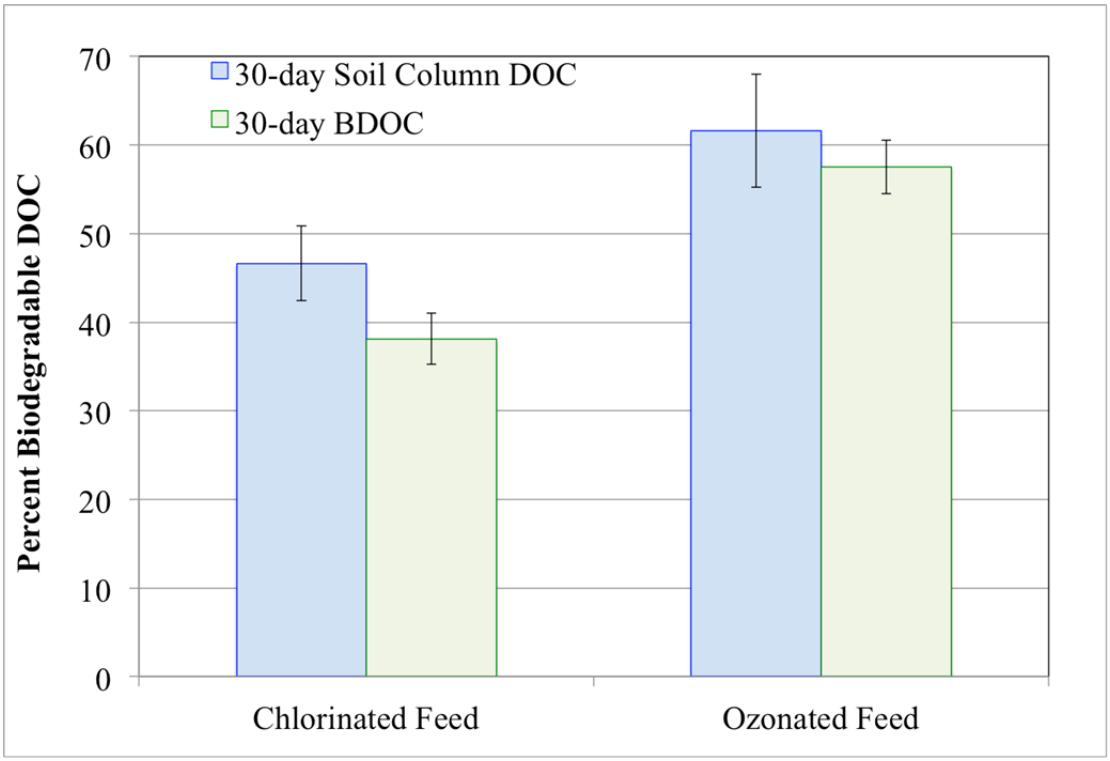

Figure 4.8. Comparison of DOC removal through soil columns and BDOC; errors bars represent standard deviation.

The data shown in Figure 4.8 indicate that BDOC is potentially an effective and economical tool that can serve as a conservative predictor of SAT performance. However, this observation is limited to DOC removal. The DOC reductions based on BDOC reactors and soil columns were indistinguishable when standard deviation is considered (i.e., the standard deviations overlapped in both the chlorine and ozone systems).

4.1.4 Miscellaneous Routine Water Quality Parameters

Miscellaneous routine water quality data from Phase 2 are summarized in Table 4.5.

Table 4.5. Miscellaneous Routine Water Quality Data

Water Quality Parameter	Cl_2 Soil Column System			O_3 Soil Column System		
	Influent	Vadose Effluent	Saturated Effluent	Influent	Vadose Effluent	Saturated Effluent
Ammonia, mg/L as N	0.79	<0.08	<0.08	0.49	<0.08	<0.08
Conductivity, µS/cm	900	903	902	912	882	937
Nitrate, mg/L as N	2.93	3.57	0.37	3.18	3.53	0.63
Nitrite, mg/L as N	0.013	<0.005	<0.005	0.011	0.006	0.008
pH, pH units	7.1	7.5	8.0	7.1	7.5	8.2
Orthophosphate, mg/L as PO4	2.48	1.90	0.44	2.44	1.73	1.88
Total Nitrogen, mg/L as N	3.73	3.57	0.37	3.68	3.54	0.64
Turbidity, NTU	0.65	0.57	0.51	0.35	0.38	0.30

As a whole, total nitrogen decreased through both Soil Column Systems, as did phosphate and turbidity. Decreases in ammonia, nitrate, and nitrite indicate the presence of both oxic and anoxic zones within the soil column. Phosphate was likely attenuated by the existence of zones supporting organisms capable of accumulating phosphate. More phosphate appears to have been attenuated in the saturated Cl_2 Soil Column System than in the saturated O_3 Soil Column System. The reason for this may have been the higher oxygen levels in the ozonated water as compared with the Cl_2 Soil Column System, thus making the O_3 Soil Column System less able to establish and support anaerobic organisms. It is well known that phosphate-accumulating organisms require anaerobic conditions (Tchobanoglous et al., 2004). The difference in phosphate attenuation could also be an analytical artifact.

4.2 DBPs

All DBPs were completely removed in both SAT Soil Column Systems. NDMA values measured in the influent of the Cl_2 Soil Column System were more variable than the values measured in the O_3 Soil Column System influent. Through the first two sampling events, the Cl_2 water collected from SJCWWRP was not quenched with sodium bisulfite (SBS) at the time of collection but rather 1 to 3 hours after collection, leading to the formation of NDMA. After observing the high NDMA concentration that resulted from this practice, subsequent Cl_2 samples were immediately quenched with SBS to remove any residual chlorine and prevent further NDMA formation. NDMA values measured from the vadose column effluent for both Soil Column Systems decreased over time, suggesting that acclimation continued to occur for the degradation of complex organic compounds. NDMA concentrations in both vadose column effluents were below the detection limit during the last two sampling events (Table 4.6).

Table 4.6. NDMA Data Collected During Phase 2

	Sampling Event								Mean
	1	2	3	4	5	6	7	8	
Secondary		95							95
Cl_2 Soil Column System									
Influent		1100			410	340	190	200	448
After Vadose		150			32	19	<2.0	<2.0	41
After Saturated Rep 1	<2.0		2.2	<2.0	<2.0				<2.0
After Saturated Rep 2		<2.0	<2.0	<2.0					<2.0
O_3 Soil Column System									
Influent		640			290	310	300	380	384
After Vadose		9.5			<2.0	<2.0	<2.0	<2.0	3.5
After Saturated Rep 1	<2.0		<2.0	<2.0	<2.0				<2.0
After Saturated Rep 2		<2.0		<2.0					<2.0

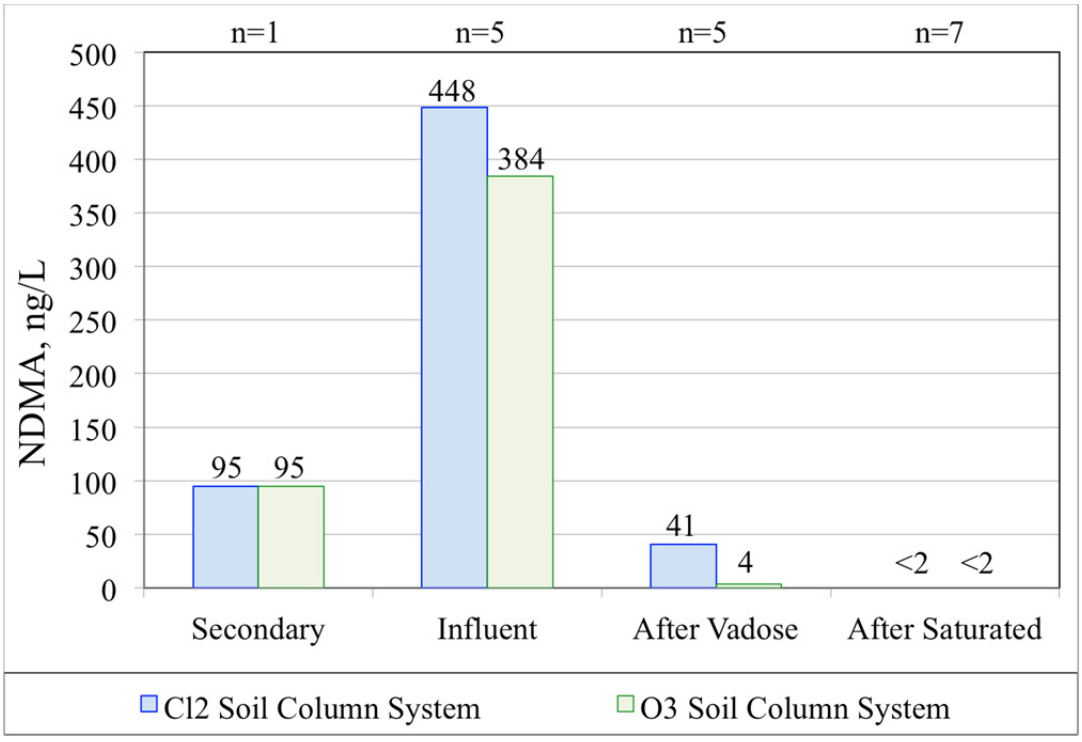

Figure 4.9. Average NDMA removal.

NDMA is reliably removed through SAT via co-metabolism by in situ soil biota (Bradley et al., 2005; Gunnison et al., 2000; Zhou et al., 2009; Nalinakumari et al., 2010; Sharp et al., 2005). Several studies, including this one, support this finding and are summarized in Table 4.7.

The average concentrations of additional DBPs specific to chloramination and ozonation are presented in Table 4.8. (The concentrations of the full suite of nitrosamines, HAAs, and THMs are provided in Appendix D.)

Table 4.7. Comparison of NDMA Removals through SAT

Test System	HRT, days	$NDMA_o$, ng/L	$NDMA_f$, ng/L	NDMA Removal	Source
Cl_2 Soil Column System (oxic + anoxic)	30	448	<2	>99.6%	This study
O_3 Soil Column System (oxic + anoxic)	30	384	<2	>99.5%	This study
Soil columns (oxic)	20	888	40	95.50%	Nalinakumari et al., 2010
Soil columns (anoxic)	20	934	248	73.40%	Nalinakumari et al., 2010
Soil columns (oxic)	6	140	<2	>98.6%	Drewes et al., 2006
Soil columns (anoxic)	25	120	<2	>98.3%	Drewes et al., 2006
Riverbank filtration	5	5.4 to 6.1	<2	>63.0%	Drewes et al., 2006

Table 4.8. DBP Concentrations

	Sampling Event 1	Sampling Event 2	Sampling Event 3	Average
Cl_2 Soil Column System				
HAAs				
Influent	30	36	15	27
Saturated	<1.0	<1.0	<1.0	<1.0
TTHMs				
Influent	34.7	24.3	15.1	24.7
Saturated	<0.50	<0.50	<0.50	<0.50
O_3 Soil Column System				
Bromate				
Influent	5.5	<0.50	1.4	2.5
Saturated	<0.50	<0.50	<0.50	<0.50

For the Cl_2 Soil Column System, HAAs and TTHMs were attenuated to non-detect (ND) levels during all sampling events. For the O_3 Soil Column System, bromate was also reduced to ND levels during all sampling events. Previous research has shown that HAAs and TTHMs are effectively removed via biodegradation (Amy et al., 1996; Wilson et al., 1995). Bromate removal requires anoxic conditions in which bromate serves as an electron acceptor similar to nitrate. As previously discussed, nitrate removal was consistently observed (Section 4.1.4), thus providing evidence that anoxic conditions developed within the biofilms of the Soil Column Systems.

4.3 CECs

The overall average value for each CEC evaluated, across all sampling events for each sample location, is provided in Table 4.9. A complete set of data depicting the occurrence of CECs and summary statistics are tabulated for each sample location in Appendices E and F, respectively.

Table 4.9. Average CEC Concentrations at All Sampling Locations

Compound Name	Concentration, ng/L						
	After Secondary	After Disinfection		After Vadose		After Saturated	
		Cl_2	O_3	Cl_2	O_3	Cl_2	O_3
4-Nonylphenol (tech mix)[1]	220	143	176	1,620	1,513	2,796	1,200
4-tert Octylphenol[1]	23	20	22	1,735	1,380	1,415	717
Acesulfame-K	1,142	411	343	521	301	171	56
Acetaminophen[1]	10	18	<10	<10	<10	<10	<10
Atenolol	140	93	<30	32	<24	<28	<28
Atorvastatin	<10	<10	<10	<10	<10	<10	<10
Azithromycin	149	127	<10	<10	<10	<10	<10
Bisphenol A[1]	<10	28	386	10	<10	<10	<10
Caffeine	<10	<10	<10	<10	<10	<10	<10
Carbamazepine	230	207	<10	262	<10	232	<10
Carisoprodol	199	269	86	238	81	349	76
Diazepam	<10	<10	<10	<10	<10	<10	<10
DEET[1]	52	198	18	155	14	140	25
Diclofenac	224	86	<10	15	<10	11	<10
Dilantin	259	157	42	165	28	156	17
Erythromycin-H2O	44	31	<10	11	<10	<10	<10
Fipronil	51	37	14	39	13	28	6
Fluoxetine	45	34	<10	<10	<10	<10	<10
Furosemide	425	176	<10	11	<10	11	<10
Galaxolide	2,817	2,400	234	<50	<50	<50	50
Gemfibrozil	150	192	<10	<10	<10	12	<10
Ibuprofen	16	11	<10	48	13	<10	<10
Iohexol	8,785	7,950	4,520	620	485	1,300	198
Iopromide[1]	224	265	104	12	11	27	10
Meprobamate	361	328	177	351	160	252	91
Metoprolol	501	385	10	194	<10	<10	<10
Naproxen	25	22	<10	<10	<10	<10	<10
Nonylphenol diethoxylate[1]	230	222	113	<25	<25	78	52
Nonylphenol monoethoxylate[1]	425	367	27	<25	<25	103	65

Table 4.9. Average CEC Concentration at All Sampling Locations (*continued*)

Compound Name	Concentration, ng/L						
	After Secondary	After Disinfection		After Vadose		After Saturated	
		Cl_2	O_3	Cl_2	O_3	Cl_2	O_3
Octylphenol diethoxylate[1]	29	29	<25	<25	<25	<27	<25
Octylphenol monoethoxylate[1]	139	115	<25	<25	<25	50	40
Primidone	211	184	47	211	46	196	32
Propranolol	63	38	<10	11	<10	<10	<10
Sucralose[1]	25,200	30,214	18,143	32,157	17,543	29,388	16,013
Sulfamethoxazole[1]	1,927	370	11	718	61	410	22
TCEP	340	365	319	369	279	358	258
TCPP[1]	2,982	3,823	3,220	676	477	1,363	551
TDCPP	545	534	503	216	163	218	126
Tonalide	109	102	<50	<50	<50	<50	<50
Triclocarban	76	68	<10	<10	<10	<10	<10
Triclosan	103	22	<10	<10	<10	<10	<10
Trimethoprim	172	57	<10	16	<10	<10	<10

Notes: Some CECs showed apparent formation after one or more of the treatment steps (disinfection, vadose, and/or saturated); statistically significant increases (student t-test with alpha = 0.05) are explored and discussed in Appendix G

Of the 42 CECs tested, 38 CECs were present in the secondary wastewater. Atorvastatin, bisphenol A, caffeine, and diazepam were not detected. Chlorine disinfection did not further reduce any CECs completely. The Cl_2 Soil Column System resulted in the complete removal of 15 CECs (acetaminophen, atenolol, azithromycin, erythromycin, fluoxetine, galaxolide, ibuprofen, metoprolol, naproxen, octylphenol diethoxylate, propranolol, tonalide, triclocarban, triclosan, and trimethoprim). Ozone disinfection reduced 18 CECs completely (acetaminophen, atenolol, azithromycin, carbamazepine, diclofenac, erythromycin, fluoxetine, furosemide, gemfibrozil, ibuprofen, naproxen, octylphenol monoethoxylate, octylphenol diethoxylate, propranolol, tonalide, triclocarban, triclosan, and trimethoprim). The O_3 Soil Column System resulted in the complete removal of 3 additional CECs (galaxolide, iopromide, and metoprolol). Nonylphenols are known to have extremely complex metabolic pathways in the environment that make data difficult to interpret (Montgomery-Brown et al., 2008) [With the soil columns, 4-nonylphenol was likely a biodegradation product as it was not present in the secondary wastewater but was present in biodegraded effluents. Drewes et al. (2011) also observed such an increase on one occasion.] The efficacy of the O_3 Soil Column System is driven by the ozone disinfection step. A comparison of overall treatment (secondary wastewater compared with final soil column effluents) is provided in Figure 4.10.

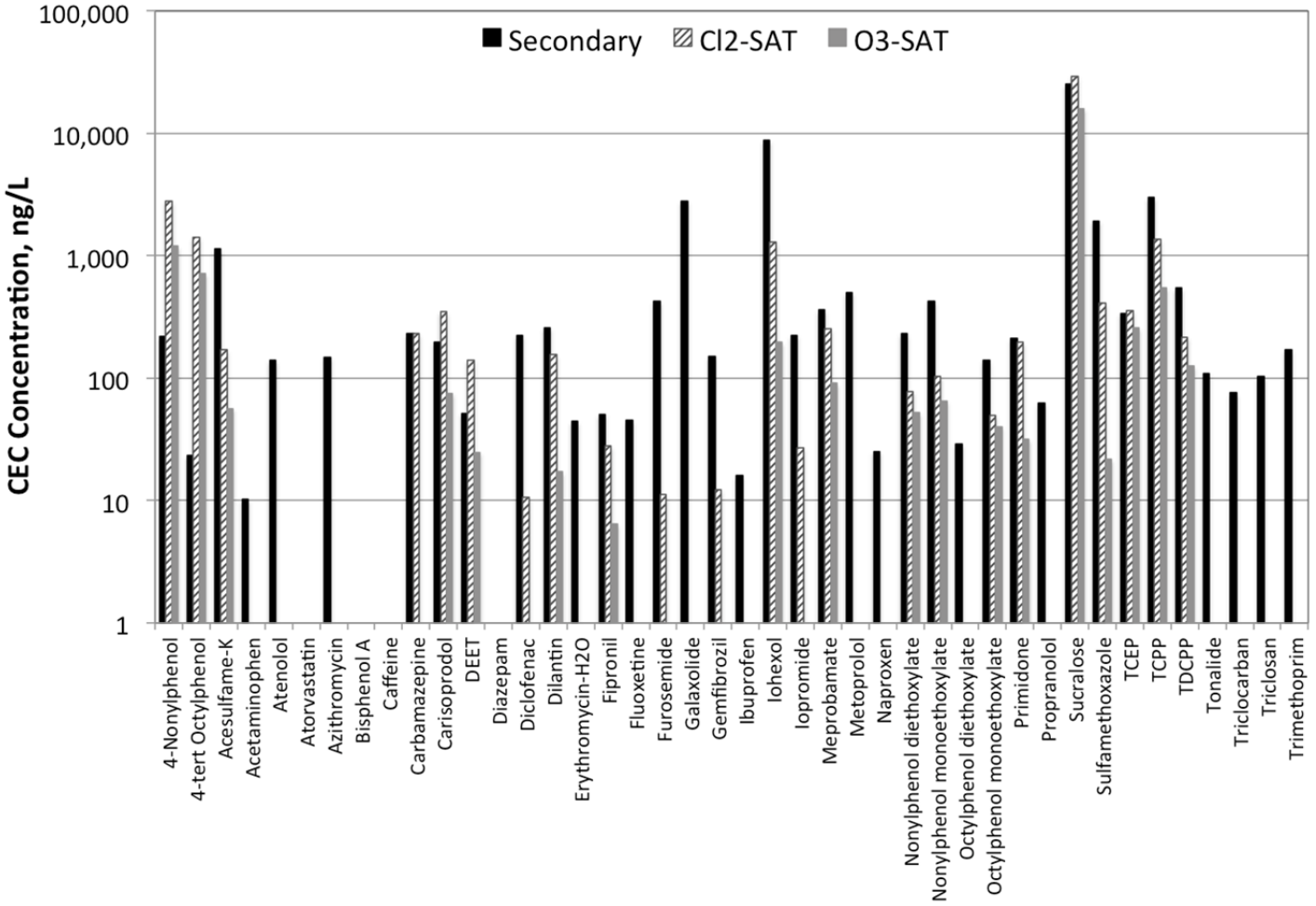

Figure 4.10. CEC concentrations in secondary wastewater and final effluents from the Cl_2 Soil Column System and O_3 Soil Column System; no bar indicates that the level was below the detection limit.

As shown in Figure 4.10, the CEC concentrations in the O_3 Soil Column System effluent were lower than the concentrations observed in the Cl_2 Soil Column System. Notably, the O_3-SAT alternative completely removed the same CECs as the Cl_2-SAT alternative in addition to carbamazepine, diclofenac, furosemide, gemfibrozil, iopromide, and octylphenol monoethoxylate.

Among the various classes of CECs explored (pharmaceuticals, antibiotics, fire retardants, detergent metabolites, artificial sweeteners, and household chemicals classes), fire retardants, artificial sweeteners, and detergent metabolites were found to be the most difficult classes of CEC to address via SAT (Figure 4.11, which is Figure 4.10 divided into six CEC categories).

