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Transfer of wastewater associated pharmaceuticals and personal care products to crop plants from biosolids treated soil

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ABSTRACT

The plant uptake of emerging organic contaminants such as pharmaceuticals and personal care products (PPCPs) is receiving increased attention. Biosolids from municipal wastewater treatment have been previously identified as a major source for PPCPs. Thus, plant uptake of PPCPs from biosolids applied soils needs to be understood. In the present study, the uptake of carbamazepine, diphenhydramine, and triclocarban by five vegetable crop plants was examined in a field experiment. At the time of harvest, three compounds were detected in all plants grown in biosolids-treated soils. Calculated root concentration factor (RCF) and shoot concentration factor (SCF) are the highest for carbamazepine followed by triclocarban and diphenhydramine. Positive correlation between RCF and root lipid content was observed for carbamazepine but not for diphenhydramine and triclocarban. The results demonstrate the ability of crop plants to accumulate PPCPs from contaminated soils. The plant uptake processes of PPCPs are likely affected by their physico-chemical properties, and their interaction with soil. The difference uptake behavior between plant species could not solely be attributed to the root lipid content.

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1. Introduction

Wastewaters are generated worldwide by anthropogenic activities, and are commonly treated in wastewater treatment facilities utilizing different processes. During the biological treatment of wastewater using the activated sludge process, large amounts of sewage sludge are unavoidably generated as a byproduct, leading to a need for appropriate and safe disposal. For health and safety purposes, sewage sludges are usually treated to meet certain regulatory criteria before disposal. Treated sewage sludges, commonly referred to as biosolids, are rich in nutrients and organic matter and can thus be used to amend soils, improving soil structure and fertility (USEPA, 1999). In the United States, over seven million tons of biosolids were generated in 2004, of which about 55% were land applied in agricultural settings (NEBRA, 2007).

The land application of biosolids provides a waste management route for sewage sludge, and in tandem amendment to the receiving soils. Although benefits exist, such as increase in organic matter and nutrients, biosolids land application has been shown to possibly increase nutrient loss to surface waters when applied beyond the beneficially calculated amounts (Tian et al., 2006; Singh and Agrawal, 2008). Also as a by-product, increases in pollutant load in the receiving soils for many wastewater associated contaminants, such as pathogens (Zaleski et al., 2005), heavy metals (McLaren et al., 2007), and toxic organic pollutants (Gottschall et al., 2010) has been documented. These pollutants, once in soils, could potentially be translocated to crop plants grown in the contaminated soil, potentially causing damage to the plants, and/or entering the food chain. The consumption of contaminated crop plants might increase both human and livestock exposure to those pollutants (Zohair et al., 2006; Zhang et al., 2007; Sridhara Chary et al., 2008).

Plant uptake and soil persistence from biosolids-amended soils of heavy metals and organic pollutants such as pesticides and persistent organic pollutants (POPs), have long been documented (Wang and Jones, 1994; O'Connor, 1996; Benítez et al., 2001; Lavado et al., 2007). More recent research suggests that a group of emerging contaminants known as pharmaceuticals and personal products (PPCPs) could also be transferred into plants as a result of biosolids reuse (Wu et al., 2010a; Eggen et al., 2011; Karnjanapiboonwong et al., 2011). PPCPs comprise a variety of chemicals differing in physico-chemical properties. Many PPCPs are ionizable and dissociation in soil solution may greatly affect their plant uptake behavior. This effect is difficult to predict as

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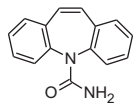
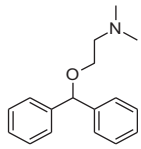
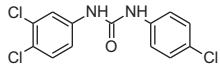
Table 1
Structures and selected properties of tested compounds.

Compound (application)	Solubility* (mg L^{-1}, °C)	Structure	pK_a	Log K_{ow}	Half-life in soil (day)
Carbamazepine (anticonvulsant)	17.66		2.3 (Nghiem et al., 2005)	2.45 (Nghiem et al., 2005)	495 (Walters et al., 2010)
Diphenhydramine (antihistamine)	362.7		9.08 (Box and Corner, 2008)	3.44 (Box and Corner, 2008)	> 1000 (Walters et al., 2010)
Triclocarban (antimicrobial)	0.65		12.7 (Loftsson et al., 2005)	4.9 (Loftsson et al., 2005)	108 (Ying et al., 2007)

* Calculated values using U.S. Environmental Protection Agency EPI Suite V 4.10.

chemicals can behave differently in ionized form from their neutral form. Thus it is important to understand these processes for a better prediction of the behavior of PPCPs.

