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Lead Concentrations in Fish and River Sediments in the Old Lead Belt of Missouri

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Although active mining activities in the Old Lead Belt of Missouri ceased in 1968, old tailings piles remain and continue to impact the lead (Pb) concentrations in sediments and fish in the Big River and Flat River Creek in Missouri. A 3-year study was conducted that examined the Pb concentrations in organic-rich sediments, fish fillets, and fish whole bodies at 13 sites in these two rivers. The results showed that the sediments had significantly elevated Pb concentrations near and beyond the tailings piles compared with control sites upstream from the mining activity. The results also showed the Pb concentration in sediments correlated well with Pb concentrations in suckers ($r = 0.86$, $p < 0.0001$) and small sunfish ($r = 0.82$, $p < 0.0001$). Annual average Pb concentrations in sucker fillets ranged up to 0.67 μg/g (wet weight basis) and exceeded the World Health Organization guidelines of 0.3 μg/g (wet weight basis) at many of the sample sites. Fillets of bass, which feed at a higher trophic level, had much lower Pb concentrations than either suckers or small sunfish, and no bass fillet samples exceeded the WHO guideline. Whole body Pb concentrations in small sunfish were also determined and correlated strongly ($r = 0.92$, $p = 0.0004$) with sediment concentrations before leveling at a maximum of approximately 20 μg/g (wet weight basis).

Introduction

Elevated lead concentrations have been observed in river sediments and fish due to erosion and runoff from surface ore deposits and tailings piles left over from previous mining activities which ceased in the Old Lead Belt of Missouri in 1968. In 1980 the Missouri Department of Conservation (MDC) presented preliminary data indicating accumulation of lead in edible tissues of black redhorse suckers (*Moxostoma duquesnei*) and longear sunfish (*Lepomis megalotis*) collected at several sites along the Big River downstream of a large tailings pile located near Desloge, MO (*1*). The Desloge tailings pile, located in a large horseshoe bend of the Big River (Figure 1), had a history of serious erosion which had resulted in the release of large quantities of fine tailings material into the adjacent river. Eroded tailings essentially covered the river bottom for a considerable distance downstream. Subsequent studies by Gale et al. (*3, 4, 6−9*), Wixson et al. (*2, 5*), and Schmitt and Finger (*10*) provided additional data regarding the extent of lead contamination of fish, mollusks, and sediments taken from a larger number of sites along the Big River and two tributaries, Eaton Creek and Flat River Creek. In general, these reports concluded that bottom-feeding suckers and small sunfish collected at many of these sites were found to contain elevated concentrations of Pb in various tissues, especially bone, scales, and intestinal contents, as compared with similar specimens taken from control sites outside the old mining district.

Public health concerns have focused on the edible fish fillets if they exceed the World Health Organization (WHO) guideline of 0.3 μg/g (ppm; wet weight basis) and a maximum daily dietary intake of 450 μg (*11*). While the United States has no legislated standard for Pb in fish that might be consumed as food, the U.S. Food and Drug Administration recently issued a guidance/action/tolerance concentration for lead in fish at 1.3 μg/g (*12*). The WHO guideline is currently recognized by the MDC as it issues advisory warnings regarding the consumption of certain types of fish taken from various contaminated streams, including the Big River and its tributaries in Missouri's Old Lead Belt. The legislated limit for Pb in commercial food in the United Kingdom (UK) is 1 μg/g (*13*). A person who consumes one pound of food containing 1 μg/g Pb would ingest 454 μg of Pb, approximately equal to the WHO maximum recommended daily intake.

The environmental nuisance created by erosion of vast quantities of tailings materials and vagrant heavy metals from abandoned industrial sites in Missouri's Old Lead Belt has been well documented in the scientific literature and widely recognized as a serious problem. Based upon available data and growing environmental awareness and concern, in October, 1992, the Old Lead Belt of Missouri was added to the list of "Superfund" sites identified by the USEPA. In July, 1994, in an agreement with the Missouri Department of Natural Resources (MDNR), the Doe Run Company initiated plans to remediate several abandoned industrial sites identified as major sources of environmental contamination. In October, 1995, remediation work began on the Desloge tailings pile, to regrade, install coarse rock cover and ripwrap and revegetate the area in an attempt to stabilize the tailings material and prevent continued erosion into the Big River. As part of this agreement, the Doe Run Company agreed to perform periodic analyses of the river sediments and metal concentrations in selected fish species within affected regions of the Big River and Flat River Creek. This paper presents the results of that study.

