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Diazinon and Permethrin Mitigation Across a Grass–Wetland Buffer

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Abstract Vegetated buffers of different designs are often used as edge-of-field treatment practices to remove pesticides that may be entrained in agricultural runoff. However, buffer system efficacy in pesticide runoff mitigation varies widely due to a multitude of factors including, but not limited to, pesticide chemistry, vegetation composition, and hydrology. Two experimental systems, a control (no vegetation) and a grass–wetland buffer system, were evaluated for their ability to retain diazinon and permethrin associated with a simulated storm runoff. The two systems were equally inefficient at retaining diazinon (mean 9.6 % retention for control and buffer). Grass–wetland buffers retained 83 % and 85 % of *cis*- and *trans*-permethrin masses, respectively, while the control only retained 39 % and 44 % of *cis*- and *trans*-permethrin masses, respectively. Half-distances (the distance required to decrease pesticide concentration by one-half) for both permethrin isomers were 26 %–30 % shorter in grass buffers (22–23 m) than in the control (32 m). The current study demonstrates treatment efficacy was a function of pesticide properties with the more strongly sorbing permethrin retained to a greater degree. The study also demonstrates challenges in remediating multiple pesticides with a single management practice. By using suites of management practices, especially those employing vegetation, better mitigation of pesticide impacts may be accomplished.

Keywords Pesticides · Runoff · Pyrethroid · Organophosphate · Insecticide

Agriculture continues to face multiple challenges in the 21st century. While the demand for food and fiber production continues to increase with an expanding global population, there remains a growing scrutiny on the degradation of surface and ground water resources from pesticide contamination. To address these issues, farmers are implementing various management practices aimed at decreasing off-site transport of pesticides. One classification of management practices targeting pesticide runoff is buffers. Dabney et al. (2006) noted buffers come in a variety of forms, but can generally be categorized as in-field, edge-of-field and after-field. Buffers generally rely on the presence of vegetation to reduce the velocity of runoff, allowing for sorption in plants and soil while also encouraging chemical breakdown through natural processes and bacteria. Otto et al. (2012) demonstrated the ability of narrow vegetative filter strips to decrease herbicide concentrations running off cultivated fields. Krutz et al. (2004) found adsorption to grass, grass thatch, and/or soil are important mechanisms for pesticide retention under saturated conditions in vegetated filter strips. Both the pesticide water phase dissipation time and organic carbon sorption coefficient, in addition to system hydraulic retention time and overall plant coverage are important factors in designing systems for pesticide retention and mitigation (Stehle et al. 2011). Lacas et al. (2005) conducted a review of pesticide mitigation with grass buffer

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strips, reporting that wide variability in mitigation results are partially explained by dynamic, site-specific factors, in addition to the many different physicochemical processes occurring during runoff events. Additionally, in a review by Zhang et al. (2010), it was reported that of pollutant removal studies using grass buffers, pesticide mitigation varied the most. Within the literature examined, buffer width alone accounted for 60 % of total variance in pesticide removal efficacy (Zhang et al. 2010).

Diazinon [*O*,*O*-diethyl *O*-(2-isopropyl-6-methyl-4-pyrimidinyl) phosphorothioate] is an organophosphate insecticide used on a variety of different crops, including, but not limited to, alfalfa, almond, carrots, cherries, lettuce, and processing tomatoes [California Department of Pesticide Regulation (CDPR) 2013]. Even though diazinon's use over the last decade has dropped from nearly 315,000 kg in 2002 to about 40,000 kg in 2011 in California alone, it is still an important pesticide being used by vegetable and stone fruit growers (CDPR 2013). Diazinon is often detected in surface waters, especially those in the Central and Imperial Valleys of California, and has been an identified source of aquatic toxicity (de Vlaming et al. 2004; Brady et al. 2006; Anderson et al. 2011). Diazinon median 96 h LC_{50} values are 25 and 602 μg/L for benthic invertebrates and fish, respectively (Munn et al. 2006). With a K_{OC} of 1750, diazinon is not expected to rapidly bind to sediment being transported during storm runoff events (Evans et al. 1998).

