

Designing OSSF Surface Application Systems: Update on Effects of Infiltration and Fate of Emerging Compounds

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INTRODUCTION

Pharmaceuticals and personal care products (PPCPs) are compounds used worldwide by individuals for health care, cosmetics, and veterinary medicines. Finding some ecological effects by exposure to PPCPs Fraker and Smith (2004) and Vajda et al., (2008) identified a concern about pharmaceuticals occurrence in the environment. However, long-term behavior of these chemicals in the environment at a very low concentration is largely unknown.

Once these compounds reach the wastewater treatment systems, they may undergo degradation and sorption processes or remain unchanged through the system. Wastewater treatment plant effluents are discharged to surface water sources or land applied and transferred to groundwater. Both sources are utilized as drinking water supplies and treated in water treatment plants. Technology developments in trace elements analysis and detection methods provided the opportunity to identify these chemicals in drinking water at part per trillion (ppt or ng/l) levels (Bhandari et al., 2009). Since then, PPCPs have been evident in a wide range of environmental media. Depending on type of media and treatment process, removal of compound may occur through aerobic and anaerobic biodegradation, adsorption (to soil, biosolids or activated carbon), photodegradation, hydrolysis and chemical oxidation. Wastewater effluent is commonly reused as irrigation water, which is able to fertilize the soil for crop growth.

PPCPs occurrence in soils and groundwater at the site of wastewater effluent land application have been discussed by a few researchers (Ternes et al. 2007, Karnjanapiboonwong et al. 2010, Gibson et al., 2010 and Xia et al., 2005) and they found PPCPs were detected in groundwater samples within the range of 0 to 1745 ng/l. When wastewater is land applied it may go through photodegradation, sorption to the soil particles, biodegradation or plant uptake. Soil components such as organic matter content, pH, clay content and presence of ions may affect the sorption process (Monteiro and Boxall, 2010). Acidic compounds ($pK_a < 7$) are not strongly adsorbed to the soil OM, which is negatively charged. But basic compounds have more tendencies to adsorb to soil OM. Clay content of the soil has a high sorption capacity due to its small pore sizes and large specific area. Since PPCPs are present in the environment at very low concentrations and sorption to soil is one of the major removal mechanisms, they are not usually bio-available to microbes (Onesios et al., 2009). Wastewater when applied on a vegetated land, plant uptake of the pharmaceuticals is one of the removal mechanisms. Liquids within the soil moves into the plant. So, solubility of the compound is an important factor for plant uptake. Also, organics with moderate hydrophobic characteristics [$0.5 < \text{LogKow} < 3$] are more likely to be taken up by plants (Wenzel et al., 1999).

ONSITE SEWAGE FACILITIES

On-Site Sewage Facilities (OSSFs) are the treatment systems installed at the site of one or a group of households developed in small communities. Wastewater is collected and discharged to a septic tank for treatment. The treated effluent is irrigated over a soil absorption site, which is either a vegetated area or an underground dripping system. Therefore, wastewater effluent is going through removal processes

within the soil such as filtration, sorption and degradation as discussed above. While these treatment systems are not commonly used in large areas, they have not been evaluated adequately for occurrence and removal of pharmaceuticals.

HYPOTHESIS

Within the last decade, a great deal of research has been conducted to investigate the occurrence, fate and removal of PPCPs. However, many of the experiments were established in laboratories with predetermined variables and conditions, so pharmaceutical behavior assessment in soil lacks pilot-scale studies where all removal procedures take place. Therefore, there are still research needs available prior to providing any regulations or guidelines for wastewater treatment plant management regarding PPCP issues. In this research, fate and removal of the pharmaceuticals in a pilot-scale OSSF system has been examined. Also, it is hypothesized that the wastewater containing PPCPs can be substantially cleaned of its PPCP load by surface application and percolation through the soil profile.

OBJECTIVES

The main objectives of this research are:

1. To investigate the PPCP removal capacity of a land treatment site soil throughout the year by using soil lysimeters
2. To determine the significance of the effect of photodegradation on PPCP removal rate
3. To determine the significance of the effect of plant existence on PPCP removal rate