Reported aqueous-phase Pb concentrations in 0.45-μm glass-fiber filtered water samples taken from the Big River in 1982 ranged from 5 to 54 μg/L, with most samples falling below 30 μg/L (*3*). Pb instead tends to associate primarily with clay minerals, Mn oxides, Fe and Al hydroxides, and decomposing organic matter (*14*). In high alkalinity waters such as the Old Lead Belt, Pb may also become concentrated in $CaCO_3$ particles or as phosphates, especially under alkaline conditions (*14*). Elevated Pb concentrations in the sediments are accessible to members of aquatic food webs, including various fish consumed by local fishermen. This provides a direct link between the particulate lead and the fish within the river ecosystem.

The objectives of this study were to determine the absolute concentrations of and the relationship between Pb concentrations in sediments and selected fish species upstream,

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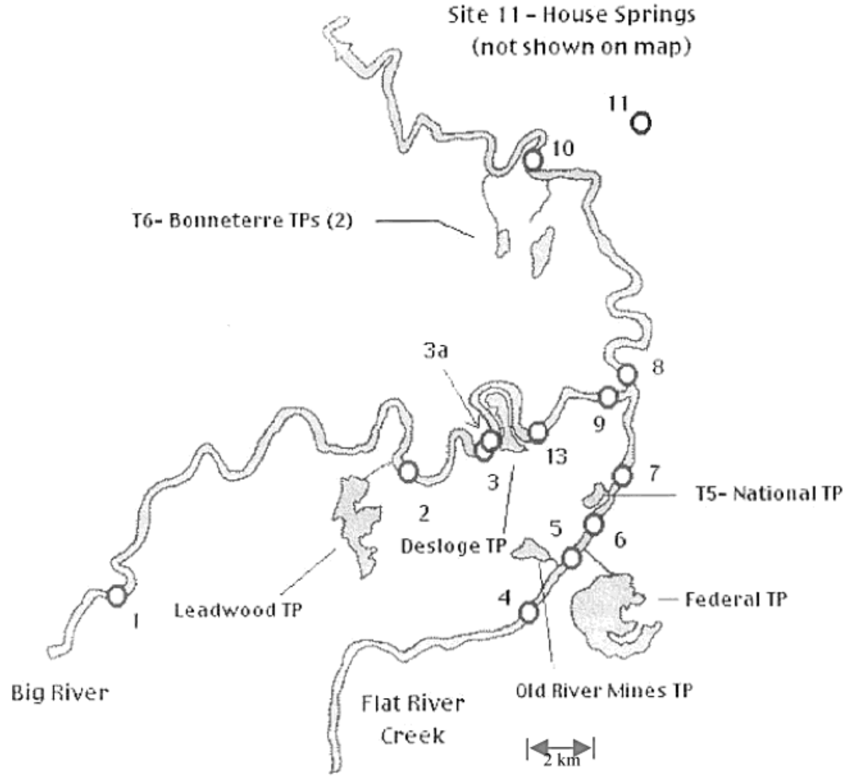

FIGURE 1. Sample site locations on Big River and Flat River Creek showing locations of major tailings piles in area.

adjacent to, and downstream of the abandoned industrial sites on the Big River and Flat River Creek in Missouri's Old Lead Belt. To accomplish this goal, sediment, fish tissue, and whole fish samples were collected at specific sites within the Old Lead Belt over a 3-year period. The sites, sampling methodology, and results are discussed below.

Site Description

Fish and sediment samples were collected from 13 sites on the Big River and Flat River Creek in the Old Lead Belt of Missouri near Desloge, MO (Figure 1). Sites BR1 and FR4 (Big River Site 1 and Flat River Site 4, respectively) were upstream of any tailings piles and served as control sites on the Big River and Flat River Creek, respectively (Figure 1). Site BR11 was approximately 130 km downstream from the nearest tailings pile and was immediately below a concrete spillway which blocked upstream migration of fish. The remaining sites were within the region directly affected by lead mining and milling (Figure 1). Additionally, it should be noted that treated effluent from the Park Hills Sewage Treatment Plant discharges immediately upstream of Site BR9 on the Big River.

