THE ENVIRONMENTAL SIGNIFICANCE OF TOXIC METALS FOUND IN FISH COLLECTED THROUGHOUT THE COALFIELDS OF PENNSYLVANIA[1]

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Abstract. Fish were collected from seven streams located within the Pennsylvania bituminous coal region to determine whether levels of toxic metals which were presently exempt from regulation could be accumulating in fish and posing a potential threat to the environment and its users. Station locations were chosen based on a review of historical information and were sampled directly downstream of National Pollutant Discharge Elimination Systems (NPDES) permitted discharge points from various active mining and processing facilities. Fish were collected by electrofishing and attempts were made to collect both adult predator species as well as bottom feeders. Whenever gamefish were included in a sample, both fillets and whole fish were submitted for analysis. Samples were analyzed for 14 selected metals based upon their suspected occurrence and 26 organochlorine pesticides and polychlorinated biphenyls (PCBs). Sampling was conducted as part of the U.S. Fish and Wildlife Service's (FWS) Environmental Contaminants Program and results for seven elements (As, Cd, Cu, Hg, Pb, Se and Zn) were compared to national background levels from a database of 112 monitoring stations described in the U.S. FWS's National Contaminant Biomonitoring Program (NCBP). Concentrations meeting or exceeding the 85th percentile for cadmium, copper, mercury and selenium were recorded for five of the seven sites sampled. Data for eleven priority pollutants listed by the Environmental Protection Agency were also compared to human health criteria for protection against consuming contaminated fish tissue. However, sufficient populations of gamefish were not sampled at any site exceeding 85th percentile concentrations to illicit public health concerns. Comparing data

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on copper and iron for organoleptic effects (avoiding consumption because of poor taste or odor), brook trout and rock bass whole specimen samples for two different sites exceeded the recommended 1 mg/L copper level and edible portions of gamefish from four sites exceeded the 1 to 2 mg/L iron level. The analytical results from the pesticide and PCB testing did not indicate any unusual problems. Samples from large developed watersheds showed signs of chlordane, DDT, dieldrin and their derivatives. Since this study was an initial screening of a small subsample of NPDES permitted minesite discharges and did reveal a potential environmental problem, future monitoring, provided available funding, will be attempted upstream and downstream of certain locations. This effort should verify the actual origin of the contaminants or document whether the problem is from another source further upstream.

INTRODUCTION

Toxic pollutant control became a national issue in 1976 when the Natural Resources Defense Council, Inc. sued the U.S. Environmental Protection Agency (EPA) for failure to control toxic pollutants as required by Section 307(a) of the 1972 Federal Water Pollution Control Act Amendments. A settlement agreement was executed and EPA susbequently established a list of 65 classes of toxic chemicals encompassing 129 compounds (priority pollutants). The court settlement also caused the Clean Water Act of 1977 to be signed into law with a major emphasis on toxic pollution control. Since publication of the original priority pollutant list, EPA has been compiling the most up-to-date scientific information on selected toxic compounds which are suspected of being a threat to public health or the environment. EPA periodically releases this information in the form of ambient water quality criteria. A summary of 64 aquatic-based and human health-based criteria documents was published in 1980 (U.S. EPA 1980). The most recent EPA criteria document, the "Gold Book", was released as a subscription service which automatically updates the user as new criteria documents become available (U.S. EPA 1986).

The 1976 settlement agreement also required EPA to develop Best Available Technology Economically Achievable (BAT) effluent limitation guidelines, pretreatment standards, and new source performance standards for the 65 priority pollutants and classes of pollutants for 21 major industries. The coal industry was included in this group since relatively little was known about toxic pollutants as constituents of coal mining-related wastewaters. EPA conducted a comprehensive study to determine the status of toxic pollutants in coal mining wastewaters. This study involved identification of raw waste and treated effluent characteristics, including: 1) the sources and volume of water used, the processes applied, and the sources of pollutants in the plant; and 2) the constituents of the wastewaters, including 129 toxic pollutants, of which 13 were toxic metals (U.S. EPA 1981). EPA concluded that the levels of toxic metals at both surface and deep mines were so low that the best practicable control technology currently available (BPT), as required by the 1972 Water Pollution Control Act for conventional pollutants, was sufficient to control the discharge of toxic pollutants from minesites. Their conclusion was based on four factors: 1) toxic metals were only found at or very near detection limits; 2) toxic metal reduction levels were insignificant beyond BPT levels; 3) BAT levels required by the 1972 Act were infeasible because of technical and cost considerations; and 4) toxic organics while sometimes found were observed at levels too low to effectively treat (U.S. EPA 1981). The study most importantly recognized that toxic metals may occasionally be present in high concentrations because of site-specific conditions. It was recognized that when high concentrations occurred, permit writers would have the authority to establish particular limitations (U.S. EPA 1981). Final rules exempting mining-related point source discharges from toxic's control were published in the Federal Register in October, 1985 (D. Sweeney, Personal Communication, 1987).

