SUPPORTING INFORMATION

Spatial distribution of old and emerging Flame Retardants in Chinese Forest Soils: Sources, Trends and Processes.

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Text S1. Chemical analysis and QA/QC.

S1.1 Chemical analysis

S1.1.1 Details on methods

About 20 grams per soil sample were (after drying and homogenization) spiked with 20ng of PCB 198 and PCB 209 as surrogate recovery standards and extracted using dichloromethane (DCM) for 36 h. Activated copper granules were added to the collection flask to remove elemental sulphur. Each sample extract was concentrated by a rotary evaporator and solvent-exchanged into hexane. Extracts were cleaned-up through an 8 mm i.d. alumina/silica column packed as follows (from top to bottom): anhydrous sodium sulphate (1cm), 50% sulfuric acid silica (3cm), neutral silica gel (3cm, 3% deactivated) and neutral alumina (1cm, 3% deactivated). FRs were eluted with 20ml of a mixture of dichloromethane:hexane (1:1, V/V), and concentrated to 0.5 ml under a gentle stream of nitrogen. A GPC column was used for the final clean-up step. 6.5g of Bio-Beads S-X3 were used in a 15cm glass column (id=2 cm). The concentrated samples were loaded and eluted with 55mL of a mixture hexane:dichloromethane (1:1, V/V). The first fraction (15mL) was discarded, the following fraction (40ml) containing the FR and the recovery standard was collected. 30μL of dodecane containing 10 ng ^{13}C-PCB 141 (internal standard) were added as "keeper". Samples were concentrated to the final volume of 30 μL under a gentle nitrogen stream and then stored for injection.

S1.1.2 Instrumental analysis

GC-ECNI-MS (Agilent GC7890 coupled with 5975C MSD) with a DB5-MS capillary column (15 m × 0.25 mm i.d. × 0.25 μm film thickness) was used for the determination of eight PBDE congeners (BDEs 28, 47, 99, 100, 153, 154, 183 and 209), six NBFRs (TBPH, PBEB, HBB, TBB, TBE and DBDPE) and DP (*anti*-isomer and *syn*-isomer). 1 μL of sample was injected in splitless mode. Helium was used as carrier gas at the flow rate of 1 mL min^{-1}. Methane was used as chemical ionization moderating gas. The temperature of transfer

line and ion source was maintained at 280 ℃ and 230℃, respectively. The GC oven temperature started at 110℃ for 1 min, increased to 200℃ at a rate of 20℃ min^{-1} (held for 1 min), then to 310℃ at a rate of 10℃min^{-1} (held for 12 min). The specific parameters for the target compounds were shown in Table S1 and the total ion chromatograms of standards and one soil sample were shown in Figure S4.

S1.2 Quality Assurance/Quality Control (QA/QC)

The stability of GC-MS was checked daily using repeated analysis of standard solution. Standards deviation of the instrument ($SD_{instrument}$) ranged from 3% to 12%, and the Instrument Detection Limit (IDL) was three times $SD_{instrument}$, ranging from 0.09 to 0.36 pg. Standards deviation of the blank samples (SD_{blank}) ranged from 0.6% to 7.4%, and the Method Detection Limit (MDL) was calculated by $3.36*SD_{blank} + average_{blank}$, ranging from 1.1 to 28.6 pg g^{-1}, depending on congeners (Table S3).

Text S2. Atmospheric back trajectories and Index of potential source influence analysis.

S2.1.1 Atmospheric back trajectories

Atmospheric back trajectories were analysed using Hysplit to evaluate, on an annual base, the main origin of air masses reaching individual sampling sites. For each sampling site, a total of 360 ten-day backward trajectories were computed covering the full year preceding the sampling. Back trajectories were released from an altitude of 100 m above ground level every 2 days at 6 and 18 UTC (2 and 14 Beijing time (CST)) in order to take into account the day/night variation of the depth of the (mixed) planetary boundary layer (BL). The information on air mass trajectory was elaborated to obtain maps of back trajectories density. The back trajectory density $f_{x,i}$ value for a given point in space i (identified by the area delimited by grid cells with 2.5' resolution) was calculated in relation to each sampling location x as:

$$f_{x,i} = \frac{BTn_{x,i}}{360}$$

where $BTn_{x,i}$ is the number of times a back trajectory originated from site x was passing over location i during the year preceding the sampling, and 360 is the total number of computed back trajectories. The rationale of such an elaboration was to obtain aggregated information showing the areas surrounding the sampling locations with the highest potential to serve as source of airborne FRs for the selected sampling areas. For example, emissions of contaminants from locations with low $f_{x,i}$ value are likely to have little influence on the contamination profile of site x.

S2.1.2 Index of potential source influence

In order to further parameterize the potential influence of primary urban sources on the regional distribution of FRs in China, a numerical index of potential source influence (IPSI) was defined combining the information on back trajectories density and human population distribution. More specifically IPSI was calculated as:

$$IPSI_x = 10^{-8} \sum_i (f_{x,i} \cdot p_i)$$

where p_i is the human population residing within a generic point in space i, and the numerical parameter is simply a scaling factor. IPSI for a given sampling site x is proportional to the product of back trajectory density and human population counts calculated for a circular geographical area (6000 km diameter) centred in x .

In the definition of IPSI, the geographical distribution of human population is assumed to reflect the distribution of FRs primary sources. The IPSI values defined here represent therefore a strategy to rank the influence of potential primary source areas on the contamination pattern in the receiving mountain soils. In addition, IPSI can be considered as an index of potential influence from sources in the subregional/local domain rather than from long-range transport. This can be derived from the fact that $f_{x,i}$ values, which obviously

tend to be higher in proximity of the back trajectory origin, were not weighted for the effect of distance. The potential influence of long-range transport was instead assessed by computing the fraction of time air masses were travelling above the BL using the Hysplit outputs for BL height and back trajectory elevations.

Table S1 GC-MS parameters

Compound	RT[a] (min)	*m/ z*
PBDEs		
BDE-28	10.482	79, 81
BDE-47	13.145	79, 81
BDE-99	15.993	79, 81
BDE-100	14.626	79, 81
BDE-153	18.245	79, 81
BDE-154	16.652	79, 81
BDE-183	18.939	79, 81
BDE-209	27.114	79, 81
NBFRs		
PBEB	11.481	79, 81
HBB	13.068	79, 81
TBB	17.659	357
TBPH	22.850	462, 464
TBE	21.943	79, 81
DBDPE	30.372	79, 81
DP		
syn-DP	22.283	653.8
anti-DP	22.687	653.8
^{13}C-PCB 141 (SS)	11.091	372

Note: RT [a] Retention time

Table S2. Information on sampling locations.

Site		Longitude	Latitude	Altitude(m)	Temperature(°C)	Precipitation(mm)
1	DX-1	123.51	49.54	445	-1.33	505
	DX-2	122.96	52.95	590	-4.79	468
	DX-3	123.61	51.84	860	-5.55	524
	DX-4	122.99	52.30	950	-6.42	523
2	XX-1	128.52	46.63	810	-1.55	714
	XX-2	128.47	46.63	1420	-4.58	798
3	CB-1	127.89	42.26	1030	1.27	740
	CB-2	128.13	42.14	1250	0.06	768
	CB-3	128.07	42.08	1700	-2.01	856
	CB-4	128.07	42.06	1995	-3.76	943
4	ME	127.68	45.39	400	1.67	676
5	HS-1	118.41	44.27	1140	1.47	396
	HS-2	118.72	44.20	1240	0.60	426
	HS-3	118.71	44.19	1400	-0.91	460
6	SHB-1	117.48	42.33	1610	0.13	481
	SHB-2	117.51	42.44	1870	-0.93	501
7	BCW-1	117.60	40.83	1120	4.45	599
	BCW-2	117.61	40.83	1360	3.67	612
	BCW-3	117.61	40.82	1580	2.19	635
	BCW-4	117.61	40.82	1710	1.78	642
8	WYZ-1	113.85	38.73	1430	3.88	527
	WYZ-2	113.84	38.72	1660	4.29	516
	WYZ-3	113.84	38.72	1770	2.96	558
	WYZ-4	113.84	38.72	1880	2.18	584
9	GD-1	111.44	37.89	1310	1.76	555
	GD-2	111.43	37.90	1870	3.61	528
	GD-3	111.43	37.89	2200	2.52	542
10	TS-1	117.61	36.75	400	13.46	640
	TS-2	117.12	36.23	880	10.00	843
	TS-3	117.11	36.25	1100	8.83	903
11	BAIMASI	113.50	35.47	950	13.76	596
12	AS-1	107.47	33.86	1720	7.65	868
	AS-2	107.42	33.89	2410	5.04	908
	AS-3	107.41	33.88	2470	4.93	915
	AS-4	107.43	33.91	3150	1.16	986
13	SN-1	110.42	31.67	1670	8.88	1225
	SN-2	110.49	31.76	1850	8.25	1236
	SN-3	110.28	31.46	2480	4.99	1443
	SN-4	110.30	31.44	2825	3.33	1510
	JG-1	114.57	33.99	285	14.56	714

14	JG-2	114.58	34.69	580	14.47	611
	JG-3	114.59	35.56	890	14.67	577
	JG-4	114.62	37.22	1230	13.53	533
15	JF	105.68	33.69	1785	9.81	692
16	GG-1	102.07	29.60	2060	11.86	883
	GG-2	102.03	29.59	2700	8.55	924
	GG-3	102.00	29.57	2900	7.49	948
	GG-4	101.99	29.57	3100	6.75	962
17	LJ-1	102.42	27.57	2200	12.63	1046
	LJ-2	102.39	27.58	3080	9.15	1098
	LJ-3	102.37	27.57	3830	3.54	1182
18	AL-1	100.99	24.50	2190	14.78	1080
	AL-2	101.03	24.53	2570	13.16	1116
19	LG-1	108.18	26.37	1535	13.12	1336
	LG-2	108.20	26.38	1715	11.42	1405
	LG-3	108.20	26.38	1844	11.42	1405
20	FJ-1	108.72	27.90	1428	12.94	1313
	FJ-2	108.70	27.91	2095	9.55	1452
21	NL-1	112.99	24.95	1378	13.45	1743
	NL-2	113.01	24.91	1432	14.03	1716
	NL-3	113.01	24.92	1545	14.03	1716
22	Jing-1	114.14	26.58	1000	13.94	1766
	Jing-2	114.17	26.51	1310	14.31	1739
23	DH-1	112.54	23.17	210	20.85	1698
	DH-2	112.53	23.17	320	20.58	1704
	DH-3	112.52	23.18	586	19.61	1738
24	DYS-1	118.22	25.64	1180	14.33	1734
	DYS-2	118.22	25.65	1510	14.33	1734
25	LQS-1	119.14	27.97	380	17.19	1796
	LQS-2	119.21	27.92	920	13.91	1984
	LQS-3	119.19	27.87	1650	12.04	2103
	LQS-4	119.19	27.89	1929	11.61	2129
26	TM-1	119.43	30.36	790	12.21	1559
	TM-2	119.71	30.58	1460	15.88	1319
27	DB-1	115.78	31.09	1050	11.20	1632
	DB-2	115.77	31.10	1630	9.08	1733
28	SWDS-1	107.91	21.90	393	20.58	2006
	SWDS-2	107.90	21.90	521	20.58	2006
	SWDS-3	107.92	21.88	708	17.96	1878
29	WGS	114.17	27.46	1175	10.17	2004
30	SYK-1	105.92	38.75	2030	4.96	245
	SYK-2	105.91	38.73	2350	3.48	265

