Supporting Information

Decabrominated diphenyl ethers (BDE-209) in Chinese and global air: Levels, gas/particle partitioning, and long-range transport: Is long range transport of BDE-209 really governed by the movement of particles?

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Contents

S1 Monitoring

S1.1 China POPs Soil and Air Monitoring Program, Phase 2 (China POPs SAMP-II)

Under the China POPs SAMP-II, focused on Chinese urban regions, there were 15 sampling sites including 11 urban centers (Beijing, Harbin, Dalian, Xi'an, Nanchang, Kunming, Chengdu, Lhasa, Guangzhou, Lanzhou, Shihezi), 1 suburban place (Shanghai (Lingang)), and 3 background/rural locations (Waliguan, Wudalianchi, and Xuancheng) (see **Figure S1**) (Yang et al. 2013). The air sampling had run 24 hours every week for 2 consecutive years started in August, 2008 and ended in July, 2010, under which the samples had been obtained using HiVol air samplers, and all the samples were treated and analyzed in the IJRC-PTS laboratories for minimizing the uncertainty. Some results have been published (Li et al., 2015a; b; Li and Jia, 2014; Yang et al. 2012; 2013; Ma et al. 2010; 2011a; b; 2013).

It is worthwhile to point out that the air monitoring at the three background/rural sites were a part of the International Polar Year (IPY) Project #327, "*INterContinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic* (INCATPA)" (Yang et al. 2013).

China POPs SAMP-II Publications

Li, Y. F.; Ma W. L.; Yang M. 2015a, Prediction of gas/particle partitioning of polybrominated diphenyl ethers (PBDEs) in global air: a theoretical study. *Atmos. Chem. Phys.*, 15, 1669-1681.

Li, Wen-Long, Hong Qi, Wan-Li Ma, Li-Yan Liu, Zhi Zhang, Mohammed .A. Mohammed, Wei-Wei Song, Zifeng Zhang, Yi-Fan Li, 2015b, Brominated flame retardants in Chinese air before and after the phase out of polybrominated diphenyl ethers, Atmospheric Environment, 117, 156–161.

Li Y. F. and Jia, H. L.: 2014, Prediction of Gas/Particle Partition Quotients of Polybrominated Diphenyl Ethers (PBDEs) in north temperate zone air: An empirical approach, Ecotoxic.

Environ. Safety, 108, 65-71.

Ma W; Li YF, Qi H, Sun D, Liu L, Wang D, 2010, Seasonal variations of sources of polycyclic aromatic hydrocarbons (PAHs) to a northeastern urban city, China. *Chemosphere* 79: 441-447.

Ma W, Liu L, Qi H, Sun D, Shen J, Wang D, Li YF, 2011a, Dechlorane plus in multimedia in northeastern Chinese urban region. *Environ Int* 37: 66-70.

Ma W, Sun D, Shen W, Yang M, Qi H, Liu L, Shen J, Li YF, 2011b, Atmospheric concentrations, sources and Gas/Particle partitioning of PAHs in Beijing after the 29th Olympic Games. *Environ Pollut* 159: 1794-1801.

Ma W, Qi H, Baidron S, Liu L, Yang M, Li YF, 2013, Implications for long-range atmospheric transport of polycyclic aromatic hydrocarbons in Lhasa, China. *Environ Sci Pollut Res* 20: 5525-5533.

Yang, M., Qi, H., Jia, H., Ren, N., Ding, Y. S., Ma, W. L., Li, Y. F.. 2013, Polybrominated Diphenyl Ethers (PBDEs) in Air across China: Levels, Compositions, and Gas-Particle Partitioning. Environmental science & technology, 47: 8978-8984

Yang, M.; Jia, H.-L.; Ma, W.-L.; Qi, H.; Cui, S.; Li, Y.-F. 2012, Levels, compositions, and gas-particle partitioning of polybrominated diphenyl ethers and dechlorane plus in air in a Chinese northeastern city. *Atmos. Environ. 55*, 73-79.

S1.2 PBDE study under China POPs SAMP-II

In this program, PBDEs in air at 15 sites continuously monitored for 2 year from August, 2008 and ended in July, 2010, and in this study, the data for one year from September of 2008 to August of 2009 were presented (Yang et al., 2013).

Concentration data of BDE-209 in gas phase at Guangzhou site is not available due to the missing of the gas samples at this site.

S2 Experiment Section

The major portion of this section is from Yang et al., (2013).

S2.1 Sample collection

Air samples with both gaseous and particulate phases were simultaneously collected using high-volume air samplers for 24 hours every week from September 2008 to August 2009 at 15 sampling sites (See **Table S2**, SI, for detailed information). There are two different kinds of air samplers were used in the sampling events under SAMP-II. One is high-volume air sampler KB-1000 (Kingstar Electronic Technology Co.,Ltd. Qiangdao) used at the 11 urban and 1 suburban sites, and another is high-volume air sampler TE-1000 (Tisch company, USA) used at the three background/rural sites. For the KB-1000, an air flow rate was set as 0.8 $m^3 \cdot min^{-1}$, the duration of sampling was a consecutive 24 h on a weekly base and daily sampling volumes were approximately 1150 $m^3 \cdot d^{-1}$. A layer of GFF glass fiber filters (20 cm×25 cm) and two layers of polyurethane foam (PUF) plugs (length 5.0 cm, diameter 9.5 cm) were set to collect air samples with particulate and gaseous phases, respectively. For the TE-1000, the air flow rate was set as 0.2 $m^3 \cdot min^{-1}$, the duration of sampling was also consecutive 24 h on a weekly base and daily sampling volumes were approximately 300 $m^3 \cdot d^{-1}$. Each sample composes of one circular GFF glass fiber filters (diameter 4 inches) and one polyurethane foam (PUF) plug (length 3 inches, diameter 7/8 inches) to trap PBDEs in particulate and gaseous phases, respectively. Prior to sampling, GFFs were baked at 450^{o}C for 7 h and PUF plugs were Soxhlet extracted for 24 h with acetone and for another 24 h with hexane to remove any organic contaminant. After sampling, exposed GFFs were wrapped with prebaked aluminum foils and sealed with double layers of polyethylene bags, and PUFs were placed in solvent rinsed aluminum shipping containers sealed with polytetrafluoroethylene tape, and sent to IJRC-PTS laboratories for further treatment and analysis. All sampled GFFs and PUFs had been stored at -20 ^{o}C until extraction.

S2.2 Chemicals, reagents and sample preparation

All solvents used were of pesticide analysis grade purity (J.T. Baker, Phillipsburg, NJ). Silica gel (100-200 mesh) was purchased from Merck (Merck, Germany). Standards for 12

PBDE congeners (Tri-BDE: BDE-17, -28; Tetra-BDE: BDE-47, -66; Penta-BDE: BDE-85, -99, -100; Hexa-BDE: BDE-138, -153, -154; Hepta-BDE: BDE-183; and Deca-BDE: BDE-209) were purchased from Accustandard Inc. (New Haven, CT). BDE-71, used as internal standards, was also purchased from Accustandard Inc. (New Haven, CT).

Details of air samples extraction and clean-up procedure can be found in our previous study (Yang et al., 2013). In brief, the PUF plugs and GFF were spiked with PCB-155 as a recovery standard, and were Soxhlet exacted with a mixture of acetone: hexane (1:1, v/v) and dichloromethane for 24 h, respectively. The extracts were concentrated by rotory-evaporationand cleaned up using active silica gel columns (0.4-m length × 10-mm i.d.) with 70 mL of dichloromethane: hexane (1:1, v/v). After concentrated to 1 mL by a gentle nitrogen flow (purity 99.999%), a known amount of BDE-71 was added as internal standard.

S2.3 Instrumental analysis

Tri- to hepta-PBDEs analysis were performed with a Thermo Trace gas chromatograph coupled with a Thermo PolarisQ mass spectrometer (GC/MS) using an electron capture negative ionization (ECNI) ion source in the selected ion monitoring (SIM) mode. A 30-m HP-5MS capillary column (250 μm i.d., 0.25μm film thickness; J&W Scientific, Folsom, CA, USA) was used for the separation of these chemicals. The ion source and the GC to MS transfer line were held at 200 ºC and 280 ºC, respectively. The oven temperature program was initiated at 110 ºC (held for 1 min), and increased to 250 ºC at rate of 20 ºC min^{-1} (held for 5min), and finally increased to 300 ºC at 25 ºC min^{-1} (held for 8 min). Methane was used as a chemical ionization moderating gas, and helium as the carrier gas at flow rate of 1mL·min^{-1}. m/z 79 and 81 were monitored for tri- to hepta- PBDEs.

Particle-bound BDE-209 was determined by Varian CP-3800 gas chromatograph coupled with an electron capture detector (GC/ECD), equipped using a 7 m (250 μm i.d., 0.25μm film thickness; J&W Scientific, Folsom, CA, USA) HP-5 MS capillary column. The column oven temperature program was: 100 ºC held 2 min, then 6 ºC to 210 ºC held 5 min, 7 ºC to 300 ºC and held 10 min; injector: 250 ºC; detector: 310 ºC. Nitrogen was used as the carrier gas at constant flow of 2.0 mL·min^{-1}.

