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RESEARCH ARTICLE

Effects of soil moisture depletion on vegetable crop uptake of pharmaceuticals and personal care products (PPCPs)

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Abstract Agricultural crops have a long history of being irrigated with recycled wastewater (RW). However, its use on vegetable crops has been of concern due to the potential prevalence of microcontaminants, such as pharmaceuticals and personal care products (PPCPs) in the latter, which represents a possible health hazard to consumers. We investigated the uptake of three PPCPs (atenolol, diclofenac, and ofloxacin), at three different concentrations in irrigation water (0.5, 5, and 25 μg L^{-1}) in relation to three varying volumetric soil moisture depletion levels of 14 % (−4.26 kPa), 10 % (−8.66 kPa), and 7 % (−18.37 kPa) by various vegetable crop species. Experiments were conducted in a split-split block completely randomized design. PPCPs were extracted using a developed method of accelerated solvent extraction and solid phase extraction and analyzed via liquid chromatography mass spectrometry (LCMS). Results indicate that all treated crops were capable of PPCP uptake at nanogram per gram concentrations independent of the applied soil moisture depletion levels and PPCP concentrations. Ofloxacin was the chemical with the highest uptake amounts, followed by atenolol and then diclofenac. Although the results were not statistically significant, higher concentrations of PPCPs were detected in plants maintained under higher soil moisture levels of 14 % (−4.26 kPa).

Keywords Pharmaceuticals and personal care products · PPCPs · Soil moisture depletion · Crop uptake

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Introduction

Water resources, in particular freshwater, can be considered as the foundation for human survival, economic development, social welfare, and ecosystem function (Agboola and Braimoh 2009; UN-WWAP 2009, 2012). During recent decades, a variety of factors such as water pollution, climate variability, land use polices, urbanization, unstable economic tendencies, and the overexploitation of freshwater resources have put an unprecedented strain on this renewable but limited resource (UN-WWAP 2009; Davies and Simonovic 2011; Steduto et al. 2012; UN-WWAP 2012). These elements, added to an expected increase in agricultural food and fiber production, the possible increase of biofuel crops, and the continuous rise in demand of water for agricultural, industrial, and urban uses further compromise available freshwater resources and overall water security (Anderson 2003; de Fraiture et al. 2008; de Fraiture and Wichelns 2010; Steduto et al. 2012). Therefore, it is imperative to adopt measures that promote an elevated level of environmental and economic feasibility in the management and utilization of water resources for present and future water demands.

It is estimated that agriculture accounts for about 70 % of total global freshwater usage (Vallee et al. 2003; FAO 2005; Steduto et al. 2012). Hence, improving water productivity in agricultural systems would have a significant impact on

curbing some of the negative impacts brought upon freshwater resources and aid in attaining future water and food security. Water productivity can be defined as the net return of benefits per unit of water used and can be increased by producing multiple goods and services with the least amount of water possible (Molden et al. 2010). A practice used worldwide to increase water productivity in agriculture and which has greatly contributed towards a sustainable management of water resources is the use of recycled wastewater for the production of goods and services (Anderson 2003; Botti et al. 2009; Chen et al. 2012). Recycled wastewater (RW) has been used for a variety of agricultural, environmental, and industrial purposes throughout the world, such as irrigation, manufacturing processes, groundwater replenishment, streamflow restoration, and the creation of recreational areas (Pescod 1992; Burkhard et al. 2000; Furumai 2008; Chen et al. 2012; Plumlee et al. 2012; Bischel et al. 2013). RW has also been used for domestic purposes, as an alternate source of water in urban areas, (Parsons et al. 2010; Furumai 2008), and has been considered an economic "drought-proof" source of water (Anderson 2003; Benotti and Snyder 2009).

The integrated use of RW has numerous environmental and economic benefits (Anderson 2006). By increasing water supply, RW reduces the amount of water that has to be extracted from existing water resources and, in some cases, even replenishes them, aiding in their conservation (Furumai 2008; Pedrero et al. 2010; Chen et al. 2012). Effectively using RW for secondary uses also helps decrease water pollution, as there is a decrease in sludge discharge into the environment (Pescod 1992; Anderson 2003; Chen et al. 2012). From an agricultural perspective, RW is also beneficial, because it not only frees up higher-quality water for alternate purposes other than irrigation, but in addition, it contains trace nutrients that can supplement the soil and therefore be beneficial for crop production (Pescod 1992; Burkhard et al. 2000; Manas Ramirez et al. 2002; Anderson 2003; Pedrero et al. 2010).

