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Septic systems as hot-spots of pollutants in the environment: Fate and mass balance of micropollutants in septic drainfields

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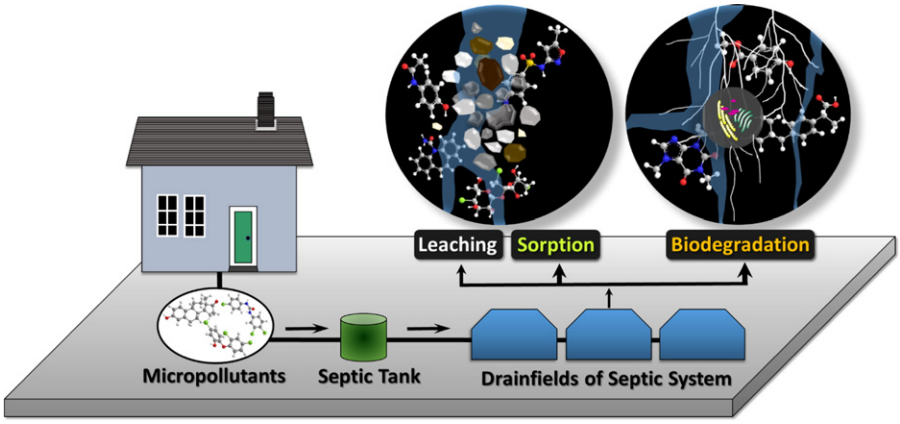
HIGHLIGHTS

- Fate and mass balance of 17 micropollutants were investigated in septic systems.
- Approximately 85% of effluent applied hormones and PPCPs attenuated in drainfield.
- Sucralose was a better indicator of septic effluent than caffeine in the leachate.
- Sorption and microbial degradation were key mechanisms of micropollutants removal.
- Vadose zone processes limited leaching of micropollutants from the drainfields.

GRAPHICAL ABSTRACT



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ABSTRACT

Septic systems, a common type of onsite wastewater treatment systems, can be an important source of micropollutants in the environment. We investigated the fate and mass balance of 17 micropollutants, including wastewater markers, hormones, pharmaceuticals and personal care products (PPCPs) in the drainfield of a septic system. Drainfields were replicated in lysimeters (1.5 m length, 0.9 m width, 0.9 m height) and managed similar to the field practice. In each lysimeter, a drip line dispersed 9 L of septic tank effluent (STE) per day (equivalent to 32.29 L/m^2 per day). Fourteen micropollutants in the STE and 12 in the leachate from drainfields were detected over eight months. Concentrations of most micropollutants in the leachate were low (<200 ng/L) when compared to STE because >85% of the added micropollutants except for sucralose were attenuated in the drainfield. We discovered that sorption was the key mechanism for retention of carbamazepine and partially for sulfamethoxazole, whereas microbial degradation likely attenuated acetaminophen in the drainfield. This data suggests that sorption and microbial degradation limited transport of micropollutants from the drainfields. However, the leaching of small amounts of micropollutants indicate that septic systems are hot-spots of micropollutants in the environment and a better understanding of micropollutants in septic systems is needed to protect groundwater quality.

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1. Introduction

Many micropollutants such as pharmaceuticals and personal care products (PPCPs) are used for personal health, hygiene, and cleaning in daily life. These micropollutants are released in domestic wastewater and frequently detected in surface waters (Kolpin et al., 2002; Lindqvist et al., 2005; Singh et al., 2010), groundwater (Fram and Belitz, 2011; Gottschall et al., 2012; Katz et al., 2010; Schaider et al., 2014), drinking water (Benotti et al., 2009; Bruce et al., 2010), soils and sediments (González-Naranjo et al., 2013; Williams and McLain, 2012; Yang et al., 2015). Many micropollutants including PPCPs are endocrine disrupting compounds and have been linked to reproductive effects in aquatic organisms (Brian et al., 2007; Miyagawa et al., 2014). Some assessments conclude that human health effects are unlikely at current exposure levels; however, there are concerns for possible cumulative effects over long exposure periods (Letzel et al., 2009; Saravanan and Ramesh, 2013).

In rural and urbanizing watersheds, wastewater is treated and disposed onsite using septic systems, a common type of onsite wastewater treatment systems. In a septic system, a septic tank receives effluent from a home or business, which is then dispersed in the soil via a network of subsurface soil infiltration trenches (i.e., soil treatment units, or drainfields) for further treatment before effluent reaches groundwater. Other types of dispersal units include mound soil systems, low-pressure pipe systems, and constructed wetlands (U.S. Environmental Protection Agency, 2002). Mound systems are used in areas with shallow groundwater table such as Florida and other coastal areas in the United States as they provide an additional 0.6–0.9 m of elevated disposal field on top of the natural soil (Gill et al., 2007). Transformation and removal of PPCPs and hormones in septic systems vary with compound properties and treatment conditions (Conn et al., 2006). Several studies have suggested that aerobic conditions promote removal of many micropollutants (Carrara et al., 2008; Swartz et al., 2006). By analyzing effluent in a septic system and pore water below the infiltrative surface, Conn et al. (2010a) observed >90% removal of caffeine, triclosan, ethylenediaminetetraacetic acid, nitrilotriacetic acid, and 4-nonylphenolmonoethoxycarboxylate and <50% removal of 4-nonylphenol.

Septic systems can be a source of micropollutants in the groundwater (Del Rosario et al., 2014; Godfrey et al., 2007; Katz et al., 2010; Phillips et al., 2015; Swartz et al., 2006; Verstraeten et al., 2005). Swartz et al. (2006) found caffeine, estrogens, and other pharmaceutically active compounds in groundwater in Cape Cod, Massachusetts, USA. A study in Nebraska reported caffeine, antibiotics, and analgesics in groundwater from 12 of the 19 domestic wells (Verstraeten et al., 2005). In the northeastern United States, total concentrations of PPCPs and plasticizers were up to 20 μg/L in groundwater influenced by septic systems (Phillips et al., 2015). These studies suggest that micropollutants are present in groundwater impacted by septic systems.

