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Kyle E. Juracek* and Andrew C. Ziegler

ABSTRACT

The historical and ongoing lead (Pb) contamination caused by the 20th-century use of leaded gasoline was investigated by an analysis of bottom sediment in eight small rural reservoirs in eastern Kansas, USA. For the reservoirs that were completed before or during the period of maximum Pb emissions from vehicles (i.e., the 1940s through the early 1980s) and that had a major highway in the basin, increased Pb concentrations reflected the pattern of historical leaded gasoline use. For at least some of these reservoirs, residual Pb is still being delivered from the basins. There was no evidence of increased Pb deposition for the reservoirs completed after the period of peak Pb emissions and (or) located in relatively remote areas with little or no highway traffic. Results indicated that several factors affected the magnitude and variability of Pb concentrations in reservoir sediment including traffic volume, reservoir age, and basin size. The increased Pb concentrations at four reservoirs exceeded the U.S. Environmental Protection Agency threshold-effects level (30.2 mg kg^{-1}) and frequently exceeded a consensus-based threshold-effects concentration (35.8 mg kg^{-1}) for possible adverse biological effects. For two reservoirs it was estimated that it will take at least 20 to 70 yr for Pb in the newly deposited sediment to return to baseline (pre-1920s) concentrations (30 mg kg^{-1}) following the phase out of leaded gasoline. The buried sediment with elevated Pb concentrations may pose a future environmental concern if the reservoirs are dredged, the dams are removed, or the dams fail.

LEAD (Pb) is an important environmental contaminant because of its known toxicity to humans and other living organisms (Forstner and Wittmann, 1981; Adriano, 2001). Over the past century, a major source of Pb in the environment was particulate emissions from vehicles. The Pb originated from the combustion of leaded gasoline, which was introduced in the 1920s and quickly became standard (Davies, 1990). From 1941 to 1970 in the USA, Pb consumption by the gasoline industry increased 457% from 45350 to 252600 metric tons (Forstner and Wittmann, 1981). During the period of maximum leaded gasoline use (i.e., the 1940s through the early 1980s), vehicle emissions became the predominant source of Pb to the environment. For example, in 1970, total national emissions of Pb from vehicles accounted for about 80% of all emissions and were about 570 times greater than the total Pb emissions from coal-fired powerplants (USEPA, 2000).

Use of leaded gasoline increased until its phase out, legislated by the Clean Air Act of 1970, began in the 1970s. The subsequent rapid decline in gasoline Pb consumption in the USA was estimated to be about 75% from 1975 to 1985 (Alexander and Smith, 1988). From 1970 to 1990, total national Pb emissions from vehicles decreased an estimated 99.8% (USEPA, 2000). Total national Pb emissions from all sources for 1900 through 1998, as well as total emissions from on-road vehicles for selected years, are shown in Fig. 1.

Numerous studies have documented increased Pb concentrations in air, soil, and vegetation located near streets and highways (e.g., Cannon and Bowles, 1962; Daines et al., 1970; Lagerwerff and Specht, 1970; Motto et al., 1970). Lead concentrations decrease rapidly in air, soil, and vegetation with distance from streets and highways and in soils with depth in the soil profile (Singer and Hanson, 1969; Daines et al., 1970; Lagerwerff and Specht, 1970; Milberg et al., 1980). Lead concentrations also are affected by prevailing wind directions (Cannon and Bowles, 1962; Page et al., 1971; Milberg et al., 1980). While much of the environmental Pb contamination from vehicular emissions was concentrated near the sources, long-distance atmospheric transport also has been documented (Boutron et al., 1991; Barrie et al., 1992; Outridge et al., 2002).

Once introduced into the environment, Pb may be transported to lakes and reservoirs via direct atmospheric deposition or by a terrestrial pathway. The terrestrial pathway involves the attachment of Pb to soil particles that are subsequently eroded and transported to lakes and reservoirs by fluvial processes. Bottom sediments within lakes and reservoirs serve as a sink for Pb and also may serve as a source of Pb to the overlying water column and biota (Baudo et al., 1990; Zoumis et al., 2001).

Previous studies have documented changes in the deposition of Pb in bottom sediment attributed to human activity. For example, Crecelius and Piper (1973) and Bertine and Mendeck (1978) documented an increase in Pb deposition in bottom sediment associated with industrialization and the use of leaded gasoline. In the former study, lead concentrations in the bottom sediment of Lake Washington (Seattle, WA) increased from a baseline of about 25 mg kg^{-1} to 400 mg kg^{-1} (1500% increase). Callender and Van Metre (1997) found an increase, and subsequent decrease, in Pb deposition for several reservoirs that reflect the history of leaded gasoline use in the USA. In the Callender and Van Metre study, the most pronounced Pb trends and largest peak concentrations were measured for the reservoirs located in urban and suburban settings. Specifically, they documented an increase in Pb concentrations from a baseline of about 18 mg kg^{-1} to 90 mg kg^{-1} (400% increase) for White Rock Lake (Dallas, TX) and an increase from a baseline of about 40 mg kg^{-1} to 150 mg kg^{-1} (275% increase) for Lake Harding (Atlanta, GA). In a study of seven Connecticut lakes, Siver and Wozniak (2001) found that Pb concentrations increased from a mean

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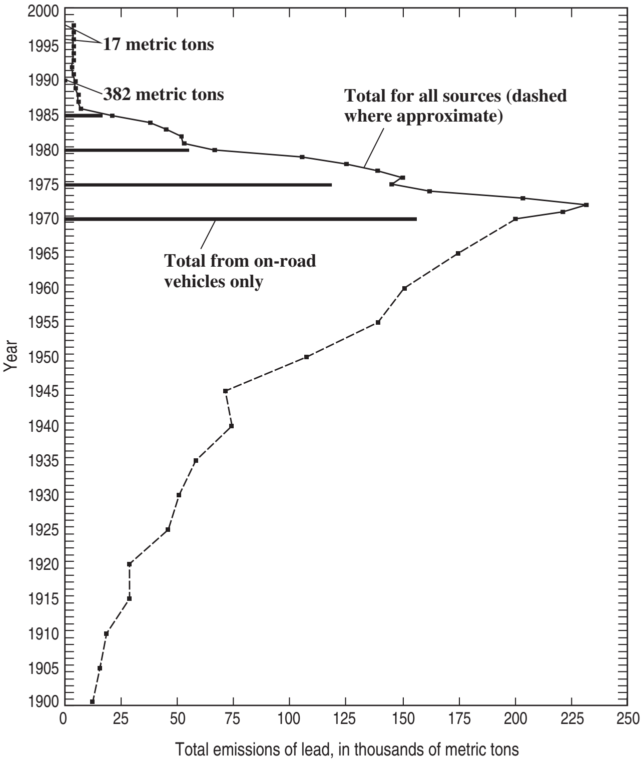

Fig. 1. Total national Pb emissions from all sources from 1900 through 1998 for the United States. Data for 1970 through 1998 from USEPA (2000). Pre-1970 emissions estimated using information for the upper Midwest from Callender and Van Metre (1997).

baseline of 31 mg kg^{-1} to a mean maximum of 310 mg kg^{-1} (900% increase).

