SUPPORTING INFORMATION
For:
Occurrences and Fates of Hydroxylated Polybrominated Diphenyl Ethers in Marine

Sediments in Relation to Trophodynamics

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This supporting information provides information on (1) chemicals and standards, (2) the formulae and selected reaction monitoring conditions of the target analytes in LC-MS/MS analysis (Table S1); (3) Concentrations of individual MeO-PBDEs detected in marine sediments (Table S2); (4) the detection frequencies of OH-PBDEs for each species analyzed in Liaodong Bay (Table S3); (5) parameters of regressions for TMFs based on different substitution methods for concentrations below detection limits (Table S4); (6) sampling locations in Liaodong Bay (Figure S1); (7) range of trophic levels of organisms collected from Liaodong Bay (Figure S2); (8) relationship between the concentrations of 6-OH-BDE47 and 6-MeO-BDE47 in sediments (Figure S3).

Chemicals and Standards. Nine OH-PBDEs (2'-OH-6'-Cl-BDE7, 6-OH-BDE47, 3-OH-BDE47, 5-OH-BDE47, 2'-OH-BDE68, 4'-OH-BDE49, 2'-OH-6'-Cl-BDE68, 6-OH-BDE90 and 2-OH-BDE123), eleven PBDEs (BDE28, BDE47, BDE66, BDE77, BDE100, BDE99, BDE85, BDE154, BDE153, BDE138 and BDE183) and twelve MeO-PBDEs (6-MeO-BDE17, 4-MeO-BDE17, 2'-MeO-BDE68, 6-MeO-BDE47, 5-MeO-BDE47, 4'-MeO-BDE49, 5'-MeO-BDE100, 4'-MeO-BDE103, 4'-MeO-BDE99, 4'-MeO-BDE101, 6-MeO-BDE90, and 6-MeO-BDE85) were selected as target compounds. ^{13}C-PBDEs and eight MeO-PBDEs standards were obtained from Wellington Laboratories Inc. (Guelph, Ontario, Canada). 3-OH-BDE47, 5-OH-BDE47 and 2'-OH-BDE68 were obtained from AccuStandard (New Haven, Connecticut, USA). 6-MeO-BDE17, 4-MeO-BDE17, 6-MeO-BDE90, 6-MeO-BDE85, 6'-OH-BDE17 and the remaining seven OH-PBDEs were synthesized in the Department of Biology and Chemistry, City University of Hong Kong. Purities of all metabolites were >98%. Pesticide residue grade dichloromethane (DCM), n-hexane, methyl *tert*-butyl ether (MTBE), acetonitrile and methanol were obtained from OmniSolv (EM Science, Lawrence, KS, USA). Sodium sulfate, silica gel (60-100 mesh size), aluminum oxide (neutral, 150 mesh size), pyridine (anhydrous, 99.8%), methyl chloroformate (MCF), potassium hydroxide (KOH), hydrochloric acid (HCl), 2-propanol and dansyl chloride were purchased from Sigma-Aldrich (St. Louis, MO, USA).

TABLE S1. The Formulae and Selected Reaction Monitoring Conditions of the Target Analytes in LC-MS/MS Analysis.

Analyte	Formula	Transition monitored (m/z)	Collision energy (V)
2'-OH-6'-Cl-BDE7	$C_{12}H_7Br_2ClO_2$	612→171[a]	48
		614→171	48
6'-OH-BDE17	$C_{12}H_7Br_3O_2$	656→171[a]	48
		658→171	48
3-OH-BDE47	$C_{12}H_6Br_4O_2$	734→171[a]	48
5-OH-BDE47		736→171	48
6-OH-BDE47			48
4'-OH-BDE49			48
2'-OH-BDE68			48
2'-OH-6'-Cl-BDE68	$C_{12}H_5Br_4ClO_2$	770→171[a]	48
		772→171	48
6-OH-BDE90	$C_{12}H_5Br_5O_2$	814→171[a]	48
2-OH-BDE123		816→171	48

a MRM transition used for quantification

TABLE S2. Concentrations of individual MeO-PBDEs detected in 23 Marine Sediments (pg/g dry weight) from Liaodong Bay, North China.

Chemicals	Concentration
6-MeO-BDE17	2.0±1.1
4-MeO-BDE17	2.7±1.8
2'-MeO-BDE68	5.5±1.9
6-MeO-BDE47	15±1.6
5-MeO-BDE47	2.1±1.2
4'-MeO-BDE101	2.2±1.6

TABLE S3. Detection Frequencies of OH-PBDEs for Each Species Analyzed in Liaodong Bay.

