The Urban Environmental Gradient: Anthropogenic Influences on the Spatial and Temporal Distributions of Lead and Zinc in Sediments

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Urban settings are a focal point for environmental contamination due to emissions from industrial and municipal activities and the widespread use of motor vehicles. As part of the National Water-Quality Assessment Program of the U.S. Geological Survey, streambed-sediment and dated reservoir-sediment samples were collected from the Chattahoochee River Basin and analyzed for total lead (Pb) and zinc (Zn) concentrations. The sampling transect extends from northern Georgia, through Atlanta, to the Gulf of Mexico and reflects a steep gradient in population density from nearly 1000 people/km^2 in the Atlanta Metropolitan Area to fewer than 50 people/km^2 in rural areas of southern Georgia and northern Florida. Correlations among population density, traffic density, and total and anthropogenic Pb and Zn concentrations indicate that population density is strongly related to traffic density and is a predictor of Pb and Zn concentrations in the environment derived from anthropogenic activities. Differences in the distributions of total Pb and Zn concentrations along the urban−suburban−rural gradient from Atlanta to the Florida Panhandle are related to temporal and spatial processes. That is, with the removal of leaded gasoline starting in the late 1970s, peak Pb concentrations have decreased to the present. Conversely, increased vehicular usage has kept Zn concentrations elevated in runoff from population centers, which is reflected in the continued enrichment of Zn in aquatic sediments. Sediments from rural areas also contain elevated concentrations of Zn, possibly in response to substantial power plant emissions for the region, as well as vehicular traffic.

Introduction

Many of the common anthropogenic pollution problems are focused in urban settings. Besides point-source inputs to the environment, such as industrial stack emissions and effluents, there are municipal-wastewater and solid-waste incinerator inputs. In addition, there is an input of contaminants related to the process of urbanization, termed "street dust" (*1*). While these are all significant sources of urban environmental contamination, recent environmental legislation has attempted to reduce the severity and amount of the industrial inputs. A population shift from urban and rural settings to the suburban environment, however, has caused sources of contaminants to aquatic systems to be contributed by larger geographic areas. Suburbia does not contribute much by way of industrial pollution, but it does serve as a source of contaminants to the environment from household and yard wastes, storm drains, construction activities and materials, and traffic.

Lead (Pb) and zinc (Zn) have been used by humans for a variety of purposes throughout the 19th and 20th century. Point-source inputs of Pb and Zn to aquatic systems (streams, lakes, and reservoirs) include industrial effluents, municipal-wastewater effluents, and stack emissions from smelting operations and fossil-fuel combustion (*2*−*4*). From 1950 to the 1970s, automobile use increased in response to economic and population growth, and the predominant source of Pb became automobile exhaust emission of tetraethyl Pb (*5*). Population growth has continued to the present (*6*), while Pb concentrations have declined due to the removal of leaded gasoline (*7*). Environmental Zn concentrations, however, may have remained elevated because of the lack of specific regulatory actions regarding Zn.

Today, most of the new anthropogenic Pb and Zn additions to the environment are derived from material sources. Lead is used in paper, plastics, and ceramics (*1*). Zinc is used in most commercial metal products (e.g., brass, bronze, castings, galvanized metal) and is added during the manufacture of automobile tires in the form of zinc oxide (ZnO), as an accelerator in the vulcanization process (*8*). In addition, Zn is a common contaminant in agricultural and food wastes. Both Pb and Zn have significant emissions from coal fly ash, although the quantity of Zn emissions is nearly double that of Pb (*9*). Fossil fuel combustion is the main contributor to worldwide anthropogenic emissions of Zn (*10*).

The National Water-Quality Assessment (NAWQA) Program of the U.S. Geological Survey is designed to describe the quality of the Nation's water resources and to provide a sound understanding of the natural and human factors that affect the quality of these resources (*11*). The NAWQA Program, which began in 1991, consists of 59 study units, defined as study areas that comprise large river basins and/or aquifer systems. Study units are spatially distributed across the conterminous United States. In general, the NAWQA Program is targeting the predominantly residential and commercial land-use settings and de-emphasizing drainage basins with abundant point sources and large areas of industrial activity.

The focus of this paper is analysis of streambed-sediment and reservoir-sediment-core data collected from the Apalachicola−Chattahoochee−Flint (ACF) River basin NAWQA study unit in Georgia, Alabama, and Florida (Figure 1). Lead and Zn concentrations in streambed sediment collected by the study unit are integrated and interpreted with Pb and Zn concentrations in age-dated reservoir-sediment cores also collected by the NAWQA program, allowing interpretation of both spatial and temporal patterns. The spatial record of streambed-sediment samples collected during 1992−1994 extends along a longitudinal gradient (the Chattahoochee River), from rural sites north of Atlanta, through Metropolitan Atlanta, and southward to rural areas in the Florida Panhandle (Figure 1). The spatial and temporal record of reservoir-

