Supporting Information

Polycyclic aromatic hydrocarbons (PAH) in Chinese forest soils: profile composition, spatial variations and source apportionment

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S 1. Statistical methods

In order to identify independent sources tracers (individual components), we used principal component analysis (PCA). The PCA was carried out with varimax rotation (Kaiser-Normalization), and initially the components having eigen values greater than 1 (following Kaiser's rule) were extracted. The influence (and association) of BC and/or TOC on PAHs (individual and sum-components) was analyzed using regression models. All the analyses were carried out with SPSS 18.0 and MS-Excel software, while the geographical distribution of PAHs concentrations in study area was plotted using ArcGIS 9.2 software.

S2. BC and TOC analysis

Total organic carbon was analyzed according to the procedure explained by Chen et al. (2009)[1]. In brief, treatment with 10% HCL was carried out for 3g of soil sample which was freeze-dried, ground and sieved, to remove the inorganic carbon, and then samples were washed three times with deionized water, and dried overnight at 60 °C. The overall standard deviation of measurements was better than 3% (n=3). TOC contents were determined by using the TOC-VCPN with the solid sample module (SSM- 5000A; Shimadzu, Japan).

For black carbon (BC) detection, the chemo-thermal oxidation (CTO-375) method described elsewhere[2,3] was used. Briefly, the dried soil samples (2-3 g) were exposed to thermal oxidation (375 C, 18 h) in a muffle furnace under constraint air flow. They were then digested with 1 N HCL[4]. The residual organic carbon content was determined as BC by using a TOC analyzer (SSM- 5000A; Shimadzu, Japan).

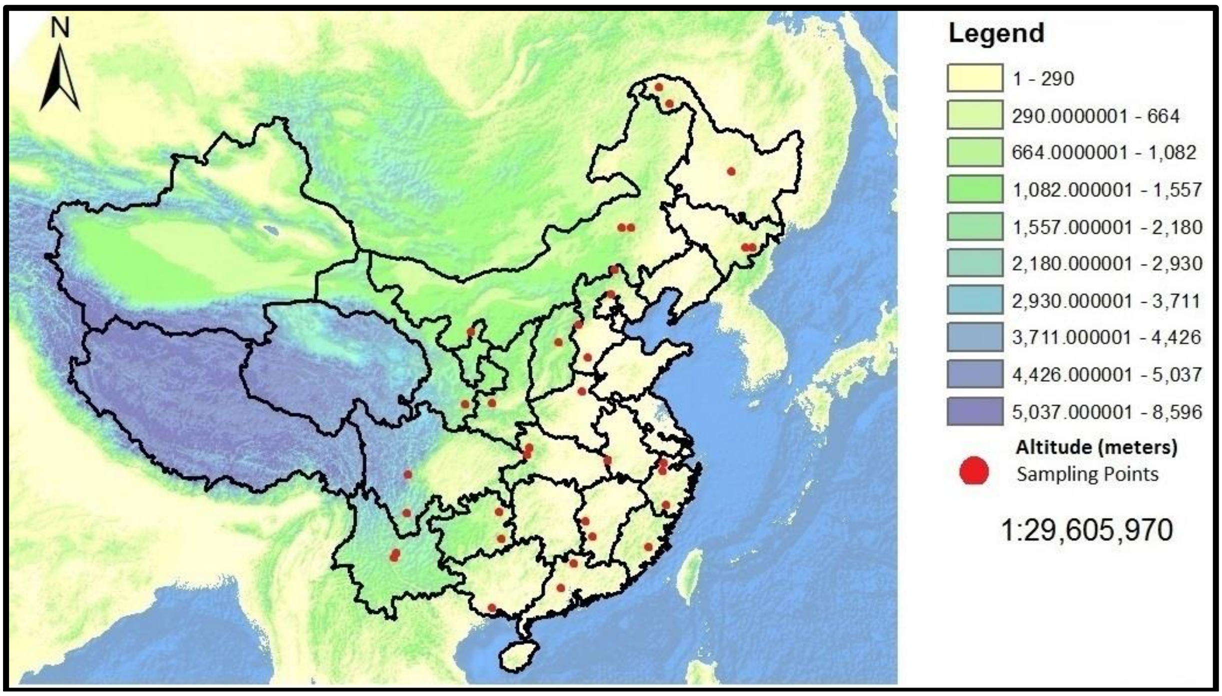

Figure S1: Altitudinal map displaying sampling sites across the forests of China. (The background map was made using ArcGIS 9.3 by one co-author)