In the United States, 25% of households and 33% of new developments use septic systems (U.S. Environmental Protection Agency, 2005). In Florida, it is estimated that over 2.67 million septic systems discharge approximately 1613 million liters of effluent per day (Meeroff et al., 2008). In areas with septic systems, groundwater is often the drinking water source for the local population. Knowledge of the fate, transport, and attenuation mechanisms of micropollutants in septic systems drainfields is needed to protect human and environmental health. Our previous study found acetaminophen, caffeine, and carbamazepine in stream sediments of an urban river due to the presence of septic systems in the watershed (Yang et al., 2015). In this study, our objectives were to (i) investigate the leaching of micropollutants, including markers of wastewater (i.e., sucralose and caffeine), PPCPs, and hormones, (ii) determine the spatial distribution inside the drainfields, and (iii) determine the potential removal mechanisms of micropollutants in the drainfields of septic systems. To our knowledge, this study is a first attempt to determine the spatial distribution and unravel attenuation mechanisms of micropollutants inside the drainfield of a septic system.

2. Materials and methods

2.1. Drainfields construction

Septic system drainfields were constructed at the University of Florida-Gulf Coast Research and Education Center (GCREC) in Wimauma, FL (latitude 27°45′29.51″ N, longitude 82°13′36.4″ W). Natural soil used in the drainfields was collected from the A horizon (Ap: 0–17 cm) and partial A/E horizons (18–30 cm) of a zolfo fine sand series (sandy siliceous, hyperthermic Oxyaquic Alorthods). The study site soil is somewhat poorly drained and seasonal water table fluctuates from 60 to 152 cm below the soil surface.

Three replicate lysimeters (1.5 m length, 0.9 m width, 0.9 m height, 1:1 side slope) were constructed using pressure treated wood to mimic septic system drainfields (Supplementary Information (SI) Fig. S1). The detailed methodology of lysimeters construction can be found in De and Toor (2015). In brief, a hole was drilled at the bottom of each lysimeter to which a floor drain strainer and a pipe was attached to collect leachate in a covered 19-L glass bottle. A section of cheesecloth and mesh fabric was placed on top of the hole to prevent any material loss. Each drainfield was packed to 7.6 cm depth with a mixture of sand and pea gravel to facilitate free drainage, followed by 30.5 cm of natural soil (loamy sand: 87% sand, 9% silt, 4% clay), and 30.5 cm of sand (commercial builder sand; 100% sand). Field bulk density of 1.6 g/cm^3 for the sand layer and 1.5 g/cm^3 for the soil layer were achieved by packing a known mass of soil and sand in each lysimeter in 5 cm increments. A drip line with three emitters was placed on top of the sand layer to disperse 9 L of septic tank effluent (STE) per lysimeter, which is equivalent to the maximum allowable rate of 32.29 L/m^2/day for Florida's sandy soils in 6-doses at 4-h intervals. The source of STE was the wastewater from the graduate student housing and business units of the GCREC (daily employee load of ~50 people). A final 15.3 cm layer of sand was placed on top of the drip line before planting St Augustine grass (*Stenotaphrum secundatum*) to mimic a residential drainfield. For the initial establishment of the grass, water was applied three times per week (7.2 L per drainfield) with a sprayer for 3–4 weeks and leachate (as background water) was collected to determine background micropollutant concentrations. The septic drainfields used in this study were not covered to prevent rain and were free standing and thus may have slightly different temperature pattern than drainfields surrounded by soil (SI Fig. S1). However, the study site has a subtropical climate with an average monthly annual air temperature ranging from 15.3 to 26.6 °C and daily extremes ranging from 0.7 to 35 °C in 2013 (Florida Automated Weather Network Website, 2013).

2.2. Sample collection

A total of 40 leaching events were conducted from January to August 2013. A leachate sample was collected every 24 h after STE application from each lysimeter for the first 15 days (n = 15). For the next 25 weeks, a weekly flow-weighted composite sample (n = 25) was made by compositing daily leachate samples collected during each week. Septic tank effluent samples (n = 40) were collected at the time of leachate collection throughout the 8-months study period. All STE and leachate samples were collected in 1-L amber glass bottles, preserved with 1 g of sodium azide and kept dark and refrigerated at 4 °C during the composite collection period each week. The composite sample holding time was less than 24 h before analysis. Conventional water quality parameters including pH and EC (using HI 9828, Hanna Instruments Inc., RI, USA) and chloride (using AQ2$_+$, Seal Analytical, Mequon, WI, USA) were determined following standard methods (U.S. Environmental Protection Agency, 1983). Soil and commercial sand

samples were collected prior to packing lysimeters and after one year of STE application (n = 36 per lysimeter; SI Fig. S2). Samples were passed through a sieve with 2 mm i.d. pore size, placed in glass amber jars, and stored at −80 °C until analysis.

2.3. Analysis of micropollutants

The analyses included two wastewater markers, four pharmaceuticals, three hormones, one plasticizer, and seven personal care products (SI Table S1). All standards and reagents were purchased in the highest purity available (see SI). Water samples were filtered through two pieces of glass microfiber filters (0.7-μm, Whatman GFF 1825-047; 2.7-μm, Whatman GF/D 1823-047, Whatman Inc. Piscatway, NJ, USA). The cleanup procedure was based on method from Vanderford and Snyder (2006). The target compounds were extracted from aqueous samples onto Oasis hydrophilic-lipophilic balance (HLB) solid phase extraction (SPE) cartridges (6 cm^3, 200 mg; Waters, Milford, MA). Prior to SPE extraction, filtered water samples were spiked with a solution of isotopically labeled standards that contained a stable isotope of analyte ($^{13}C_2$-acetaminophen-$^{15}N_1$, bisphenol-A-d_{16}, carbamazepine-d_{10}, and 17β-estradiol-d_4, caffeine-d_9, estrone-d_4, 17α-ethinyl estradiol-d_4, ibuprogen-d_3, sulfamethoxazole-d_4). Cartridges were sequentially preconditioned with 5 mL of methyl *tert*-butyl ether (MTBE), 5 mL of methanol, and 5 mL of reagent water. After the conditioning step, 1 L of STE and leachate samples were then loaded onto the cartridges at a flow rate of 10 mL/min, after which the cartridges were rinsed with 5 mL of reagent water and then dried under vacuum for 30 min. The SPE cartridges were eluted with 5 mL of methanol followed by 5 mL of 10/90 (v/v) methanol/MTBE. The eluents were evaporated to less than 0.5 mL using a RapidVap system (Model 79000-02, Labconco Co. Kansas City, MO, USA) and brought up to a final volume of 0.5 mL using methanol prior to analysis with liquid chromatography followed by mass spectrometry.

