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Effectiveness of Integrated Best Management Practices on Mitigation of Atrazine and Metolachlor in an Agricultural Lake Watershed

Richard Lizotte[1] · **Martin Locke**[1] · **Ronald Bingner**[1] · **R. Wade Steinriede**[1] · **Sammie Smith**[1]

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Abstract The study examined the influence of land-use (cropping patterns) and integrated agricultural best management practices (BMPs) on spring herbicide levels in an agricultural watershed. Atrazine and metolachlor were applied for weed control during spring of 1998–2002, 2005, and 2007–2013. Watershed-wide mass of applied herbicides ranged from 12.7 to 209.2 g atrazine and 10.9–302.2 g metolachlor with greatest application during 1998, 2009–2010 (atrazine) and 2007–2013 (metolachlor). Spring herbicide concentrations in Beasley Lake water ranged from below detection to 3.54 µg atrazine/L and 3.01 µg metolachlor/L. Multiple linear regression analyses with cropping patterns, BMPs, rainfall and time as independent variables, showed atrazine applications were associated with increases in cotton acreage and quail buffer, while metolachlor applications increased over time. Multiple linear regressions showed lake atrazine concentrations were associated with conservation tillage, rainfall, and corn, while lake metolachlor concentrations were associated with the cumulative metolachlor application and sediment retention pond installation.

Keywords Herbicides · Buffers · Conservation tillage · Cropping patterns · CEAP

The use of herbicides is a key component of modern row-crop agriculture necessary to meet food and fiber demands of a growing global population (Tilman et al. 2011). Such use, however, may come at a cost of potential impacts to non-target fish and wildlife resulting in a need for better understanding of the effectiveness of integrated conservation management strategies (Reichenberger et al. 2007). Two of the most commonly used herbicides, atrazine (2-chloro-4-ethylamino-6-isopropylamino-S-triazine) and metolachlor [2-chloro-*N*-(2-ethyl-6-methylphenyl)-*N*-(2-methoxy-1methylethyl) acetamide] (Table 1), can be used on a variety of crops in the United States but are commonly used on corn (*Zea mays* L.), sorghum (*Sorghum bicolor* L. [Moench]), and soybeans (*Glycine max* [L.] Merr.) (Geier et al. 2009; Murphy and Coats 2011; Williams et al. 2011). Both herbicides can be applied preemergence in the spring for control of a variety of broadleaf weeds and grass (Geier et al. 2009; Williams et al. 2011), and usage is likely to increase in response to increasing loss of glyphosate efficacy due to globally growing glyphosate resistance in weeds (Webster and Sosnoskie 2010).

As part of a broad national plan to address concerns of improving water quality through the use of agricultural best management practices (BMPs), in 2003 the US Department of Agriculture (USDA) agricultural research service (ARS) and the USDA Natural Resources Conservation Service (NRCS) worked in tandem to nationally assess the efficacy of BMPs in a partnership known as the conservation effects assessment project (CEAP). One such agricultural CEAP watershed is Beasley Lake, an oxbow lake located in Sunflower County, western Mississippi within the Mississippi River drainage basin known as the Delta. The Delta is intensively cultivated with about 36% of the landscape in row-crop production leading to widespread stress of numerous waterbodies resulting from influxes of suspended sediments, nutrients, and pesticides (Brown and Froemke 2012). The study watershed is especially well-suited to address CEAP goals since Beasley Lake

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✉ Richard Lizotte
richard.lizotte@ars.usda.gov

[1] USDA-ARS, National Sedimentation Laboratory, Oxford, MS 38655, USA

Table 1 Physical and chemical properties of atrazine and metolachlor applied in Beasley Lake watershed from 1998 to 2013

