Supporting Information for

Clarifying temporal trend variability in human biomonitoring of polybrominated diphenyl ethers through mechanistic modeling

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This file contains 19 pages, 5 tables and 5 figures.

Contents

Table S1. Properties of PBDE congeners used in the simulation

Property	Symbol	BDE-47	BDE-99	BDE-153	BDE-209	Ref.
Molecular weight (g mol^{-1})	MW	486	565	644	959	
Air-water equilibrium partition coefficient at 25°C (-)	$\log K_{AW}$	-3.1	-3.4	-3.7	-4.8	[1]
Octanol-air equilibrium partition coefficient at 25°C (-)	$\log K_{OA}$	10.4	11.3	12.1	16.8	[1]
Octanol-water equilibrium partition coefficient at 25°C (-)	$\log K_{OW}$	6.5	7.0	7.4	10.0	[1]
Internal energy of air-water phase transfer (kJ mol^{-1})	ΔU_{AW}	77	79	77	65	[1]
Internal energy of octanol-water phase transfer (kJ mol^{-1})	ΔU_{OW}	-20	-12	-21	-80	[1]
Internal energy of octanol-air phase transfer (kJ mol^{-1})	ΔU_{OA}	-97	-91	-98	-145	[1]
Atmospheric OH reaction rate constant (cm^3 molecules^{-1} s^{-1})	k_{OH}	1.0×10^{-12}	5.5×10^{-13}	5.5×10^{-13}	3.4×10^{-14}	[1]
Degradation rate constant in soil (h^{-1})	k_{soil}	7.5×10^{-5}	4.1×10^{-5}	2.2×10^{-5}	9.1×10^{-6}	[1]
Degradation rate constant in sediment (h^{-1})	k_{sed}	2.5×10^{-5}	1.4×10^{-5}	7.4×10^{-6}	3.0×10^{-6}	[1]
Degradation rate constant in water (h^{-1})	k_{water}	1.5×10^{-4}	8.2×10^{-5}	4.4×10^{-5}	1.8×10^{-5}	[1]
Degradation rate constant in organic phase (h^{-1})	k_{org}	1.5×10^{-4}	8.2×10^{-5}	4.4×10^{-5}	1.8×10^{-5}	[1]
Degradation rate constant in indoor surfaces (h^{-1})	k_{surf}	0	0	0	0	[2]
Activation energy for atmospheric OH reaction (kJ mol^{-1})	E_{OH}	10	10	10	10	Default
Activation energy for degradation in soil (kJ mol^{-1})	E_{soil}	30	30	30	30	Default
Activation energy for degradation in sediment (kJ mol^{-1})	E_{sed}	30	30	30	30	Default
Activation energy for degradation in water (kJ mol^{-1})	E_{water}	30	30	30	30	Default
Activation energy for degradation in organic phase (kJ mol^{-1})	E_{org}	30	30	30	30	Default
Activation energy for degradation in indoor surfaces (kJ mol^{-1})	E_{surf}	30	30	30	30	Default
Normalized (10 g) organism biotransformation half-life (h)	$t_{1/2}^{M,organism}$	9.5×10^{2}	2.2×10^{3}	2.5×10^{3}	3.5×10^{2}	[3, 4]
Normalized (70 kg) human biotransformation half-life (h)	$t_{1/2}^{M,human}$	1.5×10^{4}	4.3×10^{3}	5.3×10^{4}	2.9×10^{2}	[5]

Table S2. Information on indoor and outdoor uses of PBDE congeners

	Fraction in indoor use (1)	Indoor use		Outdoor use	
		Scale parameter (year)(2)	Shape parameter (year)(3)	Scale parameter (year)(2)	Shape parameter (year)(3)
BDE-47	100%	9.1	2.4	-	-
BDE-99	100%	10.7	2.4	-	-
BDE-153	80%	8.6	2.4	2.5	2.4
BDE-209	55%	5.8	2.4	5.5	2.4

Notes:

(1) Calculated based on the distribution of the technical PBDE mixtures among five main applications (taken from Abbasi et al.[6])

	Indoor use			Outdoor use	
	Electrical and electronic equipment	PUF Foam	Textile	Automobile	Construction
C-pentaBDE	5-10%	90-95%			
C-octaBDE	80%			20%	
C-decaBDE	35%		20%	35%	10%

(2) Averages of the lifespans of electrical and electronic equipment (9.1 years), PUF foam (11.9 years), textiles (11.2 years), automobiles (12.0 years) and construction (12.5 years), weighted by the fractions of the technical mixtures used in individual applications. The lifespans are averages of related articles taken from the “Lifespan Database for Vehicles, Equipment, and Structures (LiVES)” (www.nies.go.jp/lifespan).