Micropollutants were extracted from the soils collected from drainfields using an ultrasonic cleaner (Mettler Electronics Corp, Anaheim, CA, USA). The extraction procedure followed modified methods by Wu et al. (2011) and Vanderford and Snyder (2006). Prior to extraction, filtered water samples and soil samples were spiked with a solution of isotopically labeled standards that contained a stable isotope of analyte to assess compound recovery and accuracy. Briefly, soil samples were weighted into a 40-mL amber glass vial and ultrasonically extracted by 20 mL of methylene chloride: acetone (1:1, v/v) twice and cleaned up with method described in Vanderford and Snyder (2006) as described above. The average percent recoveries for all selected micropollutants ranged from 62% (sulfamethoxazole) to 115% (estrone) from spiked water, STE, and leachate and 75% (17α-ethinyl estradiol) to 105% (17β-estradiol) from soil samples; these values are within the reported literature values. The reporting limits ranged from 0.8 to 127 ng/L for selected micropollutants. The details of analytical methods, instruments, and analyses are provided in SI (Tables S2–S5).

2.4. Microbial degradation of micropollutants

Degradation of a mixture containing micropollutants was determined in time series batch incubations of drainfield soil/sand samples in order to assess the potential microbial degradation of micropollutants (see SI for details). In brief, sample incubations consisted of 5 g of sterilized (autoclaved for 30 min at 121 °C and 20 psi for twice) or unsterilized soil/sand in 40-mL amber glass vials and divided into two groups: (i) non-spiked soil/sand, and (ii) spiked soil/sand with micropollutants to an initial concentration of 25 ng/g. To maintain aerobic conditions, sample vials were capped loose with Teflon-lined caps and air was added by vortexing for 2 min every day. For anaerobic conditions, the vials containing spiked soil/sand were purged with N_2 for 5 min and capped tight until extraction. The moisture content (~10%) was kept constant by adding water every 2–3 days according to the weight loss during the incubation period. Three replicates samples for each treatment were incubated either under aerobic or anaerobic conditions in the dark at 22 °C for 20 days and collected at each time point and immediately frozen (−80 °C) until analysis using an ultrasonic cleaner as described above.

2.5. Quality assurance and quality control

Laboratory quality assurance and quality control (QA/QC) included spikes, field and laboratory blanks, and stable isotope standards. A summary of QA/QC data is shown in SI (SI Table S5). Fortified laboratory spikes and blanks were analyzed with each set of 10 samples. No analytes of interest were detected in blanks. Values below detection limits were set at 50% of reporting limits when calculating mean and median concentrations.

2.6. Statistical analysis

The non-parametric Wilcoxon rank test was used to examine the significance ($p < 0.05$) of measured micropollutants between STE and leachate samples. Principal component analysis (PCA) was performed for pattern recognition and identification of correlations between various measured micropollutants. Correlations between measured parameters were tested using the non-parametric Spearman's rank sum correlation test. Correlations were considered significant for $p < 0.05$. All statistical analysis was performed using the JMP statistical software package (JMP Pro 12, SAS Institute).

3. Results and discussion

3.1. Water quality parameters

In the STE, the mean values of pH, EC, and Cl were 7.2 ± 0.25, 1.2 ± 0.13 dS/m, and 133 ± 20.7 mg/L, which decreased in the leachate to 6.9 ± 0.24, 0.8 ± 0.24 dS/m, and 95 ± 47.1 mg/L, respectively due to the percolation of STE in the drainfields containing sand and soil layers (SI Table S6). Further, dilution with the infiltrating rainfall during the wet season (June–August) likely reduced Cl and EC in the leachate.

3.2. Micropollutants in the septic tank effluent

No micropollutants were found in any background water (n = 6) used in the beginning of the study to facilitate grass establishment. Fourteen of the 17 target compounds were found in the STE (n = 34), comprising 10 PPCPs (acetaminophen, carbamazepine, ibuprofen, sulfamethoxazole, triclosan, triclocarban, butyl paraben, ethyl paraben, methyl paraben, and propyl paraben), a hormone (estrone), a plasticizer (bisphenol-A), a sweetener (sucralose), and a stimulant (caffeine) (Fig. 1 and SI Table S6). Hormones (17α-ethinyl estradiol and 17β-estradiol) and preservative (heptyl paraben) were not found in any STE samples during the sampling period. Sucralose, caffeine, ibuprofen, triclosan, triclocarban, methyl paraben, and propyl paraben were found in all STE samples. Acetaminophen, estrone, ethyl paraben, and bisphenol-A were found in more than 85% of STE samples and carbamazepine, sulfamethoxazole, and butyl paraben were found in less than 60% of STE samples.