Property	Atrazine[a,b,c]	Metolachlor[d,e,f]
Pesticide use	Herbicide	Herbicide
Class	Triazine	Acetanilide
Molecular formula	$C_8H_{14}ClN_5$	$C_{15}H_{22}ClNO_2$
Density @20°C (g/mL)	1.19	1.12
Water solubility @26°C (mg/L)	34.7	488
K_{ow} coefficient	2.61	3.13
K_{oc} coefficient	11.6	1.48
pK_a value	1.6	None
Soil half-life ($t_{½}$)	>100 days	17 days
Water half-life ($t_{½}$)	58 days	47 days
1° degradation pathways	Microbial Chemical Hydrolysis	Microbial Photolysis

[a] Pubchem (2016a)

[b] EXTOXNET (1996a)

[c] Bouldin et al. (2006)

[d] Pubchem (2016b)

[e] EXTOXNET (1996b)

[f] Moore et al. (2001)

included a long-term database (7 years) prior to 2003 that included a focus on assessing lake water quality as a function of structural BMP implementation (Locke et al. 2008). In support of the goals of the national CEAP assessment, the current study assessed the effectiveness of watershed-wide integrated agricultural BMPs on spring lake atrazine and metolachlor concentrations in conjunction with known applications of both herbicides within the study watershed. The study incorporated a pesticide database of 16 years (1998–2013) concomitant with land-use database (cropping patterns) and the implementation of multiple and varied BMPs. Such a database allowed for an empirical assessment of how integrated BMPs, land-use, and pesticide use can be managed to improve and sustain lake water quality.

Materials and Methods

Beasley Lake watershed was first established as a research site in 1995 with monthly pesticide monitoring established in 1998 as part of the Mississippi Delta Management Systems Evaluation Area (MDMSEA) to assess the value of structural BMP placement on lake water quality (Smith and Cooper 2004). The study watershed is located at latitude 33°24′15″N, longitude 90°40′05″W and is comprised of a 22–27 ha isolated oxbow lake with a hydrologic drainage area of 625 ha with 141 ha of non-arable riparian bottomland hardwood forest (Fig. 1). The remaining watershed is comprised of 484 ha of arable land that has been in row-crop production for four primary crops: cotton (*Gossypium hirsutum* L.), soybeans, corn, and sorghum.

Beginning in 1996 through 2010, a total of six major independent BMPs comprising approximately 24% of arable land were voluntarily implemented by several farmers. During 1996, two structural BMPs were implemented. First, modified drainage pipes with slotted board risers at the culvert inlet directly upstream from strategic flow points. These allowed for reduced flow rates entering the pipes and settling of suspended sediment were installed (Locke 2004). In conjunction, edge-of-field vegetative buffer strips (VBS), initially encompassed 2.9 ha along the west side of the lake prior to 1995 (Fig. 1) followed by an additional 1.6 ha of VBS implemented in 1995 that was comprised of switchgrass (*Panicum virgatum* L.) or fescue (*Festuca arundinacea* Schreb.) (Locke et al. 2008). A further addition of 4.7 ha of VBS comprised of bahiagrass (*Paspalum notatum* Flugge) was planted in 2001 in the southern portion of the watershed. Second, from 2002 to 2004, 2006, and 2008–2009, conservation tillage (CT) soybeans were the primary row-crop (52–84% of arable land). Third, a 1.1 ha constructed wetland consisting of a sediment trap and two treatment cells was installed in 2002 near the east south east lake shoreline (Locke et al. 2011). Fourth, in 2003–2004, 87 ha of arable land north of the lake was removed from row-crop production and planted in eastern cottonwood trees (*Populus deltoides* Bartr. Ex. Marsh.), oak trees (*Quercus* sp.), and hickory trees (*Carya* sp.) as part of the Conservation Reserve Program (CRP) (Locke et al. 2008). Within CRP, the constructed wetland and about 1.1 ha of VBS was subsumed into CRP north and east of the lake (Fig. 1). Fifth, in 2006, about 9 ha of arable edge-of-field land south of the lake was removed from row-crop production and converted to vegetative buffer habitat to attract northern bobwhite quail (*Colinus virginianus*) (QB). In contrast to VBS, QB is comprised of a mixture of annual and perennial plants, primarily eastern cottonwood trees (*Populus deltoides* Bartr. Ex. Marsh.), oak trees (*Quercus* spp.), goldenrod (*Solidago* spp.), Canadian horseweed [*Conyza canadensis* (L.) Cronquist], and little bluestem (*Schizachyrium scoparium*) (Fig. 1). Finally, in 2010, about 1 ha of edge-of-field land was converted into a vegetated two-cell sediment retention pond (SRP) with a hydraulic retention time (HRT) of 1–2 days that received agricultural runoff from ditches draining land south and west of the lake (Fig. 1).

