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Nationwide increase of polycyclic aromatic hydrocarbons in ultrafine particles during winter over China revealed by size-segregated measurements

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Text S1 Theoretical relationship between meteorological parameters and PAHs.

PAHs are semi-volatile compounds (SVOCs) and can partition between the gas and particle phases. The gas-particle (G/P) partitioning behavior of atmospheric PAHs can be described as equations (1) and (2) (Pankow, 1994).

$$K_{p,OM} = \frac{RT}{10^6 \overline{MW_{OM}} \zeta_{OM} P_L^o} \quad (1)$$

$$P_L^o = P_L^{o,*} \exp[\frac{\Delta H_{vap}^*}{R} (\frac{1}{298.15} - \frac{1}{T})] \quad (2)$$

where $K_{p,\,OM}$ represents the absorptive G/P partitioning coefficient of individual PAH, R (m^3 Pa/ K/mol) is the ideal gas constant, T (K) is the ambient temperature. $\overline{MW_{OM}}$ (g/mol) is the mean molecular weight of organic matter (OM) and is assumed to be 200 g/mol (Xie et al., 2014), ζ_{OM} is the scale activity coefficient of each compound in the absorbing phase and is usually assumed to be unity. $P_L^{o,*}$ is the vapor pressure of each PAH at 298.15K and ΔH_{vap}^* is vaporization enthalpy of the liquid at 298.15K. Thus, for a specific PAH in a single OM phase at a fixed relative humidity, the G/P partitioning should be driven by ambient temperature only. As Figure 1 showed, the decrease of ambient temperature can cause the increase of $K_{p,\,OM}$. This means that the decrease of ambient temperature would result in the increase of individual PAH in the particulate phase assuming a constant total concentration in the air.

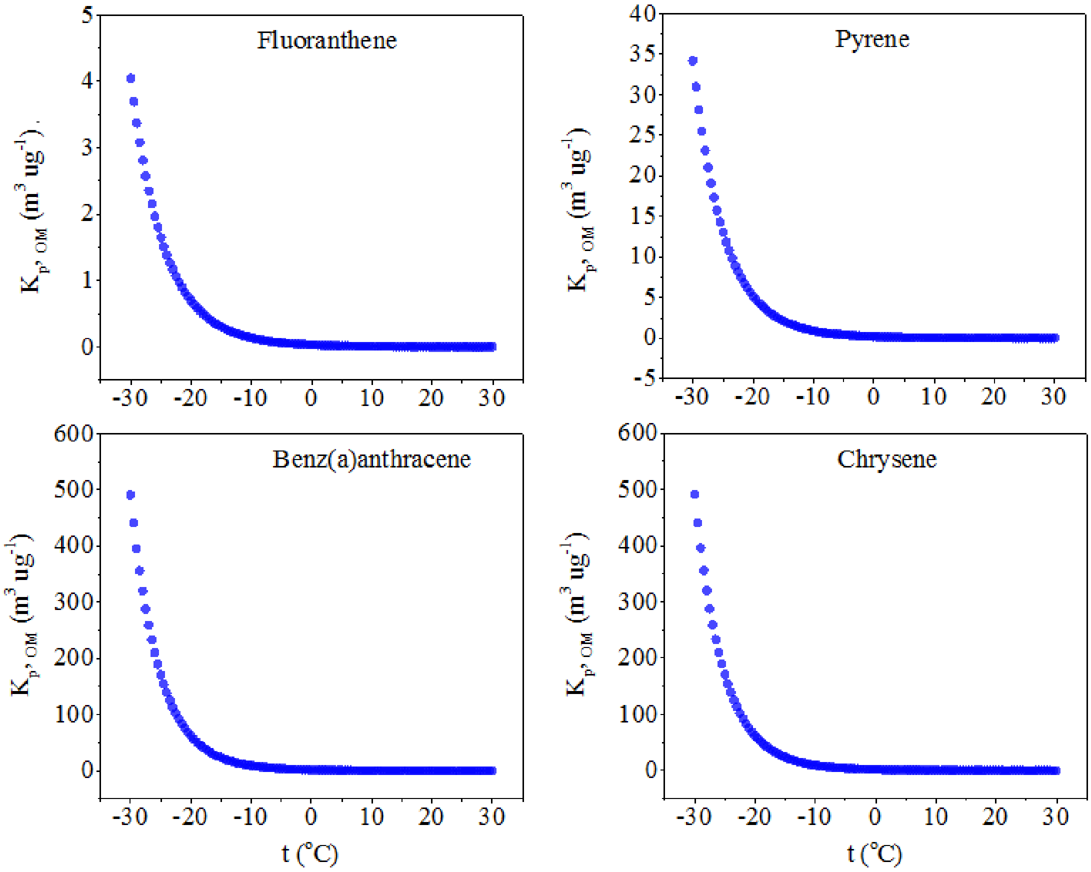

Figure 1 The $K_{p,\ OM}$ ($m^3\ ug^{-1}$) under different temperature.

In the atmosphere, PAHs removal by OH can be described as:

$$\frac{dC_{PAH}}{dt} = -k * [OH] * C_{PAH} \qquad (3)$$

where k is the rate constant for the reaction of a PAH with OH radical, C_{PAH} is the concentration of individual PAH in the air. Solar radiation (SR) directly affects photochemistry in the air. As Figure 2 showed, solar radiation values during our campaign positively correlated with the concentrations of hydroxyl radical [OH] which were estimated based on the empirical equation (4) (Ehhalt and Rohrer, 2000). Thus, the decrease of SR can indeed lower [OH] and accumulate PAHs in the air, resulting in the increase of PAHs concentrations.

$$[OH] = a(JO^1D)^{\alpha}(JNO_2{}^{\beta})\frac{bNO_2+1}{cNO_2{}^2+dNO_2+1} \qquad (4)$$

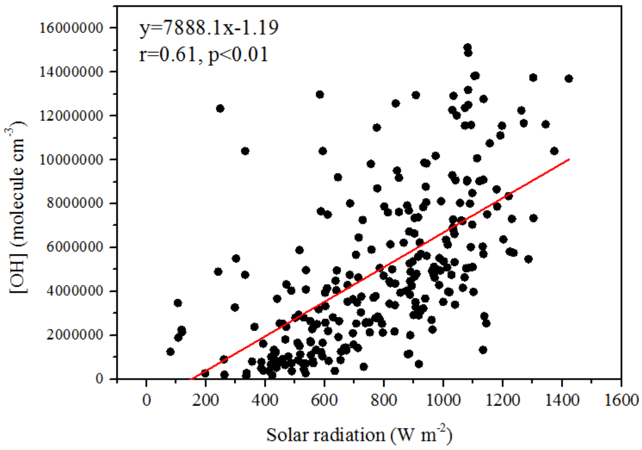

Figure 2 Correlation between OH concentration and solar radiation.