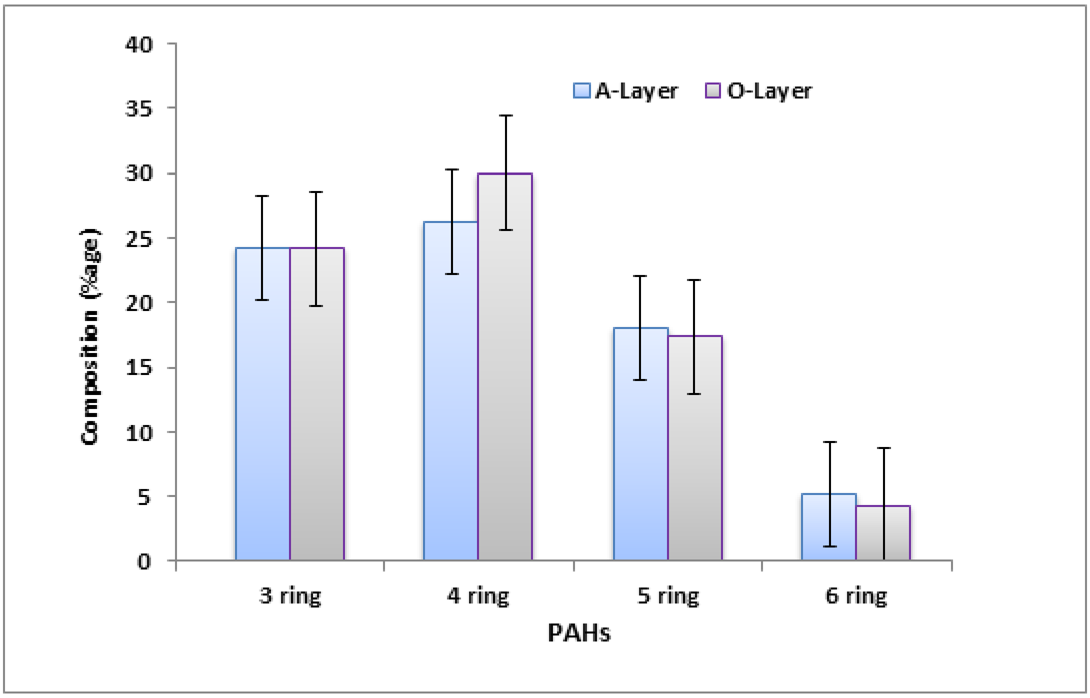

Figure S2: Composition profile of total PAHs (O- & A-horizon)

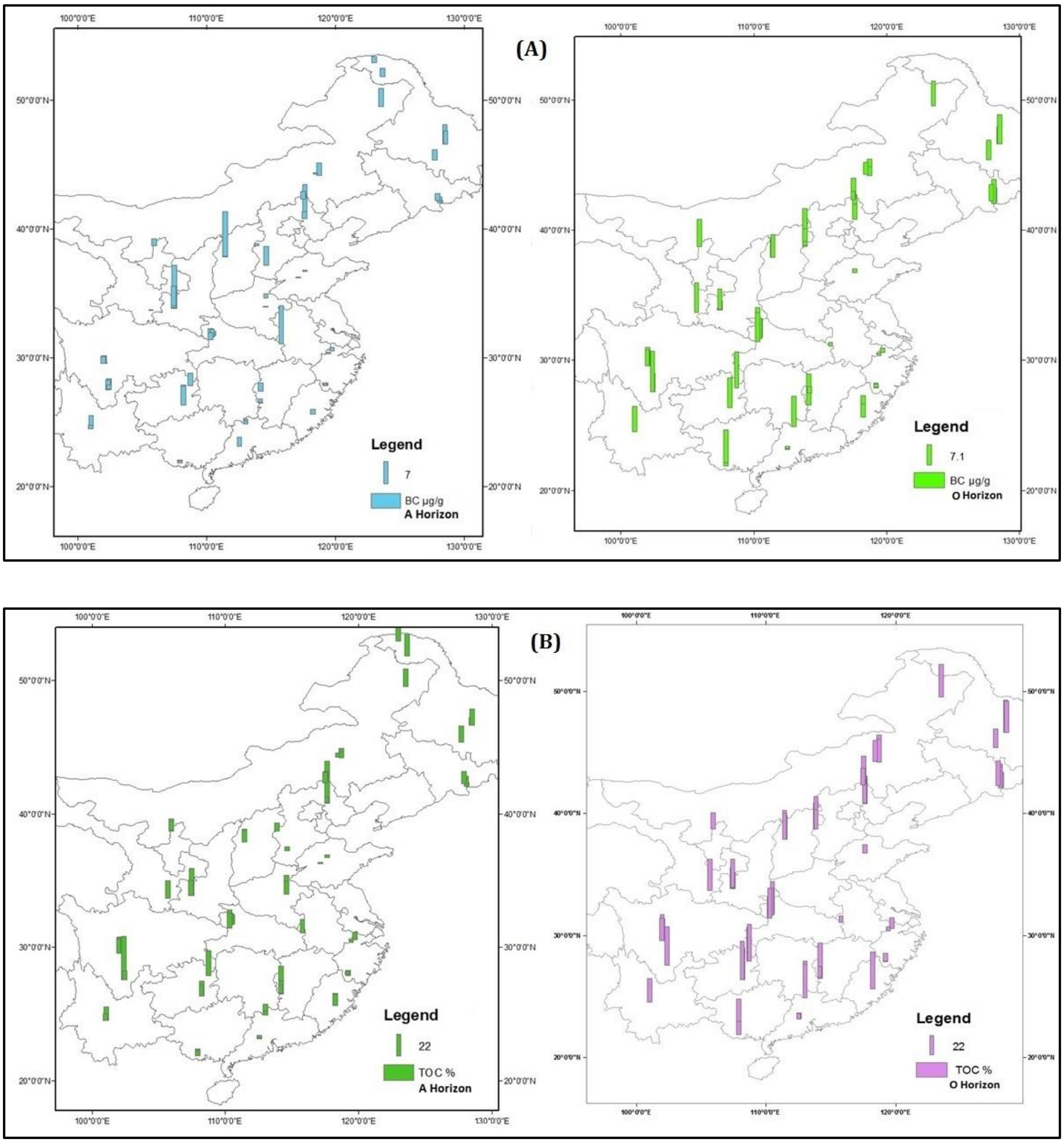

Figure S3: Spatial distribution of BC (A) and TOC (B) in the studied soil samples. (The background map was made using ArcGIS 9.3 by one co-author)

