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Mercury Concentrations in Pond Fish in Relation to a Coal-Fired Power Plant

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Abstract. Many studies have reported that atmospheric mercury is the primary cause for bioaccumulation in fish from remote lakes. Few data, however, are available on the possible effects of near-field mercury deposition on mercury concentrations in fish from local waters. Mercury concentrations were surveyed in fish from 23 ponds in the vicinity of a 543-megawatt coal-fired power plant located at Dickerson, Maryland. A stratified random sampling design was used to select ponds within zones delineated by concentric arcs mapped at 3, 7, 10, and 15 km from the plant. For each pond, mercury concentrations were measured by atomic absorption spectrometry in sunfish (bluegill or green sunfish) in all ponds, and largemouth bass, which were present in 14 of the ponds. Mean mercury concentrations in the ponds ranged from 0.01 to 0.38 ppm for sunfish and 0.04 to 0.43 ppm for bass. Stepwise multiple regression identified variables related to tissue concentrations. Differences between strata were tested with analysis of covariance, after adjusting the concentrations to account for differences in water quality. The observed pattern of mercury bioaccumulation did not match the pattern predicted by a wet deposition model.

Reports of elevated mercury concentrations in fish from lakes and reservoirs with no known local sources have led researchers to conclude that deposition of atmospheric mercury from regional and global sources is the primary route for contamination of these waters (Sorensen *et al.* 1990; Grieb *et al.* 1990; Wiener *et al.* 1990). The combustion of coal is believed to contribute approximately one-half of the anthropogenic mercury emitted to the world's atmosphere (Douglas 1991).

Few data are available, however, on the possible impacts of near-field mercury deposition on concentrations in fish from water bodies near an emission source. Anderson and Smith (1977) examined the concentrations of mercury in fishes from Lake Sangchris (near the coal-fired Kincaid power plant) and three other lakes in central Illinois. They found that average concentrations in largemouth bass (*Micropterus salmoides*) from Lake Sangchris were lower (0.07 ppm) than the average concentrations in bass from the three other lakes (0.16–0.56 ppm). Average mercury concentrations in black bullhead (*Ameiurus melas*) were also lower in Lake Sangchris (0.16 ppm) relative to two other lakes (0.18 and 0.25 ppm). Greenberg *et al.* (1992) sampled tissues of American eels (*Anguilla rostrata*) from two sites in the Pequest River, New Jersey, and from a tributary, in a study of mercury emissions from a municipal incinerator. The authors reported that there were no obvious differences between concentrations at the three sampling sites. They stated, however, that comparisons were confounded by the small number of fish, the variation in fish sizes and ages, and the mobility of the fish.

We measured mercury concentrations in fish from ponds at various distances from the 543-megawatt coal-fired power plant located at Dickerson, Maryland (Figure 1). The objective was to determine whether the geographical distribution of tissue concentrations in fish sampled from ponds in the area surrounding the plant was consistent with the pattern of mercury deposition predicted by a wet deposition plume model.

Methods

Sampling Design

The Dickerson power plant was selected because the existence of approximately 200 ponds within a 15-km radius permitted development of a statistically rigorous sampling design. A stratified random design was used so that unbiased estimates of mercury concentrations in fish tissue samples could be made. Beginning at the power plant, concentric arcs were drawn at distances of 3, 7, 10, and 15 km (Figure 1). These distances were selected based on: (1) the knowledge that wet deposition decreases with distance from the source as the inverse of the radius, (2) initial examination of topographical maps to identify distances that would contain enough ponds to permit random selection, and (3) the criterion that the areas within at least the three inner strata should be similar. The arcs cover about 270° because the plant's location on the Potomac River permitted only the Maryland area, which is generally north, east, and south of the power plant, to be studied. Preliminary meteorological data from the closest National Weather Service station at Dulles International Airport, 31 km south of the power plant, indicated that precipitation was associated with winds from the north and south; therefore, the sampling design allowed comparisons of mercury concentrations in fish sampled from ponds to the north and south of the plant with those to the east of the plant.

