

Supporting Information

**A Dynamic Multimedia Environmental and Bioaccumulation Model for
Brominated Flame Retardants in Lake Huron and Lake Erie, USA**

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1. Modeling Methods

1.1. Fugacity-based multimedia environmental model

The BFR mass balance equation for each compartment is as follows.

Air (subscript 1)

$$V_1 Z_1 \frac{df_1}{dt} = (E_1 + G_{A1} C_{B1} + D_{21} f_2 + D_{31} f_3) - f_1 (D_{12} + D_{13} + D_{R1} + D_{A1}) \quad (S1)$$

Water (subscript 2)

$$V_2 Z_2 \frac{df_2}{dt} = (E_2 + G_{A2} C_{B2} + D_{12} f_1 + D_{32} f_3 + D_{42} f_4) - f_2 (D_{21} + D_{24} + D_{R2} + D_{A2}) \quad (S2)$$

Soil (subscript 3)

$$V_3 Z_3 \frac{df_3}{dt} = (E_3 + D_{13} f_1) - f_3 (D_{31} + D_{32} + D_{R3}) \quad (S3)$$

Sediment (subscript 4)

$$V_4 Z_4 \frac{df_4}{dt} = (D_{24} f_2) - f_4 (D_{42} + D_{R4} + D_{A4}) \quad (S4)$$

where V represents the compartment volume (m^3), Z is the fugacity capacity ($\text{mol}/\text{m}^3 \cdot \text{Pa}$), f is the fugacity (Pa), E is the emission rate (mol/h), G is the advection flowrate (m^3/h), C is the concentration (mol/m^3), and D is the D-value ($\text{mol}/\text{Pa} \cdot \text{h}$). Subscript ij indicates transport from compartment i to compartment j , and the subscripts B , A and R refer to the background concentration, advection, and reaction respectively.

Given initial conditions for the chemical concentration in each compartment, the evolving fugacities in the multimedia environment for a temporally varying emission profile is determined by numerical solution to the coupled set of first-order linear ordinary differential equations (S1-S4).