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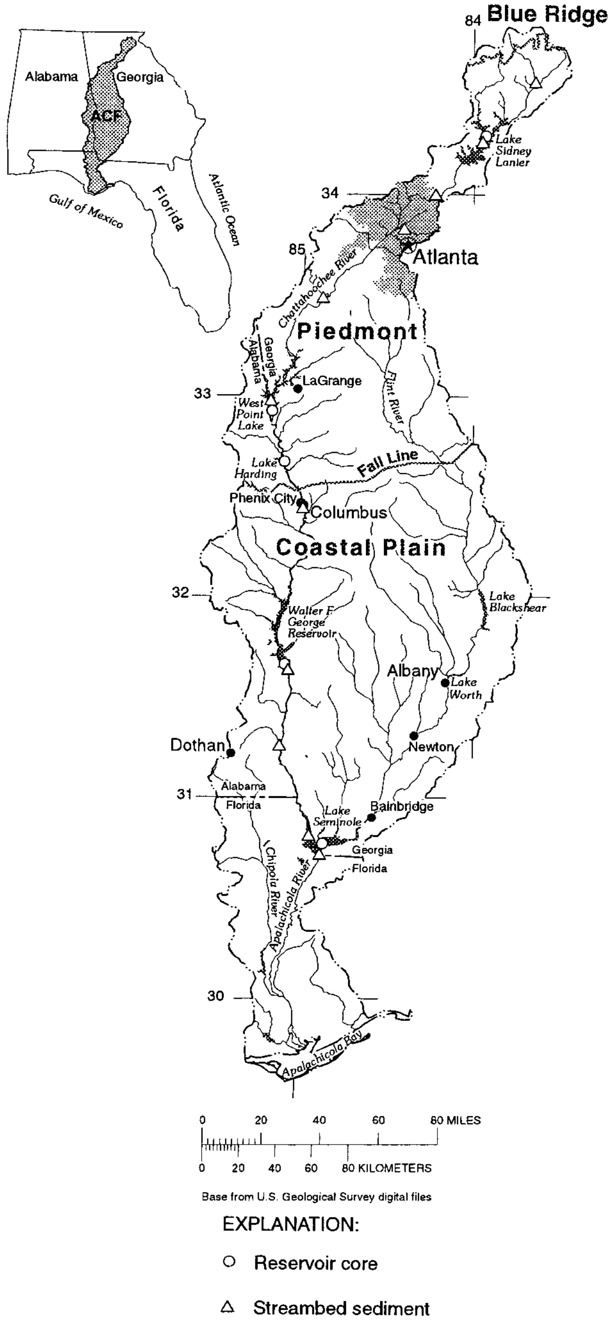

FIGURE 1. Location of study area, physiographic provinces, streambed-sediment sampling sites, and reservoir-coring sites.

sediment cores collected from five reservoirs located along this gradient extends over a period of 25 years, from 1970 to 1995.

The ACF River basin comprises 52 840 km^2 and extends from the Blue Ridge Physiographic Province in the north, through the Piedmont Province, to the Coastal Plain Province in the southern half of the basin (Figure 1). Five major hydroelectric reservoirs exist on the Chattahoochee River: Lake Sidney Lanier, West Point Lake, Lake Harding, Walter F. George Reservoir, and Lake Seminole (Figure 1). These five reservoirs account for nearly 100% of the surface-storage capacity in the ACF River basin. Population density is highest in the Upper Chattahoochee basin (835 people/km^2), which includes Metropolitan Atlanta, and decreases south of Atlanta to 90 people/km^2 around Lake Harding and fewer than 20 people/km^2 in the Lower Chattahoochee basin (*12*).

Objectives of this paper are to (1) document concentrations of Pb and Zn in streambed-sediment and reservoir-sediment-core samples; (2) show how the concentrations vary over the urban−rural gradient and are related to anthropogenic activities; and (3) demonstrate the differences in Pb and Zn concentrations as related to anthropogenic influences over both space and time.

Experimental Section

Streambed-sediment sampling sites were selected to be representative of upstream land-use activities, such as urbanization in Atlanta and Columbus, GA; surface-water impoundments; and agricultural activities in the Coastal Plain province (Figure 1; (*13*)). Samples were collected in depositional zones by wading along a cross-section of the stream. The upper 2−3 cm of sediment in five to 10 depositional zones was sampled and composited. Compositing of samples within and between depositional zones was assumed to smooth any local-scale variability and provide a sample with average concentrations of trace elements at each site (*14*). Samples were collected in the fall of one of three successive years using a hand-held core sampler or a Teflon spoon, scoop, or spatula, depending on local conditions. Sediments were sieved through a 63-μm nylon cloth, held in a plastic frame, into a glass jar. Sediment-sampling and sieving tools were cleaned between sample sites by soaking in 0.2% phosphate-free detergent, rinsing with deionized water, rinsing with a 5%, high-purity, nitric-acid solution, and rinsing multiple times with deionized water.