In this study, we re-measured BDE-209 in PUF using Agilent 6890GC-5975MS in the

electron capture negative ionization mode. A DB-5 MS capillary chromatographic column (15 m × 0.25 mm × 0.10 μm, J&W Scientific) was used to separate BDE-209 from other chemicals. The following temperature program was used, 110 °C for 0.5 min, 5 °C/min to 220°C, 20°C/min to 310 °C, and held for 15 min. Furthermore, the temperatures were 106 °C, 200 °C, and 270 °C for the quadrupole, ion source, and interface, respectively. The high-purity helium was used as the carrier gas with the flow rates of 1.4 mL/min and 1.7 mL/min for the HP5-MS column and DB5-MS column, respectively. For the identification, the peaks for samples were selected on the basis of retention time with standards. For the quantification, the five-point calibration curves (with the ranges of 50-1000 ng/mL for BDE 209, 1-25 ng/mL for other PBDEs, 8-400 ng/mL for DBDPE, and 1-50 ng/mL for other alternative BFRs) were applied, which were established with the authentic standard/internal standard concentration ratios and corresponding peak area ratios.

S2.4 Quality assurance/quality control

The identification of all target compounds were based on the GC retention time matched those of the standard compounds within ± 0.1 min. Filed blank and laboratory blank were added for every 10 samples. No target compounds were found in the blanks. Recoveries of PCB-155 were 88 ± 10% (mean ± one standard deviation). The final reported concentrations were not recovery corrected.

A spike test was performed to examine if PBDEs can be extracted effectively with the applied method. Average recoveries of spiked 12 PBDEs standards in clean PUF and GFF were 71% ± 5% and 78% ± 6%, respectively. Instrument detection limits (IDL) for PBDEs ranged from 0.02 to 0.72 $pg \cdot m^{-3}$ (**Table S3**).

Breakthrough sampling experiments were taken once a month in order to investigate whether there is a breakthrough of the target substance. During the breakthrough sampling experiments, the second 5cm PUF using in KB-1000 was divided into a 3cm and a 2cm PUFs, the 3 inch PUFs using in TE-1000 was replaced with a 2-inch and a 1 inch PUFs. After sampling, the 2 cm and 1 inch PUFs were treated and analyzed as ordinary samples, and the results showed that the concentrations of PBDEs in 2 cm and 1 inch PUFs were less than 1/5 of the total PUF content, indicating that there was not any breakthrough during the sampling time (See the figure below).

Figures

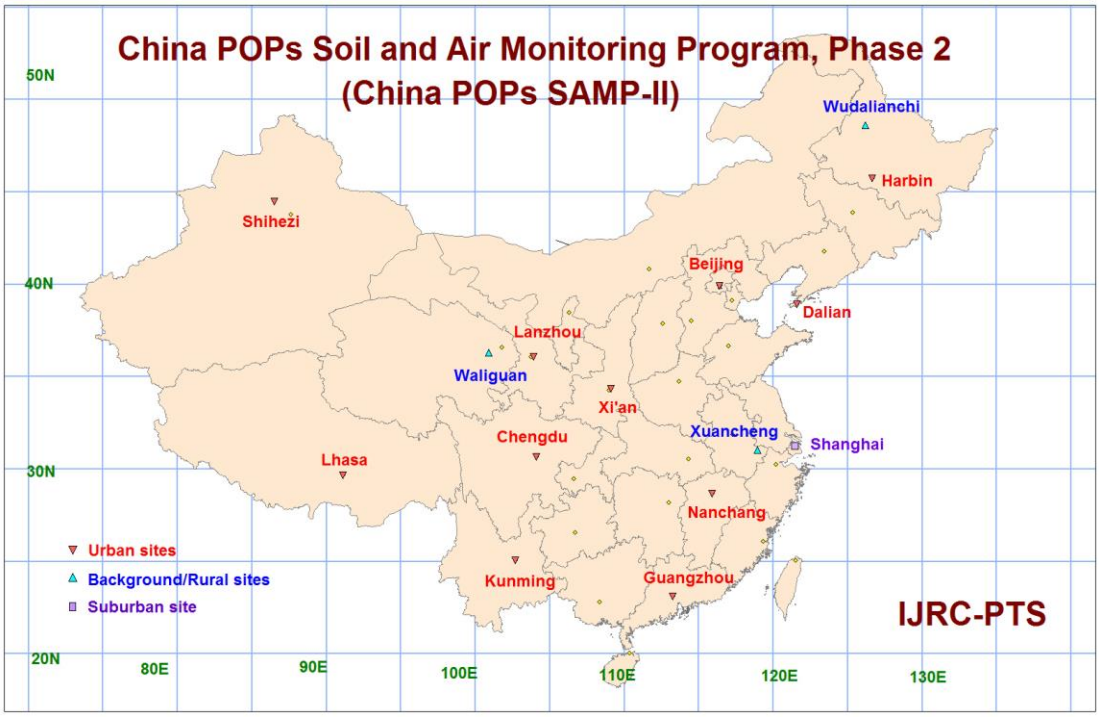


Figure S1.

Locations of sampling sites under China POPs Soil and Air Monitoring Program, Phase 2 (China POPs SAMP-II, 2008-2010). The numbers from 80 to 130 at the bottom are longitude in °E and the numbers from 20 to 50 at the left are latitude in °N (Yang et al., 2013).

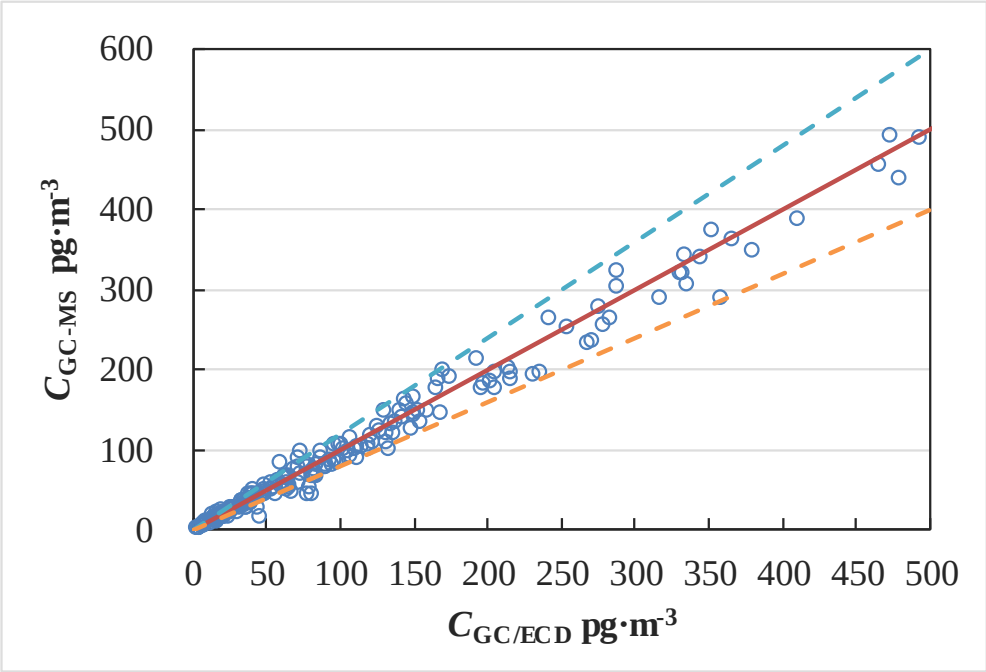


Figure S2.

The comparison between the concentrations of BDE-209 in particle phase detected by GC/ECD ($C_{GC/ECD}$) and GC/MS ($C_{GC/MS}$). The blue circles represent the concentrations of BDE209 detected by GC/ECD and GC-MS for the same samples. The red solid line shows the 1:1 agreement, the blue and orange dashed line show a deviation of ± 20%.