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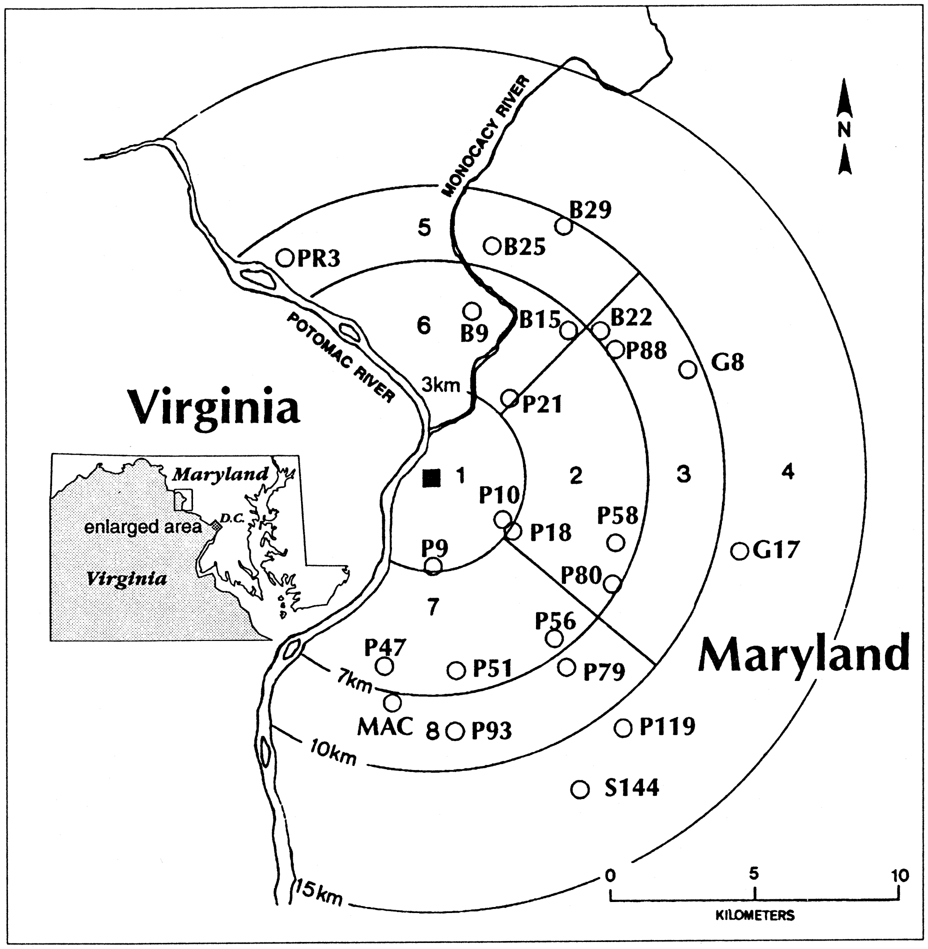

Fig. 1. Location of the Dickerson, Maryland, power plant (closed square) showing the ponds (open circles), and sampling strata (numbered 1–8)

The inner concentric arc was designated Stratum 1 and consisted of the area within 3 km of the power plant. The inner boundary of Stratum 1 was defined by the plant's property line. The areas within 3 to 7 km and 7 to 10 km of the plant were divided into thirds (three strata each) to define northern, eastern, and southern strata of approximately equal size (Figure 1). The entire area between 10 and 15 km was designated as a single stratum (Stratum 4), considered a reference area because it was the farthest from the plant. All ponds within the eight strata were identified on U.S. Geological Survey 7.5-min topographical maps. Ponds to be sampled within each stratum were chosen randomly. Pond locations and identification numbers are shown in Figure 1.

Fish Collection

Three bluegill (*Lepomis macrochirus*) and three largemouth bass were targeted from each pond. The fish were collected by angling or by dragging a small gill net through the pond. Whenever possible, crews attempted to collect fish of similar lengths. Fish were collected from 23 ponds during September and October 1992. Three ponds were sampled in all strata except Stratum 1, where only two ponds contained fish. Bluegill were collected from all ponds except pond P10, where green sunfish (*L. cyanellus*) were the only sunfish available. Largemouth bass were collected from 14 ponds; in several of these (P9, B22, S144, and P119) only two bass were collected, and in pond P79 only one bass was collected. Fish were wrapped in aluminum foil, placed in labeled plastic bags, and kept in iced coolers. Samples were kept frozen at Versar's Columbia, MD, facility and shipped on ice to Versar Laboratories, Inc., Springfield, VA, for mercury analysis.

Water Quality

Alkalinity, pH, conductivity, and hardness were measured during the survey because these parameters have been correlated with fish tissue mercury concentrations (Grieb *et al.* 1990; Sorensen *et al.* 1990; Wiener *et al.* 1990). One-liter water samples were collected in polyethylene bottles at a depth of 0.5 m in each pond and stored at 4°C until analysis.

Mercury Analysis

Prior to preparation, fish were weighed and total lengths were measured. Fillets were obtained using a stainless steel fillet knife, which was washed with detergent and water and rinsed three times with deionized water between samples. Both fillets from the sunfish and one fillet from the largemouth bass were used (see Versar, Inc., and Coastal Environmental Services, Inc. (1994) for fillet weights). Samples were digested according to USEPA (1980) methods and analyzed for total mercury by cold vapor atomic absorption (CVAA) spectrophotometry according to Method 7471 (USEPA 1986). A standard reference material, dogfish muscle tissue (DORM-1) from the National Research

Council of Canada, was analyzed. All mercury concentrations were reported in parts per million (ppm) wet weight, with a detection limit of 0.01 ppm.