For the influence of boundary layer, low height of boundary layer can inhibit the vertical diffusion of PAHs, which leads to PAHs accumulation and increased concentrations.

References

Ehhalt, D.H., Rohrer, F., 2000. Dependence of the OH concentration on solar UV. J. Geophys. Res.-Atmos. 105, 3565-3571.

Pankow, J.F., 1994. An absorption model of gas/particle partitioning of organic compounds in the atmosphere. Atmos. Environ. 28, 185-188.

Table S1 Detail information of the sampling sites in China.

Sampling sites	Type	Region	Latitude (°N)	Longitude (°E)	Sampling duration
Hailun (HL)	Suburban	Northeast China	47.45	126.92	biweekly 48-hr
Tongyu (TYU)	remote	Northeast China	44.42	122.87	biweekly 48-hr
Beijing (BJ)	Urban	North China	40.01	116.34	biweekly 48-hr
Taiyuan (TY)	Urban	North China	37.87	112.55	biweekly 48-hr
Dunhuang (DH)	Urban	Northwest China	40.13	94.71	biweekly 48-hr
Shapotou (SPT)	remote	Northwest China	37.45	104.95	biweekly 48-hr
Hefei (HX)	Urban	East China	31.86	117.27	biweekly 48-hr
Wuxi (WX)	Suburban	East China	31.40	120.22	biweekly 48-hr
Qianyanzhou (QYZ)	remote	East China	26.75	115.07	biweekly 48-hr
Kunming (KM)	Urban	Southwest China	25.04	102.73	biweekly 48-hr
Xishuangbanna (BN)	remote	Southwest China	21.92	101.25	biweekly 48-hr
Sanya (SY)	Suburban	South China	18.23	109.48	biweekly 48-hr

Table S2 The target compounds and their abbreviations.

NO.	Target compounds	Abbreviations
1	Phenanthrene	Phe
2	Anthracene	Ant
3	Fluoranthene	Flu
4	Acephenanthrylene	Acep
5	Pyrene	Pyr
6	Retene	Ret
7	Benzo(ghi)fluoranthene	BghiF
8	Cyclopenta(cd)pyrene	CcdP
9	Benz(a)anthracene	BaA
10	Chrysene	Chr
11	Benzo(b)fluoranthene	BbF
12	Benzo(k)fluoranthene	BkF
13	Benzo(j)fluoranthene	BjF
14	Benzo(e)pyrene	BeP
15	Benzo(a)pyrene	BaP
16	Perylene	Per
17	Indeno(cd)fluoranthene	IcdF
18	Indeno(cd)pyrene	IcdP
19	Dibenzo[a,h]anthracene	DahA
20	Dibenz(a,c)anthracene	DacA
21	Benzo(b)chrysene	BbC
22	Picene	Pic
23	Benzo(ghi)perylene	BghiP
24	Coronene	Cor

Table S3 Annual average concentrations of individual PAHs (ng m^{-3}) in China.

	HL		TYU		BJ		TY	
	Mean±95%CI	Range	Mean±95%CI	Range	Mean±95%CI	Range	Mean±95%CI	Range
Phe	15.63±13.49	0.72-163.73	2.86±0.79	0.69-7.48	4.96±1.94	1.00-19.99	12.76±7.95	2.54-80.52
Ant	2.82±1.91	0.15-22.78	0.57±0.22	0.03-2.73	0.41±0.14	0.13-1.28	3.51±1.20	0.26-13.04
Flu	17.09±11.63	0.43-120.43	3.37±1.69	0.27-18.19	6.3±4.09	0.55-39.53	22.99±12.25	1.38-113.95
Acep	3.06±1.90	nd-16.80	0.39±0.23	nd-2.51	0.62±0.34	nd-3.05	2.98±1.92	0.10-18.25
Pyr	13.01±8.86	0.26-89.77	2.40±1.26	0.23-13.94	4.44±2.96	0.41-29.65	16.06±8.7	1.02-83.88
Ret	1.68±0.85	0.08-6.42	1.01±0.72	0.04-5.65	1.14±0.62	nd-5.26	1.47±0.83	0.05-8.19
BghiF	3.52±2.20	0.08-21.79	0.71±0.45	0.02-4.88	1.69±1.05	0.10-8.95	3.97±2.06	0.19-17.91
CcdP	1.88±1.31	0.04-12.04	0.25±0.12	0.01-1.09	1.01±0.68	nd-5.68	2.41±1.23	0.17-9.36
BaA	6.21±3.74	0.38-35.54	1.31±0.62	0.37-6.91	3.07±1.85	0.17-16.46	12.13±6.91	0.66-59.7
Chr	7.07±4.20	0.33-42.61	1.88±1.01	0.15-10.91	4.85±2.74	0.46-23.73	18.29±9.03	1.24-74.45
BbF	7.85±4.26	0.28-42.32	1.73±0.72	0.10-7.41	7.11±3.7	0.91-32.17	23.32±9.17	3.08-76.12
BkF	4.37±2.33	0.10-26.41	0.98±0.58	0.02-6.01	2.39±1.12	0.24-8.99	9.78±4.55	0.73-36.44
BjF	1.82±0.99	nd-8.17	0.32±0.18	nd-1.60	0.68±0.41	nd-3.49	2.77±1.47	0.11-11.79
BeP	4.11±1.99	0.17-18.86	1.11±0.45	0.10-4.95	3.68±1.74	0.57-14.42	18.11±7.34	2.13-59.76
BaP	4.55±2.44	0.07-22.95	0.81±0.46	0.02-4.85	2.54±1.51	0.19-12.57	10.99±5.27	0.72-42.03
Per	0.59±0.31	0.02-3.57	0.29±0.09	nd-0.82	0.63±0.29	nd-2.46	1.75±0.74	0.24-6.05
IcdF	1.77±1.04	0.02-10.23	0.33±0.2	nd-2.23	1.14±0.62	0.05-5.13	3.46±1.51	0.35-12.16
IcdP	4.89±2.86	0.05-28.36	0.79±0.45	0.04-4.89	3.4±1.83	0.38-15.25	11.88±5.22	1.16-43.9
DahA	0.96±0.53	nd-4.65	0.24±0.12	nd-1.01	0.58±0.34	nd-2.99	3.32±1.65	0.21-13.64
DacA	0.57±0.24	nd-1.69	0.21±0.07	nd-0.37	0.38±0.16	nd-1.22	1.05±0.61	nd-5.11
BbC	0.92±0.45	nd-3.45	0.31±0.13	nd-0.75	0.55±0.24	nd-1.98	1.74±1.00	0.05-8.38
Pic	0.8±0.40	nd-3.25	0.35±0.16	nd-1.02	0.47±0.25	nd-2.17	2.3±1.24	0.11-10.44
BghiP	3.52±1.89	0.06-18.62	0.66±0.33	0.05-3.71	2.99±1.53	nd-12.85	13.23±5.62	1.26-42.06
Cor	1.50±0.97	nd-9.89	0.50±0.19	nd-1.51	1.09±0.61	nd-5.87	4.32±1.92	0.32-15.39
∑PAHs	108.34±67.74	3.99-717.96	21.92±9.83	3.15-110.8	55.23±29.63	7.53-256.75	204.57±94.07	23.75-819.9
BaP_{eq}	8.28±4.52	0.20-42.68	1.55±0.84	0.09-9.09	5.12±2.88	0.51-23.21	22.24±10.33	1.76-82.89