Reservoir-sediment cores were collected using a Benthos gravity corer fitted with a 3.05-m long, 6.3-cm diameter, plastic-lined barrel (*15*). The cores were extruded vertically and subsampled for metals and radionuclides. The sampling tools were rinsed in ambient lake water, soaked in 0.2% phosphate-free detergent, and rinsed again in ambient lake water between subsamples. All subsamples were chilled until shipped to the laboratory. Subsamples of the core were not sieved due to the small amount ($\leq$10 g) of material available. Sediment cores were collected near the reservoir dam, where silts and clays accumulate. Generally, these lacustrine sediments are characterized by uniform, fine-textured muds (*15*) and are of comparable grain size to the sieved streambed sediments.

In the laboratory, streambed-sediment samples were air-dried and ground to a fine powder. Reservoir-sediment-core subsamples were frozen, freeze-dried, and ground to a fine powder. Elemental concentrations were determined on concentrated-acid digests (nitric-hydrofluoric) using either inductively coupled plasma-atomic emission spectrometry (ICP/AES) [streambed-sediment samples; (*16*)] or graphite furnace atomic absorption spectrometry (GF/AAS) [reservoir-sediment-core samples; (*17*)]. Quality assurance was provided by determining the elemental concentrations of strong-acid digests for duplicate samples and a variety of soil, lake, and marine reference samples (*16*). Six streambed-sediment sites were sampled twice on the same day. Replicate analyses for Pb and Zn varied by ± 25% and ± 20%, respectively.

For age dating, activities of ^{137}Cs were measured by counting freeze-dried samples of reservoir-sediment cores in a fixed geometry with a high-resolution, intrinsic germanium detector γ spectrometer (*18*). Four dates can be identified in cores from reservoirs that were constructed before 1952: the reservoir impoundment date, the first occurrence of detectable ^{137}Cs in 1953, the peak ^{137}Cs concentration in 1963−1964, and the date of collection of the sediment core. Based on these four dates, mean sedimentation rates were calculated for intervening time intervals, which were then used to assign approximate deposition dates to samples from each core.