Permethrin [3-phenoxybenzyl-(1RS)-*cis, trans*-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate] is a pyrethroid insecticide with over 35 years of use in the United States (USEPA 2006). Although about only 30 % of the annual 900,000 kg applied are in agricultural settings, nearly 137,000 kg are applied on 21,000 ha in California alone (USEPA 2006, CDPR 2013). Both freshwater and estuarine aquatic organisms have a low toxicity threshold for permethrin, with median 96 h LC_{50} values of 0.17 and 6.8 μg/L for benthic invertebrates and fish, respectively (Munn et al. 2006). Because of permethrin's high affinity for organic carbon (K_{OC} 16,400–550,000) and low water solubility (0.2 mg L^{-1}), it is expected to rapidly sorb to sediment that may be transported during storm runoff events (Imgrund 2003). Arora et al. (2010) confirmed that strongly sorbed pesticides, such as those with a $K_{OC} > 1{,}000$, had an average retention of 70 % in buffer strip mitigation studies.

The objective of this research was to evaluate the efficiency of a grass-wetland buffer in the mitigation of diazinon and permethrin associated with a simulated storm runoff event. Mitigation efficiency will be determined by calculating pesticide half-lives ($t_{1/2}$), half-distances ($d_{1/2}$), and by comparing inflow and outflow loads of pesticides.

Materials and Methods

Two experimental plots at the University of Mississippi Field Station were evaluated, one unvegetated (17 m × 42.5 m) serving as a control and one vegetated (17 m × 47 m) serving as a treatment. The vegetated plot consisted of naturally established *Eleocharis obtusa* and *Carex lurida* (95 %) with scattered patches of *Paspalum urvillei* (5 %). Underlying soils were Lexington silt loam with a moist bulk density of 1.2–1.55 G cm^3, 0.5 %–2 % organic matter, and a permeability of 5–15 cm h^{-1}. Antecedent soil conditions for both plots were similar, with both being approximately 80 %–90 % saturated at the time of the experiment.

A one-time simulated 5 h runoff event was conducted to assess the efficiency of the two systems in mitigating a pesticide mixture added to each system. The pesticide mixture was prepared by combining 79.2 mL Diazinon 4E®, 10.8 mL permethrin (Hi-Yield 38 Plus®), and spring water (University of Mississippi Field Station—UMFS) into a 189 L mixing chamber at each of the two experimental plots. Amounts of pesticide used were based on the standard application rate (4.9 L/ha for diazinon; 0.37 L/ha for permethrin), an assumed standard agricultural field size of 32 ha, and 0.05 % and 0.09 % runoff for diazinon and permethrin, respectively (Spencer and Cliath 1991). Pesticide-water mixture was delivered to each experimental plot via 1.27 cm Tygon® tubing plumbed into a 17 m PVC diffuser running the width of each plot, with 2.5 cm holes drilled every 25 cm along the diffuser for even outflow. The mean discharge rate for the pesticide mixture was 0.631 L/min, while unamended UMFS spring water was also delivered at a rate of 745 L/min to each experimental plot through the same diffuser. Flows were measured using a calibrated collection system and stopwatch. Following the 5 h event, no further water or pesticide mixture was amended to the experimental plots.

Inflow (0 m), midflow (14–16 m), and outflow (42.5–47 m) sampling stations were designated for the collection of water, sediment, and where applicable, plant material on both experimental plots. Samples of all three environmental matrices were collected hourly for 10 h, then again at 15, 24, 48, 72 h, 7, 14, 21, and 28 days.

Water (500 mL) was collected in amber glass jars, while sediment (20 g) was collected from the surface 2 cm at each sampling station using an acetone-washed trowel. Approximately 20 g of plant material between the sediment and water surface were collected at each sampling site, representing the stems and leaves in direct contact with the surface water. All samples were immediately placed on ice for transport back to the USDA National Sedimentation Laboratory, Oxford, MS. Upon arrival at the laboratory, water samples (unfiltered) were immediately

fixed with 25 mL ethyl acetate and 500 mg sodium sulfate. Sediment and plant samples were air dried, ground using a Wiley Mill (Thomas Scientific, Swedesboro, NJ), then prepared for pesticide analyses according to Bennett et al. (2000). Pesticide analyses were conducted using an Agilent Model 7890A gas chromatograph equipped with dual Agilent 7683B series autoinjectors, dual split-splitless inlets, dual capillary columns, and an Agilent ChemStation (Agilent Technologies, Santa Clara, CA). Agilent electron capture detectors analyzed pesticides at 325 °C with ultra-high purity nitrogen as the makeup gas. A 1 μL injection volume was used for diazinon in addition to both *cis*- and *trans*-permethrin. Aqueous limits of detection (LOD) were 0.2, 0.21, and 0.17 μg/L for diazinon, *cis*-permethrin, and *trans*-permethrin, respectively. LODs for sediment pesticide concentrations were 20, 20.9, and 17.1 μg/kg, for diazinon, *cis*-permethrin, and *trans*-permethrin, respectively. LODs for plants were 50, 52.25, and 42.75 μg/kg for diazinon, *cis*-permethrin, and *trans*-permethrin, respectively. Further analytical descriptions on analyses are available in Lizotte et al. (2011).