Annual cropping patterns and pesticide application data were collected with the cooperation of local farmers. Available arable land within the watershed from 1998 to 2013 were converted into area (ha) using geographic information systems (GIS) Spatial Analyst extension of Arc GIS software (ESRI Inc., Redlands, CA). Beasley Lake surface

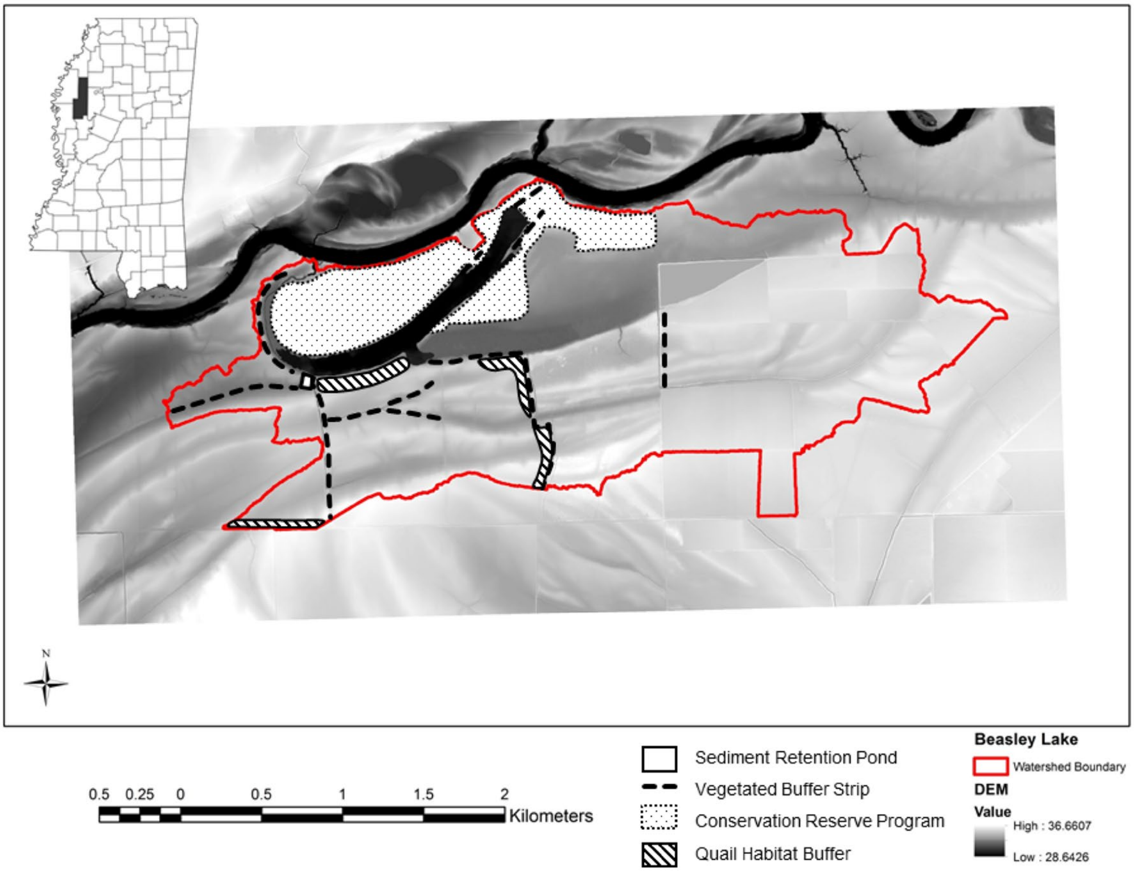

Fig. 1 Map of Beasley Lake watershed showing agricultural best management practices of vegetated buffer strips, conservation reserve program, quail buffer, sediment retention pond, and constructed wetland

