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Surface Water, Ground Water and Sediment Quality in Three Oxbow Lake Watersheds in the Mississippi Delta Agricultural Region: Pesticides

CHARLES COOPER*, SAMMIE SMITH, JR. AND MATT MOORE
USDA-ARS-National Sedimentation Laboratory, 598 McElroy Drive, P.O. Box 1157, Oxford, MS 38655-1157
*(*Corresponding Author, E-mail: ccooper@ars.usda.org*)

ABSTRACT

We measured residual and current use pesticides in shallow groundwater, surface water, and lake sediment in three oxbow lakes and their watersheds in the intensively cultivated alluvial plain of the Mississippi River, USA. The three-year study focussed on providing knowledge of pesticide movement and concentrations from intensive agricultural production into aquatic ecosystems and evaluating the degree of contaminant deposition and persistence in oxbow lake sediments. Penetration of insecticides into shallow groundwater did not represent a hazard. Herbicides and insecticides were found in surface water as a result of rainfall-related runoff from farm fields. A common herbicide, atrazine, had the largest number of detections (28% of lake water samples). Sediment cores taken for pesticide analysis were also viewed by age in each lake. Residual organochlorines dominated the pesticide sediment profile. DDT and its metabolites were the only compounds that permeated sediments at all sites. Sediments deposited in the 1960s and 1970s which contained higher concentrations of DDT have been effectively sealed by more recent material so that the sink of contaminant is unavailable to the water column. Controlled release of pyrethroid insecticides in a forested wetland showed that the wetland was totally effective in neutralizing the pyrethroids before they reached lake surface water. The lakes are designated for fish and wildlife use, but pesticides seldom exceeded more stringent drinking water standards. Acute toxicity concerns lessen as more conservation measures are installed on and off fields. Additionally, DDT metabolites continue to slowly degrade. A suite of protective measures is necessary to protect downstream water resources from runoff-related contamination.

Key Words: Ecosystem contamination, Dating, Organochlorines, Agricultural runoff, Toxicity.

INTRODUCTION

Oxbow lakes, common floodplain features around the globe, are remnants of meandering floodplain rivers that have been physically isolated from their respective main river channels and generally capture small relic drainages. In temperate and tropical zones where agriculture is prevalent, oxbows provide measures of recent and historical off-site damages from agriculture because of drainage patterns and trapping phenomena. Isolation has resulted in physical and chemical changes in lake basins and in the floral/faunal assemblages present. Over time, allochthonous organic materials have been processed and energetically depleted, resulting in the lakes being less heterotrophic and more autotrophic. If suspended sediment concentrations are low enough to allow suitable light penetration, isolated oxbow lakes provide conditions conducive to photosynthesis, primarily via phytoplankton, and may support sustainable fisheries production. However, decades of traditional agricultural practices, including clean tillage and no winter cover on land surrounding oxbow lakes, often result in continuous high lake turbidity from fine sediment transported in runoff. Hot, humid climatic conditions and a long growing season make the Mississippi Delta, USA ideal for intensive row crop production, primarily cotton (*Gossypium hirsutum* L.), soybeans [*Glycine max* (L.) Merr.], rice (*Oryza sativa* L.), and corn (*Zea mays* L.). These same conditions provide for enhanced weed growth and insect infestations, resulting in the need for intensive agrochemical pest control measures. Some areas have had pesticides applied for more than 150 years. In addition, runoff often transports agrochemicals into the lakes causing further reductions in water quality (Leonard 1988). Consequently, many

Delta oxbow lakes, long known for their fish productivity and recreational value, have become unattractive in water quality and water clarity. In the 1970s, some Delta lakes were closed to fishing because of high concentra-tions of DDT. Recently, a fish flesh advisory was issued by the Mississippi Department of Environmental Quality for selected Delta lakes because of high levels of pesticides in fish. Contamination of ground water, especially by herbicides (Williams et al. 1998; Ritter et al. 1991), is always a concern where pesticides are applied.

The Mississippi Delta Management Systems Evaluation Area project (MDMSEA) began in 1995 as part of a national water quality research program conducted by a consortium of Federal, State, and local agencies. Two objectives of the MDMSEA project include: (1) development and evaluation of economical, environmentally-sound best management practices (BMPs) and alternative and innovative farming systems for improved water quality/ecology in the Mississippi Delta and (2) assessment of the effects of environmentally-sound agricultural activities on surface/shallow ground water quality and the environment.

Development of region-specific alternative farming systems (composed of combinations of selected BMPs) is crucial to protecting the surface and ground water resources and improving ecological and environmental quality of the entire Mississippi Delta. Research is needed on contaminant routing and processing in these alluvial plain agricultural watersheds because the potential for agrichemical percolation (leaching) to relatively shallow water tables, which are hydraulically connected to nearby lakes and rivers, is poorly understood. The effectiveness of riparian zones or wetlands to trap sediment and to trap and process agrochemicals in runoff is also poorly defined. Essentially nothing is known about the ability of Delta lakes to recover and/or to sustain fisheries production after sediment and agrichemical inputs are permanently reduced. The purpose of this water quality study is to provide knowledge of pesticide detections in shallow ground water and lakes resulting from intensive agricultural production and to evaluate the degree of contaminant deposition and persistence in oxbow sediments. The ultimate goal of this research is to provide information that will lead to recommendations that foster ecologically healthy lakes and streams with sustainable fisheries.

MSEA Project Description

The project design involves a hierarchy of BMPs in small watersheds located in Sunflower and Leflore counties in west-central Mississippi, USA (Figure 1). A detailed project description, location and watershed use information, and BMP selection process is located in Rebich and Knight (2001). The watersheds are "closed systems." Unlike flow-through systems, each watershed originates flow which drains into an oxbow lake.

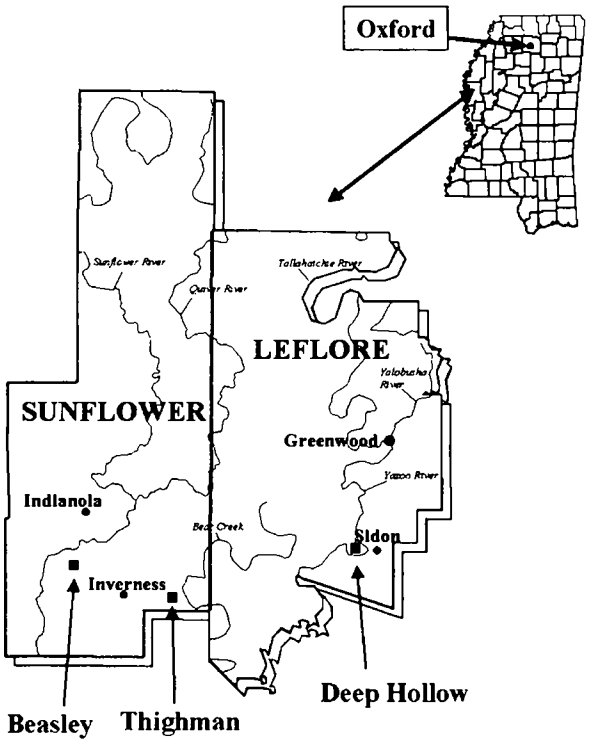

Figure 1. Map showing location of watershed sites of the MDMSEA project in west-central Mississippi.