(3) Default values in Abbasi et al.[6]

Table S3. Emission factors of PBDE congeners used in the simulation

Lifecycle stage	Receiving compartment	BDE-47	BDE-99	BDE-153	BDE-209
Industrial processes	Rural; air$^{(1)}$	1.6×10^{-3}	2.7×10^{-4}	4.6×10^{-4}	1.8×10^{-4}
	Rural; soil$^{(1)}$	8.8×10^{-5}	1.5×10^{-5}	2.5×10^{-5}	1.0×10^{-5}
	Rural; freshwater$^{(1)}$	7.0×10^{-5}	1.2×10^{-5}	2.0×10^{-5}	8.0×10^{-6}
Indoor use	Indoor; air$^{(2)}$	7.3×10^{-5}	1.5×10^{-5}	3.9×10^{-6}	$3.5\times10^{-5\,(3)}$
Outdoor use	Urban; air$^{(2)}$	7.3×10^{-5}	1.5×10^{-5}	3.9×10^{-6}	$6.3\times10^{-7\,(4)}$
Waste disposal	Rural; air$^{(5)}$	3.9×10^{-4}	3.9×10^{-5}	5.0×10^{-5}	6.1×10^{-6}
	Rural; freshwater$^{(5)}$	1.0×10^{-5}	1.0×10^{-7}	1.0×10^{-7}	1.0×10^{-9}

Notes:

(1) Emission factors to air are taken from Schenker et al.[1] (for congeners other than BDE-209) and Abbasi et al.[7] (for BDE209; the geometric mean of emission factors in their high and low scenarios). Emission factors to soil and freshwater are extrapolated based on the surveyed data documenting that 91%, 5.5% and 3.5% of PBDE (based on decaBDE) emissions enter air (including both fugitive and stack emissions), land (soil), and freshwater, respectively.[8] Note that the emission factor to freshwater represents the fraction after wastewater treatment in manufacturing facilities.

(2) Calculated using an empirical relationship, $\log EF = -0.839\times\log K_{OA}$ (20 °C) + 4.83, given in ref.[9], except for BDE-209.

(3) Average of emission factors for textiles (9.5×10^{-5})[1] and plastics (6.3×10^{-7}; average of the three measurements in Table 2 of ref.[10]), weighted by the respective fractions of regional total *indoor* consumption (55% of the total PBDE consumption) used in textiles (20%) and electrical and electronic equipment (35%).[6]

(4) Emission factor from plastics; average of the three measurements in Table 2 of ref.[10]

(5) Average of emission factors from landfills and dumpsites, incineration facilities (to air only), and recycling sites (to air only) (taken from Abbasi et al.[7]), weighted by their relative importance in Ontario (60%, 15% and 25%). All landfill leachate is assumed to enter freshwater.

Table S4. Food consumption rates and exposure factors of a 25-year-old female Canadian
The model extrapolates the food consumption rates and exposure factors to other ages based on empirical relationships with human body weight[11] (69.8 kg for the 25-year-old female Canadian[12]).

Food consumption (m^3 d^{-1})	
Planktivorous fish	4.7×10^{-6} (1)
Piscivorous fish	1.4×10^{-5} (1)
Vegetables	5.2×10^{-4} (2)
Beef	5.5×10^{-5} (3)
Dairy products	2.5×10^{-4} (4)
Ingestion of environmental media	
Air (m^3 d^{-1})	15.3[12]
Drinking water (L d^{-1})(5)	3(6)
Soil (mg d^{-1})	20[12]
Indoor dust (mg d^{-1})	20[13]
Time-activity relationship	
Fraction of time spent indoors	98%[12]

Notes:

(1) Converted from statistics of the annual fish consumption rate of 6.8 kg per person, per year by Ontario residents (obtained from www.dfo-mpo.gc.ca), assuming that the fish consumption is composed of 25% planktivorous fish and 75% piscivorous fish. According to a Statista survey (https://www.statista.com/statistics/948509/types-of-fish-eaten-at-home-canada/), salmon (66% of respondents; piscivorous) and tuna (53% of respondents; planktivorous and piscivorous) are the two most popular types of fish consumed by Canadians. According to Fisheries and Oceans Canada, the eastern Pacific is the main commercial fisheries landing region for salmon and tuna, which produces 30 times more salmon and tuna than the Atlantic (See statistics between 2015 and 2017 below; http://www.dfo-mpo.gc.ca/stats/commercial/sea-maritimes-eng.htm).

Year	Eastern Pacific	Atlantic
2017	14722 (12893 t salmon + 1829 t tuna)	892 t (0 t of salmon and 892 t of tuna)
2016	26896 (24058 t salmon + 2838 t tuna)	671 t (0 t of salmon and 671 t of tuna)
2015	24362 (20064 t salmon + 4298 t tuna)	879 t (0 t of salmon and 879 t of tuna)

(2) Taken from statistics of *the Canadian Community Health Survey: Overview of Canadians' Eating Habits*. The number is for combined vegetables and fruits.

(3) Converted from statistics of protein disappearance of animal protein sources (boneless weight) in Canada (i.e., food available per person, per year) (obtained from the Canadian Agri-food Sector Intelligence www.agr.gc.ca). Average (20.1 kg per person, per year) of yearly data for 2000–2010.

(4) Converted from statistics of annual fluid milk and cream consumption of 91 liters per person, per

year by Ontario residents (obtained from www.dairyinfo.gc.ca). Assume that the milk contains 5.5% of lipid content.