water samples (4 L) were collected monthly from 1998 to 2013 at the midpoint of the lake. Samples were extracted and preserved on site with the addition of 4 g of KCl and 400 mL (0.106 gal) of pesticide-grade ethyl acetate and manual mixing for 1 min. Samples were placed on wet ice and transported to the USDA-ARS National Sedimentation Laboratory, Oxford, MS for pesticide analysis. Samples were analyzed using two Agilent HP model 6890 gas chromatographs (Agilent Technologies, Inc.,Waldbronn, Germany) equipped with dual Agilent HP 7683 ALS autoinjectors, dual split-splitless inlets, dual capillary columns, an Agilent HP Kayak XA Chemstation, and the autoinjector set at 1.0 µL injection volume fast mode were used for target herbicides, atrazine and metolachlor. Based on fortified samples, herbicide recoveries and extraction efficiencies were ≥90%, and detection limits were 0.125 and 0.0125 µg/L for atrazine and metolachlor, respectively. A more comprehensive description of pesticide analytical methods is available in Lizotte et al. (2014).

Data analysis was conducted on herbicide data from spring samples (March 20–June 20) coinciding with periods of spring herbicide applications (Tables 2, 3). Spring herbicide application data and lake concentration data were analyzed to assess temporal and integrated spatial effects of cropping patterns, BMPs, and spring rainfall (cumulative amount 7 days prior to sampling mm and intensity mm/h). A series of linear or multiple linear regression analyses were performed. If data failed to meet assumptions of normality, log-normal transformations or rank-transformations were used prior to regression analyses (Conover and Iman 1981).

Results and Discussion

Spring atrazine applications in Beasley Lake watershed occurred primarily in April with only two applications occurring in March and May. All applications were limited to three crops, soybean, corn, and sorghum predominantly in the eastern portion of the watershed (Table 2; Fig. 1). Application rates varied from 1.17 to 5.85 L ha and area of application ranged from 8.99 to 98.91 ha. Greatest total applied atrazine mass was 209.19 g occurring on 5/15/1998 with corn across three different regions

Table 2 Spring (March 20–June 20) atrazine application dates, crops, locations, rates, area, and total mass in Beasley Lake watershed 1998–2013

Date	Crop	Location	Rate L/ha	Area ha	Total mass[a] g
4/4/1998	Corn	East	1.17	37.57	20.58
4/13/1998	Corn	East	2.34	16.40	17.92
5/15/1998	Corn	North, South, West	4.68	95.45	209.19
4/12/1999	Sorghum	East	2.34	54.96	60.22
4/13/1999	Corn	East	3.51	8.99	14.78
4/4/2001	Sorghum	East	3.12	17.65	19.98
5/1/2001	Sorghum	East	3.51	28.91	47.53
4/7/2002	Sorghum	East	3.74	32.81	57.54
4/15/2002	Sorghum	East	1.75	51.87	42.63
4/20/2005	Sorghum	East	3.74	40.64	71.26
4/20/2005	Sorghum	East	2.34	72.95	79.92
4/22/2005	Sorghum	East	1.75	33.66	27.66
4/20/2007	Sorghum	East	1.75	81.84	67.27
4/22/2007	Sorghum	East	2.34	77.61	85.03
4/27/2007	Sorghum	East	3.74	40.64	71.26
3/25/2009	Corn	South, West	2.34	98.91	108.37
4/19/2010	Corn	South, West	2.34	98.91	108.37
4/9/2011	Sorghum	East	2.34	11.63	12.74
4/25/2012	Corn, Soybean	East	2.34	51.87	56.87
3/20/2013	Soybean	East	5.85	40.64	48.67

