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Effect of Planting Covers on Herbicide Persistence in Landscape Soils

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Recent monitoring shows that the majority of urban streams in the United States are contaminated by pesticide residues, and the contamination is mainly due to runoff from residential landscapes. In this study we evaluated the effect of landscape planting on persistence of the herbicides 2,4-D and dicamba in soil under laboratory conditions. The herbicides exhibited substantially different persistence in the same soil type that had been subjected to different planting practices for about 6 years. In the 0−10 cm surface layer, the half-life of 2,4-D was 30.7 d in soil under trees, which was about 20 times longer than in soil planted with turf grass (1.6 d). The difference in 2,4-D persistence was closely correlated to the number of 2,4-D-degrading bacteria that had evolved in the soils. The half-life of dicamba was much longer in soil under a tree canopy (149 d) than in mulched soil (7.9 d). The rate of dicamba degradation was proportional to soil organic matter content. This study indicates that planting practices can modify soil chemical properties and microbial activity and may further affect pesticide runoff potential by influencing pesticide degradation. Characterizing pesticide behavior as a function of planting covers may improve our understanding of pesticide runoff in urban environments and also help to identify strategies for minimizing pesticide contamination to urban streams.

Introduction

In the United States, home lawns occupy 20−25 million acres or 8−10 million ha (*1*). Given that vast expansion has occurred over the past decade and that many residential landscapes also contain nonturf systems (e.g., shrubs, ground covers, mulches, trees, etc.), the total urban landscaped area is very large. For instance, the total area of environmental horticulture in California was estimated to be 1.4 million acres (*2*). Residential landscapes serve as the direct target of pesticides applied to home lawns and gardens and the first-tier buffer for pesticides applied around structures. However, pesticide use in residential settings has apparently led to contamination of urban streams. Surveys by the U.S. Geological Survey (USGS) have shown that 99% of the tested urban streams contain at least one pesticide, with 70% containing five or more pesticides (*3*). The presence of pesticides at trace levels may cause short- or long-term impairments to aquatic ecosystems, such as toxicity (insecticides) or phytotoxicity (herbicides) (*4, 5*). To protect water quality, regulations such as Total Maximum Daily Loads (TMDLs) are being developed for many surface waterbodies in the United States (*6*).

Source analysis and monitoring studies have shown that movement of pesticides from residential areas is mainly associated with stormwater runoff (*6*−*8*). For instance, storm flow constitutes about 80% of the water discharged to the Newport Bay in Orange County, CA (*6*). In the San Diego Creek, which serves as the main drainage channel to Newport Bay, the median diazinon (445 ng L^{-1}) and chlorpyrifos (87 ng L^{-1}) concentrations in storm flow are higher than or similar to those in the base flow (200 ng L^{-1} for diazinon and 111 ng L^{-1} for chlorpyrifos). Thus, the overwhelming majority of the pesticide load in the San Diego Creek derives from storm runoff. In a stream that receives drainage from an urban watershed in Colorado, the peak pesticide concentration consistently coincided with storm events (*7*).

While the amount of overland runoff depends on both rainfall factors and surface conditions, appearance of a pesticide in the runoff will be largely controlled by its availability at the time that a runoff event occurs. This was reflected in monitoring results, in which the most frequently detected pesticides happened to be the more persistent ones (*9*). Pesticide persistence in a soil also correlates closely with soil chemical properties and microbial activity (*10, 11*). Residential landscapes, unlike agricultural fields, are comprised of highly diversified plantings, such as turfgrass, trees, ground covers, shrubs, mulched areas, and bare surfaces. The heterogeneous planting may affect soil properties, influencing pesticide persistence and runoff potential.

The objectives of this study were to evaluate the interaction of types of urban planting with soil chemical properties and microbial activity and the effect on the degradability of 2,4-D and dicamba. The use of 2,4-D and dicamba ranked first and 5th among urban herbicides (*12*). Residue of 2,4-D was detected at 51%, while that of dicamba at 25%, of the urban stream sites surveyed by USGS (*12*). The results from this and similar studies will be useful for identifying high-risk landscape systems and for developing mitigation practices to reduce pesticide runoff.