Table S3. The measured concentrations and MDLs for selected compounds (pg/g) and Total Organic Carbon (TOC) (%) of each sampling site in O-horizon and A-horizon. (ND means not detected)

Number ID	O-horizon	BDE28	BDE47	BDE100	BDE99	BDE154	BDE153	BDE183	BDE209	*anti*-DP	*syn*-DP	TBPH	PBEB	HBB	TBB	TBE	DBDPE	TOC
1	DX-1	ND	9.5	4.4	14.2	11.7	6.2	16.4	165.5	41.1	30.4	108.49	ND	ND	228.1	56.9	826.6	37.3
	DX-2	ND	9.2	ND	4.8	4.4	2.8	ND	87.0	17.1	17.4	10.87	ND	ND	28.2	ND	196.2	28.7
	DX-3	ND	25.2	5.1	8.5	3.0	4.7	8.5	87.9	13.1	13.4	51.62	2.9	4.5	251.5	114.2	437.0	35.0
	DX-4	ND	13.3	ND	ND	3.0	3.0	ND	47.1	16.6	11.8	3.97	7.5	ND	23.3	ND	128.3	41.4
2	XX-1	ND	34.7	9.2	36.9	20.9	19.0	23.0	930.5	88.2	70.5	47.48	ND	25.7	122.6	ND	4004.9	36.3
	XX-2	8.0	29.3	11.4	58.1	25.8	23.6	33.1	674.9	113.7	97.5	40.23	11.4	62.5	56.3	10.2	3398.0	37.1
3	CB-1	ND	17.4	8.4	14.9	9.0	7.1	14.9	721.7	85.1	63.0	40.47	7.5	12.2	880.3	16.9	3099.9	29.0
	CB-2	ND	8.6	ND	5.0	4.8	7.3	ND	124.7	15.4	12.4	53.31	ND	ND	248.0	7.3	227.9	17.2
	CB-3	13.7	58.1	11.0	38.4	19.7	13.3	19.2	1356.6	149.4	91.1	32.05	1.5	25.5	467.2	30.9	4507.8	28.0
	CB-4	ND	30.0	6.4	19.3	10.1	8.8	15.4	508.1	96.1	74.4	93.14	ND	9.0	38.2	32.2	1347.0	25.0
4	ME	9.3	26.0	5.9	52.6	17.5	18.3	24.6	877.3	131.0	107.5	169.61	ND	46.0	889.8	ND	5188.2	21.5
5	HS-1	ND	22.0	ND	8.9	4.2	4.2	ND	224.6	33.1	19.9	70.56	ND	14.2	413.6	3.5	906.5	24.2
	HS-2	ND	21.1	ND	13.8	6.0	4.7	ND	77.0	28.5	22.4	50.43	ND	19.0	347.8	ND	686.0	26.7
	HS-3	ND	11.0	4.3	18.5	6.8	6.8	ND	182.5	87.6	97.8	33.97	ND	16.5	162.7	7.1	1490.4	31.2
6	SHB-1	ND	10.9	ND	17.3	7.4	8.8	13.2	289.8	78.8	65.8	44.66	3.0	33.0	1390.2	ND	903.9	19.6
	SHB-2	ND	35.1	12.5	35.4	12.1	17.4	19.8	344.7	115.2	81.0	156.49	ND	67.5	1177.9	ND	2183.3	32.2
7	BCW-1	ND	18.4	7.5	34.9	18.4	13.4	21.2	525.9	109.3	94.6	32.61	ND	87.2	470.8	10.2	2800.2	28.1
	BCW-2	29.8	121.6	74.6	135.6	373.7	17.2	46.1	823.9	278.7	182.7	222.05	4.4	345.1	126.7	21.3	4374.9	41.9
	BCW-3	21.1	239.2	15.3	74.0	24.3	32.5	54.0	1304.0	674.6	375.5	80.82	2.6	133.3	116.8	32.5	3406.3	31.6
	BCW-4	11.3	69.5	34.9	80.1	36.7	25.4	40.0	2469.7	236.8	230.8	38.75	ND	272.9	8.7	65.8	18122.5	23.5
8	WYZ-1	ND	21.1	13.5	115.1	32.2	36.2	38.1	484.5	194.0	172.0	210.28	ND	257.8	363.7	14.8	2252.6	30.4
	WYZ-2	9.4	87.0	18.8	68.6	39.1	29.7	31.9	751.5	212.7	213.9	46.13	ND	178.5	1110.3	19.7	4546.4	22.5
	WYZ-3	20.7	88.3	24.6	68.0	56.8	36.1	53.2	1676.2	287.2	244.1	48.93	1.9	106.9	388.5	50.8	9087.8	37.4
	GD-1	ND	17.0	6.0	25.6	9.5	6.5	16.5	140.1	74.0	76.5	58.68	ND	41.5	15.3	ND	41.4	28.1
9	GD-2	ND	44.7	13.9	20.8	12.3	10.0	17.3	94.7	113.8	110.6	79.66	ND	ND	10.8	ND	45.3	29.6
	GD-3	ND	15.7	5.6	19.2	7.7	5.3	9.6	626.5	81.1	62.9	25.33	ND	27.9	247.2	ND	361.3	33.1
	TS-1	14.6	48.9	11.6	58.2	17.7	22.7	36.2	3187.4	144.1	109.3	18.51	1.1	88.3	457.8	94.1	8667.5	10.1
10	TS-2	17.9	75.7	11.9	63.9	30.2	27.5	42.3	2399.8	301.0	297.9	61.26	4.9	40.6	64.1	20.1	16833.7	11.9
	TS-3	26.0	100.1	17.0	42.1	30.2	28.4	42.1	4357.3	257.7	338.5	47.23	ND	87.7	55.0	24.2	11016.8	31.3
	AS-1	ND	5.2	5.5	7.0	7.5	7.5	9.4	ND	19.9	13.9	42.00	3.0	2.6	287.6	ND	25.1	18.8
12	AS-2	6.1	16.6	ND	8.4	6.6	3.6	9.2	281.2	130.7	110.2	51.68	ND	4.9	719.2	14.8	1251.0	21.6
	AS-3	ND	13.8	8.8	7.1	5.9	4.1	10.6	66.2	68.5	51.9	169.44	ND	ND	972.1	21.1	955.2	24.0
	AS-4	7.0	30.8	7.0	12.2	4.7	2.9	ND	45.0	48.6	24.3	17.59	2.2	ND	566.0	ND	247.4	33.1
	SN-1	ND	9.5	3.6	10.9	7.0	7.0	ND	114.7	67.8	58.2	29.09	6.9	ND	192.9	10.8	251.2	31.2
13	SN-2	ND	20.1	ND	7.5	8.3	7.2	9.7	55.3	42.5	38.4	64.09	7.8	ND	16.6	8.2	296.4	37.7
	SN-3	ND	25.0	4.2	13.7	12.3	18.6	19.0	132.9	83.0	60.0	61.76	5.1	ND	306.7	6.7	576.2	24.4
	SN-4	7.6	34.6	13.7	13.7	16.3	14.8	19.8	138.2	45.1	36.5	117.06	ND	3.0	37.3	9.5	335.1	34.7