METHODOLOGY

Soil columns were developed by direct removal of soil from the ground at the site of wastewater land application as we were interested in keeping the soil profile, heterogeneity and ground cover unchanged for our data to represent the native soil behavior within the treatment period. The test columns used were 12-inch diameter soil columns 3 feet in height containing 2.5 feet of soil and 6 inches of sand to gravel at the bottom to allow water to be captured and collected as it is passed through the column of soil. Three different groups of soil lysimeters, grass-covered, plastic-covered and open columns, each with three replications, have been provided to define the effect of UV radiation and grass presence in removal rates of pharmaceuticals. Starting January 2010, synthetic wastewater was applied over the soil columns using a drip irrigation system on top of each column. Peristaltic pump was used to transport water from the tank to the irrigation system. The pumping schedule was adjusted using a timer in a way that columns were irrigated for 10-20 min at 5.00 AM to avoid any extra evaporation. Loading rates are adjusted for each month to meet the evapotranspiration (ET) rates and have at least 1 liter of effluent to be sufficient for laboratory analysis. To avoid having more variables in removal analysis, synthetic wastewater was used. Wastewater consisted of a mixture of tap water and four different pharmaceuticals, each in 100 ppb of concentration. Drugs were selected in a way to cover four main therapeutic classes, antibiotics, anti-inflammatories, anti-epileptics and anti-depressants, listed in Table 1. All columns were drained over night every month; 1 liter- volume samples were collected in amber bottles, packed and shipped in ice to Baylor University. Samples were analyzed in Dr. Bryan Brooks' laboratory for pharmaceutical concentration in the collected water. Solid phase extraction method followed by liquid chromatography-tandem mass spectrometry were used to determine the concentration of target pharmaceuticals present in leachate and influent and effluent samples.

Table 1: Physical and chemical properties of selected pharmaceuticals *

Compound name (CAS #)	Acetaminophen (103-90-2)	Carbamazepine (298-46-4)	Fluoxetine hydrochloride (54910-89-3)	Trimethoprim (738-70-5)
Therapeutic use	OTC drug used to relieve pain and fever, analgesic (Anti-inflammatory)	Used to prevent and control seizures (anti-epileptics)	Used for the treatment of depression and other mental/mood disorders (antidepressant)	Used to treat urinary tract and other infections (antibiotic)
pKa	9.38	13.9	10.06	7.12
Log kow	0.46	2.45	4.65	0.91
Solubility(in water at 25°C), mg/l	14,000	18	14,000	400
Environmental half life (hr)	16-26	>1500		>720
Other	---	Stable to hydrolysis	---	---

* The following information was generated from the hazardous Substances Data Bank (HSDB), a database of the National Library of Medicine's TOXNET system (<http://toxnet.nlm.nih.gov>) on May 12, 2010.

RESULTS AND PRIMARY INTERPRETATIONS:

Columns were loaded based on ET calculations and volume leachate required. So, the loading rate is different for each month as well as the leachate water volumes (Tables 2&3). Concentrations of target PPCPs are defined in all samples taken from column leachates and influent to the system.

Table 2: Loading water volumes (ml) for each month

	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec
Grass-covered columns	13500*	13500	6290	6362	6603	7750	11600	17600	15000	12000	9000	7500
Plastic-Covered columns	12000	12000	5844	6278	6447	6200	5220	5120	4800	4800	4500	4500
Open columns	12000	12000	6113	6337	6102	6200	7250	11200	13500	12000	8600	7067

Table 3: Leaching water volumes (ml) for each month

	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec
Grass-covered columns	762.3*	5600	1033	10	10	0	0	2210	5123	5597	4973	4843
Plastic-Covered columns	2793	4707	2483	2767	2967	1553	2100	1637	1993	3047	2185	2640
Open columns	571.7	1967	1433	486.7	520	0	0	0	2140	3070	3080	3537

*Water volumes are average of three replicates

Within the summer season, as we expected, no water was collected from some open and grass-covered columns due to the high evapotranspiration (ET) rate. Figures 1 through 4 show the leachate concentrations of acetaminophen, trimethoprim, carbamazepine and fluoxetine throughout the test period. Concentrations are the calculated average of the three replications of each column type with the standard deviation in the range of 0-99 ng/l. Results represented the removal rates of more than 95% in all columns.

Regarding Figure 1, acetaminophen concentration is higher for open columns in the month of February and April which may be a result of photodegradation in the open columns, since there are more sunny days in May compared to previous months. It can be seen that acetaminophen concentration in leachate is zero after the month of July; this may be a result of microbial population build-up in soil, since acetaminophen is highly biodegradable.

Trimetoprim concentrations (Figure 2) show a constant high removal of these compounds during the project period. If we look back at the chemical characteristics, provided in Table 1, we can see that it has low water solubility. Therefore, these compounds are more likely to adsorb onto soil particles instead of passing through the soil in a soluble form in the leached water.

Carbamazepine concentrations in leachate from grass-covered columns in Figure 3 show a varying trend throughout the year. Its presence has a reverse relation with plant uptake during seasons of the year. In fall season when plant uptake decreases, carbamazepine concentrations increase. This relation is reverse for plastic-covered columns which lacks plant presence and high variation in ET.

