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# Concentrations and Hazard Assessment of Organochlorine Contaminants and Mercury in Smallmouth Bass from a Remote Lake in the Upper Peninsula of Michigan

K. S. Henry,[1,3] K. Kannan,[1,2,3] B. W. Nagy,[3] N. R. Kevern,[3] M. J. Zabik,[1,2] J. P. Giesy[1,2,3,4]

[1] Pesticide Research Center, Michigan State University, East Lansing, Michigan 48824-1222, USA
[2] National Food Safety and Toxicology Center, Michigan State University, East Lansing, Michigan 48824-1222, USA
[3] Department of Fisheries and Wildlife, Michigan State University, East Lansing, Michigan 48824-1222, USA
[4] Department of Zoology, Michigan State University, East Lansing, Michigan 48824-1222, USA

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**Abstract.** Concentrations of PCBs, DDTs, toxaphene, chlordanes, dieldrin, and mercury were determined in smallmouth bass (*Micropterus dolomieui*) collected from Fumee Lake, a remote lake in the Upper Peninsula of Michigan. An ecological hazard assessment was conducted to determine potential impacts of contaminants on bald eagles and mink eating fish from this lake. Concentrations of organochlorines, except toxaphene, and mercury in smallmouth bass were similar to those found in fish from Lake Superior, where atmospheric inputs are the primary sources. Bioaccumulation was indicated by a positive correlation between fish weight and contaminant concentrations for organochlorines, while mercury concentrations did not appear to correspond predictably to body weight. Concentrations of mercury and PCBs in smallmouth bass were sufficiently great to be of concern regarding their consumption by eagles or mink.

Extensive use of the Great Lakes region of North America for urban, industrial, and agricultural purposes has resulted in significant contamination of wildlife with persistent organic pollutants (Baumann and Whittle 1988; D'Itri 1988; Giesy *et al.* 1994a). Although a great deal of effort has been expended studying the fate and effects of these contaminants in wildlife in the Great Lakes (International Joint Commission 1993), less effort has been directed toward inland lakes and streams around the Great Lakes region (Swackhamer and Hites 1988). However, a few studies have shown that mercury contamination in fish from regional inland lakes may be at levels detrimental to humans or wildlife (Glass *et al.* 1990; Grieb *et al.* 1990; Mason and Sullivan 1997). The Michigan Department of Public Health issued a statewide fish consumption advisory in December 1988 for all inland lakes based on levels of mercury contamination. Fish from these inland lakes can be a potential source of human (Fiore *et al.* 1989) and wildlife exposure (Giesy *et al.* 1994b,c; Meyer *et al.* 1995) to toxic contaminants. Assessing contaminant concentrations in fish from remote inland lentic systems such as Fumee Lake will not only provide information on possible contaminant sources, but also information on exposure of humans and wildlife ingesting fish from such lakes.

*Correspondence to:* J. P. Giesy

In this study, concentrations of PCBs. *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, lindane (γ-HCH), chlordanes, dieldrin, toxaphene, and mercury were determined in smallmouth bass (*Micropterus dolomieui*), a piscivorous fish collected from Fumee Lake. The smallmouth bass is a member of the sunfish family (Centrarchidae) and is a favorite sport fish among anglers. It is a common food item for fish-eating birds and possibly mammals. Therefore, an ecological hazard assessment was conducted to determine potential impacts of contaminants on bald eagles and mink eating fish from this lake. Previous studies have evaluated hazards associated with the consumption of fish from three Michigan rivers on the health of bald eagles and mink (Giesy *et al.* 1994b,c; Giesy *et al.* 1995). A similar approach was adopted to evaluate hazards associated with the consumption of Fumee lake bass by these animals.