Number ID	O-horizon	BDE28	BDE47	BDE100	BDE99	BDE154	BDE153	BDE183	BDE209	*anti*-DP	*syn*-DP	TBPH	PBEB	HBB	TBB	TBE	DBDPE	TOC
	JG-2	9.5	58.8	15.3	68.7	41.6	41.6	64.8	903.2	220.8	188.6	413.25	3.8	29.4	ND	104.9	5810.4	23.3
14	JG-3	11.2	79.8	25.7	70.5	49.4	41.4	56.1	1213.6	310.6	227.5	464.51	9.2	26.9	491.6	6.7	7990.6	30.1
	JG-4	10.3	81.9	30.8	76.4	61.3	33.9	50.2	976.8	357.8	330.6	174.17	4.0	37.8	9.7	57.5	6110.7	26.9
	JF	ND	29.1	7.1	19.7	24.9	24.9	26.8	553.8	74.0	58.0	106.69	5.3	ND	145.7	15.9	98.9	36.4
15	GG-1	ND	ND	ND	ND	2.7	ND	ND	ND	5.0	4.5	19.97	ND	ND	34.0	ND	89.7	15.5
16	GG-2	ND	100.0	30.9	138.0	20.8	40.1	59.8	366.1	226.6	110.9	94.41						25.3
	GG-3	ND	56.2	14.1	74.9	12.4	29.3	39.6	144.0	129.8	57.1	168.26	2.0	8.4	34.8	109.2	284.7	30.1
	GG-4	ND	15.1	27.6	27.6	176.8	16.6	ND	ND	133.0	52.1	28.00	ND	ND	53.2	ND	53.9	25.9
	LJ-2	ND	72.2	64.5	21.9	75.6	41.1	16.1	44.2	30.0	19.3	184.98	10.4	ND	554.3	3.5	84.3	38.1
17	LJ-3	ND	3.8	ND	ND	2.8	ND	ND	40.1	10.0	5.0	17.57	ND	ND	111.4	ND	531.4	44.2
	AL-1	ND	25.0	5.1	8.5	6.4	4.7	ND	277.4	18.1	11.6	42.38	6.1	15.9	ND	ND	107.3	27.2
18	AL-2	ND	8.8	ND	5.5	3.3	4.2	ND	285.7	14.1	13.5	186.81	92.1	52.1	282.5	97.5	371.0	27.2
	LG-1	ND	40.8	8.2	30.6	85.6	14.9	18.8	133.0	78.4	75.1	355.34	7.0	ND	ND	17.5	835.4	44.6
19	LG-2	ND	41.6	12.4	18.6	20.6	7.0	11.1	152.5	105.8	75.2	263.40	6.0	ND	15.3	19.2	948.3	37.2
	LG-3	ND	41.5	6.8	14.6	13.3	9.4	9.7	115.9	62.8	48.4	428.45	ND	4.7	29.1	ND	1005.4	29.7
	FJ-1	ND	24.8	11.3	27.7	24.8	13.5	25.2	153.7	204.2	196.2	287.03	9.0	10.4	42.1	44.6	1673.4	42.8
20	FJ-2	6.2	74.9	16.5	55.7	22.3	24.3	32.0	459.2	157.2	134.8	272.02	15.0	10.0	48.7	48.1	2682.4	39.3
	NL-2	12.9	134.7	39.9	91.6	59.5	41.9	58.7	507.2	457.9	281.6	643.23	3.7	35.5	2.8	56.8	1937.8	42.6
21	NL-3	10.3	101.8	36.1	92.9	38.8	40.9	45.4	168.6	174.9	123.6	425.62	ND	ND	8.6	36.2	1024.0	38.7
	Jing-1	6.1	32.7	11.1	22.1	17.9	17.9	20.9	102.3	105.3	82.4	58.47	ND	19.3	110.9	25.3	1625.4	21.1
22	Jing-2	ND	15.7	10.7	9.7	9.3	7.1	9.7	52.4	44.6	28.7	29.13	3.2	3.0	34.7	18.1	652.5	16.8
	DH-1	ND	34.9	21.6	42.4	28.4	19.0	34.3	2430.6	242.1	191.2	399.26	1.2	6.4	216.3	332.4	5946.9	4.3
23	DH-2	ND	74.6	48.8	108.1	70.8	62.8	74.1	5891.4	496.3	388.3	643.71	2.1	19.0	25.6	248.9	2631.5	6.9
	DH-3	ND	70.5	28.6	88.4	50.2	33.5	58.7	4680.2	363.4	295.8	83.65	2.1	8.7	ND	200.6	4085.3	6.3
	DYS-1	9.5	85.0	12.3	38.9	26.2	21.5	26.6	401.5	155.1	138.0	118.17	7.5	17.6	60.1	44.2	3147.4	32.7
24	DYS-2	16.9	143.5	24.6	79.1	42.3	46.2	54.5	705.7	260.1	195.6	81.84	5.0	10.9	39.3	99.1	3539.0	42.1
	LQS-1	ND	19.1	9.6	12.8	12.7	12.7	13.4	223.9	90.8	71.1	36.92	3.2	3.7	100.9	36.4	2163.5	16.2
25	LQS-2	ND	17.1	6.5	8.9	10.1	7.3	11.7	193.5	86.1	79.2	90.60	2.8	5.1	5.6	26.7	1098.5	42.1
	LQS-3	ND	4.8	ND	ND	3.2	ND	ND	89.2	30.3	29.8	128.86	ND	ND	31.3	ND	51.7	8.9
	TM-1	ND	25.3	8.3	15.3	17.8	11.2	18.0	608.6	102.3	95.0	99.38	3.0	ND	ND	25.3	1849.1	6.0
26	TM-2	ND	33.3	11.2	22.9	26.7	13.0	30.5	1958.3	177.5	182.2	36.17	ND	12.5	231.2	ND	9691.6	3.9
	DB-1	ND	18.1	6.2	12.0	11.6	9.4	13.5	345.6	103.7	76.9	123.24	2.9	5.4	227.0	22.8	1645.1	5.2
27	DB-2	16.1	93.3	21.1	38.3	43.9	30.8	57.4	965.7	478.7	323.9	85.67	2.7	16.2	50.6	138.9	5482.5	12.7
	SWDS-1	ND	23.3	4.1	11.3	9.9	5.8	8.2	108.9	74.7	62.6	213.69	ND	2.8	ND	11.1	1257.8	41.0
28	SWDS-2	ND	7.5	ND	8.3	5.7	6.3	7.7	150.3	89.0	68.3	125.42	2.1	ND	ND	14.1	1227.5	15.5
	SWDS-3	ND	22.6	10.2	19.5	17.5	14.1	23.8	168.9	92.9	78.7	265.25	6.8	ND	ND	44.9	1808.9	16.1
	WGS	ND	23.2	9.4	25.9	13.8	13.8	15.3	453.2	66.7	58.7	113.63	2.4	10.7	24.0	10.8	893.5	27.4
29	SYK-1	ND	5.1	ND	9.4	ND	2.4	ND	62.4	37.4	25.5	41.81	ND	7.6	ND	ND	120.2	18.5
	MDLs	5.9	1.9	3.4	4.0	1.9	1.9	7.6	28.6	2.6	1.3	3.2	1.1	2.0	2.4	3.3	1.8	
	Detection Frequency	30.7	98.7	81.3	94.7	98.7	96.0	80.0	96.0	100.0	100.0	100.0	58.7	70.7	88.0	73.3	100.0	
	Mean values	13.0	43.3	16.0	37.3	28.2	17.7	28.4	728.6	139.3	110.2	131.7	6.8	46.0	250.5	48.5	2643.0	

Number ID	A-horizon	BDE28	BDE47	BDE100	BDE99	BDE154	BDE153	BDE183	BDE209	*anti*-DP	*syn*-DP	TBPH	PBEB	HBB	TBB	TBE	DBDPE	TOC
1	DX-1	ND	5.9	ND	ND	ND	ND	ND	72.6	28.3	18.2	248.9	4.3	ND	31.6	12.2	319.5	18.0
	DX-2	ND	3.7	ND	ND	3.7	ND	ND	ND	6.0	3.8	88.5	ND	ND	42.0	ND	32.3	15.7
	DX-3	ND	40.5	ND	4.2	ND	ND	ND	56.3	17.9	10.0	38.8	11.1	ND	116.5	6.0	160.3	21.4
	DX-4	7.9	3.4	ND	ND	ND	ND	ND	66.0	2.6	2.0	17.0	ND	ND	96.1	ND	186.7	14.4
2	XX-1	ND	15.5	4.1	10.1	7.4	6.4	8.1	225.3	30.7	26.8	33.2	ND	4.3	16.4	14.0	851.4	16.3
	XX-2	ND	5.1	ND	ND	5.5	3.6	ND	52.5	21.1	10.3	15.1	ND	ND	36.7	ND	127.8	7.9
3	CB-1	ND	5.1	ND	4.8	4.1	2.5	ND	229.7	22.7	27.6	17.5	ND	ND	64.4	8.9	338.2	12.7
	CB-2	ND	ND	ND	ND	ND	ND	ND	ND	5.7	2.6	34.7	2.0	ND	42.4	4.1	207.2	3.1
	CB-3	ND	11.6	ND	4.3	3.6	3.6	8.1	74.9	16.9	15.5	17.0	15.6	3.2	63.5	11.9	184.9	10.1
	CB-4	ND	5.3	ND	ND	ND	ND	ND	ND	8.2	6.8	61.8	1.4	ND	12.0	ND	97.0	7.8
4	ME	ND	11.7	4.5	8.5	4.9	6.6	13.5	350.6	50.6	24.1	21.2	ND	8.4	159.4	9.0	822.1	16.1
5	HS-1	ND	4.0	ND	ND	ND	ND	ND	ND	8.5	4.0	18.4	ND	ND	490.4	ND	51.2	3.6
	HS-2	ND	ND	ND	ND	ND	ND	ND	ND	9.1	2.4	16.2	ND	ND	930.3	ND	85.8	6.3
	HS-3	ND	2.6	ND	ND	ND	ND	ND	28.9	6.5	4.3	34.4	2.5	ND	59.0	ND	96.1	9.5
6	SHB-1	ND	2.1	ND	ND	ND	ND	ND	29.9	8.6	6.5	27.2	ND	ND	1574.6	ND	93.5	10.6
	SHB-2	ND	2.5	ND	ND	2.5	1.9	ND	63.7	26.6	13.1	11.3	ND	ND	1318.9	ND	127.1	10.3
7	BCW-1	ND	3.8	ND	5.9	3.1	3.1	ND	125.6	16.9	12.4	5.8	ND	2.5	335.3	18.5	277.1	5.3
	BCW-2	ND	7.3	ND	11.5	5.3	2.5	ND	199.7	42.9	53.1	44.4	ND	9.7	ND	9.3	57.9	11.6
	BCW-3	ND	10.4	ND	6.0	4.3	3.8	9.0	77.3	35.9	23.0	9.0	ND	4.4	701.2	4.9	214.5	12.6
	BCW-4	ND	4.5	ND	5.2	2.7	2.7	ND	124.2	18.3	12.0	17.1	ND	3.2	768.4	9.1	410.6	12.4
8	WYZ-1	ND	3.3	ND	ND	3.3	ND	ND	33.1	15.8	16.2	13.8	ND	2.3	79.0	ND	140.2	4.7
	WYZ-2	ND	3.5	ND	ND	ND	ND	ND	ND	7.1	5.3	10.2	ND	ND	268.8	ND	38.2	7.9
	WYZ-3	ND	6.5	ND	4.6	3.4	1.9	ND	44.9	51.4	15.7	63.7	ND	2.1	340.6	9.3	72.2	3.9
	WYZ-4	ND	6.2	ND	ND	2.4	2.1	ND	ND	8.1	5.9	75.4	ND	ND	341.3	ND	54.4	4.0
9	GD-1	ND	4.5	ND	ND	ND	ND	ND	ND	7.6	7.3	5.5	ND	ND	691.0	ND	45.1	10.6
	GD-2	ND	3.7	ND	ND	2.0	2.4	ND	54.3	16.7	13.3	32.0	ND	2.8	ND	ND	15.7	11.4
	GD-3	ND	8.4	6.9	ND	ND	2.7	ND	80.3	75.1	25.0	16.8	ND	2.7	366.1	49.5	214.2	13.4
10	TS-1	ND	16.1	4.4	26.0	10.0	7.0	11.8	884.9	25.9	24.9	14.6	1.7	25.2	489.7	14.4	1351.0	2.7
	TS-2	ND	23.9	8.2	21.1	11.3	12.4	20.1	2174.0	98.7	101.6	43.0	3.2	42.2	39.2	15.1	2055.4	2.5
	TS-3	ND	8.9	ND	7.7	4.1	5.7	ND	263.4	23.7	33.5	38.3	ND	ND	63.7	11.8	223.0	2.0
11	BMS	ND	12.6	ND	14.0	7.7	5.6	8.2	120.7	35.7	31.6	33.2	21.4	ND	734.7	9.8	506.8	8.9
12	AS-1	ND	6.9	ND	7.1	ND	ND	ND	40.3	24.9	12.7	14.0	10.8	ND	180.7	ND	195.2	15.3
	AS-2	ND	4.8	ND	ND	2.0	ND	ND	ND	22.9	14.2	29.1	ND	ND	106.0	ND	229.8	15.4
	AS-3	ND	11.4	ND	4.4	2.7	2.2	ND	45.6	29.7	22.6	22.6	15.3	ND	180.8	ND	247.1	13.2
	AS-4	ND	5.4	4.9	ND	4.6	4.6	ND	ND	18.4	12.1	12.6	ND	ND	150.6	ND	147.5	26.5
13	SN-1	ND	4.6	ND	ND	2.2	2.2	ND	53.3	29.2	14.6	40.7	4.3	ND	65.7	4.2	104.4	11.9
	SN-2	ND	14.6	ND	5.5	5.3	2.9	ND	54.0	29.4	15.5	34.9	28.5	3.2	26.3	3.5	103.2	9.3
	SN-3	ND	22.0	ND	4.7	3.8	2.2	ND	73.2	27.4	22.8	54.7	19.4	ND	21.0	5.2	152.3	18.1
	SN-4	ND	17.2	ND	ND	ND	ND	ND	19.1	16.3	12.9	49.0	2.6	1.7	10.4	6.1	233.0	14.8
14	JG-1	ND	14.5	6.2	13.8	12.1	10.6	10.8	218.4	79.7	57.6	55.9	ND	2.2	76.6	21.0	1578.0	19.6
	JG-2	ND	24.0	7.1	21.4	14.6	11.5	17.9	567.6	104.3	87.4	93.5	ND	5.1	ND	35.1	2845.0	4.3
	JG-3	ND	28.8	ND	16.0	13.6	25.0	ND	208.0	62.4	44.9	103.3	6.8	3.1	9.6	34.0	811.4	9.6
	JG-4	ND	9.9	ND	10.4	6.9	3.6	ND	120.9	31.3	31.4	45.3	ND	ND	19.9	17.0	350.7	16.5
15	JF	ND	10.9	ND	12.1	8.1	5.8	ND	119.3	32.9	30.8	82.7	69.5	ND	108.0	7.1	149.7	17.3