However, there have been growing concerns regarding the use of RW for irrigation purposes due to the prevalence of microcontaminants. Given that the primary source of RW originates from municipal wastewater effluent (Sedlak et al. 2000), a large amount of compounds from commercial, industrial, and domestic origins can make their way through wastewater treatment facilities and end up in RW (Fatta-Kassinos et al. 2011). One emerging group of these compounds are pharmaceuticals and personal care products (PPCPs), which are used for personal health or cosmetic reasons and encompass a diverse collection of thousands of chemicals, including prescription and over-the-counter (OTC) drugs, fragrances, and cosmetics, among others (Fatta-Kassinos et al. 2011). PPCPs have been found to endure traditional wastewater treatment procedures (Daughton and Ternes 1999; Drewes et al. 2002; Stamatelatou et al. 2003; Carballa et al. 2004; Stackelberg et al. 2007; Zhang et al. 2008; Benotti and Snyder 2009; Zorita et al. 2009; Rosal et al. 2010; Deblonde et al. 2011; Ryu et al. 2011) and can also bioaccumulate in the environment (Vaicunas et al. 2013) with the potential of reaching humans through the food chain (Stamatelatou et al. 2003; McClellan and Halden, 2010; Fatta-Kassinos et al. 2011). Additionally, widely used quality guidelines on RW tend to focus mainly on risks from pathogens (Fatta-Kassinos et al. 2011), whereas PPCPs have not been subjected to strict scrutiny (Daughton and Ternes 1999). Hence, the potential amounts of PPCPs that have been introduced into the environment via RW, as well as their effects, cannot be accurately assessed.

Although many PPCPs do not exhibit acute toxicity, or have not been known to pose a significant risk to public health in terms of human dietary uptake (Dodgen et al. 2013; Wu et al. 2013), they do have the potential to disturb the endocrine system of non-target organisms at very low concentrations during long-term exposure (Daughton and Ternes 1999; Fatta-Kassinos et al. 2011). Chemical disturbances to the endocrine system interfere with natural hormone cycles, metabolism, development, reproduction, and growth (Jackson and Sutton 2008). Additionally, PPCPs include many antibiotic compounds, which have been found to endure traditional wastewater treatment procedures (Miao et al. 2004; Gros et al. 2007; Zorita et al. 2009; Chang et al. 2010; Gao et al. 2012). When these compounds are constantly applied at low doses to the environment, as in RW irrigation, it accelerates the global increase in antibacterial resistance (Willis 2000; Witte 2000; Schwartz et al. 2003; Kumar et al. 2005; Stine et al. 2007; Kemper 2008; Pignato et al. 2009; Barker-Reid et al. 2010; Knapp et al. 2010; LaPara et al. 2011; Jechalke et al. 2013). This, in turn, may alter the physiology of microbiotic ecosystems (Martinez 2009; Schauss et al. 2009), leading to the evolution of more drug-resistant pathogenic bacteria. Therefore, as RW is widely used for irrigation purposes, it represents an input pathway of PPCPs and antibiotics into terrestrial food webs with the likelihood of being taken up by plants and transferred to humans through consumption (Fatta-Kassinos et al. 2011; Karnjanapiboonwong et al. 2011).

There are a number of studies that have documented the uptake and accumulation of various PPCPs and antibiotics in plant tissues of food crops usually consumed raw. For example, Boxall et al. (2006) found that florfenicol, levamisole, and trimethoprim were taken up by lettuce, while enrofloxacin, florfenicol, and trimethoprim were found within carrot tissues. Similarly, Herklotz et al. (2010) determined that cabbage and rape can uptake carbamazepine, salbutamol, sulfamethoxaxole, and trimethoprim when grown under hydroponic conditions. Dolliver et al. (2007) found that the antibiotic sulfamethazine was taken up by corn, lettuce, and potato plants grown with sulfamethazine-supplemented swine manure, whereas Jones-Lepp et al.

(2010) found that clindamycin was taken up by spinach, lettuce, and carrot roots, and roxithormycin was taken up by lettuce and carrot roots. Triclosan and 17α-ethynylestradiol have also been found to bioaccumulate in bean plants grown in sand and soil (Karnjanapiboonwong et al. 2011), and carbamazepine has been shown to be absorbed by cucumber plants and bioaccumulated in the raw fruit (Shenker et al. 2011). Additionally, triclocarban, fluoxetine, triclosan, and diazepam were found at high levels in roots of lettuce, spinach, cucumber, and pepper crops, while meprobamate, primidone, carbamazepine, dilantin, and diuron exhibited active translocation from roots to leaves (Wu et al. 2013). Also, samples of citrus trees grown using reclaimed municipal wastewater indicated that they can uptake and translocate nonylphenol, bisphenol, and β-estradiol to the fruit, while hydroponic studies using lettuce indicated that these same compounds could all accumulate in the latter (Lu et al. 2012).