Beasley Lake watershed (Figure 2; south of Indianola, Mississippi in Sunflower County also) received nominal BMP treatment consisting of grade stabilization and water control structures, including slotted-board water control structures, slotted-inlet pipes (field drainage pipes that have a partial barrier in front of the inlet to slow water and deposit sediment), overfall pipes (pipes that step water down to a lower level from a field to prevent gully formation), and culverts. Numerous grass filter strips have been established along major drainages into the lake. An existing large forested wetland adjoining the lake provided additional natural water treatment. Deep Hollow Lake watershed (Figure 3; near Sidon in Leflore County, Mississippi) received an intensive BMP effort of winter wheat cover crop and conservation-tillage for all selected cotton and soybean fields. Pioneering weed sensor technology reduced

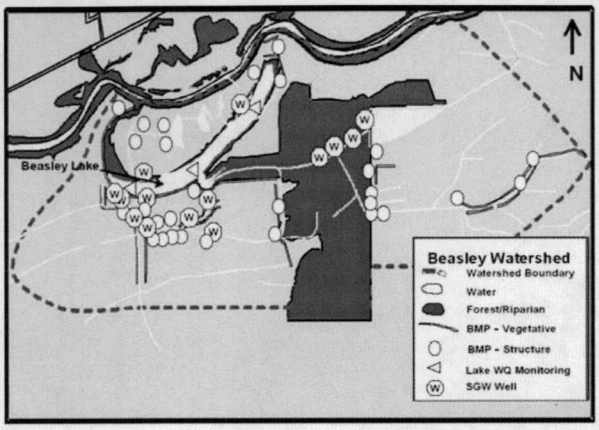

Figure 2. Shallow groundwater well locations in Beasley Lake watershed.

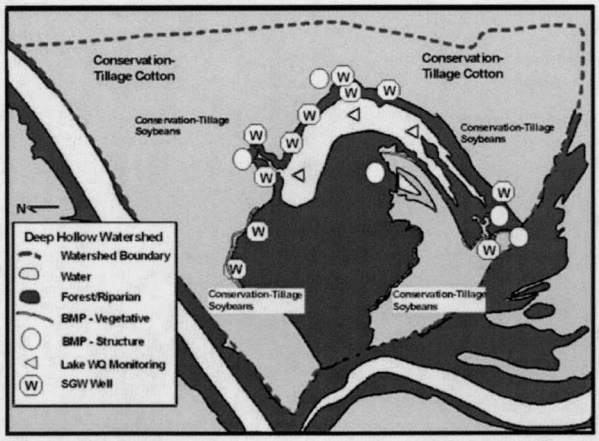

Figure 3. Shallow groundwater well locations in Deep Hollow Lake watershed.

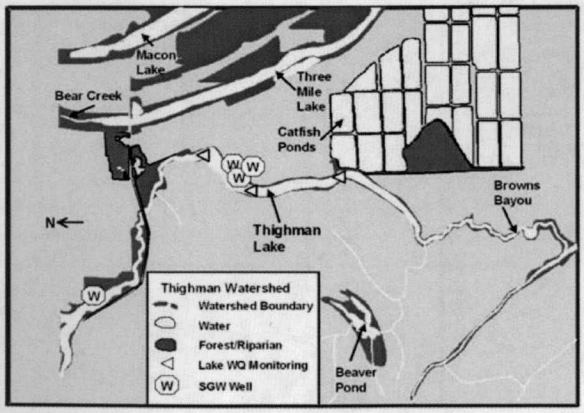

Figure 4. Shallow groundwater well locations in Thighman Lake watershed.

herbicide input. Additionally, grass filter strips and stiff grass hedges along with slotted-board riser pipes and slotted-inlet pipes were used at critical drainage locations. Thighman Lake watershed (Figure 4; south of Moorehead in Sunflower County, Mississippi) served as a control with no BMPs initially. Ultimate goals of the project were reduced sediment and agrochemical transport in runoff, improved oxbow lake water quality/ecology, sustained profitable crop yields, and enhanced, sustainable fisheries in the oxbow lakes.

MATERIALS AND METHODS

Well Placement and Installation

Observation wells for sampling shallow ground water were installed in clusters of 3 (about 1 m apart) at depths of 1.6 m, 3.2 m, and 4.8 m at critical flow areas in the watersheds (Figures 2, 3, and 4). Deep Hollow Lake watershed had 11 well sites. This watershed had the most MDMSEA project-implemented BMPs. The 11 well sites were at the structural BMPs which were designed to slow surface flow and result in increased infiltration (Figure 2). The other well sites in this watershed were deeper wells related to special hydrogeology research (Adams and Davidson 2001). The Thighman Lake watershed was originally intended to serve as reference watershed with initially no project-imposed BMPs. Thus, there were only 4 well sites (two in a small riparian area and two associated with major drainages into the lake). The Beasley Lake watershed had only structural BMPs imposed by the project. The 10 well sites (outside and west of the large forested wetland/riparian area) were primarily located at these structural BMPs and along field drainage ditches.

After 9 cm diameter holes were drilled, they were backfilled with pea gravel to the required well depth and a commercially available 5 cm diameter well (schedule 40 PVC with 30 cm of well screen) cut to a length of about 30 cm longer than the required depth was inserted into the hole. The hole around the outside of the well was backfilled first with about 30 cm of small, uniform sized gravel to encase the well screen in gravel. The rest of the hole around the outside of the well was then filled to the surface of the ground with commercially available 0.6 cm diameter bentonite pellets. Bentonite pellets were also packed around the outside of the well at the surface of the ground, covered with soil, and packed again. This provided a watertight casing around the well to prevent surface water (possibly containing dissolved agrochemicals) from seeping down the outside of the well.

Sampling Procedure

The sampling of shallow ground water was similar to that reported for a previous study (Smith et al. 1991). Usually within 24-h of a rainfall event, a 500 mL sample was collected from each well (using a battery-operated ISCO Acu-well™ model 150 portable pump fitted with a Teflon-lined intake line) in a 0.5 L amber bottle with Teflon-lined screw cap. Each well was pumped dry and the excess well water was discarded. Shallow ground water samples were placed on ice, immediately transported to the National Sedimentation Laboratory (NSL), and stored at 4°C (usually <72 h) for pesticide analyses via gas chromatography (GC).

A 4 L water sample was collected monthly from surface water at each of the oxbow lakes in a 10 L glass jar, fitted with a Teflon-lined screw cap. Samples were taken from the middle of each lake in the middle of the water column. Samples were extracted onsite by adding 4 g KCl and 400 mL pesticide grade EtOAc and shaking vigorously by hand for about 1 min, placed on ice, transported to the NSL, and stored at 4°C for GC analysis.

Initial sediment sampling was conducted with an Ekman dredge for sediment/water interface samples. Sediment cores were also taken to greatest depth possible with manual coring equipment from an anchored boat. Ten cm diameter sediment cores were driven, lifted into a clean semi-tubular ruled trough, and divided into incremental 10 cm sections by depth for detection of metals, pesticides, and other contaminants. Additional verification was done for sediment at depths to 20 cm in all three lakes.

Sediment samples were also collected from three sites in each of the three study lakes in 1997 for Cesium dating. Results allowed us to calculate sediment accumulation rates since atmospheric nuclear testing marker dates in 1954 or 1964. Cores were divided into incremental 10 cm sections by depth for cesium-dating analyses. A kg of sediment from each 10 cm depth increment was acquired for cesium dating (Ritchie et al. 1983).