The Big River is the dominant river hydraulically with an annual average flow of approximately 5.7 m^3/s at sites BR1 to BR 10, with reported ranges of approximately 0.9 to 230 m^3/s during the late summer months. Flat River Creek is a small tributary with typical flow rates of approximately 0.06 to 14 m^3/s and recorded monthly means as high as 0.6 m^3/s during the summer months (*15*).

Methods

Fish Sampling. Attempts were made each year to collect five specimens of each of three groups of fish from each site for the fillet analyses. Group A included small sunfish with longear sunfish (*Lepomis megalotis*) being preferred. Group B included bottom-feeding suckers with the black redhorse sucker (*Moxostoma duquesnei*), northern hog-sucker (*Hypentelium nigricans*), and golden redhorse sucker

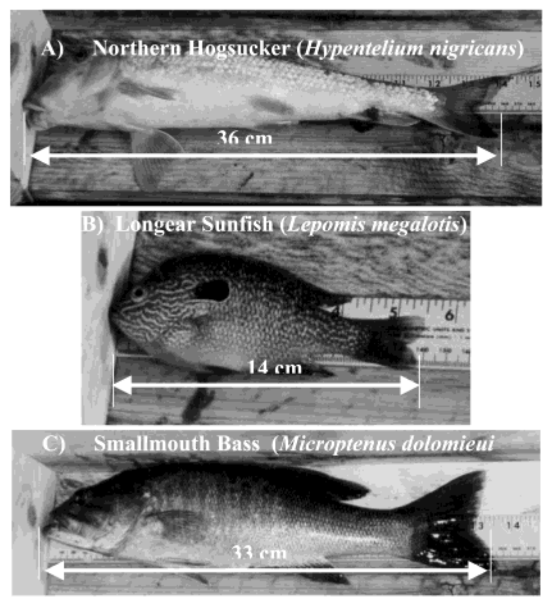

FIGURE 2. Specimens of primary species samples in study: (A) Sucker (Northern Hogsucker (*Hypentelium nigricans*)); (B) Sunfish (Longear Sunfish; *Lepomis megalotis*)); and (C) Bass (Smallmouth Bass (*Micropterus dolomieui*)).

(*Moxostoma erythrurum*), being preferred. Group C included top-feeding bass, with largemouth bass (*Micropterus salmoides*), and smallmouth bass (*Micropterus dolomieui*) being preferred. The preferred species from each group are shown in Figure 2. The total number of fillet samples collected in 1998, 1999, and 2000 were 193, 290, and 189, respectively.

In addition to fillets, five whole body specimens of Group A (small sunfish) were also collected for analysis from each sites in 1999. Whole body analyses were repeated on specimens taken from the Big River control site (BR1) and at Site BR3A at the base of the Desloge tailings pile in 2000.

These samples were important because while only fish fillets are consumed by humans, it is the whole bodies that are consumed by aquatic and terrestrial predators and, hence, contaminants may move up the food chain. Also at sites BR1 and BR3A in year 2000, five additional sunfish specimens were eviscerated, and the carcasses and internal organs were pooled separately to form two samples for analysis from each site. The purpose of these eviscerated samples was to begin to assess how much of the whole body metal load might be due to the presence of ingested sediments. All specimens were stored in ice chests and placed in chain-of-custody control until analyzed for Pb concentrations.

Fish were collected by electrofishing as authorized by the MDC. At very shallow sites on the Flat River Creek, a backpack-mounted electroshock unit was utilized to stun the fish (Coffelt Model BP-1C0). At all other sites, a larger Smith-Root Model 5 GPP 5000-W generator and shock box with accompanying Model SR-6 electrofishing tote barge was used.

Specimens were initially placed alive in clean polyethylene holding tubs and taken to a processing area established on the river's edge at each site. The same personnel conducted the same tasks at each site to reduce the chance for systematic bias in the fish processing. Live fish were categorized and segregated, measured, and weighed using a calibrated, portable, battery-operated electronic balance. Individual specimens were sacrificed by severing the spinal cord near the operculum (gill cover) with a stainless steel knife, scaled with a fishermen's scaler, and rinsed thoroughly in flowing river water upstream of disturbed sediments. Special care was always taken to remove all scales and to prevent contamination of a fish sample with sediment, fecal material, scales, or cross contamination by other samples. Each fish was filleted, using a stainless steel knife on a plastic fillet board equipped with a stainless steel clamp to secure the head of the specimen. Fillets were rinsed thoroughly in river water and double bagged in separate new plastic bags, sealed, and labeled with a unique identifying number. All materials used in the fish collection and processing procedures were carefully chosen and maintained to prevent contamination by heavy metals. Immediately upon return to the laboratory, all fish fillets, whole bodies, and eviscerated specimens were frozen prior to shipment to the analytical laboratory.