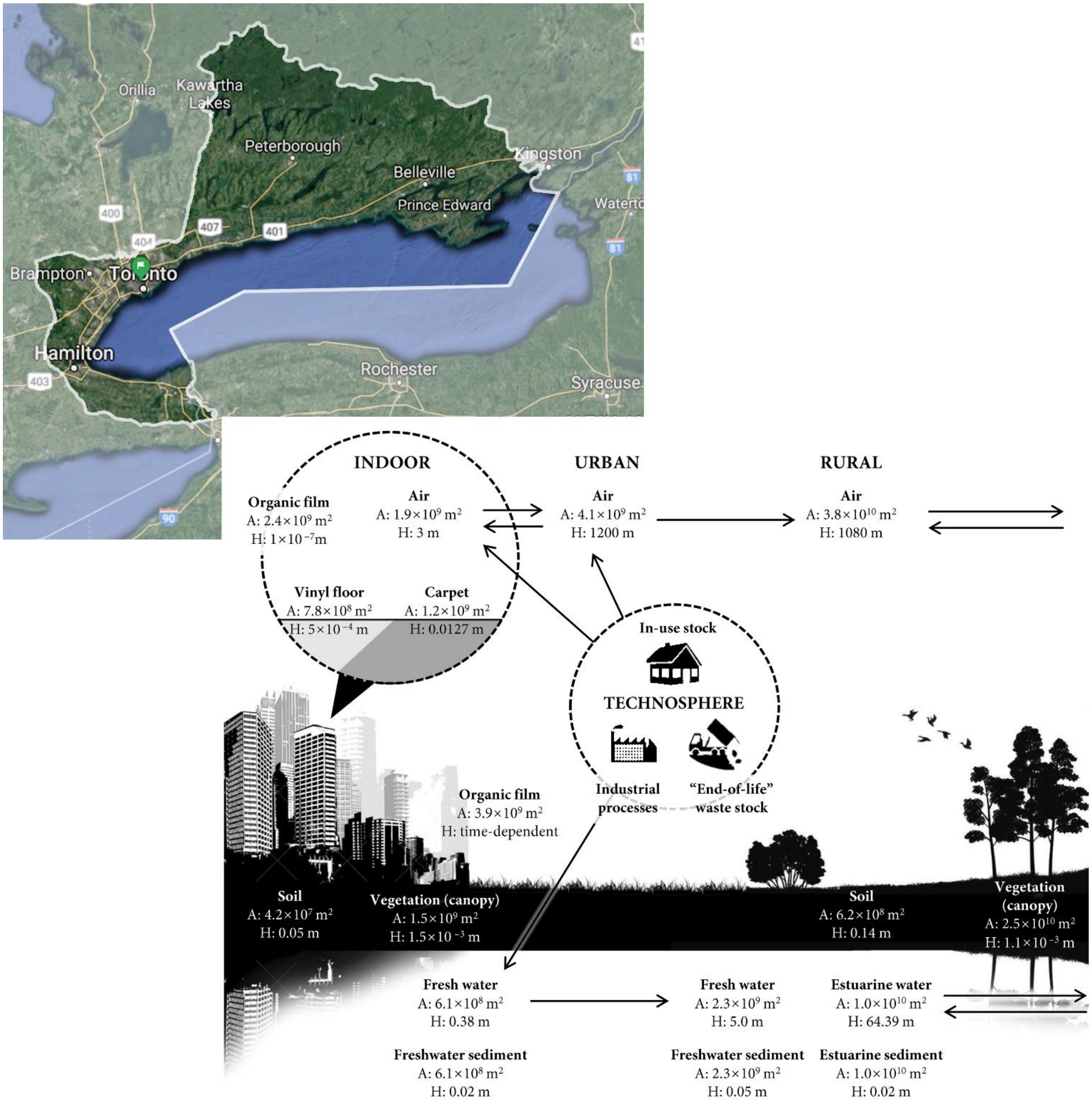
(5) We do not consider the loss of PBDEs during the treatment of drinking water.

(6) Model default value.

Table S5. Modeled daily uptake rates (μg d^{-1}) of PBDEs for a 6-year-old child and a 25-year-old adult in the years 2000 and 2010

	6-year-old child		25-year-old adult	
	Year 2000	Year 2010	Year 2000	Year 2010
BDE-47	3.88	2.61	0.89	0.60
BDE-99	1.09	0.92	0.25	0.25
BDE-153	0.09	0.05	0.10	0.07
BDE-209	2.67	2.82	0.62	0.70
Σ4PBDEs	7.73	6.40	1.86	1.62

Figure S1 Modeled region: the Canadian side of the Lake Ontario watershed



The chemical exchange between indoor and urban air is characterized using an air exchange rate of 0.75 h^{-1} (default in the RAIDAR-ICE model[14]) and that between urban and rural air is characterized using an atmospheric residence time in the urban and rural air compartments of 28.3 h (default in the ChemCAN model[15]).

Figure S2. Annual consumption (new use) of four PBDE congeners in the modeled region Extrapolated from the total consumptions in North America[7] based on the fraction of the North American population living in the modeled region.