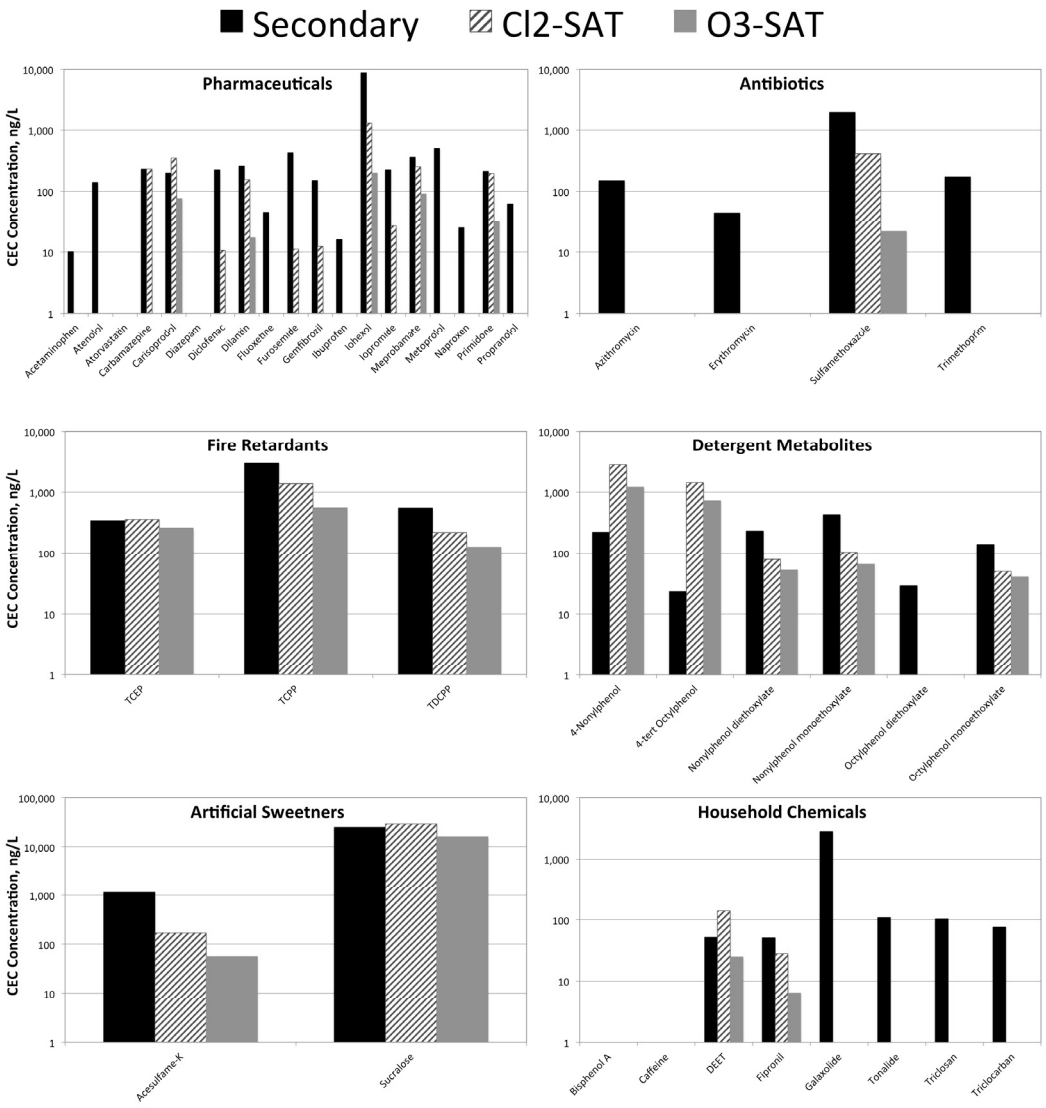

Figure 4.11. CEC concentrations in secondary wastewater and final effluents from the Cl_2 Soil Column System and O_3 Soil Column System for six different CEC categories; no bar indicates that the level was below the detection limit.

Another way to compare the two disinfection alternatives in conjunction with SAT is in graphical form. A template is provided in Figure 4.12. In such a plot, the removal of CECs is plotted for two treatment systems simultaneously; the location of a given data point indicates the effectiveness of the treatments being compared. CECs that lie on the diagonal line are those CECs that are equally removed by both treatment systems. CECs that lie above the diagonal line are those CECs that are better removed by the process on the y-axis. CECs that lie below the diagonal line are those compounds better removed by the process on the x-axis.

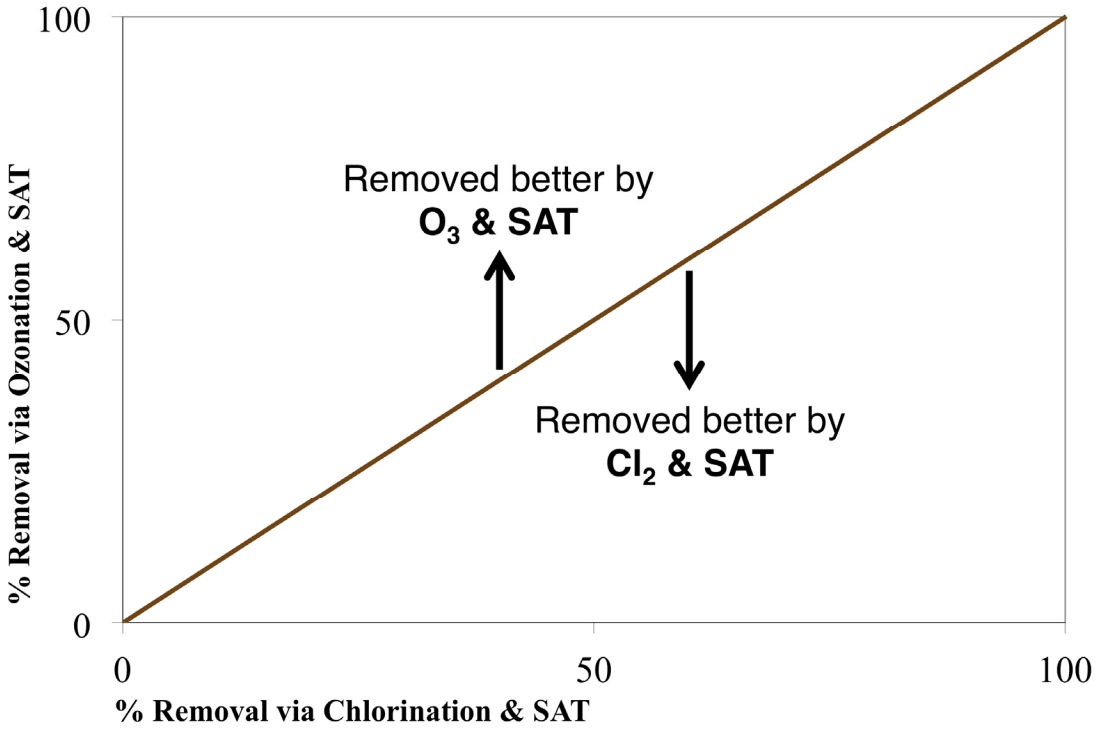

Figure 4.12. Template of a plot to compare two treatment alternatives.

Any CECs found to be ND in secondary wastewater (atorvastatin, caffeine, and diazepam) were omitted as their removals cannot be determined. In addition, compounds with complex degradation pathways (namely phenols) were omitted, and apparent increases were plotted as zero removal. The shorthand used in the comparison plot in Figure 4.13 is listed in Table 4.10.

Table 4.10. Shorthand for CECs Plotted in Figure 4.13

CEC Name	Shorthand
Acesulfame-K	ACE-K
Acetaminophen	APAP
Atenolol	ATEN
Azithromycin	AZT
Carbamazepine	CBZ
Carisoprodol	CPDL
DEET	DEET
Diclofenac	DCF
Dilantin (Phenytoin)	PHT
Erythromycin-H2O	ERY
Fipronil	FIP
Fluoxetine	FLX
Furosemide	FUR
Galaxolide	HHCB
Gemfibrozil	GFB
Ibuprofen	IBP
Iohexol	IOH
Iopromide	IPM
Meprobamate	MPB
Metoprolol	METO
Naproxen	NPX
Primidone	PRM
Propranolol	PRO
Sucralose	SUC
Sulfamethoxazole	SMX
TCEP	TCEP
TCPP	TCPP
TDCPP	TDCPP
Tonalide	AHTN
Triclocarban	TCC
Triclosan	TCS
Trimethoprim	TMP

Figure 4.13 utilizes the concept presented in Figure 4.12 to display the relative performances of ozonation in conjunction with SAT and chlorination in conjunction with SAT.

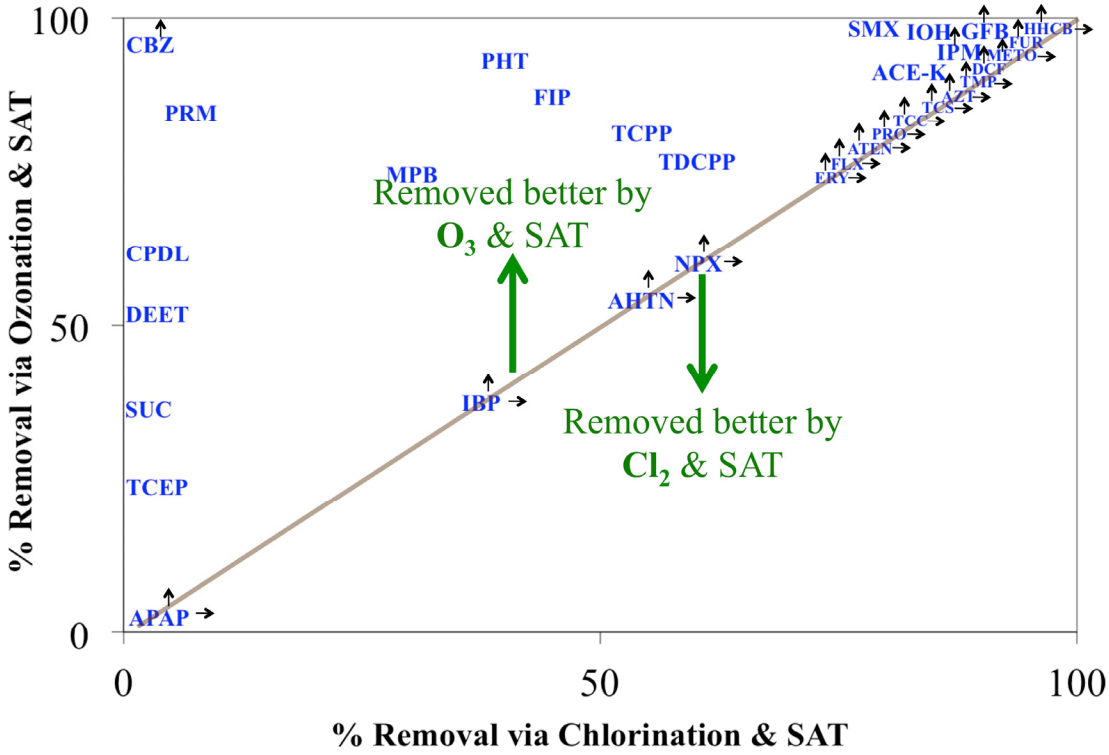

Figure 4.13. Graphical comparison of the two disinfection alternatives in conjunction with SAT.

Note: Small black arrows indicate that the removal was actually greater than the removal plotted; this is because ambient concentrations in the source water were used so that some influent concentrations were not present at sufficient levels to have detectable concentrations in the soil column effluent

Although a variety of CECs were removed by both systems, all compounds were removed equally or better by the O_3 Soil Column System, as depicted by the compounds located above the diagonal line. The absence of compounds below the diagonal line indicates that there is no CEC removed by the Cl_2 Soil Column System that is not also removed by the O_3 Soil Column System. Although it may appear that most of the CECs were equally removed by both ozone-SAT and chorine-SAT, all compounds on the line were removed to below detection level, so it is not possible to determine which system performed better. In this study, a decision was made to examine CECs in the SJCWWRF secondary effluent at their indigenous levels. As a result, several CECs were removed below their respective detection limits after treatment. Consequently, potential removals are greater than those the study was able to demonstrate. Several of the CECs removed by ozone are recalcitrant to SAT alone, indicating that ozone has the potential to supplement CEC removal by SAT. Ozonation reduced some CECs, namely carbamazepine, dilantin, fipronil, meprobamate, and primidone, which would otherwise be recalcitrant through the biological processes in SAT within the 30 day subsurface retention time investigated. Sucralose and TCEP are recalcitrant through both SAT systems and could serve as intrinsic tracers to track the recycled water. For the secondary wastewater tested, sucralose is especially interesting because it is consistently

present at high concentrations (around 30,000 ng/L), and it has a sufficiently low detection limit (40 ng/L).

Overall, CEC removal through SAT treatment is better characterized for the Cl_2 Soil Column System than for the O_3 Soil Column System. As discussed previously, this is more a function of the low levels of CECs being measured in the O_3 Soil Column System influent and of the efficacy of the ozonation disinfection than a measure of SAT. More conclusions could be drawn if the influent waters for each Soil Column System were spiked with the CECs of interest.

The data also shows that CEC removals improved as the soil columns aged and the biota in the columns further acclimated. Only three constituents were removed to a quantifiable degree (i.e., the soil column effluent concentrations were above the detection limit): iohexol, TCPP, and TDCPP. Their removal rates as the soil column matured are shown in Figure 4.14.

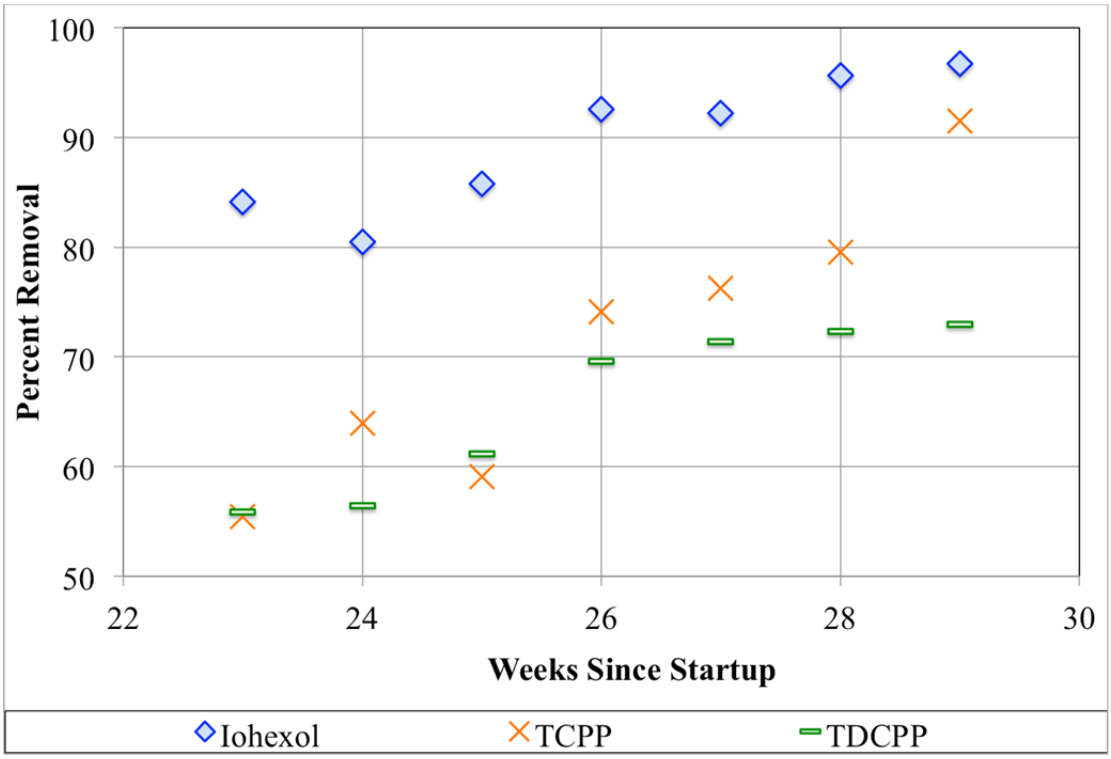

Figure 4.14. Improvements in removal rate with aging soil columns.

Laws et al. (2011) also found that travel time significantly impacted the removal of ibuprofen, sulfamethoxazole, TCEP, TCPP, trimethoprim, DEET, meprobamate, and triclosan. The practical implication of this finding is that the removal of some CECs may continue to increase over a startup period of the groundwater recharge operation as microbes acclimate to the wastewater. This finding supports work by Drewes et al. (2006).

Chapter 5

Phase 3: Microorganism Challenge

The Cl_2 Soil Column System was used for Phase 3, the microbe challenge phase. Both columns were operated independently with a 30 day HRT under saturated conditions. Cl_2 Column #1 remained downflow, and Cl_2 Column #2 remained upflow. Separate columns were used for coliform and MS-2 spiking studies to avoid interference between these organisms. Cl_2 Column #1 was used to test coliform removal, and Cl_2 Column #2 was used to test MS-2 virus and *Cryptosporidium* removal. Spiked concentrations for the three spiking events are summarized in Tables 5.1.

Table 5.1. Microbial Concentrations in Spike Used for Microbe Challenges

Round	Phase 3 Time Elapsed when Spike Introduced, days	Spike Solution Concentration		
		Cl_2 Column #1 Total Coliform, cfu/mL	Cl_2 Column #2 MS-2 Virus, pfu/mL	Cl_2 Column #2 *Cryptosporidium*, oocysts/mL
1	1	1.70E+07	3.50E+09	2.20E+05
2	22	4.50E+06	4.70E+09	5.50E+05
3	44	2.20E+07	4.20E+09	3.80E+05

Details of the procedure of the spiking are described in Section 2.4. In summary, after introducing the spiked pulse, normal operations were resumed (continuous feed of unspiked water, saturated conditions with a 30 day residence time). For *Cryptosporidium*, composite effluent samples were collected such that the duration determined during the tracer study was bracketed (i.e., for a 30-day HRT, collection of effluent needs to include collecting samples at least 24 to 36 days after spike addition). For the three microbe challenges, effluent composites were collected 21 to 36 days after Round 1 spiking, 24 to 36 days after Round 2 spiking, and 23 to 41 days after Round 3 spiking. For the entire duration of the study, MS-2 virus and total coliform were enumerated in 3- to 4-day composite samples, and samples were composited between periods when the spike was expected in the effluent.

Effluent samples from Cl_2 Column #1 were analyzed for total coliform. Effluent samples from Cl_2 Column #2 were analyzed for MS-2, and the remaining volume was concentrated and sent to BioVir for *Cryptosporidium* enumeration. Coliform, MS-2, and *Cryptosporidium* breakthroughs were not observed. As for detection limits of the methods used, total coliform was measured at less than 20 cfu/100 mL, MS-2 was measured at less than 30 pfu/1 mL, and *Cryptosporidium* was measured at less than 0.2 oocyst/1 L. Appendix G provides all TOC, UVT, and microbe data collected during this phase.

To address the potential of adsorption onto tubing, samples from Cl_2 Column #2 were also taken directly from the column (prior to entering the effluent tubing), in addition to composite samples. On days 50, 53, 59, 60, 64, 67, 71, 74, 78, 80, 85, and 88 samples were analyzed in this manner, and it was confirmed that MS-2 virus was not breaking through at detectable

levels. To confirm the levels added, the spiking solution was sampled in the field. Table 5.2 presents the average water quality of non-spiked feed, spiked feeds, and the effluents from each column.

Table 5.2. Average Water Quality of the Non-spiked Feed, Spiked Feeds, and Effluent from Each Column, as well as Values from Phase 2 for Comparison

Water Quality Parameter	Feed (Not Spiked)	Bacteria Spike Solution	Virus and *Cryptosporidium* Spike Solution	Bacteria Spiked Column Effluent	Virus and Crypto Spiked Column Effluent	Effluent Concentration during Phase 2
Ammonia, mg/L as N	0.67	1	5	<0.08	<0.08	<0.08
Conductivity, µS/cm	941	967	1204	1007	1006	902
Nitrate, mg/L as N	4.29	9.8	3.6	4.8	0.25	0.37
Orthophosphate, mg/L as PO4	5.77	18	32	1.6	0.55	0.44
pH	6.83	7.24	7.13	7.66	7.96	8
TOC, mg/L	6.45	15	128	4.49	3.24	3.3
Turbidity, NTU	0.53	12.8	1.3	0.79	0.51	0.51
UVT, %	76	33	48	82	85	86

On the basis of these findings, the absolute removal cannot be defined, but the removals were found to be greater than 7.8 log for coliform bacteria, 8.1 log for MS-2 virus, and 9.5 log for *Cryptosporidium* oocysts after a 30-day HRT in saturated flow. These results are difficult to compare against other published work because those studies did not report residence time. Exceptions are studies by Santamaria et al. (2011) and Quanrud et al. (2003) in which residence time is reported. Santamaria et al. (2011) used laboratory soil columns and spiking studies to evaluate the relationship between *Cryptosporidium* removal and travel time in sandy soil and found that log removals increased with travel time. Santamaria observed 5 log removal of *Cryptosporidium* in 58 h (2.4 days) of subsurface travel. Data from this study and Santamaria's study were widely separated in time. For MS-2, Quanrud et al. (2003) observed 2 log removal of MS-2 in 20 h (0.8 day) of subsurface travel. In light of the importance of pathogen removal in regulations for groundwater recharge with recycled water, these results deserve further investigation.