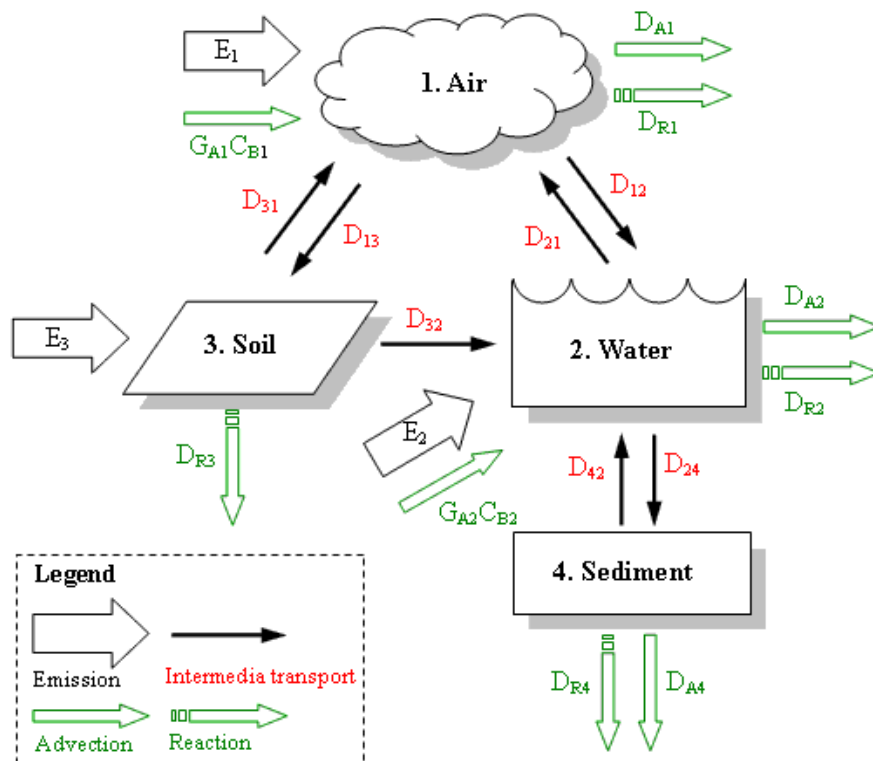
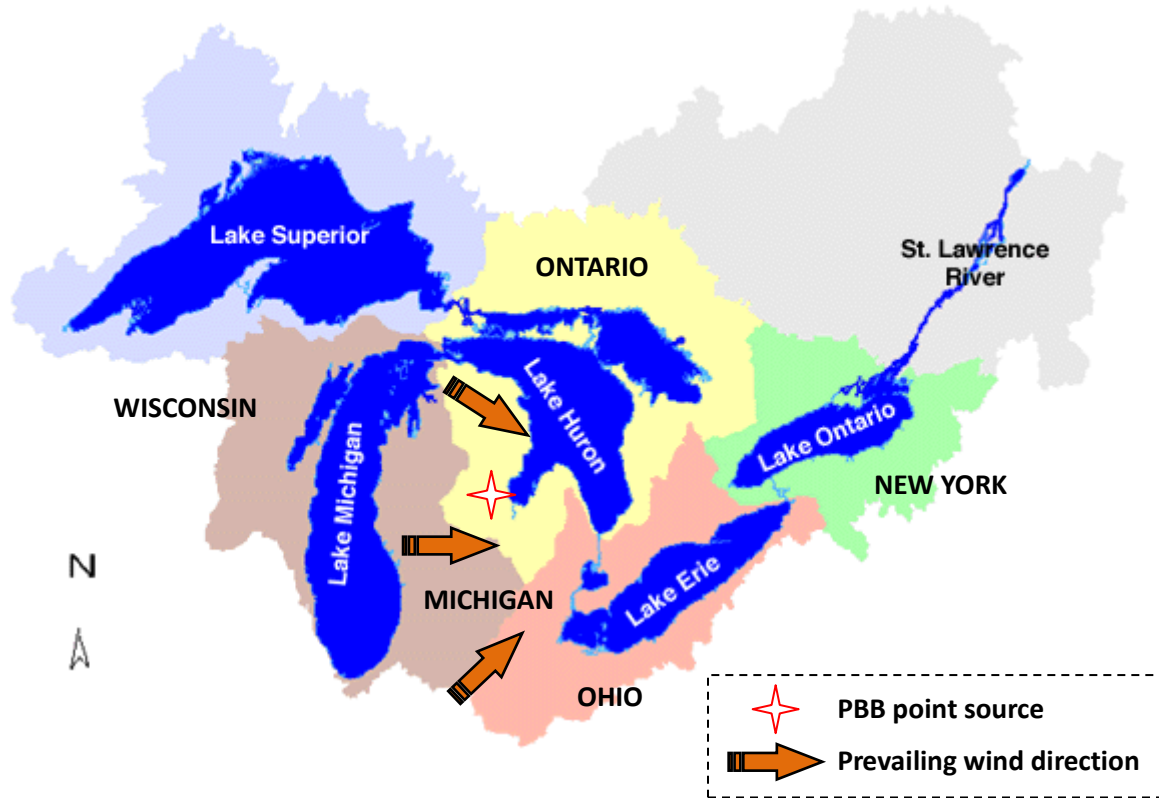


Figure S1. Schematic diagram of multimedia environmental compartment model. E is the emission rate (mol/h), G is the advection flow rate (m^3/h), C is the concentration (mol/m^3), and D is the D-value ($\text{mol}/\text{Pa}\cdot\text{h}$). Subscript ij indicates transport from compartment i to compartment j , and the subscripts B , A and R refer to the background concentration, advection, and reaction respectively.



Note:
 Figure is adapted from Reference [1].
 Prevailing wind direction is obtained from Reference [2].

Figure S2. Great Lakes watersheds. The PBB point source is located in the Saginaw Bay region of Michigan. Arrows represent prevailing wind directions during the period from 1930 to 1996.