Materials and Methods

Soils and Chemicals. Soil samples were collected from a field located at the Agricultural Experiment Station on the campus of University of California in Riverside, CA. The field consisted of multiple 8 × 8 m plots with different planting covers that were established in 1995. The soil was a Hanford fine sandy loam (coarse loamy, mixed, Thermic Haplic Durixeralf) containing 0.3% organic matter. The planting systems included Bradford pear tree (*Pyrus calleryana*), "shortcut" tall fescue grass (*Festuca arundinacea*), mulches (chipped tree branches and leaves), and a low growing ground cover, spring cinquefoil (*Potentilla tabernaemontani*). All plots had received the same fertilization (0.5 lb actual N per 1000 ft^2 every month) and pesticide (oxadiazon, pendimethalin, and glyphosate) treatments. The turfgrass plots were occasionally treated with fungicides (iprodione and mancozeb) and mowed twice a week from March through November and weekly from December through February. The plots of turfgrass and cinquefoil had 100% cover by the

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TABLE 1. Selected Properties of Surface and Subsurface Soils for the Landscape Soils Used in the Study

soil	OM (%)	clay (%)	silt (%)	sand (%)	CEC (meg/100 g)	pH
Surface Soil (0−10 cm)						
tree	0.35	10	24	66	6.3	5.4
grass	0.82	9	24	67	7.7	6.7
mulch	1.95	8	24	68	10.7	6.9
ground cover	1.16	8	26	66	8.4	6.3
Subsurface soil (11−30 cm)						
tree	0.28	10	26	64	9.1	6.2
grass	0.37	10	24	66	7.5	6.9
mulch	0.33	11	26	63	8.2	6.9
ground cover	0.34	10	24	66	7.9	6.6

vegetation, while the plots of tree had one tree at the center of the plot. Four replicated soil cores of 10 cm diameter were removed from four plots with the same planting using a soil auger. Soil was collected from the 0−10 cm layer and the 10−30 cm layer and pooled for the same planting treatment. These samples were sieved through a 2-mm sieve while still moist and were stored in plastic bags at room temperature before analysis. Chemical and physical properties of these soils were analyzed by the University of California − Division of Agricultural and Natural Resources (DANR) Analytical Laboratory (Table 1). Soil organic matter content was determined using the Walkley-Black method (*13*), and particle sizes were determined using the hydrometer method (*14*).

Standards of 2,4-D (2,4-dichlorophenoxyacetic acid, purity 99%) and dicamba (purity 98%) were purchased from Chem Service (West Chester, PA) and were used for spiking soil samples and as standards in HPLC calibration. Methanol and acetonitrile used in extraction and analysis were both of HPLC grade.

Degradation Experiments. Degradation of 2,4-D and dicamba in the different landscape soils was determined by incubating spiked soil samples at 20 °C. The initial soil water content was adjusted to 8% (w/w) by drying or adding deionized water when necessary. Thirty grams of soil (dry weight equivalent) was weighed into 150-mL flasks. Stock solutions of dicamba or 2,4-D were prepared in water at 60 μg mL^{-1} for each herbicide. For treatment, 1.0 mL of stock solution was added into each soil flask using a pipet, which resulted in an initial herbicide concentration of 2.0 mg kg^{-1}. The flasks were allowed to sit for 2 h and then were mixed thoroughly by rotation and shaking. The flasks were then loosely covered with aluminum foil and held in an incubator at 20 °C. Moisture levels were maintained during incubation by weighing the soil flasks and adding deionized water when necessary.