Therefore, in order to further understand the effects of PPCPs in agricultural systems, this study focused on examining the effects of soil moisture content, PPCP type, and PPCP concentration on the bioaccumulation of such chemicals by vegetable crops when irrigated with simulated RW.

Materials and methods

Conceptual description

We determined the PPCP uptake extent of several vegetable crops irrigated with spiked water as a proxy for RW by conducting two separate controlled experiments within an enclosed greenhouse at the University of Hawaii at Manoa (21° 18′ N, 157° 48′ W). This spiked irrigation water contained various chemical mixtures based on three different PPCPs, which have been identified in RW, at three different concentration levels. Additionally, the evaluated crops were submitted to three varying degrees of soil moisture depletion. By these means, we created several simulated RW solutions for irrigation where we evaluated if the treated crops could take up the applied PPCP compounds at each given concentration and soil moisture depletion level. A supplemental study was also performed where vegetable crops were grown in soil media that had been previously irrigated with the simulated RW, in order to examine any residual traces of the applied PPCPs within the agricultural system.

Experimental variables

The soil media used for the experimental trials was P.W. Gillibrand Co. 60 sieve pure silica sand (Table 1), which was selected in order to minimize PPCP solution interactions with soil organic matter (Hernandez-Ruiz et al. 2012). The experimental volumetric soil moisture depletion thresholds used throughout the study were set to begin irrigation at *14 %* (−4.26 kPa), *10 %* (−8.66 kPa), and *7 %* (−18.37 kPa). Irrigation was automatically terminated when the soil media reached field capacity, corresponding to a volumetric soil moisture content of *28 %* (−0.99 kPa). Soil moisture levels were measured using time domain transmissometry (TDT) soil moisture sensors (Miralles-Crespo and van Iersel 2011).

The selected PPCPs evaluated throughout the study were atenolol (ATN), ofloxacin (OFL), and diclofenac (DIC) (Table 2). ATN is a beta-blocker drug used in cardiovascular therapy, OFL is a fluoroquinolone antibiotic used to treat respiratory and urinary tract infections, and DIC is an NSAID prescribed for inflammatory and pain management. Traces of all three PPCPs have been found in RW and freshwater bodies (Mons et al. 2003; Ashton et al. 2004; Thomas and Hilton 2004; Bound and Voulvoulis 2005; Roberts and Thomas 2006; Li et al. 2013). We used the following three varying concentrations of PPCP solutions within our experiments: 1× (0.5 μg L^{-1}), 10× (5 μg L^{-1}), and 50× (25 μg L^{-1}). Our lowest 1× concentration is well within the range of values reported globally for RW effluents (Deblonde et al. 2011; Alidina et al. 2014; Teijon et al. 2010; Gao et al. 2012; Vidal-Dorsch et al. 2012). The selected crops to be evaluated were cowpea (*Vigna unguiculata*), Swiss chard (*Beta vulgaris* var. cicla) and turnip (*Brassica rapa* var. rapa) for the first experimental trial, while collards (*Brassica oleraceae* var. acephala), basil (*Ocimum basilicum*), lettuce (*Lactuca sativa*), and cilantro (*Coriandrum sativum*) were used during the second trial.

Experimental design

For the main PPCP uptake experiments, we designed a CRD split-split-plot design with three replications for each treatment. Chemical mixture concentration levels represented the main plot treatment, while soil moisture depletion percentages comprised the subplot treatment. Within each subplot, three and four plant varieties were grown for the first and second experimental trials respectively, and constitute the sub-subplots, as well as the experimental unit of the study (Table 3 and Fig. 1). This design allowed for the evaluation and comparison of the treatment effects of interest for each evaluated crop. In addition, this design provided for increased precision in measuring the treatment effect of soil moisture depletion on crop uptake of PPCPs (assigned at subplot-scale treatment) (Jones and Nachtsheim 2009), which was the main interest of the study.

Analysis of variance (ANOVA) was used to compare treatment results and significant differences were considered at 95 % significance levels ($p < 0.05$). All statistical analyses were conducted with SAS 9.3 (SAS Institute Inc., Cary, NC, USA) and all treatments were analyzed as fixed effects.