[a] Total mass was determined based upon application rate, g active ingredient per liter for the trademark product label, and application area

of the watershed. Spring Beasley Lake water atrazine concentrations above detection limits ranged from 0.132 to 3.539 µg/L. Within Europe, concentrations are not to exceed 1 µg atrazine/L (European Commission 2003), while Canadian guidelines to protect aquatic life limit atrazine to 1.8 µg atrazine/L (CCME 1999a). Lake atrazine concentrations above 1 µg/L occurred on 4/21/1999, 5/31/2000, 4/16/2007, 5/3/2010, 6/1/2010, 4/22/13, and 5/20/2013 and above 1.8 µg/L on 4/21/1999, 4/16/2007, 6/1/2010, and 4/22/13 (Fig. 2). A multiple linear regression model using spring atrazine application area showed approximately 47% of the atrazine application area could be significantly explained by a combination of cotton cropping patterns and QB ($r^2=0.467$, $p=0.005$, n = 20). The model produced a negative slope for both variables ($t=-3.33$ and -2.25). This indicated an increase in atrazine application area with a concomitant shift in cotton acreage to conservation tillage soybeans and QB as observed in the mid-western corn-belt where conservation tillage resulted in less water runoff and atrazine loss (Gourneau et al. 2001). A multiple linear regression model using spring lake atrazine concentrations showed approximately 62% of these data could be significantly explained by a combination of CT, corn acreage, rainfall amount, and rainfall intensity ($r^2=0.618$, $p=<0.001$, n = 39) with a negative slope for CT ($t=-2.61$) and rainfall intensity ($t=-3.53$) a positive slope for corn ($t=5.05$) and rainfall ($t=4.85$) indicating decreases in atrazine lake concentrations with increasing CT area and rainfall intensity and with decreased corn acreage and 7 d rainfall. In comparison, spring metolachlor applications in Beasley Lake watershed occurred in April and May with only two applications in March and one in June.

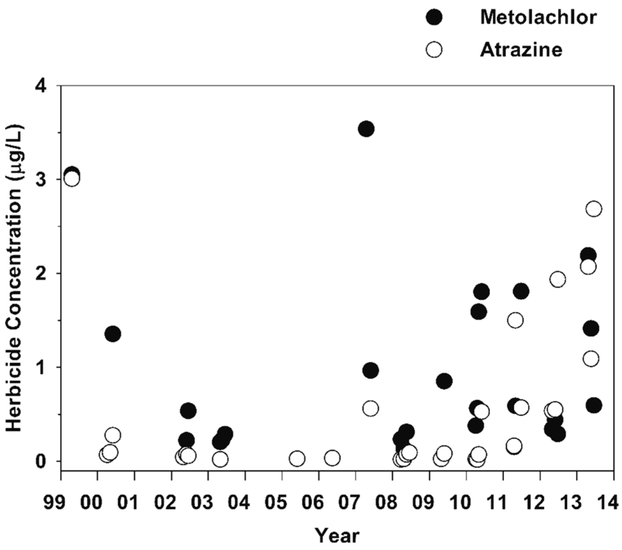

Fig. 2 Spring (March 20–June 20) atrazine and metolachlor concentrations in Beasley Lake 1998–2013

Table 3 Spring (March 20–June 20) metolachlor application dates, crops, locations, rates, area, and total mass in Beasley Lake watershed 1998–2013