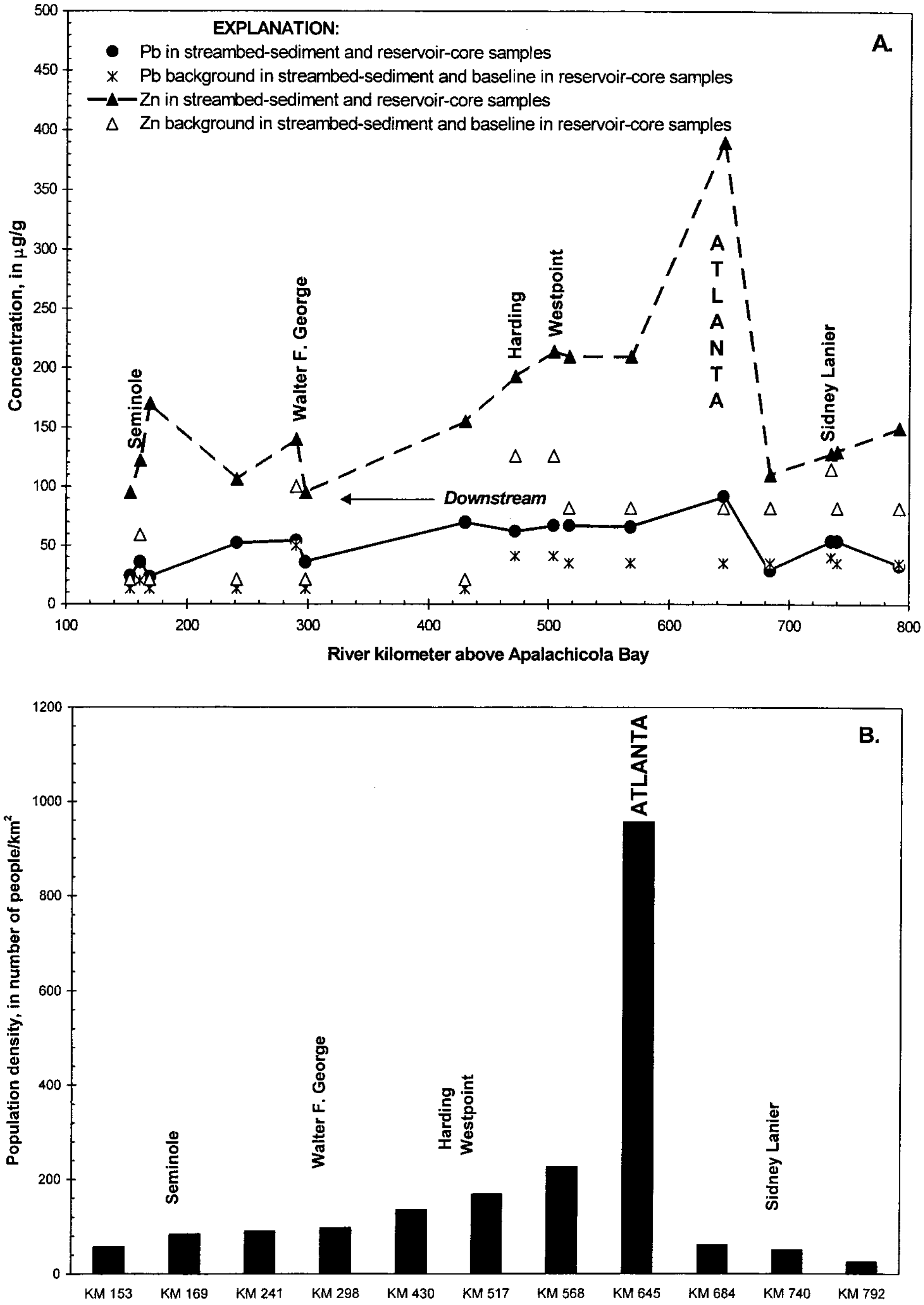

FIGURE 2. River kilometer upstream of Apalachicola Bay versus (A) total Pb and Zn concentrations in streambed sediment collected 1992–1994, reservoir sediment for the time interval 1990–1995, Pb and Zn background concentrations in streambed-sediment samples, and Pb and Zn baseline concentrations in reservoir-core samples and (B) population density of basins containing sampling sites. Population density was calculated from the 1990 census data and the watershed area.

Generally, metal concentrations that have been corrected for their *background* contribution correlate better with measures of human activity or impact (population density, traffic volume, land use, toxic release inventory, etc.) than uncorrected metal concentrations. In this study, *background* refers to metal concentrations of soil samples collected from undeveloped areas in the streambed-sediment sampling-site basins and the reservoir watersheds. Deeply weathered soils were sampled from the two major physiographic provinces in the study area: the Piedmont and the Coastal Plain. Eight and nine soil samples were collected from the Piedmont and Coastal Plain Provinces, respectively. The chemical composition of the background soils in the two provinces are substantially different (see Figure 2A) and bracket concentrations in the upper continental crust (20 ug/g Pb and 71 ug/g Zn; (*19*)). For reservoir-core sediments, the *baseline* concentration is obtained from the bottom of the core, which usually represents minimal anthropogenic activity during the early stages of reservoir development. Anthropogenic metal concentrations are calculated for sediment samples by subtracting the mean background or the baseline metal concentration from the total metal concentration.

Vehicular traffic data were obtained from the Georgia Department of Transportation (written communication, 1998) for counties that border the five reservoirs and for the Metropolitan Atlanta area. For each county, all sections of roads with average annual daily traffic greater than the 75th percentile for the county were tabulated. For those highly traveled road sections, the total average annual daily traffic was divided by the total number of miles of road to yield the traffic density, in number of vehicles per mile. Traffic density

TABLE 1. Pearson Correlation Coefficients (r Values) for Total and Anthropogenic Metal Concentrations and Population Density for Streambed-Sediment and Reservoir-Core Samples and for Total and Anthropogenic Metal Concentrations and Traffic Density for Reservoir-Core Samples[a]

	TotPb	AntPb	TotZn	AntZn	Popdn
Streambed-Sediment Samples ($n = 11$)					
TotPb	1.00				
AntPb	0.87 (0.01)	1.00			
TotZn	0.76 (0.10)	0.56 (1.00)	1.00		
AntZn	0.70 (0.25)	0.69 (0.30)	0.93 (0.00)	1.00	
Popdn	0.73 (0.15)	0.63 (0.58)	0.93 (0.00)	0.92 (0.00)	
Reservoir-Core Samples ($n = 6$)					
TotPb	1.00				
AntPb	0.79 (0.92)	1.00			
TotZn	0.94 (0.09)	0.84 (0.55)	1.00		
AntZn	0.82 (0.68)	0.92 (0.13)	0.95 (0.05)	1.00	
Popdn	0.86 (0.44)	0.73 (1.00)	0.93 (0.12)	0.87 (0.38)	1.00
Trfdn	0.64 (1.00)	0.53 (1.00)	0.80 (0.81)	0.77 (1.00)	0.93 (0.10)

[a] Level of significance shown in parentheses; TotPb, total lead concentration; AntPb, anthropogenic lead concentration; TotZn, total zinc concentration; AntZn, anthropogenic zinc concentration; Popdn, population density in the sampled basin; Trfdn, traffic density in the county upstream of the reservoir.

was calculated for the year 1995, and results were correlated with metal concentrations and population density.

The cultural enrichment factor (CEF) is used to indicate whether the entry of metal contaminants into a water body has increased in relation to an earlier period baseline or background value (*20*). As used here, the CEF is defined as the ratio of normalized (to aluminum) concentration of sediment at any depth to the normalized (to aluminum) background or baseline concentration. CEFs that are about 1 indicate no significant anthropogenic enrichment, whereas CEFs greater than 2 suggest anthropogenic contribution.

