CONCENTRATIONS OF SELECTED PESTICIDES IN TREATED AND UNTREATED DRINKING WATER FROM LAKE BRUIN, LOUISIANA, APRIL - DECEMBER 1999

Michael A. Manning
U.S. Geological Survey, WRD
Pearl, Mississippi

INTRODUCTION

Contamination of surface water in the United States from pesticide application has been a major water-quality issue during the past several decades. Nationwide use of pesticides increased more than 300 percent from 1968 to 1988. A total of 4.6 billion pounds of pesticides was applied in the United States during 1997 (U.S. Environmental Protection Agency, 2000). It is always of concern that a percentage of applied pesticides could move from the site of application and subsequently contaminate local surface waters. Even a small amount of pesticides in surface waters may represent a threat to the aquatic ecosystem and to human health. Because lakes and reservoirs store water and do not have the flush effect of rivers and streams, pesticide concentrations in lakes can have a much longer residence time. Therefore, agricultural compounds dissolved in lakes with long residence times may occur at lower concentration value due to dilution, but can persist longer in the water column because of storage (Stamer, Battaglin, and Goolsby 1998).

The purpose of this study was to determine the occurrence of a broad range of pesticides in reservoirs that supply a source of drinking water to the public. Data collected during this study expands the National Water Quality Assessment Program (NAWQA) extensive pesticide database to include surface waters used for drinking water.

From April to December 1999, the U.S. Geological Survey (USGS), National Water Quality Assessment Program in cooperation with the U.S. Environmental Protection Agency, Office of Pesticide Programs (EPA-OPP), conducted a pilot study to monitor the concentration of selected pesticides in lakes that supply water for public consumption. Lake Bruin, located in an intensive agricultural region of northwestern Louisiana, was selected as one of several lakes throughout the Nation to be monitored.

SITE DESCRIPTION AND LOCATION

Reservoirs and lakes were selected for monitoring because they are considered to be vulnerable to pesticide contamination, and often serve as a source for drinking water. Lake Bruin is one of many oxbow lakes formed by an old meander cutoff from the winding Mississippi River. The lake is located in northeastern Louisiana about 4 miles north of St. Joseph, Louisiana, and adjacent to the Mississippi River's western levee system (figure 1). Lake Bruin, which has a classical oxbow shape, has a surface area of 4.7 square miles. The lake is a popular recreational area for fishing, swimming, and boating, and provides a source of drinking water for several nearby communities. During 1998, a total of 0.61 million gallons per day of water was withdrawn from Lake Bruin for public drinking water supplies. Residential development on the extensive shoreline around the lake is becoming increasingly popular for weekend homes and fishing camps. The water level of Lake Bruin is controlled by wet and dry periods, and change in stage of the adjacent Mississippi River. In the past, studies have shown that oxbow lakes associated with the Mississippi River can exchange water with the Mississippi River through the alluvial aquifer (Harbeck, Golden, and Harvey 1961). No major rivers or streams drain into or away from the lake. Drainage into the lake may consist of both point (seepage of domestic sewage and or direct input) and non-point sources (Demas 1985). Lake Bruin receives drainage from nearly 22 square miles of mostly agricultural farmland. The major crops grown in the area include cotton, corn, and soybeans. Local residents have stated that, at times (depending on wind conditions), the lake receives direct input of pesticides from aerial applications to adjacent agricultural fields (Demas1985). Air and rain depositional studies done in Jackson, and Rolling Fork, Mississippi, have shown that many pesticide compounds can be transported by the wind, and can then be later deposited by falling rain, dew, and fog depending on various weather patterns (Coupe 1998).

SAMPLING AND METHODS

Two sites were sampled for pesticides at Lake Bruin. A total of 11 water samples were collected at each site for pesticide analysis. Sampling at each site began in April 1999 and continued through the last week of September. One additional sample was collected the last week of December 1999. One site was located on the western most shoreline of Lake Bruin. Water samples at this location were collected directly from the pump intake, which supplies raw (untreated) water from Lake Bruin to a nearby drinking water treatment facility. Samples representing the finished (treated) drinking water were collected directly from a spigot at a nearby drinking water treatment plant. These samples represent the finished drinking water after it has completed the water treatment plant process and before it reaches the public. The treatment plant is approximately 1 mile from the pump intake site at Lake Bruin.