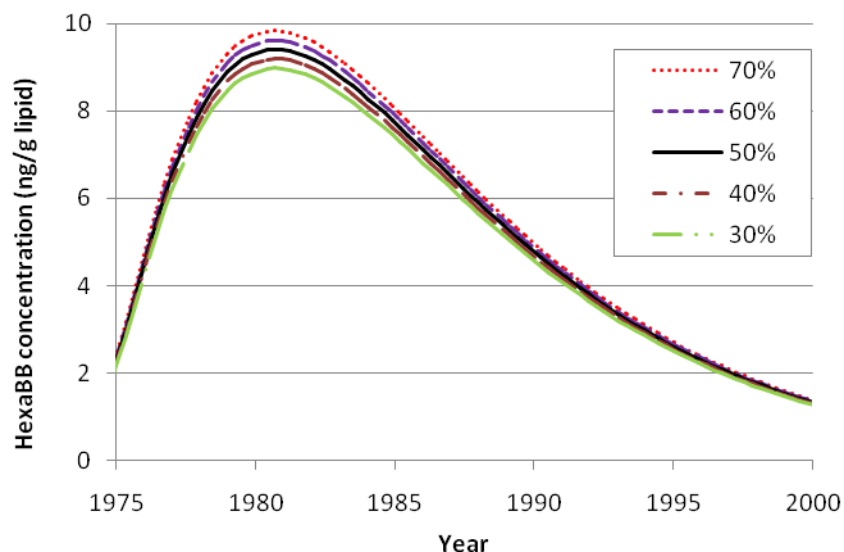


Figure S3. Comparison of model output for hexaBB concentration in Lake Erie lake trout, depending on the assumed fraction of air advection outflow of hexaBB from the Lake Huron watershed that enters the Lake Erie watershed as an air advection inflow.

Comment: In the PBB fate modeling study, 100 % of the advection losses through the water and sediment compartments and 50 % of the advection loss through the air compartment from the Lake Huron watershed were assumed to enter the Lake Erie watershed. This assumption is supported by the observation that prevailing wind directions in this region of the U.S. during the period from 1930 to 1996 are from the west-northwest (WNW) and west-southwest (WSW) in compass points [2] as shown in Figure S2.

Although the exact fraction of the PBB advection losses from the Lake Huron watershed that enters the Lake Erie watershed is unknown, as shown in Figure S3, the uncertainty in the hexaBB concentration in Lake Erie lake trout due to the choice of this fraction is less than 5.6% within the fraction range from 70% to 30%. This uncertainty is small in comparison to uncertainties associated with, for example, measurements of the observed hexaBB concentrations in lake trout.

1.2. Model inputs for landscape parameters, chemical properties, and food web properties

The water solubility and vapor pressure of hexaBB at 25 °C [3] were recalculated as $8.80 \times 10^{-4} \text{ g/m}^3$ and $5.54 \times 10^{-7} \text{ Pa}$ respectively at the mean temperature of the study region, 8.6 °C, using the van't Hoff equation. No information was available for the half-life of hexaBB in fish, and so this parameter was set equal to the hexaBB degradation half-life in the fat tissue of rats [4]. Annual average concentrations of $\text{PM}_{2.5}$ ($9.4 \text{ } \mu\text{g/m}^3$) and the aerosol density ($1,800 \text{ kg/m}^3$) were used to calculate the aerosol volume fraction in air [5, 6]. The air-side and water-side values of the air-water mass transfer coefficient (MTC) were calculated as suggested by Mackay and Yuen [7]. The air-side MTC depends on the average wind velocity, and for this parameter, Great Lake wind velocity data at 5 m height above the lake surface was used. Other MTCs were assigned using the default values for the Ontario, Canada region reported in ChemCAN Version 6.0 from the Canadian Centre for Environmental Modelling and Chemistry [8].

