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Pharmaceuticals and Personal Care Products (PPCPs) in Alfalfa Irrigated With Reclaimed Water

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Reclaiming Water for Urban Foodsheds *integrates basic scientific research with Extension outreach to examine the feasibility of using reclaimed water for irrigated agriculture in urban environments. Funded by a grant [2017-69007-26309] from the USDA National Institute of Food and Agriculture, research is conducted in University of Nevada, Reno campus laboratories and the Nevada Experiment Main Station Farm Laboratory and Greenhouse Complex. This special publication, which is part of a series, reports on research to quantify PPCPs in alfalfa irrigated with reclaimed water on farm-scale fields.*

Introduction

In water-scarce areas, diluted wastewater has been used for agricultural purposes for centuries (WWAP, 2017). In recent decades, advanced technologies have become available for treating wastewater (WWAP, 2017) so that, it is used increasingly as an agricultural irrigation water source (Calderón-Preciado et al., 2011; Chen et al., 2011; Hyland et al., 2015; Nason et al., 2019; Wu, Conkle, & Gan, 2012; Wu et al., 2014).

Wastewater treated for reuse is referred to as reclaimed water. A typical wastewater treatment process for reclamation includes:

- primary treatment to remove large objects and particles;
- secondary treatment to remove biochemical oxygen demand; and
- tertiary treatment consisting of sand filtration; and disinfection.

Truckee Meadows Wastewater Reclamation Facility (TMWRF) in Reno, Nevada, uses stringent nitrogen and phosphorus removal in addition to these treatment processes. Once the water has passed through the treatment plant, it is "reclaimed." Some TMWRF reclaimed water is used to irrigate local golf courses and the University of Nevada, Reno Experiment Station's Main Station Field Laboratory (MSFL). The remainder is released to the Truckee River to be used again elsewhere in the basin. Other wastewater treatment facilities in the U.S. similarly release reclaimed water to rivers and lakes or inject it into groundwater aquifers for storage and future reuse.

Various pollutants have been detected at trace concentrations in reclaimed water, including pharmaceuticals and personal care products (PPCP) (Chen et al., 2011; Dodgen et al., 2015; Goldstein, Shenker, & Chefetz, 2014; Guo & Krasner, 2009; Pan Liu, & Yin, 2016; Wu et al., 2014). Trace concentrations of pharmaceuticals are present because they are incompletely metabolized by humans and not entirely removed by wastewater treatment processes (Awfa et al., 2018; Pan et al., 2016). When reclaimed water is used for irrigation, soil and crops are exposed to PPCPs (Dodgen et al., 2015; Goldstein et al., 2014; Kinney et al., 2006; Wu et al., 2012; Wu et al., 2013; Wu et al., 2014). Thus, reclaimed water is a potential source of PPCPs in plants and soil when used for irrigation. Examples of pharmaceuticals found in trace amounts in reclaimed water include antibiotics, anticonvulsants and analgesic pain relievers. Personal care products found in reclaimed water include soaps, shampoos, cosmetics and insect repellents.

The presence of PPCPs in reclaimed wastewater and their translocation and accumulation in(to) irrigated crops may pose a phyto-toxicological risk (Herklotz et al., 2010), leading to decreased yields (Poustie et al., 2020), exposure to humans and livestock, or increased the proliferation of antibiotic resistance (Boxall et al., 2006). Human health risks associated with dietary intake of individual PPCPs present in reclaimed water-irrigated produce have been studied extensively and are estimated to be low (Carter et al., 2014; Wu et al., 2013). These studies compare acceptable daily intake (ADI) to consumptive exposure to determine if exposure may impact human health. However, data sources for such risk assessments typically are limited to samples from produce:

1) cultivated in hydroponic environments (Calderón-Preciado et al., 2011; Herklotz et al., 2010; Hyland et al., 2015; Nason et al., 2019);
2) grown under well-controlled greenhouse conditions (Carter et al., 2014; González-Naranjo, Boltes, & Biel-Maesto, 2013; Revitt, Balogh, & Jones, 2015); and/or
3) experimentally exposed to a single PPCP at a time (Chen et al., 2011; Grossberger et al., 2014; Kinney et al., 2006).