A determination of the effects of historical leaded gasoline use on Pb concentrations in reservoir bottom sediment is important for several reasons. The Pb in the sediment may affect sediment-dwelling organisms, and it also may enter the food chain and bioaccumulate (Wong et al., 1978; Jaworski, 1987; Wright and Welbourn, 2002). Thus, such information is fundamental for understanding the effects of vehicle emissions on habitat quality. Second, an understanding of the magnitude of Pb concentrations in bottom sediment is important for future reservoir management. For example, decisions to draw down or dredge a reservoir may include consideration of the potential remobilization of sediment-associated contaminants such as Pb and the possible environmental consequences. Also, dam removal or failure may result in the introduction of large quantities of Pb-contaminated sediment to the downstream environment. Doyle et al. (2003), citing Shuman (1995), noted that the increased recent interest in dam removal as a management option has exposed a lack of fundamental information on the physical, chemical, or biological effects of removing dams. A component of the needed information is an understanding of the quality of the impounded sediment before it is released downstream. Third, there is the question of persistence. Specifically, how long will Pb from historical vehicle emissions continue to elevate Pb concentrations in sediment above baseline (i.e., pre-1920s) levels? Finally, an understanding of changes in sediment contaminant concentrations over time has implications for the estimation of contaminant loads and yields that are used in the development of total maximum daily loads.

In the study described in this paper, the goal was to investigate the effect of highway traffic on Pb concentrations in the bottom sediment of eight small reservoirs with predominantly rural basins in eastern Kansas, USA. Reservoirs with rural basins were selected to determine the effect of highway traffic in the absence of other sources of Pb often associated with urban areas (e.g., industry, powerplants). The authors were unaware of any previous study that attempted to isolate the effect of historical vehicular Pb emissions on reservoir sediment quality. Specific objectives were to: (i) quantify and interpret temporal changes in Pb concentrations in reservoir bottom sediment relative to baseline concentrations; (ii) assess the relation between sediment Pb concentrations, highway traffic, and other variables; (iii) evaluate the effect of historical vehicular Pb emissions on sediment quality using available guidelines; and (iv) estimate the time required for Pb to return to baseline concentrations in newly deposited sediment.

METHODS

Description of Study Areas

The eight small reservoirs included in this study are located throughout eastern Kansas (Fig. 2) and have completion dates ranging from 1879 to 1993. The reservoir basins range in size from less than 2 to about 36 km^2. Original water-storage capacities range from 0.3 to 6.9 million m^3 (Table 1). Current capacities are less due to the effects of ongoing sedimentation. The reservoirs have sufficient capacity to accommodate substantial additional sediment storage. Available information indicated that none of the reservoirs have been dredged.

Physiographically, the reservoir basins can be characterized with reference to the physical divisions as defined by Fenneman (1946) and Schoewe (1949). All of the basins are located within the Central Lowland Province of the Interior Plains (Fenneman, 1946). Within the Central Lowland Province, the basins are located within three separate sections—the Dissected Till Plains, the Osage Plains, and the Arkansas River Lowlands (Schoewe, 1949) (Fig. 2).

The reservoir basins located in the Dissected Till Plains of northeast Kansas include Centralia, Gardner City, Mission, and Pony Creek Lakes (Fig. 2). The Dissected Till Plains are characterized by dissected deposits of glacial till that consist of clay, silt, sand, gravel, and boulders that overlie bedrock of primarily shale and limestone, with some sandstone (Jordan and Stamer, 1995). Slopes in the basins are typically less than 10% (USDA-SCS, 1960, 1979a, 1982b). However, in the Gardner City Lake Basin, slopes near the streams may be as much as 20% (USDA-SCS, 1979a).

Three basins are located within the Osage Plains that cover much of east-central and southeast Kansas. Within the Osage Plains, Bronson City and Crystal Lakes are located in the Osage Cuestas, whereas Otis Creek Reservoir is located in the Flint Hills Upland (Fig. 2). The Osage Cuestas generally consist of a series of irregular northeast-southwest trending escarpments between which are flat to gently rolling plains. The topography of the Flint Hills Upland is characterized as gently rolling. Throughout the Osage Plains, the underlying bedrock is primarily limestone and shale (Schoewe, 1949). Slopes in the