Number ID	A-horizon	BDE28	BDE47	BDE100	BDE99	BDE154	BDE153	BDE183	BDE209	*anti*-DP	*syn*-DP	TBPH	PBEB	HBB	TBB	TBE	DBDPE	TOC
16	GG-1	ND	ND	ND	ND	ND	ND	ND	95.2	2.9	1.8	22.266	ND	ND	10.7	ND	45.8	2.9
	GG-2	ND	28.9	15.5	152.4	6.6	122.4	ND	ND	5.9	5.4	77.2	ND	ND	16.2	ND	24.3	15.3
	GG-3	ND	12.9	ND	59.1	2.3	46.4	ND	ND	6.0	3.6	14.1	17.6	ND	13.3	4.2	35.6	10.3
	GG-4	ND	48.6	ND	5.7	4.3	3.2	ND	120.0	22.4	8.9	132.5	ND	4.7	98.3	27.9	144.7	14.1
17	LJ-1	ND	ND	ND	ND	ND	15.9	ND	ND	3.9	3.3	5.7	ND	ND	284.4	ND	265.5	8.6
	LJ-2	ND	4.1	ND	ND	2.7	2.0	ND	ND	8.4	5.7	47.5	13.2	ND	234.7	ND	108.3	17.2
	LJ-3	ND	13.1	6.3	6.3	4.8	3.7	ND	75.4	22.1	23.7	70.5	11.0	ND	350.6	10.0	227.4	43.6
18	AL-1	ND	7.2	ND	ND	ND	ND	ND	ND	5.8	3.6	29.0	ND	ND	8.0	ND	50.5	6.9
	AL-2	ND	23.0	4.0	ND	3.0	3.0	13.6	ND	9.0	5.4	144.9	5.0	2.7	31.6	ND	135.8	13.4
19	LG-1	ND	21.6	8.7	30.5	78.2	22.3	ND	67.2	33.9	34.1	94.0	11.1	4.7	6.1	4.8	72.1	14.6
	LG-2	ND	6.1	ND	5.0	3.6	2.3	ND	736.4	20.5	23.1	91.5	3.2	ND	37.3	4.6	288.6	9.1
	LG-3	11.2	54.0	13.5	52.6	4.6	109.0	ND	66.6	40.4	29.4	350.0	1.4	2.9	61.8	12.0	276.8	10.9
20	FJ-1	ND	20.8	10.1	11.9	10.8	10.8	11.0	98.7	74.8	66.8	102.7	3.1	ND	17.0	29.4	713.6	24.7
	FJ-2	ND	11.2	10.9	7.6	3.6	8.3	ND	46.5	34.3	20.7	38.1	ND	ND	4.4	9.3	180.9	16.1
21	NL-1	ND	52.9	13.5	43.2	22.8	30.4	18.9	580.0	141.5	86.3	128.6	3.5	16.9	ND	67.3	1865.5	10.4
	NL-2	ND	23.6	10.7	25.7	17.1	16.0	15.7	343.2	129.9	108.6	149.7	1.5	2.6	46.5	37.1	1551.7	8.9
	NL-3	ND	11.8	5.0	10.7	8.8	10.6	ND	140.5	30.7	23.1	122.3	ND	ND	13.1	14.3	498.9	4.1
22	Jing-1	ND	13.2	6.4	7.0	8.0	3.3	8.7	154.0	45.8	35.4	19.4	ND	21.4	41.0	7.6	34.0	8.4
	Jing-2	ND	28.3	9.5	19.6	11.1	12.3	9.5	179.7	70.9	47.9	82.2	6.5	ND	25.1	33.7	945.7	16.0
23	DH-1	ND	3.7	5.7	ND	4.1	13.9	ND	134.0	29.5	17.5	388.0	ND	ND	ND	16.0	702.5	3.1
	DH-2	ND	24.6	11.1	24.8	29.2	19.4	32.6	608.4	222.5	160.8	144.4	4.6	10.4	7.8	235.1	6893.2	2.6
	DH-3	ND	22.6	14.7	31.1	24.3	14.2	31.1	2356.5	174.9	118.7	62.3	1.2	ND	ND	114.8	12604.1	3.2
24	DYS-1	ND	55.6	5.5	20.1	17.0	10.7	18.9	157.3	143.6	83.7	293.8	5.2	14.4	107.0	34.8	188.7	12.0
	DYS-2	ND	29.4	7.8	17.9	8.0	8.0	19.0	227.3	68.9	55.8	62.3	1.8	ND	17.6	43.7	946.0	11.6
25	LQS-1	ND	2.3	ND	ND	3.1	ND	ND	50.0	16.1	12.3	94.0	ND	ND	77.4	4.1	335.3	2.9
	LQS-2	ND	7.4	ND	6.9	8.0	4.2	10.0	138.5	41.1	34.9	23.7	ND	ND	71.0	25.0	3557.8	5.2
	LQS-3	ND	5.4	5.6	4.4	2.3	4.2	ND	ND	28.5	19.4	65.8	ND	2.4	4.0	6.3	131.4	4.0
	LQS-4	ND	ND	ND	ND	ND	2.7	ND	ND	6.0	5.6	526.4	ND	ND	6.0	ND	30.3	2.4
26	TM-1	ND	8.6	ND	ND	2.5	2.5	ND	107.6	43.1	22.2	38.4	ND	ND	ND	6.6	403.8	2.9
	TM-2	ND	9.4	4.0	5.5	9.4	2.4	11.1	361.0	56.8	57.9	21.3	1.6	3.6	8.2	19.3	1467.9	7.3
27	DB-1	ND	15.8	4.9	8.2	5.4	3.2	ND	135.0	106.8	67.9	25.0	8.0	10.1	37.2	10.9	373.8	3.6
	DB-2	ND	36.8	8.4	19.6	16.1	12.7	25.2	696.0	186.9	171.8	45.2	9.2	3.3	4.8	74.6	1071.1	13.5
28	SWDS-1	ND	4.1	ND	ND	ND	ND	ND	37.0	20.2	17.8	114.2	1.7	ND	ND	ND	5.5	5.7
	SWDS-2	ND	2.6	ND	ND	2.3	6.8	ND	48.4	21.0	17.6	47.1	ND	ND	ND	5.1	283.7	4.6
	SWDS-3	ND	9.9	5.3	7.7	8.0	4.6	8.6	109.8	65.9	64.0	85.2	2.4	ND	ND	23.0	589.9	7.0
29	WGS	ND	15.2	9.1	11.2	9.9	9.9	9.6	96.7	41.8	35.0	76.9	ND	2.2	9.8	6.0	417.7	14.8
30	SYK-1	ND	1.9	ND	ND	ND	ND	ND	ND	6.1	7.5	30.0	ND	ND	ND	ND	23.3	9.7
	SYK-2	ND	ND	ND	ND	ND	ND	ND	ND	5.0	2.3	17.8	ND	ND	ND	ND	27.5	11.7
	Detection Frequency	2.4	92.7	37.8	58.5	74.4	73.2	29.3	75.6	100.0	100.0	100.0	45.1	39.0	85.4	65.9	100.0	
	Mean values	9.5	13.9	7.8	17.2	8.3	11.5	14.6	240.6	39.7	29.9	68.9	9.0	7.2	184.0	22.9	660.0	

Table S4. Spearman correlation coefficients and P-values for the correlation analysis between selected PBDEs, NBFRs used as PBDEs substitutes and DP isomers.

In particular: DBDPE, TBB (combined with TBPH) and TBE are respectively used as alternatives for penta-BDE, octa-BDE and deca-BDE formulation.

	O-horizon		A-horizon	
	r	P	r	P
BDE 209 vs DBDPE	**0.688**	<0.001	**0.670**	<0.001
BDE 183 vs TBE	**0.553**	<0.001	**0.630**	<0.001
∑Penta-BDE vs ∑ (TBPH+TBB)	0.172	0.141	-0.077	0.489
∑Penta-BDE vs TBPH	**0.406**	<0.001	**0.411**	<0.001
∑Penta-BDE vs TBB	-0.053	0.655	**-0.221**	0.046
TBPH vs TBB	**-0.254**	0.029	**-0.460**	<0.001
anti-DP vs *syn*-DP	**0.981**	<0.001	**0.967**	<0.001

Table S5. Compound ratios for TBB (f_{TBB})[a] and *anti*-Dechlorane Plus (DP) (f_{anti})[b].