Table 1. Chemical names of pesticides mentioned in this paper.

alachlor	(2-chloro-26,66-diethyl-N-methoxymethylacetanilide)
aldrin	[(1R,4S,4aS,5S,8R,8aR)-1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4:5,8-dimethanonaphthalene]
atrazine	(2-chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine) Aatrex
bifenthrin	[2-methylbiphenyl-3-ylmethyl (Z)-(1RS,3RS)-3-(2-chloro-3,3,3-trifluoroprop-1-enyl)-2,2-dimethylcyclopropanecarboxylate] Capture
chlorfenapyr	[4-bromo-2-(4-chlorophenyl)-1-(ethoxymethyl)-5-(trifluoromethyl)-1H-pyrrole-3-carbonitrile] Pirate
chlorpyrifos	(O,O-diethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate) Lorsban
cyanazine	[2-(4-chloro-6-ethylamino-1,3,5-triazin-2-ylamino)-2-methylpropionitrile]
cyfluthrin	[RS-α-cyano-4-fluoro-3-phenoxybenzyl(1RS,3RS)-cis,trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate] Baythroid
λ-cyhalothrin	{[1α(S*),3α(Z)]-cyano(3-phenoxyphenyl)methyl 3-(2-chloro-3,3,3-trifluoro-1-propenyl)-2,2-dimethyl-cyclopropanecarboxylate]} Karate
DDD	[1,1-dichloro-2,2-bis (p-chlorophenyl) ethane]
DDE	[1,1-dichloro-2,2-bis(p-chlorophenyl)ethylene]
DDT	[1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane]
deltamethrin	[(S)-α-cyano-3-phenoxybenzyl (1R,3R)-3-(2,2-dibromovinyl)-2,2-dimethylcyclopropanecarboxylate]
dieldrin	(1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a,octahydro-1,4,5,8-dimethanonaphthalene)
endosulfan	(6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiopin-3-oxide)
endrin	(1,2,3,4,10,10-hexachloro-1R,4S,4aS,5S,6,7R,8R,8aR-octahydro-6,7-epoxy-1,4:5,8-dimethanonaphthalene)
esfenvalerate	{[S-(R*,R*)]-cyano(3-phenoxyphenyl)methyl 4-chloro-α-(1-methylethyl)benzeneacetate} Asana XL
fipronil	[(RS)-5-amino-1-(2,6-dichloro-α,α,α-trifluoro-p-tolyl)-4-trifluoromethylsulfinylpyrazole 3-carbonitrile] Regent
fluometuron	[N,N-dimethyl-N'-(3-(trifluoromethyl)phenyl)-urea] Cotoran
heptachlor	(1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene)
methoxychlor	(2,2-bis(p-methoxyphenyl)-1,1,1-trichloroethane)
methyl parathion	(O,O-dimethyl-O-p-nitrophenyl phosphorothioate)
metolachlor	[2-chloro-6'-ethyl-N-(2-methoxy-1-methylethyl)acet-o-toluidide] Dual
norflurazon	[4-chloro-5-(methylamino)-2-(α,α,α-trifluoro-m-tolyl)-3(2H)-pyridazinone] Zorial
pendimethalin	[N-(1-ethylpropyl)-3,4-dimethyl-2,6-dinitrobenzenamine] Prowl
trifluralin	(α,α,α-trifluro-2,6-dinitro-N,N-dipropyl-p-toluidine) Treflan
zeta-cypermethrin	[(S)-α-cyano-3-phenoxybenzyl (1RS,3RS;1RS,3SR)-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate] Fury

Pesticide Analyses

The pesticides initially targeted for analysis in shallow ground water samples, along with their GC retention times, are shown in Table 1. Heptachlor, aldrin, endosulfan, dieldrin, endrin, methoxychlor and p,p'-DDT (metabolites p,p'-DDE and p,p'-DDD) are relatively persistent, chlorinated hydrocarbon insecti-cides with a history of common past use throughout the Mississippi Delta. The other compounds are generally less persistent herbicides and insecticides that were in current use in the MDMSEA watersheds in 1995. Analysis of ground water samples was similar to the method of Smith et al. (1994), with modifications by Bennett et al. (2000). Ground water samples were allowed to come to room temperature (about 25°C) and the volume measured and recorded. The entire sample was extracted by sonification (1 min/pulse mode/80% duty cycle) with 1-reagent-grade KCl and 100 mL pesticide-grade EtOAc, partitioning in a separatory funnel with the water phase discarded. The EtOAc phase was dried over anhydrous Na_2SO_4 and concentrated by rotary evaporation to near dryness. The extract was taken up in about 5 mL pesticide-grade hexane, subjected to cleanup by silica gel column chromatography, and concentrated to 1 mL for GC analysis. Mean extraction efficiencies, based on fortified samples, were >87% for all pesticides from shallow ground water.

Initially, Tracor™ model 540 gas chromatographs were equipped with Dynatech Precision™ GC-411V autoinjectors. A multi-level calibration procedure was used with standards and samples injected in triplicate. Calibration curves were updated every tenth sample. Tracor™ gas chromatographs were replaced in late 1998 with Hewlett Packard model 6890™ gas chromatograph systems. Both level of detection and level of quantification were reported for each pesticide (Table 2).

RESULTS AND DISCUSSION

Pesticides in Shallow Ground Water

Over the first three water years (WY) (1996-1998), a total of 622 well samples were collected. There were 103 well samples collected in WY96, 160 in WY97, and 359 in WY98. Of the 622 well samples collected, there were only five detections, and all were in WY96 (Table 3). All 5 detections were in the Beasley Lake watershed. Norflurazon was detected once at 0.4 $\mu g\ L^{-1}$, and metolachlor was detected 4 times at levels ranging from 2-8 $\mu g\ L^{-1}$. All detections were at extremely low levels and would be expected to present no water quality problems, as they were transient in nature. There were no detections in WY97 or in WY98. Thus, unlike problems encountered elsewhere (Iensee 1988), field applied compounds seldom moved into shallow ground water, and the project-imposed BMPs caused no shallow ground water quality prob-lems in any of the three MDMSEA watersheds. Pesti-cides leached into the soil profile were likely degraded/ processed in the biologically active upper soil horizons. Evaluation of shallow ground water quality in the MDMSEA watersheds continues at a reduced level. Current samples have provided no contradictory infor-mation to the results of our three-year intensive study.

Pesticides in Field Runoff and Lake Water

As part of the MDMSEA project, Shaw, Shankle, and Kingery (2001) studied the environmental fate of fluometuron in soil influenced by different best man agement practices. They concluded that soils collected by grass buffer strips have the potential to rapidly degrade fluometuron since its half-life was at least 48 days shorter than in cropped areas. Zablotowicz et al. (2001) conducted a companion study by characterizing micro-

Table 2. Targeted pesticides and their levels of detection (LOD) and quantitation (LOQ) (ng L^{-1}).