## Materials and Methods

### *Study Site*

Fumee Lake is located in the Upper Peninsula of Michigan (Figure 1), and has a surface area of 193 ha. The lake is fed by several surface springs and artesian wells. The maximum depth of the lake is 4 m, while more than half the lake is 1.5 m deep. On 15 August 1994, average specific conductance, pH, hardness, and alkalinity were 272 μmhos/cm, 8.41, 152, and 128 mg $CaCO_3$/L, respectively. Nitrogen and total P concentrations were 0.33 and 0.025 mg/L, respectively. Fumee Lake is a relatively remote lake with limited impact from anthropogenic activities, and access to this lake is by backpack trail only.

### *Fish Samples*

Smallmouth bass were collected with hook and line in August 1994. Individuals collected were divided into three size categories (<20 cm,

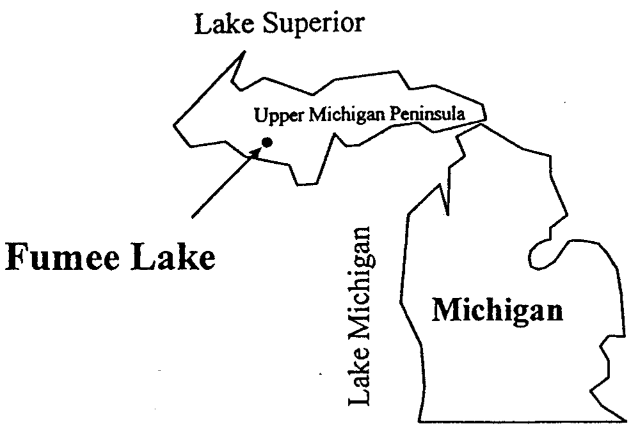


**Fig. 1.** Map of Michigan showing Fumee lake

20–30 cm, and >30 cm), and were not separated on the basis of gender. Female bass reach sexual maturity at 4 years and their length at this juncture is typically about 32 cm (Latta 1963). Males reach sexual maturity at 3 years and approximately 25 cm (Latta 1963). Individual whole fish were homogenized and frozen (−20°C) until analysis.

### *Chemical Analysis*

Organochlorines were analyzed following methods described elsewhere (Price *et al.* 1986; Zabik *et al.* 1992). Briefly, fish were homogenized with anhydrous sodium sulfate and extracted with 200 ml of 50% ethyl ether in petroleum ether. Lipid was removed by eluting through a Florisil packed glass column. PCBs and pesticides were fractionated by passing through a silica gel column. A gas chromatograph (Hewlett Packard 5890 series II) equipped with a $^{63}Ni$ electron capture detector was used to quantify organochlorines. Total concentrations of PCBs were obtained by summing corresponding peaks in samples with those in Aroclor 1254. Organochlorine pesticides including toxaphene were quantified from individually resolved peaks with corresponding standard peaks. DDTs included *p,p'*-DDT, *p,p* '-DDE, *p,p'*-DDD, while chlordanes included *cis*-nonachlor, *trans*-nonachlor, *trans*-chlordane, *cis* -chlordane, oxychlordane, and heptachlor epoxide. Recoveries of organochlorine pesticides and PCBs in spiked samples were between 90% and 110%, whereas the detection limits were 1 ng/g on a wet weight basis.

Mercury was analyzed using a cold vapor atomic absorption spectrometer (Hitachi 180-80), in which mercury ions in an oxidized sample are reduced to $Hg^0$ with $SnCl_2$. Gaseous $Hg^0$ was swept into the analysis chamber with a stream of argon (APHA 1995).

### *Hazard Indices*

Hazard indices (HI) for PCBs, DDT/DDE, dieldrin, and mercury were determined by a toxic units approach (Bowerman *et al.* 1995). One toxic unit was defined as the ratio of the concentration in the whole fish divided by the dietary no observable adverse effects concentration (NOAEC):

$$\text{HI} = \text{Concentration in fish/Dietary NOAEC}$$

An HI of greater than one (1 toxic unit) indicates that the concentration in fish was expected to be sufficiently great to equal the threshold concentrations to elicit a statistically significant response. Population level impacts at an HI of 1 may not be observable, but depending on the slope of the dose-response relationship, values of 10 to 20 are frequently related to population level effects (Bowerman *et al.* 1995). Calculated HI are based on the assumption that the diet of predator species is comprised of 100% smallmouth bass. NOAEC values have been reported for mink (Giesy *et al.* 1994c) and bald eagles (Giesy *et al.* 1995; Bowerman *et al.* 1995) and were based on laboratory feeding studies.