Figure 4 shows that high removal rates are also obtained for fluoxetine in all columns. Fluoxetine is a more soluble compound and can reach the groundwater if it is not degraded while passing through soil or not adsorbed to soil. This compound has a high log Kow, which indicates, it can be adsorbed to organic matter quickly. So, soil clay content could play an important role in the adsorption process of fluoxetine. Within June and July, fluoxetine concentrations in leachate was high, which may be a result of flushing since loading rates are higher in summer season. This would not have happened if fluoxetine was biodegraded within last 5 months.

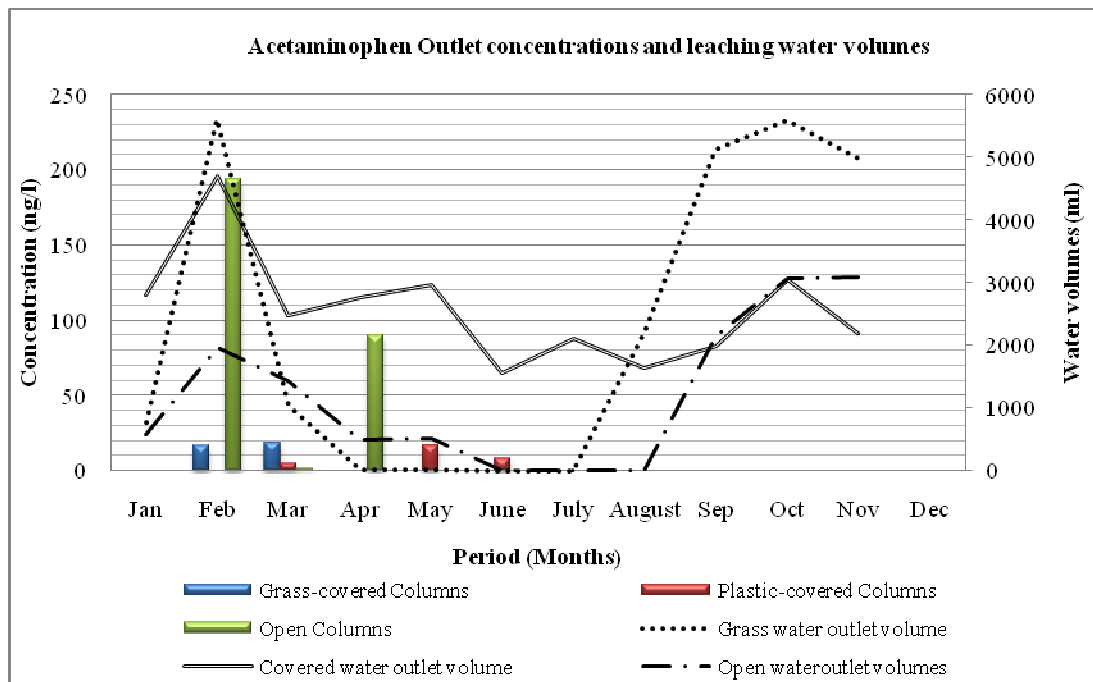


Fig. 1: Acetaminophen Concentrations in leachate in comparison with leaching water volumes