Results and Discussion

Sedimentary Pb and Zn concentrations (i.e., the metal concentration associated with solid sediment) in streambed sediment collected during 1992−1994 and in reservoir-sediment-core subsamples that correspond to the ^{137}Cs -dated time period 1990−1995 have somewhat different spatial patterns (Figure 2a). Lead concentrations are elevated (65−90 μg/g) in the Atlanta Metropolitan area but decrease to near the South Atlantic Coastal Plain background concentration of 18 μg/g (*21*) in the Lower Chattahoochee River Basin, some 300 km downstream of Atlanta. The Zn concentrations are proportionally more elevated (390 μg/g) than Pb in the Atlanta area and are substantially higher (90−170 μg/g) than the South Atlantic Coastal Plain background concentration of 35 μg/g (*21*) in the Lower Chattahoochee River Basin. The Pb concentrations do not decrease as sharply as the Zn concentrations in the urban-affected area from Atlanta to Lake Harding. Downstream of Lake Harding, the Pb concentration decreases from 65 to 20 μg/g, whereas Zn decreases by a smaller percentage, from 200 to 100 μg/g (Figure 2a). The slight increases in both Pb and Zn around Walter F. George Reservoir may be due to a smaller urban source, such as Columbus, GA and/or Phenix City, AL. We have no explanation for the increases in both Pb and Zn around Lake Seminole (Figure 2a). Both increases are substantially greater than the range of Pb and Zn concentrations in replicate samples.

One explanation for the different spatial patterns in Pb and Zn for the time period 1990−1995 is that the elevated Pb concentrations (above a background value of about 20 μg/g) originated in the Metropolitan Atlanta area, probably as street dust and urban soils with elevated Pb concentrations derived from previous use of leaded products (gasoline, paint (*22, 23*)). In contrast, there may be perpetual sources of Zn, i.e., motor-vehicle tire wear (*24*) and fossil fuel combusion (*10*). Because motor vehicles abound everywhere, there is a continual terrestrial source of Zn that could cause concentrations in sediments to be elevated, even in rural areas, above the background value, which is about 80 μg/g in the Piedmont Province and 20 μg/g in the Coastal Plain Province.

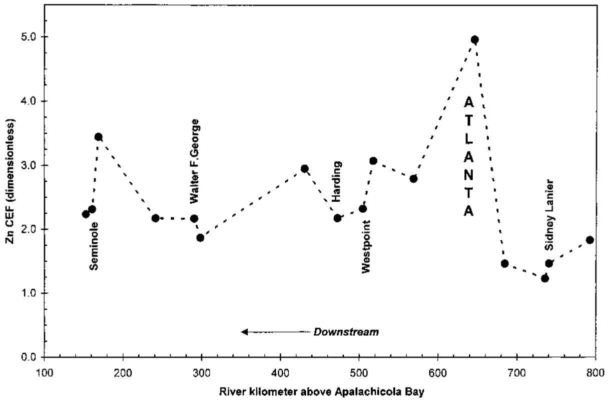

FIGURE 3. River kilometer upstream of Apalachicola Bay versus the zinc cultural enrichmentfactor (Zn CEF) for Chattahoochee River streambed-sediment and reservoir-coring sites.

For the streambed-sediment samples, total Pb and total Zn concentrations are strongly correlated with the population density of each basin (Table 1). The anthropogenic Zn concentrations vs population density also are strongly correlated, but the substantial correlation between anthropogenic Pb and population density is not significant (Table 1). Because of these correlations, the total metal concentrations, rather than the anthropogenic concentrations, are used to illustrate the urban gradient distribution (Figure 2a).

We hypothesized that the best indicator of anthropogenic activity affecting the concentration of those trace metals (Pb and Zn) that are related to motor-vehicle use would be some measure of traffic density in the basin where the streambed-sediment site or reservoir is located. Traffic-density data were obtained only for the reservoir-sediment core data set (for counties upstream of the reservoirs), because it was difficult to acquire traffic-density data for specific watersheds where streambed-sediment samples were collected. Metal-concentration and traffic-density correlations, however, are not significant (Table 1) for this small data set of five reservoirs and the Atlanta Metropolitan area. Lead and Zn concentrations are well correlated with population density ($R = 0.86$ and 0.93, respectively) for the reservoir-sediment core data set, although the correlation of Pb and population density is not significant (Table 1). The small size of the reservoir-sediment data set limits the degrees of freedom for the correlations. The metal correlations (except anthropogenic Pb) with population density in the larger streambed-sediment data set, however, are significant. Population density is strongly correlated with traffic density (Table 1) for the reservoir-sediment-core data set, and thus, we consider population density (Figure 2b) as an approximation for the quantity of automobiles that affects the watershed upstream of the sampling sites. One possible reason that anthropogenic Pb is not strongly correlated with traffic and population density is that by the mid-90s the anthropogenic Pb signal is not directly related to leaded gasoline use that has been phased out for at least 10−15 years (*25*).