Plume Modeling

The plume model assumed that most of the mercury emitted from coal-fired boiler stacks is in the gaseous mercuric chloride form, which is soluble enough to be scavenged efficiently by precipitation (Versar, Inc., and Coastal Environmental Services, Inc., 1994). Based on that assumption, wet deposition was expected to be the controlling mechanism for transport from the air to the earth's surface, especially close to the plant. Wet deposition of mercuric chloride in the study area was modeled, and rates of deposition were compared to levels of bioaccumulated mercury.

Wet deposition of stack emissions of gaseous mercuric chloride from the Dickerson power plant was predicted using the model described in Versar Inc., and Coastal Environmental Services, Inc. (1994). Representative meteorological data and stack exhaust characteristics (Brower *et al.* 1990) were input to the model. Meteorological data for 1985 to 1989 (the latest readily-accessible period), consisting of simultaneous hourly wind and precipitation measurements, were obtained from Dulles International Airport. The average wet deposition over the five-year period was calculated and expressed as isopleths of expected mercury deposition (in units of g/m^2/yr), which were normalized to a hypothetical emission rate of 1 gram per second (g/s). The model output, therefore, provided information on the expected pattern of wet deposition of mercuric chloride on a relative scale and was not intended to provide actual deposition data.

Data Analysis

Three steps were used in the data analysis. First, correlation analysis was used to identify independent (uncorrelated) environmental variables. Next, the environmental variables were used in a stepwise regression to identify the variables related to mercury tissue concentrations. Finally, these variables were used as covariates in an analysis of covariance (ANCOVA) to test for differences in mercury concentrations between strata and groups of strata.

Correlations between alkalinity, conductivity, pH, hardness, and predicted deposition rate were investigated to ensure that only independent variables were used in the models. A matrix of Pearson correlation coefficients and *t*-tests for significance was used to examine the variables for correlations (for correlation matrix, see Versar Inc., and Coastal Environmental Services, Inc., 1994). Correlations between alkalinity, hardness, and conductivity were strong ($R > 0.89$) and statistically significant ($p \leq 0.01$). Correlations of these variables with pH were not as strong but were statistically significant ($p \leq 0.05$). Thus, the regression models used only one of these four water quality parameters.

Stepwise multiple regression analysis was used to select the variables that were significantly related to tissue levels of mercury. Since fish length has been correlated frequently with mercury concentration (Grieb *et al.* 1990), length was tested as a variable in addition to the water quality parameters. For sunfish, stepwise regression identified alkalinity as the only independent variable. The resulting model was significant ($p = 0.002$, $R^2 = 0.13$). For largemouth bass, the stepwise regression identified conductivity and length as independent variables, and the resulting model was significant ($p = 0.01$, $R^2 = 0.23$). Both models included main effects without interactions.

The variables identified by stepwise regression were then used as covariates in ANCOVA for testing differences in tissue mercury concentrations between strata. Least squares means (means adjusted by the covariates) were computed for each stratum. Linear functions of the parameters were used to estimate adjusted mercury concentrations in fish sampled from strata located in the second and third concentric arcs from the power plant. *A posteriori* tests for predetermined contrasts (SAS 1987) were used to determine whether the geographic pattern of adjusted mean mercury levels among strata was consistent with predicted depositional rates. The contrasts included all comparisons between arcs of strata to determine the effect of distance, and comparisons between the two strata east of the power plant (Strata 2 and 3) and the combined strata north and south of the power plant (Strata 5, 6, 7, and 8). These contrasts were chosen based on the initial information about prevailing north–south and south–north wind directions during rain events and the general nature of wet deposition used to define the strata.

Results

Water Quality

Among the 23 ponds, there was a wide range in pH (6.11 to 9.77), alkalinity (5.7 to 84 mg/L $CaCO_3$), hardness (12 to 144 mg/L $CaCO_3$), and conductivity (25 to 280 µmhos/cm), as shown in Table 1. These water quality differences may result from differences in pond size, drainage area, geology, surrounding land use, or other factors.

Mercury Concentrations in Fish

Mean mercury concentrations in sunfish ranged from 0.01 to 0.38 ppm, and mean concentrations in largemouth bass ranged from 0.04 to 0.43 ppm (Table 1). Recovery of the standard reference material ranged from 88 to 113%.

For each species, the mean concentrations were determined for each stratum (Table 2) except for Stratum 6, where no largemouth bass were collected. For sunfish, the lowest average concentration was 0.04 ppm in the outer concentric arc (Stratum 4), which was the reference stratum. The highest average concentration was 0.19 ppm in Stratum 3, east of the plant. For largemouth bass, the lowest average concentration was 0.12 ppm in Stratum 4. The highest average concentration was 0.35 ppm in Stratum 8, south of the plant. The strata that had the highest average mercury concentrations in sunfish and largemouth bass (Strata 3 and 8, respectively) were both in the third concentric arc.