Sediment Sampling. During the second and third years of the study, two or three sediment samples were collected at each of the study sites. The samples were collected in a manner to selectively collect the organic-rich sediment near the surface of the riverbed. At each site, composite samples were obtained in a sedimentation zone (SED), a riffle zone (RIF), and at random locations along the riverbed (RAN). The samples were collected using a 0.5-L polypropylene (PP) container to scoop up grab samples of benthic sediments to a depth of 2−4 cm. The plastic container was covered with a lid and shaken vigorously. The liquid and light suspended particles were then decanted into a separate PP container. The procedure was repeated several times until a sample containing sufficient gravel-free sediment was achieved. After settling for 10−20 min, the solution above the composite sediment sample was carefully decanted off. The sediment sample in its closed plastic container was then refrigerated until processing in the laboratory. Upon return to the laboratory, each sediment sample was centrifuged at low speed for 2 min in PP centrifuge vials to dewater and concentrate the samples. The samples were then frozen and transferred to the analytical lab (along with the fish samples) for digestion and analysis.

Metal Analysis Procedure. Fish and sediment samples were handled in a strict chain-of-custody procedure at all times. Digestion and analysis of samples was performed by the Environmental Trace Substances Laboratory (UM−Rolla) in year 1. Frozen scaled fillets were ground using a blender equipped with stainless steel blades. Once ground, samples were weighed and wet weights were recorded. Samples were then refrozen and lyophilized. Thoroughly dried samples were weighed to determine moisture content and then homogenized in a blender with stainless steel blades. Aliquots of the dry samples were digested in nitric acid prior to analysis using a Perkin-Elmer Elan 5000 Induction Coupled Argon Plasma-Mass Spectrometer (ICP/MS).

In years 2 and 3, digestion and analysis of fish and sediment samples was performed by Midwest Research Institute (MRI, Palm Bay, FL). Digestion/preparation procedures for fish samples were performed according to EPA Method 200.11. This method is an alkaline hydrolysis with tetramethylammonium hydroxide (TMAH), followed by pH adjustment with nitric acid. Wet tissue is dissolved with an equal volume of TMAH at 60 °C overnight, acidified with nitric acid, and diluted 10:1 prior to analysis. Determination of metals in fish tissue was done by ICP/MS using EPA Method 200.8 (*Determination of Trace Elements in Waters and Wastes by ICP/MS*).

Digestion/preparation procedures for sediment samples were performed by MRI according to EPA Method 200.2 (*Sample Preparation for Spectrochemical Determination of Total Recoverable Elements*). This method is a hot acid leach (95 °C) with a mixture of nitric (50% v/v) and hydrochloric acids (50% and 20% v/v). The method is not a total digestion, and a significant portion of the solids in the sample remains undissolved. This digestion is intended to release the loosely bound elements from the sample matrix, as might occur in the digestive system of vertebrates. Because of the varying amounts of water in the sediment samples as delivered, all samples were dried at 60 °C before processing.

ICP/MS analysis of digested samples was performed using EPA Method 200.8 at MRI. The analysis utilized a Perkin-Elmer PE-Sciex ELAN-5000 ICP/MS with concentric nebulizer and operated in peak hopping mode using ^{72}Ge, ^{115}In, and ^{165}Ho as internal standards. Each sample was analyzed in triplicate. The method detection limit (MDL) for lead was 0.01 μg Pb/g of tissue (wet weight basis) as defined by the USEPA in 40 CFR Part 136 Appendix B.

Alkalinity and Hardness. Alkalinity was measured by acid titration to the 4.5 pH endpoint (Hach Method 8203). Calcium and total hardness were determined using the EDTA titration method (Hach Method 8204 and 8213, respectively). All values for alkalinity and hardness are reported as "mg/L as $CaCO_3$".