Median monthly total concentrations of detected micropollutants in the STE varied from 18,000 to 28,000 ng/L (SI Fig. S3). The results of PCA showed a broad gradient in concentration and distribution of micropollutants in STE, suggesting that the variable sources and usages of micropollutants (SI Fig. S4). Among all detected compounds, sucralose, caffeine, and ibuprofen were most abundant in the STE, with median concentrations of 3200, 8750, and 7370 ng/L, respectively (Fig. 1 and SI Table S6). The high concentrations and detection frequencies of some micropollutants can be related to the use and discharge of compounds. For example, the artificial sweetener (sucralose) has been used as a

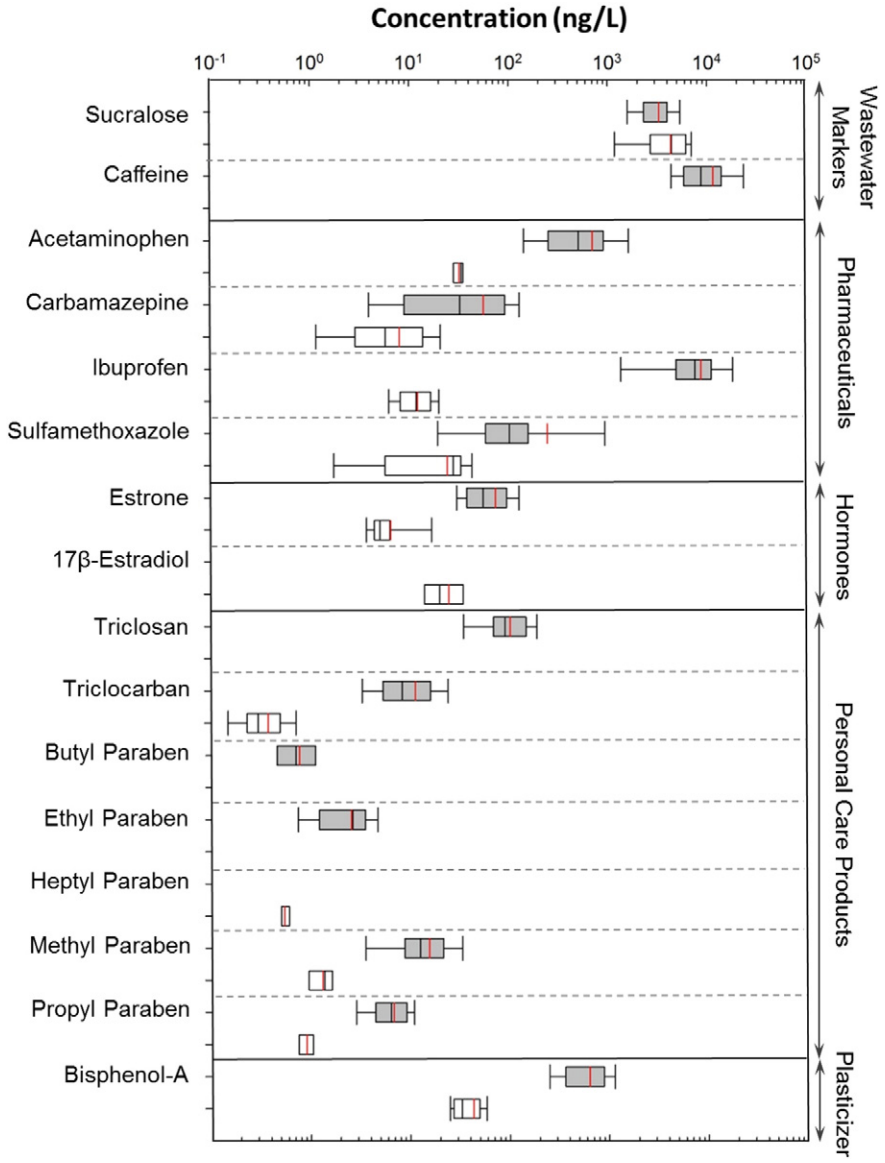

Fig. 1. Concentrations of micropollutants in the septic tank effluent (gray boxplots; n = 34) and leachate (white boxplots; n = 120) from septic system drainfields from January to August, 2013. Boxes indicate the median, mean (red), and 25th and 75th percentile values with whiskers extending to the 5th and 95th percentiles of original data points.

marker of wastewater as approximately 85% of sucralose is not absorbed in the body and the remainder 15% that is absorbed is excreted unchanged in the urine (Grotz and Munro, 2009). Sucralose was also found to not or poorly degrade during wastewater treatment and was detected in municipal wastewater effluents, surface waters, and potable water at concentrations up to μg/L level (Mawhinney et al., 2011; Oppenheimer et al., 2011; Scheurer et al., 2009; Tran et al., 2013). Likewise, caffeine in wastewater is contributed by pharmaceuticals, soft drinks, and coffee consumption. High concentrations of ibuprofen found in our STE samples can be related to its widespread use as a pain reliever and its over-the-counter availability.

The detection frequencies and concentration ranges of many micropollutants found in the STE including acetaminophen, carbamazepine, caffeine, ibuprofen, sulfamethoxazole, triclosan, estrone, and bisphenol-A are in agreement with previous literature for STE and WWTPs influent (Carrara et al., 2008; Conn et al., 2010a; Conn et al., 2010b; Du et al., 2014; Gao et al., 2012; Godfrey et al., 2007; Hinkle et al., 2005; Matamoros et al., 2009; Reyes-Contreras et al., 2012; Stanford and Weinberg, 2010; Subedi et al., 2014; Teerlink et al., 2012a; Yu et al., 2013). Overall, median concentrations of pharmaceuticals in the STE samples ranged from 32 ng/L (carbamazepine) to 7370 ng/L (ibuprofen), whereas median concentrations of personal care products ranged from 0.7 ng/L (butyl paraben) to 91 ng/L (triclosan). Median concentrations of estrone and bisphenol-A were 55 and 638 ng/L, respectively.

3.3. Micropollutants in the leachate

In the leachate samples (n = 120), 12 target compounds, including sucralose, eight PPCPs (acetaminophen, carbamazepine, ibuprofen, sulfamethoxazole, triclosan, heptyl paraben, methyl paraben, and propyl paraben), two hormones (estrone and 17β-estradiol), and a plasticizer (bisphenol-A), were found (Fig. 1 and SI Table S6). Sucralose has been determined to be a superior indicator compound in wastewater influenced surface waters due to its recalcitrance and low sorption in the soils (Mawhinney et al., 2011). In the present study, sucralose was found in more than 80% of leachate samples with a median concentration of 4100 ng/L. Ibuprofen and triclocarban were found in more than 65% of leachate samples, with median concentrations of 9 and 0.6 ng/L, respectively. Bisphenol-A and other compounds in the leachate were detected <50%, with median concentrations below 33 ng/L.