Continued

	DH		SPT		WX		HF	
	Mean ±95%CI	Range	Mean ±95%CI	Range	Mean ±95%CI	Range	Mean ±95%CI	Range
Phe	5.36 ±1.81	1.35-23.7	4.28 ±0.99	1.04-10.66	3.20 ±0.52	0.86-5.37	2.87 ±0.53	0.86-4.87
Ant	1.82 ±0.49	0.37-4.64	0.52 ±0.14	0.09-1.66	0.59 ±0.09	0.20-1.25	0.70 ±0.20	0.07-2.18
Flu	10.44 ±5.29	1.4-62.21	3.70 ±1.28	0.61-13.54	2.83 ±0.86	0.67-9.86	2.96 ±0.75	1.09-8.09
Acep	1.24 ±0.87	nd-9.76	0.24 ±0.07	nd-0.68	0.35 ±0.09	0.08-1.08	0.33 ±0.08	0.04-0.93
Pyr	7.80 ±4.85	0.73-55.13	1.99 ±0.76	0.23-7.85	1.74 ±0.48	0.42-5.49	1.78 ±0.42	0.49-4.65
Ret	2.01 ±1.2	0.05-12.05	0.27 ±0.08	0.02-0.89	0.23 ±0.07	0.02-0.63	0.52 ±0.63	0.01-7.10
BghiF	3.54 ±1.88	0.13-15.96	0.45 ±0.24	nd-2.63	0.7 ±0.28	0.08-2.81	0.55 ±0.22	0.13-2.09
CcdP	2.34 ±1.54	0.04-14.65	0.26 ±0.15	nd-1.54	0.23 ±0.08	0.03-0.69	0.19 ±0.07	0.03-0.70
BaA	8.46 ±4.65	0.41-37.88	1.35 ±0.71	0.37-8.35	1.00 ±0.31	0.38-3.42	1.02 ±0.29	0.39-2.59
Chr	8.88 ±4.63	0.41-37.07	2.03 ±1.13	0.15-11.93	2.09 ±0.75	0.32-6.87	1.72 ±0.64	0.34-5.77
BbF	14.65 ±7.2	0.64-54.53	2.91 ±1.98	0.05-22.47	3.70 ±1.24	0.38-12.28	2.59 ±0.89	0.28-7.78
BkF	5.18 ±2.78	0.19-26.27	0.94 ±0.53	nd-5.80	1.46 ±0.74	0.08-8.65	1.29 ±0.55	0.06-4.42
BjF	1.93 ±1.06	0.02-7.44	0.27 ±0.13	nd-1.37	0.32 ±0.17	nd-1.94	0.25 ±0.10	nd-0.74
BeP	7.88 ±3.78	0.41-27.5	1.76 ±1.16	0.04-13.12	2.27 ±0.77	0.25-8.61	1.75 ±0.58	0.2-4.61
BaP	5.93 ±3.14	0.08-23.09	0.90 ±0.60	nd-6.65	1.18 ±0.52	0.08-5.42	0.88 ±0.38	0.05-3.27
Per	1.02 ±0.68	0.03-5.34	0.24 ±0.10	nd-0.96	0.27 ±0.09	0.03-0.89	0.28 ±0.07	nd-0.59
IcdF	2.64 ±1.23	nd-9.09	0.45 ±0.31	nd-3.21	0.74 ±0.28	0.06-2.85	0.51 ±0.21	0.04-1.76
IcdP	7.60 ±3.8	0.18-28.03	1.27 ±0.89	nd-9.56	2.04 ±0.74	0.14-6.26	1.41 ±0.63	0.08-5.45
DahA	1.86 ±0.83	nd-6.04	0.37 ±0.25	nd-2.26	0.31 ±0.12	nd-1.11	0.31 ±0.12	nd-0.95
DacA	1.12 ±0.41	nd-3.07	0.13 ±0.07	nd-0.58	0.09 ±0.03	nd-0.22	0.11 ±0.03	nd-0.22
BbC	1.45 ±0.62	nd-4.47	0.23 ±0.14	nd-1.03	0.19 ±0.07	nd-0.61	0.21 ±0.07	nd-0.49
Pic	1.23 ±0.56	nd-4.59	0.35 ±0.20	nd-1.54	0.23 ±0.07	nd-0.6	0.29 ±0.10	nd-0.74
BghiP	5.58 ±2.67	0.21-21.62	1.16 ±0.79	nd-8.45	2.02 ±0.68	0.18-6.43	1.28 ±0.54	0.1-5.01
Cor	1.94 ±0.81	nd-6.31	0.47 ±0.33	nd-3.15	0.87 ±0.25	nd-2.20	0.46 ±0.25	nd-2.40
ΣPAHs	109.56 ±52.18	11.09-460.04	24.51 ±11.11	3.79-132.57	28.09 ±8.53	5.76-92.96	23.64 ±6.72	7.35-58.48
BaP_{eq}	11.92 ±6.19	0.31-44.98	1.79 ±1.29	0.07-15.08	2.54 ±1.02	0.21-10.62	1.91 ±0.77	0.17-6.54