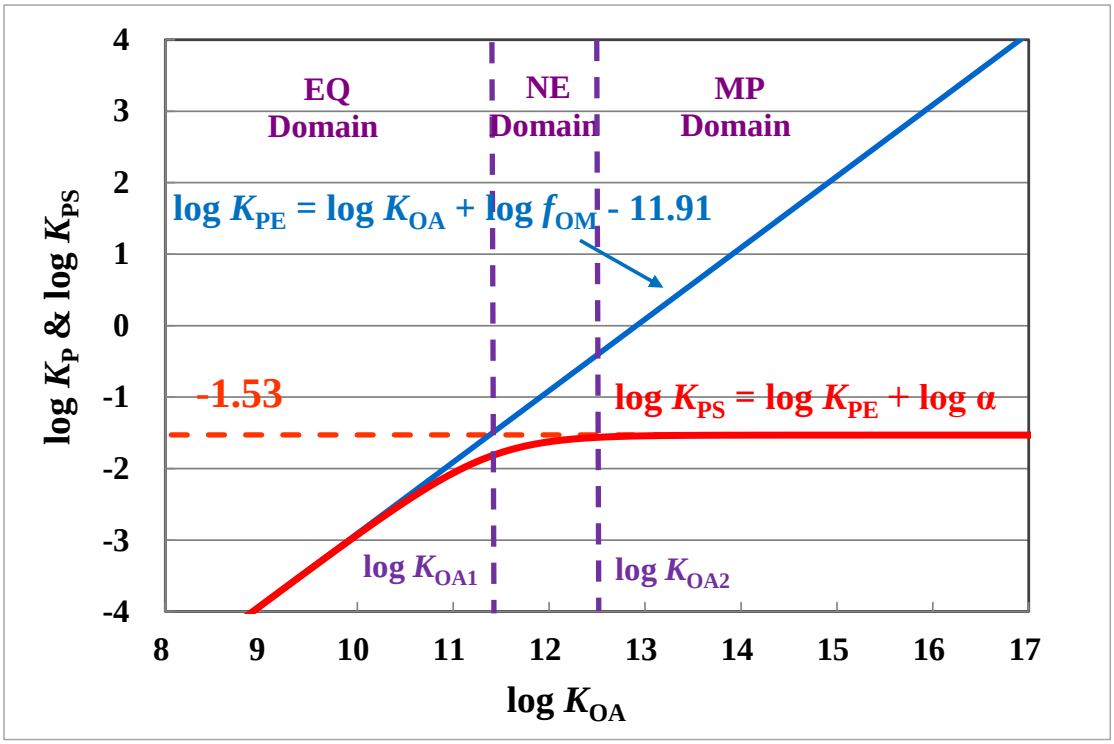


Figure S3.

The G/P partition coefficients of PBDEs as functions of log K_{OA} calculated by two equations. First one is Harner-Bidleman equation (log K_{PE}) at equilibrium (Eq. 3), presented by the dark blue straight line, and the second one is Li-Ma-Yang equation (log K_{PS}) at steady state (Eq. 5), presented by the red curve in the figure. Two threshold values of log K_{OA} (log K_{OA1} and log K_{OA2}, represented by two vertical purple dashed lines) divide the range of log K_{OA} into three domains: the equilibrium (EQ) domain, the nonequilibrium (NE) domain, and the maximum partition (MP) domain (Li et al., 2015a).

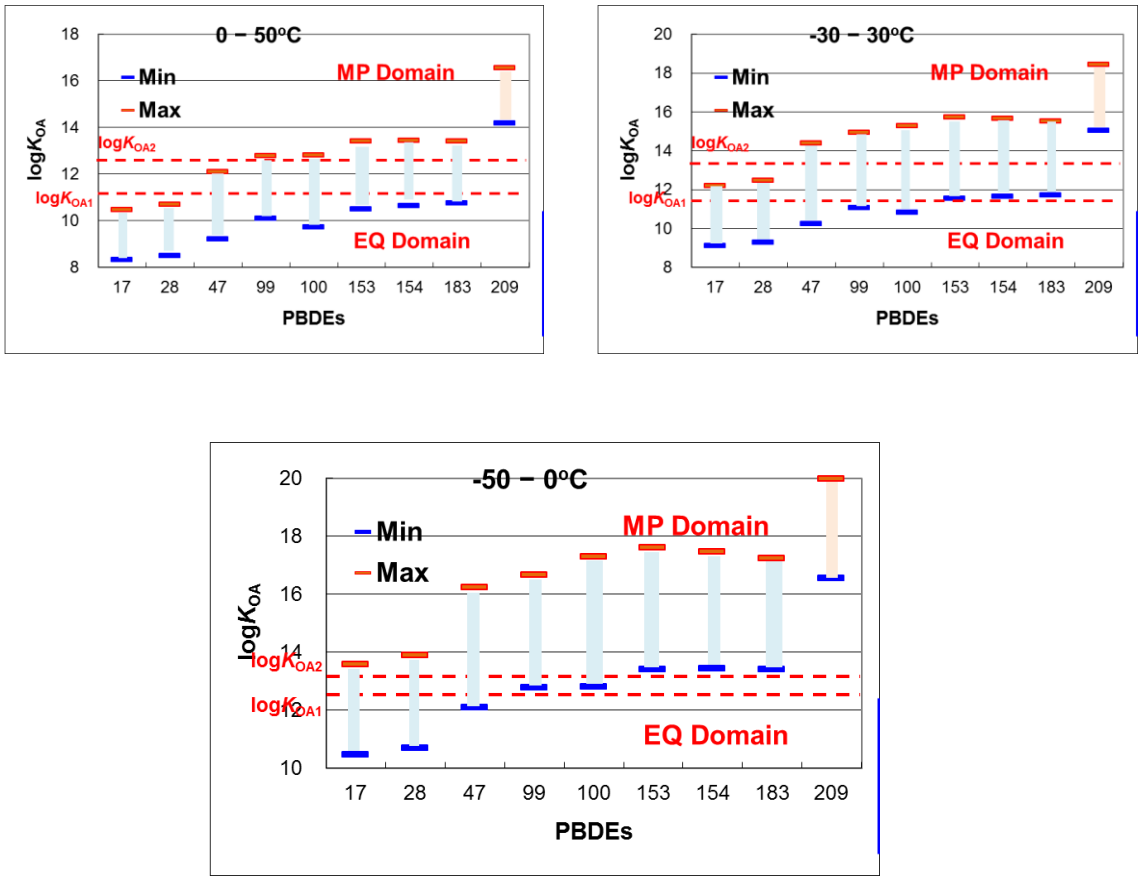
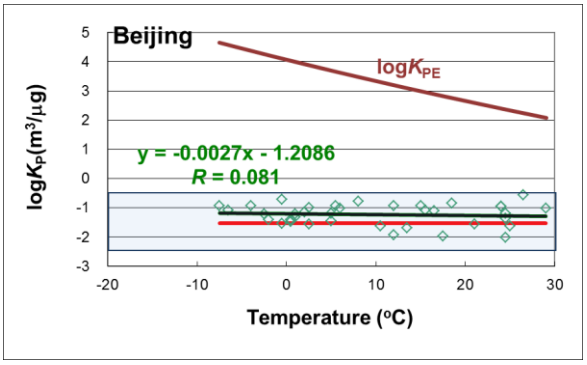
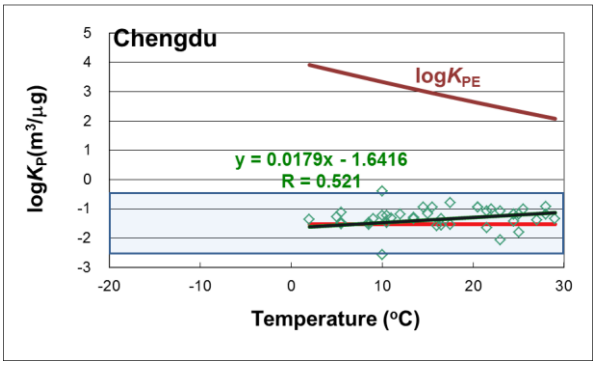
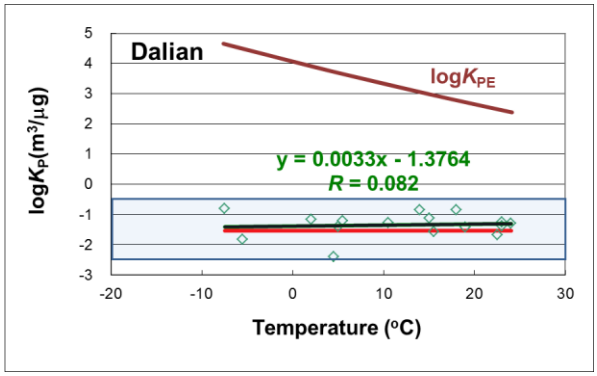


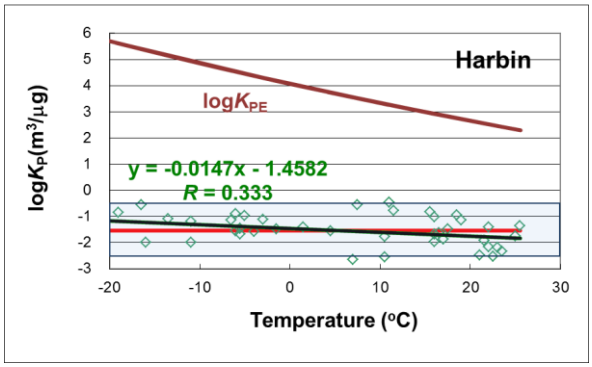
Figure S4.

The range of log K_{OA} for 9 PBDE congeners (BDE-17, -28, -47, -99, -100, -153, -154, -183, and -209) in different ranges of ambient temperature (-50°C − 0°C, -30°C − +30°C, and 0°C − +50°C). It is obvious that only BDE-209 is within the MP domain in all these temperature ranges.


