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Comparative Aquatic Dissipation Rates Following Applications of Renovate OTF Granular Herbicide and Rhodamine WT Liquid

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ABSTRACT

A field study was conducted in October 2008 to compare the dissipation rates of concurrent applications of a granular formulation of triclopyr herbicide (Renovate® OTF) and the inert dye Rhodamine WT, acting as a surrogate for a liquid herbicide. Applications were made to a relatively deep 4-ha plot (mean depth = 4.75 m) in a cove of Grandview Lake, Indiana. Renovate OTF was applied using boat-mounted, forced-air spreaders at a dose of 800 µg L^{-1} triclopyr. Rhodamine WT was applied through two long, trailing, weighted-hoses at a dose of 14 µg L^{-1} dye. Following application, both compounds rapidly mixed within the water column. Monitoring of water concentrations demonstrated relatively rapid dissipation patterns due to water exchange; 26.7 h half-life for triclopyr and 12.4 h for the dye. The results indicate the granular formulation would have a 2.2x longer exposure time and a different vertical residue profile than a subsurface injection of a liquid formulation, suggesting the potential for greater plant efficacy.

Key words: dye, formulation, granule, triclopyr.

INTRODUCTION

Rapid dissipation of aquatic herbicides due to various water exchange processes can lead to poor submersed weed control in a variety of situations. The ability to target herbicide placement and maintain the concentration in the plant mass within the 3-dimensional aquatic environment can be critical to maximize efficacy. Additional variables such as

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temperature differentials and plant density can also alter herbicide distribution. Custom subsurface injection application systems with trailing hoses have been suggested for improved delivery of liquid herbicides in deeper water areas (Haller et al. 2007). Granular formulations also have been developed to assist in delivering aquatic herbicides (Aquathol® Super K; Navigate®; Sonar® Q, PR, and SRP; and more recently Renovate OTF and SonarOne). These formulations release the herbicide in and around submersed plant communities and often sink to the bottom, depending on plant density and frequency, where the herbicide is released (Steward and Nelson 1972, Applied Biochemists 2009).

Few field studies have compared dissipation rates or residue profiles following application of a liquid herbicide to a granule formulation. Fox and Haller (1992) reported that liquid applications using long weighted-hoses or the use of granules increased potential exposure time to the herbicides compared to liquid applications using unweighted hoses. Field observations suggest that under certain conditions liquid herbicide can be rapidly diluted, resulting in poor submersed weed control. Skogerboe and Netherland (2008) reported that 80 to 90% of liquid herbicide was lost from partial-lake treatment areas within 15 h following treatment. Herbicide applications in areas such as relatively deep sites with low growing vegetation, with potential for rapid water exchange, or those adjacent to or surrounded by a large percentage of untreated water could be impacted greatly by dilution. In these situations, where rapid water exchange may be a concern, the use of a granular herbicide could theoretically maintain placement of the herbicide or increase exposure times. This effect would be dependent on the vertical placement of the herbicide, the release rate of the active ingredient from the granule, and the vertical diffusion rate from the sediment-water interface. Delayed release of the herbicide from a granule could increase potential exposure time compared to a liquid formulation when 100% of the herbicide is immediately available but vulnerable to dilution. However, herbicide release from granules has to be rapid enough to build threshold concentrations that result in an herbicidal effect.

Triclopyr (3,5,6-trichloro-2-pyridinylocyacetic acid, triethylamine salt) was approved for aquatic use in 2002 by the US Environmental Protection Agency. The liquid formulation (i.e., Renovate® 3, SePRO Corporation, Carmel, IN; hereafter referred to as SePRO) was the only formulation initially registered. In 2006, a granule formulation (Renovate OTF; SePRO) was developed to aid delivery of triclopyr to the plant mass and to minimize immediate dilution or dispersion. The granule contains 14% active ingredient (ai): 10% acid equivalent (ae). Rhodamine WT (Keystone Analine Corporation, Chicago, IL) is an inert fluorescent dye containing 20%, or 240 g ai L^{-1}. This dye is used for a variety of purposes, including water movement studies, and is commonly used to predict herbicide movement in aquatic environments (Fox et al. 1991, 1992, 1993, 2002, Turner et al. 1994).