Table 1 Physical and chemical analyses of utilized soil media

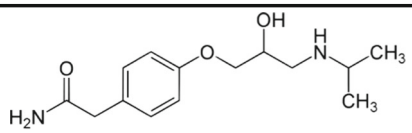
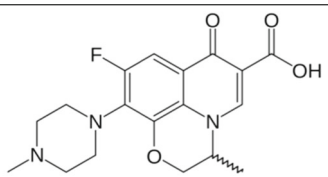
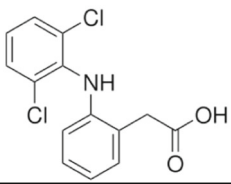
Soil texture									
Sand %		*Silt %*		*Clay %*		*USDA classification*			
99.8		0.2		0.0		Sand			
Particle size distribution (%)									
1.0–2.0 mm		*0.50–1.0 mm*		*0.25–0.50 mm*		*0.10–0.25 mm*		*0.05–0.10 mm*	
0.0		1.5		74.0		23.0		1.5	
Chemical compound analysis, LOI, and BD									
SIO_2	AL_2O_3	Fe_2O_3	Na_2O	CaO	MgO	K_2O	TiO_2	LOI	BD (g mL^{-1})
75–84 %	8–12 %	0.2–0.6 %	1–4 %	1–3 %	0–0.1 %	3–5 %	0–0.3 %	0–0.2 %	1.49

LOI loss of ignition, *BD* bulk density

Experimental setup

For each experimental trial, plants were grown in 30 polyvinyl chloride (PVC) bins (90 cm × 60 cm × 20 cm), each containing 60 kg of silica sand as soil media. Fifteen plants of each crop were planted in each bin within a randomly assigned subsubplot measuring 0.18 m^2 (Fig. 2). Each bin was irrigated using an automated microdrip irrigation system controlled by an Acclima C3500 Irrigation Controller and time domain transmissometry (TDT) soil moisture sensors (Miralles-Crespo and van Iersel 2011). When preset soil moisture depletion thresholds (14, 10, and 7 %) were reached for any given bin, the irrigation controller opened a solenoid valve corresponding to the designated bin needing irrigation and was watered until reaching estimated field capacity at 28 % volumetric soil moisture. Water, with a measured pH of 7.8,

Table 2 Physical and chemical properties of applied PPCPs

PPCP compound	Chemical structure	Formula	Molar weight (g mol^{-1})	pK_a	log K_{ow}[a]	Water solubility (25 °C)
Atenolol		$C_{14}H_{22}N_2O_3$	266.34	9.6[b]	0.16[e]	1.33 g L^{-1}[g]
Ofloxacin		$C_{18}H_{20}FN_3O_4$	361.38	5.97[c]	–0.39[e]	2.83 g L^{-1}[h]
Diclofenac		$C_{14}H_{11}Cl_2NO_2$	296.16	4.15[d]	4.51[f]	2.37 mg L^{-1}[i]

[a] Octanol-water partition coefficient

[b] O'Neil et al. (2001)

[c] Tolls (2001)

[d] Sangster (1997)

[e] Hansch and Leo (1995)

[f] Avdeef (2001)

[g] McFarland et al. (2001)

[h] Meylan et al. (1996)

[i] Fini et al. (1986)

Table 3 Sources of variation

		Experimental trial			
	Source of variation	1	2	Residual	*F* test
	Total	*df* 80	*df* 107	*df* 17	
Main plot	PPCP solution (Ch)	2	2	2	MS Ch/MS error a
	Error a	6	6	6	
	Soil moisture % (M)	2	2	1	MS M/MS error b
Subplot	Ch*M	4	4	2	MS Ch*M/MS error b
	Error b	12	12	6	
	Plant crop (P)	2	3	–	MS P/MS error c
	Ch*P	4	6	–	MS Ch*P/MS error c
Sub-subplot	M*P	4	6	–	MS M*P/MS error c
	Ch*M*P	8	12	–	MS Ch*M*P/MS error c
	Error c	36	54	–	

was supplied by municipal water lines and passed through a chemical injector (Dosatron D14MZ2) delivering a specific amount of plant nutrients (0.1 g L^{-1} 19.5-19.5-19.5 Gaviota 60 soluble fertilizer with 0.05 g L^{-1} 15.5-0-0 Yaraliva Calcinit Solution) and then towards the corresponding bin. Bins were arranged so that each row corresponded to one of the three different PPCP solutions, while every bin within each row had a different soil moisture depletion threshold (Fig. 2). Every time a solenoid valve opened in response to the soil moisture sensor reading, irrigation water with nutrient solution passed through a second chemical injector, which supplied the corresponding specific dose of PPCP solution to the running water. These spiked solutions were maintained at 20 °C in 10 L Nalgene containers under refrigeration. Water pressure was reduced at the intake point to 0.04 MPa using a Senninger preset pressure regulator, which also set the water flow to fluctuate at a maximum of 1.9 L min^{-1}. Air temperature, relative humidity, and light irradiation intensity were continuously monitored throughout the duration of the study using a HOBO U12-012 data logger.