Results and Discussion

Lead in Water and Sediments. It is appropriate to first consider the Pb in sediments because it is hypothesized that much of the total body burdens of Pb in fish may be due to contact with or ingestion of fine Pb-laden organic-rich benthic sediments or other contaminated fish lower in the food chain. Because the solubility product constant (K_{sp}) for lead carbonate is 3.3(10^{-14}) (*16*), at equilibrium lead has a very low solubility (<2 μg/L) due to complexation with carbonates in the alkaline waters of the region. Greater concentrations have been measured, however, suggesting nonequilibrium conditions may exist at times (*17*). This carbonated-buffered chemistry may be typical of what is expected for many of the streams draining or adjacent to Mississippi Valley-type ore deposits such as the Old and New Lead Belts of Missouri; the tristate district of Missouri, Oklahoma, and Kansas; the Galena District of Illinois; and east Tennessee. These types of systems are clearly differentiated from iron-sulfide-dominated systems of Colorado and other Western states subject to acid mine drainage which may lead to iron floc-laden streams.

The mean pH, alkalinity, calcium hardness, and total hardness for the Big River were 7.8, 190, 120, and 220,

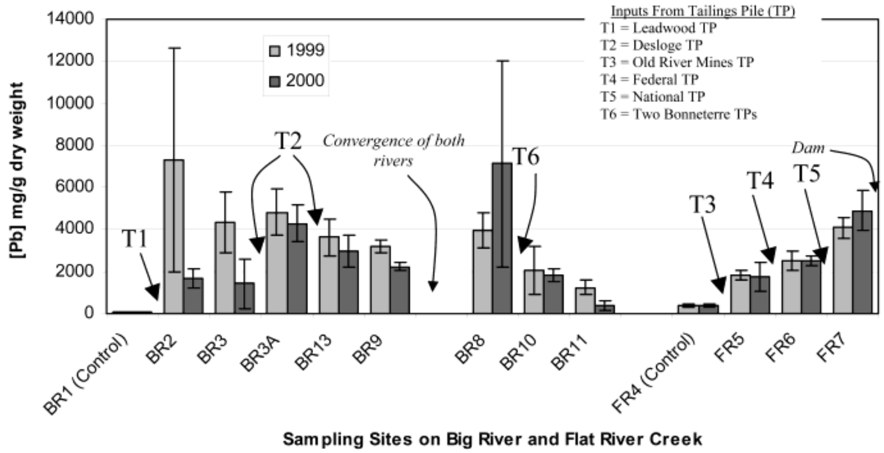

FIGURE 3. Average lead concentrations in sediments from each study site on Big River (BR) and Flat River Creek (FR) for Years 1999 and 2000. Sample sites are in order of occurrence. Location of runoff from major tailings piles in the area are indicated.

respectively. For the Flat River Creek, these values were 8.0, 200, 195, and 360, respectively. Below the confluence of the two rivers these values were 7.9, 170, 105, and 210, respectively.

Single-factor ANOVA of the sediment data showed that there were no significant differences between means of the three separate sediment samples (SED, RIF, RAN) collected at each site in either 1999 or 2000 ($\alpha = 0.05$). The mean sediment concentration (as μg/g dry weight) at each site is plotted in Figure 3 for both 1999 and 2000 showing the input locations for the major tailings piles in the region. The error bars in Figure 3 correspond to 95% confidence intervals based on pooled SED, RIF, and RAN sediment samples each analyzed in triplicate. There were no significant differences between Pb concentrations for the 2 years at most sites.

The data show that at control sites BR1 and FR4 on the Big River and Flat River Creek, the average Pb concentrations in the sediment samples were 72 and 400 μg/g (dry weight basis), respectively (Figure 3). Both of these Pb concentrations exceed the "Effects Range-Low" guideline of 47 μg/g (dry weight basis) suggested by the USEPA in its 1997 report to Congress on the Incidence and Severity of Sediment Contamination in Surface Waters of the United States (*18*). The Pb concentration in the sediment at the control site on Flat River Creek also exceeded the "Effects Range-Median" guideline of 218 μg/g (dry weight basis) suggested by the USEPA (*18*). It is unknown whether the slightly elevated concentrations of lead at control sites upstream of tailings piles is related to historic mining activities or to the natural geology of the region. While the Pb concentrations observed in sediment samples collected at the control sites exceeded the aforementioned guidelines, lead concentrations in sediments in the vicinity of and below the tailings piles were significantly larger.