Continued

	QYZ		KM		BN		SY	
	Mean ±95%CI	Range	Mean ±95%CI	Range	Mean ±95%CI	Range	Mean ±95%CI	Range
Phe	2.19 ±0.34	0.83-4.49	2.37 ±0.42	0.70-4.35	1.82 ±0.27	1.12-3.41	2.04 ±0.50	0.73-5.76
Ant	0.82 ±0.22	0.12-1.88	0.51 ±0.12	0.12-1.21	0.91 ±0.15	0.21-1.81	0.97 ±0.16	0.38-1.76
Flu	2.01 ±0.41	0.51-5.74	2.16 ±0.6	0.64-6.17	2.48 ±0.27	1.53-3.81	1.42 ±0.28	0.35-2.78
Acep	0.24 ±0.04	nd-0.47	0.19 ±0.04	nd-0.43	0.27 ±0.04	0.08-0.45	0.14 ±0.02	nd-0.24
Pyr	1.11 ±0.22	0.28-2.84	1.35 ±0.35	0.32-3.47	1.68 ±0.16	1.09-2.44	0.88 ±0.16	0.24-1.86
Ret	0.25 ±0.06	0.02-0.51	0.21 ±0.05	0.08-0.54	0.83 ±0.43	0.03-3.16	0.16 ±0.05	nd-0.42
BghiF	0.25 ±0.09	0.02-0.77	0.39 ±0.13	0.04-1.11	0.19 ±0.02	0.06-0.31	0.08 ±0.02	0.01-0.17
CcdP	0.08 ±0.03	0.01-0.29	0.17 ±0.06	nd-0.69	0.09 ±0.02	nd-0.19	0.03 ±0.01	nd-0.07
BaA	0.58 ±0.09	0.36-1.2	0.90 ±0.18	0.32-2.01	0.52 ±0.07	nd-0.85	0.44 ±0.03	0.34-0.58
Chr	0.82 ±0.26	0.21-2.41	1.41 ±0.45	0.19-4.83	0.75 ±0.11	0.25-1.43	0.34 ±0.04	0.13-0.59
BbF	1.03 ±0.37	0.09-3.14	2.80 ±1.06	0.15-11.94	0.63 ±0.13	0.22-1.56	0.27 ±0.08	0.07-0.82
BkF	0.64 ±0.3	nd-2.74	1.22 ±0.44	0.04-3.77	0.28 ±0.06	0.12-0.68	0.17 ±0.06	nd-0.54
BjF	0.17 ±0.06	nd-0.53	0.21 ±0.08	nd-0.72	0.06 ±0.01	nd-0.16	0.05 ±0.01	nd-0.10
BeP	0.72 ±0.26	0.04-2.30	2.04 ±0.73	0.12-8.34	0.44 ±0.09	0.15-0.99	0.21 ±0.07	0.04-0.61
BaP	0.44 ±0.18	nd-1.47	0.9 ±0.36	0.03-3.59	0.28 ±0.06	0.11-0.74	0.09 ±0.04	nd-0.31
Per	0.18 ±0.06	nd-0.52	0.28 ±0.09	nd-1.12	0.11 ±0.04	nd-0.39	0.19 ±0.07	nd-0.46
IcdF	0.27 ±0.11	nd-0.96	0.36 ±0.10	nd-1.14	0.15 ±0.04	nd-0.44	0.09 ±0.02	nd-0.22
IcdP	0.67 ±0.26	0.03-2.23	1.27 ±0.36	nd-3.75	0.38 ±0.10	0.12-1.08	0.17 ±0.05	nd-0.52
DahA	0.14 ±0.04	nd-0.35	0.23 ±0.07	nd-0.76	0.08 ±0.02	nd-0.2	nd	nd
DacA	0.05 ±0.01	nd-0.10	0.07 ±0.02	nd-0.16	0.10 ±0.03	nd-0.15	nd	nd
BbC	0.08 ±0.02	nd-0.12	0.09 ±0.03	nd-0.26	0.04 ±0.01	nd-0.04	nd	nd
Pic	0.09 ±0.03	nd-0.23	0.15 ±0.05	nd-0.41	0.05 ±0.01	nd-0.08	nd	nd
BghiP	0.58 ±0.22	nd-1.89	1.39 ±0.44	nd-4.02	0.4 ±0.09	nd-1.02	0.17 ±0.05	0.03-0.44
Cor	0.19 ±0.06	nd-0.53	0.44 ±0.13	nd-1.34	0.20 ±0.040	nd-0.37	0.09 ±0.01	0.08-0.11
∑PAHs	13.1 ±3.11	nd-31.9	20.66 ±5.66	4.12-61.94	12.29 ±1.29	7.49-19	7.56 ±0.94	4.35-13.13
BaP_{eq}	0.87 ±0.34	nd-3.00	1.95 ±0.71	0.11-7.44	0.56 ±0.10	0.24-1.3	0.21 ±0.06	0.07-0.57

Table S4 The annual level of atmospheric temperature, solar radiation and boundary layer height in the northern and the southern China.