Species	SED	RP	MVR	RV	ESME	UM	LH	SH	PP	TK	SN	CS	PI	LJ	LR	LC
6-OHBDE-47	100	100	100	100	0	67	67	0	100	100	100	0	100	0	57	63
2-OHBDE-68	26	100	33	100	0	67	67	0	0	0	100	0	67	0	14	38
5-OHBDE-47	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	25
4-OHBDE-49	0	0	33	0	0	0	0	0	0	0	0	0	0	33	0	0
3-OHBDE-47	0	0	0	0	0	0	67	0	0	0	0	0	0	33	14	19

SED=sediment; RP= short-necked clam (*Ruditapes philippinarum*); MVR = mactra quadrangularis (*Mactra veneriformis,Reeue*); RV= rock shell (*Rapana venosa*); ESME = Chinese mitten-handed crab (*Eriocheir sinensis H. Milne-Eswards*); UM = mole cricket (*Upogebia major*(*de Haan*)); LH = redeye mullet (*Liza haematocheila*); SH = goby (*Synechogobius hasta*); PP = small yellow croaker (*Pseudosciaena polyactis*); TK = China anchovy (*Thrissa kammalensis*); SN = Japanese spanish mackerel (*Scomberomrus niphonius*); CS = half-smooth tongue-sole (*Cynoglossus semilaevis*); PI = flathead fish (*Platycephalus indicus*); LJ = black spotfed bass (*Lateolabrax japonicas*); LR= black-headed Gulls (*Larus ridibundus*); LC= black-tailed Gull (*Larus crassirostris*) .

TABLE S4. Parameters of Regressions for TMFs Based on Different Substitution Methods for Concentrations Below Detection Limits.

		ND treatment	mean	S.D.	lower-upper 95% CI	*p*-value
6-OH-BDE 47	slope	0.5MDL	-0.682	0.225	-1.168 to -0.195	0.01
		Original	-0.587	0.206	-1.032 to-0.142	0.01
		ROS	-0.602	0.184	-0.998 to -0.205	0.006
	intercept	0.5MDL	4.518	0.657	3.099 to 5.937	<0.001
		Original	4.340	0.601	3.042 to 5.637	<0.001
		ROS	4.462	0.535	3.305 to 5.618	<0.001
2-OH-BDE 68	slope	0.5MDL	-0.815	0.286	-1.434 to -0.197	0.01
		Original	-0.595	0.240	-1.113 to -0.077	0.03
		ROS	-0.590	0.233	-1.094 to -0.087	0.02
	intercept	0.5MDL	4.466	0.835	2.662 to 6.269	<0.001
		Original	3.986	0.699	2.475 to 5.496	<0.001
		ROS	3.982	0.680	2.514 to 5.451	<0.001

FIGURE S1. Study area and sampling locations. ▲: sediment samples; ★: fish samples; ★: bird samples.
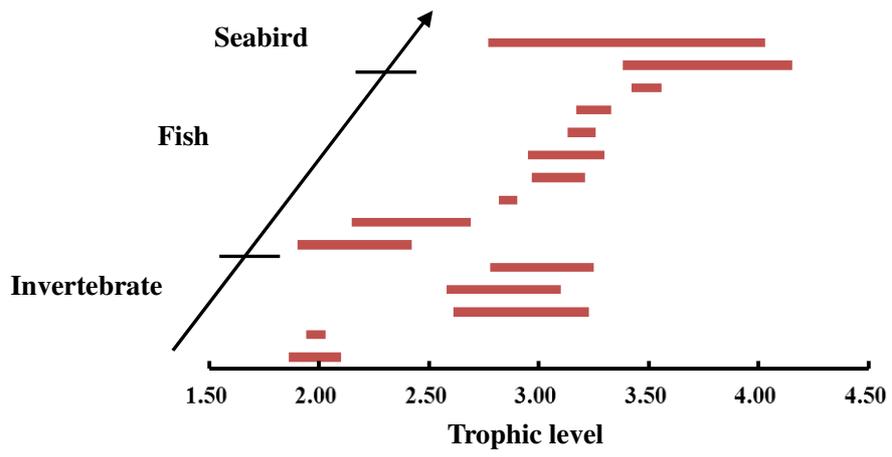
FIGURE S2: Range of trophic levels of organisms collected from Liaodong Bay (data are from reference (*1*).
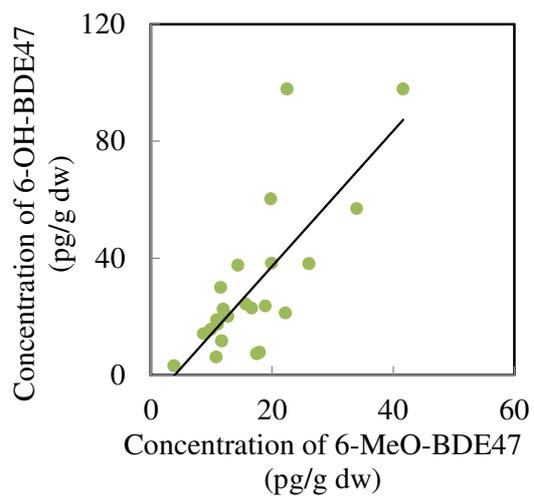
FIGURE S3: Relationships between the concentrations of 6-OH-BDE47 and the concentration of 6-MeO-BDE47 in sediments (r^2=0.571, p<0.001).

Literature Cited

(1) Zhang, K.; Wan, Y.; An, L.H.; Hu, J.Y. Trophodynamics of polybrominated diphenyl ethers and methoxylated polybrominated diphenyl ethers in a marine food web. *Environ Toxicol Chem* **2010**, *29*, 2792-2799.