Organic contaminant uptake capacities for different plant species could vary widely. Lipids content has been found to affect the root uptake of hydrophobic organic compounds (Simonich and Hites, 1995). Positive correlations between plant lipid contents and root concentration factors (RCF) have been observed in previous research (Schwab et al., 1998; Gao and Zhu, 2004). This relation is a result of hydrophobic partition governing the root uptake of hydrophobic organic compounds. For ionizable compounds, this relation may not hold.

The objective of this study was to quantify the uptake and translocation of carbamazepine, diphenhydramine and triclocarban, introduced through biosolids application to a soil, by five economical crop plants. Plants were selected to include fruit bearing, leafy greens and tuber varieties. The impact of both physico-chemical properties and plant root lipid contents on the plant uptake processes are discussed.

2. Materials and methods

2.1. Chemicals and materials

Analytical grade standards of carbamazepine (CBZ), diphenhydramine (DIP), triclocarban (TCC) and josamycin (internal standard) were purchased from Sigma-Aldrich (St. Louis, MO). Carbamazepine-D_{10}, diphenhydramine-D_5 and triclocarban-D_4 were purchased from Cambridge Isotope Laboratories (Andover, MA). Chemicals and solvents were certified ACS or HPLC grade and were purchased from Fisher Scientific (Fair Lawn, NJ). Deionized water (> 18.0 MΩ cm) was provided by a NANOpure Infinity Ultrapure Water System (Barnstead, Dubuque, IA). All prepared stock standard solutions were stored at −20 °C.

The soil used as the growing medium was an engineered soil product produced from a mixture of aged sediment dredge material from a confined open water storage facility, dewatered aged biosolids from a local WWTP and waste lime from a local fresh water treatment plant, referred to as NU-Soil (Whitehouse, OH). This soil was chosen based on availability at the research site and the presence of previous contamination derived from wastewater treatment. The soil was composed of 51% sand, 21% silt and 28% clay according to the USDA pipette analysis procedure, and is texturally classified as a sandy clay loam. Soil pH was 7.6 ± 0.1. Soil organic matter content (measured as loss on ignition at 360 °C for 2 h) was 3.0 ± 0.2% and cation exchange capacity was 30.4 ± 1.1 meq/100 g (determined by Brookside Laboratories, Inc, New Knoxville, OH).

Aerobically treated biosolids, meeting U.S. Environmental Protection Agency (EPA) 40 CFR Part 503 Rule requirements for Class B pathogen reduction were collected from a local wastewater treatment plant (WWTP) in May of 2010. Detailed information on this WWTP has been described previously by Wu et al. (2008). Percent solids were measured gravimetrically and were 34 g L^{-1} of solids. Background concentrations in the biosolids of selected compounds were not measured, but our previous data show that the concentrations of CBZ, DIP, and TCC in biosolids from the same WWTP range from 0.066 to 0.081 μg g^{-1} for CBZ, 0.788 to 0.9 μg g^{-1} for DIP, and 9.35 to 13.33 μg g^{-1} for TCC (Wu et al., 2010a).