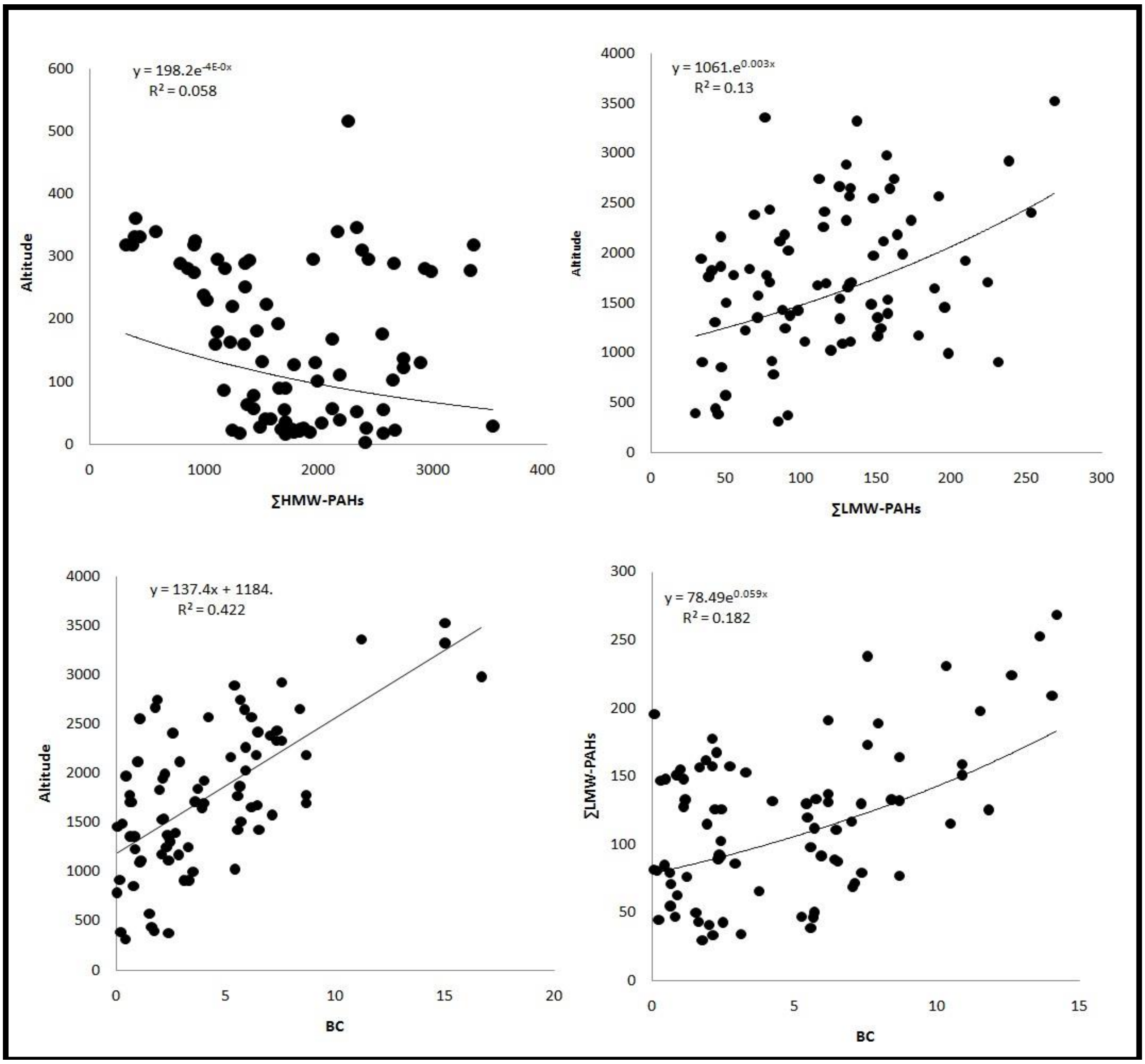

Figure S4: Topographic deposition trend of LMW vs. HMW-PAHs in Chinese forest soil