Deposition Modeling

Wet deposition modeling suggested that mercuric chloride deposition would decrease rapidly with increasing distance from the stack (Figure 2). Due to prevailing winds, depositional isopleths extend farther north and south of the stack than east and west. Predicted deposition for ponds located a given distance north or south of the stack, therefore, was higher than predicted deposition for similar ponds at the same distance to the east and west.

Examination of average mercury tissue concentrations in sunfish (Figure 3) and largemouth bass (Figure 4) for each pond (overlaid on the depositional isopleths at each pond location)

Table 1. Mercury concentrations (ppm wet weight), length data,(a) and water quality measurements at the 23 Maryland ponds

Stratum	Pond	Sunfish(b,c)	Length (mm)(d)	Largemouth Bass(e)	Length (mm)(f)	pH	Conductivity	Alkalinity	Hardness
1	P10	0.10 ± 0.02	153 ± 9	—		7.67	240	82	131
1	P9	0.10 ± 0.03	146 ± 12	0.14 ± 0.01	238 ± 4	7.02	105	31	48
2	P58	0.03 ± 0.01	132 ± 8	—		8.32	165	84	89
2	P80	0.04 ± 0.01	148 ± 7	—		7.02	130	41	60
2	P18	0.09 ± 0.10	145 ± 30	0.15 ± 0.01	327 ± 32	9.45	280	64	144
3	B22	0.38 ± 0.28	167 ± 4	0.24 ± 0.04	264 ± 52	6.96	30	8.2	15
3	G8	0.11 ± 0.04	213 ± 6	0.18 ± 0.07	273 ± 7	6.97	38	15	19
3	P88	0.11 ± 0.04	204 ± 5	0.20 ± 0.07	297 ± 10	7.64	95	47	52
4	S144	0.10 ± 0.05	198 ± 14	0.28 ± 0.17	222 ± 6	8.47	148	57	63
4	G17	0.02 ± 0.01	167 ± 2	0.04 ± 0.03	252 ± 51	7.15	75	15	26
4	P119	0.01 ± 0.01	170 ± 8	0.09 ± 0.01	275 ± 7	8.84	195	65	74
5	B25	0.17 ± 0.04	196 ± 17	0.21 ± 0.15	238 ± 46	7.06	45	14	24
5	B29	0.05 ± 0.04	183 ± 6	—		8.02	180	77	99
5	PR3	0.06 ± 0.02	209 ± 10	0.11 ± 0.08	261 ± 45	8.14	245	66	92
6	P21	0.03 ± 0.01	148 ± 6	—		8.46	150	64	84
6	B9	0.06 ± 0.02	148 ± 7	—		9.77	110	30	34
6	B15	0.06 ± 0.01	152 ± 15	—		7.35	71	28	39
7	P51	0.13 ± 0.04	169 ± 10	0.30 ± 0.11	289 ± 27	7.52	75	30	38
7	P47	0.22 ± 0.02	166 ± 29	—		6.11	25	5.7	15
7	P56	0.08 ± 0.03	138 ± 8	—		7.54	150	51	68
8	P93	0.06 ± 0.02	201 ± 5	0.43 ± 0.12	327 ± 26	6.41	45	7.0	12
8	MAC	0.06 ± 0.02	205 ± 5	0.34 ± 0.16	288 ± 27	7.34	68	23	30
8	P79	0.10 ± 0.01	136 ± 3	0.14	355	7.37	170	61	69

Units: pH (units); conductivity (μmhos/cm); alkalinity and hardness (mg/L $CaCO_3$).

a) Lengths and weights of individual fish are in Versar, Inc. and Coastal Environmental Services, Inc. (1994)

b) Green sunfish (*Lepomis cyanellus*) were sampled from pond P10; bluegill (*L. macrochirus*) from all other ponds.

c) Mean ± one standard deviation, n = 3 for all ponds

d) Mean ± one standard deviation of pond mean lengths = 169 ± 26 mm

e) Mean ± one standard deviation, n = 3 for all ponds except P9, B22, S144, P119 (n = 2), and P79 (n = 1)

f) Mean ± one standard deviation of pond mean lengths = 280 ± 38 mm

Table 2. Mean mercury concentrations in sunfish fillets adjusted by pond alkalinity (least squares mean) and mean mercury concentrations in largemouth bass fillets adjusted by fish length and conductivity (least squares mean)