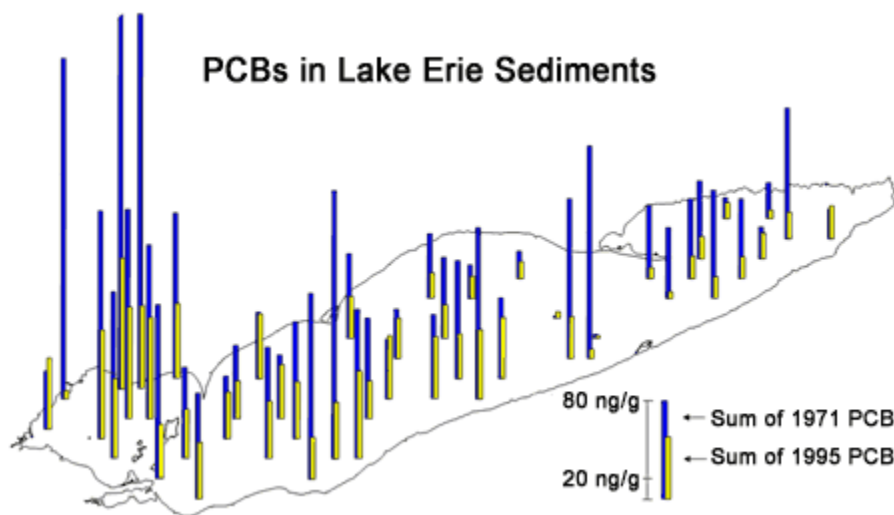
1.3. Emission rate profile

Evaporative emissions of PBDEs from principal reservoirs range from 0.01% to 0.7% [9]. Additionally, it is estimated that in the last two decades, 80% of the total reservoir of BFRs entered waste streams as municipal solid waste (MSW) [10]. Approximately 20% of the MSW was incinerated, and the remainder was disposed to landfill. Considering the structural and chemical similarities of PBBs and PBDEs [3], it is assumed that the evaporative emission factor of PBB products in use was 0.3%, and that 80% of the total reservoir of PBBs entered the waste stream. To estimate evaporative losses of PBBs from products in use and from disposed consumer goods, it was further assumed that all PBB products produced in the U.S. were used in North America, and that the evaporative emissions within the study region were proportional to the populations in the selected areas, since PBBs were principally used as fire retardant additives for the thermoplastic casings of home electronic devices. (For the resident populations of Michigan and Ontario in the Great Lakes watersheds, the combined population of these two regions is about 3% of the total population of North America.) Also, air emissions of PBBs from waste incineration were assumed to be 0.01% of the landfill waste stream, and losses of PBBs by volatilization or leaching from non-incinerated landfill waste was presumed to be comparatively small and therefore negligible [9, 11]. Using these estimates, the non-point source PBB evaporative losses from in-use and discarded consumer goods were respectively computed as 0.17% and 0.001% of the total point-source PBB emissions from the Michigan Chemical Corporation manufacturing facility. The emission contributions from in-use and landfilled products treated with PBBs were therefore small relative to direct PBB emissions from production, and are thus disregarded.

2. Results and Discussion

2.1. Pollutant contamination levels in Lake Erie

The eastern part of Lake Erie shows relatively lower pollutant contamination levels compared to other parts of the Lake Erie. National Oceanic and Atmospheric Administration (NOAA) reported PBDE sediment concentrations along Lake Erie. The eastern part of Lake Erie showed the sediment concentrations ranging from 1 to 10 ppb dry weight while the western part the sediment concentrations ranging from 11 to 88 ppb dry weight in 2004 [12]. Polychlorinated biphenyl (PCB) contamination in Lake Erie also indicates the similar trend as shown in the PBDE contamination. Figure S4 shows total PCB concentrations in sediment of Lake Erie in 1971 and 1995 (adapted from EPA [13]). The western part of Lake Erie shows relatively higher contamination than the eastern part of the lake. Painter et al. [14] also reported that the PCB sediment concentrations of the eastern part of Lake Erie were ranged from 0 to 35 ng/g while that of the western part ranged from 70 to 105 or over 105 ng/g in 1997 and 1998.



Note:

Figure is adapted from Reference [13]

Figure S4. Comparison of Lake Erie surficial sediment total PCB concentrations (ng/g dry wt.) in samples collected in 1971 and 1995.

2.1. Time trend of fugacities and PBB concentrations of aquatic organisms

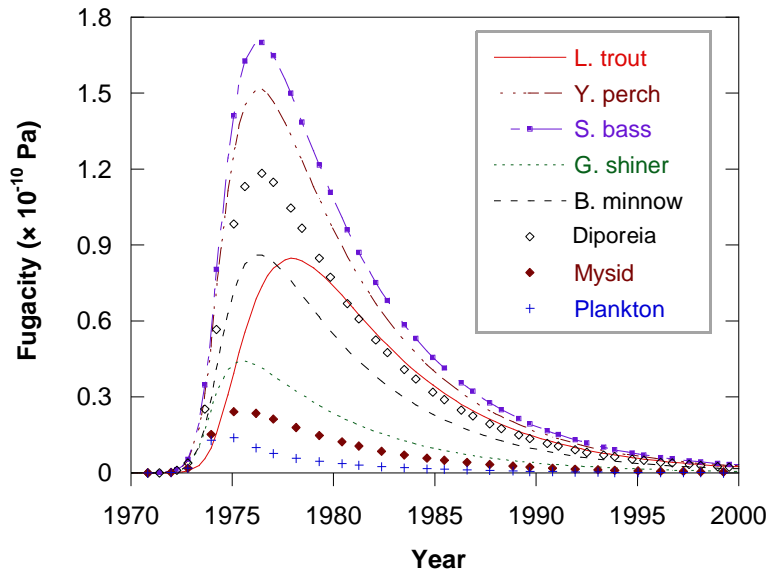


Figure S5. Time trend of fugacities of the aquatic organisms and sediment in the simulation of Lake Huron.