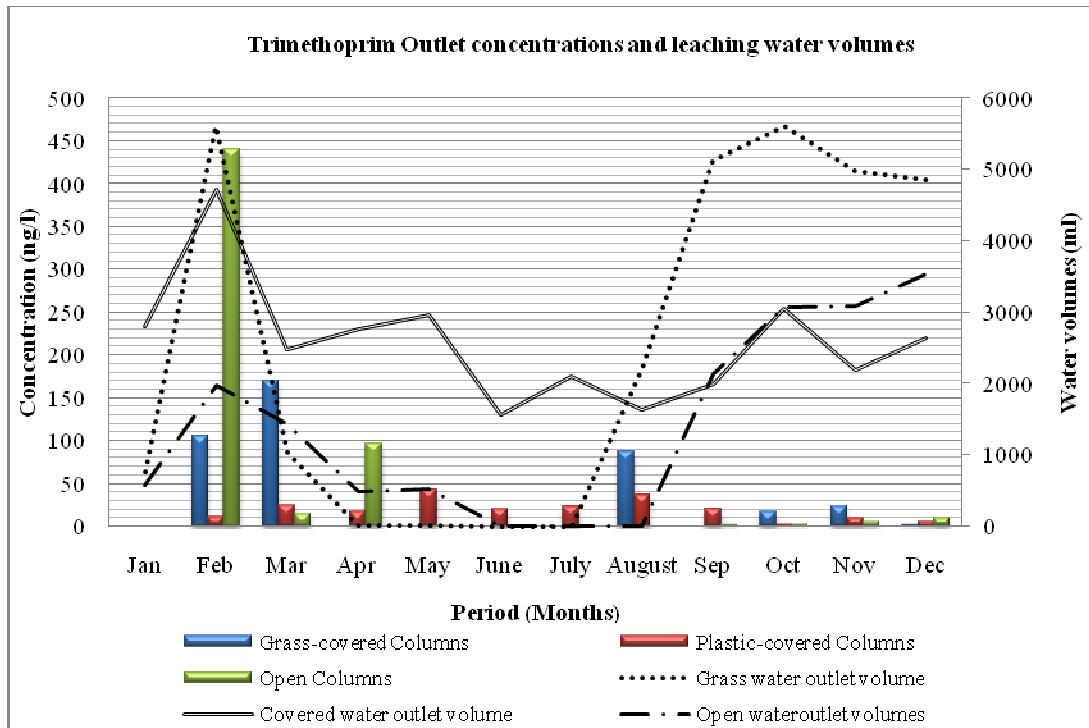


Fig. 2: Trimethoprim Concentrations in leachate in comparison with leaching water volumes

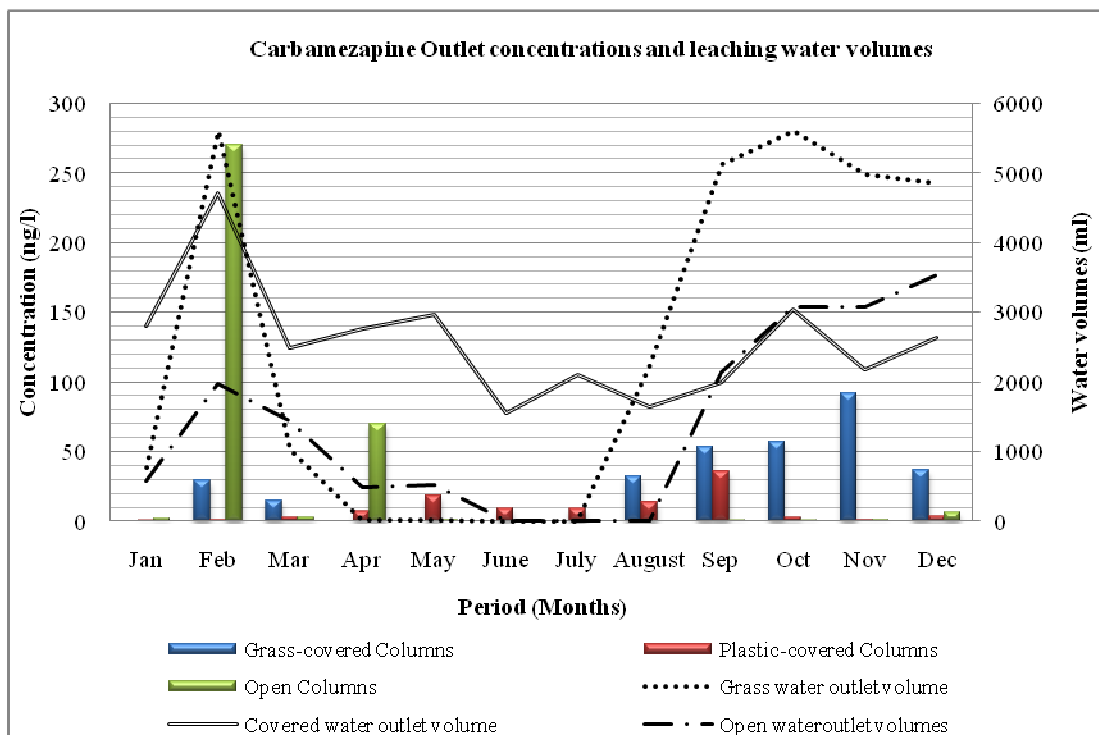


Fig. 3: Carbamazepine Concentrations in leachate in comparison with leaching water volumes