## Results and Discussion

### *Residue Concentrations*

Relative concentrations of contaminants in smallmouth bass were in the following order: PCBs > DDTs > toxaphene ≥ mercury > chlordanes > dieldrin > HCB (Table 1), similar to that observed in fish from several Great Lakes locations (Borgmann and Whittle 1991; De Vault *et al.* 1996). PCB concentrations ranged from 206 to 908 ng/g (mean 493) wet wt, values similar to those reported for lake trout (*Salvelinus namaycush*) collected in 1990–1992 from Lake Superior (mean 450 ng/g wet wt), but fivefold less than those from Lake Michigan (3490 ng/g) (De Vault *et al.* 1996). The mean DDT concentration of 339 ng/g wet wt, was twofold greater than that reported for lake trout from Lake Superior and two- to fourfold less than those reported for the other Great Lakes (De Vault *et al.* 1996). The mean concentration of toxaphene in Fumee Lake bass was approximately five- to tenfold less than that reported for trout from Lake Superior, but similar to or slightly less than that reported for trout from Lake Michigan (Glassmeyer *et al.* 1997). Elevated concentrations of toxaphene in fish from Lake Superior have been reported to be due to the presence of point sources (Glassmeyer *et al.* 1997). Concentrations of chlordanes and dieldrin in smallmouth bass were comparable to those reported for lake trout from Lake Superior, but less than those reported for Lake Michigan (De Vault *et al.* 1996). Lindane, aldrin, and heptachlor concentrations in bass were less than the detection limit of 1 ng/g, wet wt.

Few studies have reported concentrations of organochlorine compounds in fish from remote lakes around the Laurentian Great Lakes. A study conducted in 1983 measured organochlorine concentrations in fishes in Siskiwit Lake on Isle Royale, Lake Superior (Swackhamer and Hites 1988). That study indicated contamination of fish due to nonpoint sources in the Great Lakes region. The mean concentration of DDTs in smallmouth bass from Fumee Lake was twofold less than those reported for trout in Siskiwit Lake in 1983. Concentration ranges of dieldrin, HCB, and heptachlor epoxide in bass were similar to those of fish collected from Siskiwit Lake in 1983.

Because Fumee Lake is remote from point sources, the presence of organochlorine contaminants in Fumee Lake fish suggests atmospheric loading. Long-range transport of PCBs, DDTs, dieldrin, chlordanes, and toxaphene have been documented (Eisenreich *et al.* 1979; Swackhamer and Hites 1988). Concentrations of organochlorine compounds, except toxaphene, in Fumee Lake smallmouth bass were more similar to those measured in Lake Superior fish than to fish from the other Great Lakes (De Vault *et al.* 1996). The similarity in organochlorine concentrations between fish from Fumee Lake and Lake Superior was expected because atmospheric deposition is thought to be the primary source of contaminants to Lake Superior (Eisenreich *et al.* 1979).

**Table 1.** Concentrations (ng/g wet wt) of organochlorine pesticides, PCBs, and mercury in smallmouth bass collected from Fumee Lake, Upper Peninsula of Michigan