Date	Crop	Location	Rate L/ha	Area ha	Total Mass[a] g
4/1/1998	Corn	North	2.93	51.84	141.13
5/8/1998	Corn, Cotton	East	0.47	25.00	10.89
3/23/1999	Corn	East	1.76	8.99	15.91
4/12/1999	Sorghum	East	1.76	57.89	94.56
4/27/2000	Cotton	East	1.17	31.65	34.47
5/1/2001	Sorghum	East	1.17	28.91	31.37
5/3/2001	Cotton	East	0.37	40.64	13.83
4/7/2002	Sorghum	East	1.17	32.81	35.73
4/15/2002	Sorghum	East	1.17	45.32	49.18
4/20/2005	Sorghum	East	1.76	40.64	66.38
4/20/2005	Sorghum	East	0.94	75.90	71.63
3/26/2007	Corn	South, West	2.34	136.73	297.78
4/20/2007	Sorghum	East	1.17	81.84	87.43
4/22/2007	Sorghum	East	0.94	77.61	67.61
4/27/2007	Sorghum	East	1.76	40.64	66.38
5/10/2008	Soybean	South, West	2.34	139.70	304.24
4/15/2009	Soybean	South, West	2.34	98.91	215.42
5/3/2010	Soybean	South, West	2.34	26.30	57.29
4/9/2011	Sorghum	East	1.17	11.63	12.66
4/14/2011	Soybean	East	2.34	65.99	79.38
5/9/2011	Cotton	South, West	1.17	51.68	56.27
5/15/2011	Soybean	East	2.34	40.64	48.89
5/16/2011	Soybean	South, West	2.34	88.00	191.66
4/15/2012	Soybean	East	4.68	40.64	97.78
4/25/2012	Corn	East	8.18	51.87	96.37
5/9/2012	Soybean	East	4.68	25.75	61.95
5/18/2013	Soybean	South, West	0.88	139.70	114.01
6/20/2013	Soybean	East	1.46	77.61	36.41

[a] Total mass was determined based upon application rate, g active ingredient per liter for the trademark product label, and application area

Applications occurred on all four crop types (cotton, soybean, corn, and sorghum) however, most applications were with sorghum and soybeans and, as with atrazine, primarily in the eastern region of the watershed (Table 3; Fig. 1). Spring metolachlor application rates ranged from 0.37 to 8.18 L ha and application area ranged from 8.99 to 139.70 ha. Greatest total applied spring metolachlor mass was 304.24 g occurring on 5/10/2008 with soybeans across the south and west regions of the watershed. Lake water metolachlor concentrations above detection limits ranged from 0.016 to 3.01 µg/L. Canadian guidelines to protect aquatic life limit metolachlor to 7.8 µg/L (CCME1999b). Metolachlor concentrations were never >7.8 µg/L with the greatest concentration of only 3.01 µg metolachlor/L occurring on 4/21/1999 (Fig. 2). A multiple linear regression model with spring metolachlor application area showed about 41% of metolachlor application area could be significantly explained by increased metolachlor use over time and in conjunction with installment of SRP ($r^2=0.407$, $p<0.001$, $n=28$) with a positive slope for time ($t=4.04$) indicating an increase in metolachlor application area over time and a negative slope for SRP ($t=-3.53$). Such results are a response, in part, to increases in glyphosate resistant spring weed populations (Webster and Sosnoskie 2010) such as henbit deadnettle (*Lamium amplexicaule* L.) that occur in row crop acreage within Beasley Lake watershed (Locke pers. comm.) and are known to be a relatively important nuisance weed in the southeast (Riar et al. 2013). Additionally, the SRP had minimal established vegetation with a short HRT providing no measureable mitigation. A multiple linear regression model using spring lake metolachlor concentrations showed 65% of these data could be significantly explained by a combination of both metolachlor application rates and SRP ($r^2=0.650$, $p=<0.001$, $n=27$) with positive slopes for both metolachlor application rates ($t = 3.83$) and SRP ($t = 3.55$) indicating increases in lake metolachlor concentrations with increasing metoalchlor application coinciding with installment of the SRP.