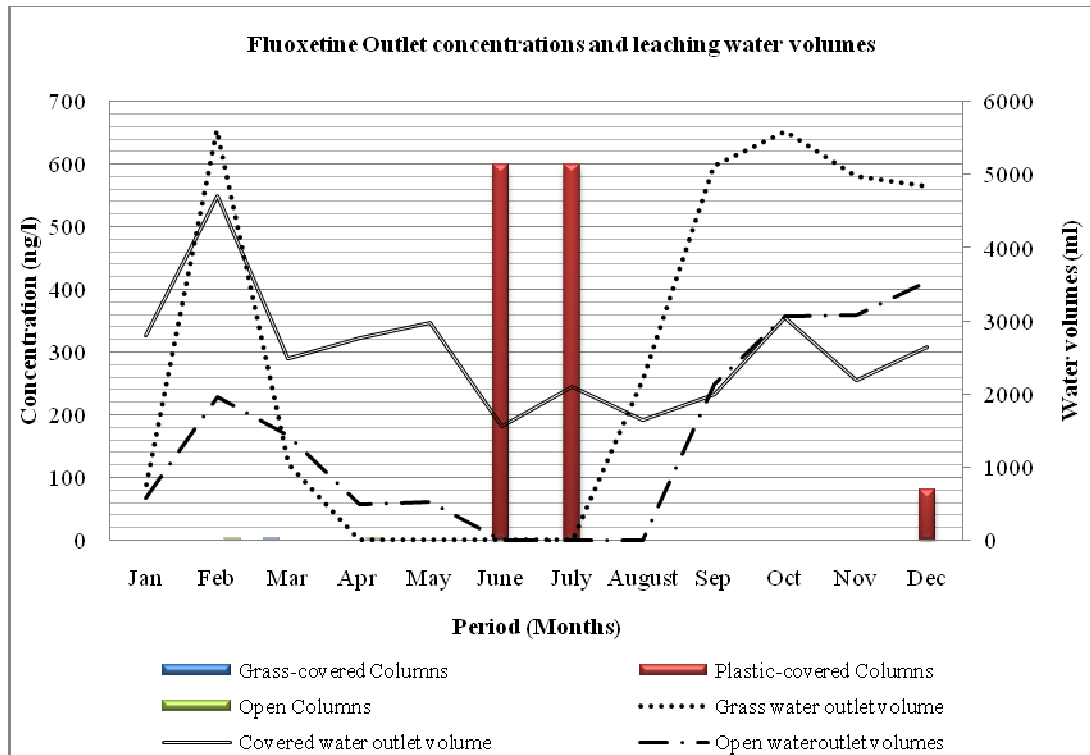


Fig. 4: Fluoxetine concentrations in leachate in comparison with leaching water volumes

Design of System Based on Infiltration

As one of the important components in the on-site sewage facility system, surface application system is the necessary approach to treat the effluent water. The core issue for the application system is to determine the soil infiltration rates at the various locations in Texas. An investigation and analysis of data from four regions in Texas have been made. It took two years to finish forty five soil infiltration rate tests using two infiltrometers and one hundred and fifty soil samples were analyzed to determine the soil classification. It is acknowledged that the base water intake rates of soils are approximately equal to the saturated hydraulic conductivity. Another source to obtain infiltration rates of soils or saturated hydraulic conductivity is from the NRCS soil survey website.

Saxton equation

Saxton et al. (1986) developed an equation that relates saturated hydraulic conductivity to soil texture and soil moisture content. The equation is:

$$K = 0.3937 \left\{ \exp \left[A + \left[\frac{B}{\theta} \right] \right] \right\}$$

Where,

$$A = 12.012 - 0.0755(\%sand)$$

$$B = -3.8950 + 0.03671(\%sand) - 0.1103(\%clay) + 8.7546 \times 10^{-4}(\%clay)^2$$

$$\Theta = 0.332 - 7.251 \times 10^{-4} (\%sand) + 0.1276 \log_{10} (\%clay)$$

K = Saturated hydraulic conductivity, inches/hr

θ = soil moisture content, ft³/ft³

Soil Survey Website

“Web Soil Survey (WSS) provides soil data and information produced by the National Cooperative Soil Survey. It is operated by the USDA Natural Resources Conservation Service (NRCS) and provides access to the largest natural resource information system in the world. NRCS has soil maps and data available online for more than 95 percent of the nation’s counties and anticipates having 100 percent in the near future”. ---NRCS soil survey website

Infiltration rate tests

Houston, Austin, Dallas and Lubbock are the test locations for the field test for different soil infiltration rates. The number of the tests distribution is illustrated in Table 1. Initial results of the data collected are provided in Table 2.

Table 1. Summary of samples collected and the relative locations.

	Houston	Austin	Dallas	Lubbock
Infiltrometer	9	9	9	20
Small ring infil	27	27	27	60
Tem, wind speed	9	9	9	20
Number of samples	27	27	27	60

Table 2. Summary of the initial results at the relative locations tested for infiltration rates as compared to that predicted by the Saxton (1986) prediction and the NRCS soil survey.