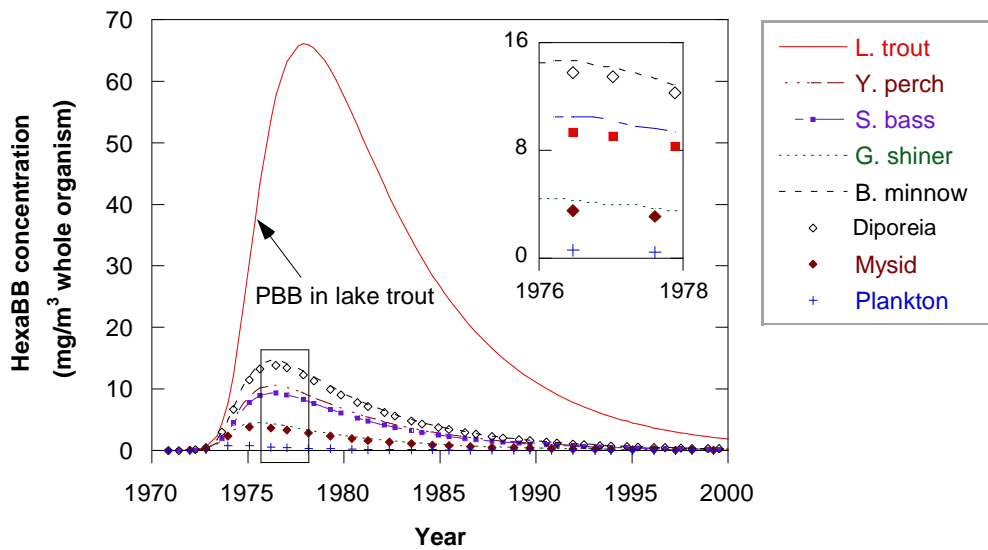


Figure S6. Time trend of hexaBB concentrations in the aquatic organisms and sediment fugacity in the simulation of Lake Huron. Detailed hexaBB concentration profiles of the organisms enclosed by a bold rectangle are shown in the right inset at the upper corner.

2.2. Estimation of BDE-47 emission by different emission scenarios

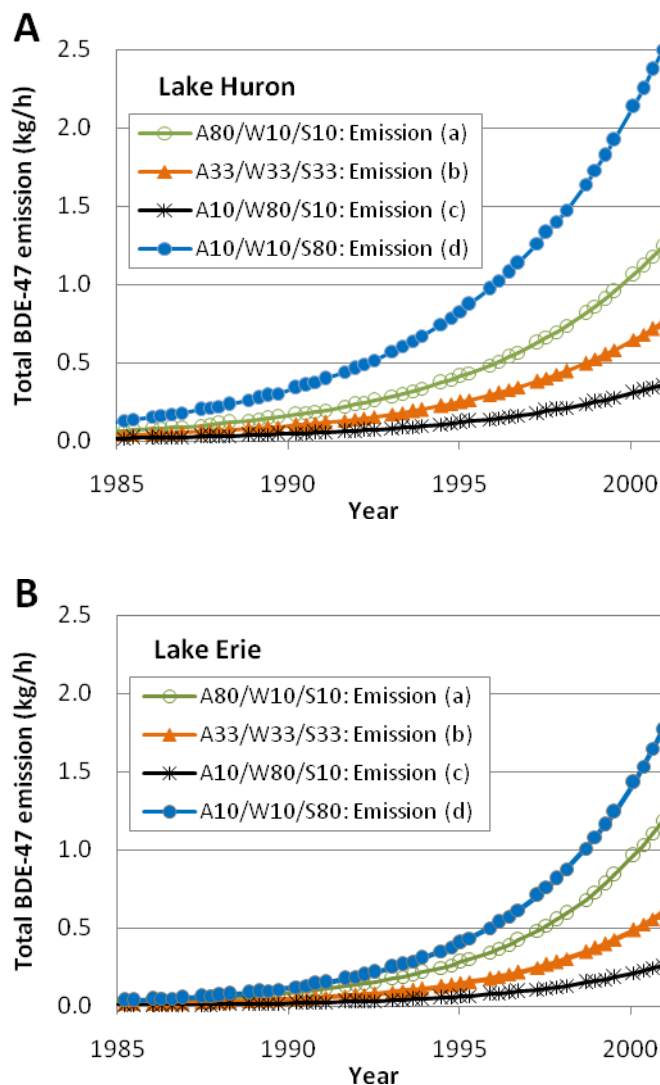


Figure S7. Estimation of total BDE-47 emission rates by different emission scenarios in Lake Huron (A) and Lake Erie (B). The legend indicates the percentage of the total BDE-47 emission released to air, water, and soil. For example, A80/W10/S10 refers to a scenario in which 80% of the BDE-47 emission is released to air, 10% is released to water, and 10% is emitted to soil.