Because of the lack of any detection of MS-2 coliphage using plaque assays or of *Cryptosporidium* oocysts using the direct count method in soil column effluents, quantitative polymerase chain reaction (qPCR) was used to further assess the water samples from the

microbe challenges. qPCR is a more sensitive method than traditional quantification methods of infectious units or intact *Cryptosporidium* oocysts because of its ability to detect the presence of genetic debris independent of viability and intactness.

Of the effluent samples collected during the microbe challenge phase, only 13 samples were still available for qPCR, which was done several months later. Of these 13, six samples were collected from the standing water above the sand, three were collected during the short duration composites, and four were collected during the longer duration composites. In the period between the originally planned experiments and qPCR, samples were stored at 4 ºC.

In the case of MS-2, two of the 13 samples had positive MS-2 genome counts. There were traces of MS-2 genetic debris in one of the short duration composites (1.08E3 copies/mL) and in one of the long duration composites (3.50E5 copies/mL). In addition, MS-2 stock solution (the same as that used during Phase 3) was used to spike fresh dechlorinated recycled water (from the same feed water source as in Phase 3) in the same manner as the preparation of spike solution used in Phase 3 and enumerated using plaque assays and qPCR. This spiked sample had 6.59E+09 pfu/mL and 1.89E+11 genome copies/mL, indicating no decay in the stock solution and supporting the idea that MS-2 virus and its genetic debris were preserved in effluent samples during storage. Although it is difficult to make definitive conclusions, specifically because biological measurements are inherently highly variable, results do indicate low to no genome counts. On the basis of the presence of substantial genetic material still in the original stock used for Phase 3, the absence of MS-2 genome in some samples is attributed to attenuation through the soil columns. These results support Phase 3 findings and show that the soil columns were effective at removing not only viable MS-2 but also MS-2 genome.

In the case of *Cryptosporidium* oocysts, all samples, including samples spiked directly with *Cryptosporidium* oocysts, were absent of genetic material. This is likely due to the irradiation (150 mJ/cm) performed on the *Cryptosporidium* oocyst stock for safety considerations, which fragments the DNA beyond the point that qPCR is able to detect.

Although these data are not sufficient to estimate removal based on genome count, they do support the microbe challenge results. The presence of MS-2 genome is indicative of the presence of spike solution moving through the Soil Column System.

Chapter 6

Conclusions

Recycled water is an increasingly significant component of our nation's potable water supply, fueled by climate change, population growth, and the resulting reduction in conventional water supplies. As we increase potable water reuse, it is crucial to continue evaluating the impacts on public health and to consider the economical and practical implications of reclaimed water treatment. Soil Aquifer Treatment (SAT) is a long-established potable reuse strategy, and coupling SAT with ozonation provides an increased level of treatment for potable reuse. Building on these promising and proven technologies, this study compared the performance of two disinfection alternatives prior to SAT: ozonation and the more conventional chlorination.

Lab-scale soil columns were constructed to simulate the SAT process. Both the vadose zone (the region between the recharge basins and the water table) and the saturated zone (the region below the water table) were reproduced with a total travel time of 30 days. This study consisted of three phases. Phase 1 consisted of system setup, acclimation, and general optimization. This phase allowed biota in the soil columns to adapt and develop for the feed waters. Phase 2 included testing the attenuation of bulk organic carbon and CECs (i.e., pharmaceuticals and personal care products) as well as DBPs and other supporting water quality parameters. Phase 3 consisted of spiking the system with MS-2 virus, coliform bacteria, and *Cryptosporidium* protozoa to evaluate the efficacy of their removal through SAT.

The salient findings for this study include the following:

- Ozone addition ahead of SAT enhances the SAT process by reducing the TOC and increasing UVT to a greater degree than when chlorine disinfection is used ahead of SAT, within the 30-day travel time investigated. These parameters (i.e., TOC and UVT) indicate the presence of DBP precursors and are particularly important to maintain at low levels to prevent further DBP formation during treatment for potable reuse. In addition, NDMA is more effectively removed through photolysis under these conditions.

- Using the EEM wastewater fingerprint as a metric, ozonation alone eliminated virtually all organic matter of wastewater origin by transforming wastewater EfOM into NOM. The EEM image's characteristics approached a state very similar to that of natural water sources.

- Using the EEM wastewater fingerprint as a metric, the SAT process with the Soil Column System fed with chlorinated water reduced the characteristic peaks of wastewater organic matter as a function of travel time.

- All DBPs were completely attenuated through SAT, including NDMA, TTHMs, HAAs, and bromate.

- SAT was beneficial for CEC removal. Both Cl_2 and O_3 Soil Column Systems exhibited attenuation of the CECs present in the secondary effluent of the recycled water treatment plant.

- Of the 42 CECs tested, 38 CECs were present in the secondary wastewater. Chlorine disinfection did not further reduce any CECs completely. The Cl_2 Soil Column System completely removed acetaminophen, atenolol, azithromycin, erythromycin, fluoxetine, galaxolide, ibuprofen, metoprolol, naproxen, octylphenol diethoxylate, propranolol, tonalide, triclocarban, triclosan, and trimethoprim. Ozone disinfection completely reduced acetaminophen, atenolol, azithromycin, carbamazepine, diclofenac, erythromycin, fluoxetine, furosemide, gemfibrozil, ibuprofen, naproxen, octylphenol monoethoxylate, octylphenol diethoxylate, propranolol, tonalide, triclocarban, triclosan, and trimethoprim. The O_3 Soil Column System completely reduced three additional CECs (galaxolide, iopromide, and metoprolol). The efficacy of the O_3 Soil Column System is driven by the ozone disinfection step.

- The O_3 Soil Column System removed all CECs that the Cl_2 Soil Column System removed and others.

- Several of the CECs removed by ozone were recalcitrant to SAT alone, indicating that ozone has the potential to supplement CEC removal by SAT. Ozonation removed some CECs, namely carbamazepine, dilantin, fipronil, meprobamate, and primidone, which would otherwise have been recalcitrant through the biological processes in SAT within the 30-day subsurface retention time investigated.

- Under conditions of saturated flow and a 30 day HRT, spiked microbes were reduced through SAT to below detection levels. These results suggest removals greater than 8.1 log for MS-2 virus, 7.8 log for coliform bacteria, and 9.5 log for *Cryptosporidium* oocysts.

Overall, findings from this study indicate that SAT is an effective, natural treatment option for the attenuation of TOC, UVA, the wastewater fingerprint, DBPs, CECs, MS-2 virus, coliform, and *Cryptosporidium*. The use of ozonation as a disinfection alternative to chlorination prior to SAT proved to be all-around beneficial, with enhanced removal of all listed constituents. Ozonation is an affordable and brine-free disinfection alternative that enhances SAT performance with substantially less energy consumption than RO.

Future studies similar in nature that will help to further our understanding of SAT and SAT pretreatment might incorporate the following:

- longer travel times (on the order of months) for situations in which project boundaries are on the order of months; it is widely held that most of the removal in the SAT process takes place early within the process, but this work supports other recent studies that suggest that when additional time is available, important benefits can result

- further assessment of pathogen removal, because this study did not observe any spiked microbes in the effluent

- exploration of SAT removal mechanisms through the use of qPCR to assess the reduction in the genome count (all organisms), continued use of direct count to quantify physical

removal (protozoa), and measurement of the reduction in infectivity through the quantification of infectious units (all organisms)

- use of a conservative tracer in the spiked matrix, which would help confirm the time at which the spike exits the system

- CEC spiking to quantify the removal potential of CECs through SAT enhanced with ozonation prior to surface application

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Appendix A

Summary of Laboratory- and Field-Scale Studies

LABORATORY-SCALE

Effect of soil

In a study by Quanrud et al. (1996b), three different soil types were tested with columns operated under aerobic conditions with 7-day wetting and 7-day drying cycles and fed chlorinated/dechlorinated secondary effluent. Similar effluent DOC concentrations were found across three soil types tested, indicating that DOC removal was independent of soil type and infiltration rates (Quanrud et al., 1996b):

Table A.1.

Soil Type	Infiltration rate during 7 days wetting, m/d	Influent DOC, mg/L	Effluent DOC, mg/L	DOC removal, %
Aqua Fria sand	0.10 to 0.76	12.6	6.9	46
Sweetwater sandy loam	0.13 to 0.76	12.5	6.7	51
North Pond silt	0.07 to 0.39	12.6	6.3	48

A study by Westerhoff and Pinney (2000) showed some difference between two soils with varying fines and organic content: Calabasas Park soil (23% fines; f_{oc} = 0.0028 g-OC/g-soil) and Kino Springs soil (41% fines; f_{oc} = 0.0045 g-OC/g-soil). The system with less organic content performed more efficiently: feed DOC = 13.4 mg/L; Calabasas Park soil column effluent DOC = 3.7 mg/L; and Kino Springs soil effluent DOC = 5.8 mg/L. The lower removal though the Kino Springs soil is attributed to higher organic content of the soil and potential leaching of organics from the soil. The authors speculated that the removals would approach a similar level after leaching subsided.

Effect of travel time

Residence time within the aquifer system is a key parameter for the level of removal achieved for most organic contaminants because the salient removal mechanism is through biodegradation (Rauch-Williams et al., 2010; Mansell and Drewes, 2004; Drewes and Fox, 1999; Drewes and Jekel, 1998; NM = Not Measured):

Table A.2.

Compound (DOC in mg/L; all others in ng/L)	Feed	HRT, days			
		3–6	10–12	15–18	21–25
Tertiary Fed, Anoxic, Saturated Columns (Rauch-Williams et al., 2010)					
DOC	6.9	4.2	3.8	3.2	3
Carbamazepine	288	NM	NM	NM	318
Diclofenac	362	75	134	110	100
Gemfibrozil	444	324	<25	<25	<25
Ibuprofen	60	40	<10	<10	<10
Ketoprofen	707	581	<10	<10	<10
Naproxen	412	327	21	19	19
Primidone	568	629	NM	NM	764
Propyphenazone	642	575	NM	62	77
TCEP	323	365	NM	314	378
TCPP	805	983	NM	NM	1009
Tertiary Fed, Anoxic, Saturated Columns (Mansell and Drewes, 2004)					
DOC	8.6	6.1	NM	NM	4.1
17-Estradiol	285	1.1	NM	NM	<0.4
Estriol	161	<0.6	NM	NM	<0.6
Testosterone	218	<0.5	NM	NM	<0.5
Tertiary (Mesa and Riverside) and Secondary (Tucson and Avra) Fed, Anoxic, Saturated Columns (Drewes and Fox, 1999)					
DOC (Mesa, AZ)	6.1	5.5	4.7	4.1	3.9
DOC (Riverside, CA)	5.2	4.5	3.4	3.0	2.3
DOC (Tucson, AZ)	11.4	5.3	3.5	3.0	2.8
DOC (Avra Valley, AZ)	4.6	NM	3.2	NM	2.8
Tertiary Fed, Anoxic, Saturated Columns (Drewes and Jekel, 1998)					
DOC	15.1	11.4	NM	NM	10.7
Secondary Fed, Anoxic, Saturated Columns (Fox et al., 2006)					
DOC	8.6	6.1	NM	NM	4.1
17β-Estradiol	285	1.1	NM	NM	<0.4
Estriol	161	<0.6	NM	NM	<0.6
Testosterone	218	<0.5	NM	NM	<0.5

CEC Removal Efficacies

Several CECs have been shown to be effectively attenuated (e.g., naproxen, gemfibrozil, and ibuprofen) through soil columns independent of oxic and anoxic operations, but some remain recalcitrant (e.g., TCPP and TCEP).

With as little as 6 days of travel time, soil columns operated under different redox conditions and nitrogen feed qualities showed excellent removal (>90%) of the following CECs: (1) oxic conditions fed with nitrified, tertiary effluent—naproxen and gemfibrozil; (2) anoxic conditions fed with nitrified, tertiary effluent—naproxen, caffeine, and ibuprofen (gemfibrozil was removed 50 to 90%); and (3) anoxic conditions with denitrified, tertiary effluent—naproxen, gemfibrozil, and ibuprofen (diclofenac was removed 50–90%). TCPP, TDCPP, and TCEP were poorly removed (less than 50% reduction) under all conditions tested (Drewes et al., 2011).

With an HRT of 11 days, Cordy et al. (2004) found that of the 27 organic compounds present in their secondary wastewater, 18 compounds were removed to below the detection limit through the soil column, and nine compounds persisted in the soil column effluent: sulfamethoxazole, carbamazepine, benzophenone, 2,6-dimethylnapthalene, 5-methyl-1*H*-benzotriazole, *N,N*-diethyltolumide, tributylphosphate, tri(2-chloroethyl) phosphate, and cholesterol.

Rauch-Williams et al. (2010) noted effective removal of gemfibrozil, ibuprofen, ketoprofen, naproxen, and propyphenazone but limited attenuation of TCPP, TCEP, primidone, and carbamazepine with a residence time of 25 days. Mansell and Drewes (2004) noted excellent removal of steroids, namely 17-estradiol, estriol, and testosterone, with a residence time of 23 days.

Westerhoff and Pinney (2010) found that TTHMFP was reduced through their soil columns. Feed TTHMFP was measured at 508 µg/L, and the matured soil columns showed 210 µg/L in the effluent of the column filled with Calabasas Park soil (23% fines; f_{oc} = 0.0028 g-OC/g-soil) and 344 µg/L in the effluent of the column filled with Kino Springs soil (41% fines; f_{oc} = 0.0045 g-OC/g-soil). Lower removal though the Kino Springs soil is attributed to the higher organic content of the soil and the potential leaching of organics.

The attenuation of NDMA has been studied (Nalinakumari et al., 2010; Drewes et al., 2006b). Nalinakumari et al. (2010) operated soil columns with a tertiary feed and 20 day HRT and found that NDMA was more readily removed under oxic conditions (feed: 888 ppt; column effluent: 40 ppt), as opposed to anoxic conditions (feed: 934 ppt; column effluent: 248 ppt). Drewes et al. (2006b) also found nitrosamines to be removed better under oxic conditions as compared with anoxic conditions but found soil acclimation to be an important component of system efficacy:

Table A.3.

Acclimation Period	Effluent (Feed), ng/L Anoxic, 6 day HRT	Effluent (Feed), ng/L Anoxic, 25 day HRT	Effluent (Feed), ng/L Oxic, 6 day HRT
4 months	120 (140)	97 (140)	<2 (140)
>8 year	3.5 (120)	<2 (120)	n/a

Field-Scale

Drewes et al. (2011) found that indicator chemicals that are primarily removed through biodegradation were removed to a similar degree at the five different field sites studied for a given travel time. DEET, diclofenac, ibuprofen, and meprobamate were found to have slow kinetics, requiring greater than 1 week to achieve 90% removal. Primidone and carbamazepine were found to be recalcitrant. In addition, the authors found that TOC, TOX, and UVA correlated with the removal of organic indicators.

In addition, Drewes et al. (2006) provided data to support that SAT is effective at transforming wastewater-derived DOC into organic matter having characteristics of natural water by using tools such as EEM, size exclusion chromatography, 13C-NMR, FTIR, and elemental analysis. The portion of the DOC remaining after SAT resembled a combination of wastewater-derived organic matter and NOM.

Mesa, AZ

Drewes et al. (2001) monitored the recharge basins used by Northwest WRP in Mesa, AZ. This facility is composed of four basins that are flooded with tertiary treated wastewater one at a time. The flooding-drying cycle is based on the individual basin characteristics, namely infiltration rates and clogging properties. Each basin typically experiences one complete cycle per month. The vadose zone within this area is shallow (less than 1.6 m), and clay lenses result in slow infiltration rates (ranging from 6–12 cm/d). The feed recycled water DOC averaged 5.2 mg/L, whereas infiltrated DOC was reduced substantially to 1.31 and 0.95 mg/L with travel times of 6 to 12 months and 12 to 18 months, respectively. The authors also demonstrated that the DOC characteristics (e.g., distribution of structural groups and molecular weights) of soil aquifer treated water approach those of the local groundwater that is not under the influence of the recycled water recharge operation. Additional findings include substantial reductions of commonly found CECs (removals: EDTA = 81%, NTA = >98%, and APEC = 99.5%).

Amy and Drewes (2007) showed that TCEP was well removed with travel times greater than 6 months, whereas TCIPP persisted even after 2 years.

Table A.4.

Travel time	DOC, mg/L	TCEP, ng/L	TCIPP
Tertiary recharge water	6.1	310–420	1085–2625
6–18 months	1.5	<10	140
6–19 months	1.8	NM	NM
6–20 months	1.5	<10	470

Mansell and Drewes (2004) reported complete removal of 17β-estradiol, estriol, and testosterone after 12 months of travel time. Significant removal of these steroids was also observed with travel times on the order of days:

Table A.5.

Constituent	Feed	Travel Time 5 days	Travel Time 12 months
DOC, mg/L	6.7	5.2	1.67
17β-Estradiol, ng/L	4.2	0.5	<0.4
Estriol, ng/L	4.9	<0.6	<0.6
Testosterone, ng/L	3.0	<0.5	<0.5

Other DOC removal monitoring studies include the following: (1) Fox et al. (2006) found that feed DOC averaged 5.2 mg/L whereas infiltrated DOC was reduced to 1.43 and 1.23–1.56 with travel times of 6 to 12 months and 12 to18 months, respectively; (2) Drewes et al. (2003) found that feed DOC was reduced from 5.6 mg/L to 1.5 mg/L after 6 to 12 months of travel in the subsurface; and (3) Drewes et al. (2006) found that 6 to 12 months of travel time translated to a DOC reduction from 5.3 mg/L to 1.7 mg/L.

Tucson, AZ

Fox et al. (2006) did not identify any relationship between vadose zone depth and treatment efficacy. Within the vadose zone, however, the redox conditions impact the rate of biodegradation of target contaminants. The redox conditions can be optimized through cyclic recharge operations. Overall, travel time is the condition that most significantly impacts the removal of organics via SAT. Soil properties have the potential to impact the infiltration rate, biofilm development, refreshment of oxygen within soil pores, and adsorption capacity. Treatment efficacy proved to be independent of these properties, making SAT a robust process. A variety of wetting and drying cycles are being used at the Tucson recharge site (typically operated with a 2-day wetting and 4-day drying cycle). It was shown that increasing the length of the drying time allowed oxygen to penetrate to greater depths.

Amy and Drewes (2007) showed that TCEP and TCIPP were persistent after a month in the subsurface:

Table A.6.

Travel Time	DOC, mg/L	TCEP, ng/L	TCIPP, ng/L
Secondary recharge water	14.1	295	770
11 days	4.84	NM	NM
35 days	0.98	175	365

Mansell and Drewes (2004) reported complete removal of 17β-estradiol, estriol, and testosterone after a 2-week travel time. Significant removal of these steroids was also observed with travel times of a single day:

Table A.7.

	Feed	Travel Time: 1 day	Travel Time: 2 weeks
DOC, mg/L	13.9	4.2	0.85
17β-Estradiol, ng/L	7.2	1.8	<0.4
Estriol, ng/L	21.3	<0.6	<0.6
Testosterone, ng/L	11.5	<0.5	<0.5

Drewes et al. (2003) found that DOC was reduced from 15 to 2 mg/L after 2 to 4 weeks of travel in the subsurface. Also, complete removal of caffeine, gemfibrozil, diclofenac, ibuprofen, ketoprofen, and fenoprofen was observed after 2 to 4 weeks of travel. Carbamazepine, primidone, and organic iodide were found to be persistent.

Whittier, CA

A study using the USGS Research Basin at the Montebello Forebay Spreading Grounds found that the majority of the decrease occurred within the first 10 h; however, further measureable removal was achieved within 60 days. Iopromide, gemfibrozil, fluoxetine, and atenolol were well removed within residence times of less than 3 days. Travel time significantly impacted the removal of ibuprofen, sulfamethoxazole, TCEP, TCPP, trimethoprim, DEET, meprobamate, and triclosan. Primidone, carbamazepine, and phenytoin remained recalcitrant (Laws et al., 2011). DOC removal and fluorescence (i.e., wastewater fingerprint) were also monitored. The authors found substantial reductions with travel time in DOC and in protein-, humic- and fulvic-like organics:

Table A.8.

Travel Time	DOC, mg/L	Quantitative Fluorescence		
		Protein	Humic	Fulvic
Basin—Tertiary WW	7.43	1.47	0.64	1.01
10 h	3.23	0.67	0.45	0.61
12 h	4.06	0.76	0.50	0.72
26 h	3.63	0.72	0.47	0.67
42 h	3.46	0.78	0.47	0.67
51 h	3.59	0.83	0.49	0.68
70 h	3.49	0.76	0.46	0.64
60 days	1.61	0.33	0.29	0.48
60 days	1.76	0.45	0.32	0.53

Note: WW = wastewater

Appendix B

Data Summary for Travel Times Up to 2 Weeks

Table B.1.