| Fish length | Weight (g) | DDT | Dieldrin | Toxaphene | Chlordanes | HCB | PCBs | Mercury |
|---|---|---|---|---|---|---|---|---|
| 0–20 cm | 56 | 138 | 26 | 93 | 43 | <1 | 366 | 490 |
| | 59 | 210 | 35 | 157 | 103 | <1 | 295 | 120 |
| | 68 | 193 | 28 | 113 | 111 | <1 | 206 | 370 |
| | 85 | 167 | 31 | 184 | 66 | <1 | 384 | 230 |
| *Mean* | *67* | *177* | *30* | *137* | *80.75* | *<1* | *313* | *303* |
| 20–30 cm | 143 | 313 | 46 | 247 | 145 | <1 | 427 | 240 |
| | 164 | 297 | 59 | 258 | 165 | 3 | 481 | 160 |
| | 235 | 309 | 52 | 231 | 164 | 5 | 519 | 240 |
| | 389 | 370 | 54 | 265 | 177 | 2 | 455 | 230 |
| | 409 | 411 | 55 | 274 | 177 | <1 | 563 | 140 |
| *Mean* | *268* | *340* | *53.2* | *255* | *166* | *3.3* | *489* | *202* |
| >30 cm | 779 | 498 | 79 | 350 | 243 | 10 | 624 | 160 |
| | 946 | 410 | 69 | 246 | 172 | 8 | 444 | 100 |
| | 1045 | 587 | 78 | 367 | 241 | 45 | 908 | 170 |
| | 1135 | 489 | 80 | 310 | 201 | 5 | 728 | 120 |
| | 1221 | 509 | 70 | 288 | 238 | 14 | 685 | 260 |
| *Mean* | *1025* | *499* | *75* | *312* | *219* | *16* | *678* | *162* |
| Grand mean | 442 | 339 | 53 | 236 | 156 | 11 | 493 | 221 |

DDT = *p,p'*-DDT + *p,p'*-DDD + *p,p'*-DDE
Chlordanes = *cis*-nonachlor + *trans*-nonachlor + *cis*-chlordane + *trans*-chlordane + oxychlordane + heptachlor epoxide
Lindane, heptachlor, and aldrin were less than the detection limit of 1 ng/g in all samples
PCBs as Aroclor 1254

Mercury concentrations in smallmouth bass from Fumee Lake were comparable to those reported for several species of fish from inland lakes in Michigan and Wisconsin (Grieb *et al.* 1990). The average mercury concentration in Fumee Lake smallmouth bass (221 ng/g wet wt) was comparable to that of the national mean value of 260 ng/g wet wt (Bahnick *et al.* 1994). As is the case for organochlorines, the primary source of mercury in Fumee Lake is considered to be the atmosphere. Earlier studies have shown the source of mercury in inland lakes in Minnesota to be atmospheric deposition, while geologic and point source contributions were thought to be less significant (Sorensen *et al.* 1990).

### *DDT and Chlordane Compositions*

Relative proportions of *p,p'*-DDT, *p,p'*-DDD and *p,p'*-DDE in total DDT concentrations in smallmouth bass were 7.2, 8.8, and 84%, respectively (Figure 2). The percentage of *p,p'*-DDT as total DDT increased, while the percentage of *p,p'*-DDE decreased with fish age. Among chlordane compounds, *trans*-nonachlor accounted for an average of 46% of total chlordane concentration, followed by *cis*-chlordane (22%) and *cis*-nonachlor (15%) (Figure 2). The concentration of *trans*-nonachlor was lower in older fish, while *trans*-chlordane, a parent compound, increased with fish age. Proportions of oxychlordane and *trans*-nonachlor in total chlordane concentrations were similar to those found in trout from Lakes Michigan and Superior, while the concentration of *trans*-chlordane was greater in Lake Michigan and less in Lake Superior than those of Fumee Lake fish (De Vault *et al.* 1996). Generally, the proportions of *p,p'*-DDT in total DDT and of *trans*-chlordane in total chlordane concentrations in fish in the Great Lakes region