Beijing
logKPE
y = -0.0027x - 1.2086
R = 0.081
logKP(m3/μg)
Temperature (°C)


Chengdu
logKPE
y = 0.0179x - 1.6416
R = 0.521
logKP(m3/μg)
Temperature (°C)


Dalian
logKPE
y = 0.0033x - 1.3764
R = 0.082
logKP(m3/μg)
Temperature (°C)


Harbin
logKPE
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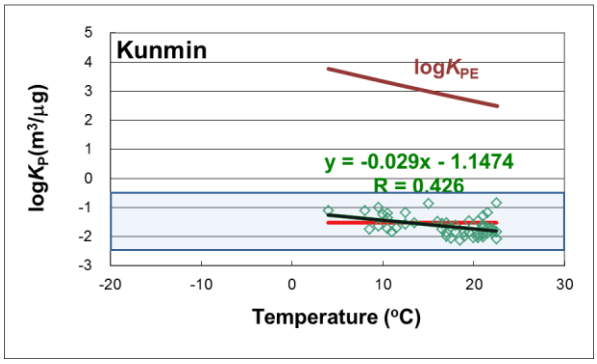
R = 0.333
logKP(m3/μg)
Temperature (°C)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logKPE
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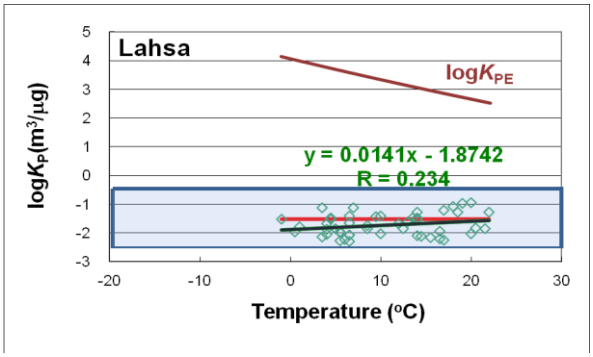
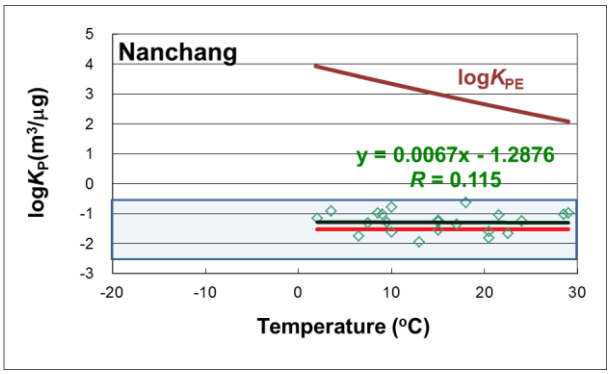
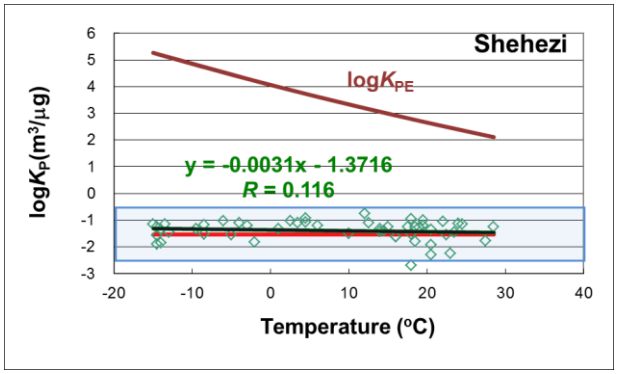
R = 0.426
logKP(m3/μg)
Temperature (°C)


Lahsa
logKPE
y = 0.0141x - 1.8742
R = 0.234
logKP(m3/μg)
Temperature (°C)


Nanchang
logKPE
y = 0.0067x - 1.2876
R = 0.115
logKP(m3/μg)
Temperature (°C)


Shehezi
logKPE
y = -0.0031x - 1.3716
R = 0.116
logKP(m3/μg)
Temperature (°C)

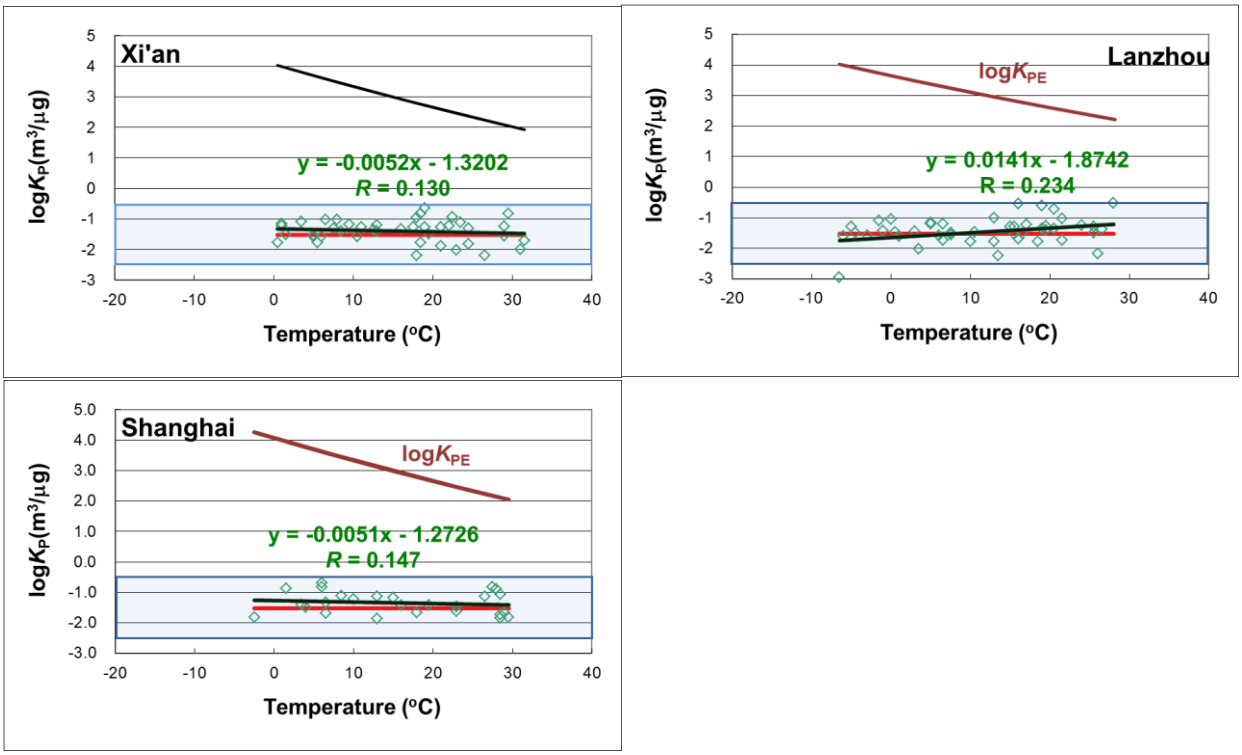


Figure S5a.

The values of log K_P of BDE-209 in different cities. The diamond marks represent the values of log K_{PM} of BDE-209 from monitoring data, the solid green lines are regression data of log K_{PM} data, the purple lines are the predicted data log K_{PE} by Equation (3), and the red lines indicate the maximum partition value of -1.53 $m^3/\mu g$ (log K_{PSM}) predicted by Equation (5).

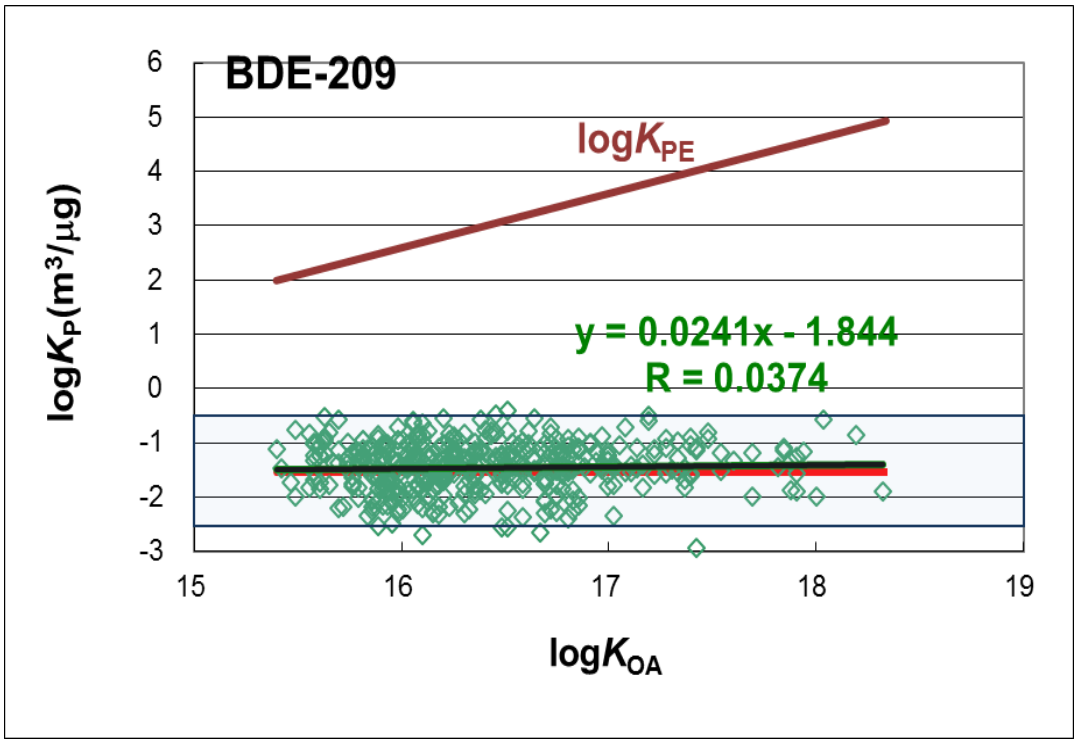


Figure S5b.

log K_{PM} of BDE-209 as function of log K_{OA} from China_POPs SAMP-II. The diamond marks represent the values of log K_{PM} of BDE-209 from monitoring data, the solid green lines are regression data of log K_{PM} data, the purple lines are the predicted data log K_{PE} by Equation (3), and the red lines indicate the maximum partition value of -1.53 $m^3/\mu g$ (log K_{PSM}) predicted by Equation (5).