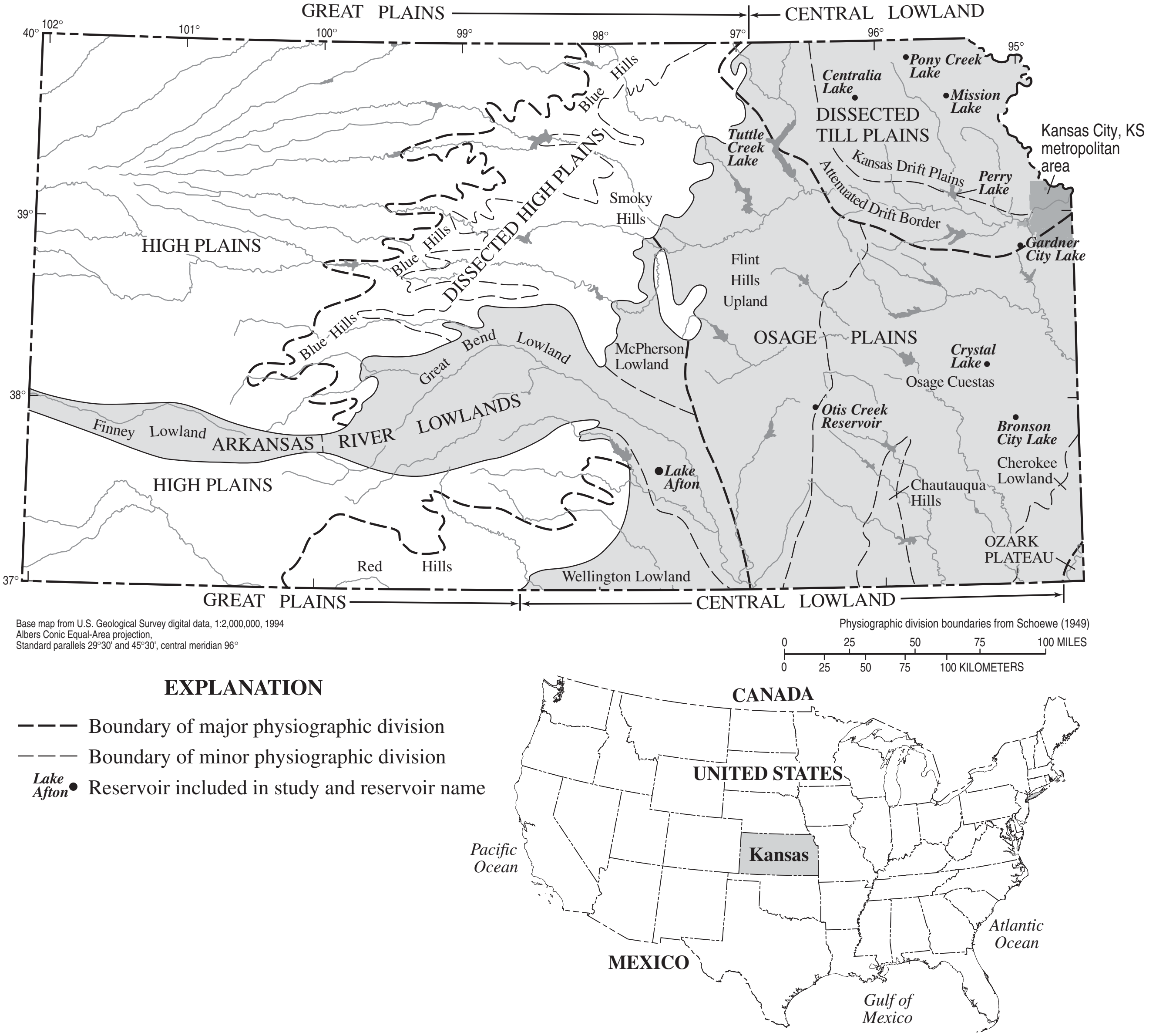

Fig. 2. Physiography of Kansas and location of small reservoirs included in study.

Bronson City and Crystal Lake Basins are generally less than 5% (USDA-SCS, 1977, 1981). In the Otis Creek Reservoir Basin slopes are generally 5 to 20% (USDA-SCS, 1982a).

Lake Afton, in south-central Kansas, is located in the Great Bend Lowland of the Arkansas River Lowlands (Fig. 2). In this area, the topography is generally flat with little relief. Typically, slopes in the basin are 3% or less (USDA-SCS, 1979b). Surficial materials are mostly sand and gravel (Schoewe, 1949).

Long-term mean annual precipitation for Centralia, Mission, and Pony Creek Lakes is about 89 cm. For Bronson City, Crystal, and Gardner City Lakes, the long-term mean annual precipitation is about 99 cm. For Lake Afton and Otis Creek Reservoir, the long-term mean annual precipitation is about 76 and 84 cm, respectively (High Plains Regional Climate Center, 2002). Most of the annual precipitation is received during the growing season (generally, April-September).

Land use (1988 through 1990) in the basins is mostly an agricultural mix of cropland and grassland (Kansas Applied Remote Sensing Program, 1993). The percentage of cropland in the basins ranges from 0.1% for Otis Creek Reservoir to 81.0% for Lake Afton. Grassland percentages range from 14.3% for Lake Afton to 96.4% for Otis Creek Reservoir. Woodland accounts for less than 7% of each basin. With two exceptions, urban land use in the basins may be considered negligible. The exceptions are the Crystal and Gardner City Lake Basins, which have 18.0 and 17.3% urban land use, respectively (Table 2). Gardner City Lake is situated on the southwestern fringe of the rapidly growing Kansas City metropolitan area. With the exceptions of Centralia Lake and Otis Creek Reservoir, every reservoir has a U.S. highway within its basin.

Site Selection and Sediment Sampling

Bottom-sediment cores were collected from each reservoir either in the fall of 2002 or the spring of 2003. With the exception of Otis Creek Reservoir, the cores were collected from a site located in the downstream one-third of the reservoir relatively close to the dam. The near-dam site was selected because it is the deepest part of each reservoir where the sediment is fine-grained and least likely to be disturbed.

Table 1. Completion date, approximate basin size, and original water-storage capacity for eight small reservoirs in eastern Kansas.

Reservoir (Fig. 2)	Year completed	Approximate basin size	Original water-storage capacity†
		km^2	million m^3
Bronson City Lake	1956	2.1	–‡
Centralia Lake	1990	32.4	5.9
Crystal Lake	1879	1.6	0.3
Gardner City Lake	1940	14.2	2.8
Lake Afton	1942	26.9	4.0
Mission Lake	1924	22.3	2.3
Otis Creek Reservoir	1971	36.3	6.9
Pony Creek Lake	1993	17.1	2.9

† Original water-storage capacity from Juracek (2004).
‡ –, not available or not determined.

The cores were collected in water depths that ranged from 3.4 to 10.7 m. With the exception of Otis Creek Reservoir, the cores were collected using a Benthos gravity corer mounted on a pontoon boat. The liner used in the gravity corer was cellulose acetate butyrate transparent tubing (with a 6.67-cm i.d.). At Otis Creek Reservoir, because of restricted boat access and relatively thin sediment deposits, sediment samples were collected from the middle of the reservoir using a Wildco box corer. The inside dimensions of the transparent plastic liner used in the box corer were 14 cm long by 14 cm wide by 20.3 cm deep. Overall, the collected cores ranged in length from 0.2 to 2.4 m.

Sediment samples for Otis Creek Reservoir were removed onsite directly from the box corer, placed in sample jars, and homogenized using a plastic stirring rod. The sediment cores collected from the other reservoirs were refrigerated (at 4 to 5°C) and processed within 1 wk after collection at the U.S. Geological Survey (USGS) laboratory in Lawrence, KS. The core liners were cut lengthwise in two places 180° apart. The cuts were completed with a 10.2-cm hand-held circular saw with its blade set at a depth to minimize penetration of the sediment cores. The cores were split in half by pulling a tightly held nylon string through the length of the cores and allowing the halves to separate. Once split, the relatively undisturbed inner parts of the cores were exposed for examination and sampling. On the basis of differences in moisture content, texture, and organic matter content (e.g., root hairs, sticks, leaves), the boundary between the bottom sediment and the underlying original (pre-reservoir) land-surface (or channel-bed) material was determined. Typically, the bottom sediment was characterized by higher moisture content, finer texture, and little if any visible organic matter as compared to the original material.

For each reservoir, the number of core intervals sampled was dependent on the age of the reservoir, the length of the core, and the amount of material required for analyses. From each interval, a representative volume of sediment was removed with care taken to avoid the sediment that came into contact with the core liner and the saw blade. The removed material was homogenized and sampled for subsequent analyses.