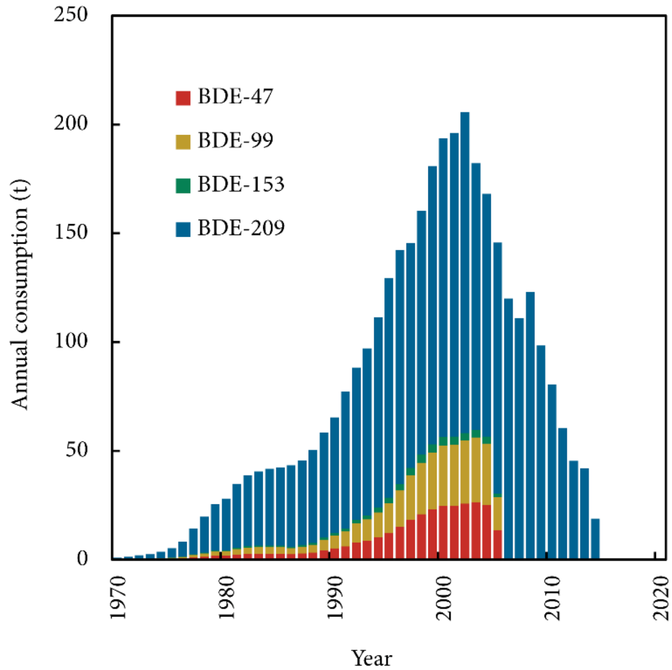
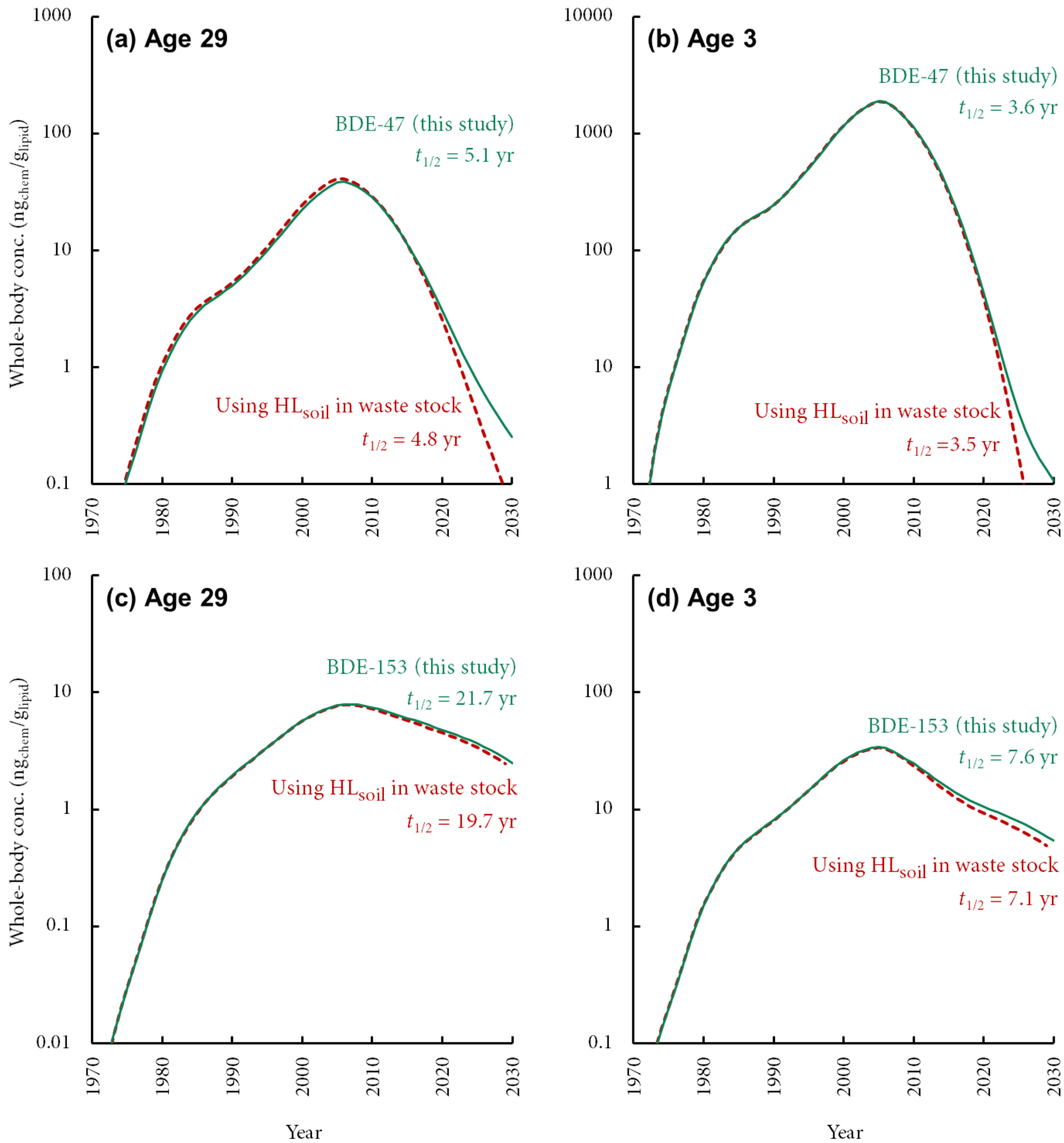


Figure S3. The impacts of model assumptions on the modeled within-age temporal trends in whole-body concentrations

(I) Comparison between modeled $t_{1/2}$ of BDE-47 (Panels a and b) and BDE-153 (Panels c and d) for adults (Panels a and c) and children (Panels b and d) under assumptions that the degradation rate constant in waste stock is the same as that in sediment (default in this study; green solid curves) or that in soil (red dashed curves). Note that using a degradation rate constant in soil, which is faster than that in sediment, leads to lower whole-body concentrations; however, the change in the calculated $t_{1/2}$ is minor.