Descriptive statistics were used to evaluate data between and within experimental systems. Mann–Whitney tests were performed for significance (0.05 level) between control and pond mass retention values. Values below detection limits were only utilized in statistical analyses 6 out of 108 possible times. This occurred when the inflow concentration was above detection and the outflow concentration was below detection limits. In these isolated instances, the system was assumed to have 100 % retention. Pesticide loads (inflow, middle, and outflow) were estimated by taking the sample concentration at the individual sampling station (inflow, middle, or outflow) and multiplying it by one-third of the volume for the entire experimental control (or buffer). Retention was determined by subtracting the outflow load from inflow load at each sampling time and determining a median value. Interquartile ranges (IQR) were likewise determined and reported along with median values in the results as median value (IQR). Pesticide depuration rate constants (k_2) were determined for water in both the control and buffer systems, as well as at the inflow, midpoint, and outflows (although no physical outflow occurred). Constants were determined by plotting the ln (maximum observed concentration) as a function of time, using linear regression analysis to determine the slope (k_2). Aqueous pesticide half-lives ($t_{1/2}$) were estimated using the equation ln (2)/k_2. R^2 values ranged from 0.7 to 0.95 for half-life determinations. Buffer half-distances ($d_{1/2}$) (the buffer distance required to decrease pesticide concentration by one-half) were estimated similarly to pesticide half-lives in that the ln (total concentration) was plotted as a function of buffer sample distance. The slope (k_2) was determined, and the half-distance was estimated using the ln (2)/k_2 equation. R^2 values ranged from 0.66 to 0.98 for half-distance determinations (Moore et al. 2008).

Results and Discussion

Diazinon, *cis*-permethrin, and *trans*-permethrin concentrations in the control and buffer experimental systems are presented in Tables 1, 2, 3. In the control (unvegetated) system, there was an overall median (IQR) aqueous diazinon load retention of 18 (41) %. During the first 5 h, median aqueous diazinon load retention was 44 (30) %, while aqueous load retention fell to 25 (32) % between the 6 and 10 h sampling. Similar aqueous retention percentages were recorded in the buffer system for diazinon. Overall median aqueous diazinon load retention was 6.7 (57) %, while median aqueous load retention for the first 5 h was 36 (46) %. Median aqueous load retention dropped to −0.66 (59) % between the 6 and 10 h sampling. No significant differences existed in median load retention between the control and grass buffer systems. Poletika et al. (2009) saw vast decreases in chlorpyrifos and atrazine removal efficiencies of vegetated filter strips when the runoff flow was concentrated (21 % and 12 %, respectively), as opposed to uniform flow (85 % and 62 %, respectively). Concentrated flow also resulted in lower infiltration (16 %). The current study also utilized a uniform flow design; however, infiltration may have been much less a factor because of the antecedent soil moisture. Additionally, the current experiment examined diazinon instead of chlorpyrifos, although both are organophosphate insecticides with some similar physicochemical properties. Another major difference between the current study and that of Poletika et al. (2009) was the drainage area ratios used. Poletika et al. (2009) compared drainage area ratios of 15:1 and 30:1, while the current study's drainage area ratio was extrapolated to be 400:1.

Different results were observed for both permethrin isomers when comparing the unvegetated control to the grass buffer system. For the control, *cis*-permethrin overall aqueous median load retention was 54 (45) %, while median aqueous load retention for the first 5 h was 36 (79) %, and median aqueous load retention between 6 and 10 h was 58 (55) %. In much smaller mesocosms of 0.6 m^3, Moore et al. (2009) reported *cis*-permethrin mass load retention in unvegetated controls of 69 ± 7 %. However, in the grass buffer system, there was an overall aqueous median *cis*-permethrin load retention of 82 (12) %, while median aqueous load retention for the first 5 h was 84 (9.0) %, and median aqueous load retention between 6 and 10 h was 76 (12) %. Significant differences existed between the control and grass buffer system when

evaluating the overall median mass retention ($p = 0$; $Z = 4.1067$); retention for the first 5 h ($p = 0.01208$; $Z = -2.5067$); and retention for 6–10 h ($p = 0.03662$; $Z = -2.0889$). Schmitt et al. (1999) reported filter strips reduced permethrin associated with filter strips 27 %–83 %. Infiltration of runoff, along with settling and dilution played a significant role in the performance differences among filter strip design and pollutants (Schmitt et al. 1999).