Age dating of the sediment samples, using cesium-137 (^{137}Cs), was performed for Crystal Lake, Gardner City Lake, Lake Afton, and Mission Lake. Age dating served the dual purpose of verifying that the bottom sediment was relatively undisturbed if the 1963 to 1964 ^{137}Cs peak was well-defined and followed by a generally uniform, exponential decrease in activity to the top of the core. For Crystal Lake, 15 intervals were analyzed for age-dating purposes, whereas 10 intervals were analyzed for each of the other three reservoirs. Due to an insufficient amount of material, age dating of the sediment was not performed for Bronson City Lake. Age dating was not performed for the sediment in Centralia Lake, Otis Creek Reservoir, and Pony Creek Lake because the reservoirs were too young for a meaningful analysis.

For Pb analyses, the objective was to analyze enough samples within each core to enable the determination of a representative median concentration as well as an indication of temporal trends. For Bronson City Lake, limited core length dictated that three intervals were sampled for analyses. For Centralia and Pony Creek Lakes, three intervals were appropriate given the young age of the reservoirs. Five intervals were sampled for Gardner City Lake, Lake Afton, Mission Lake, and Otis Creek Reservoir. For Crystal Lake, the core was of sufficient length to accommodate 10 intervals, which was justified given the older age of the reservoir. Also, for Lake Afton, a second core was sampled for 10 intervals to enable a comparison with the five-interval core to assess the effects of averaging on Pb concentrations. All sediment samples used for Pb determinations were subjected to a particle-size analysis that used standard methods (Guy, 1969; Grosbois et al., 2001).

Laboratory Analysis

For age-dating purposes, the bottom-sediment samples were analyzed for ^{137}Cs activity. Analyses were performed at the USGS National Water-Quality Laboratory in Denver, Colorado. The samples were dried and pulverized with a mortar and pestle to achieve a homogenous sample matrix. Activities of ^{137}Cs were measured by counting the samples in a calibrated geometry by gamma-ray spectrometry using a germanium detector (ASTM, 2003).

The bottom-sediment samples were analyzed for Pb content as bulk samples. Analyses were performed at the USGS Sediment Trace Element Partitioning Laboratory in Atlanta, Georgia. The samples were oven dried at 105°C. For each sample, a 500-mg aliquot was digested with a combination of HF/$HClO_4$/aqua regia in Teflon beakers at 200°C. The resulting salts were solubilized using 50 mL of 2% HCl. The Pb concentration was determined by flame atomic absorption spectrometry using mixed salt standards and background correction.

Table 2. Land-use percentages for eight small reservoir basins in eastern Kansas. Land-use data from Kansas Applied Remote Sensing Program (1993).

	Land use (% of basin)					
Reservoir (Fig. 2)	Cropland	Grassland	Woodland	Urban	Water	Other
Bronson City Lake	61.0	29.5	6.5	0	3.0	0
Centralia Lake	77.1	17.6	2.1	0	3.0	0.1
Crystal Lake	10.7	65.3	0.7	18.0	5.3	0
Gardner City Lake	30.8	43.0	3.4	17.3	4.3	1.1
Lake Afton	81.0	14.3	0.9	0	3.5	0.3
Mission Lake	69.9	25.0	1.4	0.5	3.1	0.1
Otis Creek Reservoir	0.1	96.4	0.1	0	3.4	0
Pony Creek Lake	52.8	32.8	3.5	5.6	5.2	0.1

The detection limit was 1.0 mg kg^{-1}. For quality assurance, precision was monitored by the replicate analyses of selected samples and by the concomitant digestion and analysis of standard soil reference samples available from USGS and the National Institute of Standards and Technology (Horowitz et al., 1989, 2001). In this study, six split-replicate samples (representing six different reservoirs) were analyzed. Analytical variability was ±6%. For the reference soil samples, the precision typically was within ±10% of the most probable value.

Statistical Analysis

To identify the most important variables for explaining the differences in median Pb concentrations among reservoirs, nonparametric Spearman's rho correlation coefficients (with a significance level of 0.05) were computed (Helsel and Hirsch, 1992). The explanatory variables considered included the following: reservoir age, basin size, total length of U.S. highway in basin, shortest distance from highway to reservoir, highway density in basin, average traffic volume, product of highway density and average traffic volume, and percentage of urban land use in basin (Tables 1, 2, and 3).

Analysis of Pb concentrations in relation to traffic volume was constrained by the availability of information. The Kansas Department of Transportation (KDOT) provides comprehensive data on annual average daily traffic counts for all major highways and many secondary roads in Kansas. However, complete information for all of the secondary roads in the reservoir basins was not available. Thus, for the purposes of this study, the analysis focused on the presence of major highways in each basin and the attendant traffic-volume data from KDOT. The means of annual average daily traffic counts for the years 1956, 1962, 1967, 1972, 1977, and 1982 were computed to provide representative information on traffic volume within the basins for most of the period of maximum Pb emissions from vehicles (i.e., 1940s through the early 1980s). It was assumed that the available information, although not complete, was nevertheless sufficient to enable comparisons among the reservoirs.

RESULTS AND DISCUSSION

To assess historical changes in the Pb concentrations of the reservoir bottom sediment, a baseline (i.e., pre-1920s) was established for the purpose of comparison. An accepted approach for determining the enrichment of recent sediment is comparison to a shale standard. Forstner and Wittmann (1981), citing Turekian and Wedepohl (1961), stated that the average Pb content in shale is 20 mg kg^{-1}. Davies (1990) reported an average Pb content of 23 mg kg^{-1} for shale.

In addition to values reported in the literature, Pb concentrations in sediment cores for selected reservoirs also were considered for baseline. Of the eight reservoirs included in this study, Otis Creek Reservoir was least affected by human activity. Its basin is almost exclusively grassland (Table 2) and includes no highways. Bottom sediment concentrations of Pb were very consistent over time (Table 4), and the minimal variability (within ±5% of the median value) may be due, in part, to analytical variance. For Otis Creek Reservoir, the median concentration of Pb in the bottom sediment was 23 mg kg^{-1}. Rice (1999) found streambed-sediment samples collected in two predominantly rural areas to have trace-element concentrations (including Pb) similar to the local baseline (i.e., minimally affected by human activity).

If a reservoir is sufficiently old and the bottom sediment has not been disturbed, the deepest (oldest) sediment may provide an indication of baseline conditions. Completed in 1879, Crystal Lake satisfies these criteria. The profile of ^{137}Cs activity (i.e., well-defined peak followed by a uniform, exponential decline) confirmed that the sediment in Crystal Lake has not been disturbed (Fig. 3A). The deepest two intervals (1 and 2) of the core collected from Crystal Lake, interpreted to be representative of sediment deposited mostly during the late 1800s (i.e., well before the advent of leaded gasoline), had an average Pb concentration of 30 mg kg^{-1} (Fig. 3A). Thus, when compared to Otis Creek Reservoir, regional differences in baseline Pb concentrations were apparent. Variability in baseline Pb concentrations also has been documented elsewhere. For example, in a study of seven Connecticut lakes, Siver and Wozniak (2001) reported a range in baseline Pb concentrations of 12 to 54 mg kg^{-1} with a mean of 31 mg kg^{-1}. To enable an assessment of changes in Pb deposition in and among the eight reservoirs, the Pb baseline concentration for this study was set at 30 mg kg^{-1}. Reservoirs with bottom-sediment concentrations less than the baseline were considered to be minimally affected by human sources of Pb.