Pennsylvania's coal mine regulatory program is administered by the Department of Environmental Resources (DER) through the Bureau of Mining and Reclamation (BMR). The Commonwealth gained primacy for its regulatory program from the U.S. Office of Surface Mining Reclamation and Enforcement (OSMRE) in 1982. The Commonwealth also has primacy for administering the National Pollutant Discharge Elimination System (NPDES) program for EPA and performs this function through the DER's Bureau of Water Quality Management (BWQM). Shortly after assuming primacy for the coal regulatory program, BMR took over most permitting functions for mining-related activities. However, BWQM develops water quality protection reports and advises BMR whenever more stringent water quality protection limits are necessary based upon existing state regulatory water quality standards. A toxic management plan is presently applied by BWQM in the permitting of industrial waste discharges (BWQM 1985). As a result of the 1985 EPA decision to exempt mining from toxics regulation, an attempt was made by the BMR in cooperation with BWQM to collect information on certain toxics thought to be prevalent within certain mining discharges (Bercheni 1985). Although results of this sampling effort were not available for review, permits are presently issued without prescribed toxic limits. The permittee is cautioned that should effluent standards be developed under Section 307(a) of the Clean Water Act, the permit may be amended to include these effluent standards.

The Pennsylvania Fish Commission (PFC) is an independent administrative Commission within the Commonwealth of Pennsylvania which has statutory responsibility for the protection and management of the Commonwealth's fish and aquatic life resources. The PFC provides the DER information in the mine regulatory review program to partially satisfy the Commonwealth's fish and wildlife protection responsibilities specified under the Federal Surface Mining Control and Reclamation Act (SMCRA). When the EPA and subsequent DER positions surfaced in 1985, the PFC had some immediate concern about the decisions to exempt toxics from mining regulation. Prior to 1982, certain toxic metals were routinely included by BWQM in the permitting of discharges from underground mines and coal preparation plants. Consequently to some degree, this decision for exemption was a change from existing policy. This change coupled with evidence showing that 12 out of the 14 metals on the EPA priority pollutant list accumulate in both sediments and biota (Chapman et al. 1982) gave rise to a justified concern about a potential environmental problem.

Early in 1986 the PFC was contacted by the U.S. Fish and Wildlife Service (FWS), Ecological Services Office, State College, PA, about possible candidate projects for funding in their Environmental Contaminant's program. This program includes an ongoing effort to identify contaminant "hot spots" that may be having an adverse effect on fish and wildlife. A monitoring plan was proposed by the PFC to address specific mine discharge points. A review of discharge monitoring reports to insure compliance of conventional pollutant limits was performed, DER staff was contacted for recommended monitoring sites, and a final list was developed prioritizing sites for sampling. Fairly continuous discharge types were preferred rather than short-term or intermittent discharges which are commonly associated with surface mining operations in case a chronic bioaccumulative problem existed which would result in higher, perhaps detectable concentrations in fish exposed to longer treatment periods. Funding for the analyses of 18 separate samples was approved and eventually 7 different sites were sampled (fig. 1). Site 1 was downstream from a NPDES discharge location from a refuse disposal site on Laurel Lick Run (Cambria County). Sites 2 and 3 were below NPDES discharge points from coal preparation plants on Quemahoning Creek (Somerset County) and Pine Run (Jefferson County). Sites 4, 5, 6, and 7 were downstream of NPDES permitted discharge points from underground mines on Dunkard Creek (Greene County), Tenmile Creek (Washington County), South Fork Tenmile Creek (Greene County) and Bark Camp Run (Clearfield County). The objective of the study was to determine if there was a potential residual toxic metal problem in fish located downstream of various coal mining wastewater treatment locations.