Population-density data for streambed-sediment sites along the main stem of the Chattahoochee River indicate that Metropolitan Atlanta is the main anthropogenic source of Pb and Zn (Figure 2). While the decrease in population density away from the Metropolitan Atlanta area is sharp and the decline relatively smooth, the pattern for total Zn is

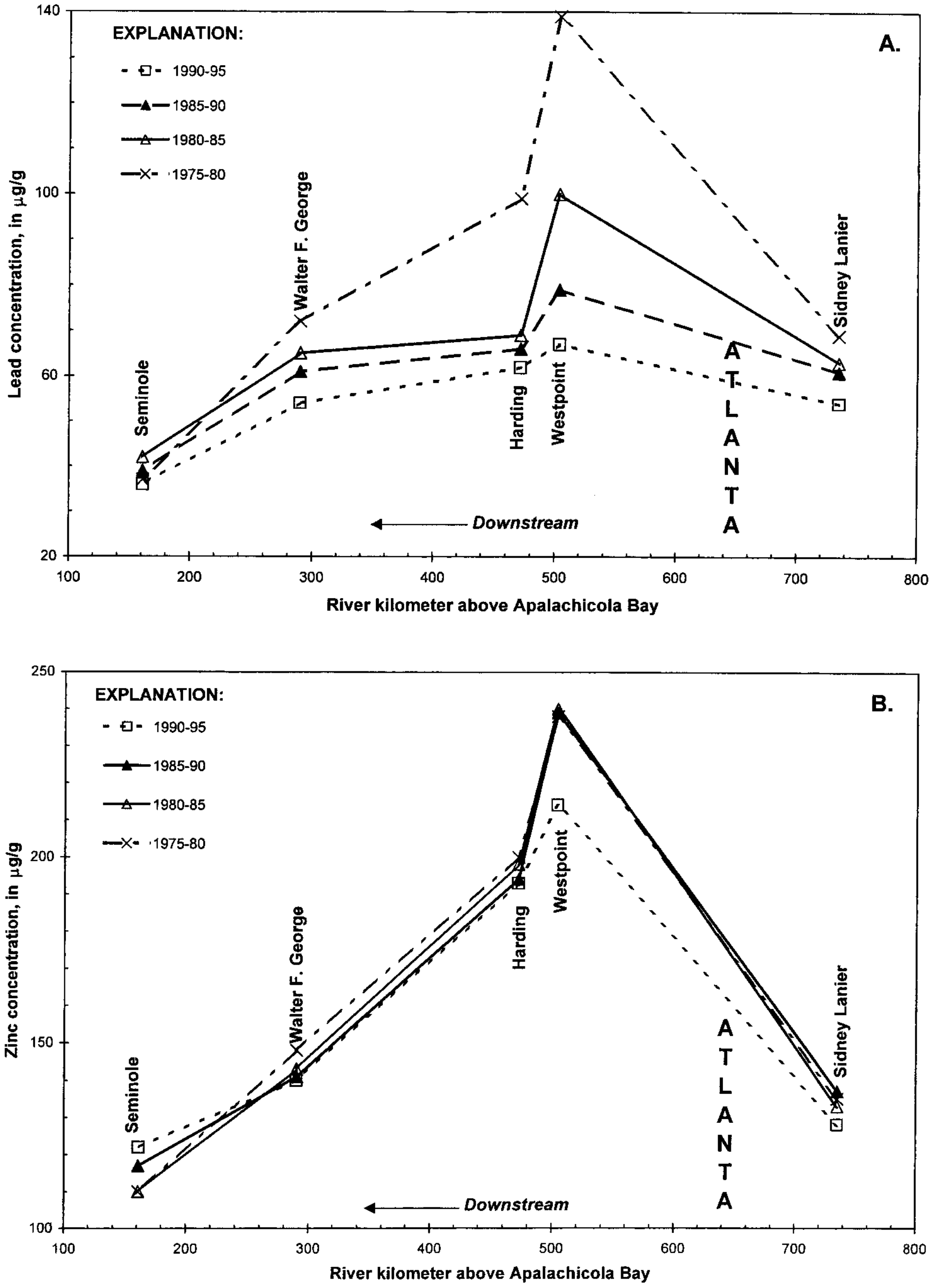

FIGURE 4. River kilometer upstream of Apalachicola Bay versus concentrations of (A) Pb and (B) Zn in dated reservoir cores from five sites along the Chattahoochee River.

somewhat more irregular than that for total Pb (Figure 2a). It should be remembered that population density is an integrated value for the basin that contains the streambed-sediment sampling site, whereas metal concentrations for specific sampling sites are not as well integrated and are subject to more variability in space and time.