Median monthly total concentrations of hormones and PPCPs (not including sucralose) in the leachate varied from 30 to 86 ng/L. In general, bisphenol-A was the most abundant compound followed by ibuprofen in the leachate (Fig. S3). Concentrations of sulfamethoxazole in the leachate were greater in the beginning of study, which then decreased. Principal component analysis based on the correlation matrix was used to assess the characteristics of leaching behavior among micropollutants and conventional parameters (SI Fig. S4). The results of PCA indicated no statistically significant correlation between measured parameters (e.g., rainfall amount) and concentrations of individual micropollutants in the leachate. A strong positive correlation ($\rho = 0.70$, $p < 0.001$) between concentrations of sucralose and Cl in the leachate was observed based on PCA. However, concentrations of hormones and other PPCPs were not related with sucralose or Cl likely due to their physicochemical properties, such as water solubility and functional groups capable of interacting with binding sites in soil. For example, hormones are generally less polar and have low water solubility at environmentally relevant pH (6.4–7.2 in this study), which can limit leaching as compared to acidic pharmaceuticals (characterized by low pK_a values). The solubility of acidic compounds such as ibuprofen and sulfamethoxazole is also low but they are mainly present in ionized forms dissolved in the aqueous phase in a pH environment higher than their pK_a, which increases their potential to leach.

Although 17β-estradiol was not found in the STE, it was present in the leachate, suggesting potential accumulation in the drainfields due to the repeated application of STE containing low levels of 17β-estradiol and/or transformation of estrone to 17β-estradiol. Further, estrogen deconjugation may occur in the drainfields due to the presence of fecal microorganisms that are capable of hydrolyzing conjugates via glucuronidase and sulfatase enzymes to unconjugated forms (Belfroid et al., 1999; Ternes et al., 1999). Concentrations of carbamazepine, ibuprofen, estrone, triclocarban, methylparaben, propyl paraben, and bisphenol-A were significantly ($p < 0.05$) lower in the leachate than the STE. The absence and low concentrations of some micropollutants in the leachate could be due to their low concentrations in the STE, (bio)transformation, and/or sorption in the drainfields. The potential removal mechanisms of micropollutants from the drainfields are discussed in the later section. Overall, the leaching potential of investigated micropollutants from the drainfield was generally low, except for sucralose. The absence and low levels of micropollutants in the leachate suggests that only a small fraction of the selected compounds leached through the 60-cm deep drainfields.

3.4. Spatial distribution of selected micropollutants in the drainfields

In order to assess whether any accumulation/sorption of micropollutants occurred, drainfields were destructively sampled ($n = 36$ per lysimeter) to collect sand and soil from different locations (SI Fig. S2). To our knowledge, this is the first study to determine the spatial distribution of micropollutants inside the septic drainfields. No residual levels of wastewater markers, hormones, or PPCPs were found in the background soil samples collected prior to the STE application. Of 17 targeted micropollutants, only three micropollutants, including acetaminophen, carbamazepine, and sulfamethoxazole, were found in the sand and soil samples in the drainfields. Concentrations of acetaminophen, carbamazepine, and sulfamethoxazole in drainfield after one year of STE application were 250–2020 pg/g, 77–594 pg/g, and 134–5100 pg/g, respectively (Fig. 2). The presence of these compounds in the sand and soil layers of drainfield suggests that sorption was the likely attenuation mechanism for these micropollutants.

Previous studies have shown that acetaminophen is reduced via fast biodegradation in WWTPs (Radjenović et al., 2009). However, Jones et al. (2006) indicated that sorption could be a possible elimination pathway for acetaminophen in a WWTP. In this study, acetaminophen was found at the bottom of the drainfield in 8% of drainfield sand and soil samples (Fig. 2), whereas carbamazepine was found in 44% of drainfield sand and soil samples, with most accumulation occurring close to the STE dispersal drip pipe and on the side of the drainfield. Elevated concentrations of sulfamethoxazole were also observed in 50% of samples close to the drip pipe. The high persistence of carbamazepine and sulfamethoxazole has been observed in WWTPs (Castiglioni et al., 2006; Göbel et al., 2007). Carbamazepine has an organic base with $pK_a = 13.9$ (SI Table S1), and hence is present predominantly as cationic specie at the experimental pH values (6.4–7.2). The high pK_a value and moderate log K_{ow} (2.25–2.45) retain carbamazepine onto solid phases (Carballa et al., 2008; Martín et al., 2012; Radjenović et al., 2009). The spatial distribution of carbamazepine inside the drainfields suggests some movement of this relatively polar compound through the vadose zone. The cationic form of sulfamethoxazole sorb more close to the environmental pH ≥ pK_{a1} (1.83) and the anionic species dominate when pH ≥ pK_{a2} (5.57) resulting in low sorption (Srinivasan et al., 2013). At experimental soil pH (6.4–7.2), sulfamethoxazole is present mostly in the anionic form (SI Table S1); therefore, sulfamethoxazole does not sorb to negatively charged soil particles. This is supported by greater concentrations of sulfamethoxazole in the leachate in the beginning of study (SI Fig. S3). Overall, the amounts of acetaminophen, carbamazepine, and sulfamethoxazole in the drainfields showed the potential for accumulation over time. To further understand the role of sorption and microbial degradation in determining the fate of these three micropollutants found in the drainfields, a short-term microcosm experiment was conducted and the potential removal mechanisms of other micropollutants are described in the next section.