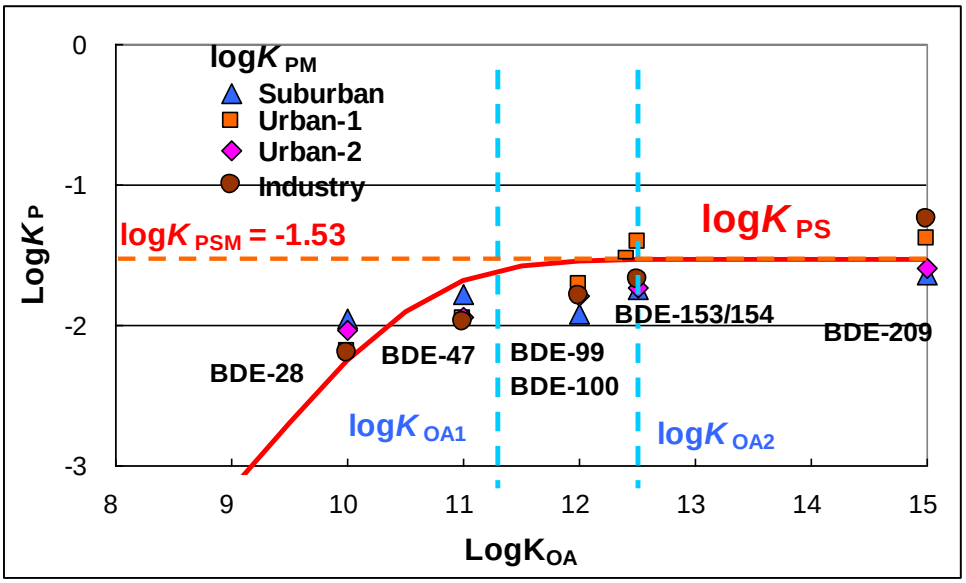


Figure S6.

Variation of log K_{PM} as functions of log K_{OA} for BDE-209 and other 6 PBDE congeners (BDE-28, -47, -99, -100, -153, and -154) at four sites (1 suburban, 2 urban, and 1 industrial) in Izmir, Turkey in summer and winter in 2004-2005. The monitoring data are from (Cetin and Odabasi, 2008). The four different colors indicate the different site types. The solid red line indicates log K_{PS} calculated using Equation (5). Two threshold values of log K_{OA} and the maximum values of log K_{PSM} are also shown. The concentration of *TSP* in the four sites were (in μg/m^3), suburban: 50.5; urban-1: 114; urban-2: 53.5; and industrial: 99.5.

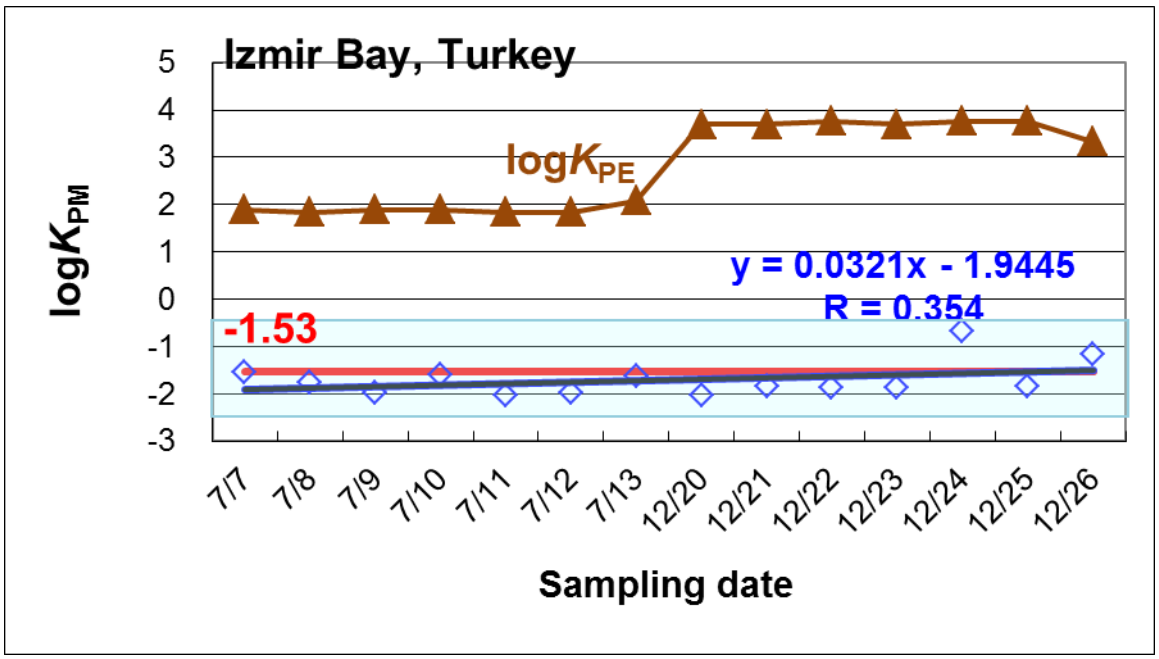


Figure S7

The values of log K_P of BDE-209 versus sampling date in Izmir Bay, Turkey in 2005. The diamond marks represent the values of log K_{PM} of BDE-209 from monitoring data, the solid green line represents regression data of log K_{PM} data, the dark purple line with triangles is the predicted data of log K_{PE} by Equation (3), and the red line indicates the maximum partition value of -1.53 $m^3/\mu g$ (log K_{PSM}) predicted by Equation (5). The monitoring data are from (Cetin and Odabasi, 2007). The values of *TSP* were assumed 30 $\mu g/m^3$ in summer and 50 $\mu g/m^3$ in winter for the study.

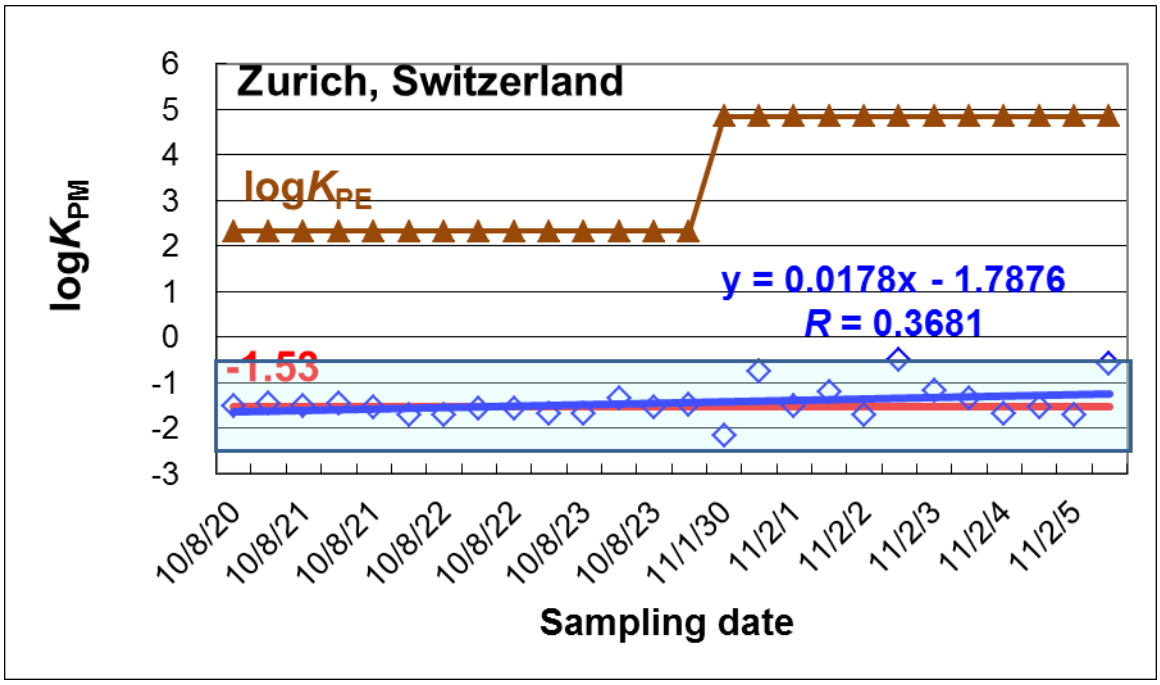


Figure S8.