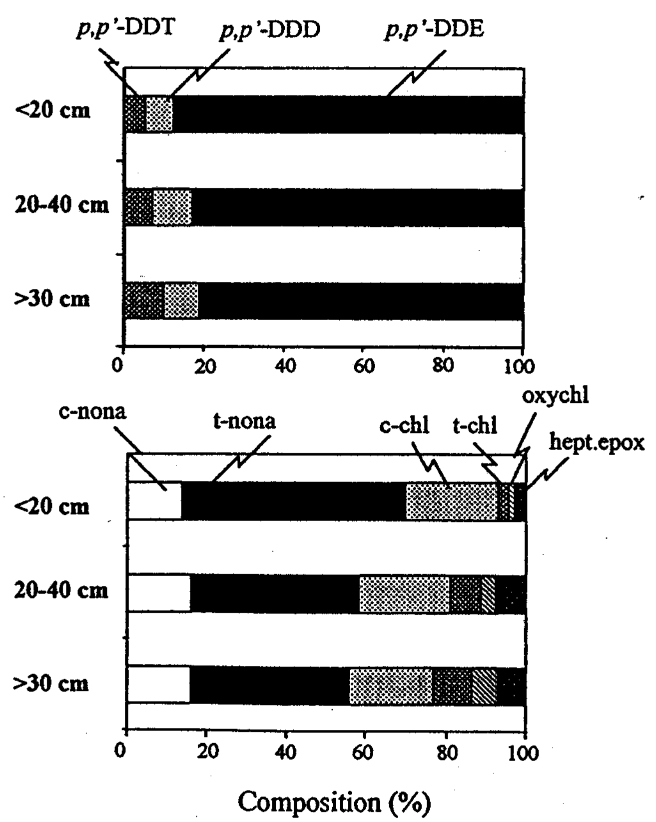


**Fig. 2.** DDT and chlordane compound compositions (%) in smallmouth bass representing three size classes from Fumee Lake

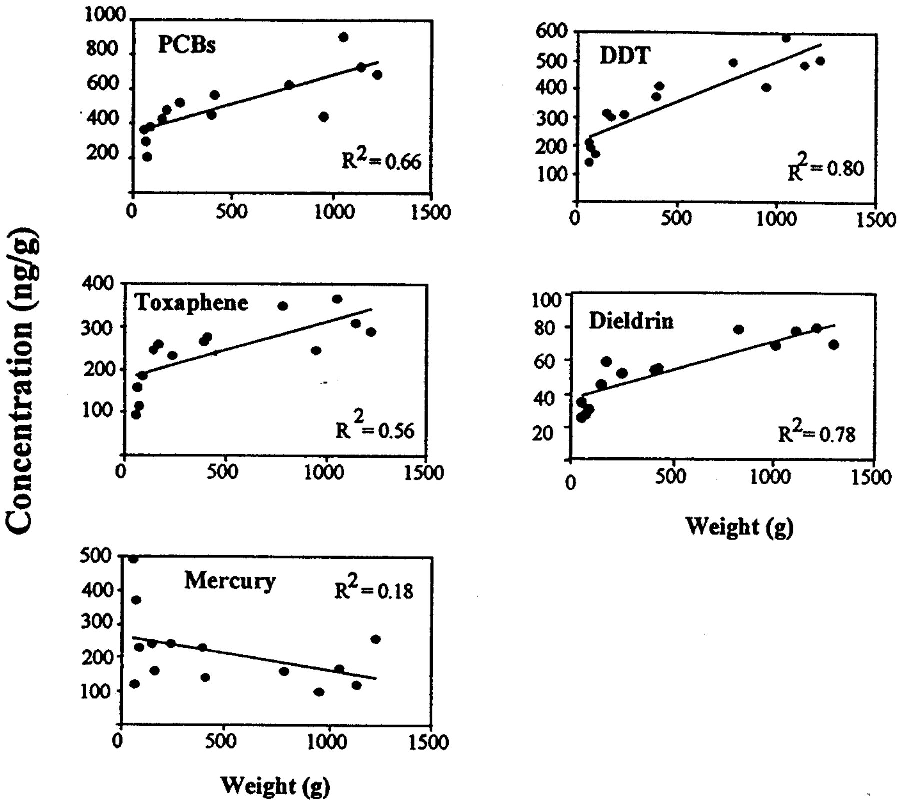


**Fig. 3.** Relationship of organochlorines and mercury concentrations (ng/g wet wt) with fish weight in smallmouth bass collected from Fumee Lake

were at least twofold less than those exposed in point source areas (Kannan *et al.* 1995; Puri *et al.* 1990).