Measured atrazine and metolachlor concentrations in Beasley Lake surface water during the 16-year study period are comparable to those in other agriculturally impacted oxbow lakes assessed within the lower Mississippi River alluvial plain and ranged up to 10 µg atrazine/L and 20 µg metolachlor/L (Senseman et al. 1997; Smith and Cooper 2004). Several variables can influence the mobility of post-application herbicides in runoff such as physico-chemical properties of the specific herbicide (Smith and Cooper 2004); rainfall timing, amount, and intensity (Rector et al. 2003; Leu et al. 2004; Shaw et al. 2006); soil properties (Alletto et al. 2010); and land-management practices (Rector et al. 2003). Atrazine is a moderately water soluble herbicide ($K_{ow} = 2.61$) with a higher likelihood of binding to soil ($K_{oc} = 11.6$). In comparison, metolachlor is highly water soluble ($K_{ow} = 3.13$) and less likely to bind to soil ($K_{oc} = 1.48$). Both herbicides have aqueous half-lives > 30 days but atrazine in soil is (>100 days) much longer than metolachlor (17 days). Primary degradation pathways for both herbicides are microbial. However, atrazine is also preferentially degraded through chemical and hydrolytic pathways while metolachlor is also degraded preferentially through photolysis (Table 1). To try to alleviate this potential problem, a number of studies within the past decade have attempted to ascertain the effectiveness of a variety of different BMPs in mitigating herbicide runoff (O'Donnell 2012) including vegetative buffers (Krutz et al. 2005; Pätzold et al. 2007; Dunn et al. 2011), conservation tillage practices (Mickelson et al. 2001; Shipitalo et al. 2006; Alletto et al. 2010), and alternative farming practices such as split applications (Harman et al. 2004; Shipitalo et al. 2006). Several studies have shown that these buffers effectively mitigate atrazine and metolachlor for three reasons. First, vegetated buffers retain herbicides through infiltration in the buffers. Second, atrazine ($K_{oc} = 11.6$) can adsorb to organic carbon which is elevated in VBS. Third, microbial degradation of metolachlor is enhanced in VBS due to greater microbial diversity in vegetated buffers versus bare tilled soil (See: Kurtz et al. 2005). The present study indicated atrazine and metolachlor in lake surface water was influenced by a combination of either CT + corn + rainfall (atrazine) or metolachlor application and SRP installation (metolachlor) however only CT showed evidence of lake herbicide mitigation (negative slope). The incongruent results for atrazine and metolachlor are likely indicative of two factors. First, although SRP was installed in 2010, it takes time to establish vegetation in recently constructed buffered areas (Krutz et al. 2005; Pätzold et al. 2007; Dunn et al. 2011). SRP in Beasley Lake watershed was not fully established until several years after installation (Locke pers. comm.) and during this time atrazine and metolachlor application area increased, including fields proximate to SRP. Second, SRP had a relatively short HRT limiting the BMPs ability to mitigate herbicides. While fully established vegetative buffers like SRP have been shown to be effective in reducing herbicide runoff in plot-level studies (Krutz et al. 2005; Pätzold et al. 2007; Dunn et al. 2011), BMP effectiveness at the watershed scale is predicated upon herbicide-laden runoff flowing directly through the buffer with a long enough duration to mitigate these pollutants. Within Beasley Lake watershed most of the herbicide applications occurred east of the lake (Tables 1, 2) where BMPs were minimal (Fig. 1) and herbicide loads were likely transported primarily via drainage ditches and circumventing most BMPs resulting in limited benefits. In contrast, watershed wide CT appeared to mitigate herbicide loads into Beasley Lake. Previous studies assessing the ability of CT to mitigate herbicide runoff reported mixed results with some studies showing CT as an effective BMP (Alletto et al. 2010) and others showing CT to be less effective or even ineffective (Mickelson et al. 2001; Shipitalo et al. 2006). Alletto et al. (2010) suggested the effectiveness of CT in mitigating herbicide runoff is influenced by a number of variables with soil properties, available crop residues, and herbicide physico-chemical properties being among the most important. Within Beasley Lake watershed, CT was clearly the most effective BMP in mitigating atrazine and metolachlor concentrations within lake surface water. Watersheds within the Mississippi Delta region with similar conditions could use CT as part of an integrated suite of BMPs to reduce herbicide loads and help improve overall lake water quality.

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