At 0, 1, 3, 7, 14, 28, 56, and 112 days after pesticide treatment, three replicate flasks were removed from each soil-pesticide treatment and transferred immediately to a freezer (−20 °C) to stop degradation. For extraction, samples were removed from the freezer and thawed at room temperature. The soil was transferred to a 250-mL centrifuge bottle. The flask was rinsed with 60 mL of methanol−water (1:1, v/v), and the rinse solution was combined with the soil. The capped centrifuge bottles were then shaken on a mechanical shaker for 2 h and centrifuged at 3500 rpm. An aliquot (2 mL) of the supernatant was filtered through a Whatman glass microfiber filter (1.0 μm pore size and 13 mm diameter) into an autosampler vial for analysis on HPLC. The recovery of the extraction method was determined to be about 100% for both 2,4-D and dicamba in all soils.

Analysis of 2,4-D and dicamba in extracts was carried out on an Agilent 1100 series HPLC system (Agilent Technologies, Wilmington, DE) with UV detection. The column was an Agilent Eclipse XDB-C8 of 4.6 × 150 mm dimension and 5 μm particle size. The mobile phase was 40% acetonitrile and 60% water acidified to pH 2.7 with phosphoric acid. The flow rate was 1.0 mL min^{-1}, and the sample injection volume was 30 μL. The wavelength of detection was 230 ± 15 nm for both herbicides.

Enumeration of Herbicide Degraders. Both 2,4-D and dicamba are known to degrade in soil through microbial transformations (*15*−*17*). In particular, many soil microorganisms have been identified to be capable of using 2,4-D as the sole carbon source and mineralizing the herbicide (*18*, *19*). The population size of 2,4-D degraders was determined in the surface soils using the most probable number (MPN) method (*20*). The same method has been used by other researchers for quantifying indigenous herbicide degraders in soils (*21*). The method is based on measurement of the presence or absence of 2,4-D degrading bacteria by using an extinction dilution procedure. Briefly, 1 g (dry weight equivalent) of fresh soil was vortexed in 9 mL of a mineral salt media (MS) in a 20-mL glass vial. The carbon-free MS solution contained 3.5 g L^{-1} of Na_2HPO_4, 0.7 g L^{-1} of $KH_2PO_4 \cdot 7H_2O$, 0.1 g L^{-1} of $MgSO_4$, 0.5 g L^{-1} of Na_2CO_3, 0.18 g L^{-1} of $CaCl_2 \cdot 2H_2O$, and 0.014 g L^{-1} of $FeCl_3 \cdot 6H_2O$. After mixing, 1.0 mL of the soil suspension was transferred to a second vial and mixed with 9 mL of clean MS solution. This dilution step was sequentially repeated for a total of seven times, yielding 10^7-fold dilution of the original 1 g of soil. One milliliter of 2,4-D aqueous solution containing the herbicide at 40 mg L^{-1} was then added into each vial, and the vials were sealed and incubated at room temperature on a platform shaker. Five replicates were used for each dilution. In one set of vials, 10 mL of deionized water was spiked with the same amount of 2,4-D, and this treatment was used as the blank control. Aliquots of samples were removed after 3 weeks and centrifuged at 14 000 rpm. The supernatant was analyzed for 2,4-D by HPLC using the same conditions as in the previous experiment. A positive detection was defined as a sample in which the 2,4-D concentration decreased by >20% compared to that in the control. Populations of 2,4-D-degrading microorganisms were calculated from the probability tables given in Alexander (*20*) using the number of positive and negative vials at inoculum-dilutions near the extinction point.

Results and Discussion

Effect of Planting on Soil Organic Matter Content. Soil OM content differed significantly among the various ground cover treatments that had been established for 6 years (Table 1). While the OM content in the soil from the tree plots remained essentially unchanged, soils from the turfgrass, ground cover, and mulch plots showed 170, 280, and 550% increases over the original level, respectively. Apparently, in turfgrass and ground cover soils, decomposition of plant detritus contributed to the increase in OM content. In the mulch soil, incorporation and decomposition of bark and other woody materials contributed to the organic matter enrichment. The very low OM content in the bare soil in the tree plot was likely caused by the lack of biomass accumulation on the soil surface. Compared to the surface layer, changes in OM content were much less noticeable in the subsurface layer (Table 1), suggesting that incorporation of organic matter due to planting or mulching was limited mostly to the surface soil. Subsurface OM content changed little from the initial conditions (Table 1). Numerous studies have shown that soil organic matter plays a critical role in soil microbial ecology and hence in the degradation of many contaminants (*22*). Thus, 2,4-D and dicamba were predicted to degrade at different rates in the different soils.