Concentration exposure-time studies have provided valuable information regarding the required duration of herbicide exposure for the effective control of many submersed weeds (Netherland et al. 1991, 1993, Netherland and Getsinger 1992). However, the influence of herbicide formulation and application technique on concentration exposure time and weed control has not been well explored, especially when comparing the effectiveness of a granular formulation to liquid application of the same herbicide. To investigate this potential difference in residue profiles due to formulation, a field dissipation study was conducted to compare the dissipation rates of concurrent applications of Renovate OTF and the inert dye Rhodamine WT. Dissipation studies investigate the movement and degradation of pesticides. They are usually conducted under "worst case" conditions, those factors that would prolong the degradation rate of the compound or maximize its off-site movement. Efficacy is not a factor in dissipation studies, which are often conducted "bare plot." Thus, for this study, a treatment scenario was chosen where herbicide would be applied to a relatively deep site with sparse vegetation found predominately near the bottom. This site was selected to represent a treatment area with potential for relatively rapid dilution from untreated water and that allowed monitoring of vertical distribution of the compounds.

MATERIALS AND METHODS

Grandview Lake is a 132-ha reservoir located in Bartholomew County, Indiana, approximately 11 km southwest of the town of Columbus and 70 km south of Indianapolis (Figure 1). This study was conducted October 2008 in a 4-ha bay in the southern portion of the lake.

A bathymetric survey was conducted along predetermined transects using a Garmin GPS 72 and a Garmin Fishfinder 160 (Olathe, KS). Depths in the bay to be treated were recorded at 366 waypoints, and an average plot depth of 4.75 m was calculated.

Four sampling locations (Figure 1; stations 1, 2, 3 and 4) were sited within the treatment plot at locations where the water depth was 5.5 m. An additional six sampling locations were sited outside the treatment area, in areas were the water

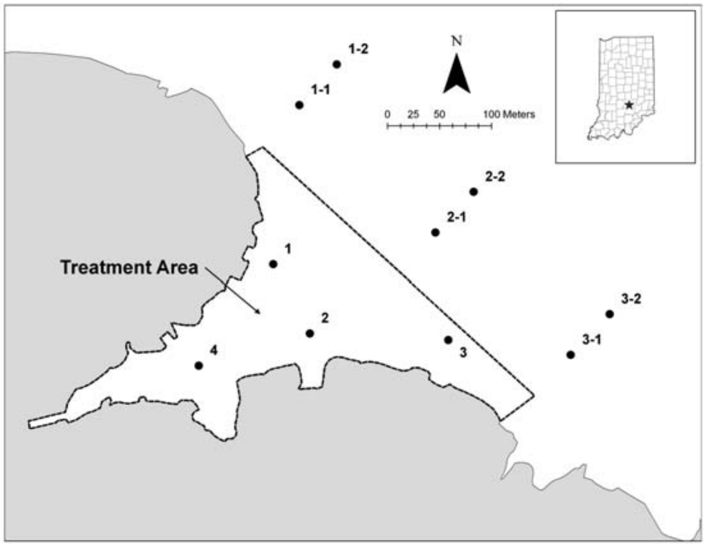

Figure 1. Plot map and location of Grandview Lake, IN.

depth was at least 12 m (Figure 1; stations 1-1, 1-2, 2-1, 2-2, 3-1, and 3-2). The off-plot sample locations were approximately 50 and 100 m from the open-water edge of the treatment area. The on-plot stations were marked with PVC pipe-mounted sampling devices (described later), and the off-plot stations were recorded as GPS waypoints.