Table 3. Median Pb concentrations in bottom sediment, total U.S. highway length in each reservoir basin, shortest distance from highway to reservoir, highway density in each basin, average daily traffic volume, and product of highway density and average traffic volume. All values rounded to two or three significant digits.

Reservoir (Fig. 2)	Median Pb concentration in bottom sediment	Total length of U.S. highway in basin	Shortest distance from highway to reservoir	Highway density in basin†	1956–82 Average daily traffic volume (number of vehicles)‡	Product of highway density and average traffic volume
	mg kg^{-1}	km	km	km km^{-2}		
Bronson City Lake	34	1.8	0.2	0.9	1660	1490
Centralia Lake	19	0	–¶	0	0	0
Crystal Lake	59§	1.2	<0.03	0.8	2710	2170
Gardner City Lake	42	1.9	2.8	0.1	4660	466
Lake Afton	45	4.0	4.6	0.1	4550	455
Mission Lake	29	4.7	0.4	0.2	1670	334
Otis Creek Reservoir	23	0	–	0	0	0
Pony Creek Lake	25	5.2	0.2	0.3	1530	459

† Highway density computed as total highway length divided by basin area.
‡ Data from the Kansas Department of Transportation (personal communication, 2003).
§ Computed using the top (most recent) seven core intervals only.
¶ –, not determined.

Table 4. Concentration of Pb in bottom sediment of eight small reservoirs in eastern Kansas. Completion date for each reservoir is indicated parenthetically.

	Pb concentration†								
Sediment-core interval	**Bronson City Lake (1956)‡**	**Centralia Lake (1990)**	**Crystal Lake (1879)‡**	**Gardner City Lake (1940)‡**	**Lake Afton (1942)‡**		**Mission Lake (1924)‡**	**Otis Creek Reservoir (1971)**	**Pony Creek Lake (1993)**
	mg kg^{-1}								
Top (most recent)	_32_	19	_51*_	_40*_	_39*_	_34_	29	23	25
	_34_	19	_59*_	_50*_	_53*_	_35_	30	23	24
	_38*_	17	_65*_	_49*_	_54*_	_43*_	_31_	23	25
	–	–	_62*_	_42*_	_45*_	_55*_	24	23	–
	–	–	_64*_	_39*_	_34_	_55*_	25	24	–
	–	–	_46*_	–	–	_50*_	–	–	–
	–	–	_36*_	–	–	_43*_	–	–	–
	–	–	_31_	–	–	_36*_	–	–	–
	–	–	_32_	–	–	_31_	–	–	–
Bottom (oldest)	–	–	28	–	–	28	–	–	–

† Concentrations in italic exceed the USEPA (1997) threshold-effects level (TEL) (30.2 mg kg^{-1}) for possible adverse biological effects. Concentrations marked with an asterisk (*) exceed the MacDonald et al. (2000) consensus-based threshold-effects concentration (TEC) (35.8 mg kg^{-1}) for possible adverse biological effects.

‡ Lead depositional profile reflects history of leaded gasoline use.

Lead Depositional Histories

Of the eight reservoirs sampled, five (Bronson City Lake, Crystal Lake, Gardner City Lake, Lake Afton, and Mission Lake) had a Pb depositional profile in the sediment core that reflected the history of leaded gasoline use (Table 4). The best example is provided by the relatively detailed core analyzed for Crystal Lake (Fig. 3A) that represents 124 yr of sediment deposition. The bottom (oldest) part of the core, intervals 1 through 3, had baseline Pb concentrations. Progressing forward in time, Pb concentrations increased and culminated with a peak concentration of 65 mg kg^{-1} for interval 8. A side-by-side comparison of the Pb and ^{137}Cs (Fig. 3A) profiles indicated that the Pb peak occurred in the 1970s. Subsequently, Pb concentrations declined to a value of 51 mg kg^{-1} for the top (most recent) interval. Over the life of the reservoir, Pb concentrations in the bottom sediment more than doubled (117% increase) from the initial baseline conditions before declining. From the peak, Pb concentrations have decreased by at least 22% to the present. Because each core interval integrates several years of deposition, the most recent sediment likely has Pb concentrations less than 51 mg kg^{-1}. Although the elimination of leaded gasoline was essentially achieved by 1990, the increased Pb concentration (relative to baseline) for interval 10 at the top of the core indicated that residual Pb was still being transported into the reservoir from the basin post-1990. Interval 10 includes sediment deposited up until the time of core collection in 2003.

Additional evidence in support of vehicular traffic as the source of Pb was provided by Zn concentrations in the sediment. Over the life of Crystal Lake, a statistically significant increasing trend in Zn deposition was indicated (Juracek, 2004). A significant source of Zn is vehicular tire wear. Callender and Rice (2000) determined that increased Zn concentrations in sediment are related to increased vehicular traffic.

A similar Pb depositional profile was indicated for Gardner City Lake, Lake Afton, and Mission Lake (Table 4). The Pb concentrations in the bottom sediment of these three reservoirs likely were affected to an unknown extent by sediment storage in multiple smaller upstream impoundments located between the highway and the reservoir in each basin. For Gardner City Lake (completed in 1940), Pb concentrations increased from an initial value of 39 mg kg^{-1} at the bottom of the core to a peak of 50 mg kg^{-1} (28% increase) before declining to 40 mg kg^{-1} (20% decrease). The larger initial Pb concentration, relative to the baseline, is consistent with the age of the reservoir, which was completed after leaded gasoline had already been in use for several years. Thus, assuming that the increased Pb concentrations were attributable primarily to historical vehicle emissions, it is anticipated that Pb concentrations in newly deposited bottom sediment of Gardner City Lake eventually will stabilize at a level less than 39 mg kg^{-1}.

For Lake Afton (completed in 1942), Pb concentrations in the five-interval core increased from an initial value of 34 mg kg^{-1} to a peak of 54 mg kg^{-1} (59% increase) before declining to 39 mg kg^{-1} (28% decrease). For the 10-interval core (Fig. 3B), Pb concentrations increased from an initial value of 28 mg kg^{-1} to a peak of 55 mg kg^{-1} (96% increase) before declining to 34 mg kg^{-1} (38% decrease). A comparison of the five- and 10-interval cores documented the expected effect of averaging when fewer intervals are used to represent the same sediment profile. Specifically, the range in Pb concentrations for the 10-interval core (28 to 55 mg kg^{-1}) was reduced in the five-interval core (34 to 54 mg kg^{-1}) by 26%. Assuming that the increased Pb concentrations were attributable primarily to historical vehicle emissions and given that the oldest (deepest) sediment had a Pb concentration of 28 mg kg^{-1}, it is anticipated that Pb concentrations in newly deposited bottom sediment of Lake Afton eventually will stabilize at a level less than 34 mg kg^{-1}.