2,4-D Persistence. The degradation of 2,4-D was followed over a period of 112 d after treatment (Figures 1 and 2). In

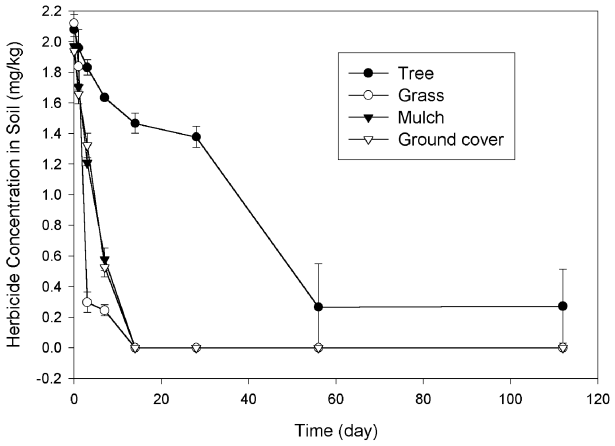

FIGURE 1. Degradation of 2,4-D in the surface soils (0−10 cm) with different landscape planting covers. Vertical lines are standard deviations of three replicates.

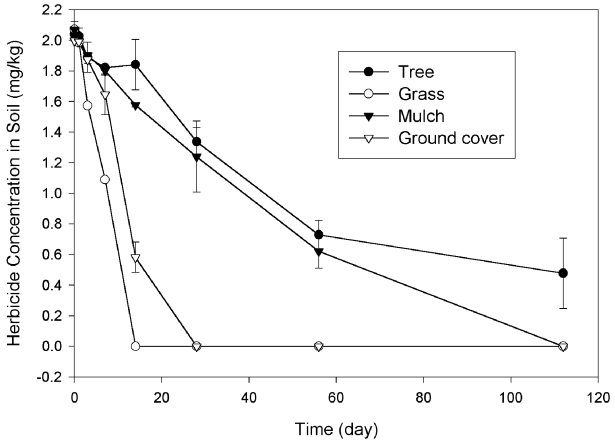

FIGURE 2. Degradation of 2,4-D in the subsurface soils (10−30 cm) with different landscape planting covers. Vertical lines are standard deviations of three replicates.

the surface soils, significantly different degradation patterns were observed among the different soils (Figure 1). The most rapid degradation occurred in the turfgrass soil, in which 2,4-D decreased to a small fraction of the original concentration within 3 d after treatment. Degradation of 2,4-D was also rapid in the ground cover soil and the mulch soil (Figure 1). The slowest degradation occurred in the tree soil, where 2,4-D concentration decreased gradually over the first 28 d and then accelerated. The two-phase pattern is typical of biodegradation and was observed for 2,4-D in previous studies (*23*). The decline of 2,4-D concentration over time was fitted to a first-order decay model to estimate the rate constant (d^{-1}) and half-life (d). The data fit the first order model well as evidenced by correlation coefficients above 0.9 (Table 2). The half-life of 2,4-D in the grass soil was estimated to be 1.6 d and was slightly longer than in the ground cover (3.9 d) or mulched (3.7 d) soil. The half-life of 2,4-D in the tree soil (30 d) was the longest among all the soils. The persistence of 2,4-D in the different soils therefore followed an order of tree soil > ground cover soil ≈ mulch soil > turfgrass soil. It may be envisioned that if a rain storm occurred following 2,4-D treatment, the potential for the herbicide to move in storm runoff would increase in the order of turfgrass soil < mulch soil ≈ ground cover soil < tree soil. Furthermore, with its large biomass and dense, fibrous root system and its ability to quickly degrade 2,4-D, grassed areas may likely act as a "filter" for 2,4-D. Grassed strips may therefore be placed on the border of residential landscapes to reduce runoff of pesticides such as 2,4-D.