MATERIALS AND METHODS

Fish were collected by backpack electrofishing at seven sites located throughout the Pennsylvania bituminous coal region (fig. 1). Sites 1, 2, and 3 were sampled on August 26-27, 1986, and sites 4, 5, 6, and 7 were sampled on October 20-21, 1986. Stations were sampled downstream of any mixing zone established in the receiving stream to insure proper exposure at least at the time of sampling. It is recognized that some species are more transient than others which could affect their temporal and spatial distributions within the watershed. Attempts were made to search downstream areas to fill three samples per site including: 1) five whole bottom feeding fish; 2) five whole predator fish; and 3) five predator fish fillets. The number of sampling sites was ultimately controlled by a funding quota of 18 individual samples. Sampling station locations are described in table 1.

Fish were collected and identified in the field, measured to the nearest 0.1 cm, weighed to the nearest 0.1 gram, wrapped in aluminum foil placed in labeled plastic

Figure 1. Map of Pennsylvania showing sampling station locations within the bituminous coalfield.

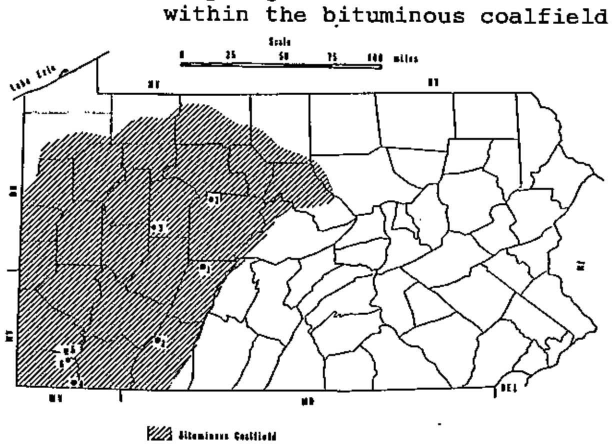


Table 1.--Sampling station locations.

SITE NUMBER	STREAM (FACILITY)	COUNTY	LOCATION
#1	Laurel Lick Run (Refuse Disposal)	Cambria	Route T-501 bridge
#2	Quemahoning Creek (Prep Plant)	Somerset	L.R. 55082 bridge
#3	Pine Run (Prep Plant)	Jefferson	L.R. 33005 bridge
#4	Dunkard Creek (Deep Mine)	Greene	3.3km downstream of L.R. 30074 bridge
#5	Ten Mile Creek (Deep Mine)	Washington	1.8km downstream of L. R. 62146 bridge in Marienna
#6	S. Fork Ten Mile Creek (Deep Mine)	Greene	1.0km downstream of L. R. 30050 bridge in Chartiers
#7	Bark Camp Run (Deep Mine)	Clearfield	0.6km upstream of PA Route 255 bridge

bags and transported on ice to the laboratory where they were frozen prior to shipment. Fish species, length, and weights of specimens collected from seven sampling sites are reported in table 2. Fish were shipped frozen on dry ice to the Mississippi State Chemical Laboratory at Mississippi State, MS for organochlorine pesticide and polychlorinated biphenyl (PCB) analysis and to the Environmental Trace Substance Research Center in Columbia, MO for selected toxic metal analysis. Both laboratories were subjected to a rigorous evaluation by a panel of U.S. FWS scientists.