For Mission Lake (completed in 1924), the Pb depositional profile indicated an analogous, although somewhat less pronounced, history. From an initial concentration of 25 mg kg^{-1}, Pb peaked at 31 mg kg^{-1} (24% increase) and then declined to 29 mg kg^{-1} (6% decrease). The fact that all of the Pb concentrations were near or less than baseline (30 mg kg^{-1}) throughout the life of the reservoir indicated that the historical use of leaded gasoline had a relatively smaller effect in this reservoir compared

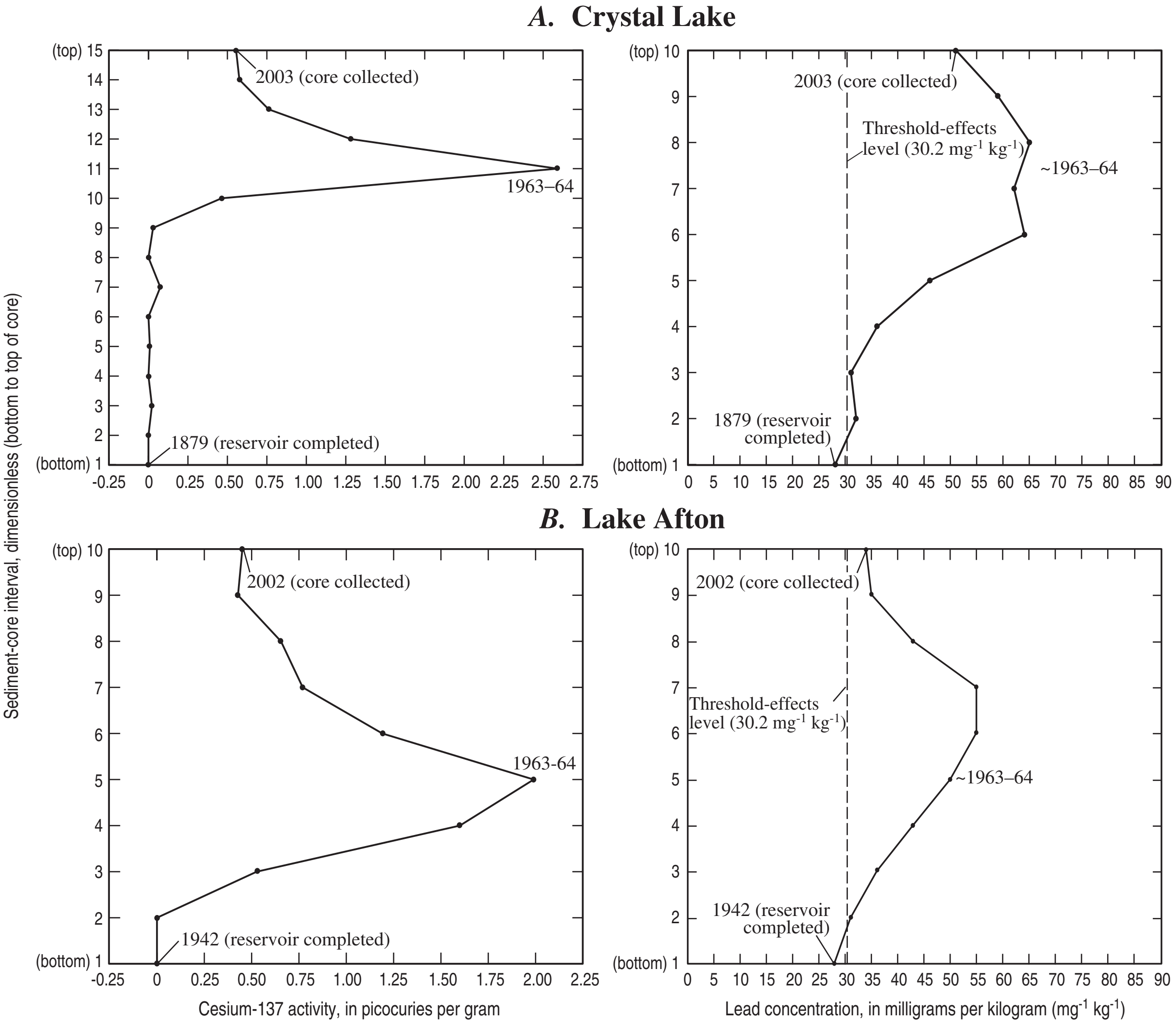

Fig. 3. Cesium-137 and Pb depositional profiles for (A) Crystal Lake and (B) Lake Afton. Sediment-quality guidelines from USEPA (1997).

to the previously discussed reservoirs. Also, given the limited range in concentrations, analytical variability may have accounted for some of the change. Because the deepest core interval corresponds to the time period that represents the first few years of leaded gasoline use, the measured Pb concentration probably is close to the true baseline for the basin.

The Pb depositional profile for Bronson City Lake (completed in 1956), although limited to three intervals, nevertheless indicated a possible decrease in Pb concentrations from 38 mg kg^{-1} at the bottom of the core to 32 mg kg^{-1} at the top. The decrease is consistent with the history of leaded gasoline use. Although, given the limited range in concentrations, analytical variability is another possible explanation.

For the three remaining reservoirs, historical leaded gasoline use has had a minimal effect on Pb concentrations in the bottom sediment. All three reservoirs had Pb concentrations that were uniform over time and less than baseline. Otis Creek Reservoir (completed in 1971) is located in a relatively remote area. The basin is almost exclusively grassland and includes no highways. Moreover, no highways are located within several kilometers of the basin in every direction. The existence of both Centralia Lake (completed in 1990) and Pony Creek Lake (completed in 1993) postdates the period during which Pb emissions from vehicle exhaust were largest (i.e., 1940s through the early 1980s). Thus, the Pb deposited in these two basins during the period of peak emissions may have partly or mostly been transported downstream by fluvial processes before construction of the dams. Also, dilution of the Pb concentrations by "clean" sediment (i.e., sediment minimally contaminated by vehicle Pb emissions) within the basins is a likely contributing factor. In all three cases, the uniformity of the Pb concentrations over time (i.e., the lack of a decreasing trend) indicates that the concentrations are at or near the true baseline for each reservoir.