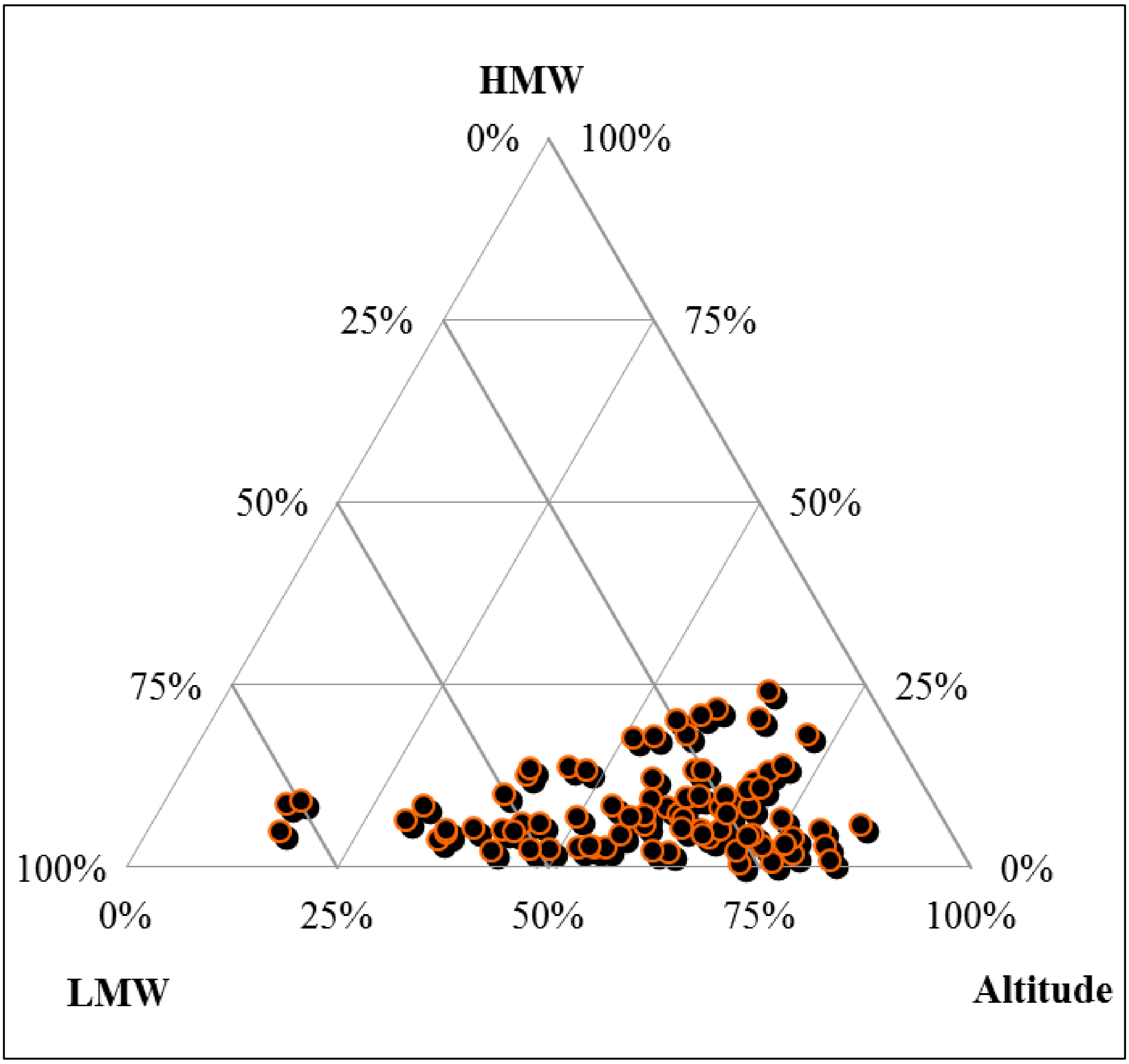

Figure S5: Triangular of Altitude distribution (in percentage) of the LMW and HMW- PAHs