2.3. Contribution fractions of emission sources to total BDE-47 emission into Lake Erie

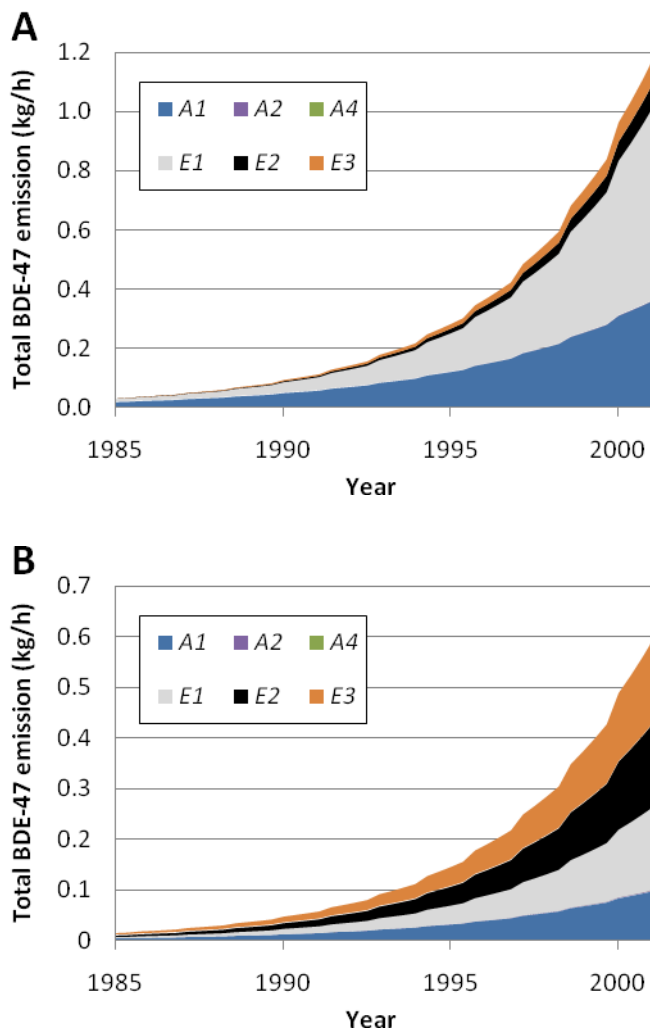


Figure S8. Input sources of BDE-47 into the Lake Erie watershed. *A* and *E* respectively represent advection input across the watershed boundary (by advective transfer from Lake Huron) and direct emission of BDE-47 from within the watershed boundary. Indices *I*, *2*, *3*, and *4* refer to the air, water, soil, and sediment compartments, respectively. (A) Emission (a) scenario with BDE-47 emission into air (80%), water (10%), and soil (10%); (B) Emission (b) scenario with BDE-47 emission equally divided into air, water, and soil (each 33.3 %).

Comment: In the case of the hexaBB fate modeling in which the point source of PBB is located in the watershed of Lake Huron, contaminant is directly delivered from the source into the each compartment of Lake Huron and is assumed to flow from the Lake Huron air, water, and sediment compartments to the corresponding media of Lake Erie. In the same model for the BDE-47 emission estimation, while the BDE-47 emission rate into Lake Huron is assumed to be only attributed to the direct emission from the Lake Huron’s watershed area into the each

compartment of Lake Huron, the emission rate into Lake Erie is assumed to be contributed by both the direct emission from Lake Erie's watershed and the advection inputs delivered from Lake Huron.

In both scenarios shown in Figure S8, the biggest BDE-47 input source to Lake Erie is direct emission from within the watershed boundary into the Lake Erie air compartment. Note that the direct emission reported for the air compartment of Lake Erie may include advection inputs originating from the adjacent areas of the Lake Erie watersheds. The contributed fraction of the advection inputs from the adjacent areas of Lake Erie was not explicitly considered in this model due to a lack of available data.

Similarly, in the case of Lake Huron, the total BDE-47 emission rate represents direct emission from within the Lake Huron watershed area in accordance with the assumed model structure. Although the total BDE-47 emission should include advection inputs from Lake Huron's neighboring watershed areas as well as direct emission from within the watershed boundary into the Lake Huron environmental compartments, the advection inputs were not explicitly considered in this model. Thus, we are unable to determine the contributed fractions of BDE-47 input from the advection inputs and from the direct emission for Lake Huron.

Table S1. Environmental input parameters and transport velocities used in Level IV multimedia compartment model.