Another process that may affect the spatial distribution of metal concentrations is the sediment-trapping effect that occurs along a series of reservoirs down river. This does happen to some degree in the Chattahoochee River system. For instance, Westpoint Lake was built in 1975 to reduce pollution loads from Atlanta that were being transported downstream to Lake Harding. A slight decrease in sedimentary Zn occurs from Westpoint Lake to Lake Harding (Figure 2a). Trapping of sedimentary metals along this series of reservoirs, however, is not the only process affecting the metal concentrations along the urban–suburban–rural gradient. Streambed-sediment samples in the Metropolitan Atlanta area exhibit an average Zn CEF of 5, whereas streambed-sediment sites and 1990–1995 dated reservoir sediments downstream of Atlanta have CEF values that range from 1.9 to 3.4, with a mean of 2.5 (Figure 3). This pattern suggests that dilution of contaminated sediment by the addition of uncontaminated earth materials may also be responsible for the attenuation of values away from the main contaminant source.

Like the spatial patterns, the temporal patterns of sedimentary Pb (Figure 4a) and sedimentary Zn (Figure 4b) concentrations in reservoir-sediment-core subsamples that are dated for four time intervals (1975–1980, 1980–1985, 1985–1990, 1990–1995) are somewhat different. Peak concentrations of Pb (140 µg/g) occur in Westpoint Lake sediments dated 1975–1980. In successive time intervals, the peak Pb concentrations decrease to about 65 µg/g in sediments dated 1990–1995 (Figure 4a). Westpoint Lake is the first reservoir on the Chattahoochee River downstream of Atlanta and thus intercepts the contaminant inputs from the Metropolitan Atlanta area. Sedimentary Pb concentrations in urban- and suburban-reservoir sediments nationwide peaked in the mid-1970s in response to maximum usage of leaded gasoline (*7*). After that, concentrations declined rapidly due to the removal of lead from gasoline (*25*). Atlanta has a very high population density relative to other areas in the Chattahoochee River basin (Figure 2b). Sediments in Westpoint Lake appear to have been most impacted by human activities, primarily vehicular traffic, in the Metropolitan Atlanta area. The decline in Pb concentrations in the sediment core from Westpoint Lake with time clearly documents the reduced burdens of Pb released to the environment by the removal of lead from gasoline. The farther downstream from the major contaminant source, the smaller the peak Pb concentration in the 1970s and the smaller the decreases in concentrations as time approaches the present (Figure 4a).

There is a prominent spatial peak in Zn in the 1975−1980 dated samples from Westpoint Lake downstream of Atlanta, similar to that of Pb (Figure 4b). Unlike Pb, however, the younger samples (1980−1990) have identically high concentrations of Zn (240 μg/g). Only the most recent set of samples (1990−1995) has an average Zn concentration (215 μg/g) that is slightly reduced from the maximum concentration present in the 1980−1990 interval. The temporal distribution of sedimentary Zn concentrations in dated reservoir cores downstream of Atlanta suggests that there is a significant source of this metal that persists to the present. A large number of coal-fired power plants are located throughout Georgia and Alabama (*26*). Total Zn concentrations and population density are strongly correlated, and population density and traffic density are strongly correlated (Table 1). These strong positive correlations suggest that automobiles as well as coal combustion may be significant terrestrial sources of Zn to sediments.

Automobile tires contain large quantities of ZnO that is used as an accelerator in vulcanization (*8*). Using an average Zn concentration of 0.73 wt %, a tire size of 65 cm in diameter and 15 cm wide and assuming that 1 cm of rubber is worn off in 32 000 km of driving, Christensen and Guinn (*8*) calculate that 3.0 mg of Zn are released to the road surface and the immediately adjacent areas. Thus, runoff from street, road, and highway surfaces can contribute significant quantities of Zn to the aquatic environment. Corrosion of galvanized metal and tire wear are frequently cited sources of Zn in urban runoff (*23*). Wiesner et al. (*24*) note that there is a direct correlation between particulate Zn concentrations in urban runoff from various cities around the world and fuel consumption (one measure of traffic density). They (*24*) suggest that this relationship indicates that tire wear may be the principal source of particulate Zn in urban runoff and that much of the global variability in particulate Zn concentrations might be explained by motor-vehicle use. Highway runoff is a major contributor to urban−suburban impoundments that are designed to catch stormwater runoff and trap sediments and sorbed contaminants (*27*).

Census data (*6*) indicate that the U.S. population has grown 29% during the period 1970 to 1995. National traffic-volume data (*28*) show that vehicle miles have increased 124% for the same time period. Thus, there is an ever-increasing supply of particulate Zn to pollute our waterways. Even though modern water-pollution control measures have reduced the loads of many contaminants to aquatic resources, they appear to have barely kept pace with Zn inputs that have been, in part, contributed by ever-increasing traffic volume. Also, it is unknown whether current use of Zn in the rubber manufacturing process is equivalent to past usage.