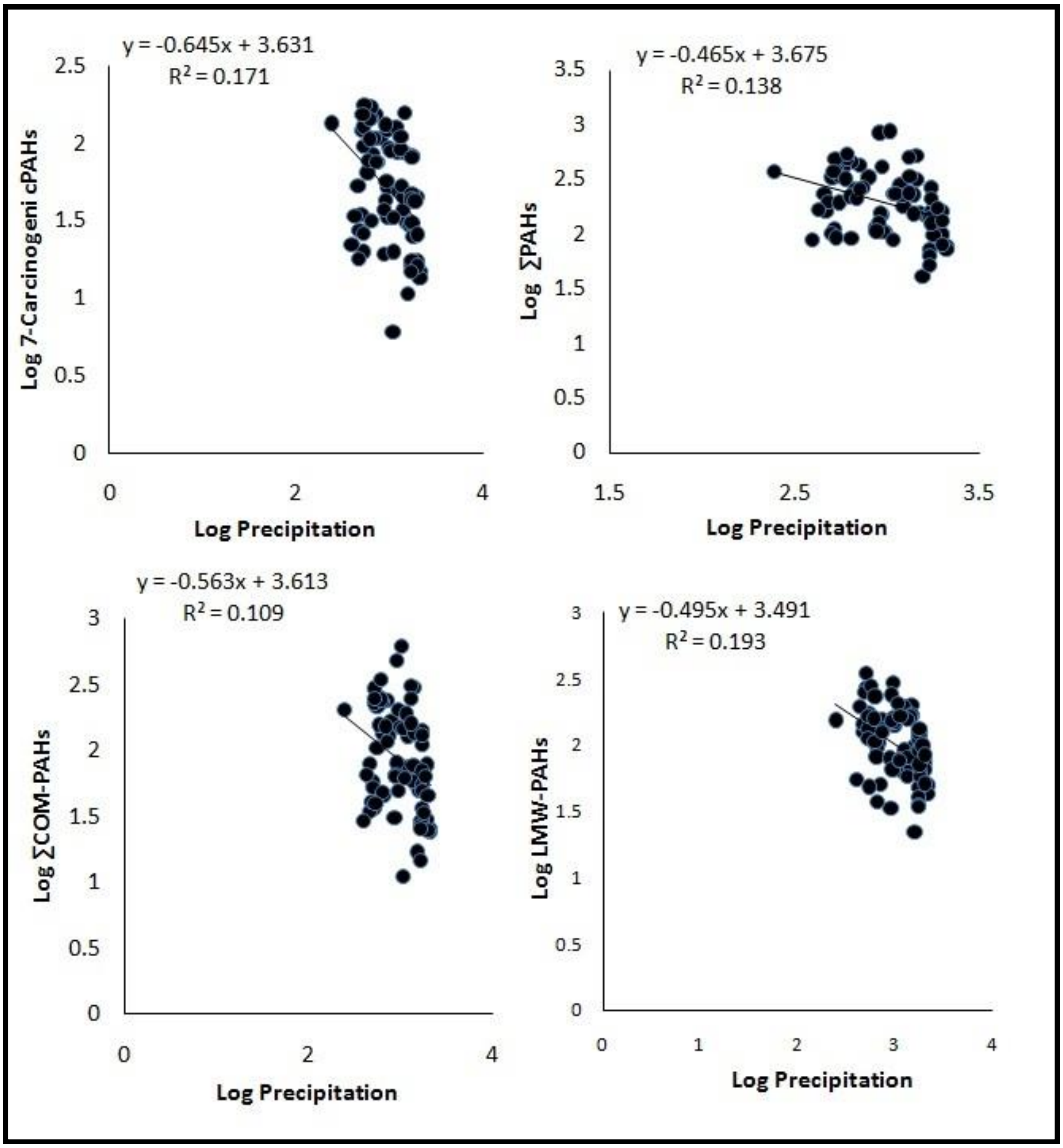

Figure S6: Influence of precipitation pattern on the distribution of PAHs

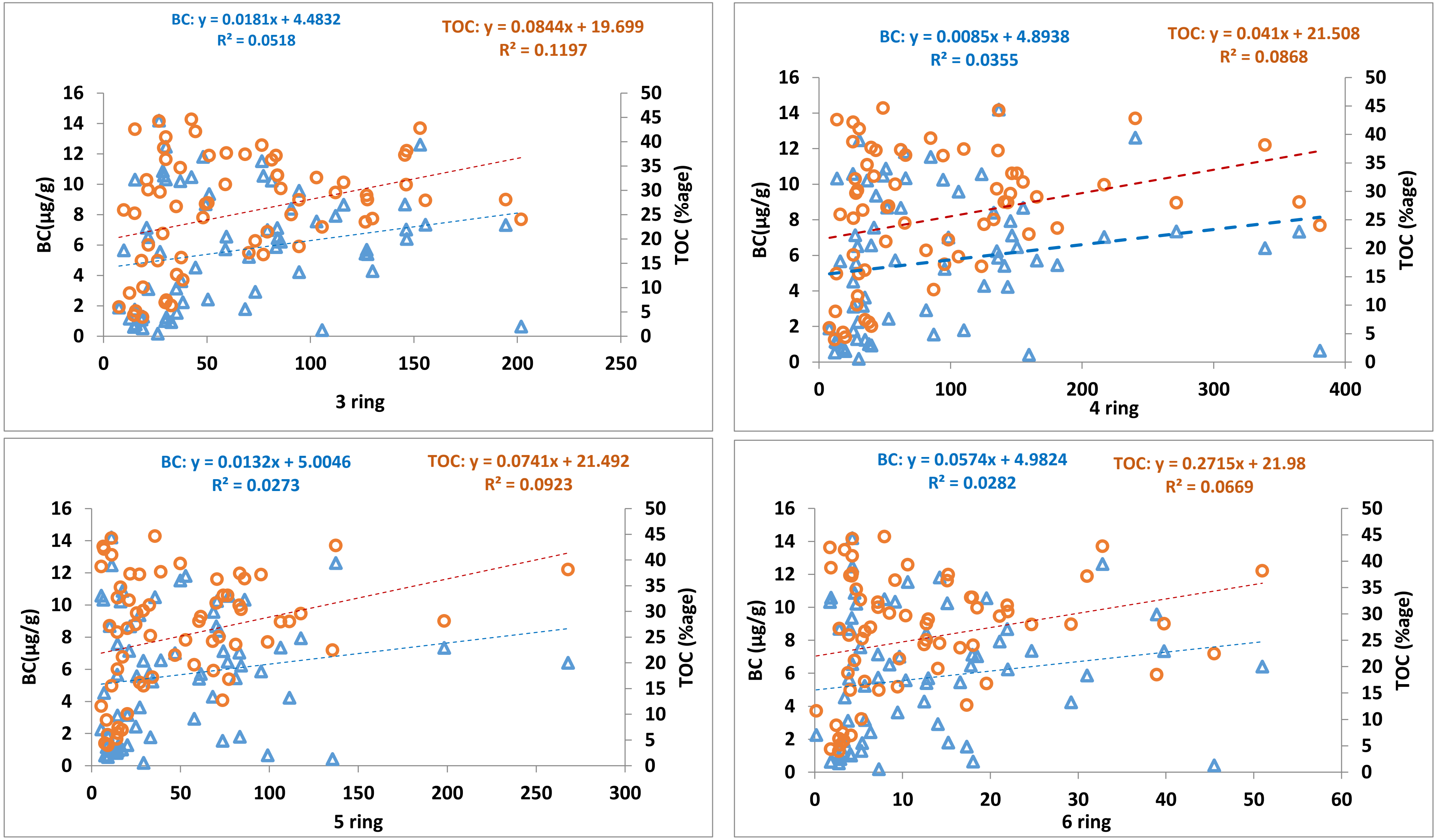

Figure S7 (a): Regression plots of TOC and BC against PAH benzene ring numbers for O- Layer soils