(f_{TBB})[a] =C_{TBB}/ (C_{TBPH}+C_{TBB}) ; (f_{anti})[b]= $C_{anti\text{-}DP}$/ ($C_{anti\text{-}DP}$+$C_{syn\text{-}DP}$); [c]: missing data due to lack of O-horizon at site.

	Site	O-horizon			A-horizon	
		f_{TBB}	f_{anti}		f_{TBB}	f_{anti}
1	DX-1	0.68	0.57		0.11	0.61
	DX-2	0.72	0.50		0.32	0.62
	DX-3	0.83	0.50		0.75	0.64
	DX-4	0.85	0.59		0.85	0.56
2	XX-1	0.72	0.56		0.33	0.53
	XX-2	0.58	0.54		0.71	0.67
3	CB-1	0.96	0.57		0.79	0.45
	CB-2	0.82	0.55		0.55	0.68
	CB-3	0.94	0.62		0.79	0.52
	CB-4	0.29	0.56		0.16	0.55
4	ME	0.84	0.55		0.88	0.68
5	HS-1	0.85	0.47		0.96	0.68
	HS-2	0.87	0.62		0.98	0.79
	HS-3	0.83	0.56		0.63	0.60
6	SHB-1	0.97	0.55		0.98	0.57
	SHB-2	0.88	0.59		0.99	0.67
7	BCW-1	0.94	0.54		0.98	0.58
	BCW-2	0.36	0.60		0.00	0.45
	BCW-3	0.59	0.64		0.99	0.61
	BCW-4	0.18	0.51		0.98	0.60
8	WYZ-1	0.63	0.53		0.85	0.49
	WYZ-2	0.96	0.50		0.96	0.57
	WYZ-3	0.89	0.54		0.84	0.77
	WYZ-4	–[c]	–		0.82	0.58
9	GD-1	0.21	0.49		0.99	0.51
	GD-2	0.12	0.51		0.00	0.56
	GD-3	0.91	0.56		0.96	0.75
10	TS-1	0.96	0.57		0.97	0.51
	TS-2	0.51	0.50		0.48	0.49
	TS-3	0.54	0.43		0.62	0.41
11	BAIMASI	–	–		0.96	0.53
12	AS-1	0.87	0.59		0.93	0.66
	AS-2	0.93	0.54		0.78	0.62
	AS-3	0.85	0.57		0.89	0.57
	AS-4	0.97	0.67		0.92	0.60
13	SN-1	0.87	0.54		0.62	0.67
	SN-2	0.21	0.53		0.43	0.66
	SN-3	0.83	0.58		0.28	0.55
	SN-4	0.24	0.55		0.18	0.56

14	JG-1	–	–		0.58	0.58
	JG-2	0.00	0.54		0.00	0.54
	JG-3	0.51	0.58		0.09	0.58
	JG-4	0.05	0.52		0.31	0.50
15	JF	0.58	0.56		0.57	0.52
16	GG-1	0.63	0.52		0.32	0.61
	GG-2	–	0.67		0.17	0.52
	GG-3	0.17	0.69		0.49	0.62
	GG-4	0.66	0.72		0.43	0.71
17	LJ-1	–	–		0.98	0.54
	LJ-2	0.75	0.61		0.83	0.60
	LJ-3	0.86	0.67		0.83	0.48
18	AL-1	0.00	0.61		0.22	0.62
	AL-2	0.60	0.51		0.18	0.62
19	LG-1	0.00	0.51		0.06	0.50
	LG-2	0.05	0.58		0.29	0.47
	LG-3	0.06	0.56		0.15	0.58
20	FJ-1	0.13	0.51		0.14	0.53
	FJ-2	0.15	0.54		0.10	0.62
21	NL-1	–	–		0.00	0.62
	NL-2	0.00	0.62		0.24	0.54
	NL-3	0.02	0.59		0.10	0.57
22	Jing-1	0.65	0.56		0.68	0.56
	Jing-2	0.54	0.61		0.23	0.60
23	DH-1	0.35	0.56		0.00	0.63
	DH-2	0.04	0.56		0.05	0.58
	DH-3	0.00	0.55		0.00	0.60
24	DYS-1	0.34	0.53		0.27	0.63
	DYS-2	0.32	0.57		0.22	0.55
25	LQS-1	0.73	0.56		0.45	0.57
	LQS-2	0.06	0.52		0.75	0.54
	LQS-3	0.20	0.50		0.06	0.60
	LQS-4	–	–		0.01	0.52
26	TM-1	0.00	0.52		0.00	0.66
	TM-2	0.86	0.49		0.28	0.50
27	DB-1	0.65	0.57		0.60	0.61
	DB-2	0.37	0.60		0.10	0.52
28	SWDS-1	0.00	0.54		0.00	0.53
	SWDS-2	0.00	0.57		0.00	0.54
	SWDS-3	0.00	0.54		0.00	0.51
29	WGS	0.17	0.53		0.11	0.54
30	SYK-1	0.00	0.60		0.00	0.45
	SYK-2	–	–		0.00	0.69

Table S6. Spearman correlation coefficients (r) and p value between the log (concentrations) of target compounds and environmental variables in (a) O-horizon and (b) A-horizon. Significant correlations are highlighted in bolt.

a.

	IPSI		TOC		Precipitation		Temperature		Latitude	
	r	p	r	p	r	p	r	p	R	p
BDE 28	0.081	0.678	**0.235**	0.041	-0.092	0.431	0.000	0.998	0.088	0.453
BDE 47	**0.307**	0.007	**0.242**	0.035	0.049	0.674	0.167	0.152	-0.097	0.409
BDE 99	0.198	0.097	0.136	0.242	-0.083	0.481	0.167	0.151	-0.042	0.724
BDE 100	**0.288**	0.024	0.079	0.499	0.138	0.238	**0.279**	0.015	-0.234	0.043
BDE 153	**0.415**	0.001	0.107	0.357	0.065	0.577	**0.268**	0.020	-0.147	0.210
BDE 154	**0.374**	0.001	0.126	0.279	0.117	0.318	**0.298**	0.009	-0.207	0.075
BDE 183	**0.414**	0.001	0.011	0.923	0.129	0.270	**0.277**	0.016	-0.147	0.209
BDE209	0.063	0.601	-0.139	0.232	-0.002	0.983	0.134	0.253	0.068	0.562
anti-DP	**0.299**	0.009	-0.031	0.791	0.121	0.300	**0.295**	0.010	-0.160	0.171
syn-DP	**0.264**	0.022	-0.069	0.556	0.120	0.307	**0.299**	0.009	-0.128	0.273
TBPH	**0.486**	0.007	0.130	0.268	**0.328**	0.004	**0.463**	<0.001	**-0.444**	<0.001
PBEB	-0.210	0.177	0.066	0.574	0.101	0.387	0.122	0.296	-0.122	0.298
HBB	**-0.315**	0.023	0.101	0.387	**-0.360**	0.002	-0.134	0.251	**0.289**	0.012
TBB	**-0.386**	0.015	0.173	0.138	**-0.400**	<0.001	**-0.470**	<0.001	**0.443**	<0.001
TBE	0.148	0.285	-0.137	0.242	**0.329**	0.004	**0.386**	0.001	**-0.271**	0.019
DBDPE	0.133	0.253	-0.062	0.595	0.062	0.597	0.148	0.204	0.063	0.591

b.

	TOC		Precipitation		Temperature		Latitude	
	r	p	r	p	r	p	r	P
BDE 28	0.114	0.306	-0.020	0.857	-0.11	0.326	0.089	0.427
BDE 47	**0.298**	0.006	**0.290**	0.008	**0.288**	0.009	**-0.310**	0.005
BDE 99	0.120	0.284	**0.256**	0.020	**0.394**	<0.001	**-0.330**	0.003
BDE 100	0.056	0.617	**0.470**	<0.001	**0.451**	<0.001	**-0.480**	<0.001
BDE 153	0.071	0.525	**0.370**	0.001	**0.494**	<0.001	**-0.480**	<0.001
BDE 154	0.038	0.736	**0.374**	0.001	**0.498**	<0.001	**-0.400**	<0.001
BDE 183	-0.051	0.650	**0.328**	0.003	**0.400**	<0.001	**-0.280**	0.013
BDE209	-0.005	0.963	**0.238**	0.031	**0.337**	0.002	-0.157	0.159
anti-DP	0.150	0.895	**0.424**	<0.001	**0.463**	<0.001	**-0.360**	0.001
syn-DP	0.140	0.903	**0.448**	<0.001	**0.505**	<0.001	**-0.390**	<0.001
TBPH	0.021	0.851	**0.484**	<0.001	**0.478**	<0.001	**-0.534**	<0.001
PBEB	**0.335**	0.002	0.079	0.483	0.072	0.522	-0.143	0.200
HBB	-0.075	0.501	0.088	0.432	0.173	0.12	-0.069	0.541
TBB	0.182	0.102	**-0.450**	<0.001	**-0.460**	<0.001	**0.419**	<0.001
TBE	0.040	0.723	**0.385**	<0.001	**0.460**	<0.001	**-0.320**	0.003
DBDPE	0.003	0.981	**0.298**	0.007	**0.387**	<0.001	**-0.220**	0.046

Figure S1. The distribution of thirty mountains across China.