In this paper we showed that total Pb and Zn concentrations in sediments correlate equally well with population density and traffic density as do anthropogenic concentrations, eliminating the need for data manipulation of total metal concentrations. The facts that streambed sediment is sieved to less than 63-μm grain size and that reservoir sediments are generally in the silt-clay grain-size class are major factors contributing to this relationship (*29*). The differences in the spatial and temporal distributions of metal concentrations along the urban−suburban gradient in the Atlanta Metropolitan area are related to anthropogenic activities such as the use and subsequent removal of leaded gasoline and the wear of rubber tires on motor vehicles. Elevated Zn concentrations in rural settings may be related to fossil-fuel combustion, notably from power plants that dominate the energy-producing landscape in Georgia and Alabama (*26*) as well as rubber tire wear.

Acknowledgments

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Literature Cited

(1) In *The Biogeochemistry of Lead in the Environment*; Nriagu, J. O., Ed.; Elsevier: Amsterdam, 1978; Part A, pp 1−14.
(2) Forstner, U.; Wittmann, G. T. W. *Metal Pollution in the Aquatic Environment*; Springer-Verlag: New York, 1979; pp 39−42.
(3) Campbell, W. J. *Environ. Sci. Technol.* **1976**, *10*, 436.
(4) Crecelius, E. A.; Bothner, M. H.; Carpenter, R. *Environ. Sci. Technol.* **1975**, *9*, 325.
(5) *National Air Pollutant Emission Trends, 1900−1992*; EPA/454/R-933/032; Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency: Washington, DC, 1993.
(6) United States Census Bureau 1998. *U.S. Census Bureau The Official Statistics;* U.S. Bureau Census, accessed December 23, 1998@ URL http://www.census/gov/population/estimates/nation.
(7) Callender, E.; Van Metre, P. C. *Environ. Sci. Technol.* **1997**, *31*, 424A.
(8) Christensen, E. R.; Guinn, V. P. *Environ. Engineer. Div., EE1* **1979**, *105*, 165.
(9) Thornton, I. *Metals in the Global Environment: Facts and Misconceptions*; Int'l Council Metals Environment: Ottawa, 1995; p 35.
(10) In *Zinc in the Environment*; Nriagu, J. O., Ed.; Wiley: New York, 1980; Part 1, pp 113−159.
(11) Leahy, P. P. et al. *U.S. Geol. Surv. Open-File Report* **1990**, *90−174*, 10.
(12) Frick, E. A. et al. *U.S. Geol. Surv. Water-Res. Invest. Report* **1996**, *96−4101*, 120.
(13) Couch, C. A. et al. *U.S. Geol. Surv. Water-Res. Invest. Report* **1996**, *95−4678*, 58.
(14) Shelton, L. R.; Capel, P. D. *U.S. Geol. Surv. Open-File Report* **1994**, *94−458*, 20.
(15) Van Metre, P. C.; Callender, E. *J. Paleolimnol.* **1997**, *17*, 239.
(16) Arbogast, B. F. (Ed.) *U.S. Geol. Surv. Open-File Report* **1990**, *90−668*, 311.
(17) Aruscavage, P. J.; Crock, J. G. In *Methods for Geochemical Analysis*; Baedecker, P. A., Ed.; *U.S. Geol. Surv. Bull.* 1987; Vol. 1770, pp Cl−C6.
(18) Van Metre, P. C. et al. *Environ. Sci. Technol.* **1997**, *31*, 2339.
(19) Taylor, S. R.; McClennan, S. M. *The Continental Crust: its Composition and Evolution*; Blackwell: London, 1985; p 46.
(20) Heit, M. et al. *Water, Air, Soil Pollut.* **1981**, *15*, 441.
(21) Shacklette, H. T.; Boerngen, J. G. *U.S. Geol. Surv. Professional Paper* **1984**, *1270*, 105.
(22) Thornton, I. *Palaoegeo. Palaeoclim. Palaeoecol.* **1990**, *82*, 121.
(23) Mielke, H. W. *Appl. Geochem., Suppl. Issue* **1993**, *1*, 257.
(24) Wiesner, M. R. et al. In *Metals in Surface Waters*; Allen, H. E., Garrison, A. W., Luther, G. W., Eds.; Ann Arbor Press: Chelsea, MI, 1998; pp 2−35.

(25) United States Environmental Protection Agency. *EPA J.* **1995**, *21*, 33.
(26) Georgia Power, accessed October 14, 1999 at URL http://www.georgiapower.com/newsroom/plants.asp.
(27) Yousef, Y. A. et al. *Sci. Tot. Environ.* **1990**, *93*, 433.
(28) *Traffic Volume Trends, May 1997*; FHWA-PL-97−004; Federal Highway Administration, U.S. Department of Transportation: Washington, DC, 1997.
(29) Rice, K. C. *Environ. Sci. Technol.* **1999**, *33*, 3, 2449.

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