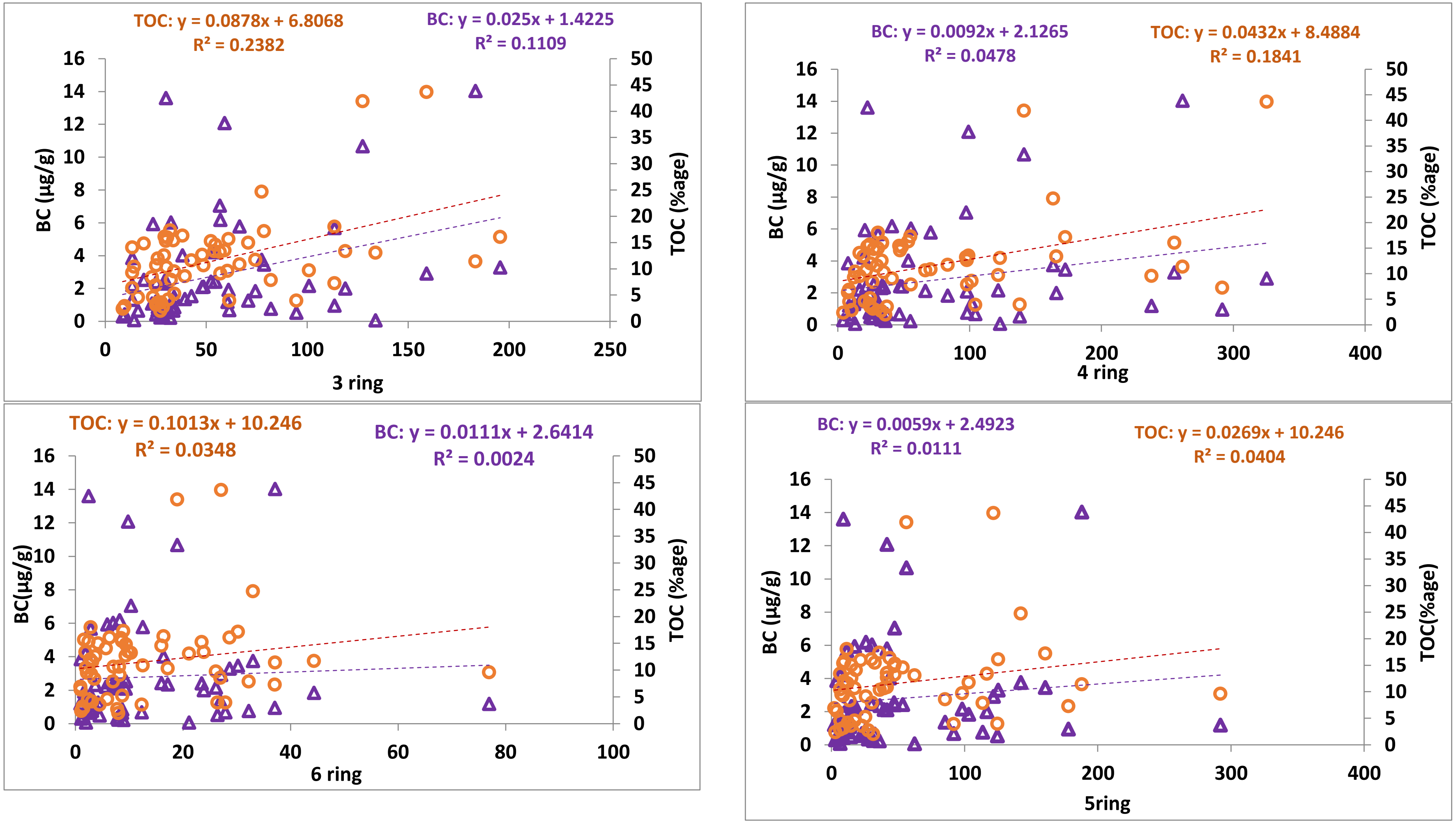

Figure S7 (b): Regression plots of TOC and BC against PAH benzene ring numbers for A- Layer soil