The values of log K_{PM} of BDE-209 versus sampling date in Zurich, Switzerland in 2010. The diamond marks represent the values of log K_{PM} of BDE-209 from monitoring data, the solid green line represents regression data of log K_{PM} data, the dark purple line with triangles is the predicted data of log K_{PE} by Equation (3), and the red line indicates the maximum partition value of -1.53 $m^3/\mu g$ (log K_{PSM}) predicted by Equation (5). The monitoring data are from Bogdal et al., (2014). The values of *TSP* in this city were assumed 30 $\mu g/m^3$ in summer and 50 $\mu g/m^3$ in winter for the study.

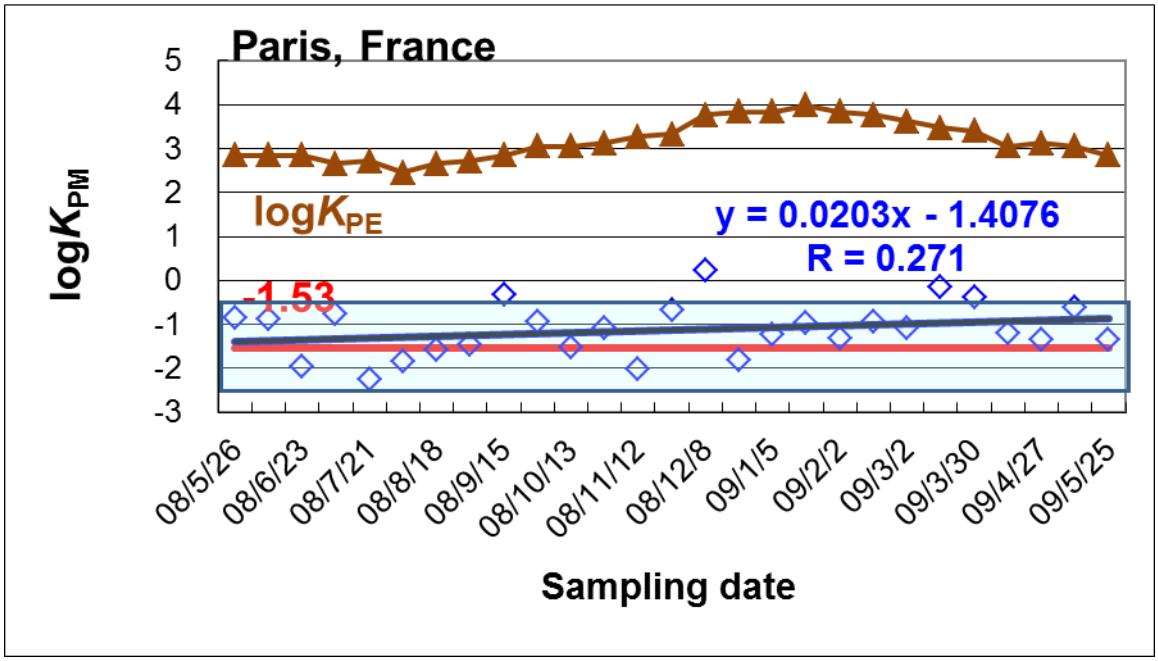


Figure S9.

Variation of log K_{PM} for BDE-209 versus sampling date in downtown Paris, France. The diamond marks represent the values of log K_{PM} of BDE-209 from monitoring data, the solid green line represents regression data of log K_{PM} data, the line with triangles is the predicted data of log K_{PE} by Equation (3), and the red line indicates the maximum partition value of -1.53 $m^3/\mu g$ (log K_{PSM}) predicted by Equation (5). The monitoring data are from Tlili et al., (2011). The values of *TSP* in this city were assumed 50 $\mu g/m^3$ in summer and 100 $\mu g/m^3$ in winter for the study.

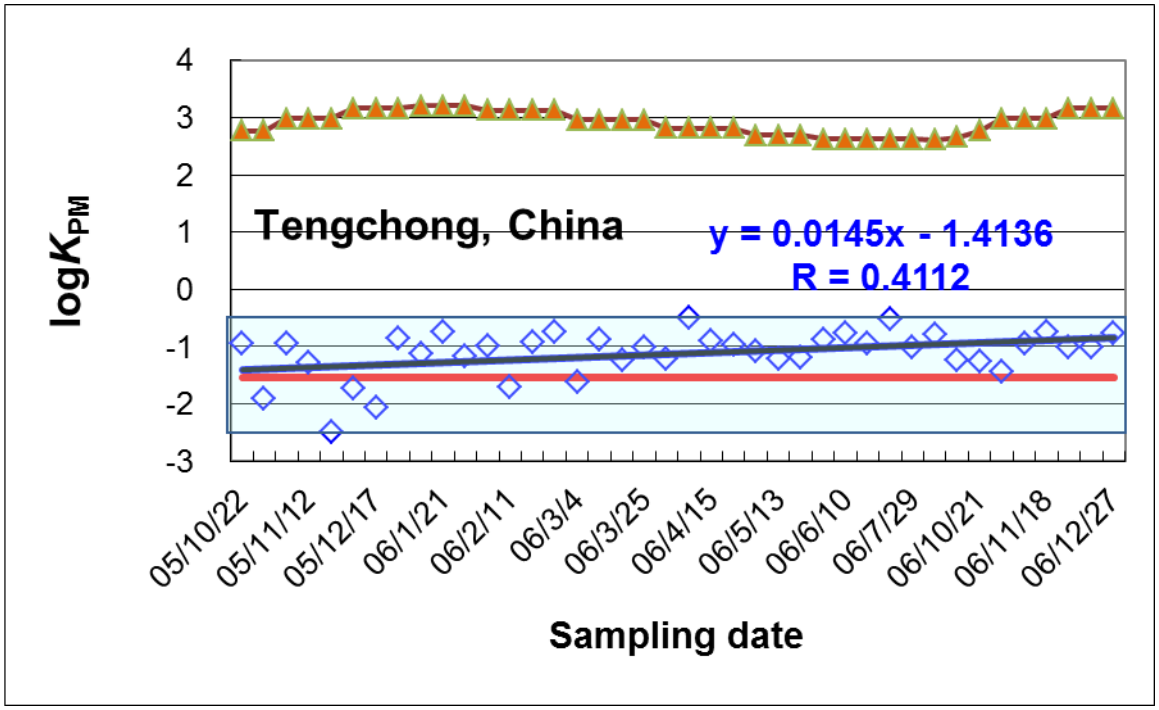


Figure S10..

Variation of log K_{PM} for BDE-209 versus sampling date at rural sites in Yunnan, China in 2006. The diamond marks represent the values of log K_{PM} of BDE-209 from monitoring data, the solid green line represents regression data of log K_{PM} data, the line with triangles is the predicted data of log K_{PE} by Equation (3), and the red line indicates the maximum partition value of -1.53 $m^3/\mu g$ (log K_{PSM}) predicted by Equation (5). The monitoring data are from Xu et al., (2011). The values of *TSP* in these sites were assumed as 100 $\mu g/m^3$ in whole year.

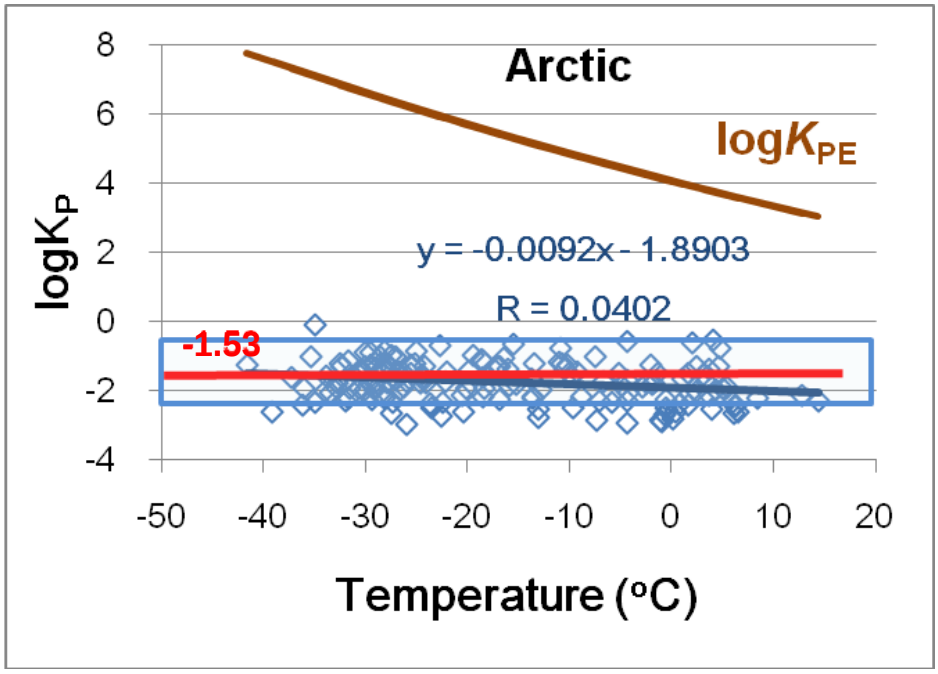


Figure S11.