2.2. Experimental setup

Plant uptake experiments were carried out in the summer of 2010 at the Stranahan Arboretum, a University of Toledo field research station. Biosolids were applied to the soil representing a typical agricultural application rate of 1.04 kg $(m^2)^{-1}$ (L1 treatment). In addition a biosolids-free (L0 treatment) and biosolids-applied plant-free control were also included. All treatments contained three replicate containers. CBZ, DIP, and TCC were selected for the study due to their high environmental stability and previously detailed potential to accumulate in plants (Wu et al., 2010b). Their selected properties are listed in Table 1. Prior to biosolids application a 1000 mg L^{-1} PPCP standard solution was prepared in methanol and thoroughly mixed with the liquid biosolids for each replicate in treatment L1 and plant-free control. Fortified biosolids for each replicate were then incorporated into the soil one at a time by thoroughly mixing using hand shovels. The biosolids/soil was mixed at a ratio of 1:7.3 in order to match the target application rate. For plant growth, two container sizes, both cylindrically shaped, were used. Large 23 L containers (28.5 cm diameter, 35.5 cm length) and smaller 10 L containers (24.0 cm diameter, 23.0 cm height) were filled evenly with either 20 or 8 L of soil, based on container size, Nominal concentrations for individual compounds, without accounting for background residual, were 21.6 μg g^{-1} in biosolids and 100 ng g^{-1} in biosolids amended soil. The biosolids were fortified with standards to ensure the detection. The concentrations used here are comparable to those detected on agricultural fields receiving biosolids application for those compounds (Gottschall et al., 2012; Xia et al., 2010). After biosolids application soil cation exchange capacity increased to 34.4 ± 1.2 meq/100 g, organic matter content increased to 3.8 ± 0.1%, and soil pH decreased to 7.5 ± 0.1.

Five plant species were examined in the plant uptake experiment. Young plants and seeds were purchased from The Andersons Store Nursery Center (Toledo, OH). The selected plants were pepper (*Capsicum annuum*, Hungarian wax hot variety), tomato (*Lycopersicon esculentum*, Supersweet 100 variety), collard (*Brassica oleracea*, Vates variety), lettuce (*Lactuca sativa*, Buttercrunch variety) and radish (*Raphanus sativus*, White icicle variety). Young plants of pepper and tomato were transplanted into the larger containers, while young plants of collard and lettuce were transplanted into the smaller containers. For these four species, one plant was planted in each container. Radish was planted from seed directly into the smaller containers. Ten seeds per container were initially planted, then thinned to three once sprouted. Plants were grown outdoors under a transparent roof and were irrigated according to each plant need using groundwater from a local shallow aquifer. The groundwater was found to not contain any of the targeted contaminants above the instrumental detection limits. Irrigation volume was kept consistent for each plant/replicate/treatment set. No attempt was made to collect leachate. All plants were allowed to reach maturity and bear fruit if applicable. Tomato and pepper plants were harvested 54 day after planting, while the collard, radish and lettuce were harvested 29 day after planting.

2.3. Sample preparation and analysis

2.3.1. Harvest and pretreatment

At harvest, root tissue and soil were collected for each treatment/plant/replicate after first harvesting aboveground biomass. Initial sub sampling was

accomplished by emptying each container onto a tarp and thoroughly mixing the contents by trowel. Samples from various locations were then collected in order to fill a 250 mL container. Soil samples were then freeze-dried using a using a Labconco Freezone 6 freeze-drier (Kansas City, MO), crushed and homogenized using a mortar and pestle and sieved through a 2-mm brass sieve. Above ground biomass was collected for each treatment/plant/replicate/part combination at harvest. Within each combination, the fruit and shoots (stems and leaves) were collected in the field, then cleaned with nanopure water, patted dry with paper towel and the biomass weight recorded. All plant tissues collected were then cut into smaller pieces, freeze-dried, and ground and homogenized using an IKA analytical mill (Wilmington, NC). All prepared samples were stored at −20 °C until analysis.

2.3.2. PPCPs extraction, clean up and LC-MS/MS analysis

After spiking with surrogate standards, plant and soil samples were extracted using pressurized liquid extraction (ASE) using a Dionex ASE200 system (Sunnydale, CA). Extracts were then further cleaned and concentrated using Biotage Evolute ABN solid phase extraction (SPE) cartridges (Charlottesville, VA). Final extracts were then analyzed using a Varian 1200L (Walnut Creek, CA) liquid chromatography-tandem mass spectrometer with electro-spray ionization interface (LC-ESI-MS/MS) and were separated using a Phenomenex Luna C8(2) column (Torrance, CA). Quantification was achieved using an internal standard calibration curve and adjusted with recoveries of isotope labeled surrogate standard. Complete details of extraction procedures, instrumental analysis and quantification are described in the Supporting Information.