All water samples were collected and processed using laboratory ultra-pre-cleaned sampling equipment. U.S. Geological Survey standards for sampling (Shelton 1994) were used to ensure uncontaminated and accurate representative samples. Each liter of sampled water was filtered through baked 0.7 m glass fiber filters using an aluminum filter plate. All samples were shipped on ice overnight to the U.S. Geological Survey National Water Quality Laboratory (NWQL) in Denver, Colorado, for final analysis. The NWQL analyzed each sample for nearly 100 pesticides and pesticide metabolites by either Gas Chromatography/Mass Spectrometry (GC/MS), or High Performance Liquid Chromatography (HPLC) (tables 1 and 2). Quality assurance samples consisted of nine blank samples, six spikes, and three replicates. Field blanks were processed to test for contamination, replicate samples were run to measure precision, and field spikes were run to measure recovery. All field blanks analyzed were free of any measurable contamination.

LAKE BRUIN INTAKE SAMPLES

Nine commonly used pesticide compounds were detected in water samples taken from the intake that feeds raw (untreated) water to the drinking water treatment plant. Atrazine, cyanazine, metolachlor, and simazine were detected in all of the samples collected from the intake. Atrazine, cyanazine, and simazine are all part of the triazine family of selective grass herbicides. Concentrations of some metabolite compounds from the triazine family, in some samples, were reported as estimated by the NWQL. However, these estimated values were used by the NWQL to suggest that some value may exist lower than the detection limit, but were also lower than the lowest calibration standard used by NWQL during the analysis procedure. These estimated values reported by the NWQL were not considered for this paper.

Atrazine can be used as an herbicide for weed control on corn and sorghum. Atrazine can also be used (usually at much lower concentrations) on residential lawns in some types of weed and feed type applications. In most agricultural areas, atrazine is applied on corn crops early in the planting season as a pre-emergent, and again during the growing season as a post-emergent (Meister and Sine 1996). The water sample collected on May 13,1999, had the maximum detected concentration of atrazine, 0.998 micrograms per liter (g/L) (table 3). This value was also the maximum concentration of any pesticide compound detected in any of the samples collected during this study. The median concentration of atrazine in the untreated water from Lake Bruin was 0.960 g/L. The sample collected on December 28 had the lowest detection for atrazine at 0.688 µg/L. The chronic criterion for the protection of aquatic life or wildlife from Canadian water- quality guidelines for freshwater aquatic life for atrazine is 1.8 g/L (AQCR-CAN)(U.S. Environmental Protection Agency 1999).

Cyanazine is also an agricultural herbicide mostly used for pre-plant applications, but can be used for post-emergence control of weeds on corn and cotton. The highest concentration of cyanazine in the untreated water from the Lake Bruin was detected on July 13 at 0.332 g/L (table 3). The median concentration for cyanazine was 0.166 g/L. The AQCR-CAN for cyanazine in freshwater is 2 g/L (U.S.Environmental Protection Agency 1999).

Metolachlor is also a selective herbicide used for pre-emergent and pre-plant weed control applications on corn, soybeans and grain sorghum. Metolachlor had a maximum concentration in the untreated water collected from Lake Bruin of 0.033 g/L on April 29. The median concentration for metolachlor was 0.021 g/L. The AQCR-CAN for metolachlor is 7.8 µg/L.

Simazine is a commonly used herbicide that is mostly applied in urban areas on golf courses and lawns and gardens to control broadleaf weeds. The highest detection for simazine was 0.184 g/L from the sample collected on July 13. The median concentration of simazine at the intake was 0.156 g/L. The aquatic life criterion, AQCR-CAN, for simazine is 10 g/L. Fluometuron and diuron are herbicides that were also detected in a few samples, but in low concentrations.

Chlorpyrifos, an insecticide used for control of crawling insects such as fire ants and termites, was detected in three of 11 of the samples from the water at the pump intake. The maximum chlorpyrifos concentration detected was 0.008 g/L. The median value for chlorpyrifos in samples with detectable concentrations was 0.006 g/L. The aquatic life criteria, AQCR-CAN, for chlorpyrifos is 0.041 g/L. Malathion is an insecticide used to control insects such as aphids (weevils) that destroy plants by sucking and chewing their leaves. Malathion was detected in 3 of 11 samples. The median concentration for malathion in samples with detections was 0.040 g/L. The aquatic criteria for malathion is 0.1 g/L. The insecticide 2,4-D was detected in a few samples, but all concentrations were just above the detection limits of the analytical method.