Recharge Location	Montebello Forebay Spreading Grounds (USGS/WRD Test Basin & San Gabriel Coastal Basin)	Inland Empire Utilities Agency (8th St. Basin)	Inland Empire Utilities Agency (Brooks Basin)	Inland Empire Utilities Agency (Hickory Basin)	Sweetwater Recharge Facility (Research Basin-1)	Soil Column Study	Soil Column Study	Soil Column Study	Regional Rapid Infiltration Basin	Sweetwater Recharge Facility (Research Basin-1)
Travel Time, Days	**0.5 to 3**	**2 to 3**	**2 to 3**	**2 to 3**	**2 to 3**	**6**	**6**	**6**	**7**	**14**
WRRF 05-04 Table Number (Drewes et al., 2011)	**3.1**	**3.2**	**3.2**	**3.2**	**3.3**	**3.5**	**3.6**	**3.6**	**3.4**	**3.3**
Indicator Compounds	**Percent Removal Through Soil Aquifer Treatment**									
Atenolol	>90	>90	>90	>90	>90				>90	>99
BHA					>90				>90	>90
Caffeine		>90	>90		>90			>90		>90
Diclofenac	90 to 50				90 to 50		90 to 50		>90	>99
Fluoxetine	>90	>90	>90		>99				>90	>99
Gemfibrozil	>90				90 to 50	>90	>90	90 to 50	>99	>99
Ibuprofen	50 to 25	50 to 25	50 to 25		90 to 50		>90	>90		>90
Naproxen	90 to 50				90 to 50	>90	>90	>90	>90	>99
Triclosan		90 to 50			>90				>90	>90
Trimethoprim					>90				>90	>99
Atorvastatin					>99					>99
Benzophenone		50 to 25	50 to 25	50 to 25	90 to 50					>90
DEET	<25	<25	<25	<25	50 to 25					>90
Dilantin (Phenytoin)	50 to 25	<25	<25	<25	50 to 25				90 to 50	90 to 50
Dioctyl phthalate					>90					>90
EDTA					<25					>90
Enalapril					>90					>90
Galaxolide					>99					>99
Iopromide	>90		90 to 50		50 to 25					>90
Meprobamate	<25	<25	<25		<25				>99	>90
Nonylphenol					>90					>99
Norfluoxetine										>90
Octylphenol					90 to 50					>90
Salicylic Acid					>90					>90
Simvastatin hydroxy acid					>90					>90
Sulfamethoxazole	<25	<25	<25		<25					>90
TCEP	<25	50 to 25	<25	50 to 25	50 to 25	<25	<25	<25	90 to 50	90 to 50
TCPP	<25	50 to 25	<25		<25	50 to 25	<25	<25	90 to 50	90 to 50
Tonalide					90-50					>90
Carbamazepine	<25	<25	<25	<25	<25				<25	<25
Primidone	<25	<25	<25	<25	50 to 25				50 to 25	<25
TDCPP		50 to 25			50 to 25	50 to 25	<25	<25		50 to 25

Source: Drewes et al. (2011)

Appendix C

Details of Soil Column Configurations from Past Studies

Table C.1

Reference(s)	Feed water (DOC (mg/L))	Column inner diameter (cm)	Total column height (m)	Soil type (d_{50})	Loading percolation rate (m/day)	Acclimation strategy
Drewes et al., 2011	Various tertiary WWs (not reported)	15.2	Anoxic/saturated: 4.9 Oxic/unsaturated: 1.2	Native alluvial groundwater recharge site material with >2 mm sieved out	Anoxic/saturated and oxic/unsaturated (0.08)	Existing, well-established setup at CSM
Linlin et al., 2011	Unsaturated: Tertiary 0.45 µm filtered 0.6 to 1 mg of O_3/mg of TOC ozonated tertiary effluent WW (6.4) Saturated: Effluent from unsaturated (2.6)	25	1.7	0.4–0.8 mm	Unsaturated: 3 days on/1 day off, downflow (2.9) Saturated: Continuous, upflow (0.04)	Fed continuously for 6 months
Nalinakumari et al., 2010	Tertiary WW (6.2)	12	4.5	Soil from a recharge site	Anoxic for 567 days, then aerobic for 201 days (0.07)	Adapted using tertiary WW
Rauch-Williams et al., 2010	Secondary WW spiked with TOrC (6.9)	15	4	Aquifer material (0.8 mm)	Anoxic, saturated, upflow, continuous (0.065)	Fed continuously for over 6 years with secondary or tertiary WW and for 5 months with the secondary WW used for this study

Reference(s)	Feed water (DOC (mg/L))	Column inner diameter (cm)	Total column height (m)	Soil type (d_{50})	Loading percolation rate (m/day)	Acclimation strategy
Debroux and Drewes, 2007	Dechlorinated tertiary WW (14.2)	Saturated: 10.2 and 15.2 Unsaturated: 30.5	Saturated: 1.4 to 1.7 Unsaturated: Not reported	Russian River with > 51 mm sieved out	Saturated: Upflow, continuous (1.4 to 2.7) Unsaturated: Downflow batch mode (not reported)	Fed continuously for 30 days prior to use
Fox et al., 2006; Mansell and Drewes, 2004	Secondary WW spiked with hormones (8.6)	14	4	Not reported	Anoxic, saturated, upflow, continuous (0.044)	Not reported
Gungor and Unlu, 2005	Secondary (COD: 40–45)	13.5	0.88	Sandy loam (not reported) Loamy sand (not reported) Sandy loam (not reported)	Downflow, saturated, 2.5 cm head, two cycles: 7 days on/7 days off and 3 days on/4 days off (0.045–0.33)	Not reported
Cordy et al., 2004	Secondary (TOC: 8.9)	32.5	2.4	Loamy sand > 2 mm (not reported)	Downflow, saturated for 23 days (2 pore volumes; 10 cm head), drained 2 days (0.053)	174 L secondary for 2 weeks; then drained for 60 days
Mansell et al., 2004	Secondary WW spiked with hormones (8.0)	8	0.3	Silica sand (d_{10}=1 mm)	Anoxic, saturated, upflow, continuous (0.115)	Not reported
Rauch and Drewes, 2004	Secondary WW spiked with TOrC (8.7)	14	4	Aquifer material (0.8 mm)	Anoxic, saturated, upflow, continuous (not reported)	Fed continuously with secondary or tertiary WW since 1997
Quanrud et al., 2003	Dechlorinated secondary WW (13)	7.6	1	Sweetwater sandy loam <3 mm Aqua Fria sand <3 mm	30 cm head, 7 days on/7 days off (3–4)	9 cycles

Reference(s)	Feed water (DOC (mg/L))	Column inner diameter (cm)	Total column height (m)	Soil type (d_{50})	Loading percolation rate (m/day)	Acclimation strategy
Drewes et al., 2001	Tertiary WW (5.2)	14	4	Aqua Fria River aquifer material (not reported)	Aerobic, saturated, upflow, continuous (0.190)	Adapted using secondary WW
Drewes et al., 2001b	Secondary WW (15.8) Ozonated secondary WW; 0.9 mg of O_3/mg of TOC (14.8) Ozonated secondary WW; 1.9 mg of O_3/mg of TOC (13.0)	14	2	Aquifer material with grain size 1–2 mm	Aerobic and anoxic, continuous	Not reported
Drewes and Fox, 2000	Various tertiary WWs (3.3–25)	14	4	Aqua Fria River aquifer material (not reported)	Anoxic, saturated, upflow, continuous (30 day residence time)	Fed continuously over 2 months with secondary WW
Westerhoff and Pinney, 2000	Aerated lagoon WW (13)	7.6	0.91	From potential recharge sites; sandy loam collected below root zone; 4.75 mm screened out	Downflow, saturated, 1 cm head, 7 days on/7 days off (0.11)	Three stages: Ripening 0–10 weeks; acclimation 10–35 weeks; maturation 35–64 weeks
Drewes and Fox, 1999	Tertiary denitrified WW (6) Secondary WW (11.5) Oxidation ditch denitrified WW (4.5) Tertiary partially denitrified WW (5.25)	14	4	Aqua Fria River aquifer material (not reported)	Anoxic, saturated, upflow, continuous (0.2)	Fed continuously over 2 months with tertiary WW

Reference(s)	Feed water (DOC (mg/L))	Column inner diameter (cm)	Total column height (m)	Soil type (d_{50})	Loading percolation rate (m/day)	Acclimation strategy
Drewes and Jekel, 1998	Tertiary WW (17)	14	2	Aquifer material with grain size 1–2 mm	Saturated (0.1–0.9) Unsaturated (0.6–1.3)	Not reported
Quanrud et al., 1996	Chlorinated secondary WW (9.8) Tertiary WW (8.1)	8.6	0.97	Sweetwater loamy soil using <2 mm	Unsaturated, 7 days on/7 days off, no ponding occurred (0.9)	15 cycles
Quanrud et al., 1996b	Chlorinated secondary WW (12.4) Dechlorinated secondary WW (12.4)	8.6	1	Sweetwater poorly graded silty sand Aqua Fria poorly graded sand North Ponding silty sand	7 days on/7 days off, ponding (0.1–3)	5–7 cycles

Appendix D

Full Suite DBP Concentrations

Table D.1.

			HAAs						Nitrosamines							THMs				
Location	Event	Bromate	HAA5	DBAA	DCAA	MBAA	MCAA	TCAA	NDPA	NDBA	NDEA	NDMA	NMEA	NPIP	NPYR	TTHMs	TBM	TCM	BDCM	DBCM
After Secondary	1								<0.0020	<0.0020	<0.0020	0.095	<0.0020	<0.0020	<0.0020					
After Cl2 disinfection	1								<0.0020	<0.0020	<0.0020	1.1	<0.0020	0.0038	<0.0020					
	3		30	<0.99	11	<0.99	<2.0	19								34.7	<0.5	22.6	10.3	1.8
	4		36	<0.99	15	<0.99	3	18	<0.0020	<0.0020	<0.0020	0.41	<0.0020	<0.0020	<0.0020	24.3	<0.5	15.5	7.4	1.4
	5		15	<0.99	7.8	<0.99	<2.0	7.2	<0.0020	<0.0020	<0.0020	0.34	<0.0020	0.0073	<0.0020	15.1	<0.5	10.6	3.8	0.71
	6								<0.0020	<0.0020	0.0082	0.19	<0.0020	<0.0020	<0.0020					
	7								<0.0020	<0.0020	<0.0020	0.2	<0.0020	<0.0020	<0.0020					
After Cl2 vadose	1								<0.0020	<0.0020	<0.0020	0.15	<0.0020	0.0023	<0.0020					
	4								<0.0020	0.026	<0.0020	0.032	<0.0020	0.003	<0.0020					
	5								<0.0020	<0.0020	<0.0020	0.019	<0.0020	0.0025	<0.0020					
	6								<0.0020	<0.0020	0.0028	<0.0020	<0.0020	<0.0020	<0.0020					
	7								<0.0020	<0.0020	0.0036	<0.0020	<0.0020	<0.0020	<0.0020					
After Cl2 saturated	0								<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020					
	1								<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	0.0039					
	2								<0.0020	<0.0020	0.0025	0.0022	<0.0020	<0.0020	<0.0020					
	3		<1.0	<1.0	<1.0	<1.0	<2.0	<1.0	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.5	<0.5	<0.5	<0.5	<0.5
	4		<1.0	<0.99	<0.99	<0.99	<2.0	<0.99	<0.0020	<0.0020	0.0022	<0.0020	<0.0020	<0.0020	<0.0020	<0.5	<0.5	<0.5	<0.5	<0.5
	5		<1.0	<1.0	<1.0	<1.0	<2.0	<1.0								<0.5	<0.5	<0.5	<0.5	<0.5
After O3 disinfection	1								<0.0020	<0.0020	<0.0020	0.64	<0.0020	<0.0020	<0.0020					
	3	5.5																		
	4	<0.50							<0.0020	<0.0020	0.002	0.29	<0.0020	<0.0020	<0.0020					
	5	1.4							<0.0020	<0.0020	<0.0020	0.31	<0.0020	0.0098	<0.0020					
	6								<0.0020	<0.0020	<0.0020	0.3	<0.0020	0.01	<0.0020					
	7								<0.0020	<0.0020	<0.0020	0.38	<0.0020	0.011	<0.0020					
After O3 vadose	1								<0.0020	<0.0020	<0.0020	0.0095	<0.0020	<0.0020	<0.0020					
	4								<0.0020	0.026	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020					
	5								<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020					
	6								<0.0020	<0.0020	0.0027	<0.0020	<0.0020	<0.0020	<0.0020					
	7								<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020					
After O3 saturated	0								<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	0.002					
	1								<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020					
	2								<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020					
	3	<0.50							<0.0020	0.007	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020					
	4	<0.50							<0.0020	<0.0020	0.0096	<0.0020	<0.0020	<0.0020	<0.0020					
	5	<0.50																		

Note: All concentrations are in μg/L

Appendix E

Raw CEC Data and General Statistics

This appendix provides CEC concentrations at all locations assessed for this study. All concentrations are reported in ng/l.

Table E.1. Concentrations after Secondary

		Secondary						
Compound Name	**Results**	**Mean**	**Standard Deviation**	**Sample Size**	**Min**	**Max**	**Median**	**95th Percentile**
Acesulfame-K	148.5, 52.6, 101.1, 4227.4, 1229.8, 1093.2	1142	1599	6	53	4227	621	3478
Acetaminophen	<10, <10, 11, 10, <10, <10	10	0	6	10	11	10	11
Atenolol	210, 49, 49, 291, 130, 108	140	95	6	49	291	119	271
Atorvastatin	<10, <10, <10, <10, <10, <10	10	0	6	10	10	10	10
Azithromycin	139, 132, 108, 205, 177, 133	149	35	6	108	205	136	198
Bisphenol A	<10, <10, <10, <10, <10, <10	10	0	6	10	10	10	10
Caffeine	<10, <10, <10, <10, <10, <10	10	0	6	10	10	10	10
Carbamazepine	240, 227, 209, 205, 265, 235	230	22	6	205	265	231	259
Carisoprodol	409, 159, 169, 98, 227, 130	199	112	6	98	409	164	364
DEET	12, 28, 136, 35, 48, 52	52	44	6	12	136	42	115
Diazepam	<10, <10, <10, <10, <10, <10	10	0	6	10	10	10	10
Diclofenac	206, 406, 250, 61, 242, 177	224	112	6	61	406	224	367
Dilantin (Phenytoin)	278, 269, 277, 241, 216, 273	259	25	6	216	278	271	278
Erythromycin-H2O	55, 25, 34, 62, 44, 46	44	13	6	25	62	45	60
Fipronil	36, 52, 52, 47, 64, 54	51	9	6	36	64	52	62
Fluoxetine	49, 42, 44, 50, 42, 44	45	3	6	42	50	44	50
Furosemide	481, 447, 476, 117, 636, 390	425	171	6	117	636	462	597
Galaxolide	3000, 3200, 2900, 2900, 2800, 2100	2817	376	6	2100	3200	2900	3150
Gemfibrozil	189, 47, 42, 86, 331, 206	150	113	6	42	331	138	300
Ibuprofen	<10, <10, 14, <10, 30, 22	16	8	6	10	30	12	28
Iohexol	11700, 5480, 6600, 11800, 5230, 11900	8785	3335	6	5230	11,900	9150	11,875

Compound Name	Results	Mean	Standard Deviation	Sample Size	Minimum	Maximum	Median	95th Percentile
Meprobamate	444, 315, 313, 370, 353, 369	361	48	6	313	444	361	426
Metoprolol	636, 420, 455, 576, 461, 458	501	85	6	420	636	460	621
Naproxen	32, <10, <10, 13, 38, 48	25	16	6	10	48	23	46
Nonylphenol diethoxylate	166, 184, 256, 229, 271, 276	230	46	6	166	276	243	275
Nonylphenol monoethoxylate	340, 352, 578, 438, 404, 439	425	86	6	340	578	421	543
Octylphenol diethoxylate	37.3, <25.0, 28.7, 28.5, 27.6, 27	29	4	6	25	37	28	35
Octylphenol monoethoxylate	238, 106, 140, 141, 113, 98.7	139	51	6	99	238	127	214
Primidone	263, 190, 193, 224, 199, 198	211	28	6	190	263	199	253
Propranolol	76, 49, 46, 79, 69, 56	63	14	6	46	79	63	78
Sucralose	26400, 28300, 27400, 13800, 27700, 27600	25,200	5619	6	13,800	28,300	27,500	28,150
Sulfamethoxazole	2460, 1600, 1250, 2780, 1890, 1580	1927	583	6	1250	2780	1745	2700
TCEP	239, 421, 423, 239, 428, 288	340	94	6	239	428	355	427
TCPP	2390, 4060, 5620, 2250, 2080, 1490	2982	1552	6	1490	5620	2320	5230
TDCPP	488, 643, 622, 507, 570, 441	545	80	6	441	643	539	638
Tonalide	96, 100, 130, 120, 100, 110	109	13	6	96	130	105	128
Triclocarban	71, 83, 90, 32, 88, 90	76	23	6	32	90	86	90
Triclosan	110, 127, 117, <10, 134, 122	103	46	6	10	134	120	132
Trimethoprim	201, 72, 82, 315, 210, 154	172	91	6	72	315	178	289
4-Nonylphenol (tech mix)	199, 154, 240, 156, 292, 278	220	60	6	154	292	220	289
4-*tert* Octylphenol	47.6, 13.1, 16.6, 22.9, 23.1, 17.1	23	12	6	13	48	20	41

Table E.2. Concentrations after Chlorination

	Chlorinated							
Compound Name	**Results**	**Mean**	**Standard Deviation**	**Sample Size**	**Min**	**Max**	**Median**	**95th Percentile**
Acesulfame-K	104.7, 72.9, 139.5, 432.9, 1235.5, 525.9, 364.8	411	404	7	73	1236	365	1023
Acetaminophen	<10, 13, <10, 23, 12, 26, 29	18	8	7	10	29	13	28
Atenolol	40, 54, 137, 221, 58, 82, 61	93	65	7	40	221	61	196
Atorvastatin	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Azithromycin	69, 78, 139, 215, 82, 172, 131	127	54	7	69	215	131	202
Bisphenol A	<10, <10, <10, <10, <10, 123, 20	28	42	7	10	123	10	92
Caffeine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Carbamazepine	203, 202, 218, 212, 99, 279, 237	207	55	7	99	279	212	266
Carisoprodol	648, 401, 154, 214, 53, 249, 162	269	198	7	53	648	214	574
DEET	13, 112, 88, 1050, 19, 65, 41	198	377	7	13	1050	65	769
Diazepam	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Diclofenac	<10, 53, <10, 51, 220, 186, 72	86	84	7	10	220	53	210
Dilantin (Phenytoin)	136, 150, 187, 182, 86, 164, 191	157	37	7	86	191	164	190
Erythromycin-H2O	22, 20, 18, 50, 19, 43, 42	31	14	7	18	50	22	48
Fipronil	32, 36, 11, 42, 43, 55, 40	37	14	7	11	55	40	51
Fluoxetine	34, 33, 39, 42, 18, 37, 36	34	8	7	18	42	36	41
Furosemide	44, 74, 16, 121, 592, 317, 66	176	209	7	16	592	74	510
Galaxolide	2500, 2400, 2500, 2400, 2500, 2600, 1900	2400	231	7	1900	2600	2500	2570
Gemfibrozil	25, 57, 180, 377, 376, 246, 83	192	147	7	25	377	180	377
Ibuprofen	<10, <10, <10, 16, 10, 10, 13	11	2	7	10	16	10	15

Compound Name	Results	Mean	Standard Deviation	Sample Size	Min	Max	Median	95th Percentile
Iohexol	10400, 9500, 6450, 7140, 6680, 4380, 11100	7950	2436	7	4380	11,100	7140	10,890
Iopromide	380, <10, 20, 219, 249, <10, 966	265	341	7	10	966	219	790
Meprobamate	362, 317, 340, 368, 168, 359, 380	328	73	7	168	380	359	376
Metoprolol	351, 342, 464, 543, 216, 401, 376	385	103	7	216	543	376	519
Naproxen	<10, <10, <10, 15, 87, 14, <10	22	29	7	10	87	10	65
Nonylphenol diethoxylate	119, 184, 230, 359, 212, 221, 229	222	72	7	119	359	221	320
Nonylphenol monoethoxylate	291, 306, 315, 500, 411, 358, 390	367	74	7	291	500	358	473
Octylphenol diethoxylate	33.7, <25.0, <25.0, 44.5, 26, 25.5, 26.6	29	7	7	25	45	26	41
Octylphenol monoethoxylate	192, 80.9, 97.8, 141, 122, 85.2, 87	115	40	7	81	192	98	177
Primidone	211, 187, 206, 193, 101, 193, 195	184	37	7	101	211	193	210
Propranolol	41, 34, 32, 44, 23, 50, 44	38	9	7	23	50	41	48
Sucralose	28900, 29000, 31400, 32500, 29600, 30100, 30000	30,214	1308	7	28,900	32,500	30,000	32,170
Sulfamethoxazole	284, 256, 92, 319, 487, 871, 278	370	249	7	92	871	284	756
TCEP	336, 398, 432, 444, 144, 494, 306	365	117	7	144	494	398	479
TCPP	3410, 3810, 4170, 10100, 1230, 2400, 1640	3823	2978	7	1230	10,100	3410	8321
TDCPP	537, 579, 647, 647, 273, 589, 469	534	131	7	273	647	579	647
Tonalide	91, 89, 82, 130, 110, 100, 110	102	16	7	82	130	100	124
Triclocarban	52, 54, 60, 70, 82, 77, 82	68	13	7	52	82	70	82
Triclosan	<10, <10, <10, <10, 91, <10, <10	22	31	7	10	91	10	67
Trimethoprim	21, 24, 49, 95, 43, 116, 50	57	36	7	21	116	49	110
4-Nonylphenol (tech mix)	71.6, 60, 50, 131, 144, 325, 218	143	100	7	50	325	131	293
4-*tert* Octylphenol	42.4, 7.6, 10.3, 20.6, 24.2, 20.3, 15.9	20	11	7	8	42	20	37