The greatest Pb concentrations observed among the various sites were at sites BR2 (below the Leadwood tailings pile) and BR8 (below the convergence of the Big River and Flat River Creek). The maximum individual sediment concentrations observed were 10 550 and 12 400 μg/g (dry weight basis), at Sites BR2 and BR8, respectively. Variability among the three sediment samples taken at both these sites also lead to larger confidence intervals than for other sites.

On both rivers, at sites downstream of tailings piles, the concentrations of lead in the sediments increased significantly (Figure 3). For the Big River, the mean Pb concentrations in the fine sediments appear to peak near the Desloge Tailings pile (BR3a) and then again immediately after convergence with Flat River Creek. The average sediment concentrations tended to then decrease to 805 μg/g at Rockport Public Access near House Springs (site BR11) 130 km downstream from the convergence of the two streams. This value is still 11 times the sediment Pb concentrations in the Big River control site. These data indicate considerable horizontal transport from the industrially affected regions of the Big River and Flat River Creek. Future studies are needed to determine Pb concentration depth profiles in sedimentation zones of the Big River from point sources in the Old Lead Belt to the point of convergence with the Meramec River approximately 150 km downstream near Eureka, MO.

It is hypothesized that heavily contaminated sediments could provide a significant dietary and contact exposure to bottom-feeding aquatic organisms. Data supporting this hypothesis are presented below.

Lead in Fish. The concentration of Pb in fish fillets is of specific concern because the fillet is the portion of the fish that is typically consumed by humans. Over the 3 years of the study, the range of Pb concentrations observed in suckers, sunfish, and bass fillets was <0.002−1.07, 0.003−0.960, and <0.002−0.185 μg/g, respectively. The mean Pb concentrations in fish fillets for 1998, 1999, and 2000 in suckers, small sunfish, and bass are presented in Figures 4−6, respectively.

In sucker fillets, there were few significant differences between years for Pb concentrations in fillets at any given site (Figure 4). There were, however, significantly higher Pb concentrations at all industrially affected sites than at the control site on the Big River (BR1) (Figure 4). Of the total 239 sucker fillets collected from industrially affected sites and analyzed over the 3-year study, 133 (56%) exceeded the WHO guideline of 0.3 μg/g (*11*). Two (<1%) exceeded the UK limit of 1.0 μg/g (*13*), and none exceeded the U.S. Food and Drug Administration guidance concentration of 1.3 μg/g (*12*). All sites except the control site BR1 had at least one sucker fillet that exceeded the WHO guideline. At nine of the total 13 sites (69%), the mean Pb concentration in sucker fillets exceeded the WHO guideline in at least 1 year (Figure 4).

In sunfish fillets, there was no significant trend for Pb over the 3-year period at any specific site although some significant differences ($\alpha = 0.05$) were observed (Figure 5). At both control sites, samples collected in 1999 had higher Pb concentrations than those in 1998 and 2000, though the difference was not significant at site BR1. The results show that there were greater Pb concentrations observed in sunfish fillets at all industrially affected sites compared with control sites, though the differences were not always significant. The sunfish fillet Pb concentrations in each year generally increased and then decreased in a pattern very similar to that shown by sucker fillets, except that the average concentrations were on the order of half as great in the sunfish (Figures 4 and 5). Fillets of sunfish taken from Sites BR3A

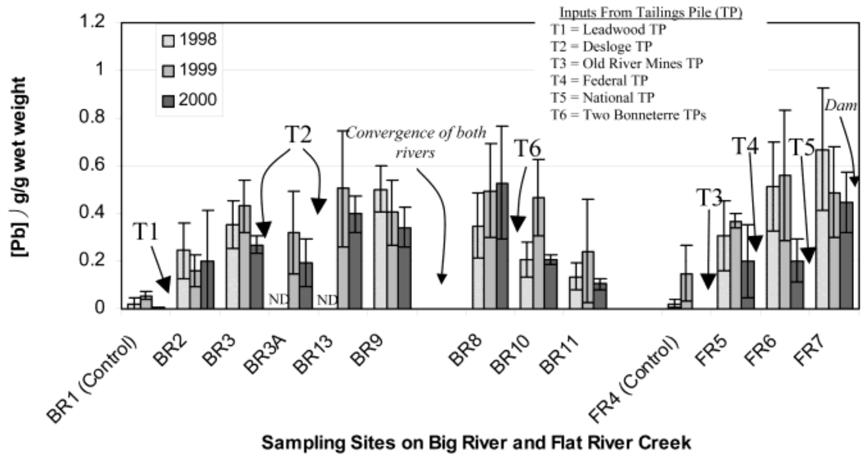

FIGURE 4. Average lead concentrations in sucker fillets from each study site on Big River (BR) and Flat River Creek (FR) for years 1998−2000. (ND = no data).