Parameters	Lake Huron	Lake Erie
Mean temperature (°C)	8.6 ^a	8.6 ^a
Air volume (m ³)	1.93E+14 ^b	1.04E+14 ^b
Atmospheric height (m)	1000 ^c	1000 ^c
Density (kg/m ³)	1.185 ^d	1.185 ^d
Wind velocity at 5m height (m/s)	5.45 ^e	5.45 ^e
Residence time (hr)	22.4	16.4
Aerosol volume fraction	5.218E-12 ^f	5.218E-12 ^f
Aerosol density (kg/m ³)	1800 ^g	1800 ^g
Aerosol dry deposition rates (m/h)	1.804E+01 ^c	1.804E+01 ^c
Water volume (m ³)	3.54E+12 ^b	4.88E+11 ^b
Depth (m)	59.4 ^b	19.0 ^b
Density (kg/m ³)	1000 ^d	1000 ^d
Residence time (hr)	1.98E+05 ^b	2.28E+04 ^b
Aquatic biota fraction	1.0E-06 ^{g,h}	1.0E-6 ^{g,h}
Aquatic biota lipid fraction	0.05 ^{g,h}	0.05 ^{g,h}
Aquatic biota density (kg/m ³)	1000 ^{g,h}	1000 ^{g,h}
Water runoff (m/h)	3.72E-05 ^g	4.05E-05 ^g
Soil volume (m ³)	9.24E+09	1.96E+10
Depth (m)	0.25 ⁱ	0.25 ⁱ
Density (kg/m ³)	Solid (2400) ^{g,h}	Solid (2400) ^{g,h}
Organic carbon fraction	0.02 ^{g,h}	0.02 ^{g,h}
Solid / air / water fraction	0.5 / 0.2 / 0.3 ^{g,h}	0.5 / 0.2 / 0.3 ^{g,h}
Solids runoff (m/h)	2.283E-08 ^{g,h}	2.283E-08 ^{g,h}
Sediment volume (m ³)	5.96E+08	2.57E+8
Depth (m)	0.01 ^{g,h}	0.01 ^{g,h}
Density (kg/m ³)	Solid (2400) ^{g,h}	Solid (2400) ^{g,h}
Organic carbon fraction	0.04 ^{g,h}	0.04 ^{g,h}
Solid / water fraction	0.3 / 0.7 ^{g,h}	0.3 / 0.7 ^{g,h}
Residence time (hr)	9.90E+06 ^h	1.14E+06 ^h
Suspended Sediment fraction	5.0E-6 ^{g,h}	5.0E-6 ^{g,h}
Suspended Sediment density (kg/m ³)	2400 ^{g,h}	2400 ^{g,h}
Suspended Sediment organic carbon fraction	0.2 ^{g,h}	0.2 ^{g,h}
Sediment deposition rate	4.57E-07 ^{g,h}	4.57E-07 ^{g,h}
Sediment resuspension rate	1.14E-07 ^{g,h}	1.14E-07 ^{g,h}
Sediment net deposition (burial)	3.43E-07 ^{g,h}	3.43E-07 ^{g,h}
Rain rate (m/h)	9.30E-05 ^g	1.01E-04 ^g
Scavenging ratio (dimensionless)	200000	200000
Transport velocities (m/h)		
Air side air-water MTC ^j	41.85	41.85
Water side air-water MTC	0.0801	0.0801
Soil-air phase diffusion MTC	0.04 ^k	0.04 ^k
Soil-water phase diffusion MTC	1.00E-05 ^k	1.00E-05 ^k
Soil-air boundary layer MTC	1.0 ^k	1.0 ^k
Sediment-water diffusion MTC	1.00E-04 ^k	1.00E-04 ^k

^a Reference [15]; ^b Reference [16]; ^c Reference [17]; ^d Reference [18]; ^e Reference [19];

^f Reference [5]; ^g Reference [6]; ^h Reference [20]; ⁱ Reference [21];

^j MTC: mass transfer coefficient;

^k Transport velocities from the properties of Ontario area, Canada, used in ChemCAN Version 6.0 [8]

Table S2. Physicochemical properties and degradation half-lives of hexabromobiphenyl^a and tetrabromodiphenyl ether^b.