Region	Sites	Temperature	Solar radiation	Boundary layer height
Northern China	HL	1.4	736.3	474.2
	TYU	3.7	734.9	571.0
	BJ	12.4	651.2	425.1
	TY	11.1	719.9	518.2
	DH	10.9	786.8	453.8
	SPT	10.3	754.0	669.5
Southern China	WX	17.3	729.6	486.6
	HF	17.0	736.3	501.1
	QYZ	20.0	788.7	571.0
	KM	16.6	970.3	658.1
	BN	23.0	988.0	506.6
	SY	23.1	1051.5	520.5

Table S5 The monthly average temperature (℃) in each sampling site in the northern and the southern China.

	Northern China						Southern China					
	HL	TYU	BJ	TY	DH	SPT	HF	WX	QYZ	KM	BN	SY
October	14.0	5.8	14.0	10.6	9.4	9.4	17.8	18.2	20.9	16.9	23.7	23.4
November	3.5	-6.5	3.5	1.5	-1.7	0.0	8.0	8.8	12.0	13.4	21.9	22.9
December	-6.5	-19.7	-6.5	-6.8	-6.4	-5.7	2.1	2.2	5.5	9.8	18.6	19.3
January	-4.5	-20.7	-4.5	-5.3	-10.0	-6.5	1.9	3.6	6.2	9.9	18.9	18.7
February	-2.1	-14.8	-2.1	-1.5	-3.7	-2.7	3.9	5.1	13.7	13.5	20.4	21.3
March	7.3	-5.4	7.3	9.0	8.7	9.1	13.4	13.4	14.3	14.6	21.7	20.3
April	15.2	5.0	15.2	15.5	18.3	15.4	19.8	18.4	21.7	18.3	23.9	24.0
May	21.4	17.9	21.4	21.4	20.2	19.4	24.6	24.7	28.2	21.9	26.2	26.3
June	22.1	20.3	22.1	19.9	24.6	20.9	23.0	23.0	26.0	19.6	24.4	25.9
July	25.8	24.4	25.8	22.3	24.4	21.3	30.0	32.5	31.4	20.5	25.3	24.3
August	27.9	22.8	27.9	25.3	25.1	23.7	31.3	31.8	32.0	20.4	25.6	25.5
September	21.9	14.0	21.9	19.4	19.3	18.0	26.5	25.1	27.4	19.4	25.3	25.2

Table S6 The monthly average solar radiation (w/m^2) in each sampling site in the northern and the southern China.

	Northern China						Southern China					
	HL	TYU	BJ	TY	DH	SPT	HF	WX	QYZ	KM	BN	SY
October	540.2	538.7	606.2	583.2	713.0	667.7	535.7	636.5	691.7	847.7	1099.0	1098.0
November	310.5	476.0	493.5	525.5	536.3	562.3	611.3	655.3	877.0	899.0	922.8	1076.8
December	298.8	426.0	389.5	370.8	491.3	424.5	497.5	569.3	406.8	765.0	857.0	939.3
January	412.8	408.0	424.3	374.5	536.0	505.3	249.5	257.8	416.8	777.8	780.5	922.8
February	585.0	603.0	559.8	560.0	649.5	601.5	287.8	309.3	624.5	949.8	857.5	1067.5
March	824.5	886.0	683.8	849.5	788.0	847.3	729.3	766.5	893.0	982.0	952.5	807.3
April	903.5	925.2	894.5	997.7	904.8	939.8	928.8	953.2	666.8	1203.0	914.8	1177.3
May	1088.3	1001.5	772.5	1036.0	953.0	994.0	952.0	994.0	1122.0	1181.8	1159.8	1187.3
June	946.5	852.0	769.0	1116.8	1077.0	1098.5	747.3	499.3	835.0	990.8	741.8	1082.8
July	944.5	727.8	650.3	492.5	952.5	833.5	1078.3	958.3	1153.5	980.3	1032.0	895.5
August	1071.8	932.5	846.3	950.8	953.8	780.5	1104.5	1158.0	1032.0	1200.3	1213.3	1213.5
September	924.0	915.5	625.3	711.8	864.3	744.0	1117.2	932.5	854.5	811.7	1306.7	1064.5

Table S7 The monthly average boundary layer height (m) in each sampling site in the northern and the southern China.

	Northern China						Southern China					
	HL	TYU	BJ	TY	DH	SPT	HF	WX	QYZ	KM	BN	SY
October	438.2	392.5	447.2	499.4	298.3	601.7	520.7	514.4	392.5	448.5	440.1	608.0
November	398.4	555.1	478.9	688.4	132.9	375.5	301.8	459.7	555.1	671.3	347.2	538.2
December	198.5	235.2	155.3	145.4	96.5	140.9	372.4	448.4	235.2	553.4	438.3	635.3
January	108.2	128.4	169.2	232.9	106.6	181.4	322.4	423.4	128.4	704.0	565.0	443.5
February	187.6	256.9	378.5	406.9	133.4	314.9	586.7	577.3	256.9	792.5	612.5	369.0
March	267.1	769.3	481.8	690.4	356.1	743.8	303.7	343.5	769.3	839.3	725.6	527.6
April	663.9	841.7	772.6	813.9	541.7	931.0	661.4	431.4	841.7	980.3	818.3	420.0
May	746.0	1000.3	520.4	590.4	843.3	1095.1	608.6	619.6	1000.3	846.9	611.5	399.2
June	848.0	721.8	486.4	846.1	938.4	1216.6	367.0	355.6	721.8	587.2	342.2	519.6
July	692.0	622.3	314.2	341.1	793.0	784.9	611.5	499.4	622.3	520.6	242.0	666.4
August	476.0	764.3	382.6	471.5	728.9	798.3	650.3	564.1	764.3	523.4	451.5	660.5
September	589.3	517.8	328.8	353.0	510.0	753.5	616.5	616.1	517.8	374.2	362.6	464.7