2.3.3. Total lipid analysis

Total lipids were analyzed using a gravimetric method following a pressurized liquid solvent extraction as described in (Moreau et al., 2003). Briefly 11 mL ASE extraction cells were packed with 1–2 g dried ground plant material and Ottawa sand (Fisher Scientific), and capped with cellulose filters. Cells were then extracted using hexane as the extract solvent with the following parameters: 1000 psi pressure, 100 °C oven temp, 5 min heating ramp, 10 min static cycle, 3 cycles, 120 s purge, 60% flush. The vial volume was then measured, dried in oven at 105 °C for 4 h, cooled and weighed. Total lipids were calculated as the mass of material left after drying for 4 h relative to the total sample mass.

2.4. Statistical analysis

Student *t*-tests were used to test the significance of differences between the treatments. Pearson correlation analysis was used to test the relationship between root concentration factors and root lipid contents. Statistical analyses were performed using Excel 2007 or SPSS Ver. 17.0 (IBM, Armonk, NY).

3. Results and discussion

The concentrations of selected PPCPs in harvested plant tissues for biosolids-amended soils (L1) are presented in Fig. 1. Selected compounds were detected in all plant tissues with concentrations ranging from 4.8 to 1287 ng g^{-1}. TCC had the highest root concentrations in all plants followed by CBZ and DIP. The high root concentrations of TCC can be attributed to the high TCC background residuals in the soil (768 ± 86 ng g^{-1}), providing for increased availability. In the shoots, CBZ had the highest concentrations, suggesting translocation potential from root to above ground tissues for CBZ is the highest. However, the concentrations of CBZ in the fruits of pepper and tomato were relatively low. Similar results were observed in previous studies of the soybean plant (Wu et al., 2010b). This observed trend suggests that the translocation of CBZ from stem to fruit may be limited. More likely CBZ might undergo degradation in the fruit. Metabolization of CBZ by *Typha spp.* has been previously observed, with CBZ metabolites being detected in leaf tissues (Dordio et al., 2011). Recently, degradation of pharmaceuticals (ibuprofen, naproxen, clofibric acid, etc.) in spath and lettuce was also observed, likely due to the plant detoxification systems (Calderón-Preciado et al., 2012).

To evaluate the transport of the compound of interest into the plant tissues from amended soil, root concentration factors (RCF) and shoot concentration factors (SCF) were calculated. RCF and SCF were calculated as the ratio of concentration at harvest in root to that in soil and that in shoot to that in soil, respectively

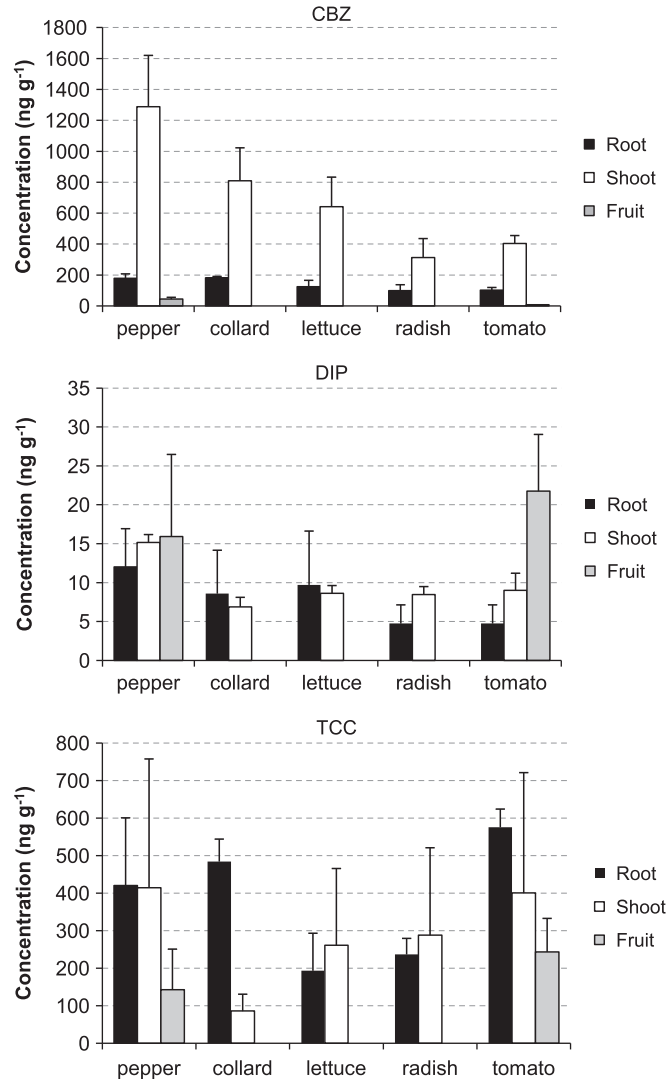


Fig. 1. Concentrations of selected PPCPs in tissues harvested from plants grown in biosolids amended soil (L1 treatment). Bars indicate mean and standard deviation (n=3). Fruit data is analyzed only for pepper and tomato plants.