(II) Comparison between modeled $t_{1/2}$ of BDE-47 (Panels a and b) and BDE-153 (Panels c and d) for adults (Panels a and c) and children (Panels b and d) under assumptions that all aquatic food is sourced from the coastal eastern Pacific, i.e., cell #52 of BETR-Global (default in this study; green solid curves) or from the Atlantic, i.e., cell #58 of BETR-Global (red dashed curves). Note that assuming all aquatic food to be sourced from the Atlantic, which is more contaminated than the coastal eastern Pacific, leads to higher whole-body concentrations; however, the change in the calculated $t_{1/2}$ is minor.

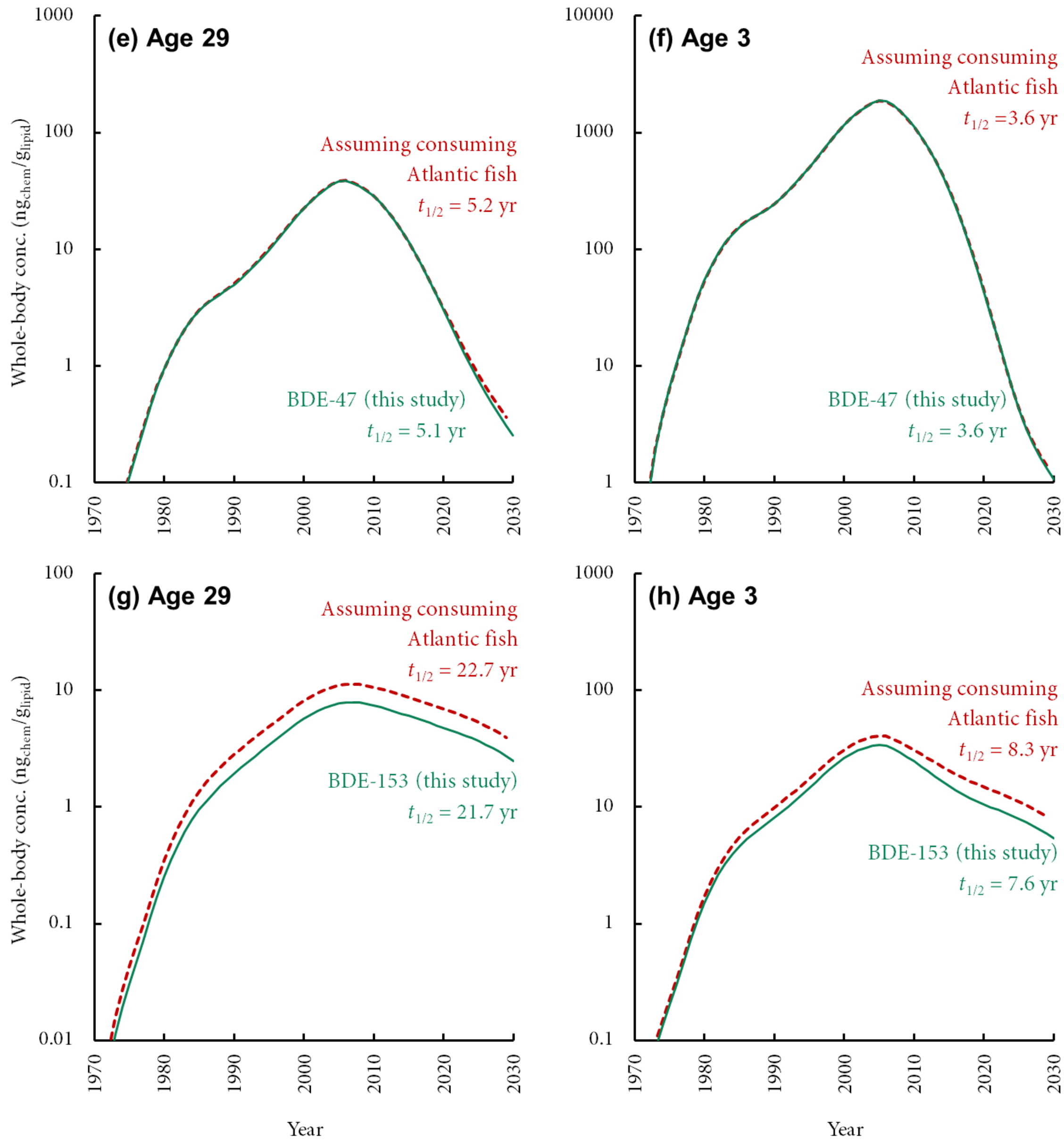
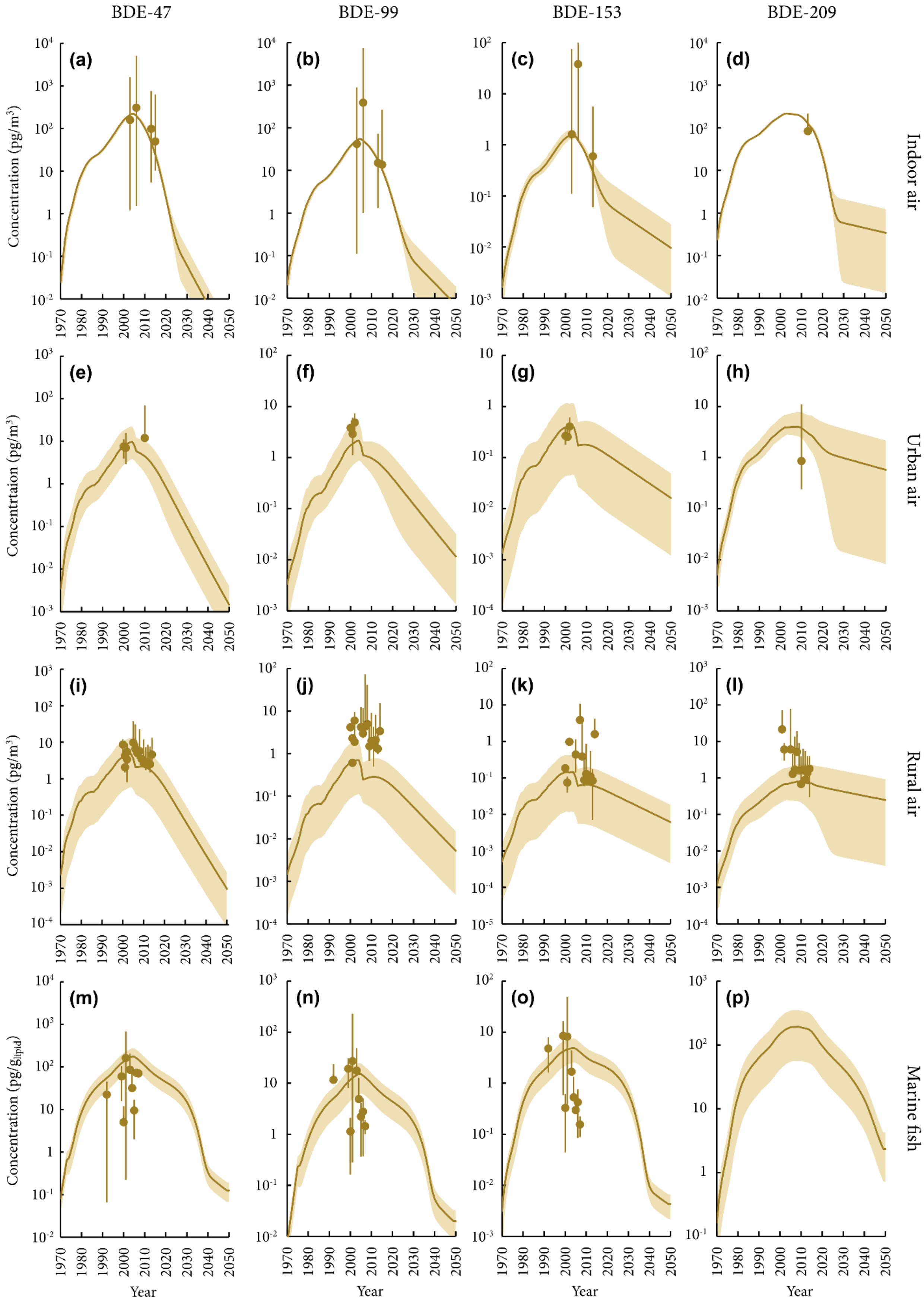