While results from such highly controlled experimental environments are useful, they do not represent the actual conditions of landscape-scale irrigated agriculture using reclaimed water. Little landscape-scale data exist because of the difficulty in conducting large field experiments where plants and soils are simultaneously exposed to multiple PPCPs from irrigation with reclaimed water. Eleven compounds were selected to represent diverse physicochemical, environmental transport, degradation and occurrence in reclaimed water. The objectives of this research were to:

1) determine the concentrations of various PPCPs in reclaimed water;
2) determine PPCP concentrations in soil, soil pore water, and the shoots and leaves of alfalfa irrigated with reclaimed water for a period greater than 10 years; and
3) determine the fate via mass balance of applied PPCPs that were present in soil up to 60 cm deep, and in the alfalfa shoots and leaves.

Research Methods

Fields located at MSFL in Reno-Sparks, Nevada, have been irrigated for more than 10 years, with reclaimed water provided by TMWRF. TMWRF treats wastewater from homes, offices, industry and storm drains located in the Truckee Meadows region. Between March and April 2018, water samples from TMWRF were collected once every week following the disinfection treatment i.e., reclaimed water. The collected samples were analyzed at the University of Nevada, Reno laboratories, including the Harry Reid Engineering Laboratory and Nevada Proteomics Center. To validate the methods used, researchers arranged for the samples to be analyzed by an external laboratory not associated with the university.

Soil samples at three depths (zero-20 cm, 20-40 cm and 40-60 cm) were collected in April 2018, and alfalfa shoots and leaves were sampled in July and September 2018. Lysimeters were installed in October 2018 at a depth of 15 inches and were separated by approximately 80 feet. Lysimeters were installed to capture the soil pore water during non-irrigating periods to determine if PPCPs remain in the soil after irrigation has stopped. Prior to the start of the irrigation season in late March 2019, the lysimeters were removed from the field. Lysimeters were removed during the growing/irrigating season because they are difficult to protect from grazing animals, and because the region is too arid to produce lysimeter samples of sufficient volume for analysis during this season. Irrigation started in mid-March and continued through the end of September.

All samples were transported on ice to university laboratories. Water samples were stored at 4 C, and soil and plant samples at -20 C, until processed, typically less than two weeks. The sample clean-up methods included filtration of water samples, freeze-drying soil and plant samples, and grinding and sieving the plant samples. The materials to be analyzed were extracted using solvents and an accelerated solvent extraction system (soil), centrifugation, sonication and decanting (alfalfa), and solid phase extraction (water, soil and alfalfa). Extracted samples were stored at -20 C until analyzed. All samples were analyzed with liquid chromatography tandem mass spectrometry in positive and negative electrospray ionization mode.

Results

PPCPs measured in this study were caffeine (stimulant), carbamazepine and primidone (anticonvulsants), DEET (insect repellent), diphenhydramine (antihistamines), fluoxetine (antidepressant), ibuprofen and ketoprofen (anti-inflammatories), meprobamate (anti-anxiety), and sulfamethoxazole and trimethoprim (antibiotics). Over a four-week period, the levels of PPCPs in the reclaimed water were monitored (Figure 1). Eleven PPCPs were identified and were in the range of 26±9 ng/L to 1,539±1,899.

The variability in the reclaimed water PPCP data likely indicates that the compounds were not removed at all or reflect variability in the influent (incoming) concentration of compounds in wastewater entering the treatment facility. While the treatment processes removed some of the measured compounds effectively, others persisted in the reclaimed water (Figure 1) in trace amounts.