Characteristics	Hexabromobiphenyl (HexaBB)	Tetrabromodiphenyl ether (BDE-47)
Synonym(s)	FireMaster BP-6 FireMaster FF-1	
Chemical formula	C ₁₂ H ₄ Br ₆	C ₁₂ H ₆ Br ₄ O
Molecular weight	627.4	485.8
Melting point	72 °C	84.1 °C
Water Solubility	1.10E-02 g/m ³ (at 25 °C) 8.80E-04 g/m ³ (at 8.6 °C)	9.47E-02 g/m ³ (at 25 °C) 1.03E-02 g/m ³ (at 8.6 °C)
Log Kow	6.39	6.39
Vapor pressure	6.93E-06 Pa (at 25 °C) 5.54E-07 Pa (at 8.6 °C)	2.15E-04 Pa (at 25 °C) 2.33E-05 Pa (at 8.6 °C)
Henry's law constant	0.395 Pa· m ³ /mol	1.099 Pa· m ³ /mol
Degradation Half-lives of hexabromobiphenyl (units: hours)^c		
Air	906	256
Water	4,320	3,600
Soil	8,640	3,600
Sediment	38,900	14,400
Fish (Fat)	11,592 ^d	1,000 ^e

^a Reference [3].

^b Reference [22].

^c Estimated by EPIWIN software provided by U.S. Environmental Protection Agency [23].

^d Reference [4] (assumed based on the biological half-life of FireMaster BP-6 in rat fat).

^e Reference [24].

Table S3. Aquatic organisms and their properties used in the bioaccumulation model in food webs of Lake Huron and Lake Erie.

Species	Vol (cm ³)	LF ^a	QD ^b	TMD ^c	GRRD ^d	GIPV ^e	GAO ^f	GAW ^g	XW ^h	XS ⁱ
(1) Plankton	0.0005	0.015	3	500,000	0.025	0	4	5.3E-8	1	0
(2) Mysid	0.1	0.04	3	50,000	0.02	20	3.5	5.3E-8	1	0
(3) Diporeia	0.002	0.03	3	50,000	0.02	0	4	5.3E-8	0	1
(4) Bluntnose minnow	30.0	0.0435	3	5,000	0.005	4	1.5	5.3E-8	1	0
(5) Golden shiner	49.8	0.0265	3	5,000	0.005	4	1.5	5.3E-8	1	0
(6) Smallmouth bass	1474	0.0141	3	5,000	0.004	3.5	1.5	5.3E-8	1	0
(7) Yellow perch	312	0.0179	3	5,000	0.005	4	1.5	5.3E-8	1	0
(8) Lake trout	2346	0.2	3	5,000	0.002	2	1.2	5.3E-8	1	0

^a LF: lipid volume fraction.

^b QD: digestion factor.

^c TMD: metabolic half-life (days).

^d GRRD: growth rate (fraction of volume per day).

^e GIPV: feeding rate (percent of body mass per day).

^f GAO: gut absorption efficiency parameter (organic).

^g GAW: gut absorption efficiency parameter (water).

^h XW: fraction of respiration from water.

ⁱ XS: fraction of respiration from sediment.

Reference for Vol and LF of species (1~3): obtained from FoodWeb Model [8]

Reference for Vol of species (4 & 5) and LF of species (4~7): obtained from Reference [25]

Reference for Vol of species (6 & 7): obtained from Reference [26].

Reference for Vol and LF of species (8): obtained from Reference [27].

Reference for b ~ i: obtained from Reference [8, 28].

Note:

Suspended solid concentration

$$= \text{SuspendedParticle Fraction} (5 \times 10^{-6}) \times \text{SuspendedParticle Density} (2400 \text{ kg} / \text{m}^3) = 12 \text{ g} / \text{m}^3$$

Fraction of organic carbon content on suspended solids in the water = 0.2

Table S4. PBB production amount in Michigan from 1970 to 1974.

Year	Production (kg)
1970	9,480
1971	83,916
1972	1,007,446
1973	1,764,050
1974	2,214,475
1975 ^a	262,188
1976 ^a	8,193
1977 ^a	2,731
Total	5,352,480

^aThe production amounts in 1975, 1976, and 1977 were assumed 96 %, 3 %, and 1 %, respectively, of the discrepant amount between the total production from 1970 to 1977 and the sum of total annual productions from 1970 to 1974. These percentages were estimated based on ratios of PBB loading discharged to the Pine River by Michigan Chemical Corporation, Michigan [29]

Table S5. Standard deviations of lognormal distributions for degradation half-lives of hexaBB. Numbers in parentheses under each compartment are degradation half-lives of hexaBB used as medians in the lognormal distributions.

	Air (906 hrs)	Water (4,320 hrs)	Soil (8,640 hrs)	Sediment (38,900 hrs)	Note
$Cf = 2$	362	1,728	3,456	15,560	median \times 0.4
$Cf = 5$	1,268	6,048	12,096	54,460	median \times 1.4
$Cf = 10$	3,080	14,688	29,376	132,260	median \times 3.4

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