The pesticide compound that were detected in all of the samples collected at the intake pump were found to be fairly uniform in concentration with little to no variation throughout the sampling period. Although some of the pesticide compounds were consistently found in each sample, all of the concentrations were relatively low. This confirms other studies which have shown that once pesticide compounds are transported into lakes and reservoirs they can persist due to storage of water, but at low concentrations due to high dilution factors created by the volume of water in the lake (Stamer, Battaglin, Goolsby 1998, Scribner and others 1996).

FINISHED WATER SAMPLES

Data from water samples taken at the drinking water treatment plant show detections of five pesticides. These pesticides, all herbicides, were atrazine, cyanazine, metolachlor, simazine, and 2,4-D. Chlorpyrifos, malathion, fluometuron, and diuron, which were detected in the lake water, were not detected in the finished drinking water at the plant. Atrazine, cyanazine, metolachlor, and simazine were detected in all of the samples collected from the pump intake in Lake Bruin, and from the drinking water treatment plant (table 4).

Atrazine had a concentration of 0.851 g/L on April 29, the highest detection at the treatment plant during the study. This concentration, however, does not coincide with the highest atrazine concentration in samples taken from the intake. The median concentration of atrazine in the (treated) drinking water from the treatment plant was 0.768 g/L. EPA maximum contaminant level (MCL) for atrazine in drinking water is 3 g/L (U.S. Environmental Protection Agency 1999).

The sample collected on July 13 had the highest concentration of cyanazine, and simazine at 0.355, 0.173 g/L, respectively. These concentrations coincide (in time) with the highest cyanazine and simazine concentrations found in the raw (untreated) water at the pump intake. The median concentrations of cyanazine and simazine in the finished (treated) drinking water were 0.117, and 0.113 g/L, respectively. EPA health advisory level (HAL) for cyanazine is 1 g/L, and the MCL for simazine is 4 g/L, (U.S. Environmental Protection Agency 1999).

Metolachlor was also detected in all of the samples from the finished drinking water at the treatment plant. The highest concentration detected was 0.027 µg/L in the water sample collected on April 29. The median concentration for metolachlor was 0.021 g/L. The HAL for metolachlor is 70 g/L (U.S. Environmental Protection Agency 1999).

Pesticide detection in the finished (treated) drinking water from the plant basically mirrored the detections in the raw (untreated) water from the lake. Although concentrations in the finished drinking water at the treatment plant were slightly less than concentrations in the lake, all of the compounds were detected in the same order of magnitude. All of the pesticide compounds detected in the finished (treated) drinking water at the treatment plant, like detections in the untreated lake water, were found to be fairly uniform with little to no variation in concentration throughout the sampling period. This is also in agreement with previous studies that have proven that conventional water treatment for drinking water, such as sand filtration, coagulation, and chlorination is relatively ineffective at removing certain herbicides such as atrazine, cyanazine, metolachlor and simazine. These same studies suggest that additional water treatment processes such as ozone and or activated carbon filtration may adequately reduce these pesticides in drinking water (Stamer, Battaglin, and Goolsby 1998). In 1996, the EPA classified atrazine, cyanazine and metolachlor as possible carcinogens (U.S. Environmental Protection Agency 1999).

DISCUSSION

Between April and December 1999, water samples were collected for pesticide analysis from a drinking water pump intake located at Lake Bruin. Samples were also collected at the drinking water treatment plant near Lake Bruin that supplies drinking water to local residences. Nine pesticides were detected at the intake pump site within Lake Bruin. Four of these compounds (atrazine, cyanazine, metolachlor, and simazine) were detected in all of the samples. Five pesticides were detected in finished water samples from the drinking water treatment plant. Four of these compounds (atrazine, cyanazine, metolachlor, and simazine) were detected in all of the samples from the treated drinking water. Atrazine had the highest concentration of any pesticide analyzed for in both the untreated, and treated water with concentrations of 0.998 g/L and 0.851 g/L, respectively. All of the compounds detected at either site were below the EPA's maximum contaminate level and the Canadian's water-quality guidelines for protection of aquatic life in freshwater. Samples collected during this study show that small amounts of agricultural and residential pesticides are transported into the lake. All of the pesticide compounds detected in the lake were found to be fairly uniform in concentration with little variation throughout the sampling period. Although some of the pesticides were consistently found in each sample, all of the concentrations were relatively low.