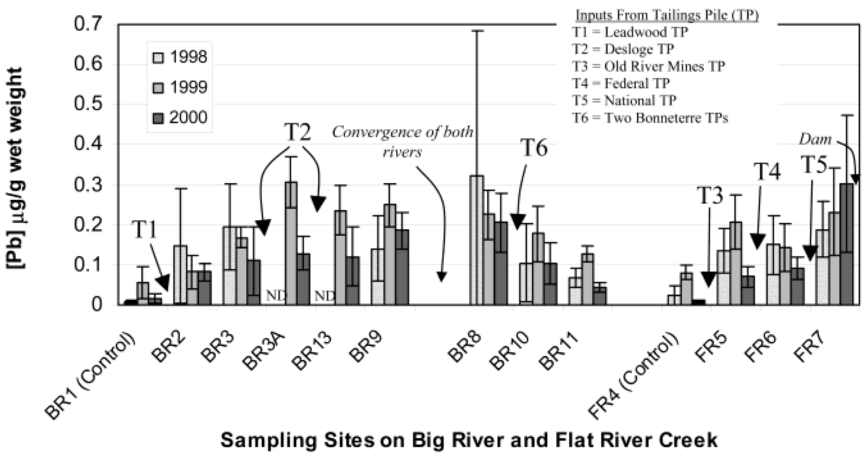

FIGURE 5. Average lead concentrations in sunfish fillets from each study site on Big River (BR) and Flat River Creek (FR) for years 1998−2000. (ND = no data).

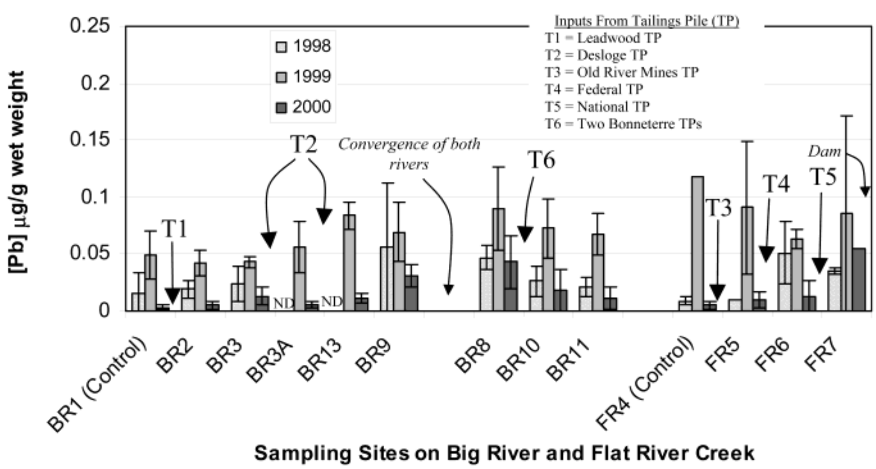

FIGURE 6. Average lead concentrations in bass fillets from each study site on Big River (BR) and Flat River Creek (FR) for years 1998−2000. (ND = no data).

and BR8 in 1998 and from Site FR7 in 2000 showed means slightly above the WHO guideline (Figure 5).

In bass fillets, Pb concentrations were much lower than those found in small sunfish or suckers. Even at sites with higher concentrations of Pb in sediment, sucker fillets, and sunfish fillets, the average bass fillet Pb concentration never exceeded the WHO guideline of 0.3 μg/g in any of the 3 years. The highest individual Pb concentration in a bass fillet was 0.185 μg/g from site BR9 in 1998.