	Alachlor	Aldrin	Atrazine	Bifenthrin	Chlorfenapyr	Chlorpyrifos	Cyanazine	Cyfluthrin	Deltamethrin
LOD	0.5	1	1	0.1	0.5	0.1	0.5	0.1	0.1
LOQ	5	10	10	1	5	1	5	1	1
	Dieldrin	Endosulfan	Endrin	Esfenvalerate	Fipronil	Fipronil sulfone	Fluometuron	Heptachlor	l-Cyhalothrin
LOD	0.1	1	1	0.1	0.1	0.1	0.5	1	0.1
LOQ	1	10	10	1	1	1	5	10	1
	Methoxychlor	Methyl Parathion	Metolachlor	Norflurazon	p,p'-DDD	p,p'-DDE	p,p'-DDT	Pendimethalin	Tralomethrin
LOD	1	1	1	0.5	0.1	0.1	1	0.5	0.1
LOQ	10	10	10	5	10	1	10	5	1

Table 3. Pesticides in MDMSEA shallow ground water in water year 1996. Bold values are detections. Data columns (analytes) or rows (samples) with all values below detection levels have been omitted. Complete data table included 17 analytes and 103 samples (see text). ND = no detection. All concentrations in $\mu g\ L^{-1}$.

Lake	Date	Site	Depth (m)	Metolachlor	Norflurazon
Beasley	01/05/1996	B2	1.6	ND	0.409
Beasley	08/12/1996	B2	3.2	2.652	ND
Beasley	09/19/1996	B7	3.2	5.496	ND
Beasley	09/19/1996	B10	4.8	8.352	ND
Beasley	09/19/1996	B2	3.2	5.580	ND
Beasley	09/30/1996	B10	4.8	4.764	ND

biological activities that contribute to removal of pesticides in forested riparian wetlands and lake water.

As a major collaborator in MDMSEA, Mississippi United States Geological Survey (USGS) collected runoff samples from selected edge-of-field concentrated flow sites in all three watersheds in 1995. Their primary objective was to assess the effects of conventional and conservation tillage on runoff water quality (Rebich 1997, Rebich et al. 2001). Thus, they measured runoff concentrations of compounds that were applied in the fields that they sampled. In 1996, the Soil and Water Research Unit, Agricultural Research Service, Baton Rouge, LA joined them in analysis of specific organophosphates and pyrethroids. In 1998, they began to place focus on pyrethroids in field runoff. No λ-cyhalothrin was measured in 1996 or 1997. In 1998 and 1999, all concentrations were <50 ng L^{-1}. Cypermethrin concentrations were <400 ng L^{-1} in 1996, and none was detected in 1997. The greatest concentration measured in 1998 or 1999 was <400 ng L^{-1}. Cyfluthrin concentrations were < 600 ng L^{-1} in 1996 and 1997. They did not reach 125 ng L^{-1} in 1998 and 1999. Deltamethrin concentrations remained below 125 ng L^{-1} in 1998 and 1999 as well. The study attempted to measure two organophosphates from the MSEA watersheds in 1996-1998. Measured methyl parathion concentrations were never above the quanti-tative detection limit (<200 ng L^{-1}) and azinphosmethyl was only detected 3 times; all were present in traces (<500 ng L^{-1})(Rebich et al. 2001). Of the 5 herbicides measured at 9 sites (fluometuron, norflurazon, cyanazine, atrazine, and metolachlor), they found that fluometuron and cyanazine occasionally exceeded drinking-water criteria in runoff (Rebich 2001). The results of USGS runoff-generated pesticide findings measured from edge-of-field samples were consistent with our lake surface water detections. Their measurements showed only minor concentrations of pesticides in runoff during the project.

Results of our pesticide analyses on monthly lake water samples (1998-2000) collected from the three oxbow lakes are presented in Table 4. During the period 1998-2000, 104 monthly lake water samples were collected. There were 90 pesticide detections above the 0.10 $\mu g\ L^{-1}$ level. Of these, 78 detections were herbicides, and 12 were pyrethroid insecticides. Atrazine was measured in 28 lake water samples, alachlor in 2, metolachlor in 22, and cyanazine in 26. With regard to the pyrethroid insecticides in lake water samples, λ-cyhalothrin was detected once, cyfluthrin once, zeta-cypermethrin 7 times, and esfenvalerate 3 times.

Annual variation of pesticides in surface water was dependent on rainfall pattern. The local weather pattern produced little runoff from agricultural fields during the 1998 cropping year. Runoff in October and November of 1998 produced the year's only contamination-producing events (Figure 5). The 1999 and 2000 cropping years had more runoff events spaced throughout the year. Concentration peaks followed spring and fall rainfall events in 1999. In 2000, many compounds were present almost all year in low (ng L^{-1}) concentrations. Atrazine concentrations in the surface water of Beasley Lake presented an example of how amount and dispersal of rainfall affected lake concentrations. We measured atrazine on 2 of 10 sampling days in 1998, 7 of 12 sampling days in 1999, and 12 of 13 sampling days in 2000.

Atrazine, a corn and cotton herbicide, was detected in both Beasley and Thighman Lake surface water. In 1998, it was measured in the fall after runoff (highest concentration was 1.57 $\mu g\ L^{-1}$). In 1999 and 2000, it was present in surface water during the cropping season (highest concentration was 7.183 $\mu g\ L^{-1}$) and after fall runoff (highest concentration was 2.433 $\mu g\ L^{-1}$) in both lakes. Deep Hollow Lake had no corn production and, thus, no atrazine. Cyanazine showed a pattern of detection consistent with runoff for all

Table 4. Pesticides in MDMSEA Lake Water. Bold values are >0.10 $\mu g\ L^{-1}$. Data columns (analytes) or rows (samples) with all values >0.01 $\mu g\ L^{-1}$ have been omitted. Complete data included 21 analytes and 104 samples (see text). ND = no detection. All concentrations in $\mu g\ L^{-1}$.