Figure S4. Modeled temporally variant concentrations of four BDE congeners between 1970 and 2050 and comparisons with observations documented in the literature

(Panels a to d for indoor air, panels e to h for urban air, panels i to l for rural air, and panels m to p for marine fish)



Notes:

For modeled concentrations, the curves represent the annual means and the shaded bands represent the ranges due to seasonal variation.

Below tabulate observed and modeled concentrations used for comparisons in Figure S4. Observed concentrations are given in "mean ± standard deviation (minimum – maximum)" (n.d. = not detected or below the limit of detection/determination, whichever is reported in the literature); modeled concentrations are given in "mean (minimum – maximum)".

Indoor air (in pg/m^3)

Year		BDE-47	BDE-99	BDE-153	BDE-209
2002-2003	Observed[16]	160 ± 290	42 ± 110	1.6 ± 8.7	
		(n.d.–1600)	(n.d.–890)	(n.d.–74)	
	Modeled	210	51	1.5	216
		(192–217)	(46–52)	(1.3–2.0)	(213–218)
2006	Observed[2]	310 ± 1100	390 ± 1700	38 ± 150	
		(1.5–5100)	(1.0–7500)	(1.2–670)	
	Modeled	197	50	1.2	208
		(194–217)	(49–53)	(1.1–1.4)	(208–210)
2013	Observed[17]	99 ± 27	1.5 ± 3.3	0.06 ± 0.17	83 ± 31
		(5.4–759)	(1.3–73)	(0.06–5.7)	(n.d.–217)
	Modeled	47	17	0.3	126
		(45–61)	(15–21)	(0.2–0.5)	(123–144)
2015	Observed[18*]	94 ± 120	26 ± 52		
		(10–630)	(N.D.–370)		
	Modeled	24	10	0.2	97
		(22–33)	(9–13)	(0.1–0.3)	(94–108)

* Concentrations based on PUF passive air samplers.