### *Contaminant Concentrations Versus Fish Weight*

Concentrations of PCBs, DDTs, toxaphene, and dieldrin increased as a function of fish weight. This is in agreement with relationships observed in some earlier investigations (Bergmann and Whittle 1991; Gutenmann *et al.* 1992), and suggestive of the bioaccumulation potential of organochlorines. Based on the assumption that weight is directly correlated to age, longer exposure times should lead to greater concentrations. Mercury concentrations were not linearly correlated with fish weight (Figure 3). While several studies have reported positive correlations between mercury concentrations and fish length or weight (Grieb *et al.* 1990; Sorensen *et al.* 1990; Gutenmann *et al.* 1992; Lange *et al.* 1994; Kannan *et al.* 1997), a few studies have found no correlation (Cope *et al.* 1990; Driscoll *et al.* 1994). Lower mercury concentrations in older, faster-growing perch or trout have been attributed to growth dilution, in which the rate of tissue elaboration exceeds the rate of mercury accumulation (Driscoll *et al.* 1994; Kim 1995). Furthermore, a linear relationship between mercury concentrations and fish length was not found in fish from lakes with low geothermal inputs of mercury (Kim 1995). A number of studies, focused primarily on smaller lakes, have sought to correlate concentrations of mercury in fish with environmental parameters (*e.g.* Grieb *et al.* 1990; Driscoll *et al.* 1994; Horwitz *et al.* 1995).

### *Hazard Assessment*

Concentrations of mercury in smallmouth bass from Fumee Lake were less than the United States Food and Drug Administration's (FDA) action level of 1 µg/g wet wt. Concentrations of mercury exceeded or were close to the thresholds for Minnesota and Wisconsin health advisories, which are 0.16 and 0.5 µg/g wet wt, respectively. Similarly, the concentrations of organochlorines in bass currently do not exceed the FDA tolerance limits.

Because concentrations of contaminants were measured in whole fish, the values cannot be used as a direct estimate of the exposure to humans who typically eat only the muscle in a fillet. However, whole-body analysis can be useful in assessing wildlife exposure. Researchers have reported adverse effects on mink which were fed fish from the Great Lakes (Wren 1991). Similarly, the productivity of bald eagles along the Great Lakes shoreline and inland areas of the Great Lakes region is less than that in other parts of North America (Best 1994; Bowermann *et al.* 1994). Therefore, hazard assessments were conducted for mink and bald eagles by comparing concentrations of PCBs, DDTs, dieldrin, and mercury in smallmouth bass with dose-response relationships for effects of these compounds. Although the concentrations of toxaphene and chlordanes in fish were sufficiently great to possibly cause some chronic effects, no controlled laboratory studies of their effects on the mink or bald eagles were available to obtain NOAEC values. Therefore, toxaphene and chlordanes were not included in the hazard assessment.

Hazard indices (HI) were estimated for three length classes of smallmouth bass based on the concentrations of PCBs, DDTs/

**Table 2.** Hazard indices[a] of PCBs, DDTs/DDE, dieldrin, and mercury for mink and bald eagles ingesting smallmouth bass from Fumee Lake

| | Mink | | | Bald Eagle | | |
|---|---|---|---|---|---|---|
| | <20 cm | 20–30 cm | >30 cm | <20 cm | 20–30 cm | >30 cm |
| PCBs | 4.3 | 6.8 | 9.4 | 2.2 | 3.5 | 4.8 |
| DDTs[b] | <0.1 | <0.1 | <0.1 | 0.97 | 1.8 | 2.5 |
| Dieldrin | <0.1 | <0.1 | <0.1 | 2.1 | 3.8 | 5.4 |
| Mercury | 6.1 | 4.0 | 3.2 | 0.61 | 0.40 | 0.32 |

[a]NOAECs were obtained from Giesy *et al.* (1994c, 1995)
[b]Only DDE values were used for eagles