Table E.3. Concentrations after Ozonation

	Ozonated							
Compound Name	**Results**	**Mean**	**Standard Deviation**	**Sample Size**	**Minimum**	**Maximum**	**Median**	**95th Percentile**
Acesulfame-K	146.7, <50.0, <50.0, <50.0, 1593.5, 356.7, 154.4	343	562	7	50	1594	147	1222
Acetaminophen	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Atenolol	<30, <30, <30, <30, <30, <30, <30	30	0	7	30	30	30	30
Atorvastatin	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Azithromycin	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Bisphenol A	153, 309, 1130, 399, 206, 366, 142	386	343	7	142	1130	309	911
Caffeine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Carbamazepine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Carisoprodol	163, 94, 63, 86, 49, 100, 48	86	40	7	48	163	86	144
DEET	<10, <10, 14, 54, 12, 14, 10	18	16	7	10	54	12	42
Diazepam	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Diclofenac	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Dilantin (Phenytoin)	42, 22, 35, 45, 62, 46, 41	42	12	7	22	62	42	57
Erythromycin-H2O	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Fipronil	6.5, 8.5, 13, 19, 20, 20, 8.8	14	6	7	7	20	13	20
Fluoxetine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Furosemide	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Galaxolide	160, 100, 250, 360, 390, 270, 110	234	116	7	100	390	250	381
Gemfibrozil	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Ibuprofen	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Iohexol	5220, 4090, 2880, 3680, 7450, 2680, 5640	4520	1698	7	2680	7450	4090	6907
Iopromide	84, <10, <10, 70, 143, <10, 398	104	139	7	10	398	70	322

Compound Name	Results	Mean	Standard Deviation	Sample Size	Min	Max	Median	95th Percentile
Meprobamate	194, 124, 172, 177, 218, 195, 160	177	30	7			177	211
Metoprolol	<10, <10, <10, <10, 12, <10, <10	10	1	7	10	12	10	11
Naproxen	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Nonylphenol diethoxylate	106, 102, 116, 123, 120, 119, 106	113	8	7	102	123	116	122
Nonylphenol monoethoxylate	<25.0, <25.0, <25.0, 39.5, <25.0, <25.0, <25.0	27	5	7	25	40	25	35
Octylphenol diethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Octylphenol monoethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Primidone	45, 26, 43, 53, 74, 52, 38	47	15	7	26	74	45	68
Propranolol	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Sucralose	15400, 15200, 20200, 19900, 20600, 18200, 17500	18,143	2234	7	15,200	20,600	18,200	20,480
Sulfamethoxazole	<10, <10, <10, 11, 15, <10, <10	11	2	7	10	15	10	14
TCEP	225, 319, 429, 373, 231, 384, 269	319	80	7	225	429	319	416
TCPP	2280, 3580, 5220, 5860, 1900, 2220, 1480	3220	1720	7	1480	5860	2280	5668
TDCPP	444, 506, 567, 598, 484, 519, 405	503	67	7	405	598	506	589
Tonalide	<50, <50, <50, <50, <50, <50, <50	50	0	7	50	50	50	50
Triclocarban	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Triclosan	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Trimethoprim	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
4-Nonylphenol (tech mix)	156, 131, 145, 186, 212, 221, 184	176	34	7	131	221	184	218
4-*tert* Octylphenol	38.9, 14.8, 12, 21.6, 33.6, 23.7, 10.7	22	11	7	11	39	22	37

Table E.4. Cl_2 Soil Column System: Concentrations after Vadose

	Cl_2: After Vadose							
Compound Name	**Results**	**Mean**	**Standard Deviation**	**Sample Size**	**Minimum**	**Maximum**	**Median**	**95th Percentile**
Acesulfame-K	165.1, 80.2, 160.5, 263.3, 2205.8, 361.6, 412.3	521	752	7	80	2206	263	1668
Acetaminophen	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Atenolol	51, 37, <30, 31, <30, 26, 20	32	10	7	20	51	30	47
Atorvastatin	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Azithromycin	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Bisphenol A	13, <10, <10, <10, <10, <10, <10	10	1	7	10	13	10	12
Caffeine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Carbamazepine	330, 222, 207, 266, 223, 293, 296	262	46	7	207	330	266	320
Carisoprodol	144, 482, 197, 210, 146, 243, 243	238	115	7	144	482	210	410
DEET	291, 105, 94, 352, 129, 61, 52	155	118	7	52	352	105	334
Diazepam	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Diclofenac	<10, 21, <10, <10, 32, <10, <10	15	9	7	10	32	10	29
Dilantin (Phenytoin)	233, 152, 147, 152, 147, 156, 165	165	31	7	147	233	152	213
Erythromycin-H2O	<10, <10, <10, 14, <10, 12, <10	11	2	7	10	14	10	13
Fipronil	21, 28, 36, 37, 56, 44, 54	39	13	7	21	56	37	55
Fluoxetine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Furosemide	<10, 14, <10, <10, 12, <10, <10	11	2	7	10	14	10	13
Galaxolide	<50, <50, <50, <50, <50, <50, <50	50	0	7	50	50	50	50
Gemfibrozil	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Ibuprofen	273, <10, <10, <10, <10, <10, <10	48	99	7	10	273	10	194
Iohexol	596, 1250, 589, 251, 1280, 193, 179	620	474	7	179	1280	589	1,271
Iopromide	22, <10, <10, <10, <10, <10, <10	12	5	7	10	22	10	18

Compound Name	Results	Mean	Standard Deviation	Sample Size	Minimum	Maximum	Median	95th Percentile
Meprobamate	453, 329, 331, 321, 361, 333, 330	351	47	7	321	453	331	425
Metoprolol	167, 164, 131, 177, 180, 275, 267	194	55	7	131	275	177	273
Naproxen	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Nonylphenol diethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Nonylphenol monoethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Octylphenol diethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Octylphenol monoethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Primidone	323, 186, 194, 203, 209, 184, 180	211	50	7	180	323	194	289
Propranolol	18, <10, <10, 10, <10, 11, <10	11	3	7	10	18	10	16
Sucralose	47300, 28000, 31200, 31200, 27200, 30900, 29300	32,157	6865	7	27,200	47,300	30,900	42,470
Sulfamethoxazole	348, 569, 246, 577, 1600, 1030, 658	718	462	7	246	1600	577	1429
TCEP	564, 371, 343, 305, 285, 361, 352	369	91	7	285	564	352	506
TCPP	926, 810, 592, 1390, 255, 420, 338	676	399	7	255	1390	592	1251
TDCPP	314, 163, 250, 211, 171, 228, 177	216	54	7	163	314	211	295
Tonalide	<50, <50, <50, <50, <50, <50, <50	50	0	7	50	50	50	50
Triclocarban	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Triclosan	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Trimethoprim	18, 17, 14, 21, 10, 19, <10	16	4	7	10	21	17	20
4-Nonylphenol (tech mix)	1400, 5660, 1550, 600, 858, 626, 645	1620	1823	7	600	5660	858	4427
4-*tert* Octylphenol	1770, 6590, 2150, 429, 499, 295, 409	1735	2266	7	295	6590	499	5258

Table E.5. Cl_2 Soil Column System: Concentrations after Saturated

	Cl_2: After Saturated							
Compound Name	**Results**	**Mean**	**Standard Deviation**	**Sample Size**	**Minimum**	**Maximum**	**Median**	**95th Percentile**
Acesulfame-K	373.6, 268.8, 132.2, 76.6, 71.9, 109, 150.3, 183.2	171	104	8	72	374	141	337
Acetaminophen	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Atenolol	<30, <30, <30, <30, <30, <30, <30, <10	28	7	8	10	30	30	30
Atorvastatin	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Azithromycin	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Bisphenol A	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Caffeine	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Carbamazepine	244, 244, 231, 226, 237, 215, 224, 232	232	10	8	215	244	232	244
Carisoprodol	118, 253, 443, 593, 526, 402, 232, 226	349	166	8	118	593	328	570
DEET	324, 157, 123, 63, 106, 115, 105, 128	140	79	8	63	324	119	266
Diazepam	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Diclofenac	14, 11, <10, <10, <10, <10, <10, <10	11	1	8	10	14	10	13
Dilantin (Phenytoin)	159, 164, 166, 144, 146, 157, 150, 158	156	8	8	144	166	158	165
Erythromycin-H2O	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Fipronil	2.2, 39, <2.0, 30, 49, 31, 38, 33	28	17	8	2	49	32	46
Fluoxetine	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Furosemide	15, <10, 14, <10, <10, <10, <10, <10	11	2	8	10	15	10	15
Galaxolide	<50, <50, <50, <50, <50, <50, <50, <50	50	0	8	50	50	50	50
Gemfibrozil	26, 13, <10, <10, <10, <10, <10, <10	12	6	8	10	26	10	21
Ibuprofen	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Iohexol	3650, 1650, 2030, 1350, 704, 502, 281, 233	1300	1156	8	233	3650	1027	3083
Iopromide	47, 39, 44, 36, 19, <10, <10, <10	27	16	8	10	47	28	46
Meprobamate	356, 337, 341, 301, 279, 199, 80, 120	252	106	8	80	356	290	351

Compound Name	Results	Mean	Standard Deviation	Sample Size	Minimum	Maximium	Median	95th Percentile
Metoprolol	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Naproxen	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Nonylphenol diethoxylate	<25.0, 154, 42, <40.0, 125, 93.1, 91, 56.5	78	45	8	25	154	74	144
Nonylphenol monoethoxylate	<25.0, 153, 47, <40.0, 168, 149, 150, 95.9	103	59	8	25	168	122	163
Octylphenol diethoxylate	<25.0, <25.0, <25.0, <40.0, <25.0, <25.0, <25.0, <25.0	27	5	8	25	40	25	35
Octylphenol monoethoxylate	<25.0, 48.7, <25.0, <40.0, 72.5, 70.3, 68.6, 47.7	50	19	8	25	73	48	72
Primidone	203, 222, 198, 191, 187, 184, 189, 196	196	12	8	184	222	194	215
Propranolol	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Sucralose	28100, 30300, 30700, 27700, 28700, 28400, 30100, 31100	29,388	1307	8	27,700	31,100	29,400	30,960
Sulfamethoxazole	292, 430, 485, 559, 526, 424, 279, 288	410	112	8	279	559	427	547
TCEP	372, 368, 360, 316, 362, 374, 368, 340	358	20	8	316	374	365	373
TCPP	2910, 1520, 1230, 1560, 987, 991, 854, 855	1363	684	8	854	2910	1111	2438
TDCPP	331, 237, 234, 225, 176, 185, 179, 175	218	53	8	175	331	205	298
Tonalide	<50, <50, <50, <50, <50, <50, <50, <50	50	0	8	50	50	50	50
Triclocarban	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Triclosan	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Trimethoprim	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
4-Nonylphenol (tech mix)	360, 6550, 2010, 199, 4130, 4060, 3180, 1880	2796	2130	8	199	6550	2595	5703
4-*tert* Octylphenol	360, 3840, 1270, 130, 1980, 1640, 1230, 871	1415	1157	8	130	3840	1250	3189

Table E.6. O_3 Soil Column System: Concentrations After Vadose

O_3: After Vadose								
Compound Name	**Results**	**Mean**	**Standard Deviation**	**Sample Size**	**Minimum**	**Maximum**	**Median**	**95th Percentile**
Acesulfame-K	<50.0, <50.0, <50.0, 255.5, 934.6, 454.1, 309.7	301	320	7	50	935	256	790
Acetaminophen	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Atenolol	<30, <30, <30, <30, <30, <10, <10	24	10	7	10	30	30	30
Atorvastatin	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Azithromycin	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Bisphenol A	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Caffeine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Carbamazepine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Carisoprodol	35, 96, 77, 84, 71, 108, 96	81	24	7	35	108	84	104
DEET	<10, 12, <10, 26, 18, <10, <10	14	6	7	10	26	10	24
Diazepam	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Diclofenac	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Dilantin (Phenytoin)	17, 17, 23, 43, 41, 29, 26	28	11	7	17	43	26	42
Erythromycin-H2O	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Fipronil	7.2, 8.2, 9.7, 19, 22, 16, 10	13	6	7	7	22	10	21
Fluoxetine	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Furosemide	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Galaxolide	<50, <50, <50, <50, <50, <50, <50	50	0	7	50	50	50	50
Gemfibrozil	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Ibuprofen	30, <10, <10, <10, <10, <10, <10	13	8	7	10	30	10	24
Iohexol	170, 361, 176, 453, 2000, 135, <100	485	680	7	100	2000	176	1536
Iopromide	<10, <10, <10, <10, 19, <10, <10	11	3	7	10	19	10	16

Compound Name	Results	Mean	Standard Deviation	Sample Size	Min	Max	Median	95th Percentile
Meprobamate	122, 146, 151, 186, 207, 152, 153	160	28	7	122	207	152	201
Metoprolol	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Naproxen	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Nonylphenol diethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Nonylphenol monoethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Octylphenol diethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Octylphenol monoethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	7	25	25	25	25
Primidone	28, 30, 39, 60, 71, 48, 43	46	16	7	28	71	43	68
Propranolol	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Sucralose	15000, 14900, 17700, 19100, 20100, 18400, 17600	17,543	1965	7	14,900	20,100	17,700	19,800
Sulfamethoxazole	12, 19, 47, 98, 115, 73, 64	61	38	7	12	115	64	110
TCEP	207, 276, 288, 363, 268, 275, 277	279	46	7	207	363	276	341
TCPP	167, 390, 244, 1110, 797, 296, 334	477	345	7	167	1110	334	1016
TDCPP	133, 132, 162, 176, 191, 190, 159	163	24	7	132	191	162	191
Tonalide	<50, <50, <50, <50, <50, <50, <50	50	0	7	50	50	50	50
Triclocarban	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Triclosan	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
Trimethoprim	<10, <10, <10, <10, <10, <10, <10	10	0	7	10	10	10	10
4-Nonylphenol (tech mix)	1830, 5220, 1550, 589, 480, 493, 431	1513	1731	7	431	5220	589	4203
4-*tert* Octylphenol	1570, 5160, 1780, 372, 302, 206, 270	1380	1792	7	206	5160	372	4146

Table E.7. O_3 Soil Column System: Concentrations After Saturated

		O_3: After Saturated						
Compound Name	**Results**	**Mean**	**Standard Deviation**	**Sample Size**	**Min**	**Max**	**Median**	**95th Percentile**
Acesulfame-K	<50.0, 51.5, 62.2, 66.9, 56.5, <50.0, <50.0, 64.8	56	7	8	50	67	54	66
Acetaminophen	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Atenolol	<30, <30, <30, <30, <30, <30, <30, <10	28	7	8	10	30	30	30
Atorvastatin	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Azithromycin	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Bisphenol A	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Caffeine	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Carbamazepine	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Carisoprodol	25, 59, 89, 125, 101, 83, 53, 70	76	31	8	25	125	77	117
DEET	15, 16, 14, 25, 34, 30, 29, 36	25	9	8	14	36	27	35
Diazepam	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Diclofenac	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Dilantin (Phenytoin)	<10, 15, 16, 19, 16, 17, 20, 24	17	4	8	10	24	17	23
Erythromycin-H2O	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Fipronil	<2, 5, 4.6, 6, 7.6, 7.5, 10, 8.4	6	3	8	2	10	7	9
Fluoxetine	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Furosemide	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Galaxolide	<50, <50, <50, <50, <50, <50, <50, <50	50	0	8	50	50	50	50
Gemfibrozil	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Ibuprofen	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Iohexol	155, 224, 314, 355, 215, 120, <100, <100	198	97	8	100	355	185	341

Compound Name	Results	Mean	Standard Deviation	Sample Size	Min	Max	Median	95th Percentile
Iopromide	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Meprobamate	99, 126, 138, 150, 125, 59, <10, 19	91	55	8	10	150	112	146
Metoprolol	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Naproxen	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Nonylphenol diethoxylate	<25.0, <25.0, <25.0, <25.0, 83.6, 104, 80.9, 47.7	52	33	8	25	104	36	97
Nonylphenol monoethoxylate	<25.0, <25.0, <25.0, <25.0, 94.4, 134, 105, 88.5	65	45	8	25	134	57	124
Octylphenol diethoxylate	<25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0, <25.0	25	0	8	25	25	25	25
Octylphenol monoethoxylate	<25.0, <25.0, <25.0, <25.0, 49.9, 62.5, 56.8, 53.3	40	17	8	25	63	37	61
Primidone	13, 29, 34, 38, 30, 32, 38, 41	32	9	8	13	41	33	40
Propranolol	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Sucralose	12500, 15300, 15300, 19200, 14800, 14900, 17400, 18700	16,013	2248	8	12,500	19,200	15,300	19,025
Sulfamethoxazole	<10, 14, 11, 14, 18, 29, 41, 37	22	12	8	10	41	16	40
TCEP	311, 210, 221, 216, 250, 268, 296, 293	258	40	8	210	311	259	306
TCPP	670, 390, 413, 610, 590, 553, 525, 660	551	105	8	390	670	572	667
TDCPP	167, 141, 127, 123, 106, 109, 110, 123	126	20	8	106	167	123	158
Tonalide	<50, <50, <50, <50, <50, <50, <50, <50	50	0	8	50	50	50	50
Triclocarban	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Triclosan	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
Trimethoprim	<10, <10, <10, <10, <10, <10, <10, <10	10	0	8	10	10	10	10
4-Nonylphenol (tech mix)	287, 132, 103, 257, 2260, 2770, 1990, 1800	1200	1111	8	103	2770	1044	2592
4-*tert* Octylphenol	459, 151, 106, 279, 1310, 1570, 1030, 827	717	553	8	106	1570	643	1479

Appendix F

CEC Occurrence at Each Sample Location

Table F.1. Occurrence Data for Secondary

Secondary	
Detects	**Non-Detects**
4-Nonylphenol (tech mix) (6)	Acetaminophen (4)
4-*tert* Octylphenol (6)	Atorvastatin (6)
Acesulfame-K (6)	Bisphenol A (6)
Acetaminophen (2)	Caffeine (6)
Atenolol (6)	Diazepam (6)
Azithromycin (6)	Ibuprofen (3)
Carbamazepine (6)	Iopromide (1)
Carisoprodol (6)	*N*-Nitroso-*n*-propylamine (1)
DEET (6)	*N*-Nitrosodi-*n*-butylamine (1)
Diclofenac (6)	*N*-Nitrosodiethylamine (1)
Dilantin (Phenytoin) (6)	*N*-Nitrosomethylethylamine (1)
Erythromycin-H2O (6)	*N*-Nitrosopiperidine (1)
Fipronil (6)	*N*-Nitrosopyrrolidine (1)
Fluoxetine (6)	Naproxen (2)
Furosemide (6)	Octylphenol diethoxylate (1)
Galaxolide (6)	Triclosan (1)
Gemfibrozil (6)	
Ibuprofen (3)	
Iohexol (6)	
Iopromide (5)	
Meprobamate (6)	
Metoprolol (6)	
N-Nitrosodimethylamine (1)	
Naproxen (4)	
Nonylphenol diethoxylate (6)	
Nonylphenol monoethoxylate (6)	
Octylphenol diethoxylate (5)	
Octylphenol monoethoxylate (6)	
Primidone (6)	
Propranolol (6)	
Sucralose (6)	
Sulfamethoxazole (6)	
TCEP (6)	
TCPP (6)	
TDCPP (6)	
Tonalide (6)	
Triclocarban (6)	
Triclosan (5)	
Trimethoprim (6)	

Note: (N) = sample number

Table F.2. Occurrence Data for Chlorination

Chlorinated	
Detects	**Non-Detects**
4-Nonylphenol (tech mix) (8)	Acetaminophen (2)
4-*tert* Octylphenol (8)	Atorvastatin (7)
Acesulfame-K (8)	Bisphenol A (5)
Acetaminophen (6)	Caffeine (7)
Atenolol (8)	Diazepam (7)
Atorvastatin (1)	Dibromoacetic Acid (3)
Azithromycin (8)	Diclofenac (2)
Bisphenol A (3)	Ibuprofen (3)
Caffeine (1)	Iopromide (2)
Carbamazepine (8)	Monobromoacetic Acid (3)
Carisoprodol (8)	Monochloroacetic Acid (2)
DEET (8)	*N*-Nitroso-*n*-propylamine (5)
Diazepam (1)	*N*-Nitrosodi-*n*-butylamine (5)
Dichloroacetic Acid (DCA) (3)	*N*-Nitrosodiethylamine (4)
Diclofenac (6)	*N*-Nitrosomethylethylamine (5)
Dilantin (Phenytoin) (8)	*N*-Nitrosopiperidine (3)
Erythromycin-H2O (8)	*N*-Nitrosopyrrolidine (5)
Fipronil (8)	Naproxen (4)
Fluoxetine (8)	Octylphenol diethoxylate (2)
Furosemide (8)	Triclosan (6)
Galaxolide (8)	
Gemfibrozil (8)	
Haloacetic acids (HAA5) (3)	
Ibuprofen (5)	
Iohexol (8)	
Iopromide (6)	
Meprobamate (8)	
Metoprolol (8)	
Monochloroacetic Acid (1)	
N-Nitrosodiethylamine (1)	
N-Nitrosodimethylamine (5)	
N-Nitrosopiperidine (2)	
Naproxen (4)	
Nonylphenol diethoxylate (8)	
Nonylphenol monoethoxylate (8)	
Octylphenol diethoxylate (6)	
Octylphenol monoethoxylate (8)	
Primidone (8)	
Propranolol (8)	
Sucralose (8)	
Sulfamethoxazole (8)	
TCEP (8)	
TCPP (8)	
TDCPP (8)	
Tonalide (8)	
Total Trihalomethanes (3)	
Trichloroacetic acid (3)	
Triclocarban (8)	
Triclosan (2)	
Trimethoprim (8)	