TABLE 2. First-order Rate Constants and Correlation Coefficient for 2,4-D Degradation in Various Landscape Soils

soil	k (day^{-1})	$T_{1/2}$ (day)	R
Surface (0−10 cm)			
tree	0.0226	30.7	0.97
grass	0.4256	1.6	0.97
mulch	0.1851	3.7	0.96
ground cover	0.1798	3.9	0.99
Subsurface (10−30 cm)			
tree	0.0151	45.9	0.97
grass	0.1318	5.3	0.98
mulch	0.0215	32.3	0.99
ground cover	0.0757	9.2	0.98

Conversely, 2,4-D applied to exposed soil surfaces such as in areas around trees or bushes may be highly susceptible to runoff, and inadvertent application in these areas should be avoided when possible.

In all subsurface soil layers, degradation of 2,4-D was much slower than in surface layer soils, but some differences were still evident among soils with different covers (Figure 2). The fastest degradation occurred in the turfgrass soil, which was followed by the ground cover soil. The slowest degradation of 2,4-D was found in the subsurface soil of tree and mulched plots. The overall order of 2,4-D persistence in the subsurface soils was therefore tree soil ≈ mulch soil > ground cover soil > grass soil. The relatively rapid degradation of 2,4-D in the turfgrass and ground cover soils may be attributed to the fact that some roots might have reached the subsurface soil, which could increase the indigenous microbial activity and hence 2,4-D degradation. In contrast, due to a lack of root activity, the high degradability of 2,4-D in the mulched soil did not extend beyond the 10 cm depth (Figure 2; Table 2). In the tree plots, root distribution was sparse and apparently did not stimulate soil microbial activity in the subsoil.

Dicamba Persistence. The degradation of dicamba was generally slower than that of 2,4-D in the same soils. In the surface soils, the fastest degradation occurred in the mulched soil, which was followed by the ground cover soil and then the turfgrass soil. The slowest degradation for dicamba was again found with the tree soil (Figure 3). The decline of herbicide concentration over time was fitted to a first-order decay model, and the estimated first-order rate constant and half-life values are given in Table 3. Good correlation was found for all soils, with R above 0.93. The half-life of dicamba in the turfgrass, mulch, and ground cover soils ranged from 7.9 to 19.6 d, which was considerably longer than that for 2,4-D in the same soils (1.6−3.9 d). The overall ranking of dicamba persistence in the different soils was tree soil > turfgrass soil > ground cover soil > mulch soil. This order was different from that for 2,4-D, indicating that different factors influenced degradation of the two herbicides in the landscaped soils. The prolonged persistence in the tree soil suggests again that dicamba applied on exposed soil such as in the area around trees or bushes may represent an increased runoff risk and such applications should be discouraged. The overall longer persistence of dicamba than 2,4-D implies that different pesticides may have different runoff risks following similar applications. The use of relatively persistent products should be reduced or avoided during the rainy season when surface runoff is more frequent.

The persistence of dicamba in the subsurface soils was significantly prolonged compared to that in the surface soils (Figure 4). The shortest persistence was found in the ground cover soil, but the half-life was still 46 d or 1.5 months. Dicamba was highly persistent in the subsurface layer of turfgrass and mulch soils, and the half-life was >100 d or 3 months. In the subsurface tree soil, dicamba became

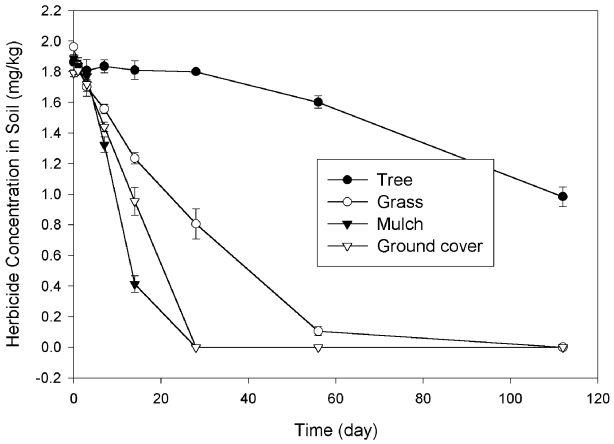

FIGURE 3. Degradation of dicamba in the surface soils (0−10 cm) with different landscape planting covers. Vertical lines are standard deviations of three replicates.