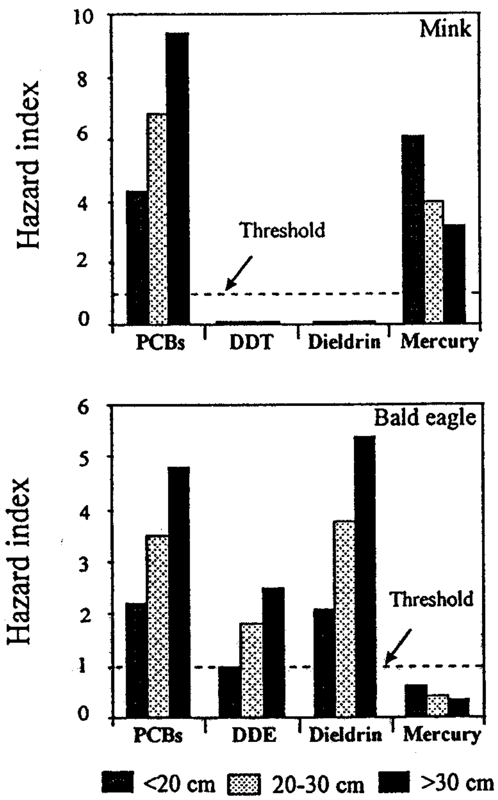


**Fig. 4.** Hazard indices of PCBs, DDTs/DDE, dieldrin, and mercury for mink and bald eagles consuming smallmouth bass representing three size classes from Fumee Lake

DDE, dieldrin, and mercury (Table 2). Among these contaminants, the two most likely to present a hazard to mink were PCBs and mercury. HI for mink based on mercury ranged from 3.2 to 6.1 (Table 2). Mercury in smaller fish represents a greater hazard to mink than it does in larger fish (Figure 4). In addition to mercury, the average HI for PCBs were between 4.3 and 9.4. Although it is unlikely that mink would eat smallmouth bass exclusively, mercury and PCB concentrations in fish are currently greater than the NOAEC for mink. Mercury concentrations of 120 to 1400 ng/g wet wt in fish were associated with adverse effects in mink and otter (Halbrook *et al.* 1994). The lowest observed adverse effect concentration (LOAEC) of PCBs in fish was 720 ng/g wet wt (Heaton 1992), similar to the concentration measured in some older bass from Fumee Lake.

The results of the hazard assessment indicate that current concentrations of PCBs in smallmouth bass of Fumee Lake could present a low-level hazard to mink and bald eagles. Similarly, the HI estimated for bald eagles and mink based on contaminant concentrations in fish from three Michigan rivers indicated that PCBs were the critical contaminants of concern (Giesy *et al.* 1994c, 1995). In addition to PCBs, mercury in mink and dieldrin in bald eagles are of concern.

## Conclusions

Contaminant concentrations, except toxaphene, in Fumee Lake smallmouth bass were more similar to concentrations in fish from Lake Superior (MDEQ 1994), where atmospheric inputs are the primary source, than to concentrations in fish from Lake Michigan, which typically receives contaminants by direct input from point sources as well as atmospheric deposition. Bioaccumulation was indicated by a positive correlation between fish weight and residue concentrations for organochlorine compounds; mercury concentrations were not correlated with body weight. Current concentrations of dieldrin are unlikely to cause population-level effects in mink, but may be near a level that could begin to cause effects in eagles, if they ate smallmouth bass exclusively. Concentrations of mercury and PCBs in the smallmouth bass from Fumee Lake were sufficient to be of concern regarding their consumption by eagles, mink, or other upper trophic-level predators. However, concentrations of neither organochlorines nor mercury have exceeded advisory guidelines established by the Michigan Department of Environmental Quality or the US Environmental Protection Agency. Mink, eagles, and other upper-trophic level predators may be at moderate risk from consumption of Fumee Lake fish, depending on their proportion in the diet, but no population level effects could be expected based on smallmouth bass contaminant concentrations.

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