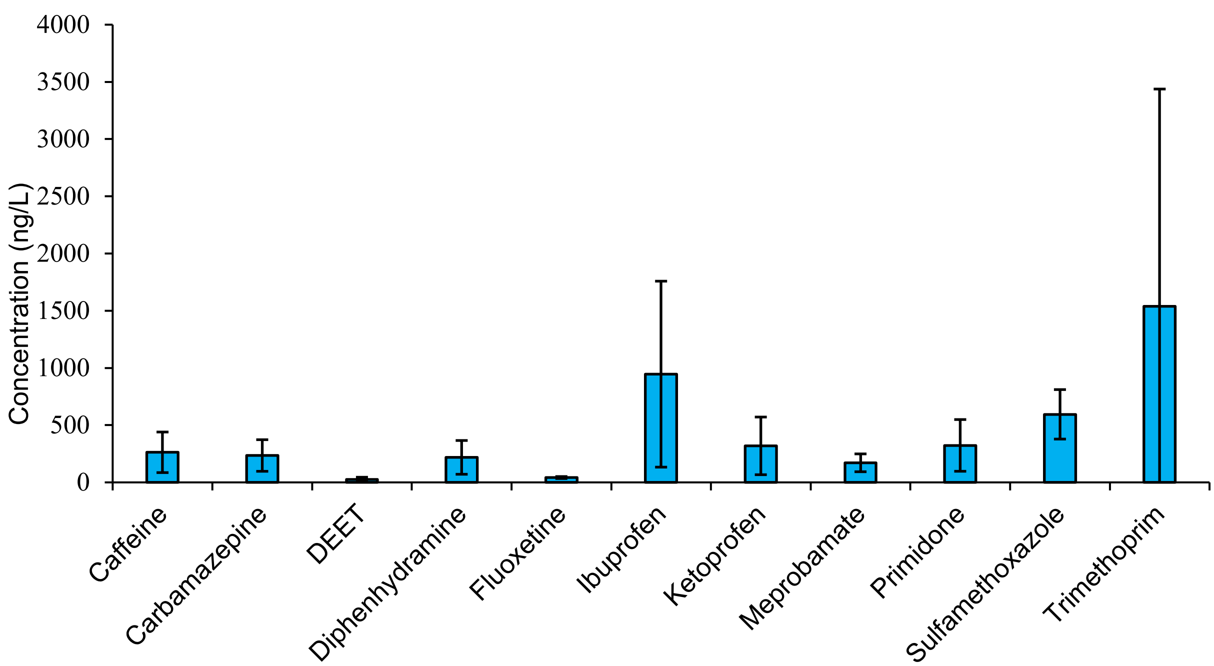

Figure 1. PPCPs in reclaimed water, treated at TMWRF. Error bars show the standard deviation of four samples analyzed by University laboratories and one separate off-campus laboratory. ng/L is nanograms per liter.

Soil samples were collected from three depths: zero-20 cm, 20-40 cm and 40-60 cm (Figure 2). Of the 11 PPCPs measured, nine PPCPs were present above detection limits. The concentrations ranged from 0.03 to 328 ng/gDW (nanogram per gram, dry weight) and were likely derived from reclaimed water applied to fields.

From zero-20 cm to 40-60 cm, the concentration of PPCPs decreased approximately 10% for all compounds, except carbamazepine and meprobamate. Carbamazepine concentration increased 2,700% from the surface to 40-60 cm, and meprobamate, which was present at five ng/g at 40-60 cm, was not detected at shallower depths. At 40-60 cm depth, carbamazepine was present at 1 ng/g, 10 ng/g and 976 ng/g because it tends to percolate and accumulate at depth (Chefetz, Mualem, & Ben Air, 2008; Gibson et al., 2010; Yu, Liu, & Wu, 2013). Carbamazepine does not sorb to soil, and therefore has potential to leach to deeper depths in soil via flow of water.