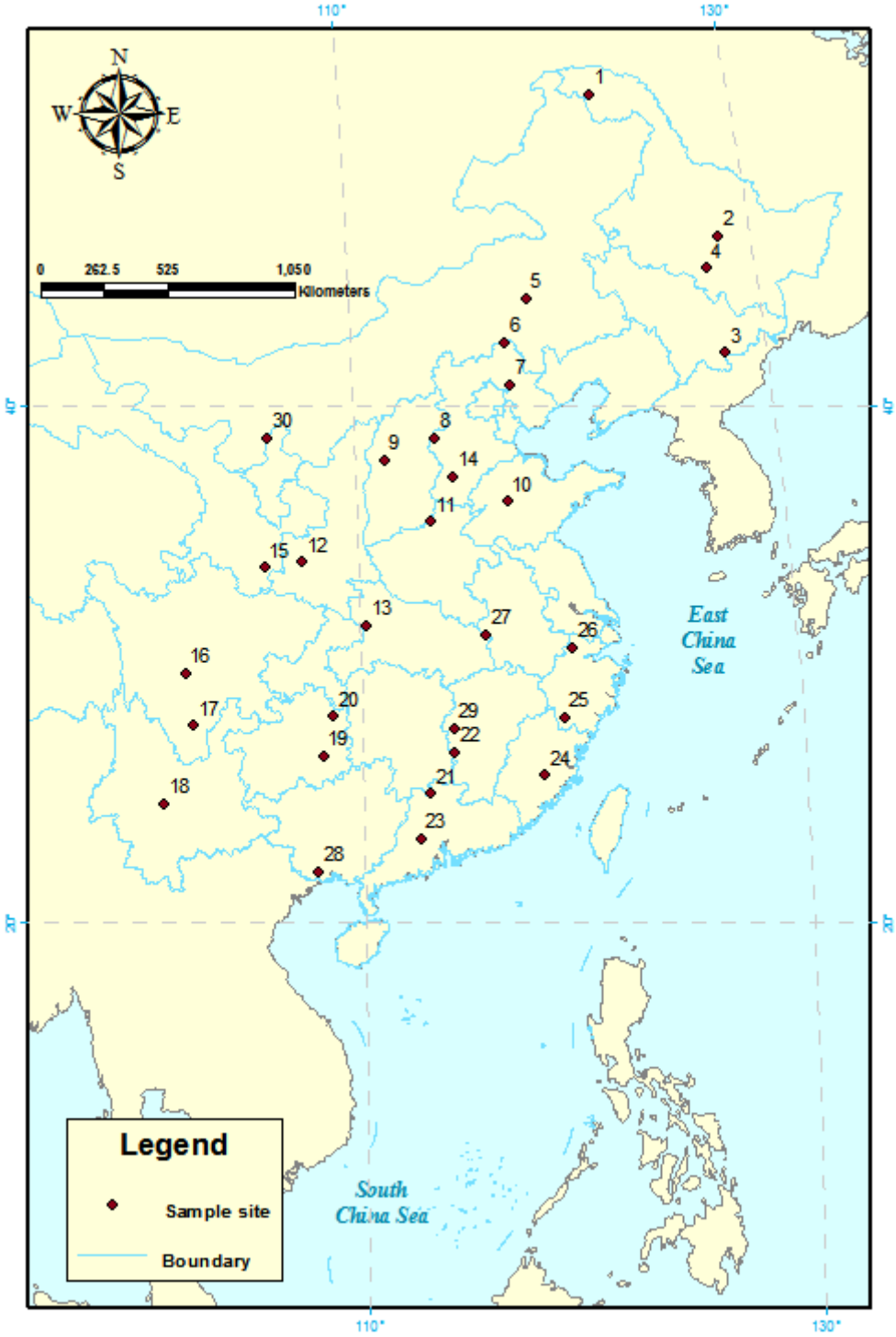


Figure S2. Correlations of natural logarithms of the concentrations of *syn*-DP concentrations plotted against *anti*–DP concentrations in O-horizon and A-horizon.

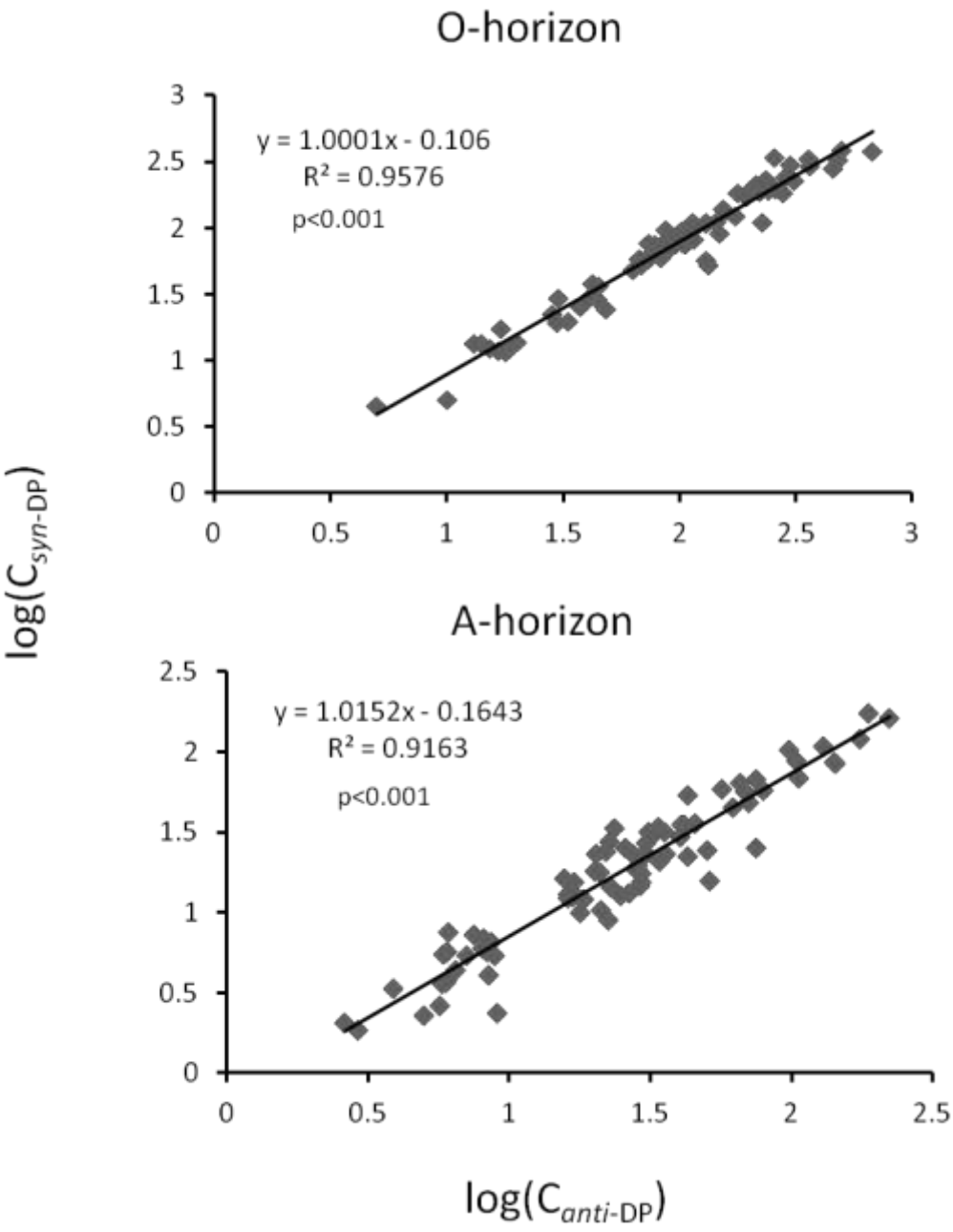
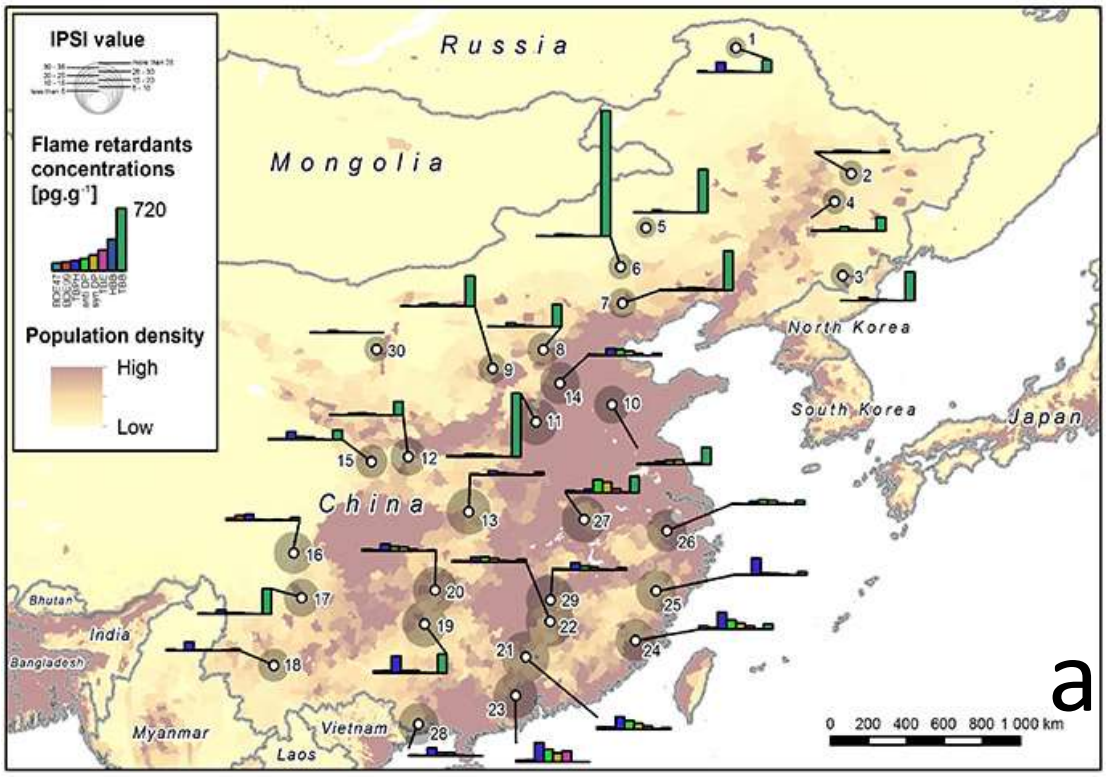
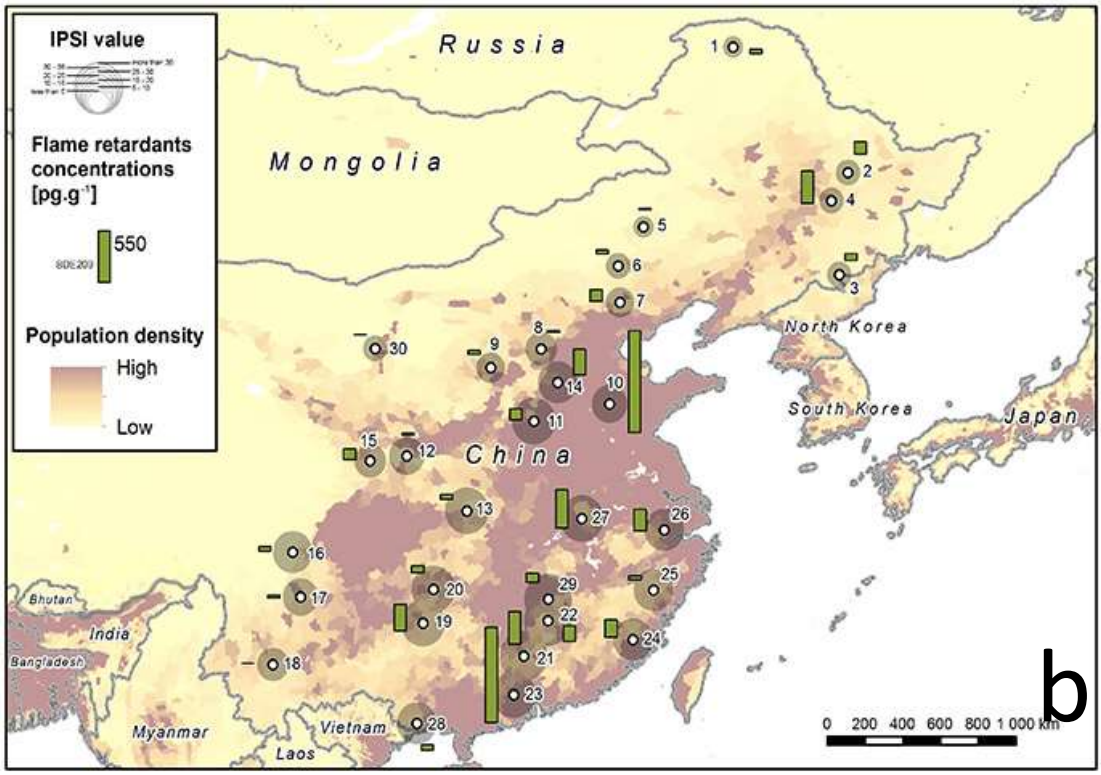