ID	Saxton hydraulic K,in/hr	NRCS, in/hr	Infiltration rates, in/hr
LBB090201-090205	0.12	0.03	0.06
LBB101-505	0.26	0.30	0.31
LBB-090101-090105	0.35	3.96	3.2
Hou-sugarland-A	0.10	0.0014-0.0595	0.30
Hou-sugarland-B	0.07	0.0014-0.0595	2.9
Hou-sugarland-C	0.09	0.0014-0.0595	0.13
Hou-9225-A	1.05	0.57-1.98	0.57
Hou-9225-B	0.95	0.57-1.98	0.16
Hou-9225-C	0.47	0.57-1.98	0.13
Hou-9527-A	1.06	0.35	0.89
Hou-9527-B	0.21	0.35	0.78
Hou-9527-C	0.23	0.35	0.18
Austin-A1-1	0.08	0.06-1.98	24.30
Austin-A1-2	0.42	0.06-1.98	2.42

Austin-A1-3	0.68	0.06-1.98	3.90
Austin-A2-1	0.12	0.57-1.98	0.09
Austin-A2-2	0.133	0.57-1.98	0.138
Austin-A2-3	0.137	0.57-1.98	0.65
Austin-A3-1	0.18	0.02-0.57	0.89
Austin-A3-2	0.13	0.02-0.57	1.84
Austin-A3-3	0.11	0.02-0.57	3.17

Fate of Nitrogen

The fate of nitrogen in a surface applied system was studied using triplicate columns for grass, open, and covered systems from December 2009 to December 2010. Influent water with 25 mg/L of total nitrogen was applied everyday through a simulated surface applied system. Bermuda grass was grown on three columns, another three columns had no grass but was open to the atmosphere, and other three columns were covered to restrict all sunlight from hitting the surface. Effluent from each column was drained each month. The collected effluent samples were analyzed for total nitrogen in the laboratory using standard DR/4000 procedure from HACH. The research and results from the column study over a period of 12 month cycle show that a proper mass balance on the nitrogen can achieve the desired results from a surface applied OSSF system. The nitrogen balance from this research provides us with the foundation needed for designing a proper surface applied system. This design also can be a good option for small towns and communities where soil can be used to renovate wastewater. The difference in sample mean is significant ($p > 0.001$) for the three different treatment columns (Figures 1 to 3). The difference in sample mean for the first three months ($p > 0.001$) between the grass and open column is not significant as it took about three months for the system to stabilize. This column study gives us a better understanding of the factors that contribute to nitrogen percolation into the groundwater and how that movement can be controlled. However a more detailed column study for the fate of nitrogen in different types of soil, coarse and fine soil, would provide useful data for modeling effective removal of nitrogen using an OSSF surface applied system.

TN Consumed by Grass Columns

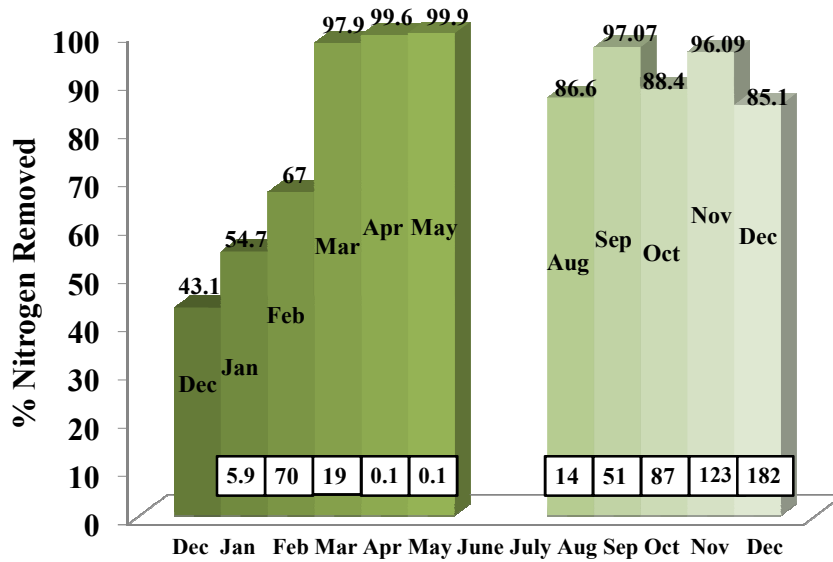


Figure 1. Percent nitrogen removal from a spray field growing Bermuda grass. Number in box represents the percent of excess water applied compared to estimated evapotranspiration.