Concurrent applications of the granular triclopyr and liquid dye took place on the morning of October 7 beginning at 08:30 and complete by 10:00. The granular product was applied from a boat with 2 Echo PB755H landscape blowers (Lake Zurich, IL) customized for granular application. The blowers were mounted on each side of the application boat and applied the granules at 45° from the bow of the boat. We applied 3400 lbs of formulated material evenly across the treatment area, which resulted in a nominal water concentration of 800 µg ae triclopyr L^{-1}. The dye was deep injected by trailing two 13.7-m weighted hoses from a boat traveling ≤8 kph equipped with a high-pressure Hypro D30 diaphragm pump (New Brighton, MN) run by a Honda Power Equipment Group DX160 motor (Alpharetta, GA). Each injection hose weighed 5.92 kg including the weighted/nozzle section, which weighed 2.02 kg. We applied 11.4 L of dye formulation in six tank mixes consisting of 1.9 L of dye in 189 L of water. This resulted in a nominal water concentration of 14 µg ai L^{-1} dye.

Water was sampled from the on-plot stations at 21 discrete events, beginning with pre-application samples collected the evening before application and continuing at approximately 0.5, 1, 2, 4, 6, 8, 12, 16, 20, 24, 28, 32, 36, 40, 44, 48, 56, 64, 72, and 96 h after treatment. The off-plot stations were sampled on the same schedule, although early events were omitted for some stations.

At the on-plot stations, water was collected from six discrete depths at each station. These depths were +0.15, +0.30, +0.60, +1.2, and +2.4 m from the bottom of the lake, representing the lower half of the column, and a near-surface sample collected at approximately +5.0 m, or about 0.5 m below the water surface. The lower 5 samples were collected via an automated sampling device consisting of submersible 12-volt bilge pumps connected at the specified depths to a PVC pipe anchored into the lake bottom. Each bilge pump had a 1.3-cm dia PVC feed pipe running to approximately 1 m above the water surface, capped with a gate valve. The power leads for the bilge pumps were taped to the feed pipes and wired into a common switch. To collect samples, a 12-volt battery was connected to the common switch, and the pumps were allowed to run for a minimum of 30 to 60 sec to purge the feed lines. The pumps were then run one at a time, starting with the lowest, and water samples of approximately 200 ml were collected into pre-labeled 250-ml amber Nalgene® bottles. The near-surface sample was collected by lowering a bottle by hand to depth and allowing water to enter the bottle.

The off-plot water samples were collected from an approximate 3-m depth by lowering a bilge pump connected to a 1.9-cm PVC feed pipe and pumping the water sample up into the pre-labeled sample bottle.

Water samples were stored in coolers with ice during the collection, holding, and shipping periods. The sample labels contained only a sample ID number keyed to the sample collection sheet, which led to blind laboratory analyses.

Water temperature and dissolved oxygen (DO) measurements were taken in 1-m increments to the bottom of the water column each morning and afternoon during the study. These measurements were taken at a marked location within the treatment plot near sample location 2.

The water samples were received at the SePRO Research and Technology Campus (Whitakers, NC) on 15 October 2008. The samples were placed into frozen storage and thawed in lots of 100 for analysis via the immunoassay (ELISA) method for triclopyr (FasTEST - SePRO; Poovey et al. 2004). The limit of detection (LOD) was 10 µg L^{-1}. When analysis of each lot was complete they were returned to the freezer and the next lot was allowed to thaw. Analyses were completed on 15 November 2008.

After completion of the ELISA testing for triclopyr residues, the samples were shipped under ambient conditions on 3 December 2008 and were received at the University of Florida Center for Aquatic and Invasive Plants (Gainesville, FL) on 8 December 2008. The samples were allowed to remain at ambient temperature, and dye concentrations were analyzed using a Turner Designs Model 10 field fluorometer and Model 8000-010 handheld fluorometer (Sunnyvale, CA). The LOD for dye was 0.1 µg L^{-1}, and the fluorometers were periodically calibrated using Rhodamine WT standards ranging from 1.2 to 9.6 µg L^{-1}.

First-order dissipation half-lives of triclopyr and dye were calculated using Microsoft Excel software statistical functions, using the full precision of the values entered into the spreadsheet. The calculations were performed using the average whole-plot concentrations.