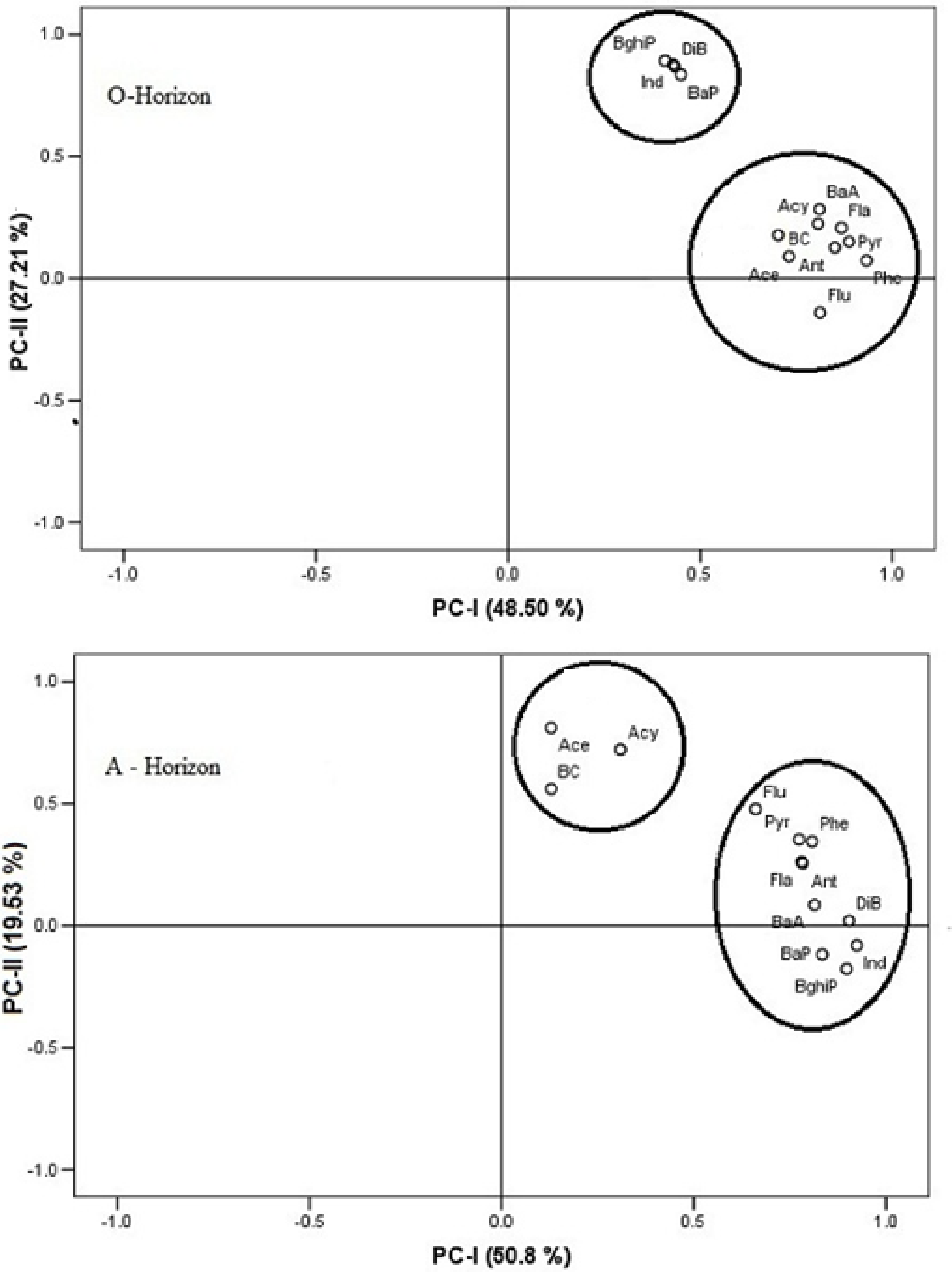

Figure S8: PCA bi-plot of PAHs congeners and BC contents in O- & A-horizon in soils of Chinese forests

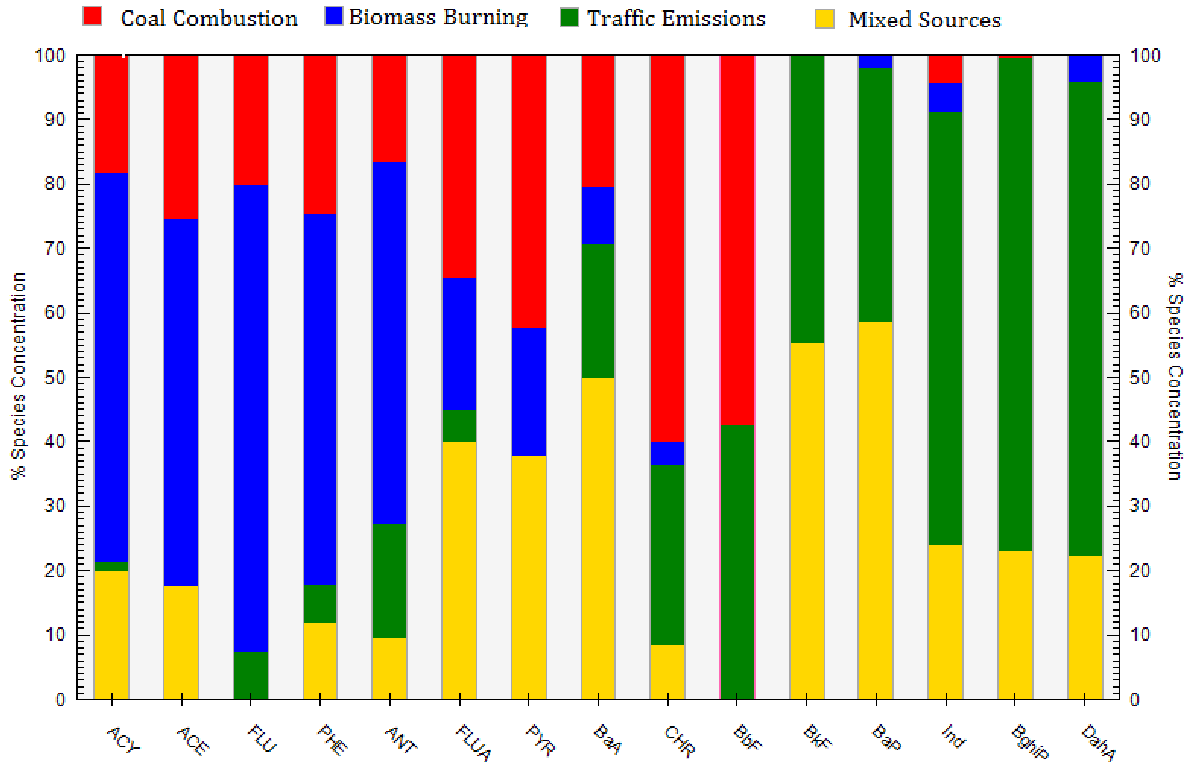

Figure S9: Fingerprints of four factor profiles on each PAHs congener in Chinese forests