3.5. Removal of micropollutants in the drainfields

Mass loading (ng) of individual micropollutants was determined by multiplying the STE amount (L) and concentrations (ng/L) of micropollutants. Strong positive correlations were found among the sum of leached micropollutant loads, leachate pore volume, and chemical effluent indicators (SI Fig. S5), suggesting that septic systems are hot-spots of micropollutants in the environment. We found that greater than 85% of the added hormones, PPCPs, and caffeine were attenuated within 60-cm deep drainfields (Fig. 3 and SI Fig. S6). The micropollutants mass found in the leachate ranged from 0 (caffeine) to 12% (methyl paraben) of the amount added with STE. The variability in micropollutant mass among three replicate drainfields was 0–100% for wastewater markers, <1–16% for pharmaceuticals, 0 ≤ 5% for hormones, 0–17% for personal care products, and 2 ≤ 5% for plasticizer (SI Fig. S6). No significant differences in micropollutant mass among three replicate drainfields were observed. The presence or absence of micropollutants in the drainfields provides some insights into the dominant factors controlling their transport. In general, the mechanisms and removal pathways of micropollutants are not completely understood in the literature. However, the retention and removal of micropollutants in the environment is expected to be largely the result of a combination of sorption and microbial degradation.

3.5.1. Markers of wastewater

The mass of sucralose in the leachate was similar to the mass applied in the STE (~100% recovered) likely due to the fact that sucralose is recalcitrant in the environment, which suggests that sucralose is a robust environmental tracer (Fig. 3). In contrast, caffeine was found in all STE samples but not detected in any leachate samples, likely due to degradation in the drainfields. This finding is consistent with previous studies that observed >99% removal of caffeine in drainfield of septic systems (Godfrey et al., 2007; Hinkle et al., 2005). The results of mass balance of two markers suggested that sucralose is more recalcitrant in the drainfields and thus is more suitable than caffeine as an indicator of septic systems impact in the groundwater.

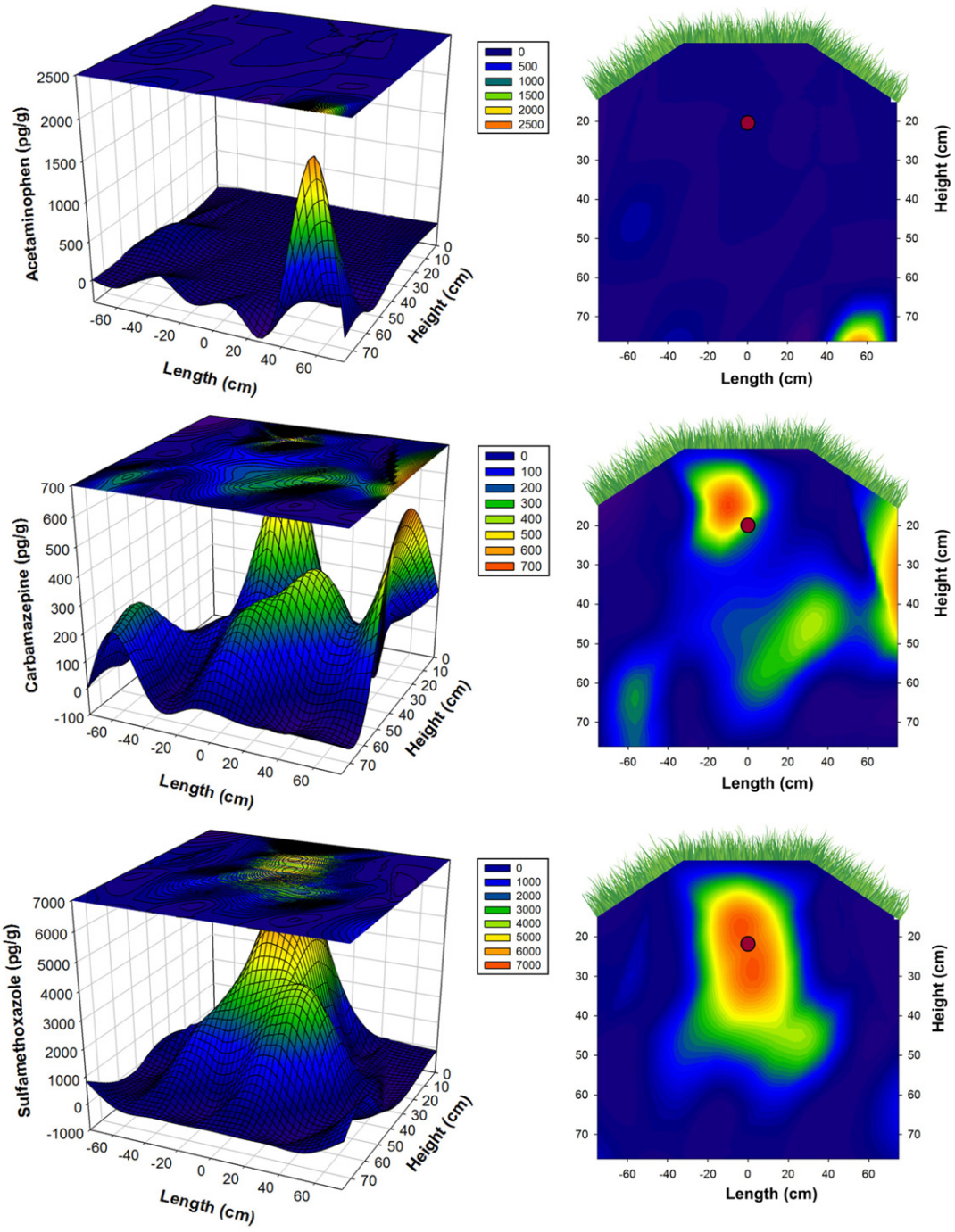

Fig. 2. Spatial distribution of micropollutants within septic system drainfields (36 locations). Drip pipe was installed at a height of 22 cm from the top (red dot). Negative numbers on *x* axis represent the left side of drainfield.