When comparing the *trans*-permethrin aqueous load retention capacities between the control and grass buffer, similar results as *cis*-permethrin were obtained. For the control, the median overall load retention was 61 (48) %, while median load retention for the first 5 h was 38 (82) %, and median load retention between 6 and 10 h was 60 (56) %. The overall mass load retention was similar to that reported by Moore et al. (2009) who found 82 ± 2 % retention in a 0.6 m^3 unvegetated control. In the grass buffer, overall *trans*-permethrin aqueous median load retention was 85 (17) %, while median retention for the first 5 h was 87 (9.4) %. *Trans*-permethrin median aqueous load retention for the grass buffer from 6 to 10 h was 79 (12) %. As with *cis*-permethrin, significant differences existed between the control and grass buffer system when evaluating the overall median mass retention ($p = 0.0005$; $Z = 3.4811$); retention for the first 5 h ($p = 0.01208$; $Z = -2.5067$); and retention for 6–10 h ($p = 0.02144$; $Z = -2.2978$).

In the control system, only 6 of the 57 sediment samples collected in conjunction with water samples (inflow, middle, and outflow) were above the limit of detection (20 µg/kg) for diazinon. Similarly, only 11 of 57 collected sediment samples in the grass buffer were above the limit of detection for diazinon. All collected sediment samples in both the control and grass buffer systems were below the limit of detection for both *cis*- and *trans*-permethrin (20.9 and 17.1 µg/kg, respectively). Multiple studies have demonstrated permethrin's rapid sorption to sediment (Sharom and Solomon 1981; Solomon et al. 1985; Imgrund 2003; Liu et al. 2004); however, the low concentrations applied in the current study may have prevented detection in collected sediment samples from either the control or grass buffer systems.

Plant samples (only available in the grass buffer system) yielded little data, with only seven out of 57 samples above diazinon's limit of detection (50 µg/kg). Of the seven detectable samples, all were collected at the 15 h or later sampling times. With respect to *cis*-permethrin, 12 out of 57 plant samples were above the limit of detection (52.25 µg/kg), with the majority coming after the 24 h sampling. Only three out of 57 plant samples were above the 42.75 µg/kg limit of detection for *trans*-permethrin, with all coming within the first 5 h of sampling at the

Table 1 Diazinon concentration (µg/L) in the sampling areas of the experimental systems

Time (h)	Control (unvegetated)			Buffer		
	Inflow	Middle	Outflow	Inflow	Middle	Outflow
1	66.2	–	26.6	146	–	50.0
2	124	117	69.3	98.6	90.7	63.3
3	85.9	103	48.2	133	117	57.3
4	160	93.9	75.8	77.5	110	73.6
5	114	122	104	107	128	80.9
6	76.9	83.2	95.7	54.0	130	54.4
7	117	80.2	82.9	81.5	110	85.2
8	107	74.1	79.4	106	86.1	48.2
9	81.5	82.2	59.5	24.1	134	68.9
10	93.9	96.6	77.6	95.3	83.9	65.8
15	84.2	107	73.6	98.1	134	93.7
24	86.6	71.4	84.2	83.7	106	76.7
48	49.5	45.8	36.4	46.6	64.5	64.5
72	36.5	51.9	41.5	34.0	60.5	44.3
168	17.5	16.1	14.1	16.4	19.4	28.5
336	3.71	4.24	7.57	5.34	8.27	6.22
504	3.12	3.02	3.31	4.10	2.58	2.24
672	1.25	1.41	1.88	1.80	1.12	1.23

– no sample collected

Table 2 *Cis*-permethrin concentration (µg/L) in the sampling areas of the experimental systems

Time (h)	Control (unvegetated)			Buffer		
	Inflow	Middle	Outflow	Inflow	Middle	Outflow
1	4.80	–	1.56	4.42	–	1.09
2	3.74	3.92	1.36	4.13	1.68	0.87
3	3.67	7.16	2.35	4.27	2.25	0.62
4	4.61	4.40	3.98	3.66	2.49	0.58
5	2.93	2.19	4.15	3.59	–	0.48
6	10.4	2.86	2.98	2.22	1.80	0.54
7	3.58	1.66	1.27	2.42	1.58	0.43
8	2.47	1.35	1.04	2.10	1.80	0.44
9	1.68	1.21	1.92	1.80	1.45	0.56
10	1.55	1.07	0.93	1.36	1.02	0.41
15	1.17	0.74	0.49	1.93	0.70	0.22
24	0.56	0.23	0.28	0.44	0.29	bd
48	bd	bd	bd	0.29	bd	bd
72	bd	bd	bd	bd	bd	bd
168	bd	bd	bd	bd	bd	bd
336	bd	bd	bd	bd	bd	bd
504	bd	bd	bd	bd	bd	bd
672	bd	bd	bd	bd	bd	bd