Pesticides in the water from the treated drinking water at the plant basically mirrored the detections in the untreated water from Lake Bruin. Although concentrations in the finished drinking water at the treatment plant were slightly less than concentrations in the lake, all of the compounds that were detected in all of the samples at both sites were detected in the same order of magnitude. All of the pesticide compounds detected in the finished (treated) drinking water at the treatment plant, like detections in the untreated lake water, were found to be fairly uniform with little to no variation in concentration throughout the sampling period. This is in agreement with other studies that have shown that conventional water treatment processes used for drinking water, such as sand filtration, coagulation, and chlorination is relatively ineffective at removing certain herbicides such as atrazine, cyanazine, metolachlor and simazine. These same studies suggest that additional water treatment processes such as ozone, activated carbon filtration, and other methods may adequately reduce these pesticides in drinking water.

Lake Bruin is a moderately deep lake (for its geographical location), and can store large amounts of water. This feature, combined with having no major streams or rivers draining into or out of the lake, creates a longer residence time for water. Therefore, pesticides compounds detected in Lake Bruin during this study were found to persist at consistent concentrations because of residence time (storage), but were detected at low concentration values due to dilution generated by the volume of water in the lake.

REFERENCES

Coupe, R.H., M.A. Manning, W.T. Foreman, D.A. Goolsby, and M.S. Majewski. 1998. Differences in Distribution of Pesticides in Air and Rain in Urban and Agricultural Areas of Mississippi. In Proceedings of the 28^{th} Mississippi Water Resource Conference, April 7-8, 1998, edited by Jean Daniel 329-3321. Mississippi State University.

Demas, C. R., 1985. A Limnology Study of Lake Bruin, Louisiana. U.S. Geological Survey, Water Resources Technical Report No. 38

Goolsby, D.A, and others. 1993. Persistence of Herbicides In Selected Reservoirs in the Midwestern United States: Some Preliminary Results. Selected Papers on Agricultural Chemicals in Water Resouces of the Midcontinental United States. U.S. Geological Survey Open File Report 93-418, p 51-63.

Harbeck, E. G. Jr., H.G. Golden, and E. J. Harvey. 1961. Effect of Irrigation Withdrawals on Stage of Lake Washington, Mississippi. U.S. Geological Survey Water Supply Paper 1460-I.

Larson, S. L., P. D. Capel, and M.S. Majewski. Pesticides in Surface Waters, Distrbution, Treads, and Governing Factors. U.S. Geological Survey National Water Quality Assessment Program. Pesticides in the Hydrologic Cycle Vol. 3.

Meister R.T. and Sine C. 1996. Farm Chemical Handbook. Volume 82.

Scribner, E.A. and others. 1996. Concentrations of Selected Herbicides, Herbicide Metabolites, and Nutrients in Outflow from Selected Midwestern Reservoirs, April 1992 through September 1993. U.S. Geological Survey Open File Report 96-393.

Shelton, L.R., 1994. Field Guide for Collecting and Processing Stream-Water Samples for the National Water Quality Assessment Program. U.S. Geological Survey Open File Report 94-455.

Stamer, J. K., W.A. Battaglin, and D.A. Goolsby. December 1998. Herbicides in Midwestern Reservoir Outflows, 1992-93. U.S. Geological Survey Fact sheet, FS-134-98.

United States Environmental Protection Agency (USEPA): Office of Water. Drinking Water Standards and Health Advisories: Organics. Last updated February 1999. Accessed March 2000. online at http://www.epa.gov/OST/Tools/dwstds.html

United States Environmental Protection Agency (USEPA): Office of Pesticide Programs. Pesticide Industry Sales And Usage: 1996 and 1997 Market Estimates (733-R-99-001) last updated January 2000. Accessed March 2000. online at http://www.epa.gov/oppbead1/pestsales/

Table 1. Dissolved pesticides analyzed by Gas Chromatography/Mass Spectrometry

Acetochlor	Methyl Parathion
Alachlor	Metolachlor**
Alpha BHC	Metribuzin
Atrazine**	Molinate
Atrazine Deethyl	Napropamide
Azinphos-Methyl	Parathion
Benfluralin	Pebulate
Butylate	Pendimenthalin
Carbaryl[1]	Permethrin-cis
Carbofuran[1]	Phorate
Chlorpyrifos*	Prometon
Cyanazine**	Pronamide
DCPA	Propachlor
DDE-P'P	Propanil
Diazinon	Propargite
Dieldrin	Simazine**
2,6-Diethylaniline	Tebuthiuron
Disulfoton	Terbacil
EPTC	Terbufos
Ethalfluralin	Thiobencarb
Ethoprop	Triallate
Fonofos	Trifluralin
Lindane	
Linuron[1]	
Malathion*	