Lake	Date	Atrazine	Methyl Parathion	Alachlor	Metolachlor	Cyanazine	Pendimethalin	pp'-DDE	Bifenthrin	l-Cyhalothrin	Cyfluthrin	Z-Cypermethrin	Esfenvalerate
Beasley	08/18/1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.003	**0.180**
Beasley	10/09/1998	**0.275**	ND	ND	**0.314**	**0.757**	ND	ND	ND	ND	ND	0.003	0.006
Beasley	11/28/1998	0.121	ND	ND	0.048	**0.506**	ND	ND	ND	ND	ND	0.003	0.001
Beasley	01/06/1999	ND	ND	ND	ND	**0.100**	ND	ND	ND	ND	ND	0.001	0.008
Beasley	04/21/1999	**3.054**	ND	ND	**3.010**	0.007	ND	0.000	ND	ND	ND	ND	0.028
Beasley	06/22/1999	**0.388**	ND	ND	**10.046**	0.006	ND	0.001	ND	ND	ND	0.070	0.007
Beasley	07/29/1999	**0.648**	ND	ND	ND	0.005	ND	0.070	0.000	0.043	ND	ND	0.001
Beasley	08/24/1999	**0.258**	ND	ND	**1.293**	0.069	ND	ND	ND	0.033	ND	ND	ND
Beasley	09/22/1999	**0.298**	0.018	0.005	**1.060**	0.100	0.009	0.007	0.037	0.022	ND	ND	0.009
Beasley	10/20/1999	ND	0.008	0.001	0.279	0.057	ND	0.003	0.040	0.025	ND	0.005	0.006
Beasley	11/08/1999	**2.433**	0.025	ND	**0.216**	0.036	**0.108**	0.007	ND	0.020	ND	0.001	0.008
Beasley	12/16/1999	0.085	0.011	ND	**0.355**	0.076	0.007	0.002	0.038	0.023	ND	0.008	0.022
Beasley	01/13/2000	0.050	0.006	ND	0.079	0.028	0.005	0.001	0.018	0.008	ND	0.022	0.001
Beasley	02/09/2000	0.059	0.010	0.007	**0.228**	0.072	0.009	0.002	0.029	0.021	ND	0.003	0.008
Beasley	03/09/2000	ND	0.008	ND	**0.187**	0.048	0.011	0.004	0.035	0.032	ND	0.003	0.022
Beasley	05/31/2000	**1.357**	ND	ND	**0.276**	**0.112**	ND	0.005	ND	0.003	ND	0.100	0.022
Beasley	06/22/2000	**0.928**	0.035	0.009	**0.156**	**0.281**	0.006	0.008	ND	0.003	ND	0.040	0.007
Beasley	07/21/2000	**0.760**	0.038	0.011	0.100	**0.585**	ND	0.004	0.019	0.004	ND	0.050	0.001
Beasley	08/18/2000	**0.591**	0.045	0.009	0.082	**0.536**	0.005	0.006	0.042	0.004	ND	0.030	ND
Beasley	09/20/2000	**0.712**	0.033	0.014	0.015	0.549	0.005	0.010	0.076	0.042	ND	ND	0.009
Beasley	10/18/2000	**0.493**	0.035	0.013	0.015	**0.371**	0.005	0.007	0.021	0.012	ND	ND	0.006
Beasley	11/08/2000	**0.245**	0.051	**0.110**	**0.138**	**0.211**	0.010	0.004	0.009	0.002	ND	ND	ND
Beasley	12/06/2000	**0.154**	0.090	0.012	0.013	**0.137**	0.006	0.004	0.018	0.012	ND	ND	0.028
Deep Hollow	05/20/1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	**0.830**	0.015
Deep Hollow	06/24/1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	**1.268**	0.002
Deep Hollow	07/22/1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	**1.645**	0.000
Deep Hollow	08/18/1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.493	0.003
Deep Hollow	06/22/1999	ND	ND	**12.550**	**0.386**	ND	ND	0.100	0.032	ND	ND	ND	0.002
Deep Hollow	07/29/1999	ND	ND	0.002	0.100	0.012	ND	0.005	0.099	0.059	ND	ND	0.000
Deep Hollow	08/24/1999	ND	ND	ND	ND	0.010	ND	0.005	0.089	0.083	**0.340**	ND	0.003
Deep Hollow	09/22/1999	ND	0.012	ND	0.068	**0.392**	0.018	0.008	0.061	0.039	ND	ND	0.760
Deep Hollow	10/20/1999	ND	0.014	0.012	ND	**0.270**	ND	0.008	0.048	0.031	ND	0.001	0.058
Deep Hollow	11/08/1999	ND	0.029	0.001	0.009	0.025	ND	0.000	0.100	**0.508**	ND	0.008	0.036
Deep Hollow	12/16/1999	ND	0.012	0.007	0.033	**0.131**	0.007	0.009	0.018	0.014	ND	0.022	0.029
Deep Hollow	02/09/2000	ND	0.017	0.001	ND	0.100	0.006	0.005	0.018	0.004	ND	0.008	0.003
Deep Hollow	04/04/2000	ND	0.030	0.013	0.011	0.005	0.009	0.006	0.017	0.012	ND	1.338	0.010
Deep Hollow	05/03/2000	ND	0.031	0.014	0.045	0.004	0.011	0.004	0.009	0.004	ND	1.045	0.015
Deep Hollow	05/31/2000	ND	0.031	0.009	0.012	0.011	ND	0.008	0.003	0.002	ND	1.622	0.002
Deep Hollow	07/21/2000	ND	0.033	0.013	0.013	**6.819**	0.022	0.007	0.024	0.071	ND	ND	0.000

Table 4. Continued.

Lake	Date	Atrazine	Methyl Parathion	Alachlor	Metolachlor	Cyanazine	Pendimethalin	pp'-DDE	Bifenthrin	l-Cyhalothrin	Cyfluthrin	Z-Cypermethrin	Esfenvalerate
Deep Hollow	08/18/2000	ND	0.037	0.013	0.009	**3.199**	0.006	0.008	0.023	0.079	ND	ND	0.001
Deep Hollow	09/20/2000	ND	0.038	0.015	0.012	**2.338**	0.007	0.013	0.040	0.009	ND	ND	0.099
Deep Hollow	10/18/2000	ND	0.036	0.016	0.009	**1.932**	0.007	0.012	0.017	0.011	ND	ND	0.058
Deep Hollow	11/08/2000	ND	0.035	0.013	ND	**1.293**	0.006	0.009	ND	0.004	ND	ND	0.036
Deep Hollow	12/06/2000	ND	**4.130**	0.012	0.009	**0.715**	0.008	0.009	0.025	0.020	ND	ND	0.029
Thighman	09/10/1998	**1.566**	0.023	ND	ND	ND	ND	0.006	ND	ND	ND	ND	0.022
Thighman	10/09/1998	ND	ND	0.071	0.078	0.004	ND	0.024	0.046	ND	ND	0.001	0.282
Thighman	04/21/1999	**7.183**	ND	ND	**1.363**	0.011	ND	0.000	0.003	ND	ND	ND	0.003
Thighman	05/25/1999	**4.571**	ND	ND	**2.594**	0.003	ND	0.002	ND	ND	ND	ND	0.015
Thighman	06/22/1999	ND	ND	ND	**1.222**	0.019	ND	0.003	0.011	ND	ND	ND	0.001
Thighman	09/22/1999	0.038	0.011	0.002	0.007	**0.372**	0.006	0.005	0.025	0.010	ND	0.001	0.022
Thighman	10/20/1999	**0.578**	0.012	0.005	ND	**0.279**	ND	0.006	0.033	0.008	ND	0.008	ND
Thighman	11/08/1999	**0.381**	0.011	ND	ND	**0.234**	0.003	0.005	0.041	0.008	ND	0.022	0.006
Thighman	12/16/1999	0.042	0.006	ND	0.100	**0.107**	0.008	0.007	0.068	0.032	ND	0.001	0.001
Thighman	04/04/2000	0.087	0.032	0.016	**0.448**	0.012	0.007	0.005	0.000	0.003	ND	ND	ND
Thighman	05/03/2000	0.060	0.041	0.013	0.937	0.012	0.009	0.005	0.009	0.005	ND	ND	ND
Thighman	05/31/2000	**1.274**	0.033	0.011	**13.006**	0.004	ND	0.005	0.001	0.002	ND	ND	ND
Thighman	06/22/2000	**2.401**	0.029	0.013	**1.026**	**0.101**	0.005	0.008	0.003	0.003	ND	ND	ND
Thighman	07/21/2000	**0.124**	0.038	0.015	**0.231**	**0.222**	0.005	0.009	0.025	0.005	ND	ND	ND
Thighman	08/18/2000	**0.231**	0.039	0.013	0.062	0.095	0.005	0.009	0.013	0.014	ND	ND	ND
Thighman	09/20/2000	**0.543**	0.031	0.011	0.036	0.084	0.007	0.014	0.036	0.027	ND	ND	ND
Thighman	10/18/2000	**0.191**	0.030	0.015	0.013	0.017	0.006	0.010	0.018	0.007	ND	ND	ND
Thighman	11/08/2000	**0.299**	0.043	0.011	0.036	0.014	0.006	0.011	0.025	0.009	ND	ND	ND