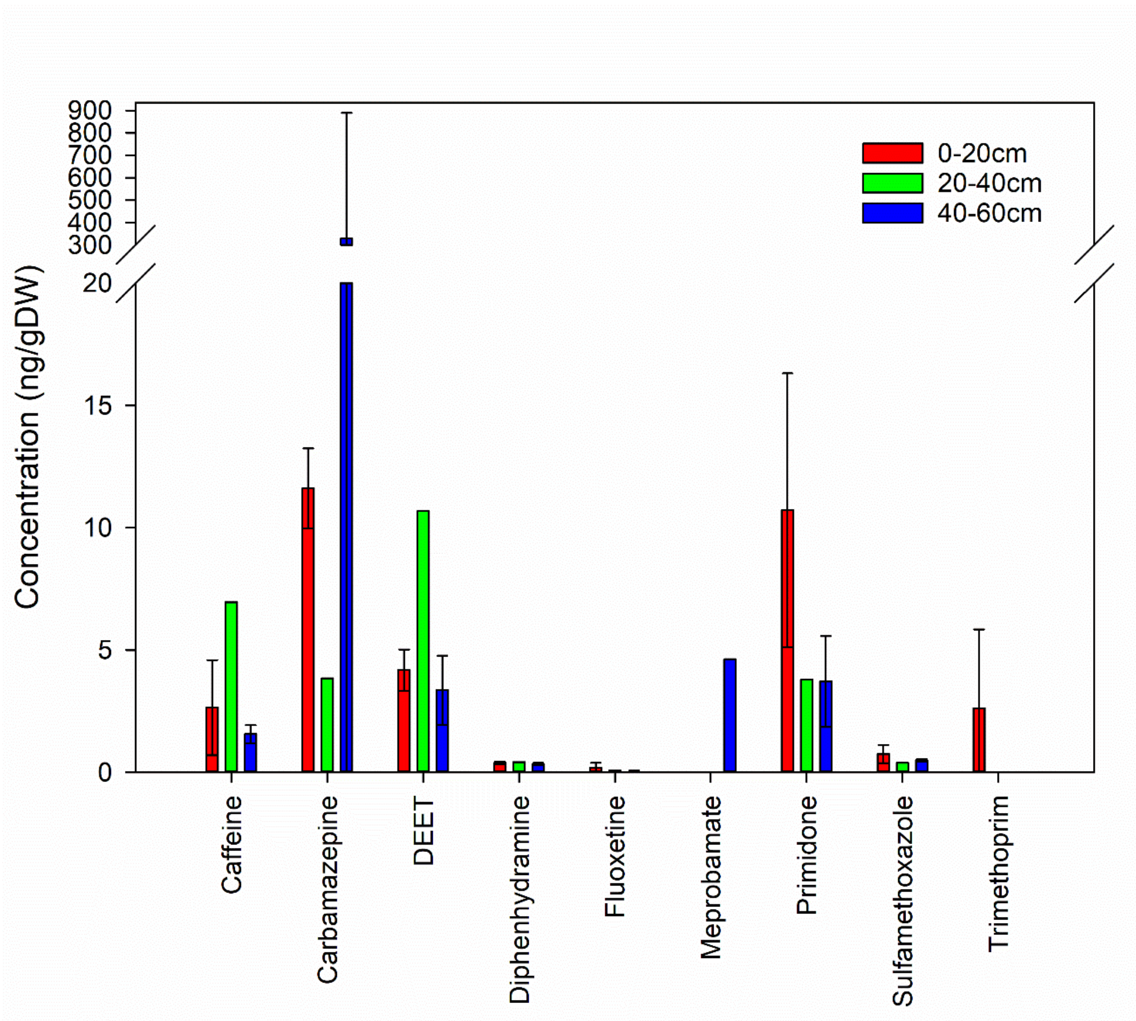

Figure 2. PPCP concentration in MSFL soil samples at three depths: zero-20 cm, 20-40 cm and 40-60 cm. No bar means below the detection limit. Error bars represent the standard deviation among triplicate samples. ng/g is nanogram per gram and DW stands for dry weight.

These results reflect landscape-scale field conditions when reclaimed water is used over 10 years to irrigate alfalfa.

Nine of 11 compounds were measured in the alfalfa plant tissues (shoots and leaves) irrigated with reclaimed water (see Figure 3). Leaves and shoots sampled in July contained higher concentrations of most compounds than leaves sampled in September. Compounds with measurable concentrations in July included carbamazepine (49 ng/g), primidone (20 ng/g), trimethoprim (22 ng/g) and sulfamethoxazole (13 ng/g). Only trimethoprim was observed at more than 10 ng/g in leaves sampled in September. Caffeine was below the detection limit in all alfalfa samples.