Table 2. Dissolved pesticides analyzed by High Performance Liquid Chromatography

Acifluorfen	DNOC
Aldicarb	Esfenvalerate
Aldicarb Sulfone	Fluometuron**
Aldicarb Sulfoxide	3 Hydroxy Carbofuran
Bentazon	Linuron
Bromacil	MCPA
Bromoxynil	MCPB
Carbaryl	Methiocarb
Carbofuran	Methomyl
Chloramben	1-Napthol
Chlorothalonil	Neburon
Chlopyalid	Norflurazon
2,4,-D**	Oryzalin
2,4,5-DB	Oxamyl
2,4,5-T	Picloram
Dacthal Mono-acid	Propham
Dicamba	Propoxur
Dichlobenil	Silvex
Dichlopro	Triclopyr
Dinoseb	
Diuron*	

[1] Note: Both methods have a few analytes in common.
* Pesticide detected in water samples collected from the intake at Lake Bruin.
** Pesticide detected in water samples collected from the treatment plant and from the intake at Lake Bruin.

Table 3. Herbicide and insecticide concentrations detected in samples collected from raw (untreated) water from Lake Bruin, Louisiana, April through December 1999.

[All pesticides and aquatic criteria levels are reported in micrograms per liter; <, less than; CAN, Canadian water-quality guideline for freshwater aquatic life (for constituents in water); AQCR, chronic criterion for the protection of aquatic life or wildlife. This applies to constituents in water, sediment (for protection of fish-eating wildlife).]

Compound	Sampling Date 1999											Median	Aquatic criteria (CAN-AQCR)
	April 29	May 13	June 2	June 16	July 1	July 13	July 27	Aug. 12	Aug. 26	Sept. 8	Dec. 28		
Atrazine*	0.962	0.998	0.927	0.974	0.972	0.918	0.991	0.841	0.863	0.960	0.688	0.960	1.8
Cyanazine*	0.136	0.131	0.124	0.109	0.166	0.332	0.248	0.227	0.192	0.238	0.151	0.166	2
Metolachlor*	0.033	0.031	0.026	0.023	0.021	0.025	0.020	0.017	0.016	0.018	0.009	0.021	7.8
Simazine*	0.151	0.168	0.152	0.164	0.160	0.184	0.159	0.124	0.154	0.156	0.137	0.156	10
Chlorpyrifos**	0.008	0.005	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	0.006	<0.004	0.005	0.04
Malathion**	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	0.106	0.023	0.040	<0.005	0.040	0.1

*Herbicide

**Insecticide

Table 4. Herbicide concentrations detected in samples collected from the finished drinking water from Lake Bruin, Louisiana, April through December 1999.

[All pesticides and advisory levels are reported in micrograms per liter; <, less than]

Compound	Sampling Date 1999											Median	Health Advisory Level (HAL)* (MCL)**
	April 29	May 13	June 2	June 16	July 1	July 13	July 27	Aug. 12	Aug. 26	Sept. 8	Dec. 28		
Atrazine	0.851	0.817	0.745	0.831	0.832	0.849	0.768	0.758	0.754	0.711	0.733	0.768	3**
Cyanazine	0.104	0.108	0.091	0.094	0.117	0.355	0.154	0.154	0.175	0.109	0.170	0.117	1*
Metolachlor	0.027	0.026	0.023	0.021	0.021	0.023	0.018	0.016	0.015	0.008	0.014	0.021	70*
Simazine	0.129	0.138	0.119	0.142	0.137	0.173	0.132	0.102	0.138	0.119	0.133	0.113	4**

*HAL, health-advisory level, lifetime,70-kg adult, assumes consumption of 2 L water per day over a 70-year lifetime; USEPA, 1999, Drinking water regulations and health advisories. Office of water.

**MCL, maximum contaminant level in drinking water; USEPA, 1999, Drinking water regulations and health advisories, Office of water.

Figure 1. Location of Lake Bruin, Louisiana.