Urban air (in pg/m^3)

Year		BDE-47	BDE-99	BDE-153	BDE-209
2000	Observed[19†]	9.5	4.3	0.2	
		(7.7–11.2)	(3.9–4.6)	(0.18–0.25)	
	Modeled	7.7	1.7	0.4	3.4
		(2.8–18)	(0.6–4.1)	(0.1–1.2)	(2.3–6.0)
2001	Observed[19†]	7.3 ± 3.8	3.1 ± 1.3	0.25 ± 0.13	
		(3.1–15)	(1.5–5.8)	(n.d.–0.39)	
	Modeled	8.3	1.9	0.4	3.6
		(3.1–19)	(0.7–4.3)	(0.1–1.2)	(2.5–6.4)
2010-2011	Observed[20‡]	12			0.86
		(2–58)			(0.62–10.1)

	Modeled	4.4	1.0	0.2	3.8
		(1.8–10.2)	(0.5–2.1)	(n.d.–0.5)	(2.4–7.6)

† Concentrations reported at sampling sites Gage, Junction, and S. Riverdare.

‡ Numbers are read from Figure 1 of Shoeib et al.[20] because no numeric data are available.

Rural air (in pg/m^3)

Year		BDE-47	BDE-99	BDE-153	BDE-209
2000	Observed[19§]	4.7	3.0	0.12	
		(4.1–5.3)	(2.4–3.6)	(0.06–0.18)	
	Modeled	4.0	0.6	0.1	0.6
		(0.5–11)	(0.1–1.7)	(0.01–0.4)	(0.2–1.6)
2001	Observed[19§]	6.5	1.9	0.2	
		(1.5–15.2)	(0.5–3.5)	(0.04–0.31)	
	Modeled	4.0	0.6	0.1	0.6
		(0.5–11)	(0.1–1.7)	(0.01–0.4)	(0.2–1.7)
2002	Observed[21]	5.5	1.9		16
		(n.d.–10.3)	(n.d.–5.9)		(n.d.–105)
	Modeled	4.4	0.7	0.1	0.7
		(0.5–12)	(0.1–1.9)	(0.01–0.4)	(0.2–1.9)
2005	Observed[22]	7.7 ± 6.7	3.7 ± 4.2	0.88 ± 0.97	1.1 ± 1.3
		(1.6–26)	(1.0–23)	(0.2–2.0)	(0.11–6.5)
	Modeled	3.6	0.5	0.1	0.7
		(0.5–9.6)	(0.1–1.3)	(0.01–0.3)	(0.2–2.1)
2006	Observed[22]	6.7 ± 6.2	4.1 ± 3.8	0.88 ± 0.30	1.5 ± 2.2
		(1.3–31)	(1.3–17)	(0.67–1.2)	(0.084–12)
	Modeled	2.1	0.2	0.06	0.8
		(0.4–5.5)	(0.1–0.6)	(0.01–0.2)	(0.2–2.2)
2007	Observed[22]	17 ± 54	22 ± 92	16 ± 22	0.75 ± 0.68
		(0.6–290)	(1.0–500)	(0.87–49)	(0.092–3.4)
	Modeled	2.1	0.3	0.06	0.8
		(0.3–5.6)	(0.1–0.7)	(0.01–0.2)	(0.2–2.2)
2008	Observed[22]	17 ± 32	17 ± 53	7.5 ± 8.9	1.7 ± 4.6
		(0.14–170)	(0.2–900)	(0.52–23)	(0.1–25)
	Modeled	2.1	0.3	0.06	0.8
		(0.3–5.8)	(0.1–0.7)	(0.01–0.2)	(0.2–2.4)
2009	Observed[22]	11 ± 14	11 ± 20	0.95 ± 2.0	0.81 ± 0.78
		(0.82–54)	(0.65–66)	(0.028–6.3)	(0.024–3.0)
	Modeled	2.2	0.3	0.07	0.8
		(0.3–5.8)	(0.1–0.7)	(0.01–0.2)	(0.2–2.3)

2010	Observed[22]	10 ± 14	11 ± 24	1.2 ± 2.5	0.5 ± 0.58
		(0.9–48)	(0.58–110)	(0.029–10)	(0.024–1.7)
	Modeled	2.1	0.3	0.07	0.8
		(0.3–5.8)	(0.1–0.8)	(0.01–0.2)	(0.2–2.3)
2011	Observed[22]	20 ± 34	27 ± 66	5.6 ± 9.4	1.2 ± 0.84
		(1.8–140)	(1.1–260)	(0.047–24)	(0.28–3.0)
	Modeled	2.0	0.3	0.07	0.8
		(0.2–5.6)	(0.1–0.8)	(0.01–0.2)	(0.2–2.3)
2012	Observed[22]	7.5 ± 6.1	5.3 ± 10	0.48 ± 1.1	0.34 ± 0.24
		(19–24)	(0.72–40)	(0.011–3.7)	(0.069–0.8)
	Modeled	1.9	0.3	0.06	0.7
		(0.2–5.3)	(0.1–0.8)	(0.01–0.2)	(0.1–2.3)
2013	Observed[22]	6.9 ± 6.9	11 ± 12	1.4 ± 1.1	0.12
		(0.74–14)	(0.58–21)	(0.068–2.1)	
	Modeled	1.8	0.3	0.06	0.7
		(0.2–4.9)	(0.1–0.8)	(0.01–0.2)	(0.1–2.3)

§ Concentrations reported at sampling sites Aurora and Egbert.