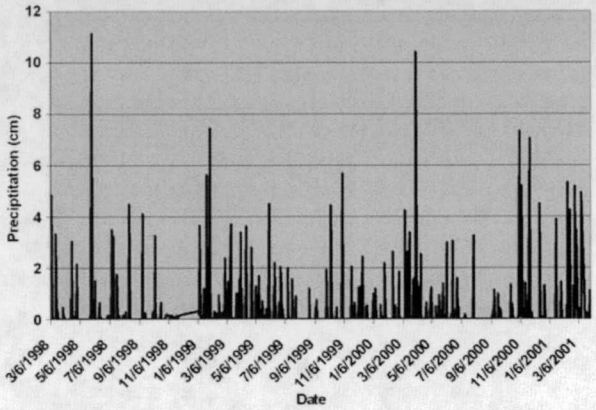

Figure 5. Precipitation chart from weather data collected at Thighman Lake.

all three years that we examined. In the dry year of 1998, little cyanazine was detected. In 1999 and 2000, detections were more common throughout the year from different runoff patterns and greater total rainfall. Greatest concentrations of cyanazine were detected in the summer and fall, 2000 in Deep Hollow Lake where they reached 6.82 $\mu g\ L^{-1}$. Other concentrations were one to two orders of magnitude less. Metolachlor produced a contamination pattern consistent with rainfall in Beasley and Thighman in 1999 and 2000. Highest concentrations were measured during April through June in both years. The low concentrations of measured metolachlor in Thighman and Beasley lakes during 1998 were attributed to lack of runoff. However, concentrations in excess of 1 $\mu g\ L^{-1}$ were detected in both 1999 and in Thighman in 2000. Residual concentrations of DDT revealed only old material as judged by metabolite concentrations; no DDT parent compound was measured. In Beasley Lake, the greatest concentration for DDE was 0.07 $\mu g\ L^{-1}$; other values were an order of magnitude less (Table 4). The largest concentration of DDD in Thighman Lake was 0.015 $\mu g\ L^{-1}$, and the greatest concentration in Deep Hollow Lake surface water was 0.08 $\mu g\ L^{-1}$ in a single sample in June, 1999. Other detections were lower by at least an order of magnitude. The low concentrations of DDT metabolites and the absence of the parent compound in lake water showed that degradation is reducing DDT to concentrations that are harmless to aquatic organisms. In a 1977-1979 study in Bear Creek, the stream bordering Thighman Lake on the east side, Cooper et al. (1987) detected the parent compound DDT and metabolites in all surface water samples for 3 years where concentrations of ΣDDT varied from <0.01-0.1 $\mu g\ L^{-1}$. They also noted that concentration levels had not declined since a previous study was completed in 1970.

Pesticide Deposition Over Time in Lake Sediments

We calculated recent sediment accumulation rates from radioactive cesium by dating 10 cm increments of sediment cores from three sites in each of the three lakes. Beasley Lake had the lowest rate of sediment accumulation (Figure 6). While the upper end of the lake accumulated sediment at a rate of >1.57 cm yr^{-1} since 1964, the remainder of the lake had <0.5 cm yr^{-1} since 1964. Thighman Lake (Figure 7) had accumulated >1.5 cm yr^{-1} -2.0 cm yr^{-1} since 1964. Deep Hollow Lake (Figure 8) had the greatest rates of sedi-

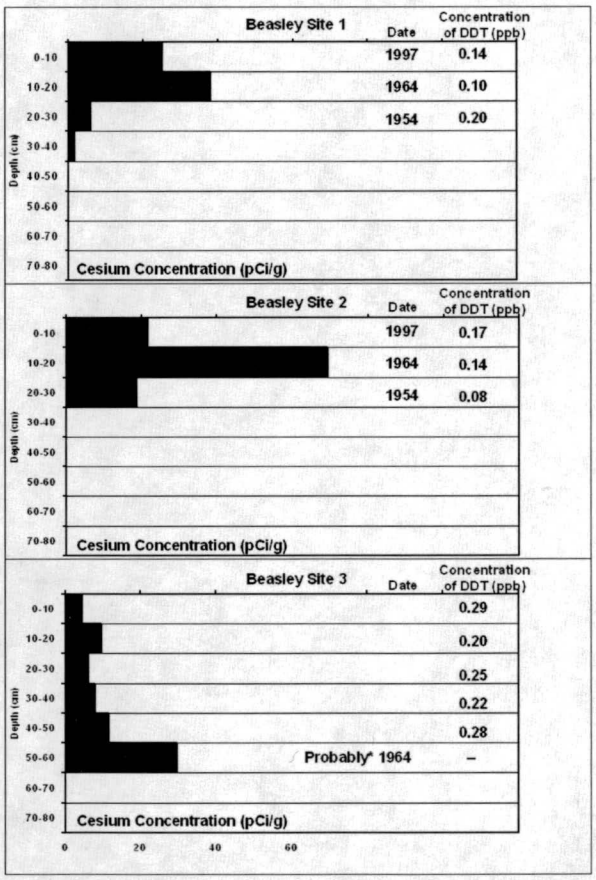

Figure 6. Cesium and ΣDDT concentrations plotted with depth in sediment profile and approximate age for Beasley Lake.

ment accumulation; its three sites averaged >1.6 cm yr^{-1}, 1.7 cm yr^{-1} and 2.1 cm yr^{-1} since 1964. The first Deep Hollow site (>1.6 cm yr^{-1}) could have a more excessive rate of sediment accumulation since the depth of coring (50 cm) did not reach a definitive cesium marker of 1964. A dense layer of large woody debris at 50 cm restricted sampling depth.

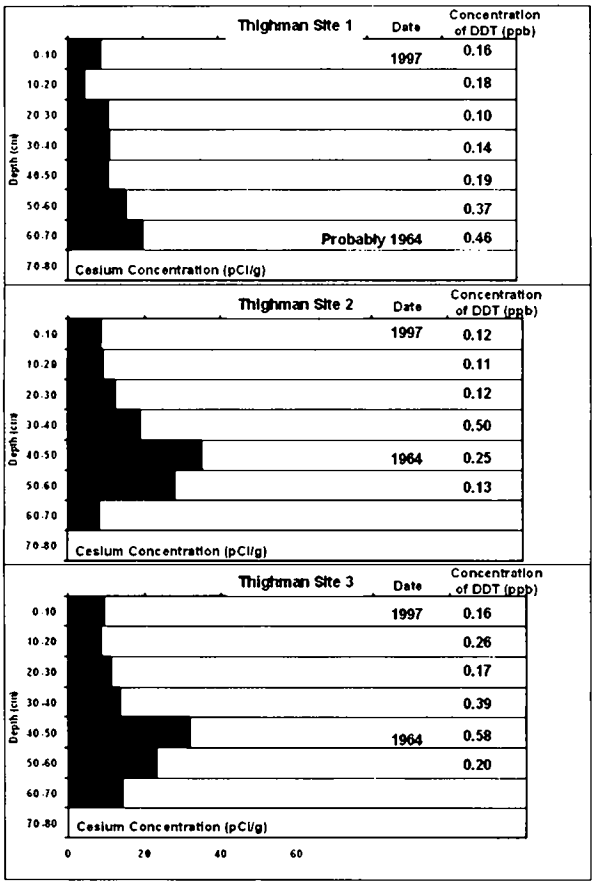

Figure 7. Cesium and ΣDDT concentrations plotted with depth in sediment profile and approximate age for Thighman Lake.