RESULTS AND DISCUSSION

Results from the fish samples submitted for analysis included 26 organochlorine pesticides, polychlorinated biphenyls (PCB's) and 14 metals. The organochlorine pesticide group was either not detected or was detected at levels barely above analytical detection limits (0.01ppm) at Sites 1, 2, 3, 4, and 7. These sites were located in watersheds with limited drainage areas and sparse anthropogenic activity. However, detectable concentrations of either the parent compound or its derivatives were found for chlordane, DDT, and dieldrin at Site 5 on Tenmile Creek and Site 6 on the South Fork Tenmile Creek. Both station locations were in portions of the watershed downstream from various types of human development including agricultural activities. The Food and Drug Administration (FDA) has established action levels for chlordane, DDT and its derivatives, and dieldrin to control levels of contaminants in human food and animal food. Action levels are limits at or above which FDA will take action to remove contaminated products from the market. Prescribed limits for fish for chlordane, DDT, and dieldrin are 0.3ppm, 5.0ppm, and 0.3ppm, respectively (FDA 1981). Total chlordane concentrations were calculated by adding all isomers. Chlordane levels at Sites 5 and 6 ranged from 0.10ppm in a whole rock bass sample at Site 6 on the South Fork Tenmile Creek to the 0.30ppm action level in a smallmouth bass fillet sample at Site 5 on Tenmile Creek. Total DDT concentrations were computed by adding DDT isomers plus those of its derivatives, DDD and DDE. A maximum total DDT concentration of 0.11 was measured in the smallmouth bass fillet sample collected at Site 5 on Tenmile Creek, but did not approach the 5.0ppm action level. The maximum reported dieldrin concentration, 0.07ppm, was for a smallmouth bass whole fish sample from Site 6 on the South Fork Tenmile Creek. PCB testing revealed undetectable concentrations at all sites except Sites 5 and 6 in the Tenmile Creek watershed (table 3). The present FDA action level of 2.0ppm was not approached by any of the samples (U.S. EPA 1984). A maximum 0.40ppm was measured from a smallmouth bass whole fish sample collected at Site 6 in the South Fork Tenmile Creek.

Table 2.--Fish species, lengths and weights of specimens collected from seven sampling sites, August through October, 1986.

SITE	SPECIES (NUMBER)	SIZE RANGE (mm)	TOTAL WEIGHT (g)
#1	White Sucker(5) *Catostomus commersoni*	116-276	432
#2	Creek Chubs (5) *Semotilus atromaculatus*	93-158	114
#3	White Suckers (5)	164-193	276
	Creek Chubs (5)	130-184	174
#4	Smallmouth Bass (W)* (5) *Micropterus dolomieui*	247-302	1213
	Smallmouth bass (F)* (5)	211-304	1127
	N. Hogsuckers *Hypentilum nigricans*	167-252	465
#5	Smallmouth Bass (W) (5)	92-308	974
	Smallmouth Bass (F) (3)	264-294	864
	Rock Bass (4) *Ambloplites rupestris*	140-174	318
	Golden Redhorse (5) *Moxostoma erythrurum*	258-335	1206
#6	Smallmouth Bass (W)(5)	170-242	432
	Smallmouth Bass (F)(4)	220-324	766
	Rock Bass (4)	154-175	302
	N. Hogsuckers (5)	205-242	574
#7	Brook Trout (3) *Salvelinus fontinalis*	146-155	84
	Creek Chubs (5)	149-192	254
	White Suckers(5)	139-193	244

*-(W) signifies whole fish, (F) signified fillet.

Toxic metal analysis included testing for silver (Ag), arsenic (As), cadmium (Cd), copper (Cu), iron (Fe), mercury (Hg), molybdenum (Mo), nickel (Ni), lead (Pb), antimony (Sb), selenium (Se), thallium (Tl), vanadium (V), and zinc (Zn). Measurable concentrations were reported by the laboratory for all metals except Ag, Pb, or Tl and are presented in Table 3. Data were compared to 85th percentile numbers developed as part of the U.S. Fish and Wildlife Service's National Contaminant Biomonitoring Program (NCBP) which is an ongoing program where fish were collected at 112 monitoring stations throughout the U.S. from 1978 through 1981 to monitor temporal and geographic trends of chemical contaminants (Lowe et al. 1985). Organoleptic data (avoiding consumption because of poor taste or odor) from available literature, established ambient water quality criteria (U.S. EPA 1986), and FDA action levels (FDA 1981) were also used for comparisons. The 85th percentile values are only available for As, Cd, Cu, Hg, Se, and Zn. Levels were exceeded for Cd and Cu in creek chubs at Site 2; Cd and Cu in white suckers and creek chubs at Site 3; Hg in smallmouth bass fillets at Site 4; Cu in rock bass at Site 6; and Cu

Table 3.--Contaminant residues (ppm wet weight) in fish tissue collected at seven sampling locations throughout the Pennsylvania bituminous coalfields, August through October 1986.