		Sunfish		Largemouth Bass	
Stratum	Arc/Direction(a)	Mean	LS Mean	Mean	LS Mean
1	Inner-all	0.10	0.12	0.14	0.13
2	2nd-east	0.06	0.08	0.15	0.22
3	3rd-east	0.19	0.17	0.20	0.18
4	Outer-all	0.04	0.05	0.12	0.12
5	3rd-north	0.09	0.10	0.16	0.17
6	2nd-north	0.05	0.05	(b)	(b)
7	2nd-south	0.14	0.13	0.30	0.29
8	3rd-south	0.08	0.06	0.35	0.30
	2nd-mean	0.08	0.09	0.22	0.26
	3rd-mean	0.11	0.11	0.24	0.22

(a) Concentric arc location of the strata from the inner (first) to the outer (fourth) arc and the direction of the strata from the power plant

(b) No largemouth bass were obtained from this stratum

does not suggest a clear relationship between mercury concentrations and predicted wet deposition. For example, the highest average mercury concentration in largemouth bass (0.43 ppm) occurred in pond P93, which was near the 0.0008-g/m^2/yr isopleth, the second lowest isopleth on the graph. Also, the highest average mercury concentration in sunfish (0.38 ppm) was in pond B22, which was outside of the lowest (0.0004 g/m^2/yr) isopleth.

Water Quality and Mercury Concentrations in Fish

For sunfish, the ANCOVA detected statistically significant differences in mercury concentrations among strata when the data were adjusted for alkalinity ($p = 0.002$). The adjusted means for each stratum or group of strata were calculated as least squares means. The average adjusted mercury concentration was lowest in Stratum 4 and highest in Stratum 3 (Table 2). Adjusting for least squares means altered the strata mean concentrations slightly but did not change the strata with the highest and lowest mean values. A series of *a posteriori* analyses were performed to evaluate the significance of the differences in least squares mean concentrations between the strata. For the effect of distance from the power plant, where distance was represented by arcs of strata, only one contrast was statistically significant: The average adjusted mercury concentration was significantly lower ($p = 0.05$) in the outer arc (Stratum 4) than in the third arc (Strata 3, 5, and 8). Sunfish from Strata 2 and 3, east of the power plant, had somewhat higher mercury concentrations than sunfish from Strata 5 and 6, to the north, and Strata 7 and 8 to the south; however, the difference was not statistically significant ($p = 0.11$).

For largemouth bass, the ANCOVA detected statistically significant differences in mercury concentrations between strata after the data were adjusted for conductivity and fish length ($p = 0.03$). The average adjusted mercury concentration was lowest in Stratum 4 and highest in Stratum 8 (Table 2). Adjusting for least squares means altered the strata mean concentrations slightly but did not change the strata with the