Table 2
Root concentration factors (RCF) and shoot concentration factors (SCF) of selected PPCPs in different plants from L1 treatment (mean ± standard deviation, n=3).

Compound	Plant type				
	Pepper	Collard	Lettuce	Radish	Tomato
RCF					
CBZ	3.34 ± 1.20	1.62 ± 0.37	1.66 ± 0.36	1.12 ± 0.50	1.06 ± 0.14
DIP	0.18 ± 0.09	0.06 ± 0.04	0.06 ± 0.04	0.03 ± 0.02	0.23 ± 0.21
TCC	0.73 ± 0.24	0.67 ± 0.08	0.34 ± 0.26	0.31 ± 0.10	0.72 ± 0.10
SCF					
CBZ	23.37 ± 7.34	8.28 ± 4.00	7.42 ± 1.51	3.42 ± 1.03	4.16 ± 0.39
DIP	0.22 ± 0.03	0.03 ± 0.00	0.05 ± 0.01	0.05 ± 0.01	0.07 ± 0.04
TCC	0.73 ± 0.56	0.12 ± 0.11	0.25 ± 0.21	0.36 ± 0.27	0.48 ± 0.36

(Table 2). In general RCF values for all three compounds were greatest for pepper plants, followed by collard and lettuce, then radish and tomato, with the exception of DIP and TCC in tomato. SCF followed similar trends. The RCF and SCF values for CBZ were found to be the highest, followed by TCC then DIP. The low RCF values observed for DIP are likely attributed to the ionization. DIP is a weak base with a pK_a value of 9.08 (Box and Corner, 2008).

Thus DIP exists primarily as a cation in the biosolids amended soil (pH 7.5). Previously, model results have shown that the ion trap effect keeps moderate bases out of the root cell (Trapp, 2009). In addition, cations can strongly bond in soils by ion exchange, possibly indicated by the strong adsorption observed for this soil for DIP ($K_d > 1000$ L kg^{-1}, unpublished data from our lab). At pH 7.5, CBZ and TCC exist in their neutral from. Thus, their accumulation in roots is mainly governed by the hydrophobic partition process. The SCF is at least three times higher than the RCF for CBZ in all plants, suggesting readily available transport from root to shoot, possibly through plant transpiration stream-flow. Previously, the uptake of hydrophobic compounds by plants was found to follow a bell-shaped curve, with higher uptake efficiency for moderately hydrophobic compounds (Briggs et al., 1987; Hsu et al., 1990). The Log octanol–water partitioning coefficients (Log K_{ow}) for CBZ, DIP, and TCC are 2.45, 3.44, and 4.9, respectively (Table 1). Their ability to translocate into plants generally agrees with previous observations for hydrophobic compounds. The relatively high Log K_{ow} of TCC might limit transport from roots to shoots (Briggs et al., 1982). Whereas, DIP is ionized in soil and thus the root uptake of DIP is kinetically limited. As a result the shoot uptake should be limited by the concentration of DIP in root tissues.