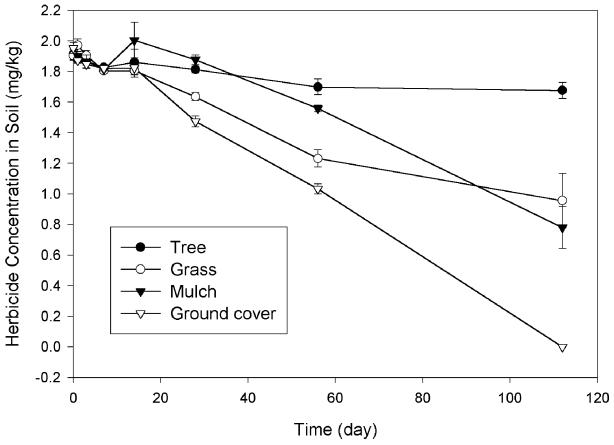

FIGURE 4. Degradation of dicamba in the subsurface soils (10−30 cm) with different landscape planting covers. Vertical lines are standard deviations of three replicates.

TABLE 3. First-Order Rate Constants and Correlation Coefficient for Dicamba Degradation in Various Landscape Soils

soil	k (day^{-1})	$T_{1/2}$ (day)	R
	Surface (0−10 cm)		
tree	0.0047	147	0.95
grass	0.0354	19.6	0.99
mulch	0.0873	7.9	0.98
ground cover	0.0620	11.2	0.98
	Subsurface (10−30 cm)		
tree	0.0012	592	0.93
grass	0.0068	102	0.99
mulch	0.0063	110	0.93
ground cover	0.0148	46.8	0.96

essentially nondegradable, and the half-life was estimated to be 590 d or over 1.5 years. The persistence of dicamba in the subsurface soils followed a ranking of tree soil > mulch soil > turfgrass soil > ground cover soil. The relatively short persistence of dicamba in the ground cover soil may be attributed to the fact that roots of the ground cover, *spring cinquefoil*, were distributed deeper in the soil than those of the turfgrass. This may have resulted in some microbial activity in the deeper soil layers. The long persistence of dicamba in subsurface soils implies that there is an increased risk for dicamba to leach through soil and contaminate groundwater under conducive conditions.

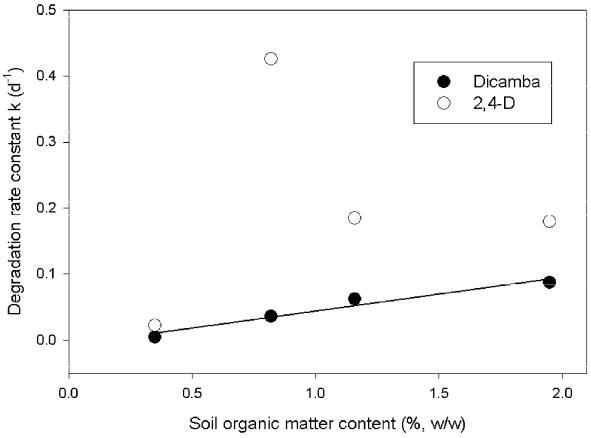

FIGURE 5. Relationship between degradation rates of 2,4-D and dicamba and soil organic matter content in the surface soils (10−30 cm) with different landscape planting covers.

Correlation with Soil Properties and Microbial Activity. Significant differences in herbicide persistence were observed in the landscape soils, especially in the surface layer. While many factors, alone or collectively, may have contributed to the differences, we attempted to understand these differences from the variation in soil OM content and in the population density of herbicide-degrading microorganisms caused by the different planting practices over time.