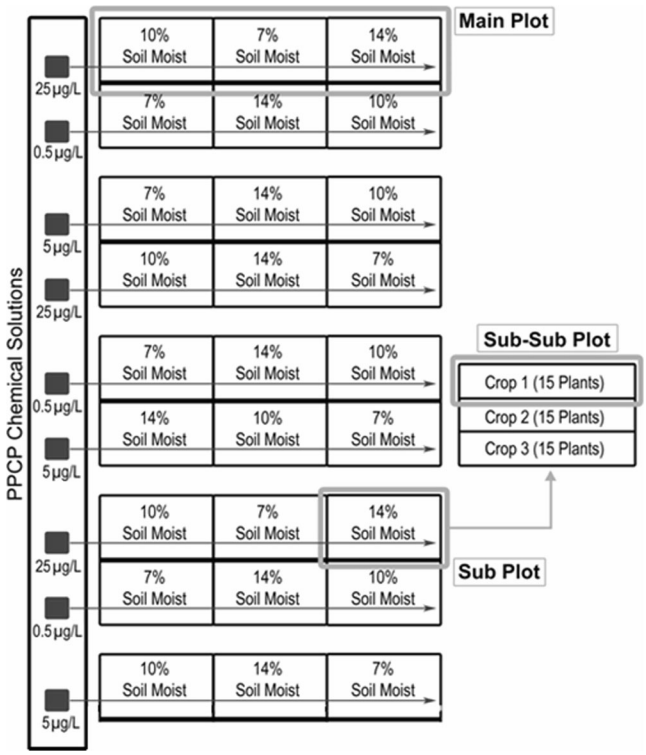


Fig. 1 Experimental design layout for the greenhouse experiments

The first experimental trial lasted a total of 98 days, with crops being harvested at 64, 84, and 98 days for cowpea, turnip, and Swiss (S.) chard, respectively. PPCP solution application and soil moisture regimes began 20 days after planting and continued until crops were harvested. Average recorded environmental parameters are as follows: air temperature, 27.5 °C; relative humidity, 61.7 %; and light intensity, 6958.13 lx. The second trial lasted a total of 92 days, with crops being harvested at 47 days for cilantro, 59 days for lettuce, 88 days for basil, and 92 days for collards. PPCP solution application and soil moisture regimes began 14 days after planting and continued until crops were harvested. Average recorded environmental parameters for this trial were air temperature, 26.3 °C; relative humidity, 68.4 %; and light intensity, 6280.74 lx.

Residual PPCP study

The residual PPCP experiment consisted of growing crops in the same soil media used for the second experimental trial, which were irrigated with 25 μg L^{-1} of PPCPs under the established soil moisture depletion regimens, without any additional PPCP inputs. The objective of this study was to

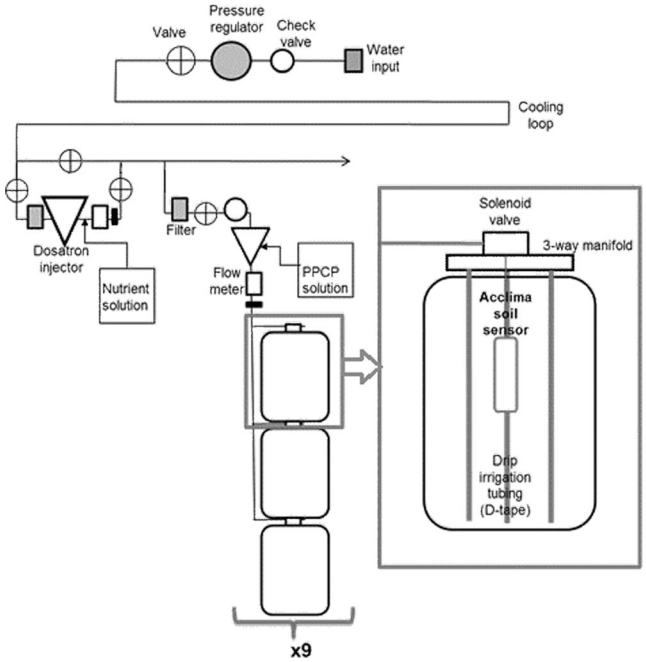


Fig. 2 Experimental setup with sensors and controls

observe the persistence of the applied PPCPs in the soil and the ability of subsequent crop cycles to take them up. The experiment was performed within a simplified factorial treatment split-plot CRD with three repetitions (Table 3). The treated crops were cilantro and S. chard.

Sample preparation and chemical analysis

Edible plant tissue was harvested, weighed, and freeze-dried using a Virtis SP Scientific benchtop lyophilizer. Finely ground plant samples (3 g cowpea or turnip; 1.5 g chard or collards; 1 g basil, lettuce, or cilantro) were used for accelerated solvent extraction (ASE). ASE cells were prepared with a cellulose filter in the bottom, followed by a spoonful (~9.6 g) of ASTM 20/30 washed Ottawa sand after which plant material was added and then the remainder of the cell was filled to the top with Ottawa sand. A methylene chloride/methanol (75/25) solution was used as the extraction solvent. The programmed extraction was set to run for three cycles at 50 °C, heating for 5 min, then at static temperature for 10 min, and followed by a 60-s purge. The total extract volume at the end of three cycles was 70–75 mL. The extract was evaporated to dryness under a stream of nitrogen. Samples were then reconstituted in 250 μL of 5 % acetonitrile for liquid chromatography mass spectrometry (LCMS) analysis.