TN Consumed by Open Columns

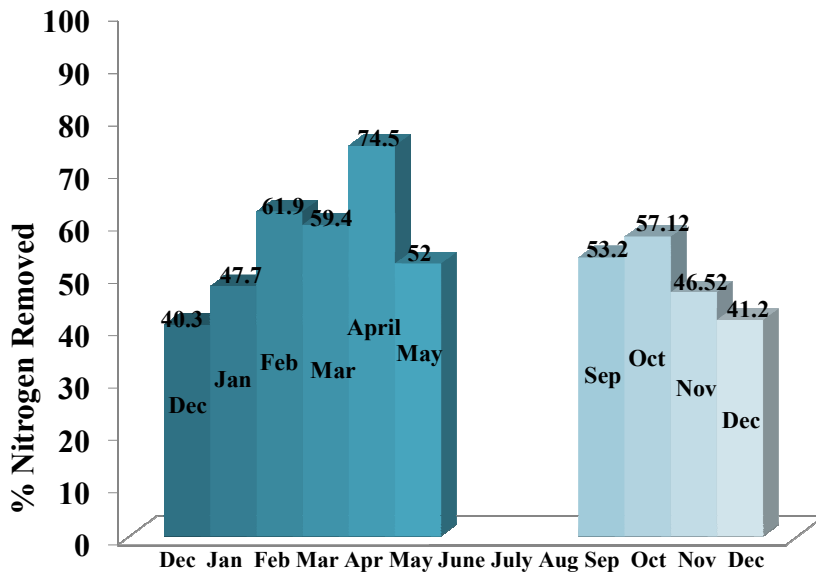


Figure 2. Percent nitrogen removal from columns receiving nitrogen where no crop was grown.

TN Consumed by Covered Columns

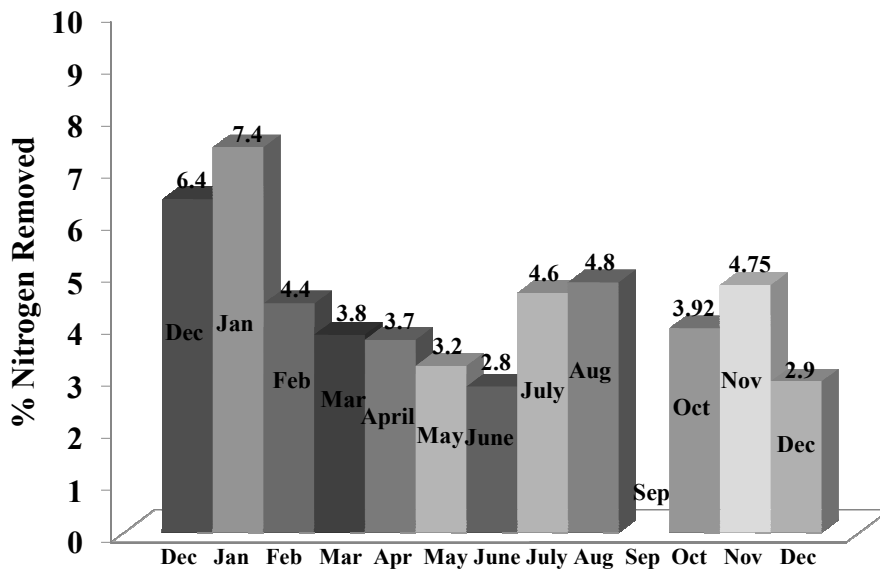


Figure 3. Percent nitrogen removal from columns covered to receive no sunlight. This could be used to estimate nitrogen removal from a leachfield receiving wastewater effluent.

CONCLUSIONS

From the preliminary interpretations, it can be found that the presence of plant, photodegradation and season of the year will affect the removal rates of PPCPs through land application of primary treated wastewater. Statistical analysis will be conducted to determine the significance of the effects of these parameters on removal rates and target compounds concentrations in the leachate which represents groundwater contamination. Some of the compounds such as fluoxetine represent accumulation and desorption pattern within the one year study. Further research would be necessary to study the accumulation of several compounds in soil media and their desorption behavior under a flushing event.

The variation in infiltration between that measured in the field compared to either the predictive approach or from a national database is considerable. Even though high variations in infiltration are expected, developing a new approach to designing system with such variations is needed.

When it comes to nitrogen removal in OSSF systems, it is apparent that a proper design based on the mass balance principles is necessary and provides the appropriate results. When no crop is present to remove the nitrogen, much larger systems will be required, but even then the design principles are not well understood. OSSF leachfields will remove little nitrogen, thus use of those systems should be carefully considered in location such as near lakes or in areas of high groundwater.

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Executive Summary for Nitrogen Study Add-on