3.5.2. Pharmaceuticals, hormones, personal care products, and plasticizer

Although none of the previous research has investigated the fate and mass balance of micropollutants in the drainfields, the limited research comparing the concentrations from the inlet and outlet of septic systems has suggested the degradation and sorption of acetaminophen, carbamazepine, sulfamethoxazole, triclosan, and hormones in the soil absorption systems and aerobic sand filters (Conn et al., 2010b; Godfrey et al., 2007; Stanford and Weinberg, 2010). For example, in a conventional soil treatment unit of septic system, more than 90% removal of triclosan in 60-cm unsaturated sandy loam soil was reported (Conn et al., 2010b). Godfrey et al. (2007) observed that the percolation through a 2-m-deep sand vadose zone lowered concentrations by more than 75% for acetaminophen, carbamazepine, and sulfamethoxazole in septic systems. The results of our incubation study showed greater microbial degradation of acetaminophen under aerobic than anaerobic conditions in both sand and soil samples, whereas no sorption of acetaminophen (sterile) was observed after 1 day of incubation (Fig. 4). These observations suggest that transport of acetaminophen through the drainfield occurred (as supported by our leaching data) and microbial degradation attenuated a major part of acetaminophen in the drainfields. Further, the acetaminophen concentration decreased by >80% within 3 days in the upper sand layer and within 5 days in the bottom soil layer in the drainfields (Fig. 4). De and Toor (2015) found that due to the textural differences in two layers of the drainfield, water

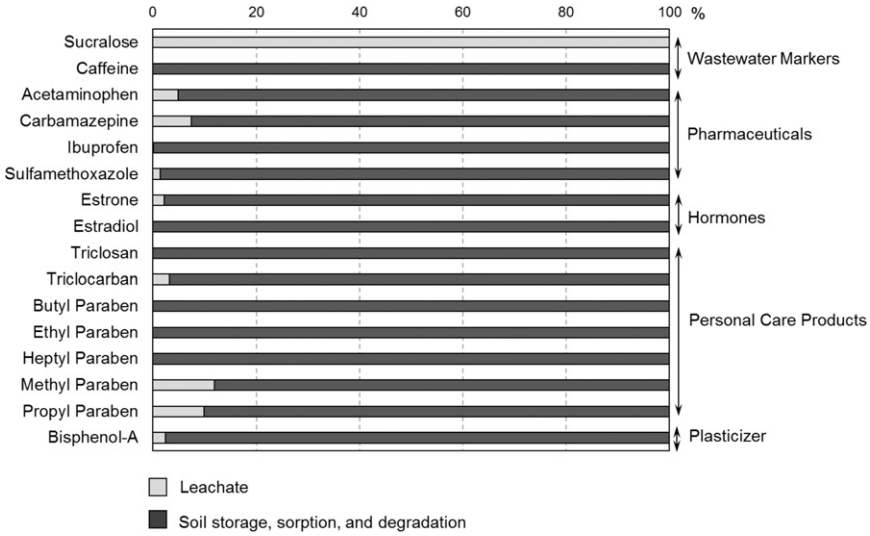

Fig. 3. Average percent reduction of micropollutants loads from the septic tank effluent (n = 34) to leachate (n = 120) from septic system drainfields.

filled porosity was greater in soil layer (mean: 0.78) than sand layer (mean: 0.22), which likely created differential oxygen availability. These findings are in line with previous studies that showed acetaminophen degradation in septic system drainfields (Teerlink et al., 2012b) and oxygen availability was a limiting factor for the elimination of micropollutants (Carrara et al., 2008; Godfrey et al., 2007; Göbel et al., 2007).

Some micropollutants such as carbamazepine, ibuprofen, estrone, 17β-estradiol, triclosan, triclocarban, and bisphenol-A with log K_{ow} values ranging from 2.25 to 4.99 could be partially attenuated by sorption (Scheytt et al., 2005). For example, triclosan and triclocarban have shown resistance to biotransformation and tend to partition into soil or sediment in the environment (Heidler and Halden, 2007; Ying et al., 2007). In contrast, microbial degradation is likely the removal