Figure S3. Spatial distribution of selected FRs in organic (A-horizon) soil samples. (a:BDE47, BDE99, anti-DP, syn-DP, TBPH, TBB, HBB and TBE; b:BDE 209;c:DBDPE)





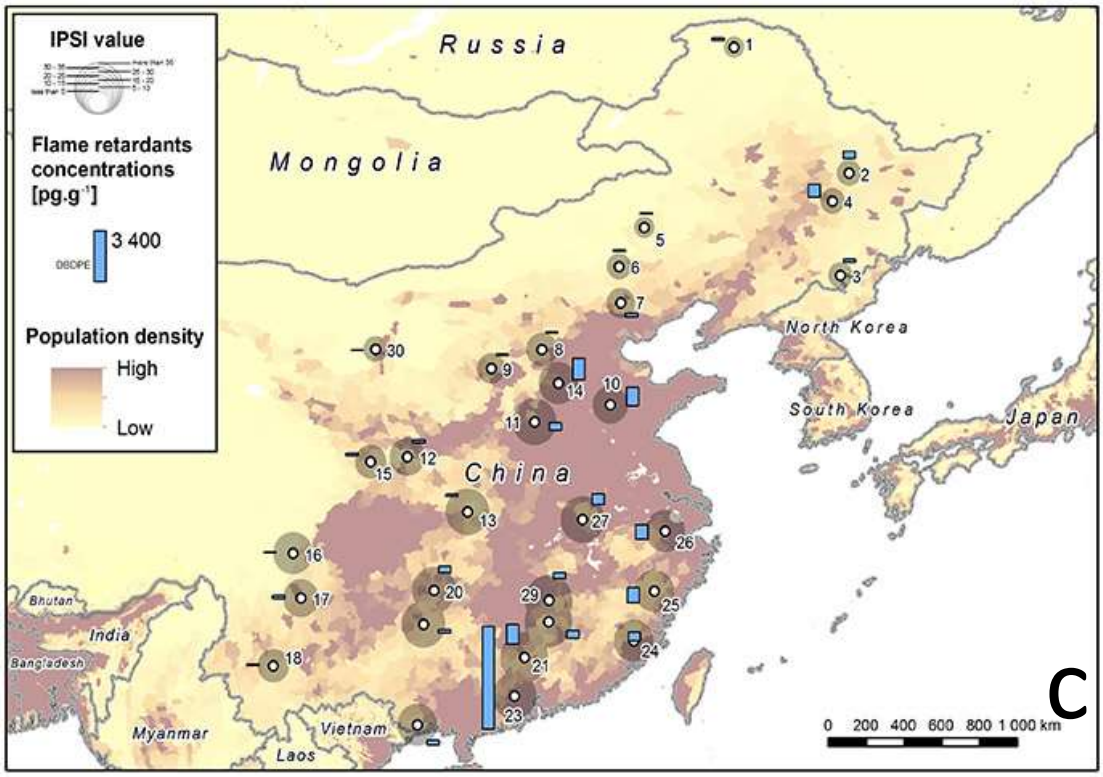
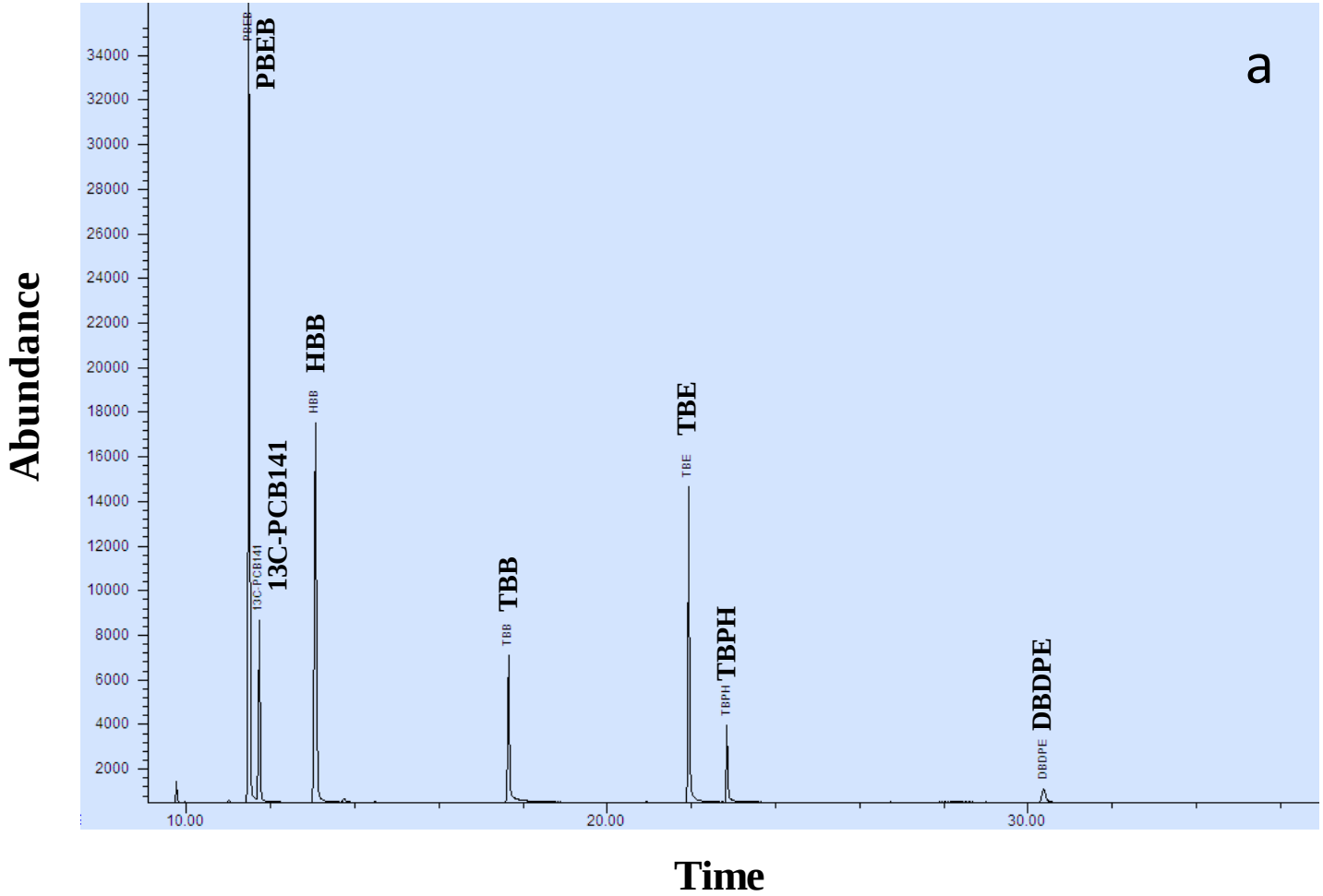
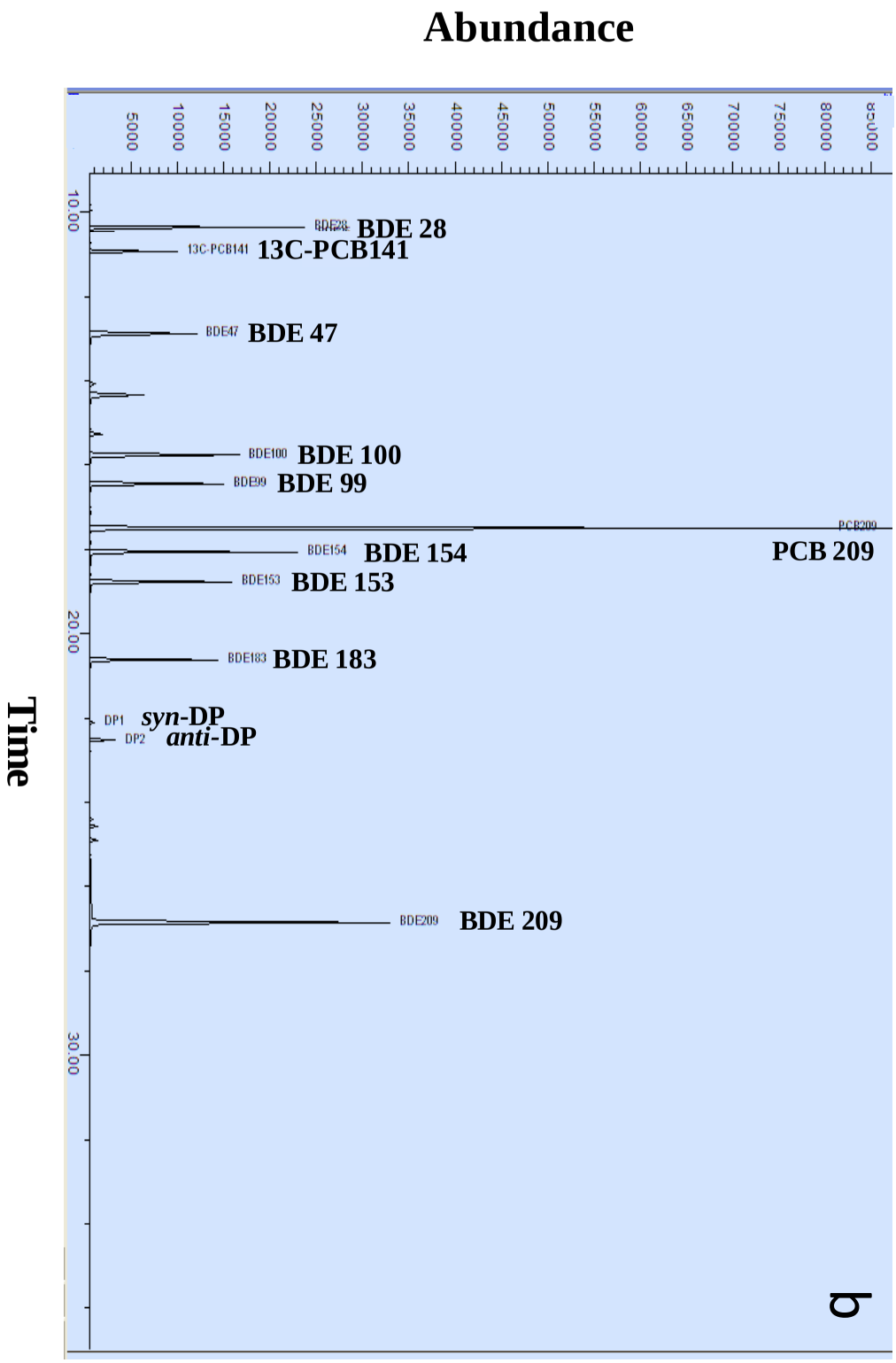
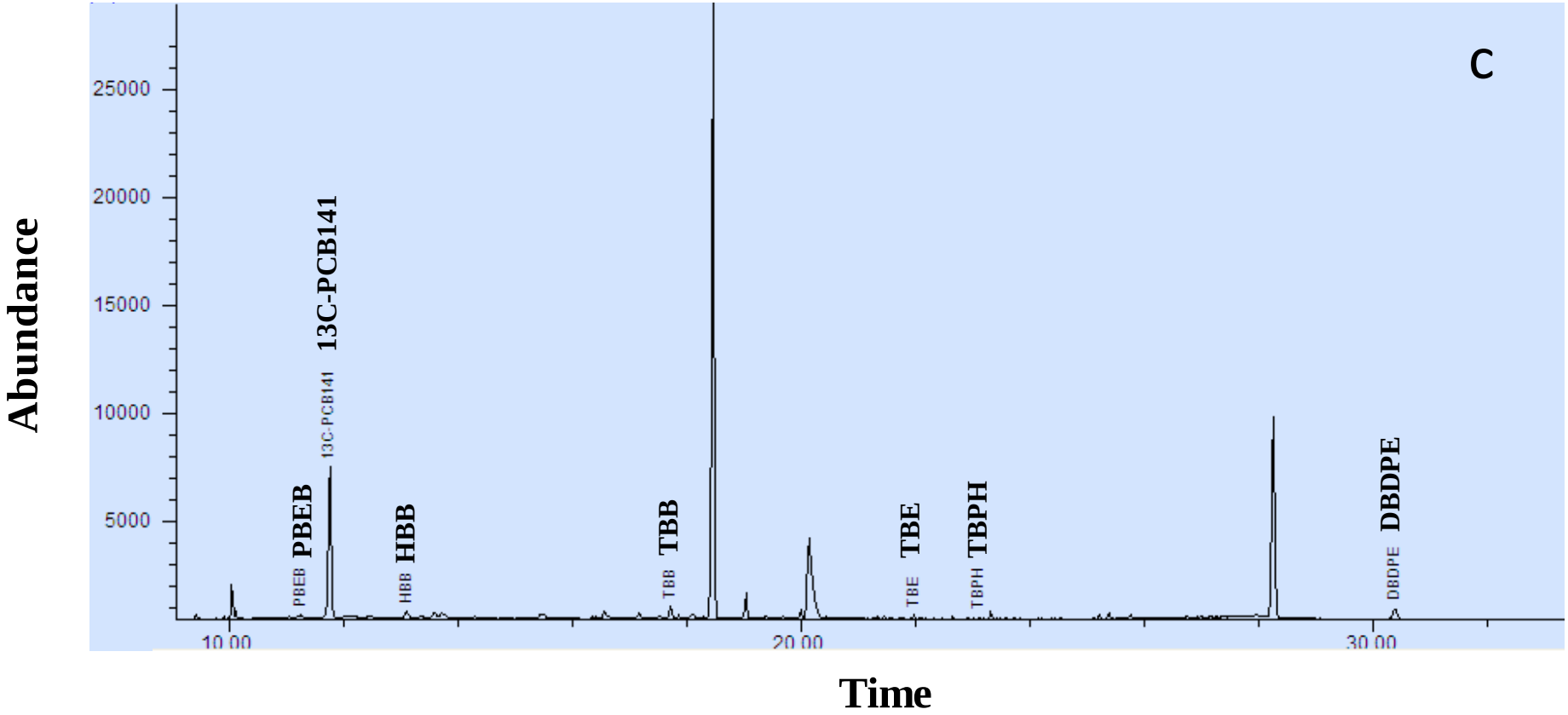
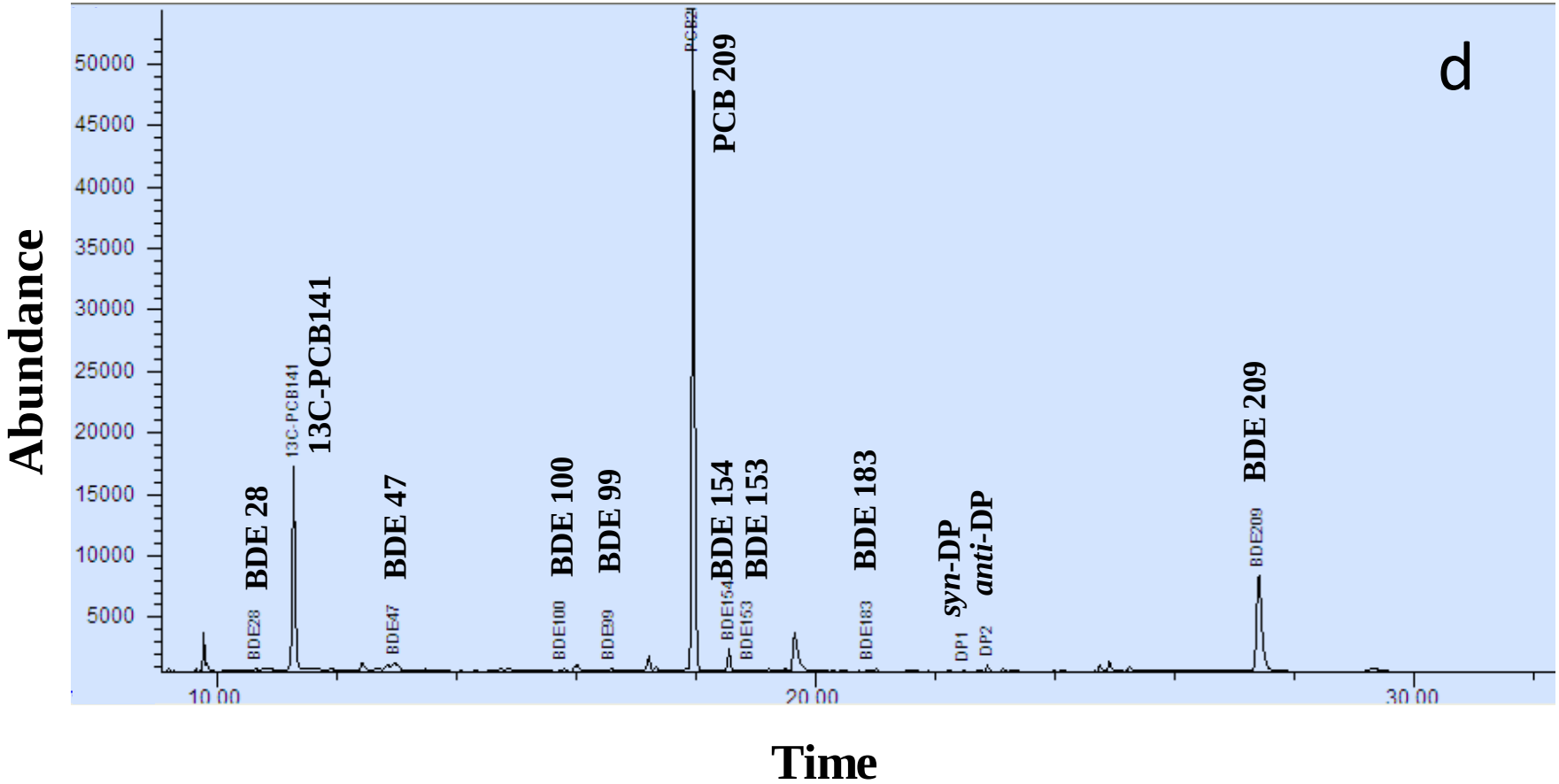

IPSI value
Flame retardants concentrations [pg.g-1]
3 400
DBDPE
Population density
High
Low
Russia
Mongolia
China
North Korea
South Korea
Japan
Bhutan
India
Bangladesh
Myanmar
Vietnam
Laos
0 200 400 600 800 1 000 km
C

Figure S4. The total ion chromatograms of standards and one soil sample.




Abundance
5000
10000
15000
20000
25000
30000
35000
40000
45000
50000
55000
60000
65000
70000
75000
80000
85000
10.00
20.00
30.00
Time
BDE28
BDE 28
13C-PCB141
13C-PCB141
BDE47
BDE 47
BDE100
BDE 100
BDE99
BDE 99
PCB209
PCB 209
BDE154
BDE 154
BDE153
BDE 153
BDE183
BDE 183
DP1
syn-DP
DP2
anti-DP
BDE209
BDE 209
b


C
Abundance
25000
20000
15000
10000
5000
PBEB
13C-PCB141
HBB
TBB
TBE
TBPH
DBDPE
10.00
20.00
30.00
Time



Note: We used two standard samples to make working curves. a and b are NBFRs and PBDEs ion chromatograms of standards (200 ppb), respectively. c and d are NBFRs and PBDEs ion chromatograms of a soil sample from site 6, respectively.