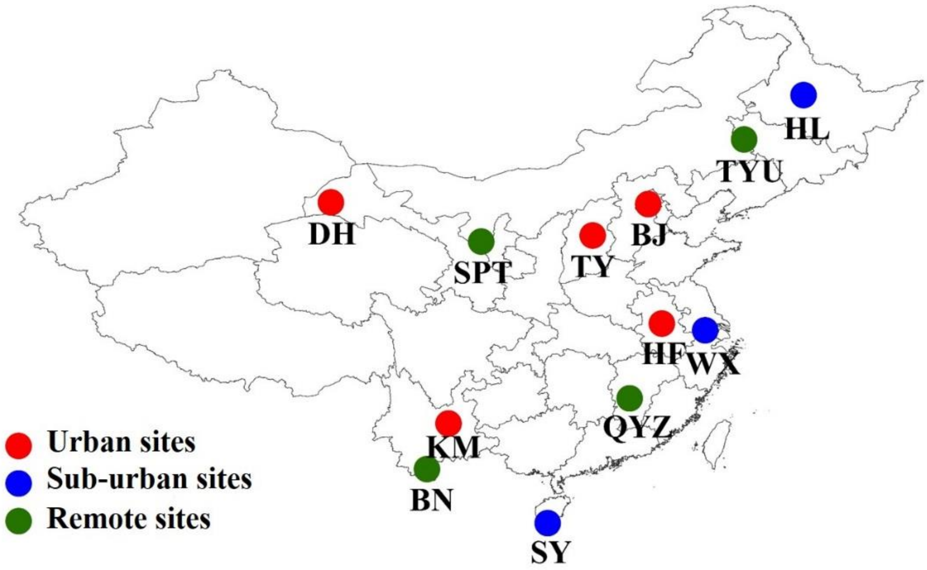

Figure S1 Sampling sites in China, including five urban sites: Beijing (BJ), Taiyuan (TY), Hefei (HF), Kunming (KM), and Dunhuang (DH), three suburban sites: Hailun (HL), Wuxi (WX), and Sanya (SY), four remote sites: Tongyu (TYU), Shapotou (SPT), Qianyanzhou (QYZ) and Xishuangbanna (BN).

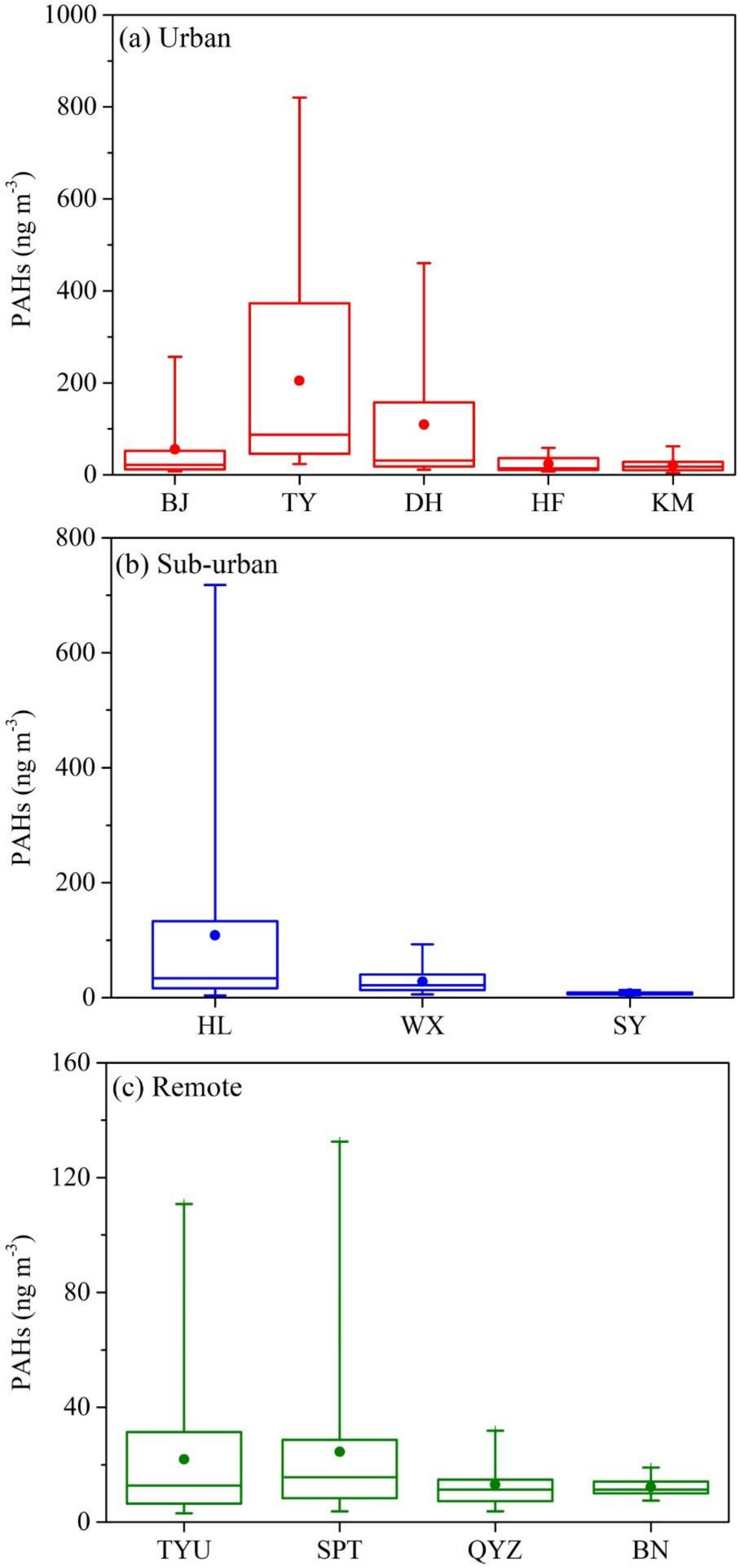

Figure S2 Concentration of $\sum_{24}$PAHs at urban, suburban and remote sites.