Table S2: Comparison of PAH concentrations (ng g^{-1}dry weight) in the mountainous soils of different regions

Country	Sampling site	Sampling year	Total PAH	PAHs (ng g^{-1})	Reference
UK **Norway**	Remote woodland	1998	15	580 (42–4850) 243 (42–750)	(Nam et al., 2008)[5]
Austria	Remote forest	1993	16	210[a] (68–1342)	(Weiss et al., 2000)[6]
Switzerland	Forest	2002	16	251 (98–578)	(Bucheli et al., 2004)[7]
Brazil	Tropical Forest Top-soil	2001	21	32	(Krauss et al., 2005)[8]
Germany	Forest Soils	2013	16	105-14889	(Ainer et al., 2013)[9]
Pearl River Delta, Southern China	Urban, Sub-Urban, Rural-Forests	2013	16	152, 74, 26	(Xiao et al., 2014)[10]
China	Forest Soils O-layer A-layer	2013	15	222 (28-804) 168 (10-670)	This study

[a] median of PAHs Concentrations

Table S3: General Topographic distributions of PAHs and BC in the forest cover of China

Elevation	>2000m		500-2000m		<500m	
	Mean±SD	**Min-Max**	**Mean±SD**	**Min-Max**	**Mean±SD**	**Min-Max**
Acy	2±1	0.7-5.5	3.1±1.7	0.6-15.5	2.2±0.9	0.4-8
Ace	3.3±1.7	0.5-16.6	3±2.9	0.4-19.6	2.3±2.8	0.2-13.7
Flu	6.4[a]±1.8	1.3-21	11.7±7.5	2.1-32	5.6±1.8	0.8-276
Phe	38[a]±16.9	6.3-163	55 [b] ±21	7.6-148	31±17.3	4.9-144
Ant	2.8[a]±1	0.5-9	5 [b] ±3.3	0.7-15	2.6±1.4	0.6-10
Fla	45[a]±23	4.3-216	39 [b] ±8	3.3-149	27.9±11	3.6-132
Pyr	19.7±150	2.6-110	22 [b] ±13	1.9-77	15.2±6.8	1.8-72.5
BaA	9±5.4	1-43.7	13.5 [b] ±5	1.1-64	8.1±5.4	0.5-62.3
Chr	16.7±11	0.8-91	16 [b] ±12	1.4-96	8.4±3.8	0.5-46.5
BbF	22[c]±16	0.6-120	19 [b] ±11	1.6-94	9.6±4.4	0.7-42
BkF	9±1.8	0.1-49	9 [b] ±7.2	0.1-26	5.7±3.7	0.5-52
BaP	12[c]±7.1	0.2-69	10 [b] ±8.4	0.1-29	6.2±2.7	0.5-62
Ind	15±6.8	0.9-77	14.9 [b] ±7	0.2-46	7.9±4	0.9-37
BghiP	7.8[c]±4	0.3-47	6.9 [b] ±6.2	0.1-22	3.7±1.8	0.5-21
DahA	11.9±4	0.6-71	11.9 [b] ±8.8	0.2-49	6.3±2.3	0.7-35
$\sum_{16}$PAHs	276±56	87.9-865	296 [b] ±163	53.9-870	189±60	24.9-721
∑LMW-PAHs	107[c]±221.9	33.6-340	131 [b] ±58	29.9-268	90±221.7	8.2-312
∑HMW-PAHs	124[c]±48	7.4-516	125 [b] ±94	17.9-454	71.1±276.7	7.5-375
TOC	20.5±6	2.9-44	21.5 [b] ±5.9	3.6-45	15.2±5.2	2.4-42.6
BC	4.8±1.4	0.1-14	4.9 [b] ±1.5	0.5-14	3.5±0.8	0.1-12.5

[a]Significantly higher in >2000m, [b] Significantly higher at altitude between 500 and 2000 m, [c]Significantly higher at >2000 than at 500

Table S4: Forest based ∑PAHs (ng/g) distribution in Chinese forest soils

	Coniferous forest	Broad-leaved	Theropencedrymion	Birch	Mesophorbium	Macrophanerophytes	Bamboo	Dahurian larch
Min	751	25	72	191	-	-	-	-
Max	4539	3601	2684	2587	-	-	-	-
Mean	2163	1509	1566	1600	1763	143	831	2356
SD	900	945	598	793	-	-	-	-

Table S5: Pearson correlations based on molecular weight PAHs versus BC and TOC

O-Layer	**TOC**	**BC**	**LMW**	**HMW**	**Precipitation**	**Temperature**
TOC	1					
BC	0.767**	1				
LMW	0.497**	0.411**	1			
HMW	0.307*	0.186	0.769**	1		
A-Layer	**TOC**	**BC**	**LMW**	**HMW**		
TOC	1					
BC	0.370**	1				
LMW	0.189	0.630**	1			
HMW	0.332*	0.155	0.158	1		
Precipitation	-0.1	-0.144	0.156	0.8	1	
Temperature	0.560**	-0.65	0.142	0.15	0.672**	1

** p< 0.01 * p< 0.05

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