In our previous research investigating the uptake of PPCPs by soybean, the RCFs for CBZ, DIP and TCC were 2.8, 0.39 and 2.0, respectively (Wu et al., 2010b). These RCFs are higher that those from this current study. This can mainly be attributed to the differences in the soils used in the two experiments. In this study, the soil is less sandy and has a high organic matter content (3.0% vs. 2.7% in the previous study). Herklotz et al. (2010) studied uptake of carbamazepine, salbutamol, sulfamethoxazole, and trimethoprim in cabbage and Wisconsin Fast Plants using hydroponic systems. All compounds were detected in plant tissues and carbamazepine and salbutamol were found in the seedpods. For cabbage, RCF and SCF ranged from 7.037 to 10.92 and from 0.0451 to 0.0805, respectively. Their CBZ RCF for cabbage is much high than results from this study, likely because nutrient solution was used for the experiment, in which no adsorption occur. Interaction of compounds with soil can significantly reduce the plant uptake potential. However, SCF is much lower than our current study, suggesting that the translocation of CBZ from root to aboveground in cabbage may be limited. Shenker et al. (2011) studied the uptake of CBZ by cucumber irrigated with reclaimed wastewater. CBZ was primarily accumulated in the leaves but the concentration was relatively low in fruits, agreeing well with the results found in our study.

Root lipid content has previously been found to be a good indicator of the root uptake ability of different plant species for many hydrophobic compounds. Gao and Zhu (2004) studied the uptake of phenanthrene and pyrene by 12 different plant species in treated soil and found significantly positive correlations between root concentration factors (RCFs) and plant root lipid contents. Huang et al., 2009 studied the uptake of Decabromodiphenyl ether (BDE-209) by ryegrass, alfalfa, pumpkin, squash, maize and radish. The root concentrations of BDE-209 were also found positively related to plant root lipid contents. Here the RCFs of tested PPCPs from L1 treatment are plotted against root lipid contents in Fig. 2. Positive correlation was only observed for CBZ ($p < 0.005$), while no significant correlation was observed for DIP and TCC. This relationship indicates that hydrophobic partition primarily controls the root uptake of CBZ. This is likely due to the factor that CBZ exists in its neutral form in the soil. TCC also exists in its neutral form but no statistically significant relation was observed. This may be attributed to its high hydrophobicity and low water solubility (0.65 mg L^{-1}). Root uptake of TCC can be kinetically limited and equilibrium may

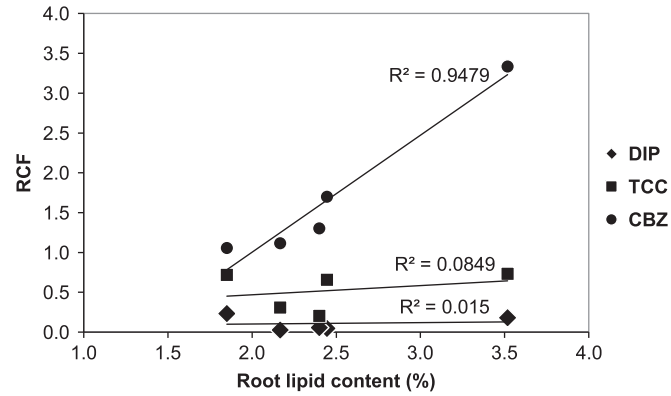


Fig. 2. Relationship between root concentration factors (RCF) and root lipid content across all plant species studied.

have not reached between the soil and plant roots. Relatively water-insoluble compounds less efficiently approach equilibrium than water-soluble ones (Chiou et al., 2001; Li et al., 2005). No positive correlation between RCF and root lipid content for DIP, likely because it is fully dissociated in the soil (ionized), indicating that root lipid content may not be a determining factor for root uptake of ionized compounds. Based on this dataset, lipid contents may not be a good predictor of the root accumulation of PPCPs, especially for those ionized in the substrate.

The concentrations of selected PPCPs in soils applied with biosolids were determined before plants were seeded and after harvest (Table 3). The concentration of TCC is considerably higher than spiked theoretical values. This is due to the high background residual in the biosolids/soil mixture, particularly the residual found in the NU-soil before biosolids application (1.4 ± 0.2, 30.1 ± 2.1 and 768 ± 86.1 ng g^{-1}, respectively for CBZ, DIP and TCC). The soil used (NU-soil) is a mixture of dried aged biosolids, aged sediments and waste lime. The background residuals are likely coming from both the aged biosolids and dredge sediment and the biosolids used for application in this experiment. Previously, TCC has been detected in sediment with concentrations varied from 0.7–1.6 μg g^{-1} (Miller et al., 2008) and in the biosolids used for application in this study (13.33 μg g^{-1}, Wu et al., 2010a). After harvest, the concentrations of CBZ dropped 21.5% and 39.5% in soils with radish and tomato, respectively. The concentrations of DIP and TCC dropped 62.5% and 41.3%, respectively, in soils growing pepper. No decreasing trend was observed for other treatments. This suggests that all three compounds are very persistent in the soil, which agrees with previous research (Ying et al., 2007; Walters et al., 2010). Plants may accelerate dissipation of chemicals by absorption from contaminated soils. However, the amount of tested compounds in the plants accounts for $<1\%$ of initially added mass. Thus, the loss due to plant uptake is negligible in this study. Here, biodegradation, or possible irreversible sorption, are more important process affecting their dissipation in these treatments. The role of plant species and the effect on soil microbial activities may play an important rule, but was beyond the scope of this study.