For bean, turnip, and S. chard samples, an additional SPE cleanup step was implemented: The ASE extract was dissolved in 3 mL of methanol and then 97 mL of water was added. The solution was loaded onto a pre-wetted Waters Oasis HLB cartridge (3 mL, 540 mg) using a Fisher PrepSep vacuum manifold. The vacuum was applied using a Brinkmann B-169 vacuum aspirator so that 15–20 drops per minute were collected into the waste receptacle. Up to nine samples could be simultaneously processed by this slow drip method. After the 100 mL solution was loaded onto the cartridge, 10 mL of water was used to rinse out the bottle and tubing and was also loaded onto the cartridge. The cartridge was then transferred to the Millipore vacuum manifold and eluted with 10 mL of methanol, followed by 3 mL of methylene chloride to strip the cartridge clean. The extracts were combined and evaporated to dryness under a stream of nitrogen. Extracts were then reconstituted in 250 μL of 5 % acetonitrile for LCMS analysis.

LCMS conditions

All samples were analyzed with a Waters (Micromass) Quattro Micro API mass spectrometer using the MassLynx v4.1 software, Shimadzu SIL-HTc autosampler, and dual SPD-10AVvp HPLC pumps. The mobile phases consisted of 0.1 % formic acid in water for pump one and 0.1 % formic acid in 2 % methanol/98 % acetonitrile for pump two. The column used was a Waters XTerra MS C18 2.5 μm, 2.1 × 50 mm, fitted with a guard column and pre-filter frit. Sample injections were 20 μL; the flow rate was set at 0.250 mL min^{-1} and sample run times were 10 min each. All materials used for sample transfer were borosilicate glass or PTFE/PP to minimize adsorption.

Results

First experimental trial

LCMS results indicate that all treated crops were capable of PPCP uptake and able to transport them to their raw edible parts at ng g^{-1} (ppt) concentration levels. Plant tissue concentrations of PPCPs were consistently highest for OFL, followed by ATN and then DIC. When analyzed by applied PPCP concentrations (Table 4), the highest mean PPCP uptake values were obtained in crops irrigated with 25 μg L^{-1}, followed by those irrigated with 5 μg L^{-1} and then the ones treated with 0.5 μg L^{-1}. When comparing PPCP uptake mean values by soil moisture depletion levels, plants treated under the 14 % soil moisture regimen had the highest mean levels of all the three applied PPCPs, followed by those treated under the 10 % regimen and then those treated under the 7 % soil moisture depletion regimen (Table 4). Whereas, when comparing by plant crop, cowpea was the crop that showed the highest mean levels of all the three applied PPCPs (Table 4). Turnip showed the second highest levels for OFL and DIC, while S. chard had the second highest mean level of ATN.

Although residual levels of PPCPs were detected within the analyzed plant tissues, no significant levels of PPCP uptake were observed in response to the applied treatment variables of PPCP concentration, soil moisture depletion levels, and crop species (Table 5).

Second experimental trial

As before, all analyzed crops were capable of taking up PPCP and able to transport them to their raw edible parts at ng g^{-1} (ppt) concentration levels. Of the three applied PPCPs, OFL was the one detected in the largest amounts, followed by ATN. DIC did not seem to be taken up by any of the treated plants. When analyzed by applied PPCP concentrations (Table 4), the highest mean PPCP uptake values were obtained in crops irrigated with 25 μg L^{-1}, followed by those irrigated with 0.5 μg L^{-1} and then the ones treated with 5 μg L^{-1}. Comparing PPCP uptake mean values by soil moisture depletion levels, plants treated under the 10 % soil moisture regimen had the highest mean levels of OFL. ATN was detected at the same amounts for plants treated under the 10 and 7 % soil moisture regimens. Plants treated below the 14 % soil moisture regimen had the second highest mean values for OFL and ATN (Table 4). Detected DIC levels were virtually nonexistent for all treatments. When comparing by plant crop species (Table 4), lettuce was the crop that showed the highest mean levels of ATN and OFL, followed by basil. Cilantro had the third highest detected mean value for OFL uptake, and the least high for ATN. Collards showed the third highest detected mean value for ATN uptake and the least high value for OFL uptake.

No significant effects of PPCP uptake were observed in response to the applied treatment variables of PPCP concentration, soil moisture depletion levels, or crop species (Table 5).