Since identical cores were taken for pesticide analysis, we were able to view sediment pesticide concentrations by age in each lake. Results were largely predictable. Residual organochlorines dominated the pesticide-sediment profiles. Older residual compounds that have been discontinued were found at greater depths in layers that corresponded to the 1950s, 1960s and 1970s. For example, ΣDDT in the Deep Hollow sediment from the 1960s reached 436.0 $\mu g\ L^{-1}$ in sediment from the 60-70 cm depth (Figure 8). Pesticides such as atrazine and trifluralin that were first formulated during the 1950s and are still actively used. They were widely dispersed in sediments. Because coring disperses small amounts of sediment downward as the sampler pushes through accumulated sediment, small concentrations of prevalent pesticides can be measured at any depth. Given this coring artifact, we limited observations to concentrations that were obviously associated with actual sediment accumulation.

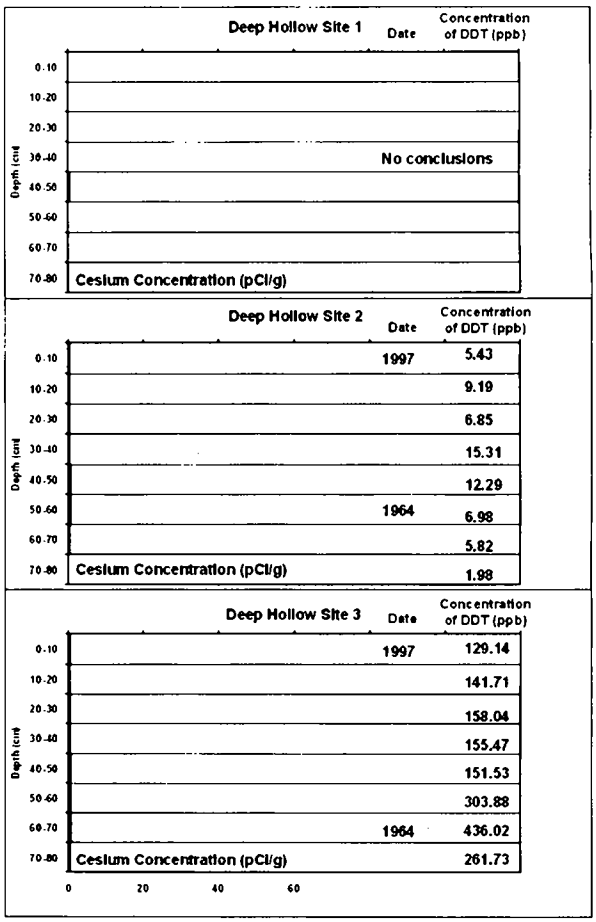

Figure 8. Cesium and ΣDDT concentrations plotted with depth in sediment profile and approximate age for Deep Hollow Lake.

Sediment pesticide levels allowed observations on availability from the sink of material in sediments. Watershed soil and lake sediment represent the two

greatest potential sources of older residual compounds in agricultural watersheds (Cooper 1991). ΣDDT was the only compound that permeated sediments at all sites. Concentrations of ΣDDT in surface sediments at Beasley and Thighman lakes ranged from 0.12 $\mu g\ L^{-1}$ to 0.29 $\mu g\ L^{-1}$ of ΣDDT. This level of ΣDDT would be expected to result in little or no contamination of the water column or biota. Deep Hollow Lake had more ΣDDT associated with its sediment. Surface sediment concentrations were 101.64 $\mu g\ L^{-1}$, 5.43 $\mu g\ L^{-1}$, and 129.14 $\mu g\ L^{-1}$ of ΣDDT at sites in the upper, center, and lower sections of the lake. Cooper et al. (1987) measured concentrations as high as 353.7 $\mu g\ L^{-1}$ for DDT, 380.8 $\mu g\ L^{-1}$ for DDE and 541.0 $\mu g\ L^{-1}$ for DDD in Bear Creek, Mississippi sediment. We observed that sediments deposited in the 1960s and 1970s which contained higher concentrations of DDT have been effectively sealed by more recent material so that the sink of contaminant is unavailable to be re-suspended.

Beasley Forested Wetland

In addition to routinely sampled groundwater wells in the forested wetland (Figure 2), we conducted a controlled release experiment in 1997, using the pyrethroid insecticides λ-cyhalothrin and cyfluthrin (each at a dose rate of 0.0336 kg/ha) (Harrington 1998; Smith et al. 2001). We conducted the experiment because routine sampling detected no pyrethroids in water leaving the 800 m long forested wetland although they were routinely applied to the fields draining the wetland and measured in runoff. After the initial controlled release, samples were routinely taken in sediment traps placed in the wetland, in shallow groundwater wells, and at a USGS sampling site at the lower end of the wetland. Sediment samples taken after one hour at the injection point contained about 3 mg L^{-1} of each insecticide. Sediment samples after four months contained <10 $\mu g\ L^{-1}$ of both pesticides. By March, 1998, λ-cyhalothrin and cyfluthrin were reduced to mean concentrations of 5 $\mu g\ L^{-1}$ and 1 $\mu g\ L^{-1}$, respectively. λ-cyhalothrin was detected in sediment samples from October, 1998 at about 2 $\mu g\ L^{-1}$; no cyfluthrin was detected. Additional sediment samples taken in 1999 did not detect either chemical.

No shallow groundwater appeared in the wells within the wetland until January, 1998. Initial concentrations were about 0.02 $\mu g\ L^{-1}$ of each insecticide. Concentrations remained extremely low (0.001 to 0.0019 $\mu g\ L^{-1}$) for several months. In August, 1998, cyfluthrin could not be detected. Cyhalothrin was detected sporadically until December, 1998. Nearly 300 well samples were collected during 1998 and 1999.

Pyrethroids were detected in shallow groundwater until December, 1998 and from transported surface sediments collected within the wetland until October, 1998. No insecticides were detected at the USGS surface water sampling site at the lower end of the wetland at any time during the study. Thus, the forested wetland was totally effective in neutralizing the pyrethroids before they reached Beasley Lake. Routine inflow samples at the upper end of the wetland revealed that pyrethroids in edge-of-field runoff were also being processed in vegetated drainage ditches that composed the main drainage flow path from the agricultural fields to the forested wetland.

Toxicity Implications

Concentrations of herbicides found in lake water were well below values reported to be harmful to aquatic organisms (EXTOXNET 2001). LC50 (96 hour) values for fish are in the range of 2.4-18 mg L^{-1}, whereas the highest herbicide concentration found in the lake water samples was about 13 $\mu g\ L^{-1}$ (3 orders of magnitude lower). Of the 21 pesticides and metabolites analyzed in lake water, DDD, dieldrin, cyanazine, trifluralin, chlorpyrifos, esfenvalerate, chlorfenapyr, pendimethalin, and bifenthrin were not of sufficient individual concentrations to cause impairment, based on available laboratory and field studies (EXTOXNET 2001).

Potential transient toxicity concerns occurred for alachlor, λ-cyhalothrin, and methyl parathion in Deep Hollow Lake. On the June 1999 sampling date of Deep Hollow Lake, alachlor concentrations exceeded the maximum contaminant level (MCL) of 2 $\mu g\ L^{-1}$. In November 1999, Deep Hollow Lake concentrations of λ-cyhalothrin (0.51 $\mu g\ L^{-1}$) were sufficient to potentially impair fish and aquatic invertebrates. LC_{50}s for λ-cyhalothrin exposure to bluegill sunfish [*Lepomis macrochirus* (Rafinesque)] and the water flea [*Daphnia magna* Strauss)] were >0.2 $\mu g\ L^{-1}$ and 0.36 $\mu g\ L^{-1}$, respectively (EXTOXNET 2001). Methyl parathion concentrations in Deep Hollow Lake during December 2000 indicated a concentration (4.13 $\mu g\ L^{-1}$) sufficient to cause potential impairment to aquatic invertebrates. The 96 h LC_{50} for *Daphnia magna* exposed to methyl parathion is 0.14 $\mu g\ L^{-1}$ (Johnson and Finley 1980).