Table F.3. Occurrence Data for Cl_2 Soil Column System: After Vadose

Cl_2: After Vadose	
Detects	**Non-Detects**
4-Nonylphenol (tech mix) (7)	Acetaminophen (7)
4-*tert* Octylphenol (7)	Atenolol (2)
Acesulfame-K (7)	Atorvastatin (7)
Atenolol (5)	Azithromycin (7)
Bisphenol A (1)	Bisphenol A (6)
Carbamazepine (7)	Caffeine (7)
Carisoprodol (7)	Diazepam (7)
DEET (7)	Diclofenac (5)
Diclofenac (2)	Erythromycin-H2O (5)
Dilantin (Phenytoin) (7)	Fluoxetine (7)
Erythromycin-H2O (2)	Furosemide (5)
Fipronil (7)	Galaxolide (7)
Furosemide (2)	Gemfibrozil (7)
Ibuprofen (1)	Ibuprofen (6)
Iohexol (7)	Iopromide (6)
Iopromide (1)	*N*-Nitroso-*n*-propylamine (5)
Meprobamate (7)	*N*-Nitrosodi-*n*-butylamine (4)
Metoprolol (7)	*N*-Nitrosodiethylamine (3)
N-Nitrosodi-*n*-butylamine (1)	*N*-Nitrosodimethylamine (2)
N-Nitrosodiethylamine (2)	*N*-Nitrosomethylethylamine (5)
N-Nitrosodimethylamine (3)	*N*-Nitrosopiperidine (2)
N-Nitrosopiperidine (3)	*N*-Nitrosopyrrolidine (5)
Primidone (7)	Naproxen (7)
Propranolol (3)	Nonylphenol diethoxylate (7)
Sucralose (7)	Nonylphenol monoethoxylate (7)
Sulfamethoxazole (7)	Octylphenol diethoxylate (7)
TCEP (7)	Octylphenol monoethoxylate (7)
TCPP (7)	Propranolol (4)
TDCPP (7)	Tonalide (7)
Trimethoprim (6)	Triclocarban (7)
	Triclosan (7)
	Trimethoprim (1)

Note: (N) = sample size

Table F.4. Occurrence Data for Cl_2 Soil Column System: After Saturated

Cl_2: After Saturated	
Detects	**Non-detects**
4-Nonylphenol (tech mix) (8)	Acetaminophen (8)
4-*tert* Octylphenol (8)	Atenolol (8)
Acesulfame-K (8)	Atorvastatin (8)
Carbamazepine (8)	Azithromycin (8)
Carisoprodol (8)	Bisphenol A (8)
DEET (8)	Caffeine (8)
Diclofenac (2)	Diazepam (8)
Dilantin (Phenytoin) (8)	Dibromoacetic Acid (3)
Fipronil (7)	Dichloroacetic Acid (DCA) (3)
Furosemide (2)	Diclofenac (6)
Gemfibrozil (2)	Erythromycin-H2O (8)
Iohexol (8)	Fipronil (1)
Iopromide (5)	Fluoxetine (8)
Meprobamate (8)	Furosemide (6)
N-Nitrosodi-*n*-butylamine (1)	Galaxolide (8)
N-Nitrosodiethylamine (3)	Gemfibrozil (6)
N-Nitrosodimethylamine (1)	Haloacetic acids (HAA5) (3)
N-Nitrosopyrrolidine (1)	Ibuprofen (8)
Nonylphenol diethoxylate (6)	Iopromide (3)
Nonylphenol monoethoxylate (6)	Metoprolol (8)
Octylphenol monoethoxylate (5)	Monobromoacetic Acid (3)
Primidone (8)	Monochloroacetic Acid (3)
Sucralose (8)	*N*-Nitroso-*n*-propylamine (7)
Sulfamethoxazole (8)	*N*-Nitrosodi-*n*-butylamine (6)
TCEP (8)	*N*-Nitrosodiethylamine (4)
TCPP (8)	*N*-Nitrosodimethylamine (6)
TDCPP (8)	*N*-Nitrosomethylethylamine (7)
	N-Nitrosopiperidine (7)
	N-Nitrosopyrrolidine (6)
	Naproxen (8)
	Nonylphenol diethoxylate (2)
	Nonylphenol monoethoxylate (2)
	Octylphenol diethoxylate (8)
	Octylphenol monoethoxylate (3)
	Propranolol (8)
	Tonalide (8)
	Total Trihalomethanes (3)
	Trichloroacetic acid (3)
	Triclocarban (8)
	Triclosan (8)
	Trimethoprim (8)

Note: (N) = sample size

Table F.5. Occurrence Data for Ozonation

Ozonated	
Detects	**Non-Detects**
4-Nonylphenol (tech mix) (7)	Acesulfame-K (3)
4-*tert* Octylphenol (7)	Acetaminophen (7)
Acesulfame-K (4)	Atenolol (7)
Bisphenol A (7)	Atorvastatin (7)
Bromate (2)	Azithromycin (7)
Carisoprodol (7)	Bromate (1)
DEET (5)	Caffeine (7)
Dilantin (Phenytoin) (7)	Carbamazepine (7)
Fipronil (7)	DEET (2)
Galaxolide (7)	Diazepam (7)
Iohexol (7)	Diclofenac (7)
Iopromide (4)	Erythromycin-H2O (7)
Meprobamate (7)	Fluoxetine (7)
Metoprolol (1)	Furosemide (7)
N-Nitrosodiethylamine (1)	Gemfibrozil (7)
N-Nitrosodimethylamine (5)	Ibuprofen (7)
N-Nitrosopiperidine (3)	Iopromide (3)
Nonylphenol diethoxylate (7)	Metoprolol (6)
Nonylphenol monoethoxylate (1)	*N*-Nitroso-*n*-propylamine (5)
Primidone (7)	*N*-Nitrosodi-*n*-butylamine (5)
Sucralose (7)	*N*-Nitrosodiethylamine (4)
Sulfamethoxazole (2)	*N*-Nitrosomethylethylamine (5)
TCEP (7)	*N*-Nitrosopiperidine (2)
TCPP (7)	*N*-Nitrosopyrrolidine (5)
TDCPP (7)	Naproxen (7)
	Nonylphenol monoethoxylate (6)
	Octylphenol diethoxylate (7)
	Octylphenol monoethoxylate (7)
	Propranolol (7)
	Sulfamethoxazole (5)
	Tonalide (7)
	Triclocarban (7)
	Triclosan (7)
	Trimethoprim (7)

Note: (N) = sample size

Table F.6. Occurrence Data for O_3 Soil Column System: After Vadose

O_3: After Vadose	
Detects	**Non-Detects**
4-Nonylphenol (tech mix) (7)	Acesulfame-K (3)
4-*tert* Octylphenol (7)	Acetaminophen (7)
Acesulfame-K (4)	Atenolol (7)
Carisoprodol (7)	Atorvastatin (7)
DEET (3)	Azithromycin (7)
Dilantin (Phenytoin) (7)	Bisphenol A (7)
Fipronil (7)	Caffeine (7)
Ibuprofen (1)	Carbamazepine (7)
Iohexol (6)	DEET (4)
Iopromide (1)	Diazepam (7)
Meprobamate (7)	Diclofenac (7)
N-Nitrosodi-*n*-butylamine (1)	Erythromycin-H2O (7)
N-Nitrosodiethylamine (1)	Fluoxetine (7)
N-Nitrosodimethylamine (1)	Furosemide (7)
Primidone (7)	Galaxolide (7)
Sucralose (7)	Gemfibrozil (7)
Sulfamethoxazole (7)	Ibuprofen (6)
TCEP (7)	Iohexol (1)
TCPP (7)	Iopromide (6)
TDCPP (7)	Metoprolol (7)
	N-Nitroso-*n*-propylamine (5)
	N-Nitrosodi-*n*-butylamine (4)
	N-Nitrosodiethylamine (4)
	N-Nitrosodimethylamine (4)
	N-Nitrosomethylethylamine (5)
	N-Nitrosopiperidine (5)
	N-Nitrosopyrrolidine (5)
	Naproxen (7)
	Nonylphenol diethoxylate (7)
	Nonylphenol monoethoxylate (7)
	Octylphenol diethoxylate (7)
	Octylphenol monoethoxylate (7)
	Propranolol (7)
	Tonalide (7)
	Triclocarban (7)
	Triclosan (7)
	Trimethoprim (7)

Note: (N) = sample size

Table F.7. Occurrence Data for O_3 Soil Column System: After Saturated

O_3: After Saturated	
Detects	**Non-Detects**
4-Nonylphenol (tech mix) (8)	Acesulfame-K (3)
4-*tert* Octylphenol (8)	Acetaminophen (8)
Acesulfame-K (5)	Atenolol (8)
Carisoprodol (8)	Atorvastatin (8)
DEET (8)	Azithromycin (8)
Dilantin (Phenytoin) (7)	Bisphenol A (8)
Fipronil (7)	Bromate (3)
Iohexol (6)	Caffeine (8)
Meprobamate (7)	Carbamazepine (8)
N-Nitrosodi-*n*-butylamine (1)	Diazepam (8)
N-Nitrosodiethylamine (2)	Diclofenac (8)
N-Nitrosopyrrolidine (1)	Dilantin (Phenytoin) (1)
Nonylphenol diethoxylate (4)	Erythromycin-H2O (8)
Nonylphenol monoethoxylate (4)	Fipronil (1)
Octylphenol monoethoxylate (4)	Fluoxetine (8)
Primidone (8)	Furosemide (8)
Sucralose (8)	Galaxolide (8)
Sulfamethoxazole (7)	Gemfibrozil (8)
TCEP (8)	Ibuprofen (8)
TCPP (8)	Iohexol (2)
TDCPP (8)	Iopromide (8)
	Meprobamate (1)
	Metoprolol (8)
	N-Nitroso-*n*-propylamine (6)
	N-Nitrosodi-*n*-butylamine (5)
	N-Nitrosodiethylamine (4)
	N-Nitrosodimethylamine (6)
	N-Nitrosomethylethylamine (6)
	N-Nitrosopiperidine (6)
	N-Nitrosopyrrolidine (5)
	Naproxen (8)
	Nonylphenol diethoxylate (4)
	Nonylphenol monoethoxylate (4)
	Octylphenol diethoxylate (8)
	Octylphenol monoethoxylate (4)
	Propranolol (8)
	Sulfamethoxazole (1)
	Tonalide (8)
	Triclocarban (8)
	Triclosan (8)
	Trimethoprim (8)

Note: (N) = sample size

Appendix G

CEC Measurement Anomalies

Some CECs showed apparent increases after a treatment step. These increases are explored in the following for the situations in which the concentrations at the two sampling locations being compared were statistically significant (using the student t-test with an alpha of 0.05). Each treatment step is discussed in the following.

Chlorination

Acetaminophen and sucralose apparently increased after chorine disinfection.

Acetaminophen

There was a small increase observed after chlorination, with concentrations slightly above the detection limit. It is unlikely that acetaminophen is being created by chlorination, and its apparent increase is likely linked to the analytical method. Methods designed to enumerate CECs are typically detecting them at extremely low concentrations in the nanogram range; thus, the quality of the water may impact the levels measured. The apparent increase after chlorination may be due to increased efficiency in the recovery step during extraction from the cleaner water source.

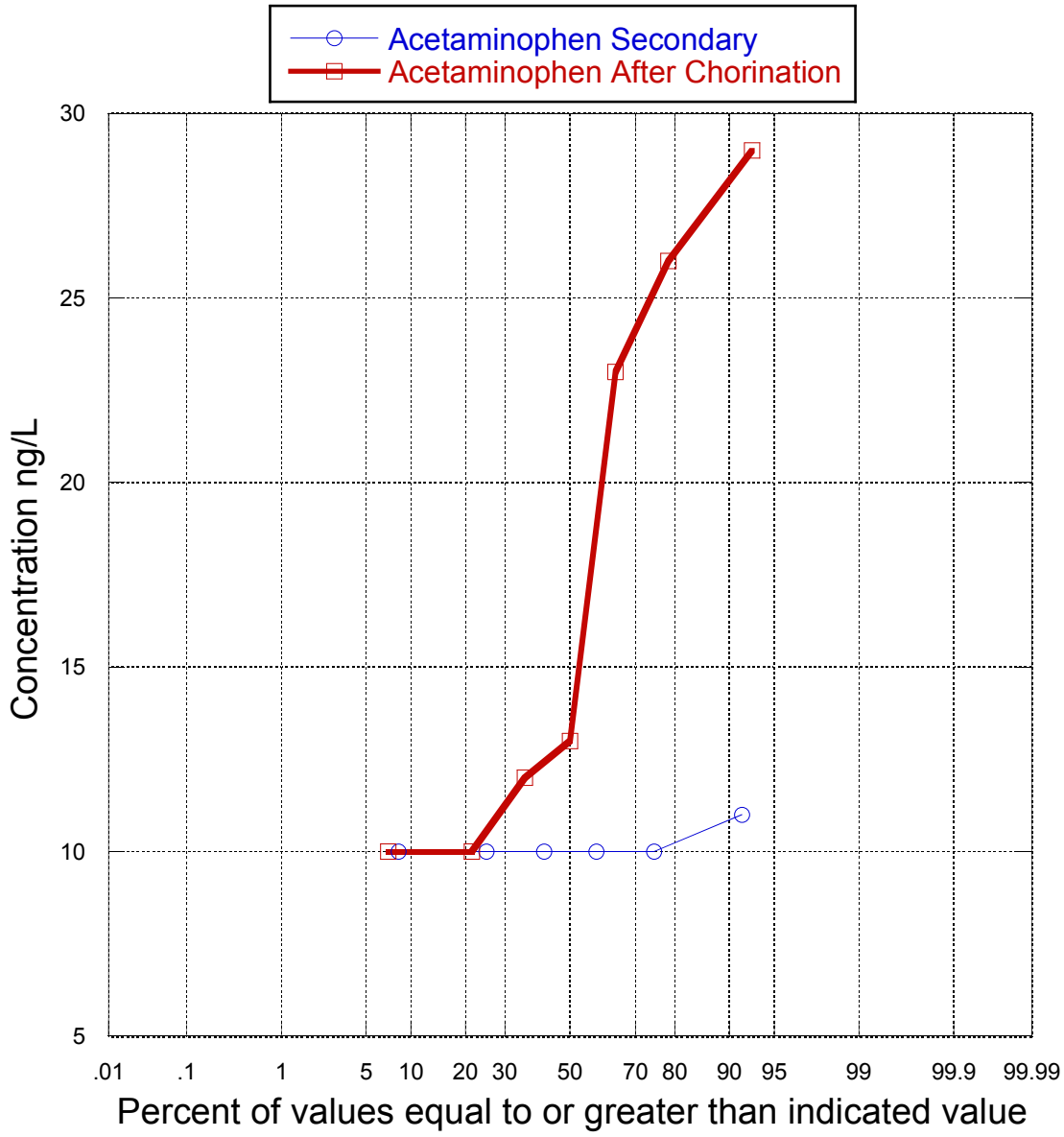

Figure G.1. Probability plot of acetaminophen concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Sucralose

Sucralose is a chlorinated sugar, but it is unlikely that chlorination formed this complex molecule. Visually, if the concentrations of sucralose in the secondary and chlorinated waters were looked at collectively, one would likely conclude that they are from the same data set. This might also be a situation in which sucralose was more efficiently recovered in the cleaner, disinfected water.

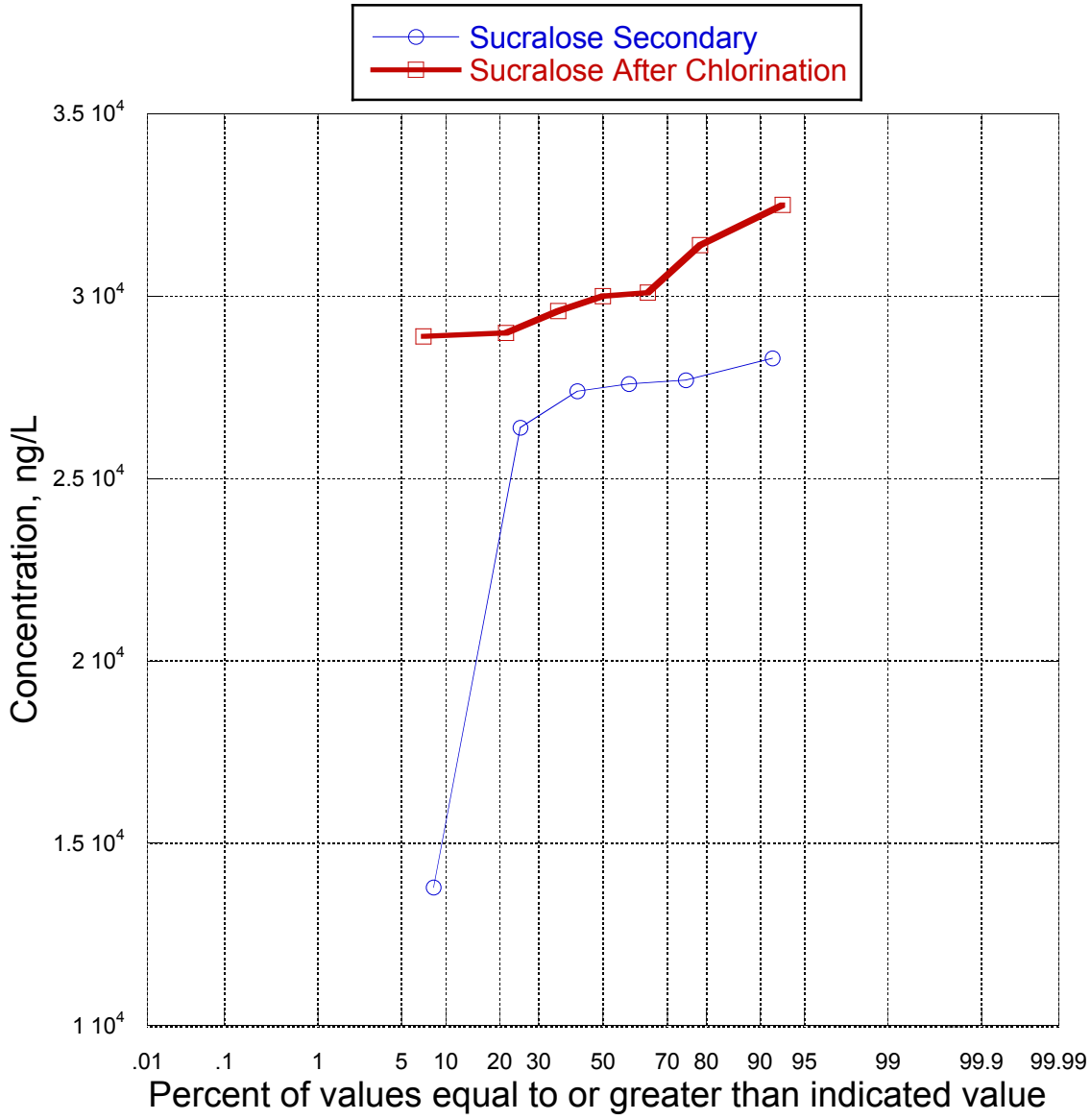

Figure G.2. Probability plot of sucralose concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Ozonation:

Bisphenol A

Bisphenol A is seemingly generated by ozonation. This increase is likely due to contamination issues. Drewes et al. (2011) also reported contamination issues around the detection of bisphenol A and speculated that the contamination was introduced during sampling and/or analysis.