RESULTS AND DISCUSSION

Measurements of water temperature and DO demonstrated that both parameters were nearly homogeneous through depth and time (Tables 1 and 2). This indicated there was no stratification that might inhibit distribution of the herbicide or dye through the water column.

The concentration data derived by the lab analyses were summarized by averaging the results from the on-plot stations by depth increment (Tables 3 and 4), and also by whole-plot (Figure 2), for each sampling period. There were no detectable residues of either triclopyr or dye in the pre-application samples.

The target whole-plot concentrations for triclopyr and dye were 800 µg L^{-1} and 14 µg L^{-1}, respectively. The whole-plot average concentrations (Figure 2) indicate the target concentration of triclopyr was not achieved on a whole-plot basis. Data indicate that the concentration of triclopyr at the lower depths in the early time periods did meet this targeted dose (Table 3). As expected, the granule application resulted in release of the active ingredient over time from the bottom of the water column where the majority of granules had settled. A study conducted under laboratory conditions (without sediment) demonstrated that in 4 h Renovate OTF had released 94% of the triclopyr acid equivalent and achieved 99% of the theoretical concentration after 24 h (Hahn 2006). Data indicate that the concentration of dye (applied via long-trailing hose) met the nominal application rate in the mid-depths of the water column, but as with triclopyr, vertically dissipated

Table 1. Water temperature (C) measured through the water column twice daily during the study period.

Depth (m)	Oct 6 PM	Oct 7 AM	Oct 7 PM	Oct 8 AM	Oct 8 PM	Oct 9 AM	Oct 9 PM	Oct 10 AM
0	21.2	20.1	20.3	20.0	20.2	19.6	20.7	19.6
1.0	21.2	20.2	20.3	20.0	20.2	19.7	20.7	19.6
2.0	20.5	20.2	20.3	20.0	20.2	19.6	19.8	19.5
3.0	20.3	20.2	20.1	20.0	20.0	19.6	19.8	19.5
4.0	20.2	20.2	20.1	20.0	20.0	19.6	19.7	19.5
5.0	20.2	20.2	20.1	20.0	20.0	19.5	19.6	19.5
6.0	20.1	20.1	20.1	20.0	19.9	19.5	19.6	19.5
7.0	20.1	20.1	20.0	19.9	19.9	19.5	19.5	19.5
7.5	—	20.0	19.9	19.8	19.8	19.4	19.5	19.5
8.0	20.1	—	—	—	—	—	—	—

Table 2. Dissolved oxygen (mg L^{-1}) measured through the water column twice daily during the study period.

Depth (m)	Oct 6 PM	Oct 7 AM	Oct 7 PM	Oct 8 AM	Oct 8 PM	Oct 9 AM	Oct 9 PM	Oct 10 AM
0	8.71	8.42	8.52	8.10	8.80	8.10	8.67	8.73
1.0	8.02	8.10	8.68	7.82	8.39	7.88	8.40	8.57
2.0	8.50	8.33	8.60	7.87	8.38	8.28	8.62	8.23
3.0	8.54	8.20	8.35	7.95	8.42	8.00	8.54	8.47
4.0	8.53	7.98	8.03	8.15	8.69	7.36	8.25	8.48
5.0	8.27	8.02	7.80	7.35	8.34	7.83	7.88	8.49
6.0	8.03	8.19	7.99	7.39	8.22	7.75	7.94	8.56
7.0	8.52	8.31	8.20	7.70	8.10	7.94	8.10	8.65
7.5	—	7.82	8.01	7.45	8.48	8.13	8.04	8.55
8.0	8.52	—	—	—	—	—	—	—

Table 3. Average on-plot triclopyr concentrations (µg ae L^{-1}) by depth increment (indicated by distance above the lake bottom) following a nominal 800 µg ae L^{-1} application (n = 4; HAT: hours after treatment).