Correlation with soil OM content was performed for the surface soils for 2,4-D and dicamba. Excellent correlation was found between the degradation rate and soil OM content for dicamba (R = 0.98) but not for 2,4-D (Figure 5). The relationship between soil OM content and the rate constant for dicamba degradation was $k = -0.0077 + 0.0515 \times \text{OM}$, where k is the first-order rate constant in d^{-1}. Soil organic matter supports the growth of soil microorganisms and contains a great number of chemically active functional groups. Dependence of pesticide degradation on soil OM content has been also observed by other researchers (*10, 24*). However, similar dependence on soil OM content was not found for 2,4-D, and the correlation coefficient R was only 0.16. The lack of correlation suggests that some other factors other than soil OM content might play a more significant role in 2,4-D dissipation in the landscape soils.

The population density of herbicide degraders was enumerated for the surface soils using the most probable number (MPN) method, in which the herbicide served as the carbon source to stimulate biodegradation. No significant dissipation was observed in dicamba-treated tubes even 7 weeks after the treatment, suggesting that dicamba degradation in the soils was likely caused by mechanisms other than biodegradation. However, dissipation of 2,4-D occurred 1 week after the treatment, and data after 3 weeks of incubation were used to estimate the population density of 2,4-D degraders in the surface soils. The mean population and 95% confidence limits of 2,4-D degrading microorganisms were estimated to be 2300 (1000−5500), 230 000 (89 000−600 000), 49 000 (19 000−126 000), and 13 000 (5000−33 500) cells g^{-1} soil, for the tree, turfgrass, mulch, and groundcover soils, respectively. Regression analysis showed that there was a linear relationship between the mean density of 2,4-D degraders and the degradation rate constant k (d^{-1}) for the surface soils (R = 0.94). Linear relationship between degrader populations has been previously observed in agricultural soils for 2,4-D (*10, 11*) and atrazine (*21*). Organic contaminants have been observed to disappear more quickly from rhizosphere soils than from nonrhizosphere soils (*25*). It also appears that monocot species with fibrous rooting systems are more effective in enhancing degradation of organic

contaminants in planted soils than dicot species (*25*, *26*). In this study, the highest 2,4-D-degrader population was found in the turfgrass (monocot) soil, which was an order of magnitude greater than that in the ground cover (dicot) soil. The increased population of 2,4-D degraders in the turfgrass soil may be caused by the root exudates secreted by the tall fescue turfgrass, which might have selectively enriched 2,4-D degrading microorganisms.

Pesticide use in residential environments contributes to contamination of urban surface waterbodies. Watershed-scale mitigation measures are urgently needed to reduce pesticide load to meet water quality standards. Our study showed that landscape planting covers played a critical role in controlling the persistence of 2,4-D and dicamba in soil. After about 6 years, the soil chemical and microbial properties became significantly different under the different planting covers. Changes in microbial degrader population abundance were associated with herbicide degradation rate. The difference in 2,4-D persistence among the different landscape soils was seemingly associated with variation in population abundance of 2,4-D degraders, but the difference in dicamba persistence was better associated with variations in soil OM content. Of all the landscape systems tested, herbicide persistence was consistently prolonged in the soil collected around the tree landscape, which had comparatively the lowest OM content and abundance of herbicide degrading microorganisms. Therefore, under conducive conditions, higher runoff risks may be expected for 2,4-D and dicamba in such landscape systems. Dependence of pesticide persistence on planting covers may be used by city planners, developers, landscape architects, and professional landscapers for designing landscapes that are more resistant to pesticide runoff. Such information may be also used for educating the general public (e.g., homeowners) for reduced or guided pesticide use in sensitive planting systems. The influence of landscape planting on pesticide behavior should be further studied under field conditions, as photolysis, plant uptake, or volatilization also contribute to pesticide dissipation. Such influences may be considered along with other processes, such as surface infiltration and sediment retention, in devising management practices to reduce urban pesticide runoff.

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