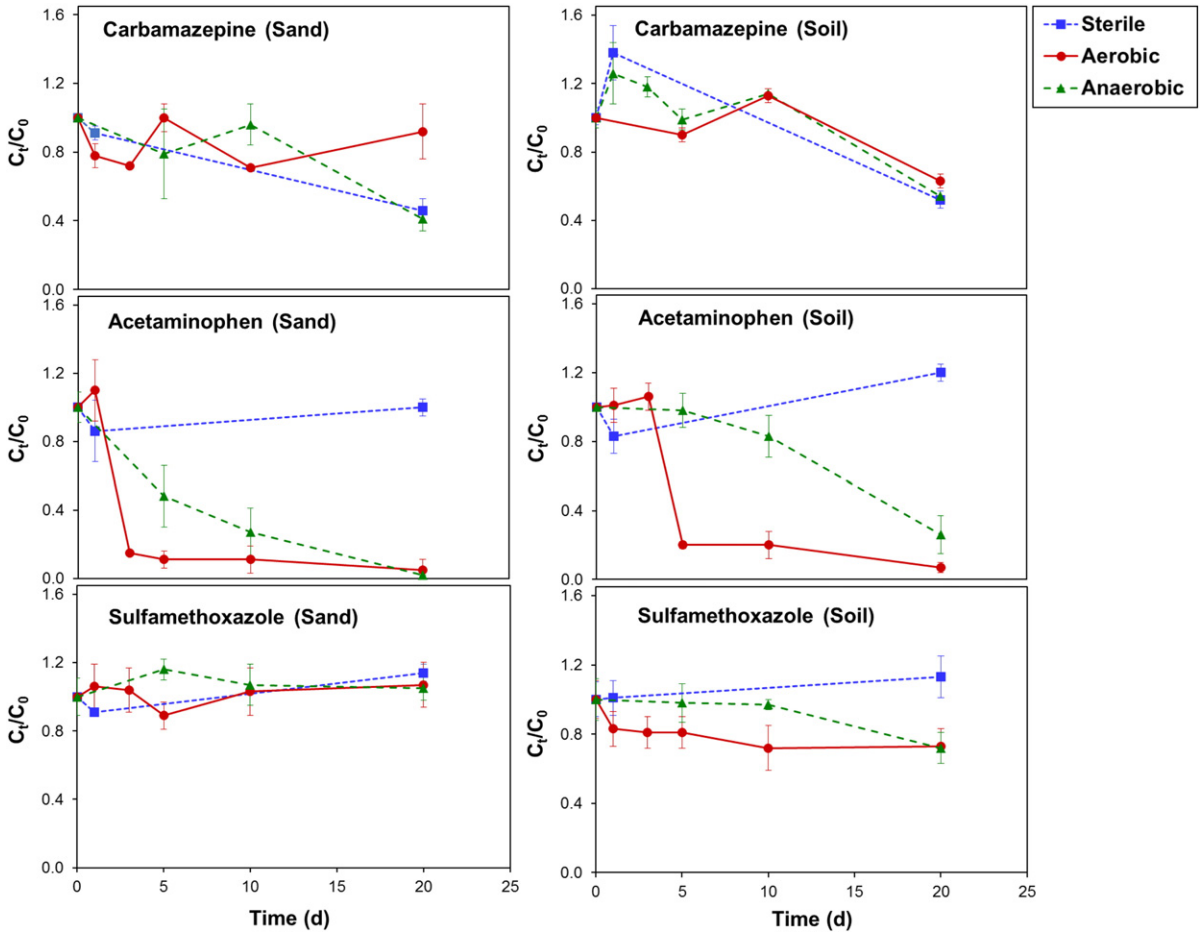

Fig. 4. Relative concentration (C_t/C_0) of the micropollutants during the incubation study in soil and sand layers of septic drainfields. Sterile represents no microorganisms in the system. Error bars represent the standard deviation of three replicate samples.

mechanism for the compounds that are more hydrophilic and known not to bind to organic matter present in the drainfields. Martín et al. (2012) reported that 99% removal rate of ibuprofen from four WWTPs was likely due to the biodegradation instead of sorption onto sludge. In addition, biological removal of parabens was found to be >97% in a household wastewater system (Andersen et al., 2007) and more than 99% by using activated-sludge treatment methods (Yamamoto et al., 2007). Bisphenol-A was found not likely to be stable, mobile, or bioavailable in soils (Fent et al., 2003).

The mass of carbamazepine and sulfamethoxazole was found to be <15% in the leachate, and these two compounds were detected in the drainfields (Figs. 2 and 3). Further, our incubation study showed no significant microbial degradation of carbamazepine and sulfamethoxazole (Fig. 4). Previous studies have observed variable removal efficiencies (22–89%) of sulfamethoxazole due to the microbial degradation (SI Table S1), whereas some work has shown that sulfamethoxazole was highly resistant to microbial degradation (Suarez et al., 2010). Dalkmann et al. (2014) observed that microbial degradation did not substantially contribute to sulfamethoxazole dissipation in soils after long-term irrigation with untreated wastewater. Du et al. (2014) reported 8 and 48% removal rates of sulfamethoxazole from various types of septic systems. Given that the sulfamethoxazole primarily exists as an anion in the drainfields, most of the detected sulfamethoxazole would be associated with the soil surface by cation bridging, with minor contributions from van der Waals force (Srinivasan et al., 2013; Thiele-Bruhn and Aust, 2004). These findings suggest that sorption was the main removal mechanism of carbamazepine, which partially affected transport of sulfamethoxazole in the drainfield.

There is also a possibility of micropollutants transfer from soil to plants via root systems. Such uptake by plants has been reported for acetaminophen (Tanoue et al., 2012), carbamazepine (Herklotz et al., 2010; Shenker et al., 2011; Wu et al., 2014), sulfamethoxazole (Sabourin et al., 2012; Tanoue et al., 2012), ibuprofen (Calderón-Preciado et al., 2012), 17β-estradiol, and caffeine (Sabourin et al., 2012; Wu et al., 2014). Plant uptake of micropollutants in the current study was considered to be relatively insignificant based on previous research that demonstrated <2% of micropollutants applied to soil are taken up by plants (Dolliver et al., 2007; Kumar et al., 2005; Colon and Toor, 2016). Volatilization is not likely to be a removal mechanism of selected micropollutants due to their small Henry's law constants (3.99×10^{-19}–1.5×10^{-7} atm m^3/mol) with low vapor pressures (3.25×10^{-14}–5×10^{-3} mmHg at 25 °C) (United States Environmental Protection Agency, U.S. EPA, 2012). Overall, our data suggest that vadose zone processes, sorption and microbial degradation, were effective at removing and transforming micropollutants in the septic system drainfields.

4. Conclusions

This study investigated the fate and transport of 17 micropollutants including wastewater markers (i.e., sucralose and caffeine), hormones, and PPCPs in septic system drainfields. The removal of many micropollutants was more than 85% in 60-cm deep drainfields containing aerobic sand and relatively anaerobic soil layers. Vadose zone processes such as sorption, transformations, and microbial degradation likely limited transport of micropollutants from the drainfields. Results from this study provided insights on the fate of micropollutants inside drainfields and improved our understanding of the transport of micropollutants in septic systems. The absolute transport/removal mechanisms of micropollutants studied here could not be determined as a detailed analysis of transformation products of each micropollutant is needed. However, given the evidence generated in this study pertaining to the efficacy of vadose zone processes at removing micropollutants, it seems obvious that maintaining aerobic conditions in parts of the drainfields are needed to effectively remove micropollutants and protect groundwater. The presence of micropollutants in the leachate also raises concerns about potential implications as many investigated micropollutants (e.g., carbamazepine and sulfamethoxazole) have been detected in the groundwater and surface water. As many septic systems in coastal states of United States are located within 60-cm of groundwater, there may be concerns of leaching of micropollutants to shallow groundwater and eventually to surface waters. Further, it is possible that micropollutants can accumulate in the drainfields and eventually leach in the long-term operation (>20+ years) of septic systems. Future research should target transport of micropollutants in septic systems over longer time periods, which would allow to better characterize micropollutant transport pathways and attenuation mechanisms. More research is also needed to determine the possible ecological and human risks associated with individual micropollutants and their mixtures leaching from septic systems.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.06.043.

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