		Triclopyr Concentration µg L-1					
Date	HAT	+0.15 m	+0.30 m	+0.60 m	+1.2 m	+2.4 m	+5.0 m
7-Oct	0.5	750.3	492.3	434.4	273.5	203.4	124.2
7-Oct	1	693.6	607.6	511.5	388.7	257.7	112.7
7-Oct	2	861.5	762.2	518.2	210.6	372.8	231.1
7-Oct	4	671.2	526.4	331.5	231.8	133.4	131.5
7-Oct	6	608.2	350.4	323.6	137.6	115.5	310.8
7-Oct	8	484.5	461.5	219.9	162.9	60.9	290.4
7-Oct	12	552.4	447.5	381.1	173.5	145.4	156.8
8-Oct	16	275.5	171.9	129.0	150.3	168.6	131.8
8-Oct	20	212.4	315.9	322.1	196.1	223.3	124.6
8-Oct	24	288.9	289.5	172.0	150.4	196.2	145.3
8-Oct	28	184.0	172.3	189.5	133.6	177.0	167.0
8-Oct	32	234.6	172.1	208.6	75.9	112.0	145.1
8-Oct	36	207.3	167.0	170.0	152.2	164.8	147.4
9-Oct	40	196.0	185.8	171.8	167.1	181.4	174.8
9-Oct	44	195.5	198.7	189.7	236.3	172.7	232.5
9-Oct	48	180.4	172.6	152.8	124.6	115.1	117.4
9-Oct	56	114.0	72.6	76.8	87.9	94.1	76.0
10-Oct	64	129.3	103.4	83.1	67.0	64.5	58.0
10-Oct	72	85.5	60.4	60.0	49.8	69.9	40.6
11-Oct	96	27.7	26.0	22.5	22.6	19.6	20.5

relatively rapidly (Table 4). Triclopyr concentrations demonstrate that as the triclopyr acid was released from the granules, it quickly dispersed through the water column and did not result in abnormally high or long-lived concentrations at the sediment/water interface (Table 3). There was no evidence of accumulation of triclopyr at the bottom of the lake. There was a concentration gradient by depth for the majority of the sample events. The triclopyr and dye began dispersing

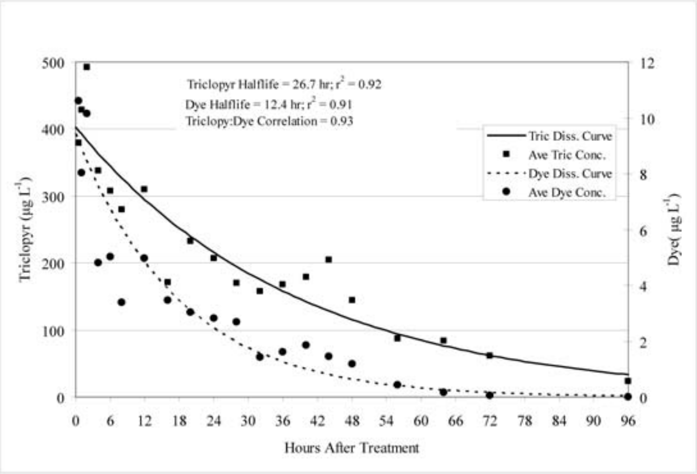

Figure 2. First-order dissipation curves of triclopyr (Tric.) and rhodamine dye (Dye; µg L^{-1}) following concurrent applications in Grandview Lake, IN.

vertically through the water column immediately after application and were well mixed by 28 h after application.

Triclopyr dissipated from the plot with a calculated half-life of 26.7 h ($r^2 = 0.92$), and Rhodamine WT dye dissipated with a calculated half-life of 12.4 h ($r^2 = 0.91$; Figure 2). The correlation coefficient between the triclopyr and dye dissipations was 0.93, which indicates that the two compounds had strongly correlated patterns of dissipation from the test plot. This result is in agreement with other lake studies in which Rhodamine WT was applied concurrently with liquid triclopyr (Turner et al. 1994, Petty et al. 1998, Fox et al. 2002). A factor influencing the post-application concentrations was the wind pattern noted during and after application. Observations recorded during the application state that the winds were calm, <8 km h^{-1}, and from the east to northeast, which would have reduced water exchange from the treatment plot bay and the rest of the lake (Figure 1). Soon after application the wind speeds increased and came from a southerly direction. These observations are confirmed by hourly weather data recorded at the nearby Columbus Municipal Airport, which recorded no wind from 8:00 to 11:00 on the day of application, followed by winds averaging 16.6 kph from the southeast to southwest from 12:00 through 18:00 the following day. These winds would increase chemical loss from the treatment plot through increased water-exchange rates, primarily pushing the material to the northwest shoreline (supported by visual observation of dye movement) and off-plot to the north. Winds remained negligible for the majority of the remainder of the study.