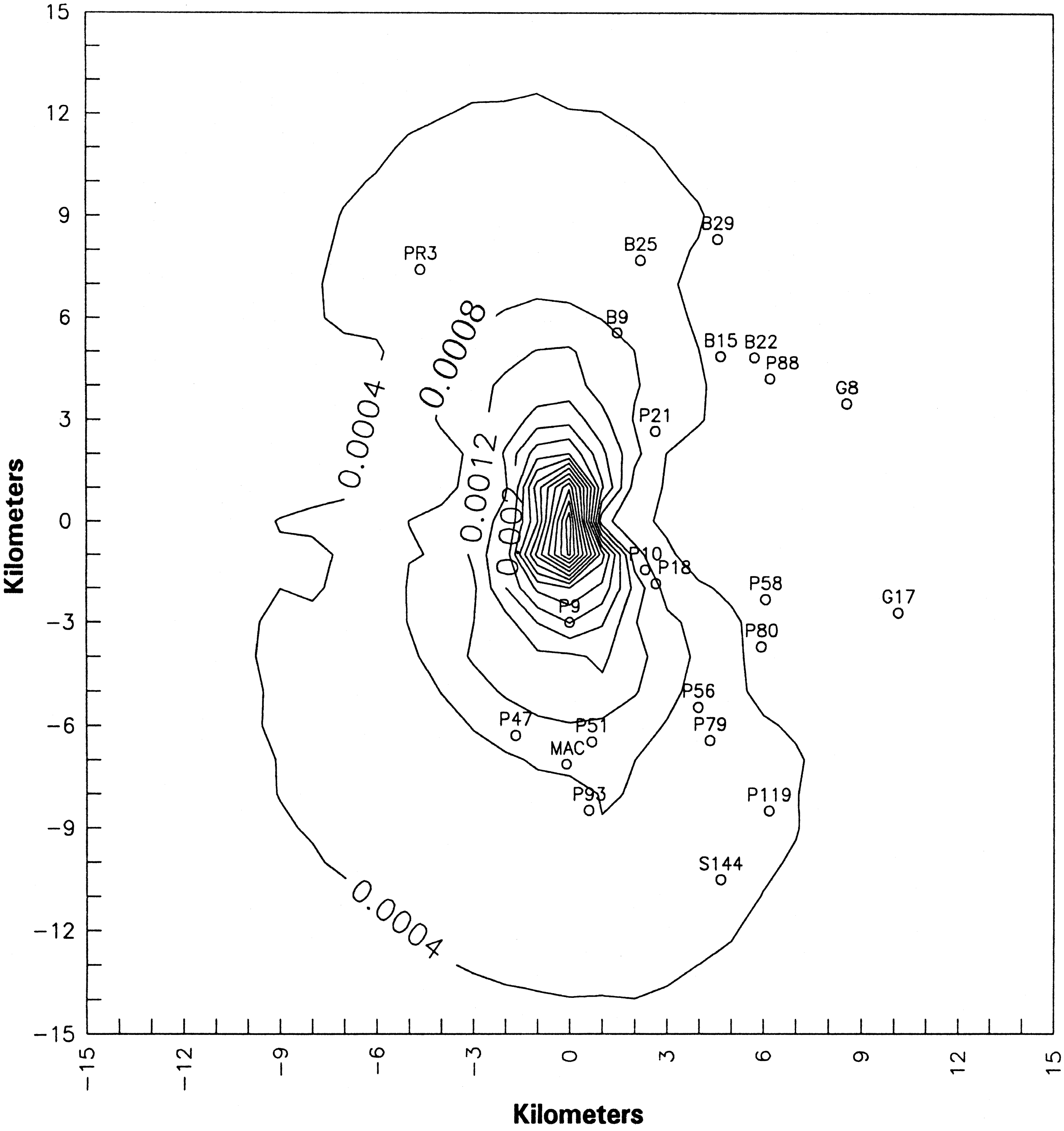

Fig. 2. Locations of sampled ponds in relation to model-predicted rates of wet deposition ($g/m^2/yr$) of mercuric chloride

highest and lowest mean values. In tests for differences between arcs of strata, no contrast was statistically significant at $p = 0.05$, but two were significant at $p = 0.10$. The average adjusted mercury concentration was significantly lower in the outer arc (Stratum 4) contrasted to the second ($p = 0.07$) and third arcs ($p = 0.06$).

Discussion

Bioaccumulation of mercury by sunfish and largemouth bass did not decrease with distance from the power plant, as did the predicted wet deposition of mercuric chloride. Largemouth bass from the combined north and south strata had higher mercury concentrations than bass from the eastern strata (Table 2), as might be predicted if wet deposition of mercury emitted from the power plant was a major source. The differences, however, were not statistically significant, even when the concentrations were adjusted to account for the effects of biotic and abiotic covariates. Although concentrations in fish sampled from the concentric arc farthest from the plant were the lowest for both species, as might be predicted if wet deposition from the power plant were the major source of mercury, concentrations in the adjacent third arc were the highest. Based on these observa-