Potential seasonal impairment concerns were noted in all three lakes for DDT. DDT concentrations in Beasley Lake collected in January, February, April, June, and August of 1999 exceeded the 10d LC_{50} for the amphipod *Hyalella azteca* (Saussure) (0.07 $\mu g\ L^{-1}$) (Hoke et al. 1994). During the collection timeframe of April 2000 through February 2001, Beasley Lake samples again exceeded this DDT concentration. Deep Hollow Lake samples collected in January and February

1999, and the timeframe of April 2000 through February 2001, likewise exceeded the DDT 10d LC_{50} for *Hyalella azteca*. DDT concentrations measured in Thighman Lake from September 1998 through February 1999, May 1999, and September 1999, revealed DDT concentrations that exceeded the 0.07 $\mu g\ L^{-1}$ 10d *Hyalella azteca* LC_{50}. Because of these prolonged exposures, potential existed for impairment of aquatic invertebrates.

Zeta-cypermethrin potential seasonal impairment was also noted in all three lakes. Potential impairment at Beasley Lake existed from June 1998-August 1998; June 1999-August 1999; May 2000-August 2000; and again on the sampling date from December 2000. Elevated pesticide concentrations at Deep Hollow Lake were observed from May 1998-September 1998; May 1999-September 1999; April 2000-May 2000; and September 2000. Thighman Lake concentrations capable of potentially causing impairment were collected from May 1998-July 1998; April 1999-June 1999; May 2000-June 2000; and February 2001. These potential impairment concentrations were based on the cypermethrin LC50 for Daphnia magna (0.2 $\mu g\ L^{-1}$) (EXTOXNET 2001).

While these individual pesticide concentrations raise concern, potential synergistic toxicity effects from a suite of pesticides present in water samples also exist. Denton et al. (2003) reported on the synergistic effects of an organophosphate (diazinon) and pyrethroid (esfenvalerate) in water. When atrazine ($\geq 40\ \mu g\ L^{-1}$) was present with chlorpyrifos, methyl parathion, or diazinon, there was a significant increase in *Hyalella azteca* toxicity, as well as significant decreases in acetylcholinesterase activity when compared to the organophosphates dosed individually (Anderson and Lydy 2002). Jin-Clark et al. (2002) reported synergistic effects of atrazine and cyanazine on chlorpyrifos toxicity of the midge, *Chironomus tentans*. Atrazine has also been shown to elicit synergistic toxicity effects on *Chironomus tentans* when mixed with methyl parathion (Pape-Lindstrom and Lydy 1997). Chemical synergy has been reported between atrazine and alachlor after 96 h exposures on amphibian larvae (Howe et al. 1998). Not all pesticides act synergistically, however, as described by Hoagland et al. (1993).

The US EPA has derived water quality standards for some of the legacy pesticides that are no longer in use. DDT has a freshwater acute water quality standard of 1.1 $\mu g\ L^{-1}$ and a freshwater chronic water quality standard of 0.001 $\mu g\ L^{-1}$. The human health DDT water quality standard is 0.00059 $\mu g\ L^{-1}$ (water and organism). Dieldrin's freshwater acute and chronic water quality standards are 0.24 $\mu g\ L^{-1}$ and 0.056 $\mu g\ L^{-1}$, respectively, while its human health water quality standard is 0.00014 $\mu g\ L^{-1}$. Likewise, drinking water standards have been established by the US EPA for alachlor (2 $\mu g\ L^{-1}$), atrazine (3 $\mu g\ L^{-1}$), and endrin (2 $\mu g\ L^{-1}$). Only three out of more than 100 lake samples had concentrations that exceeded the atrazine MCL for drinking water. The freshwater acute water quality standard for DDT was exceeded only once; however, the chronic water quality standard was exceeded in approximately 86 (82.7 %) collected samples. Based on this information, there is potential aquatic faunal risk due to chronic residual DDT exposure. Dieldrin's freshwater acute water quality standard was never exceeded during the sampling period, and only once, during a transient event, did it exceed the chronic water quality standard.

CONCLUSION

In summary, an intensive sampling of surface water, shallow ground water, and sediments was coupled with field and runoff phenomena to present a picture of historical and current common pesticide contamination in closed-system oxbow lakes in the Mississippi Delta. Lake contamination by current use pesticides was reduced by on-field and edge-of-field best management practices. Selected herbicides measured in concentrated flow zones were present in ng L^{-1} levels. Water that penetrated soil did not represent a shallow ground water impact. Measurement of lake surface water resulted in 90 pesticide detections above the 0.10 $\mu g\ L^{-1}$ level. Of these, 78 detections were herbicides and 12 were pyrethroid insecticides. Atrazine was most common and was found in 28 (26.9 %) lake water samples. With regard to the pyrethroid insecticides found in lake water samples at concentrations $>0.01\ \mu g\ L^{-1}$, λ-cyhalothrin was found once, cyfluthrin once, zeta-cypermethrin 7 times, and esfenvalerate 3 times.

Sediment cores analyzed for pesticides were also viewed by age in each lake. Predictably, residual organochlorines dominated the pesticide-sediment profiles. Older discontinued but residual compounds were found mainly at greater depths in layers that corresponded to the 1950s, 1960s and 1970s. ΣDDT was the only compound that permeated sediments at all sites. We observed that sediments deposited in the 1960s and 1970s which contained higher concentrations of DDT have been effectively sealed by more recent material so that the sink of contaminant is unavailable to be re-suspended.

Pyrethroid studies in a forested wetland substantiated that the wetland was totally effective in neutralizing the pyrethroids before they reached Beasley Lake. Routine inflow samples at the upper end

of the wetland also revealed that pyrethroids in edge-of-field runoff were also being processed in vegetated drainage ditches that composed the main drainage flow path from the agricultural fields to the forested wetland.

While the lakes were not drinking water supplies, pesticides seldom exceeded drinking water standards. Acute toxicity concerns lessen as more conservation measures are installed on and off-field. Potential seasonal impairment concerns for aquatic invertebrates were noted in all three lakes for DDT because of prolonged exposure. Zeta-cypermethrin potential seasonal impairment was also noted in all three lakes. Surface water conditions continue to improve as legacy pesticides slowly degrade. Oxbow lakes in agriculturally-productive floodplains serve as sinks for DDT in sediments while most current use pesticides pose little problem when applied properly. Edge-of-field vegetation, ranging from grass buffers to vegetated drainage ditches or forested wetlands, provides valuable, inexpensive ways to reduce pesticides to acceptable levels.

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Mention of a pesticide in this paper does not constitute a recommendation for use by the United States Department of Agriculture nor does it imply registration under FIFRA as amended. Names of commercial products are included for the benefit of the reader and do not imply endorsement or preferential treatment by the U.S. Department of Agriculture. All programs and services of the United States Department of Agriculture are offered on a nondiscriminatory basis without regard to race, color, national origin, religion, sex, marital status, or handicap.

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