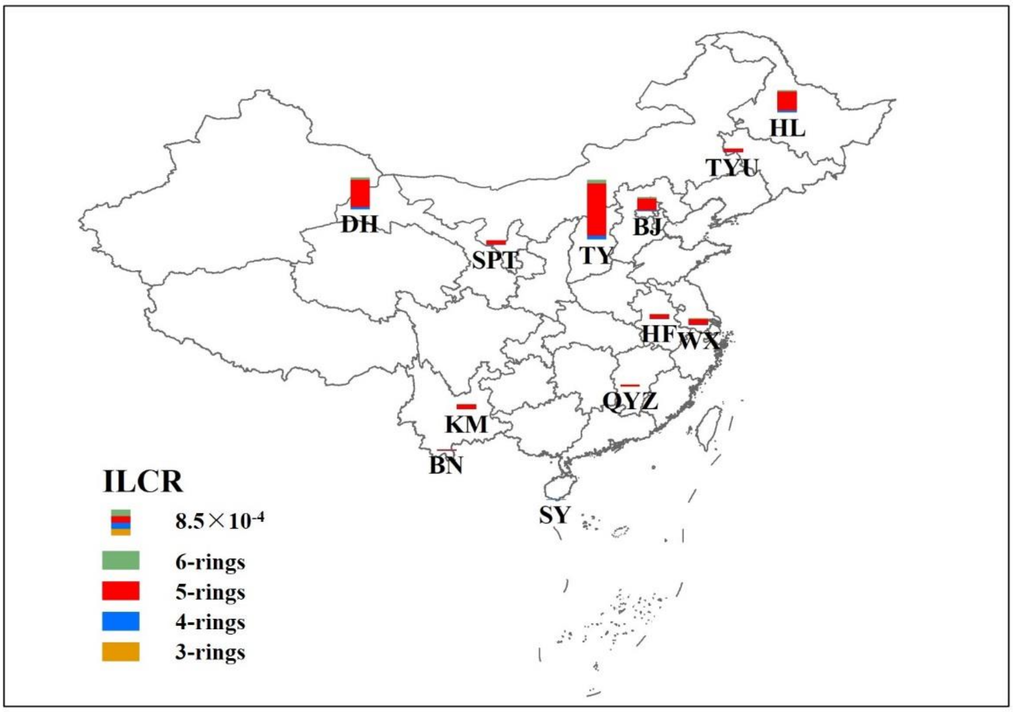

Figure S3 Annual averages of ILCR at 12 sites over China.

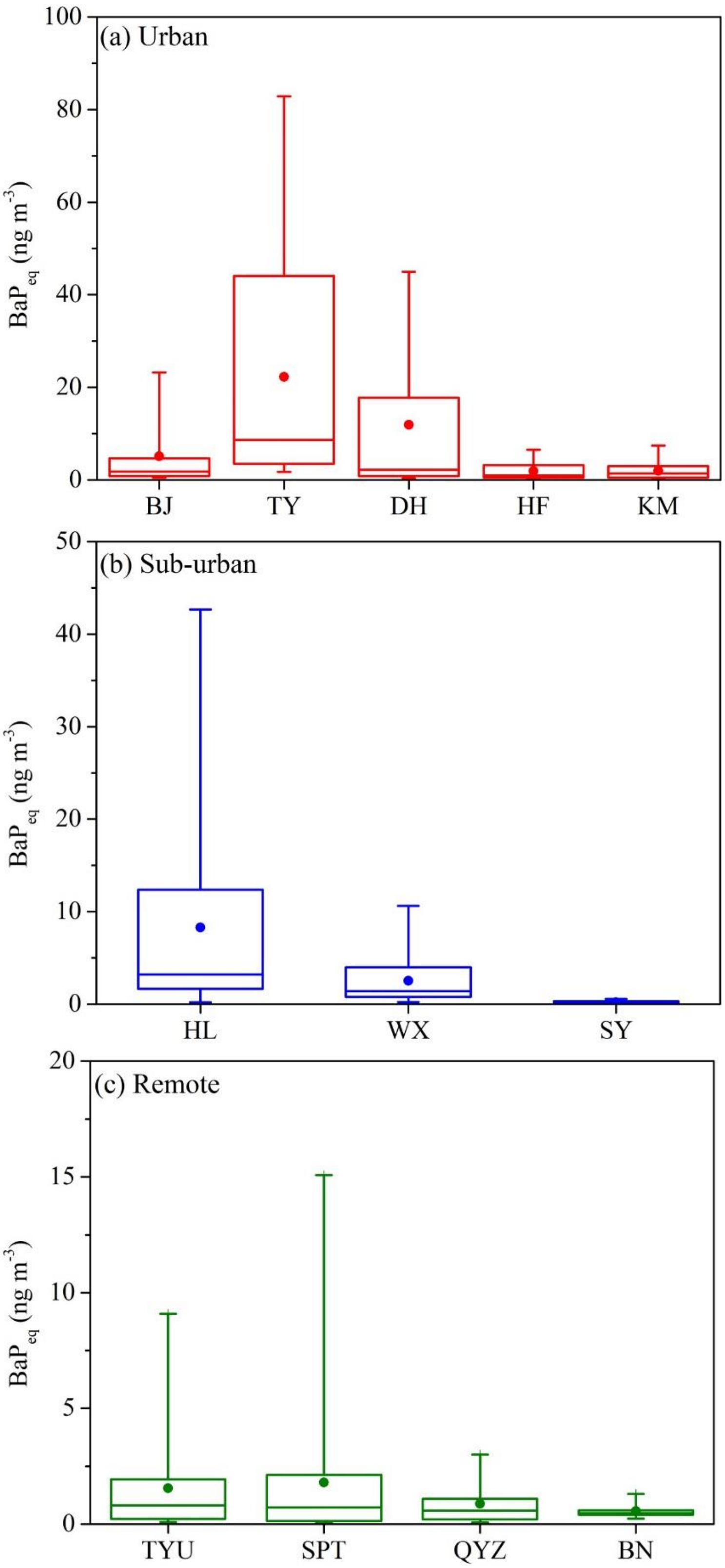

Figure S4 Concentration of BaP$_{eq}$ at urban, suburban and remote sites.