Sediment versus Fish. Figure 7 shows the Pb concentrations in sediments versus Pb in fish fillets for 1999 and 2000. There was significant correlation between Pb concentrations in the sediment (to 4000 μg/g Pb) and Pb concentrations in fish tissue of both bottom-feeding suckers ($r = 0.86$, $p < 0.0001$) and sunfish ($r = 0.82$, $p < 0.0001$) (Figure 7). This is consistent with the known feeding habits of these fish. The data show that Pb concentrations in sucker fillets increase much more sharply with sediment Pb concentration than

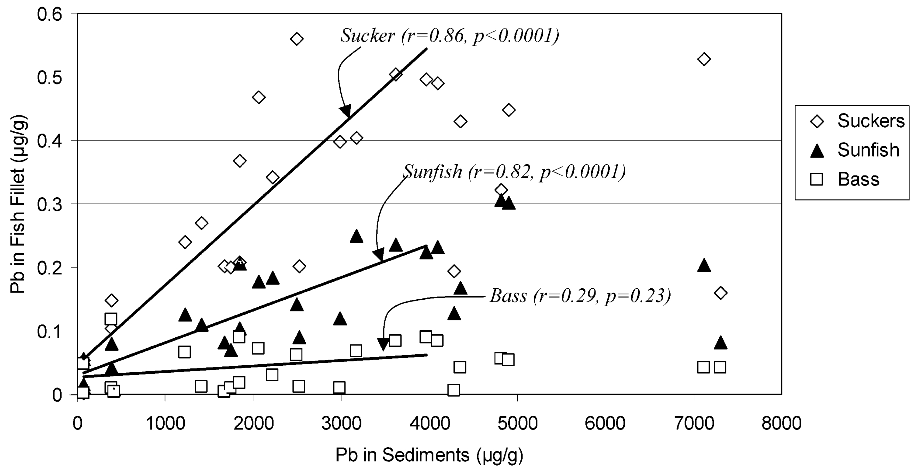

FIGURE 7. Correlation between Pb concentrations in sediment versus fish fillets from all sites for years 1999 and 2000.

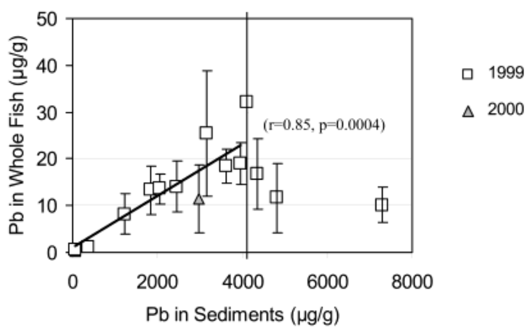

FIGURE 8. Correlation between Pb concentrations in sediment versus whole fish (sunfish) from all sites for years 1999 and 2000. (Error bars represent 95-confidence intervals based on five replicate whole body specimens.)

sunfish (Figure 7) due probably to their more exclusive bottom-feeding nature. There was not significant correlation between Pb in sediment and in bass fillets ($r = 0.29$, $p = 0.23$) (Figure 7) due potentially to their higher level in the food chain.

Figure 8 shows the relationship between Pb concentrations in sediments and Pb in whole-body sunfish. There was a significant correlation between whole body burden of Pb in small sunfish and the Pb concentration of the sediments ($r = 0.92$, $p = 0.0004$). At higher sediment concentrations (>4000 μg/g Pb), however, the Pb concentrations did not increase further. The reason for this is not certain.

As discussed previously, five sunfish were eviscerated at both BR1 (control) and BR3a near the Desloge Tailings Pile. The pooled internal abdominal organs and carcasses had mean Pb concentrations of 0.795 and 0.217 μg/g, respectively, at BR1 (control) and 63.4 and 12.2 μg/g, respectively, at BR3a. These preliminary data clearly suggest that much of the body burden of Pb may be associated with the abdominal viscera, possibly the intestines resulting from recently ingested sediments. Further investigation is needed to verify this hypothesis.

The results of this study have shown that there still exists a significant impact of the historical Pb mining activities in the Old Lead Belt in Missouri on Pb concentrations in river sediments and fish in the Big River and Flat River Creek. Average Pb concentrations in suckers and sunfish exceeded WHO guidelines (0.3 μg/L wet basis) at certain sites along these rivers. Concentrations of Pb in fish appear to be strongly correlated with the concentrations in the organic-rich sediments for species which feed primarily (suckers) or partially (sunfish) off the river bottom. The impact of sediment Pb concentrations on bass is much more limited due probably to their higher level in the food chain. More study is needed on the fate and impacts of Pb within the food chain in the rivers in historic and active Pb mining districts. Assessment of the long-term effectiveness of ongoing remediation efforts in Missouri's Old Lead Belt will require continued surveillance of Pb concentrations in the sediments and aquatic organisms of the Big River and Flat River Creek.

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