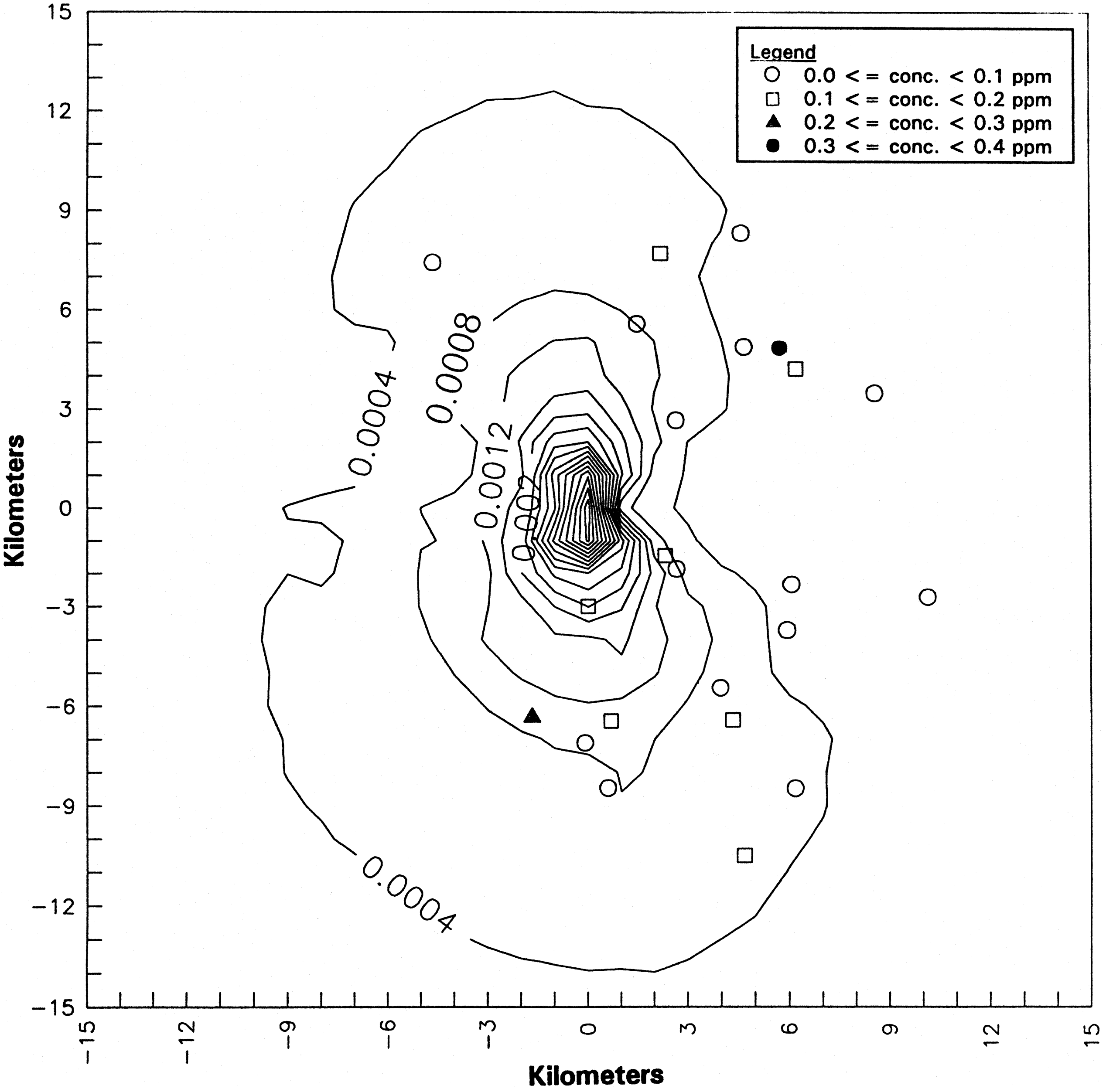

Fig. 3. Average concentration of mercury in sunfish fillets sampled from 23 ponds in relation to model-predicted rates of wet deposition ($g/m^2/yr$) of mercuric chloride

tions, the predicted pattern of local wet deposition of mercuric chloride did not match the observed pattern of mercury bioaccumulation in fish.

There are several possible explanations for the difference between the predicted pattern of wet deposition of mercuric chloride and the observed patterns of mercury accumulation by fish. Mercury in the fish may not have originated from localized wet deposition of mercuric chloride from the power plant. Other types of transport or other sources not related to the power plant may be more important. Determination of mercury speciation in power plant emissions and the measurement and modeling of local deposition are subjects of intensive research by government and industry scientists. Although this study measured several environmental variables frequently correlated with mercury uptake by fish, other unmeasured chemical variables such as sulfate concentration and dissolved organic carbon (Grieb *et al.* 1990; Winfrey and Rudd 1990; Gilmour and Henry 1991), or biological variables such as age and diet of the fish (Wren and MacCrimmon 1986) may also influence mercury bioavailability and bioaccumulation. In addition, the presence of selenium is theorized to reduce mercury bioaccumulation in fish (Turner and Rudd 1983; Wren and Stokes 1988). It is possible that selenium, which is present in trace amounts in coal (Klauda 1986), may also be emitted to the atmosphere during combustion and may be deposited locally, thus affecting mercury bioaccumulation in fish from the various pond locations.