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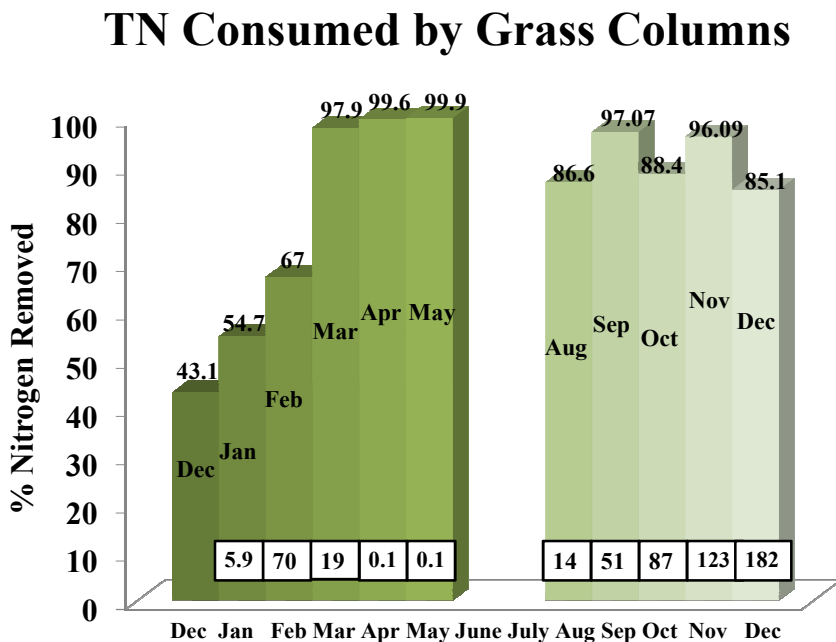


Figure 4. Percent nitrogen removal from a spray field growing Bermuda grass. Number in box represents the percent of excess water applied compared to estimated evapotranspiration.

TN Consumed by Open Columns

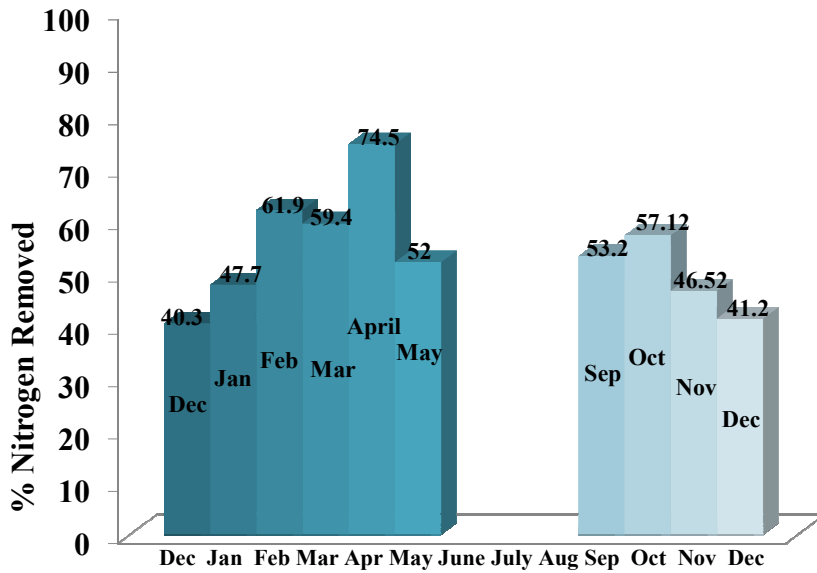


Figure 5. Percent nitrogen removal from columns receiving nitrogen where no crop was grown.

TN Consumed by Covered Columns

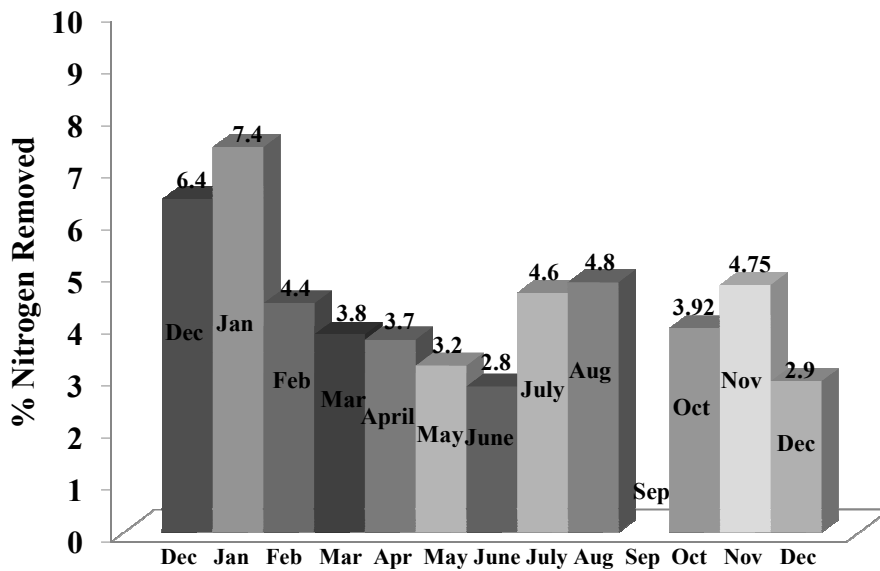


Figure 6. Percent nitrogen removal from columns covered to receive no sunlight. This could be used to estimate nitrogen removal from a leachfield receiving wastewater effluent.