Site / Species	As	Cd	Cu	Fe	Hg	Mo	Ni	Sb	Se	V	Zn	PCBs
					ppm Wet Weight							
Site #1												
White Sucker	0.02	0.05	0.80	64.1	0.05	ND	0.23	0.04	0.61	0.11	18.7	ND
Site #2												
Creek Chub	ND	0.10	1.55	23.8	0.02	0.12	0.09	0.04	0.36	0.01	22.9	ND
Site #3												
White Sucker	0.04	0.07	1.21	164.8	0.07	0.35	0.35	ND	0.68	0.21	27.3	ND
Creek Chub	ND	0.11	1.88	33.9	0.06	0.07	0.17	0.07	0.50	0.02	27.4	ND
Site #4												
Smallmouth Bass	0.08	ND	0.58	18.9	0.16	0.05	ND	0.10	0.58	0.02	18.1	ND
Smallmouth Bass (F)	0.05	ND	0.32	>.3	0.24	ND	ND	0.17	0.49	0.01	13.9	ND
N. Hogsucker	ND	0.01	0.61	23.3	0.05	0.02	0.07	0.11	0.52	0.02	19.1	ND
Site #5												
Smallmouth Bass	0.08	0.02	0.55	36.4	0.11	0.08	0.18	0.13	0.66	0.02	13.0	0.29
Smallmouth Bass (F)	0.05	ND	0.37	7.3	0.17	ND	ND	0.05	0.54	ND	9.9	0.14
Rock Bass	ND	ND	0.82	690.7	0.10	ND	0.25	0.14	0.57	0.09	23.3	0.15
Golden Redhorse	0.03	0.01	0.52	50.7	0.06	0.17	0.18	0.10	0.55	0.03	19.8	0.20
Site #6												
Smallmouth Bass	0.05	ND	0.51	20.5	0.09	ND	ND	0.15	0.71	0.02	15.2	0.40
Smallmouth Bass (F)	0.07	ND	0.51	26.7	0.09	0.08	ND	0.12	0.71	0.02	14.4	ND
Rock Bass	0.05	0.01	1.31	26.1	0.08	0.05	0.05	0.11	0.54	0.05	21.2	0.24
N. Hogsucker	0.04	0.03	0.82	53.5	0.08	ND	0.16	0.07	0.47	0.08	30.9	0.19
Site #7												
Brook Trout (F)	0.02	0.03	1.23	35.5	0.05	0.60	0.17	0.10	0.41	0.05	28.8	ND
Creek Chub	ND	0.02	2.04	23.1	0.06	0.08	0.12	0.09	0.46	ND	20.3	ND
White Sucker	0.04	0.01	0.92	138.9	0.05	ND	0.54	ND	0.47	0.19	22.7	ND
85th Percentile	0.22	0.06	0.90		0.18				0.71		40.09	

Note: Whole Fish Values Unless (F) Indicating Fillets.
ND - Not Detectable.

in brook trout, creek chubs, and white suckers at Site 7. Although the 85th percentile numbers are not an established regulatory standard, they do serve as a warning system to alert resource managers to possible "hotspots" of environmental contamination. While figures 2 and 3 do not offer any statistical support to the significance of the problem because of the small sample size, they do illustrate that Cu and Cd are present in high enough concentrations at selected stations to be on concern when compared to the national database.

Figure 2. Copper concentrations (ppm wet weight) in selected whole fish samples.