Residual PPCP study

Crops within this trial were harvested at 47 days after planting. Based on LCMS analyses, the treated crops were capable of residual PPCP uptake at the ng g^{-1} (ppt) concentration level. Just as in the previous experimental trials, OFL was the compound detected in the largest amounts, followed by ATN and then DIC. When analyzed by applied soil moisture depletion rates (Table 4), the highest mean OFL values were found in crops treated at the 14 % soil moisture depletion rate, followed by those treated at 7 % and then those treated at 10 %. The highest ATN concentrations were found in crops irrigated under the 10 % soil moisture depletion regime, followed by those under the 14 % regime and then those under the 7 % regime. DIC was again detected in very low concentrations with the highest values being detected in crops treated at 7 %, then those under 14 %, and finally those under the 10 % soil moisture depletion treatment. When distributed by crop type, higher levels of all three PPCPs were detected in S. chard (Table 4). However, none of these values were of significance when analyzed with the developed ANOVA model (Table 5).

Discussion

Our results suggest that applied treatments did not significantly alter or hinder the transpiration pathways of the treated crops, allowing them to freely uptake the applied PPCPs and implying that PPCP uptake is likely attributable to transpiration and mass flow transport. This coincides with results obtained by Briggs et al. (1982), Chiou et al. (2001), Su and Zhu (2007), Winker et al. (2010), Karnjanapiboonwong et al. (2011), and Shenker et al. (2011), where they also determined that the plant's transpiration pathway is the main driver in the uptake, transport, and translocation of plant-available soil compounds. Our results also coincide with the theoretical framework of transpiration-driven uptake (Kramer 1945; Slatyer 1960; Kramer et al. 1967) where, if soil moisture content is retained at favorable levels, plants will maintain their stomata open, maximizing photosynthesis, and soil water absorption will occur at elevated levels due to increased transpiration rates. Subsequently, as more water moves through the plant, more photosynthesis occurs, more biomass is allocated, and there is a higher probability that higher levels of the dissolved PPCPs will be taken up, transported, and retained in plant tissue due to this continuous flux of water.

It appears that treated plants take up PPCPs at ng g^{-1} (ppt) levels independently of applied soil moisture or added drug concentrations. Even though the applied soil moisture depletion regimens did not significantly affect crop uptake of PPCPs, during the first experimental trial, a slight trend can be observed in which PPCP uptake increases with added soil moisture (Table 4). Also, within this experimental trial, another incremental tendency can be observed in terms of applied PPCP solution concentrations, where there were higher levels of uptake in crops irrigated with higher concentrations (Table 4). These trends suggest that higher levels of applied PPCP concentrations and increased irrigation would result in higher levels of PPCPs. However, these trends were not observed in the second experimental trial or in the additional treated crops, where uptake tendencies were irregular and not defined by any of the applied treatments (Table 4). Of the three applied PPCPs, OFL was detected in the highest amounts, followed by ATN and then DIC. DIC was not detected in many of the analyzed samples, although detected levels are similar to those reported in previous hydroponic studies (Dodgen et al. 2013). In terms of crop effect, no distinct trend was identified and detected levels for crop edible parts were fairly consistent throughout all experimental trials,

Table 4 Detected mean PPCP amounts in plant tissue distributed by experimental variable

Experimental variable	Experimental trial								
	1			2			Residual		
	Detected mean PPCP amounts in plant tissue (ng g^{-1})								
	ATN	DIC	OFL	ATN	DIC	OFL	ATN	DIC	OFL
PPCP concentration (μg L^{-1})									
0.5	9.50	0.53	16.94	3.88	0.01	23.29	–	–	–
5	11.93	0.99	32.86	2.70	0.00	17.02	–	–	–
25	37.01	5.17	65.69	3.89	0.01	26.06	–	–	–
Soil moisture (%)									
7	5.93	0.49	16.40	3.53	0.01	20.73	4.13	0.10	37.07
10	13.59	0.89	22.69	3.53	0.01	23.55	5.36	0.02	30.53
14	36.00	4.83	71.42	3.42	0.00	22.09	4.35	0.08	41.07
Crop									
Cowpea	23.00	4.25	59.52	–	–	–	–	–	–
S. Chard	13.70	1.00	30.57	–	–	–	4.26	0.07	39.60
Turnip	19.72	1.12	22.68	–	–	–	–	–	–
Cilantro	–	–	–	2.03	0.00	16.08	4.84	0.07	32.84
Lettuce	–	–	–	5.51	0.00	37.82	–	–	–
Basil	–	–	–	3.74	0.02	21.41	–	–	–
Collards	–	–	–	2.70	0.00	13.19	–	–	–

further suggesting that PPCP uptake is driven by the transpiration process and the factors that influence it.