– no sample collected, *bd* below detection

Table 3 *Trans*-permethrin concentration (μg/L) in the sampling areas of the experimental systems

Time (h)	Control (unvegetated)			Buffer		
	Inflow	Middle	Outflow	Inflow	Middle	Outflow
1	4.09	–	1.26	3.72	–	0.86
2	3.20	3.29	1.11	3.42	1.34	0.66
3	3.13	6.02	1.94	3.53	1.79	0.45
4	3.90	3.71	3.36	3.06	1.98	0.41
5	2.50	1.79	3.58	3.03	–	0.33
6	9.03	2.45	2.71	1.79	1.36	0.38
7	3.12	1.40	1.09	1.97	1.24	0.30
8	2.14	1.13	0.86	1.63	1.44	0.30
9	1.46	0.96	1.72	1.40	1.14	0.27
10	1.32	0.88	0.76	1.03	0.78	bd
15	0.96	0.58	0.36	1.46	0.49	bd
24	0.35	bd	bd	0.17	0.17	bd
48	bd	bd	bd	bd	bd	bd
72	bd	bd	bd	bd	bd	bd
168	bd	bd	bd	bd	bd	bd
336	bd	bd	bd	bd	bd	bd
504	bd	bd	bd	bd	bd	bd
672	bd	bd	bd	1.80	1.12	1.23

– no sample collected, *bd* below detection

inflow site. Due to the sparse number of detectable sediment and plant pesticide samples, all mass calculations, half-lives, and half-distances were calculated with only aqueous data.

The half-life for diazinon in the control system was 106 ± 9 d, while the grass buffer system had a diazinon half-life of 103 ± 7 d. For *cis*-permethrin, half-life in the control was 5.3 ± 0.5 d, while it increased to 8.3 ± 2 d in the grass buffer. A similar *trans*-permethrin half-life was noted in the grass buffer system of 7.5 ± 2 d, compared to the control system's *trans*-permethrin half-life of just 4 ± 0.8 d. These half-lives are comparable to those reported by Lutnicka et al. (1999) ranging from 1.1 to 3.6 d. Because diazinon and both permethrin isomers have low Henry's Law Constants (5.1×10^{-8}–1.4×10^{-6}), the pesticides are less volatile than water, leaving them to concentrate, rather than volatilize, as water evaporates from the system (Crosby 1998).

The half-distance was 68.6 m for diazinon in the control system and 73 m in the grass buffer. Although the concept and design of grass buffers is different than agricultural drainage ditches, Moore et al. (2008) reported diazinon half-distances in vegetated and unvegetated California ditches to be 55 m and 158 m, respectively. For *cis*-permethrin, half-distances were 31.7 m in the control and 23.3 m in the grass buffer. Similar results were obtained for *trans*-permethrin, with a half-distance of 31.5 m in the control and 22.1 m in the grass buffer. Half-distances for *cis*- and *trans*-permethrin in vegetated California ditches were 22 and 21 m, respectively, while half-distances for unvegetated ditches were 50 and 55 m, respectively (Moore et al. 2008).

Results from the current study demonstrate challenges in using single management practices to sufficiently mitigate pesticides to levels that do not impair aquatic receiving systems. The same practice that is successful for one pesticide may be much less efficient for pesticides with different physicochemical characteristics. Runoff containing diazinon was only partially remediated after transport through aquatic vegetation in a California study by Anderson et al. (2011). An additional practice (enzyme addition) was required to effectively remove remaining diazinon concentrations (Anderson et al. 2011). Approximately 60 % pesticide removal has been reported across the literature although vegetated buffer system efficiencies vary from 0 % to 100 % based on study design and pesticides targeted for removal (Bereswill et al. 2013). Results from the current study may also be used for future calibration of pesticide buffer models. As agriculture continues to develop management practices to mitigate pesticides associated with cropland runoff, it is critical that multiple practices be incorporated in systems to maximize pesticide removal.

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