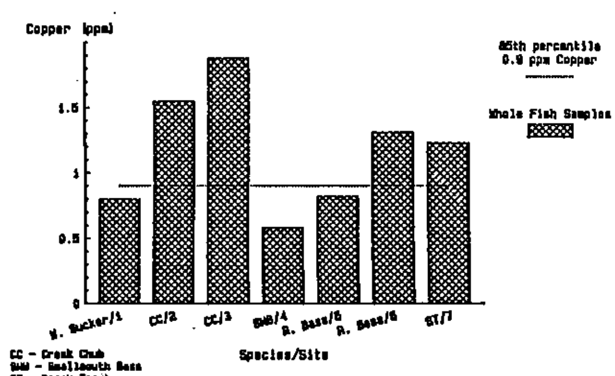
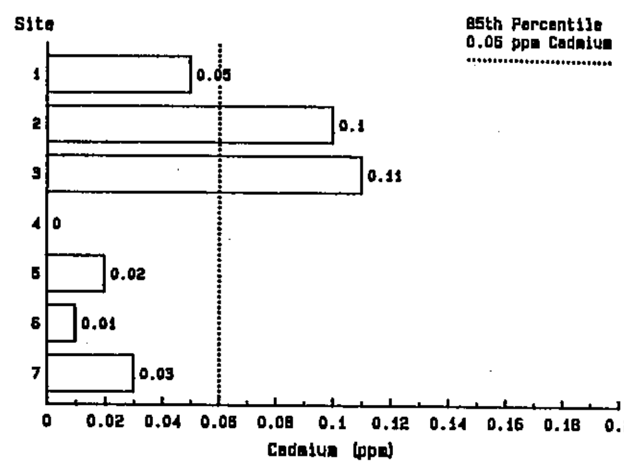


Figure 3. Cadmium concentrations (ppm wet weight) in selected whole fish samples.



Organoleptic data are scarce to nonexistent within the literature for most metals, and where it does exist, it relates primarily to drinking water and is difficult to interpret when found in fish tissue. This is especially true for certain metals such as Cu, Fe, Mo, Se, V, and Zn which can often have essential and beneficial uses in trace amounts in plant and animal nutrition (McKee and Wolf 1963). However, little is known about quantities, what forms are metabolized, how and where the elements are stored, and even less about overall mechanisms of action. These uncertainties combined with the wide array of various salts and oxides that can exist in the environment make the data difficult to interpret based on total metal concentrations. Copper, for example, has been reported to affect the taste of drinking water at concentrations in the range of 1.0-2.0 mg/L with 5.0-7.5 mg/L making water completely undrinkable (McKee and Wolf 1963). The brook trout (Site 7) and rock bass (Site 6) samples were the only two gamefish samples that might have possibly been affected by the palatability test. Comparing edible portions of gamefish samples collected at Sites 4, 5, 6, and 7 for Fe concentrations, all four sites exceed the 1.0ppm drinking water maximum which would make water unpallatable to drink (McKee and Wolf 1963). It is interesting to note that the smallmouth bass fillets collected at Sites 4 and 5 only contained 7.3ppm Fe whereas those collected at Site 6 contained 26.7ppm and the brook trout found at Site 7 were measured to have 35.5ppm.

Other nonessential nutritional metals such as As, Cd, Hg, Ni, Sb, and Tl pose more significant human health risks and are much more conspicuous by their presence in the sample analysis. The EPA (1986) developed ambient water quality criteria for all six of these priority pollutants, and sufficient information was available so that fish tissue consumption limits were developed for As, Hg, Ni, Sb, and Tl. These criteria can be used to assess the human health risks associated with the consumption of contaminated fish. Since As is the only metal of the group which is a known carcinogen, an incremental cancer risk factor of 10^{-6} was chosen by BWQM in Pennsylvania which places the criteria for consumption of aquatic organisms at 17.5 nanograms (ng)/L. A basic exposure assumption of a 70-kg male person consuming an average of 6.5g/day must be made for these criteria to be appropriate. Other criteria for the protection of public health against the ingestion of contaminated fish tissue include Hg at 146 ng/L, Ni at 100 micrograms (ug)/L and Sb at 45ppm.

These concentrations are extremely low, and all but Sb are exceeded in the edible samples collected in this study; however, sufficient populations of gamefish were not found downstream of any discharge point to be harvested in significant quantities to validate the exposure assumption. Additionally a 1.0ppm FDA action level exists for Hg which was not exceeded in any sample (FDA 1981).

CONCLUSIONS

Although the data presented in this study do not demonstrate an immediate public health threat from toxic metals, they do indicate the presence of an environmental problem which could be manifesting itself within resident aquatic communities based upon levels in excess of certain 85th percentile concentrations. More detailed study upstream and downstream of these discharges is necessary to define actual sources and quantify existing biological populations. Should further biological assessments verify that select discharges are affecting the biological uses of receiving streams by exceeding conventional pollutant loadings or introducing unregulated toxics, the regulatory authorities should address the problem by establishing site-specific limitations.

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