Furthermore, consistent with previous research, our results indicate that the applied PPCPs have the potential to persist in sand media during and between cropping cycles. This is most likely due to their gradual release from being adsorbed to soil particles and decomposing organic matter, especially in the case of ATN and OFL. ATN has been reported to have a high soil-sorption affinity (Yamamoto et al. 2009; Burke et al. 2013) and interact with soil organic matter (Chefetz et al.

Table 5 Summary statistics of main fixed effects on PPCP uptake

Fixed effects	*df*	ATN uptake		DIC uptake		OFL uptake	
		First experimental trial					
		F value	*p* value	*F* value	*p* value	*F* value	*p* value
PPCP concentration	2	2.46	0.1273	1.51	0.3251	2.06	0.2427
Soil moisture level	2	5.28	0.0755	1.70	0.2238	2.81	0.0998
Crop	2	0.46	0.6329	0.46	0.6383	0.43	0.6519
		Second experimental trial					
		F value	*p* value	*F* value	*p* value	*F* value	*p* value
PPCP concentration	2	1.19	0.3926	2.29	0.2178	0.93	0.4668
Soil moisture level	2	0.002	0.9980	1.75	0.21530	0.02	0.9802
Crop	3	1.87	0.1456	1.69	0.1802	2.24	0.0940
		Residual experimental trial					
		F value	*p* value	*F* value	*p* value	*F* value	*p* value
Soil moisture level	2	0.00	0.9958	1.12	0.3867	0.11	0.8962
Crop	1	3.46	0.1124	0.11	0.7502	0.61	0.4653

2008; Yamamoto et al. 2009). OFL has been seen to adsorb to silica due to the formation of electrostatic repulsion forces (Goyne et al. 2005) and interact with mineral and organic soil particles (Pan et al. 2012; Zhou et al. 2014), whereas DIC has demonstrated mobility in soil while having low sorption coefficients (Scheytt et al. 2005; Xu et al. 2009), possibly explaining its low detection amounts in the residual study.

Results from the residual PPCP study are of particular interest from an agricultural and environmental management perspective, as they suggest that the evaluated compounds can continuously accumulate in the soil by means of irrigation residue and belowground plant biomass incorporation, with the potential to be further absorbed by subsequent crop cycles and/or be transported across the landscape. This low-exposure and long-term permanence of PPCPs within the soil could have further ecological consequences, such as evolving into an energy source by bacteria prompted to adapt to these novel and readily available inputs, as in the case of atrazine (Rousseaux et al. 2001; Vibber et al. 2007; Sagarkar et al. 2014). Although the PPCPs in our study were detected in extremely low amounts when compared to the amounts of atrazine already present in agricultural systems, PPCPs could follow a similar trend in the future, providing alternate energy sources to soil microorganisms and potentially causing significant changes in the soil microflora, which could then expand to more ample ecological changes.

Finally, there was an overall noted variability among detected PPCP values in relation to treatment means. This variability could be attributed to parameters not considered within the experiments, such as biotic activity within the soil, crop-specific physiological pathways and anatomical structure, and PPCP compound degradation or transformation throughout the irrigation process.

Conclusion

Results from our study indicate that the treated plant crops were capable of PPCP uptake into their raw edible parts at ng g^{-1} (ppt) concentration levels, independent of the prescribed soil moisture depletion regimens and the differing levels of applied PPCP concentrations. The uptake of these PPCP compounds can be attributed to the transpiration process and mass flow transport, not root-exposed concentration. OFL was the studied compound detected at the highest levels, followed by ATN and DIC, which were not detected in all analyzed plant tissues. PPCPs were also detected in nonedible plant root tissues and in crops grown on soil previously irrigated with PPCPs (residual experiment). The results from these studies pose concerns in terms of agricultural management and the prevalence of these compounds in the soil and throughout growing cycles. Even though the dietary uptake of these PPCP by humans seems to be negligible, as the detected amounts are significantly lower than reported therapeutic doses (HSDB 2015), the ecological concerns of a continuous input of these compounds could potentially lead to unwanted outcomes, such as becoming a readily available energy source for soil microorganisms or accumulating in off-site aquatic sources due to run-off, provoking a chain reaction of further ecological change.

Our results also suggest that by reducing water usage in a coordinated manner and tapping into unconventional water resources for expanded applications, overall water productivity can be improved without significantly compromising agricultural yields; however, the potential long-term ecological implications of using such resources should be taken into consideration. Finally, there are many variables our study did not assess, such as soil biotic interactions and other factors apart from soil moisture that directly influence plant transpiration. Therefore, there still exists a huge potential to further investigate, not only the uptake of PPCPs by plants but also the many still unknown ecological effects and interactions of employing RW for irrigation purposes.

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