Lead Concentrations and Explanatory Variables

Several variables may account, in part, for the Pb concentrations measured in the reservoir bottom sediment. Sediment characteristics that can affect Pb concentrations include particle size (e.g., sand content) and organic matter content (Horowitz, 1991). All sediment samples analyzed for this study (i.e., the individual core intervals) had a silt and (or) clay content of 98% or greater and a total organic carbon (TOC) content generally in the range of 2.0 to 3.5% (Juracek, 2004). Thus, particle size likely is not important for explaining differences in Pb concentrations within the individual reservoir sediment cores. Normalization using TOC did not substantially affect the relative magnitudes and trends of the Pb concentrations within the individual cores.

Statistical analysis indicated significant positive correlations between median Pb concentrations and average traffic volume (1956 through 1982), the product of highway density and average traffic volume, and reservoir age (Table 5). Of these three variables, average traffic volume was the best predictor of median Pb concentration. The strong correlation between traffic volume and median Pb concentrations was not surprising because the primary source of Pb was from vehicular emissions. The correlation between reservoir age and median Pb concentrations was indicative of the fact that the amount of Pb deposition was directly related to the amount of overlap between the existence of the reservoir and the period of maximum Pb emissions.

A negative correlation (significant at the 0.06 level) was indicated between median Pb concentration and basin size (Table 5). This relation is possibly suggestive of the importance of basin size as an indicator of the availability of "clean" sediment to dilute Pb concentrations in reservoir bottom sediment. Alternatively, the relation may be a result of the small sample size and (or) sediment storage in the basin.

No statistically significant correlations were indicated between median Pb concentrations and total length of U.S. highway in each basin, shortest distance from highway to reservoir, highway density in each basin, and percentage of urban land use in each basin (Table 5). However, for Crystal Lake, distance from highway to reservoir apparently was important. Crystal Lake had the largest median Pb concentration despite a traffic volume that was about 40% less than the traffic volume for the highways in the Gardner City Lake and Lake Afton Basins. The shortest distance from the highway to the shore of Crystal Lake is less than 0.03 km, whereas the respective shortest distances for Gardner City Lake and Lake Afton are about 2.8 and 4.6 km, respectively (Table 3). Thus, Crystal Lake may have received more Pb via direct atmospheric deposition. Also, basin size may be an important factor. In this case, the larger Pb concentrations in the Crystal Lake sediment may be partly because of the small basin size (relative to the other two reservoirs, see Table 1) and the associated limited potential for dilution by "clean" sediment and (or) sediment storage.

Table 5. Correlations between median Pb concentration in bottom sediment and several explanatory variables.

Explanatory variable	Spearman's rho	Two-sided *p*-value
Reservoir age	0.76	0.02800
Basin size	−0.69	0.05799
Total length of U.S. highway in basin	0.25	0.54794
Shortest distance from highway to reservoir	0.03	0.95653
Highway density in basin	0.53	0.17651
Average traffic volume (1956–82)	0.87	0.00451
Product of highway density and traffic volume	0.78	0.02287
Percentage urban land use in basin	0.49	0.21270

Overall, it is apparent that several factors can affect the magnitude and temporal variability of Pb concentrations in the bottom sediment of small rural reservoirs in eastern Kansas. Statistically significant factors (at the 0.06 level or better) included traffic volume, reservoir age, and basin size. Also, Pb concentrations likely are affected by a relatively substantial percentage of urban land use in the Crystal Lake and Gardner City Lake Basins (Table 2).

The presence or absence of temporal trend in Pb concentrations documented in this and other studies suggests that basin size may be an important determinant of Pb concentrations in the bottom sediment of rural reservoirs. In this study, sediment cores collected from five of eight small rural reservoirs displayed a Pb depositional profile that reflected the history of leaded gasoline use. The basins for these five reservoirs are less than 30 km^2 in size (Table 1). However, for rural reservoirs with larger basins, the Pb concentrations attributable to historical leaded gasoline use may be diluted by a large contribution of "clean" sediment (Callender and Van Metre, 1997). For example, a depositional trend for Pb was absent in a 10-interval core analyzed for Perry Lake, Kansas (completed in 1969) (Fig. 2) (Juracek, 2003). Mission Lake (for which a Pb depositional trend was measured) is located within the 2890-km^2 Perry Lake Basin. Elsewhere, a depositional trend for Pb was absent in a 15-interval core analyzed for Tuttle Creek Lake, Kansas (completed in 1962) (Fig. 2) (Juracek and Mau, 2002) and in a 20-interval core analyzed for Coralville Lake, Iowa (completed in 1958) (Callender and Van Metre, 1997). The respective basin sizes for Tuttle Creek and Coralville Lakes are 24 900 and 8070 km^2. Additional research is required to ascertain the threshold basin size at which the Pb contribution from vehicular emissions may be overwhelmed by "clean" sediment in a rural reservoir.

Lead Concentrations and Sediment Quality

The U.S. Environmental Protection Agency (USEPA) has adopted nonenforceable sediment-quality guidelines (SQGs) in the form of level-of-concern concentrations for several trace elements including Pb. These level-of-concern concentrations were derived from biological-effects correlations made on the basis of paired onsite and laboratory data to relate incidence of adverse biological effects in aquatic organisms to dry-weight sediment concentrations. Two such level-of-concern guidelines adopted by USEPA are referred to as the threshold-effects level (TEL) and the probable-effects level (PEL). The TEL is assumed to represent the concentration below which toxic biological effects rarely occur. In the range of concentrations between the TEL and PEL, toxic effects occasionally occur. Toxic effects usually or frequently occur at concentrations greater than the PEL. For Pb, the TEL and PEL are 30.2 and 112 mg kg^{-1}, respectively (USEPA, 1997).

The USEPA cautions that the TEL and PEL guidelines are intended for use as screening tools for possible hazardous levels of chemicals and are not regulatory criteria. This cautionary statement is made because, although biological-effects correlation identifies level-of-concern concentrations associated with the likelihood of adverse organism response, the procedure may not demonstrate that a particular chemical is solely responsible. In fact, biological-effects correlations may not indicate direct cause-and-effect relationships because coring sites may contain a mixture of chemicals that contribute to the adverse effects to some degree. Thus, for any given site, the guidelines may be over- or underprotective (USEPA, 1997). Nevertheless, the guidelines provide a means for assessing the potential toxicity of Pb concentrations in bottom sediment and for comparing reservoirs.

In this study, Pb concentrations that exceeded the TEL (30.2 mg kg^{-1}) were measured only in sediment from the five reservoirs that had Pb depositional profiles that were indicative of the effect of historical leaded gasoline use. These five reservoirs were all completed before or during the period of maximum Pb emissions from vehicles (i.e., the 1940s through the early 1980s). The Pb concentrations exceeded the TEL for all intervals of the sediment core analyzed for Bronson City and Gardner City Lakes. For Crystal Lake and Lake Afton, Pb concentrations exceeded the TEL for all but the deepest (oldest) interval. For Mission Lake, only the peak Pb concentration exceeded the TEL. No measured Pb concentrations exceeded the PEL (112 mg kg^{-1}) (Table 4).