The effect of biosolids application on the different plants was also evaluated using biomass change. The aboveground biomass for the different plant species in the L0 (no application of biosolids) and L1 treatments (biosolids application) is presented in Table 4. For lettuce and radish the biomass changes are significantly higher from L0 to L1 treatment ($p < 0.05$, $n = 3$). This is likely due to the beneficial nutrient amendment provided to the soil by the biosolids application. However, no statistically significant difference was found for pepper, collard and tomato plants, suggesting that the differing plant species react differently

Table 3
Concentrations (ng g^{-1}, dry weight) of selected PPCPs in L1 treatment soils before planting and after harvest (mean ± standard deviation, $n=3$).

Compound	Plant type					Plant-free control
	Pepper	Collard	Lettuce	Radish	Tomato	
Pre seeding or pre planting period						
CBZ	89.3 ± 8.95	132 ± 30.7	103 ± 25.4	115 ± 9.54*	160 ± 12.0*	94.7 ± 13.7
DIP	182 ± 22.9*	166 ± 42.1	159 ± 16.6	148 ± 12.6	104 ± 9.63	157 ± 10.7
TCC	957 ± 48.5*	901 ± 157	935 ± 61.0	887 ± 51.5	942 ± 81.8	820 ± 88.0
Post harvest period						
CBZ	60.5 ± 31.8	109 ± 17.6	98.3 ± 7.95	90.3 ± 12.0*	96.8 ± 4.36*	90.0 ± 3.76
DIP	68.2 ± 5.55*	188 ± 19.5	170 ± 1.96	188 ± 46.5	136 ± 51.7	168 ± 13.6
TCC	562 ± 123*	742 ± 133	932 ± 74.0	785 ± 103	810 ± 124	733 ± 94.9

* Significant difference at 95% confidence interval.

Table 4
Aboveground biomass ((g), fresh weight) of different plant types from L0 and L1 treatment (mean ± standard deviation, $n=3$).

Treatment	Plant type				
	Pepper	Collard	Lettuce	Radish	Tomato
L0	475.27 ± 17.62	100.07 ± 8.11	143.30 ± 7.21*	64.43 ± 3.50*	757.43 ± 16.03
L1	399.43 ± 58.65	113.67 ± 18.35	218.35 ± 12.94*	100.47 ± 12.24*	653.57 ± 153.08

* Significant difference at 95% confidence interval.

to the biosolids addition. The current data does not allow for determination of toxic effects due to the addition of PPCP.

4. Conclusions

As a group of emerging contaminants, the plant uptake of PPCPs has not been well studied, but is receiving growing concern due to their potential toxic effects. Results from this study and previous researches show that PPCPs of varying classes and chemical properties could potentially translocate from contaminated soils to plants. A recent field study has demonstrated that under normal farming conditions, 8 out of the 141 PPCPs were detected in vegetables grown in the biosolids amended soil, most in the lower ng g^{-1} range (Sabourin et al., 2012). This shows that even in actual field conditions, human and livestock exposure to PPCPs by way of the consuming of crops grown on biosolids land-applied fields is possible, especially when heavily contaminated biosolids are used as amendments. Existing data also show that the uptake behavior of some PPCPs might differ from that of hydrophobic organic contaminants. This is likely due to the ionization character of many PPCPs. Speciation and sorption by hydrogen binding or complexing with soils may greatly affect their plant uptake processes. Future studies are necessary to further characterize the impact of these mechanisms in order to fully understand the transport and fate of PPCPs from source to ecosystem.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.ecoenv.2012.08.007.

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