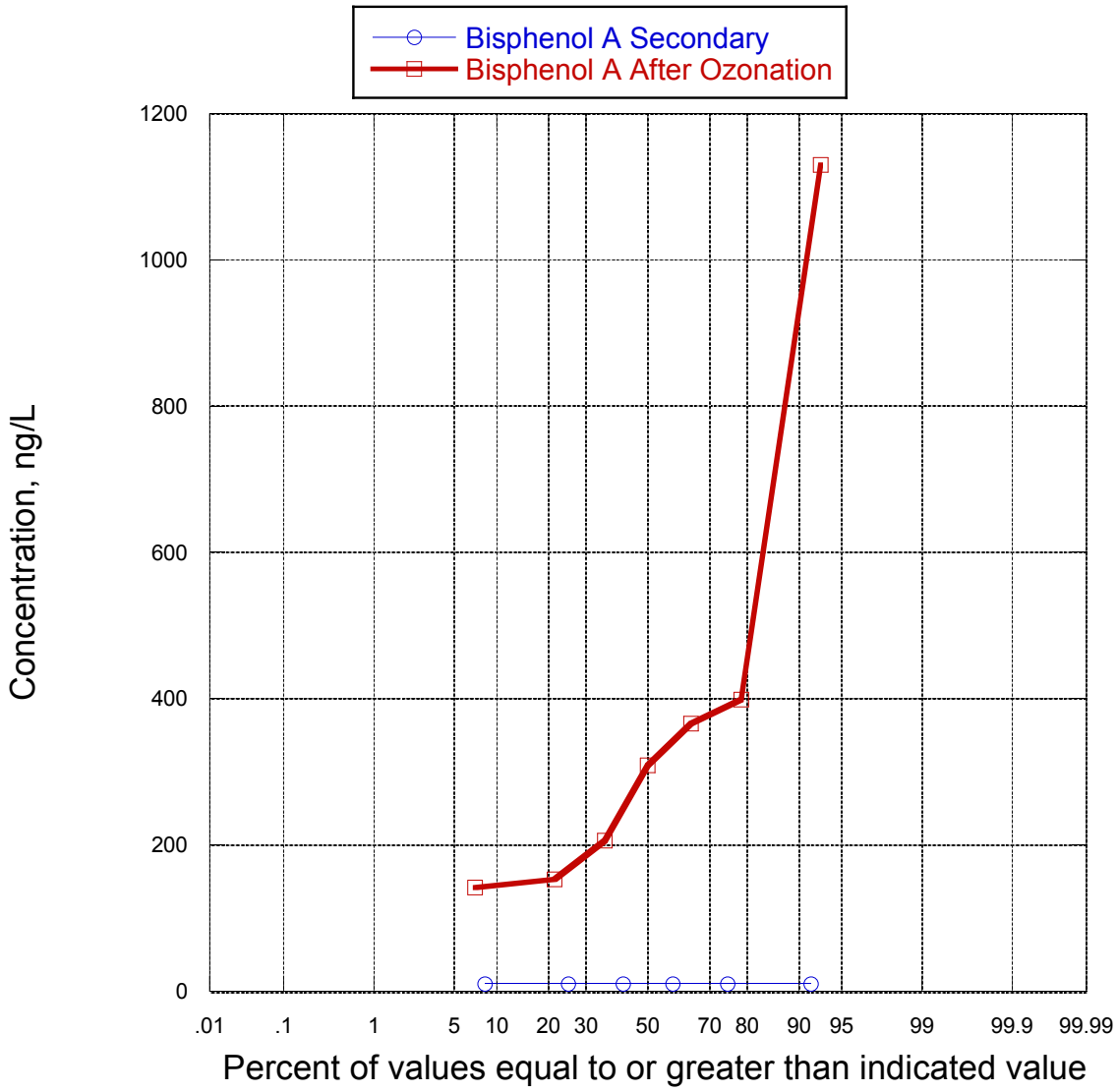

Figure G.3. Probability plot of bisphelol a concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Vadose

4-Nonylphenol (tech mix)

4-Nonylphenol increased through the vadose zone of both the Cl_2 and O_3 Soil Column Systems. Nonylphenols are known to have extremely complex metabolic pathways in the environment that make data difficult to interpret (Montgomery-Brown et al., 2008). With the soil columns, 4-nonylphenol was likely a biodegradation product, as it was not present in the secondary wastewater but was present in biodegraded effluents. Drewes et al. (2011) also observed such an increase on one occasion.

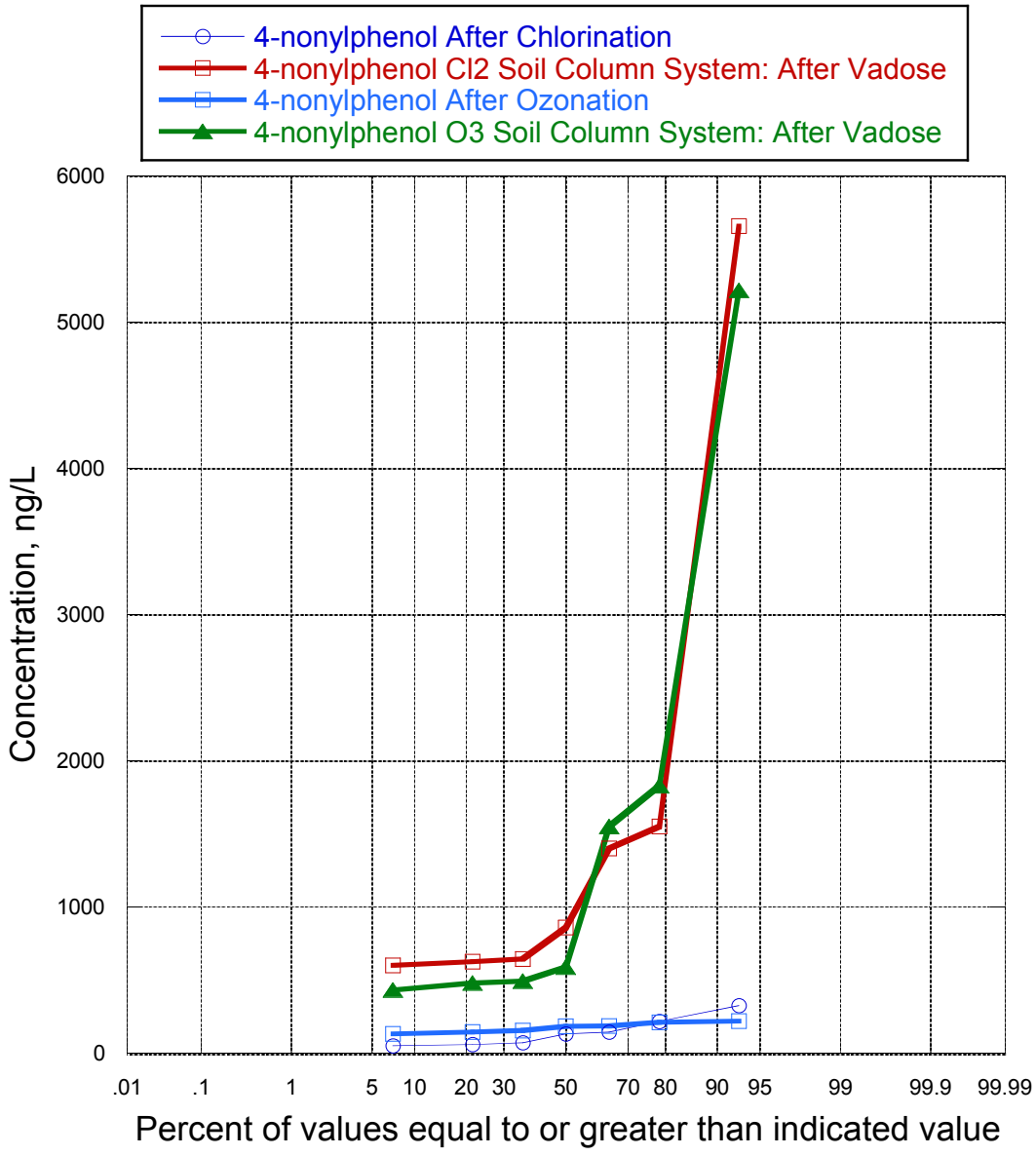

Figure G.4. Probability plot of 4-nonylphenol concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

4-tert Octylphenol

4-*tert* Octylphenol increased through the vadose zone of both the Cl_2 and O_3 Soil Column Systems. See the explanation on biodegradation byproducts provided previously for 4-nonylphenol (tech mix).

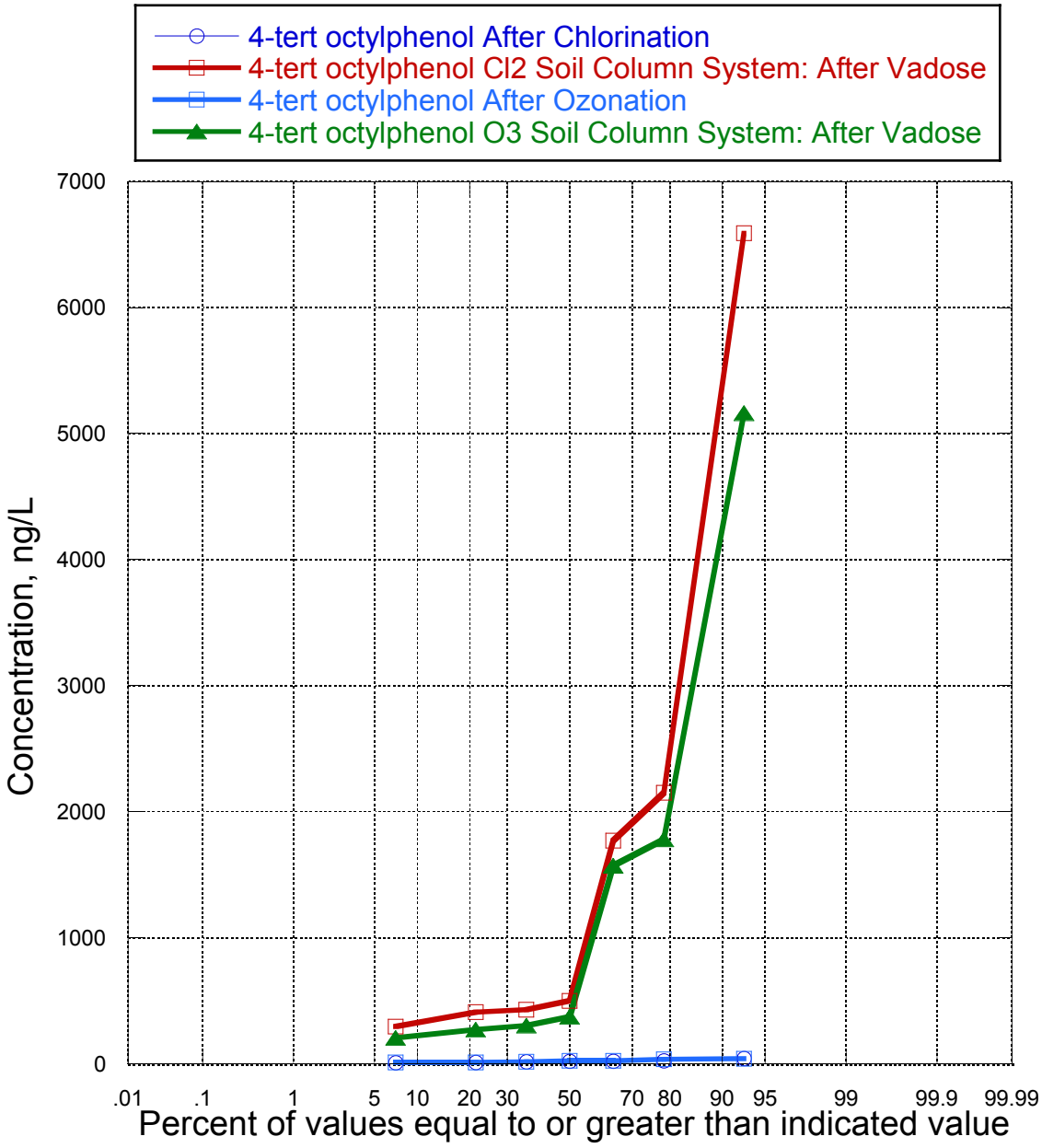

Figure G.5. Probability plot of 4-*tert* octylphenol concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Sulfamethoxazole

Sulfamethoxazole showed an apparent increase after SAT, particularly SAT preceded by ozonation. It is unlikely that the bioprocesses in the vadose zone of the ozone-fed water reconstructed a molecule like sulfamethoxazole. The increase in sulfamethoxazole may be a result of it being weakly absorbed to the soil and leaching out over time. Drewes et al. (2011) also noticed increases on two occasions during their SAT testing.

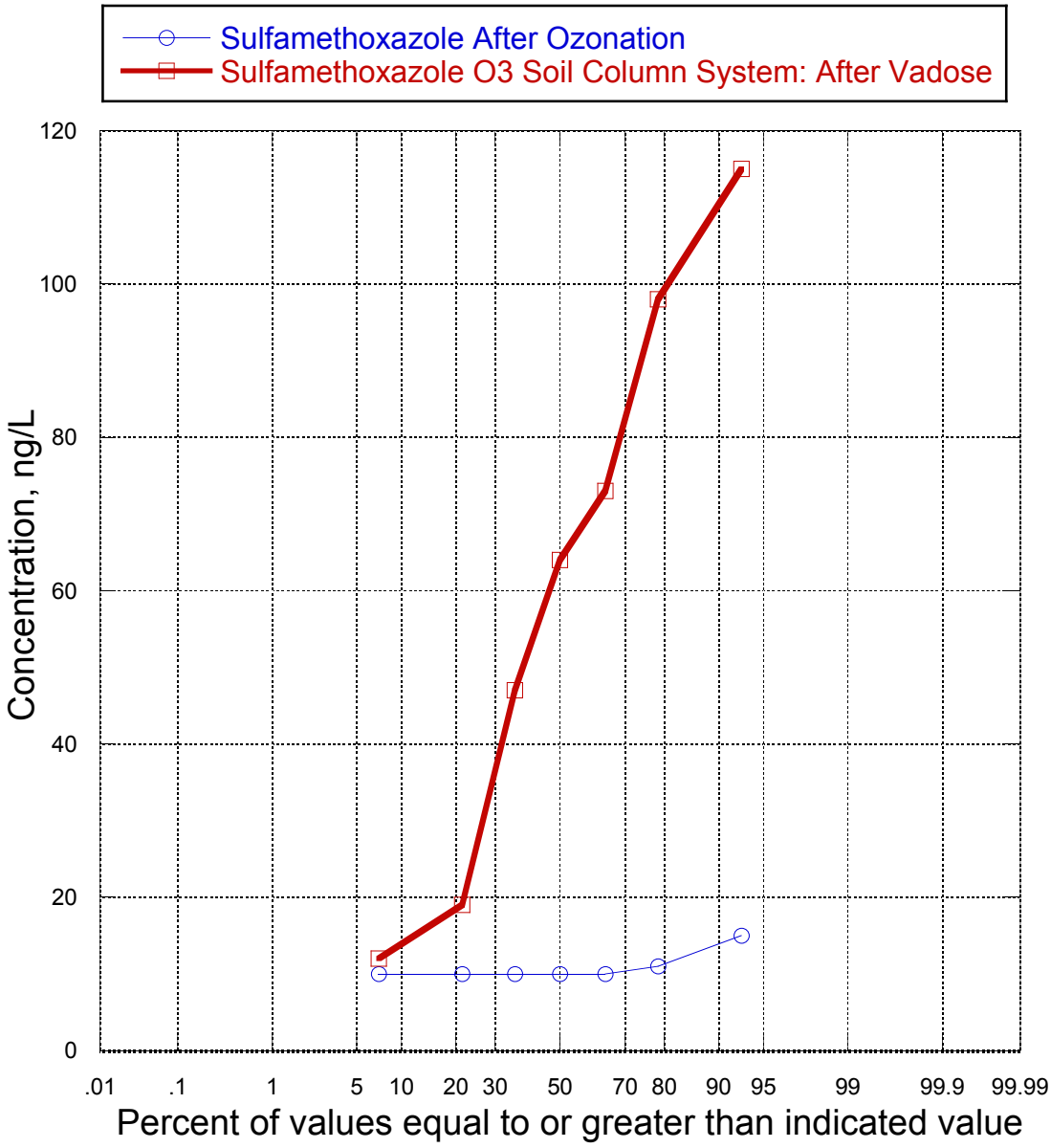

Figure G.6. Probability plot of sulfamethoxazole concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Saturated

DEET

DEET seemingly increased through the saturated column in the O_3 Soil Column System. It is possible that DEET was weakly absorbed to the soil and leached out over time. Drewes et al. (2011) also noticed increases on multiple occasions during their SAT testing.

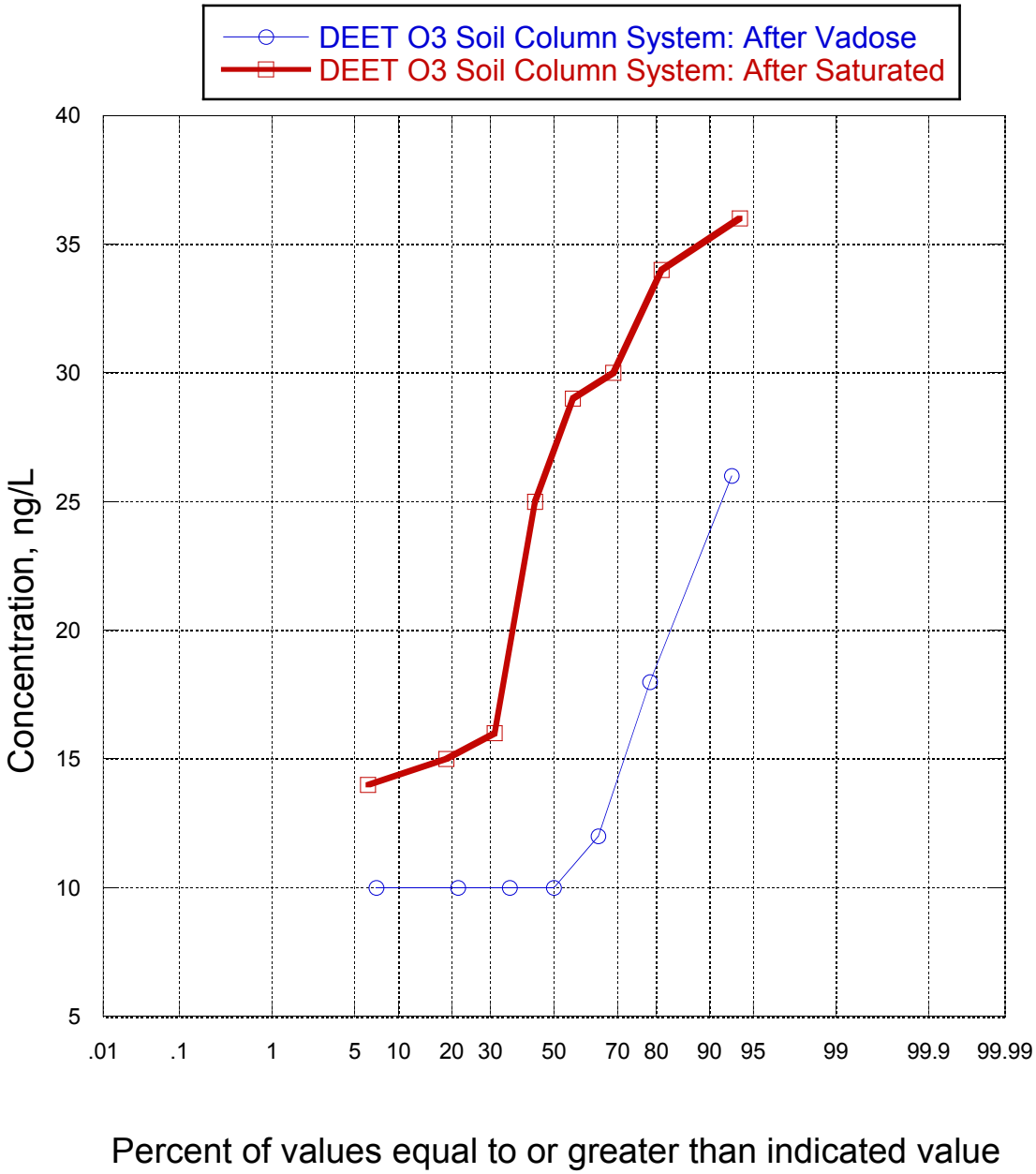

Figure G.7. Probability plot of DEET concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Iopromide

A small increase in iopromide was noticed after water percolated through the saturated column in the Cl_2 Soil Column System. Again, this may be because of the increased resolution with cleaner samples; thus, the increase may be related to the analytical method. Drewes et al. (2011) also observed some level of variability, noting both increases and decreases in concentration with travel time.