For Pb (Table 4), as well as As, Cr, Cu, Ni, and Zn (Juracek, 2004), the sediment concentrations at the bottom of the Crystal Lake core were either similar to, or substantially larger than, the respective TELs (USEPA, 1997). Because the bottom of the Crystal Lake core likely is representative of baseline conditions, the results indicated that, for certain trace elements in certain areas, baseline concentrations may equal or exceed the TELs before the effects of substantial human activity.

MacDonald et al. (2000) developed consensus-based SQGs that were computed as the geometric mean of several previously published SQGs. The consensus-based SQGs consist of a threshold-effects concentration (TEC) and a probable-effects concentration (PEC). The TEC represents the concentration below which adverse biological effects are not expected to occur, whereas the PEC represents the concentration above which adverse biological effects are expected to occur more often than not. An evaluation of the reliability of the SQGs indicated that most of the individual TECs and PECs (including those for Pb) provide an accurate basis for predicting the presence or absence of sediment toxicity. For Pb, the TEC and PEC are 35.8 and 128 mg kg^{-1}, respectively (MacDonald et al., 2000). In the present study, Pb concentrations that exceeded the TEC were measured for four of the five reservoirs that had Pb depositional profiles indicative of the effect of historical leaded gasoline use. No measured Pb concentrations exceeded the PEC (Table 4).

Estimated Recovery Time

The relatively detailed information available for Crystal Lake and Lake Afton was used to estimate the time required for newly deposited sediment to achieve baseline Pb concentrations (i.e., 30 mg kg^{-1}). These projections required the following assumptions: (i) a uniform sedimentation rate over time; (ii) minimal postdepositional disturbance; (iii) an exponential rate of decline of Pb concentrations; and (iv) no new sources contribute to the deposition of Pb in the reservoirs. The location of the peak in the ^{137}Cs profile (Fig. 3A) supports the assumption of a uniform sedimentation rate for Crystal Lake. Likewise, the ^{137}Cs profile for Lake Afton (Fig. 3B) generally was indicative of uniform sedimentation.

For both reservoirs, a first-order rate model was fit to the Pb concentrations vs. time in years (Van Metre et al., 1998). The model is expressed as $C_t = C_o e^{-kt}$, where C_t is the Pb concentration at time t, C_o is the initial Pb concentration, k is the time constant, and t is time in years. Because each core interval represents several years of deposition, the midpoint date was assigned to each interval for the purpose of estimating the time required for Pb concentrations to return to baseline. For Crystal Lake and Lake Afton, each core interval represented about 12 and 6 yr of deposition, respectively.

For Crystal Lake, it was projected that newly deposited sediment will contain baseline Pb concentrations in about the year 2050. Thus, the total recovery time for Pb concentrations to return to baseline following the phase out of leaded gasoline (circa 1980) was estimated to be at least 70 yr.

Lake Afton was completed after the introduction of leaded gasoline and, therefore, the baseline Pb concentration was not known with certainty. However, because the Pb concentrations in the bottom (oldest) two core intervals are relatively uniform (similar to the bottom three intervals of the Crystal Lake core), it was decided that an assumed baseline of 30 mg kg^{-1} was reasonable for use in the estimation of recovery time. For Lake Afton, it was projected that newly deposited sediment contained baseline concentrations of Pb in about the year 2000. Thus, the total recovery time for Pb concentrations to return to baseline following the phase out of leaded gasoline was estimated to be at least 20 yr. Given that the true baseline for Lake Afton may be smaller than the 30 mg kg^{-1} assumed, the actual recovery time may be longer. Moreover, additional complicating factors (e.g., implementation of best management practices, sediment releases associated with the failure or removal of small upstream impoundments) may lengthen the recovery time further.

An understanding of recovery time has important management implications. In this study it was estimated that a recovery time of at least 20 to 70 yr was required for Pb concentrations in the bottom sediment of two small rural reservoirs to return to baseline following the phase out of leaded gasoline. Although based on limited data, the estimated recovery time is consistent with the decadal response time of riverine systems to changes in the regulation of persistent, sediment-associated contaminants as proposed by Van Metre et al. (1998). The lengthy recovery period is a result of several factors including the amount of Pb originally introduced to the basin, the distance from source (highway) to sink (reservoir), the time required for sediment transport (including sediment storage) from source to sink, and the availability of "clean" sediment within the basin. Results of this study indicate that attempts to restore sediment quality in small rural reservoirs to baseline conditions by the elimination of the source of persistent, sediment-associated contaminants may not achieve the desired improvement for at least several decades.

CONCLUSIONS

In what possibly represents the first such effort, this study attempted to isolate the effect of historical vehicular Pb emissions on reservoir sediment quality. For the small rural reservoirs that were completed before or during the period of maximum Pb emissions from vehicles (the 1940s through the early 1980s) and had a major highway in the basin, Pb concentrations in the bottom sediment reflected the historical use of leaded gasoline. That is, Pb concentrations increased substantially above baseline (pre-1920s) concentrations (30 mg kg^{-1}) and also exceeded the threshold-effects level (TEL) (30.2 mg kg^{-1}) for possible adverse biological effects. Likewise, the increased Pb concentrations typically also exceeded the consensus-based threshold-effects concentration (TEC) (35.8 mg kg^{-1}) for possible adverse biological effects. Subsequently, with the phase out of leaded gasoline, Pb concentrations have declined but still remain above baseline and above the TEL and TEC for some of the affected reservoirs. For at least some of these reservoirs, residual Pb is still being delivered from the basins.

For the small rural reservoirs completed after the period of maximum Pb emissions and (or) located in relatively remote areas with little or no highway traffic, there was no evidence of increased Pb deposition in the bottom sediment. In the former case, it is proposed that the highway-derived Pb deposited in the basin was partly or mostly transported downstream before construction of the dams. Also, the Pb concentrations may have been diluted by "clean" sediment within the basins.

The results of this study indicated that several factors may affect the magnitude and variability of Pb concentrations in the bottom sediment of small rural reservoirs including traffic volume, reservoir age, and basin size. A relation between Pb concentrations and total length of highway in basin, shortest distance from highway to reservoir, highway density in basin, and percentage of urban land use in basin was not indicated.

In sum, the legacy of leaded gasoline in the bottom sediment of the affected small rural reservoirs is one of increased Pb concentrations and ongoing recovery. The bottom sediment containing the largest Pb concentrations is now capped by overlying sediment with progressively smaller Pb concentrations. Eventually, given enough time (i.e., at least several decades following the phase out of leaded gasoline) and assuming no new sources of Pb, it is anticipated that Pb in the newly deposited sediment will return to baseline concentrations. However, the buried sediment with increased Pb concentrations may pose a future environmental concern if the reservoirs are dredged, the dams are removed, or the dams fail.

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