Overall, off-plot concentrations of triclopyr and dye were quite low. From 36 to 64 h, triclopyr and dye concentrations were <10% of the dose applied, and from 72 to 96 HAT concentrations were 3% or less of the concentration applied.

Under the conditions of this pilot study, triclopyr released from the granules remained in the treatment plot 2.2 times as long as the liquid dye, as determined by the individual half-lives. This would indicate that the exposure time of triclopyr to target plants resulting from the granular application would be twice that of an equivalent application of liquid herbicide. The factors influencing these differences are likely the release of the active ingredient from the granules and its mode of delivery, placing it at the bottom of the water column where it is likely less susceptible to wind-driven or surface-current water exchange with untreated water.

TABLE 4. AVERAGE ON-PLOT DYE CONCENTRATIONS (µG L^{-1}) BY DEPTH INCREMENT (INDICATED BY DISTANCE ABOVE THE LAKE BOTTOM) FOLLOWING A NOMINAL 14 µG L^{-1} APPLICATION (N = 4; HAT: HOURS AFTER TREATMENT).

		Dye Concentration µg L^{-1}					
Date	HAT	+0.15 m	+0.30 m	+0.60 m	+1.2 m	+2.4 m	+5.0 m
7-Oct	0.5	11.1	8.3	8.0	9.5	9.8	17.0
7-Oct	1	6.5	5.7	5.2	6.3	13.3	11.2
7-Oct	2	4.9	4.5	7.0	12.8	20.1	11.6
7-Oct	4	4.8	1.8	0.2	5.3	9.6	7.1
7-Oct	6	1.5	2.0	4.8	5.3	5.7	11.0
7-Oct	8	1.7	3.7	1.7	1.9	0.9	10.4
7-Oct	12	6.1	6.3	5.4	4.8	4.0	3.3
8-Oct	16	3.3	3.1	3.0	3.2	3.4	4.9
8-Oct	20	2.9	3.4	2.8	2.8	3.1	3.2
8-Oct	24	0.9	3.2	3.0	3.0	3.9	3.0
8-Oct	28	2.9	2.9	2.6	2.6	2.4	2.7
8-Oct	32	1.9	1.7	1.8	0.7	0.8	1.7
8-Oct	36	1.9	1.6	1.6	1.6	1.5	1.6
9-Oct	40	2.0	1.7	1.7	1.5	1.6	2.7
9-Oct	44	1.6	1.5	1.7	1.5	1.4	1.0
9-Oct	48	1.4	1.5	1.3	1.1	1.0	0.9
9-Oct	56	0.4	0.5	0.4	0.6	0.5	0.2
10-Oct	64	0.2	0.2	0.1	0.2	0.2	0.2
10-Oct	72	0.1	0.1	0.1	0	0	0
11-Oct	96	0	0	0	0	0	0

Application techniques and/or herbicide formulations need to be further developed to maximize concentration retention under various field conditions and dilution patterns. Additional research needs to be conducted to compare the dissipation pattern of herbicides from different delivery systems and application techniques to optimize concentration and exposure times under field conditions. Research should include evaluating the influence of plant density or height on vertical herbicide distribution and dissipation from the treated area; treatment areas that are exposed to immediate dilution from untreated water (e.g., shoreline areas or open water areas) versus those that are more protected from dilution (e.g., coves); and impacts of temperature stratification. Various delivery systems should be evaluated, including controlled-release and fast- or quick-release herbicide formulations; and liquid herbicide surface application, sub-surface injection, or deep-water injection.

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