Variation of log K_{PM} for BDE-209 versus temperature at Alert, Canada from 2006 to 2012 (Xiao et al., 2012; NCP 2013) (*TSP* = 10 μg·m^{-3} was assumed). The diamond marks represent the values of log K_{PM} of BDE-209 from monitoring data, the dark purple line with triangles is the predicted data of log K_{PE} by Equation (3), and the red line indicates the maximum partition value of -1.53 m^3/μg (log K_{PSM}) predicted by Equation (5).

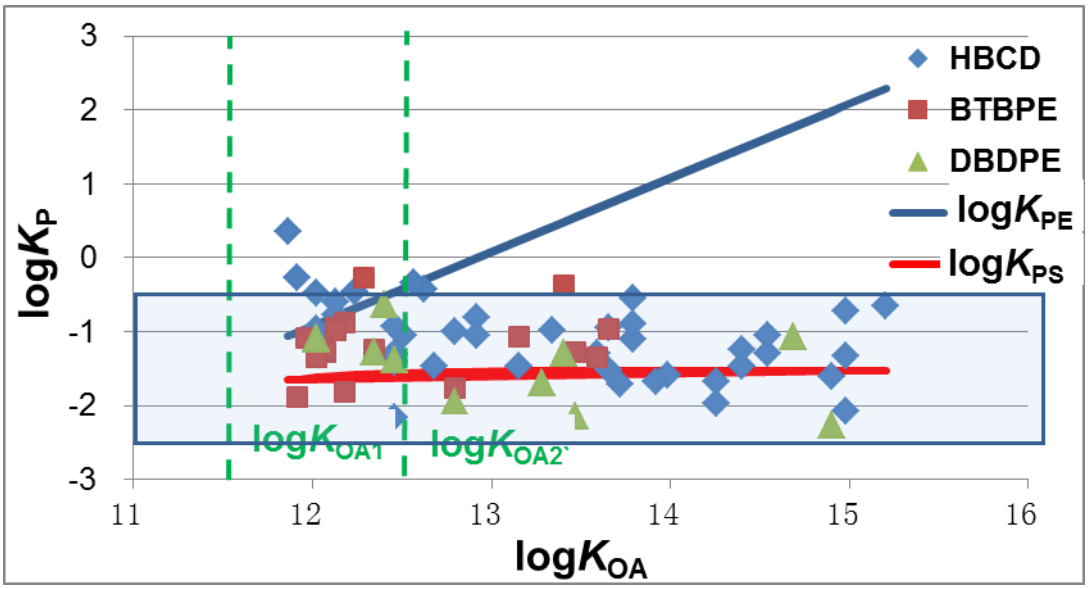
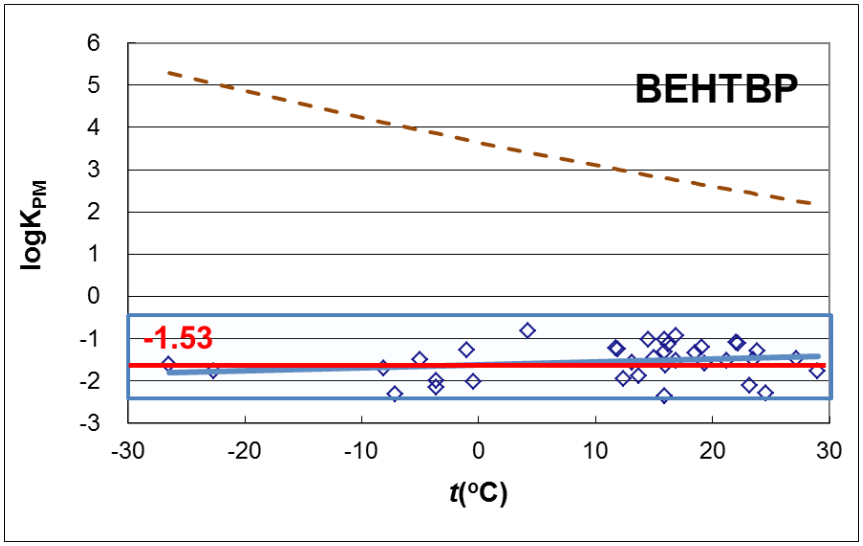
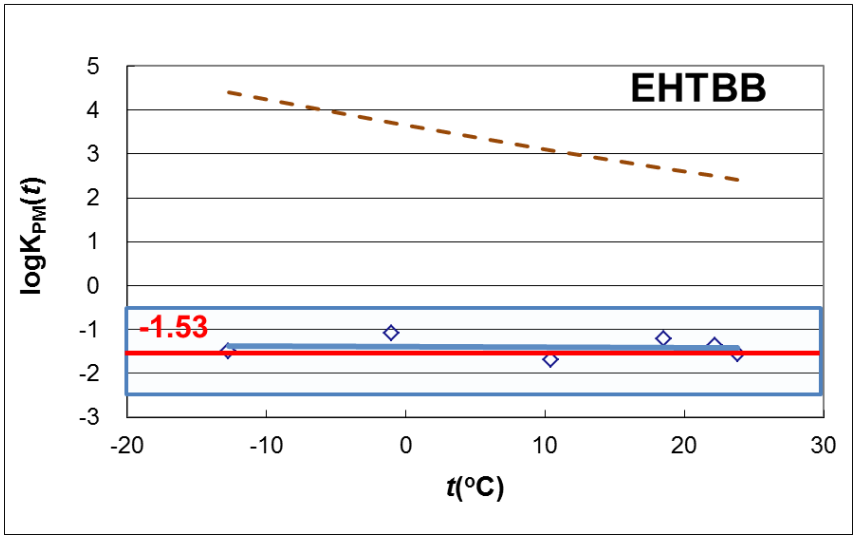


Figure S12.

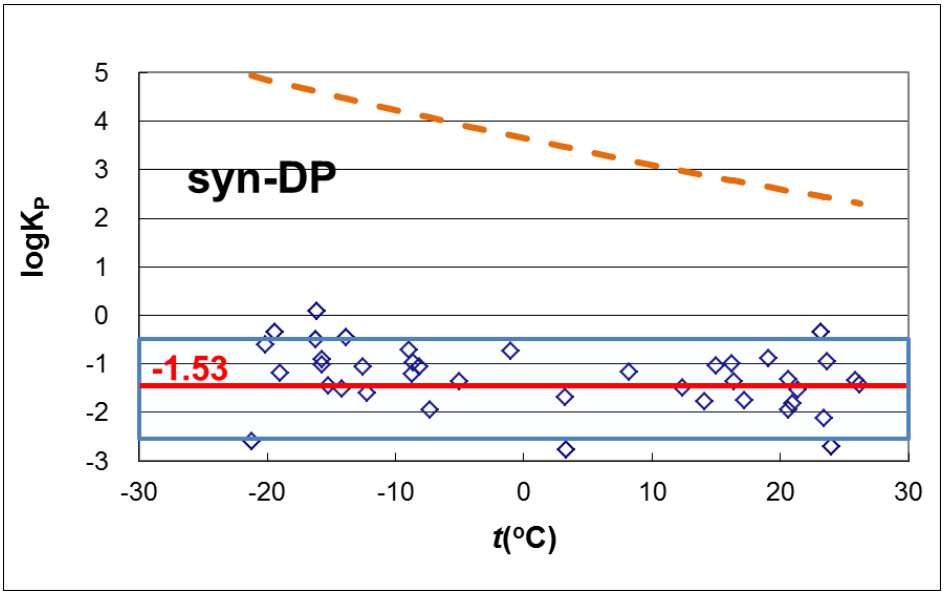
Partition quotients (log K_{PM}, log K_{PE}, and log K_{PS}) versus log K_{OA} for high brominated HBCD, BTBPE**,** and DBDPE. The values of log K_{PE} and log K_{PS} were calculated using the log K_{OA} values of BDE-183 (Li, et al, 2016a).


BEHTBP
-1.53
logK_PM
t(°C)
6
5
4
3
2
1
0
-1
-2
-3
-30
-20
-10
0
10
20
30

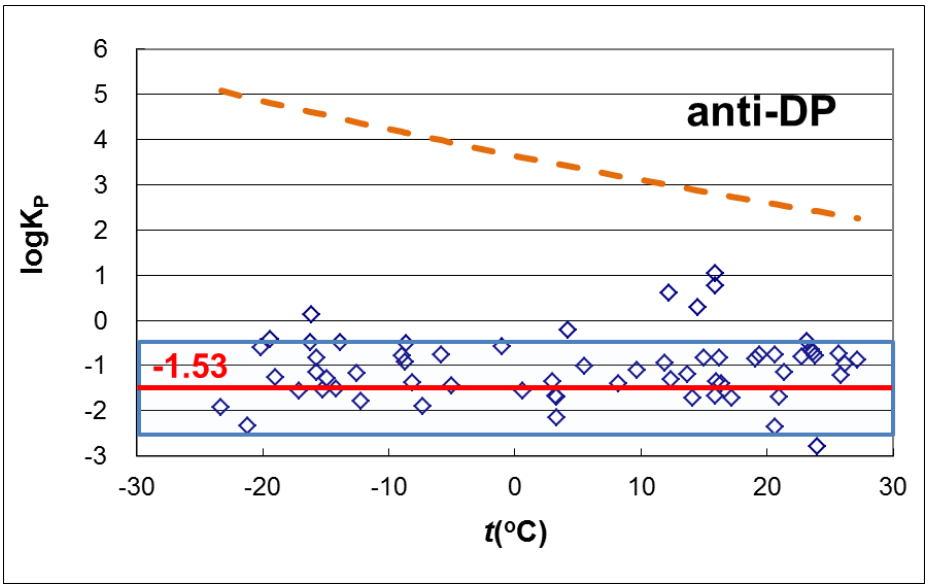
(A)


EHTBB
-1.53
logK_PM(t)
t(°C)
5
4
3
2
1
0
-1
-2
-3
-20
-10
0
10
20
30

(B)



(C)



(D)

Figure S13. Variation of log K_{PM} for BEHTBP,(A) EHTBB (B), syn-DP (C), and anti-DP (D) versus temperature (Li et al., 2016b)

Tables

Table S1. Statistics of concentrations for $\sum_9$PBDEs (BDE-17, -28, -47, -99, -100, -153, -154, -183, and -209) and BDE-209 in gaseous and particulate phases (pg·m^{-3}).