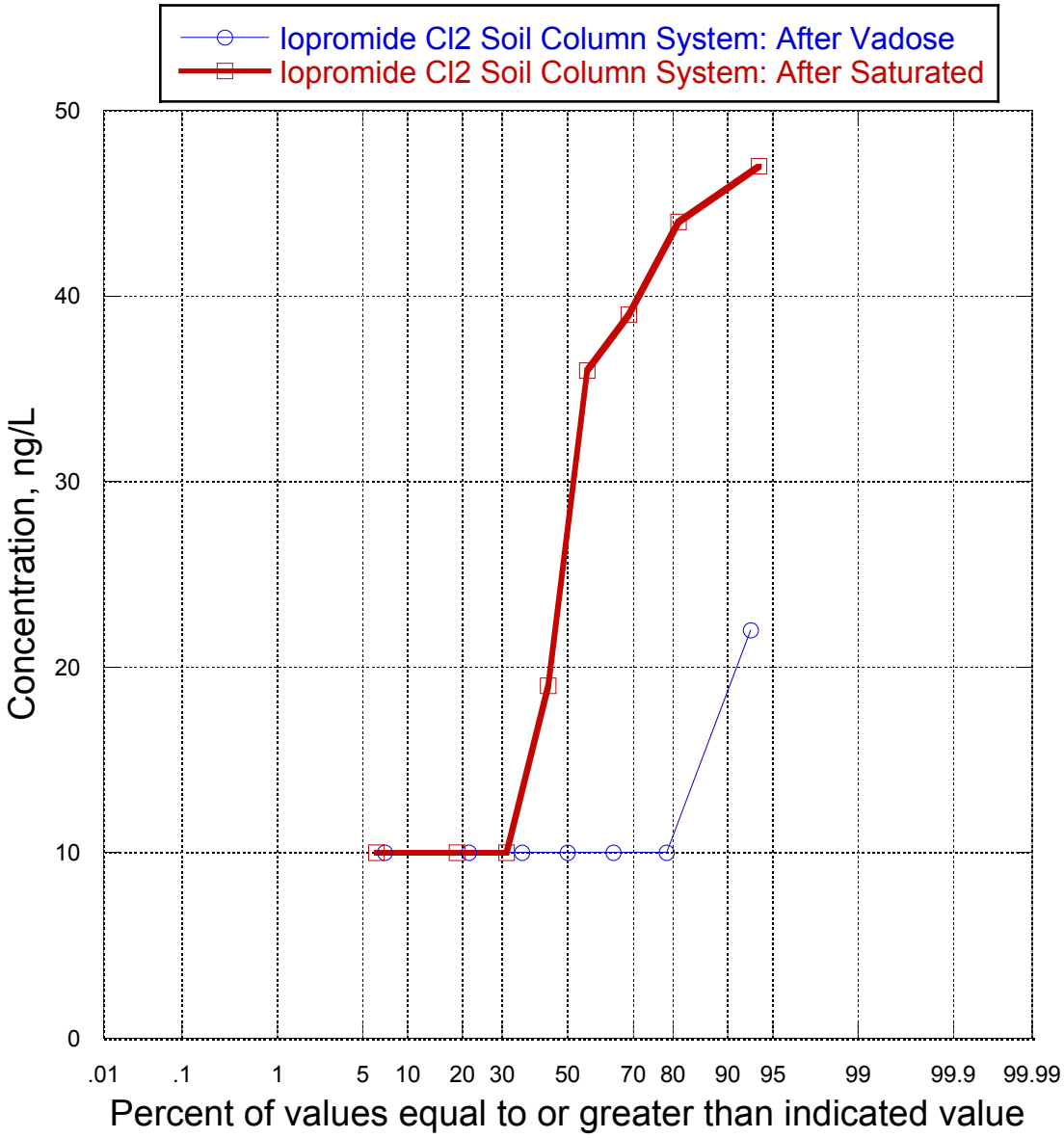

Figure G.8. Probability plot of iopromide concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Nonylphenol diethoxylate

Nonylphenol diethoxylate increased through the saturated zone of both the Cl_2 and O_3 Soil Column Systems. See the explanation on biodegradation byproducts provided earlier for 4-nonylphenol (tech mix).

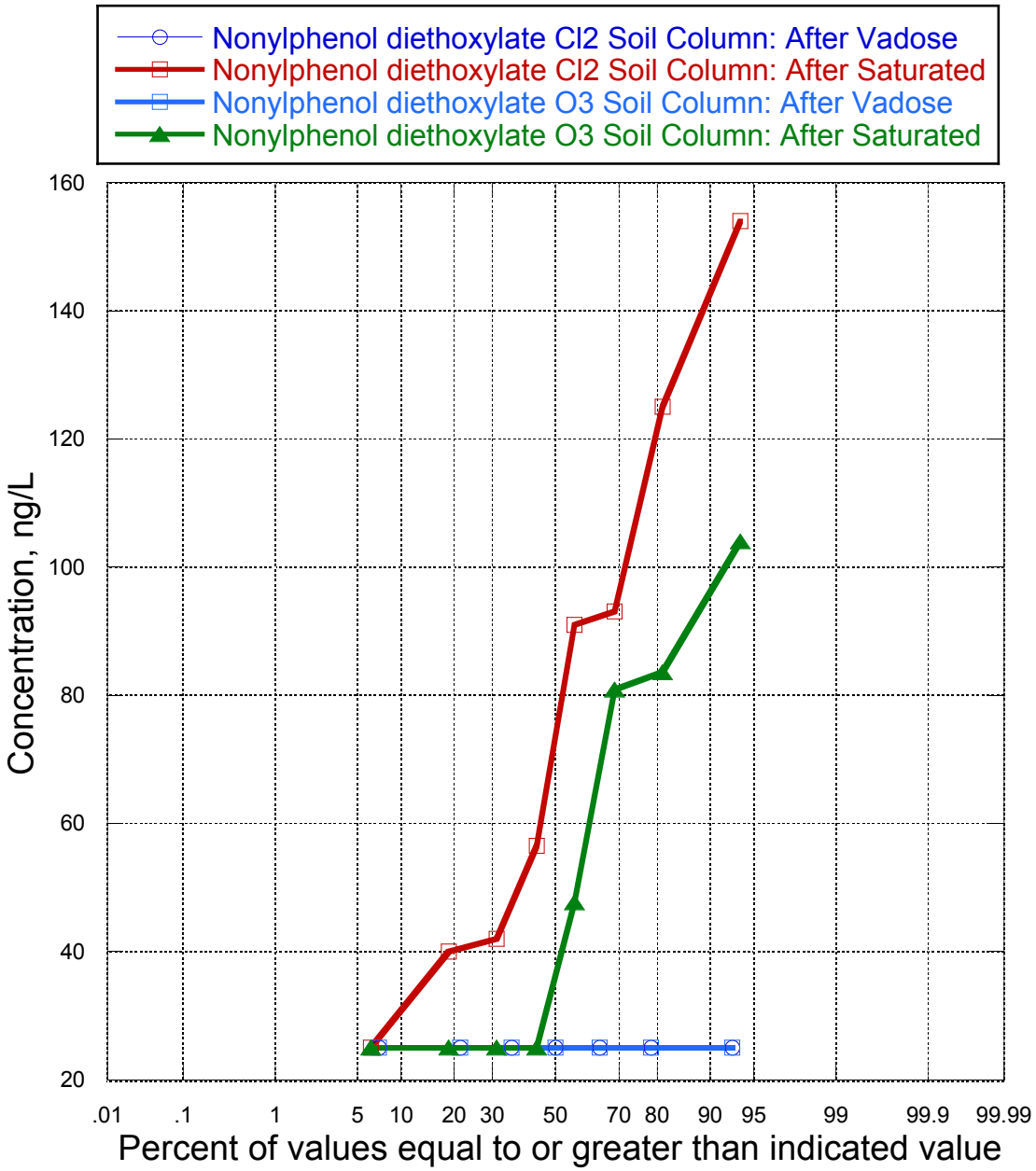

Figure G.9. Probability plot of nonylphenol diethoxylate concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Nonylphenol monoethoxylate

Nonylphenol monoethoxylate increased through the saturated zone of both the Cl_2 and O_3 Soil Column Systems. See the explanation on biodegradation byproducts provided earlier for 4-nonylphenol (tech mix).

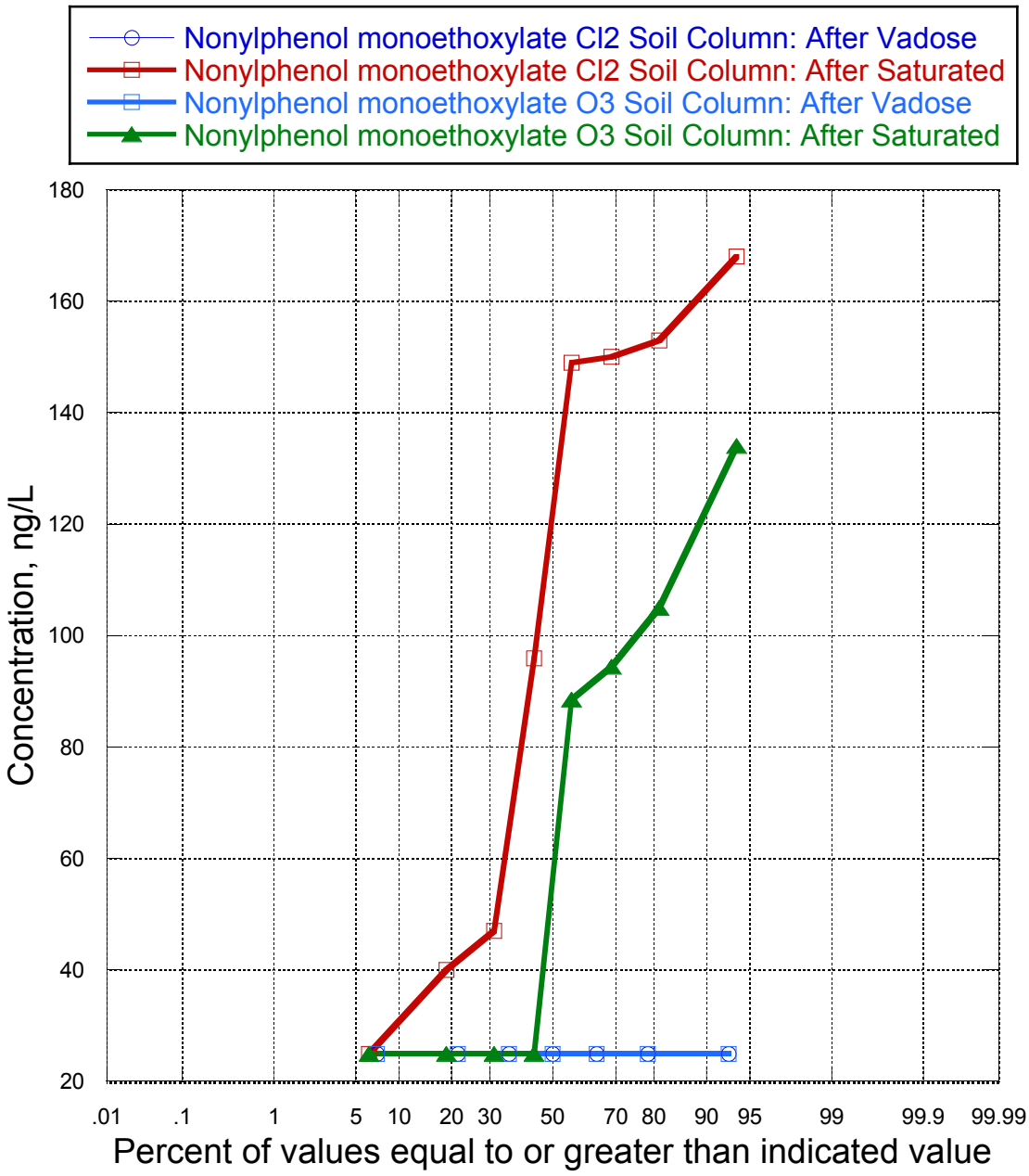

Figure G.10. Probability plot of nonylphenol monoethoxylate concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Octylphenol monoethoxylate

Octylphenol monoethoxylate increased through the saturated zone of both the Cl_2 and O_3 Soil Column Systems. See the explanation on biodegradation byproducts provided earlier for 4-nonylphenol (tech mix).

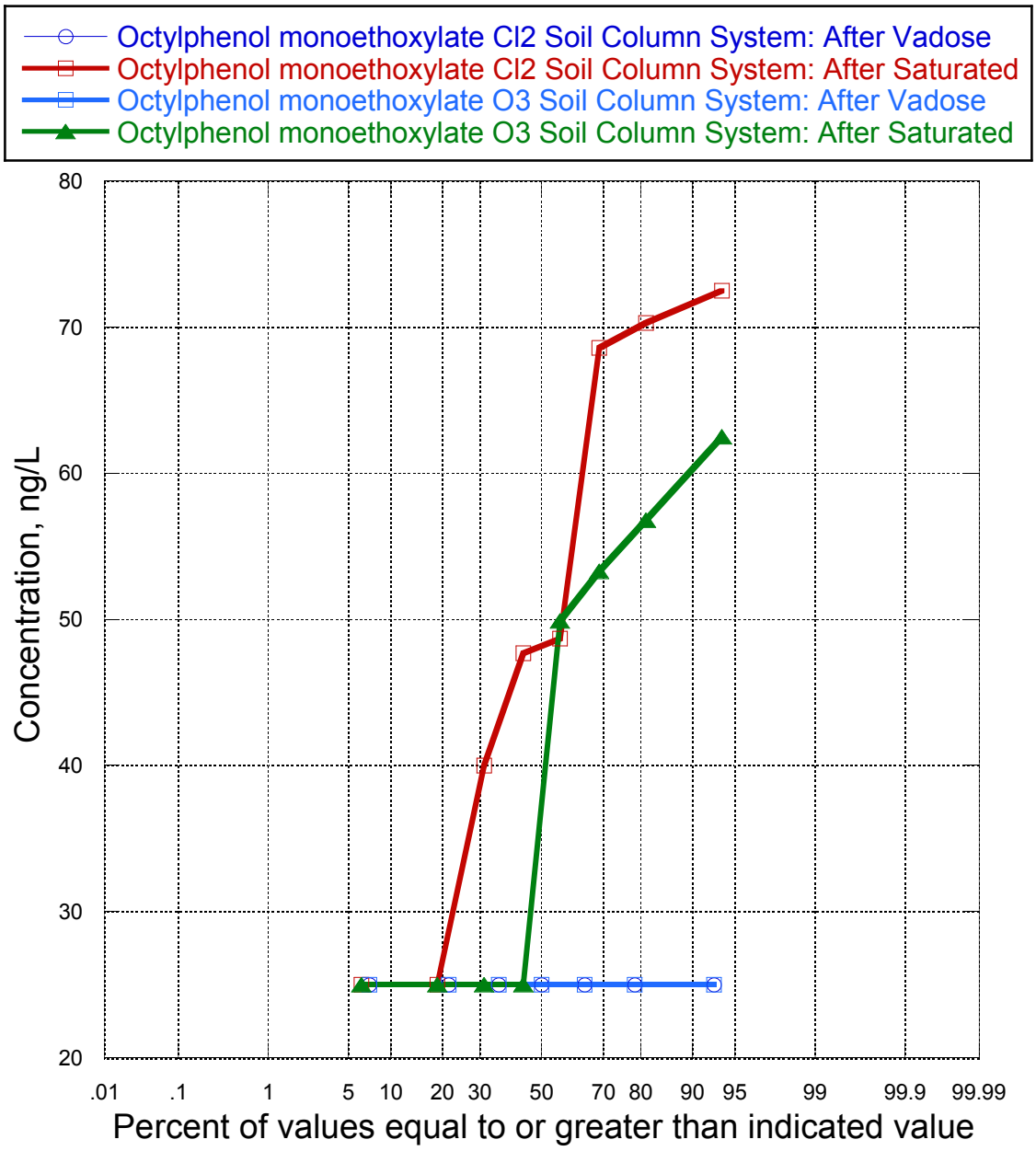

Figure G.11. Probability plot of octylphenol monoethoxylate concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

TCPP

TCPP seemingly increased through the saturated column in the Cl_2 Soil Column System. It is possible that TCPP was weakly absorbed to the soil and leached out over time. Drewes et al. (2011) also noticed increases on multiple occasions during their SAT testing.

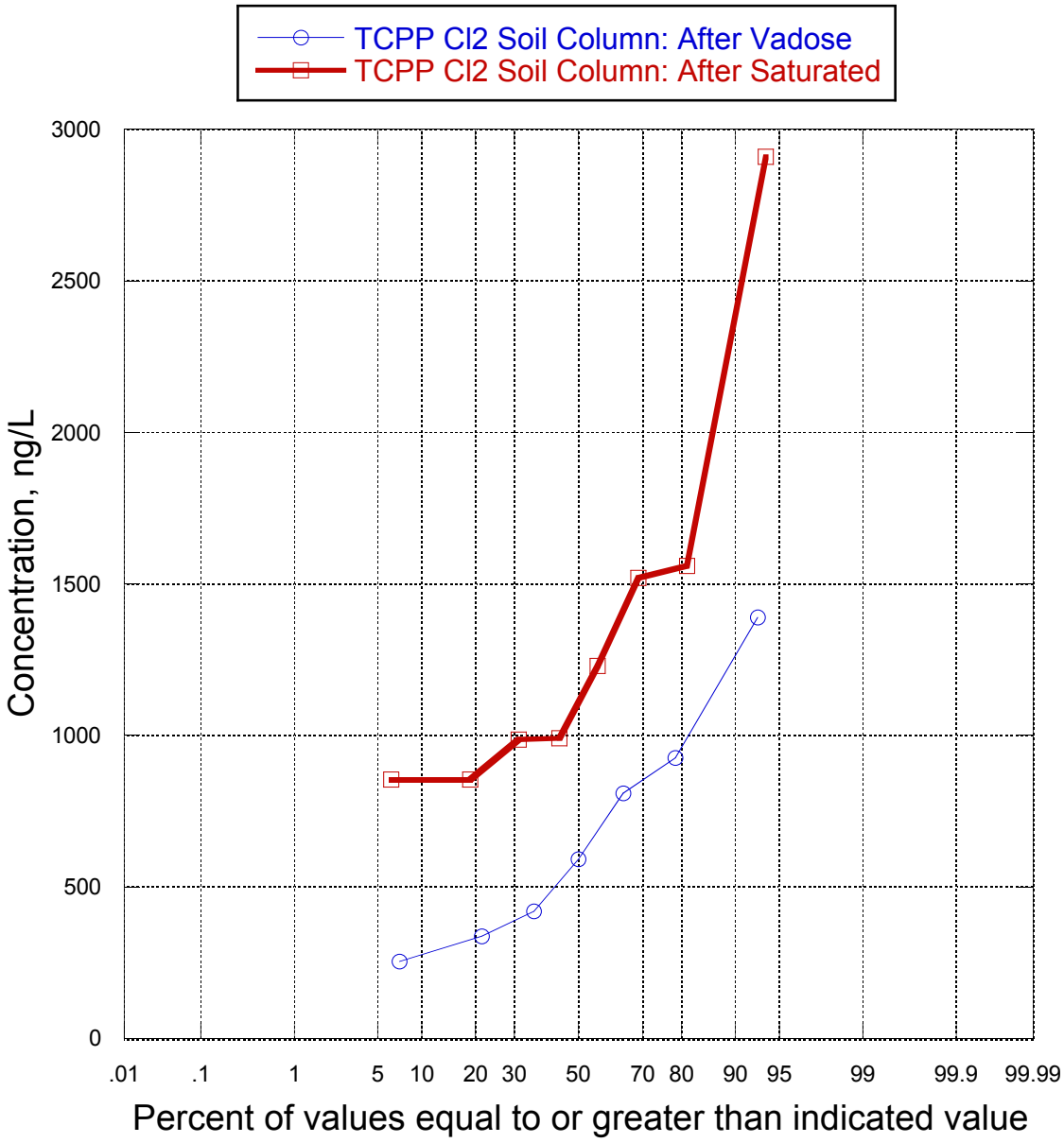

Figure G.12. Probability plot of TCPP concentrations for situations in which the concentrations at the two sampling locations being compared were statistically significant.

Appendix H

Raw Data from the Microbe Challenge Phase

Table H.1. TOC, UVT, and Microbe Concentrations during the Microbe Challenge Phase

Days into Phase 3	Cl_2 Column #1 Effluent			Cl_2 Column #2 Effluent			
	TOC, mg/L	UVT, %	Total Coliform, cfu/100 mL	TOC, mg/L	UVT, %	MS-2 Virus, pfu/mL	*Crypto*, oocysts/L
4		68	<20	3	85	<30	
5	5.04	84		3.03	86		
6	4.15	85	<20	3.33	86	<30	
7			<20			<30	
8							
9	5.92	83		2.99	86		
10	5.48	83	<20	2.97	85	<30	
11	4.77	84	<20	3.87	85	<30	
12	4.87	83	<20	3.03	86	<30	
13			<20			<30	<0.2
14							
15	4.57	83		2.98	85		
16	4.49	83	<20	3.39	83	<30	
17	4.56	84	<20	3.24	85	<30	
18	4.50	84	<20	3.14	86	<30	
19			<20			<30	
20							
21							
22	4.20	82		3.02	86		
23			<20			<30	<0.1
24	2.39						
25							
26	4.38	83		2.91	86		
27			<20			<30	
28							
29	4.15			2.95			
30			<20			<30	
31							
32	4.39	80		3.26	86		
33			<20			<30	
34							

Days into Phase 3	Cl_2 Column #1 Effluent			Cl_2 Column #2 Effluent			
	TOC, mg/L	UVT, %	Total Coliform, cfu/100 mL	TOC, mg/L	UVT, %	MS-2 Virus, pfu/mL	*Crypto*, oocysts/L
35							
36	4.41	82		3.18	85		
37			<20			<30	
38	4.45	82		3.08	86		<0.15
39	4.45	81	<20	3.33	84	<30	
40	4.93	81	<20			<30	
41							
42			<20			<30	
43							
44	4.72	82		3.00	86		
45			<20			<30	
46	4.56	81		2.98	85		<0.13
47							
48			<20			<30	
49							
50	4.31	82		3.05	86		
51							
52			<20			<30	
53	4.84	84		3.66	82		
54	4.69	83	<20			<30	
55							
56			<20			<30	
57	4.92	83		3.04	85		
58							
59	3.85	82	<20	3.05	85	<30	<0.2
60	4.42	83		3.06	85		
61							
62			<20			<30	
63							
64	4.40	84		3.30	87		
65							
66			<20			<30	
67	4.44	81		3.17	83		<0.09

Days into Phase 3	Cl_2 Column #1 Effluent			Cl_2 Column #2 Effluent			
	TOC, mg/L	UVT, %	Total Coliform, cfu/100 mL	TOC, mg/L	UVT, %	MS-2 Virus, pfu/mL	*Crypto*, oocysts/L
68			<20			<30	
69							
70							
71	5.36	82		3.36	85		
72			<20			<30	
73							
74	5.45	79		3.74	85		
75			<20			<30	
76							
77							
78	5.63	79		3.78	84		
79			<20			<30	
80	4.39	80		3.07	84		
81			<20			<30	
82							
83							
84							
85	5.15	79		4.29	85		
86			<20			<30	
87							
88	5.71	79		4.10	84		




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