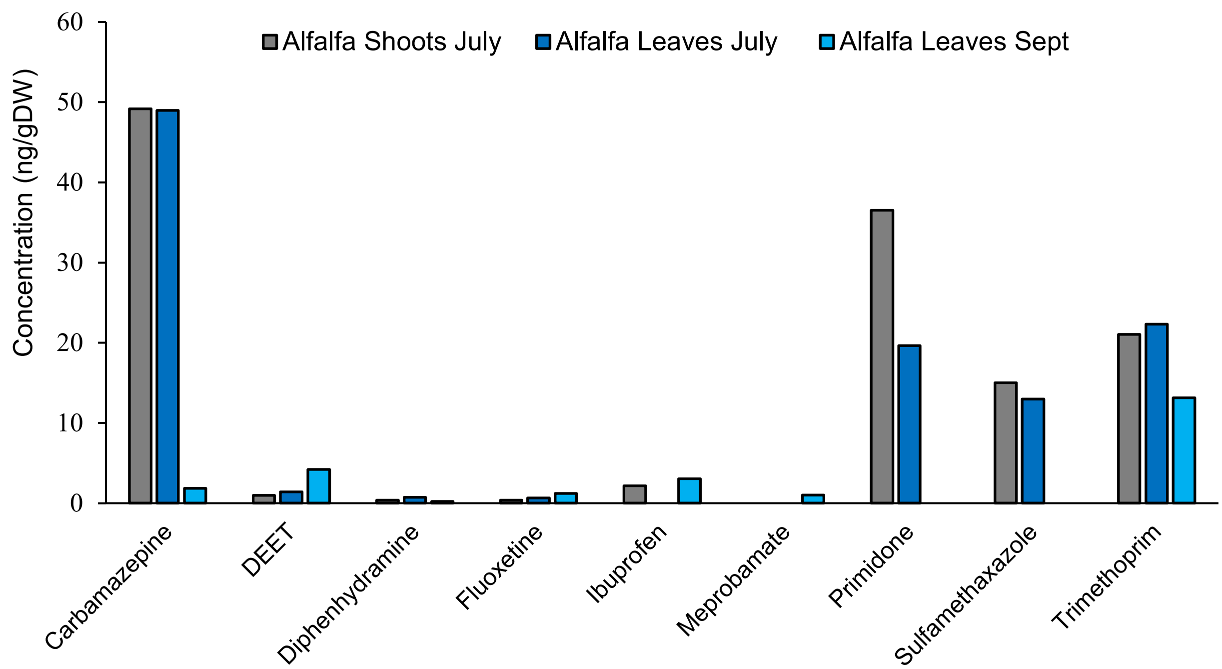

Figure 3. PPCP concentrations during July and September 2018 in alfalfa crops at MSFL irrigated with reclaimed water from TMWRF.

PPCP concentrations in the leachate samples ranged from 1 to 2,414 ng/L. Primidone and carbamazepine were present at the highest concentrations. These two highly persistent compounds are strong indicators of the reclaimed water use several months after application has ceased.

A mass balance of pharmaceuticals was conducted for the soil down to 60 cm, alfalfa shoots and leaves, and leachate to calculate the percentage of PPCPs lost over a season after applying irrigation water. The personal care product, DEET, was not included in the calculations because field workers applied the insect repellent during sampling. We estimated that only 0.2% to 5% of the original PPCP load accumulated in soil and 0.4% to 13% in alfalfa shoots and leaves. The leachate contained 1 to 54% of the applied mass of individual PPCPs. The exception was carbamazepine, which appeared to accumulate in soil over longer time periods.

Summary and Conclusion

This field-scale study at MSFL was conducted to evaluate the impact of long-term alfalfa irrigation with reclaimed water on PPCP concentrations in soils, alfalfa and soil pore water. PPCPs accumulated in soil at varying depths depending on the individual compound's physicochemical properties, even after irrigation with reclaimed water ceased. PPCP concentrations in soil and alfalfa were low (ng/g), even after being irrigated with reclaimed water for over 10 years.

Based on the presence and consumption (2.5% of 690 kg body weight of alfalfa (Selk, 2018), cattle may be exposed to a maximum of approximately 0.85 mg carbamazepine per day, or 0.05 mg/kg-day, compared to the human therapeutic dose of 11.4 mg/kg-day. Therefore, there are likely to be minimal or no health impacts to livestock. Future research is needed to assess human health risk by determining the uptake of PPCPs through the food chain to human consumption of meat and produce, but the risk is expected to be low to negligible. Considering the advantages of using reclaimed water for irrigation in water-scarce regions, the low risks from residual PPCPs after treatment appear sustainable.

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