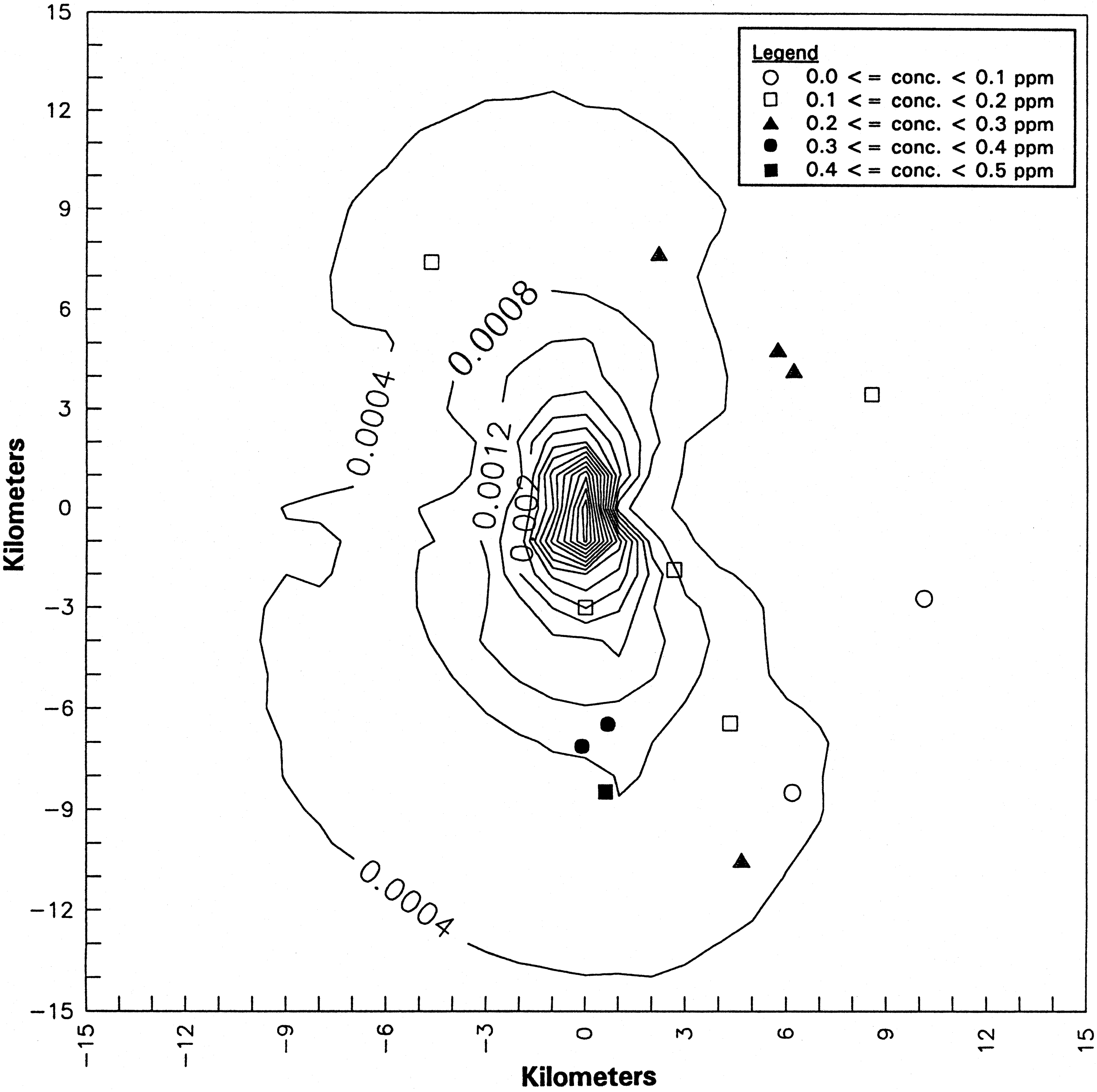

Fig. 4. Average concentration of mercury in largemouth bass fillets sampled from 23 ponds in relation to model-predicted rates of wet deposition (g/m^2/yr) of mercuric chloride

Additional (nonatmospheric) sources of mercury cannot be ruled out. For example, many of the ponds were on agricultural land. Fertilizers and lime can add mercury to cultivated soils (Andersson 1979), which can drain into ponds. Past use of mercury-based pesticides might also have resulted in historical local sources. Recently flooded reservoirs frequently contain fish with high mercury concentrations (Gilmour and Henry 1991), possibly due to the addition of nutrients that accelerate the rate of mercury methylation by soil bacteria (Stokes and Wren 1987). A similar process might also occur in ponds. No data about pesticide use and pond ages and histories were available.

Adjusted mean mercury concentrations in fish fillets from the various strata (Table 2) show generally higher mercury concentrations in largemouth bass than in sunfish. This difference is consistent with other investigators' observations that fish in higher trophic levels accumulate more mercury (Phillips *et al.* 1980; Wren and MacCrimmon 1986). Mean pond mercury concentrations for the two species, however, were not closely correlated ($R = 0.24$, $p = 0.41$). The poor correlation for mean pond mercury concentrations in fish may be due to the role of diet in mercury uptake (Wren and MacCrimmon 1986; Sorensen *et al.* 1990). It is probable that, since the ponds were isolated from one another and would be expected to support different insect and fish communities, the two species may have had different diets in the ponds where they co-occurred.

Finally, age and size of fish have been found to affect mercury accumulation (Phillips *et al.* 1980; Grieb *et al.* 1990).

Although fish length was related to mercury in largemouth bass fillets, it was not related in sunfish fillets. Stunting of green sunfish and bluegill populations due to overcrowding and competition for food is well documented (Smith 1985), and, if different ponds had different levels of sunfish crowding, the choice of length as an explanatory variable may have only partially accounted for age. The effects of size and age on bioaccumulation are complex, however, and involve not only the time for the substance to accumulate, but also dilution of the accumulated substance and changes in diet and behavior as fish grow and age (deFreitas and Hart 1975).

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