Marine fish (in pg/g_{lipid})

Year		BDE-47	BDE-99	BDE-153	BDE-209
1992	Observed[23#]	23	12	6.4	
		(6.1–64)	(3.1–32)	(n.d.–8.9)	
	Modeled	55	5.1	2.2	74
		(27–86)	(2.0–8.3)	(1.1–3.4)	(21–135)
1999-2000	Observed[24★]	5.0 ± 5.0	1.1 ± 0.9	0.3	
		(1.5–14)	(n.d.–2.4)	(n.d.–0.3)	
	Modeled	123	11	4.2	137
		(61–193)	(4.1–18)	(2.1–6.5)	(38–251)
2000	Observed[23#]	60	19	8.5	
		(27–160)	(7.7–46)	(3.4–26)	
	Modeled	135	12	4.4	148
		(67–212)	(4.5–20)	(2.2–6.9)	(41–271)
2003	Observed[25¶]	87 ± 62	18 ± 16	1.7 ± 1.4	
	Modeled	166	15	4.8	178
		(85–262)	(5.7–23)	(2.5–7.5)	(51–324)
2003	Observed[25¶]	32 ± 23	4.9 ± 4.0	0.5 ± 0.4	
	Modeled	166	15	4.8	178
		(85–262)	(5.7–23)	(2.5–7.5)	(51–324)
2005	Observed[25¶]	9.5 ± 3.8	2.2 ± 0.1	0.3 ± 0.2	

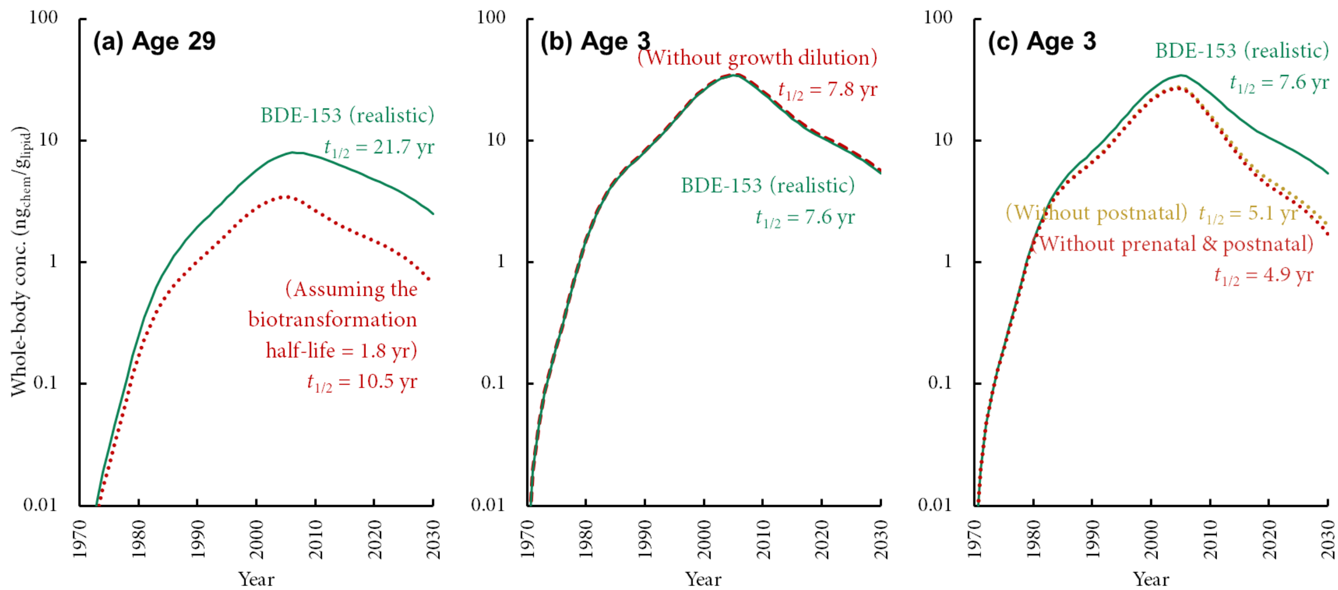
	Modeled	174	15	4.8	189
		(94–274)	(6.3–25)	(2.5–7.4)	(54–345)
2005	Observed[25¶]	73 ± 14	2.8 ± 2.3	0.4 ± 0.3	
	Modeled	174	15	4.8	189
		(94–274)	(6.3–25)	(2.5–7.4)	(54–345)
2007	Observed[25¶]	71 ± 2.2	1.4 ± 0.2	0.2 ± 0.0	
	Modeled	148	13	4.1	191
		(79–233)	(5.7–21)	(2.2–6.3)	(56–350)

Concentrations measured in Sole fish.

★ Concentrations measured in wild Salmons. Covered to lipid weight-based concentrations from wet weight-based concentrations reported in Easton et al.[24]

¶ Covered to lipid weight-based concentrations from wet weight-based concentrations reported in Ikonomou et al.[25] Data from sampling sites of West Coast Vancouver Island, Robson Bight Johnstone Strait, and Fraser River are used.

Figure S5. Additional calculations: Modeled within-age temporal trends in whole-body concentrations (Panel a) adults; assuming that BDE-153 has a shorter biotransformation half-life of 1.8 years; (Panel b) children; assuming that the rate of growth is zero; (Panel c) children; assuming no postnatal (lactation transfer) and prenatal (*in utero* trans-placental transfer) exposures.



References for the Supporting Information

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