Sampling Sites	PBDE	Gas Phase			Particle Phase		
		Mean	Min	Max	Mean	Min	Max
		Urban					
Beijing	$\sum_9$PBDEs	95.1	0.492	538	766	1.03	2930
	BDE-209	77.0(0.81[(1)])	BDL	439	728	BDL	2890
Chengdu	$\sum_9$PBDEs	94.6	1.71	681	425	55.7	1292
	BDE-209	34.9(0.37)	BDL	223	346	52.6	1280
Dalian	$\sum_9$PBDEs	22.4	0.626	321	85.8	5.73	520
	BDE-209	10.2(0.46)	BDL	261	62.5	5.44	442
Guangzhou	$\sum_9$PBDEs	74.4[(2)]	5.76	227	750	58.9	3540
	BDE-209	NA[(3)]	NA	NA	714	56.7	3500
Harbin	$\sum_9$PBDEs	57.8	BDL	570	95.2	BDL	580
	BDE-209	44.0(0.76)	BDL	517	87.8	BDL	574
Kunming	$\sum_9$PBDEs	38.0	BDL	262	112	9.04	663
	BDE-209	20.0(0.53)	BDL	132	83.0	9.04	430
Lhasa	$\sum_9$PBDEs	34.9	BDL	88.0	11.7	BDL	38.9
	BDE-209	16.0(0.46)	BDL	80.2	10.7	BDL	38.0
Lanzhou	$\sum_9$PBDEs	18.3	2.27	118	137	8.95	465
	BDE-209	6.42(0.35)	0.84	78.3	105	6.51	381
Nanchang	$\sum_9$PBDEs	65.9	BDL	352	106	20.7	540
	BDE-209	15.6(0.24)	BDL	100	91.0	19.8	529
Shihezi	$\sum_9$PBDEs	35.0	2.69	282	96.1	7.07	408
	BDE-209	16.3(0.47)	2.60	112	85.6	7.08	404
Xi'an	$\sum_9$PBDEs	56.8	3.45	301	434	12.1	1743
	BDE-209	37.6(0.66)	BDL	229	388	8.92	1710

		Suburban					
Shanghai	$\sum_9$PBDEs	16.7	BDL	118	56.2	BDL	517
	BDE-209	8.30(0.3)	BDL	77.0	46.4	BDL	489
		Rural/Background					
Waliguan	$\sum_9$PBDEs	6.77	BDL	18.5	3.98	BDL	7.11
	BDE-209	BDL	BDL	BDL	1.06	BDL	5.30
Wudalianchi	$\sum_9$PBDEs	7.93	0.61	25.6	6.75	BDL	83.3
	BDE-209	BDL	BDL	BDL	2.94	BDL	81.8
Xuancheng	$\sum_9$PBDEs	6.25	BDL	23.5	8.72	BDL	93.3
	BDE-209	BDL	BDL	BDL	6.20	BDL	87.6

BDL: Below detection limit. NA: Not available

(1) (BDE-209/$\sum_9$PBDEs). (2) BDE-209 is not included. (3) Concentration data of BDE-209 in gas phase at Guangzhou site is not available due to the missing of the gas samples at this site.

Table S2. Detail information for 15 sampling sites (Yang et al., 2013)

Type	Sampling Site	Population (ten thousand)	gross industrial output (billion Yuan)	Annual mean temperature (°C)
Urban	Beijing	1299.85	1041.31	13.61
	Chengdu	1124.96	409.21	17.71
	Dalian	583.37	508.21	12.32
	Guangzhou	784.14	1162.76	23.71
	Harbin	989.86	182.48	5.76
	Kunming	528.52	179.16	17.00
	Lanzhou	322.28	137.62	12.16
	Lhasa	47.72	3.24	11.01
	Nanchang	494.73	178.30	19.16
	Shihezi	63.20	19.00	8.05
	Xi'an	772.30	193.65	15.92
Suburban	Shanghai (Lingang)	15.00		18.43
Rural	Wudalianchi	36.70		5.00
	Xuancheng	276.75	59.46	17.69
Background	Waliguan			-0.63

Table S3 Instrument detection limits (IDL) for PBDEs ranged from 0.02 to 0.72 pg·m^{-3}.

Congener	IDL (pg·m^{-3})	Congener	IDL (pg·m^{-3})
BDE-17	0.03	BDE-100	0.02
BDE-28	0.03	BDE-138	0.13
BDE-47	0.03	BDE-153	0.04
BDE-66	0.03	BDE-154	0.04
BDE-85	0.04	BDE-183	0.16
BDE-99	0.03	BDE-209	0.72

Table S4. Parameters *A* and *B* for PBDEs, used to calculate log K_{OA} (log K_{OA} =*A*+*B*/*T*). (Harner, T. and Shoeib, M, 2002)

PBDEs	*A*	*B*
BDE-17	-3.45	3803
BDE-28	-3.54	3889
BDE-47	-6.47	5068
BDE-66	-7.88	5576
BDE-77	-5.69	4936
BDE-85	-6.22	5331
BDE-99	-4.64	4757
BDE-100	-7.18	5459
BDE-126	-8.41	6077
BDE-153	-5.39	5131
BDE-154	-4.62	4931
BDE-156	-5.8	5298
BDE-183	-3.71	4672
BDE-209[(1)]	**1.25**	**4179**

(1) Data are from this study (see the main text).

Table S5. Concentrations for BDE-209 at gas- and particle-phases in different cities and places worldwide ($pg \cdot m^{-3}$).

Sampling Site	Site type	Sampling Year	Gas-phase (Mean±SD)	Particle-phase (Mean±SD)	References
			Asia		
6 urban and 11 rural sites in north China	urban+rural	2010	4.0±6.1	37±69	(1)
Shanghai, China	urban	2011	6.72	65.8	(2)
Shanghai, China	rural	2011	13.2	3.86	(2)
Yunnan, China	rural	2006	2.17±2.46	15.90±10.46	(3)
11 urban sites across China	urban	2008-2009	**27.8±21.3**	**246±263**	this study
1 suburban site, China	suburban	2008-2009	**8.3**	**46.4**	this study
3 remote/rural sites, China	remote/rural	2008-2009	**BDL**	**BDL**	this study
			North America		
Alert, Canada	remote	2007-2008	0.83±0.94	0.20±0.15	(6)
Chicago, USA	urban	2005-2009	3.4±0.9	13±2	(7)
Cleveland, USA	urban	2005-2009	1.8±0.5	56±15	(7)
Sturgeon, USA	rural	2005-2009	0.7±0.3	1.9±0.2	(7)
Sleeping Bear Dunes, USA	rural	2005-2009	0.8±0.3	2.5±0.9	(7)
Eagle Harbor, USA	remote	2005-2009	0.5±0.1	1.3±0.4	(7)
Uclielet, Canada	remote	2004-2005	0.20±0.05	0.71±0.26	(8)
Saturna, Canada	suburban	2004-2005	0.11±0.03	1.17±0.06	(8)

		Europe			
Izmir Bay, Turkey	urban	2005	49±15	36±19	(4)
Izmir Bay, Turkey	urban	2005	52±52	46±34	(4)
Izmir,Turkey	suburban	2004-2005	6.2±4.7	9.5±5.9	(5)
Izmir,Turkey	urban	2004-2005	4.3±3.8	21.6±14.8	(5)
Izmir,Turkey	industrial	2005	6.8±5.1	39.8±24.5	(5)
Etang de Thau, France	coastal site	2007-2008	0.12±0.04	1.98±1.28	(9)
Paris, France	urban	2008-2009	4.61±6.26	21.79±38.68	(10)
Zurich, Switzerland	urban	2010	35.73±42.95	33.92±29.72	(11)

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(1) Wang et al., **2012**; (2) Li et al., **2015c;** (3) Xu et al., **2011;** (4) Cetin and Odabasi, **2007a;** (5) Cetin and Odabasi, **2007b**; (6) Xiao et al., **2012;** (7) Salamova and Hites, **2011;** (8) Noel et al., **2009;** (9) Castro-Jiménez et al., **2011;** (10) Tlili et al., **2012;** (11) Bogdal et al., **2014**

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