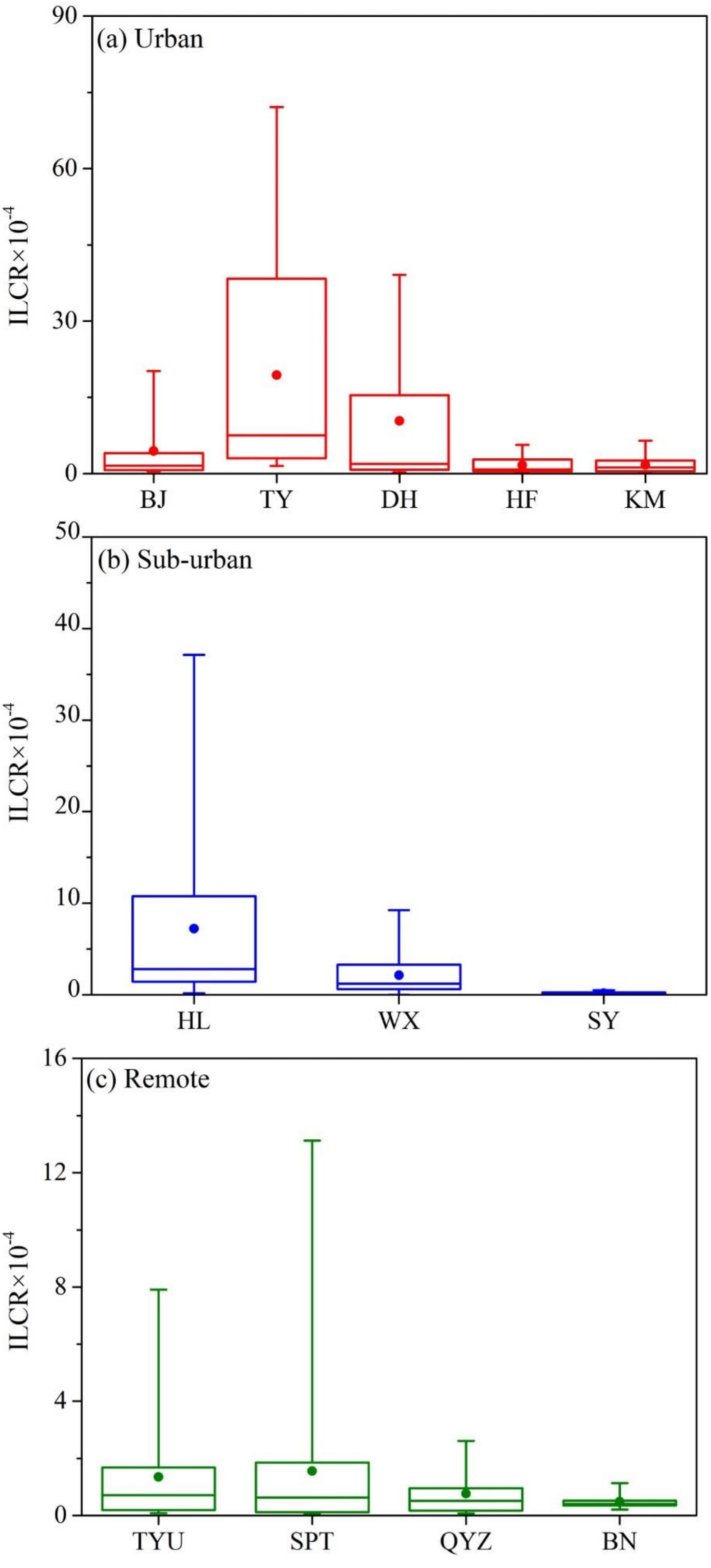

Figure S5 ILCR at urban, suburban and remote sites.

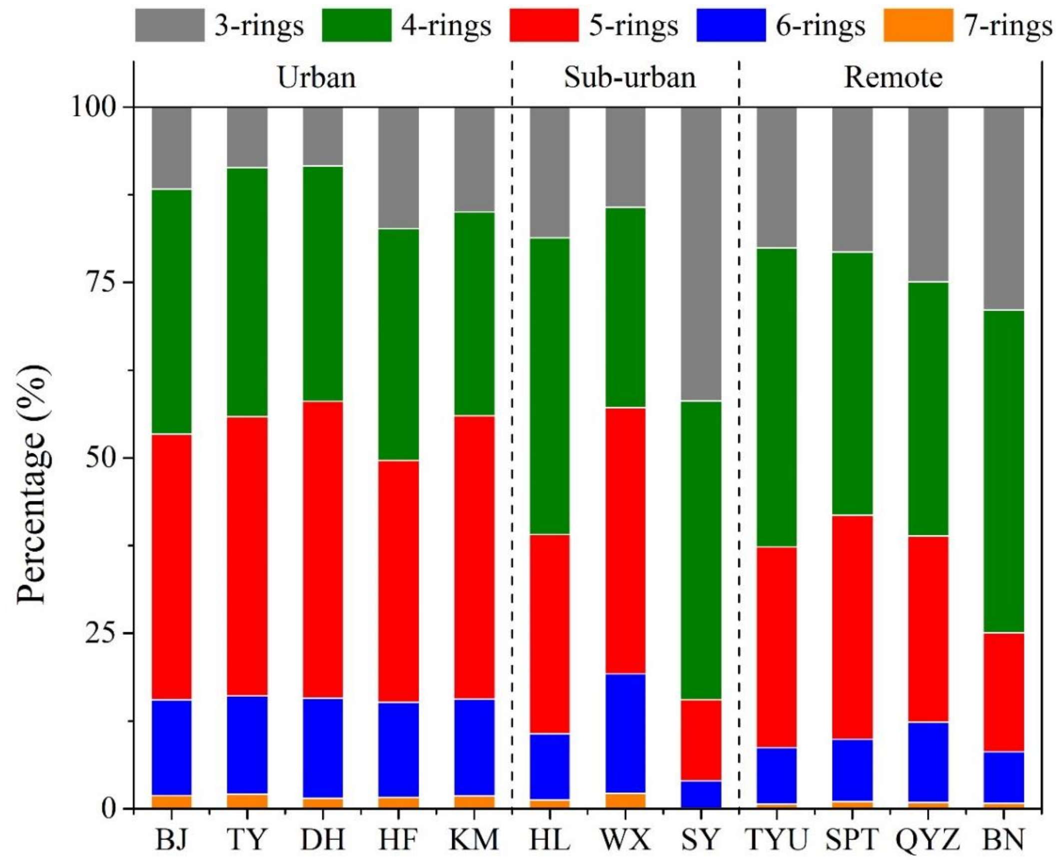

Figure S6 PAHs composition at urban, suburban and remote sites.

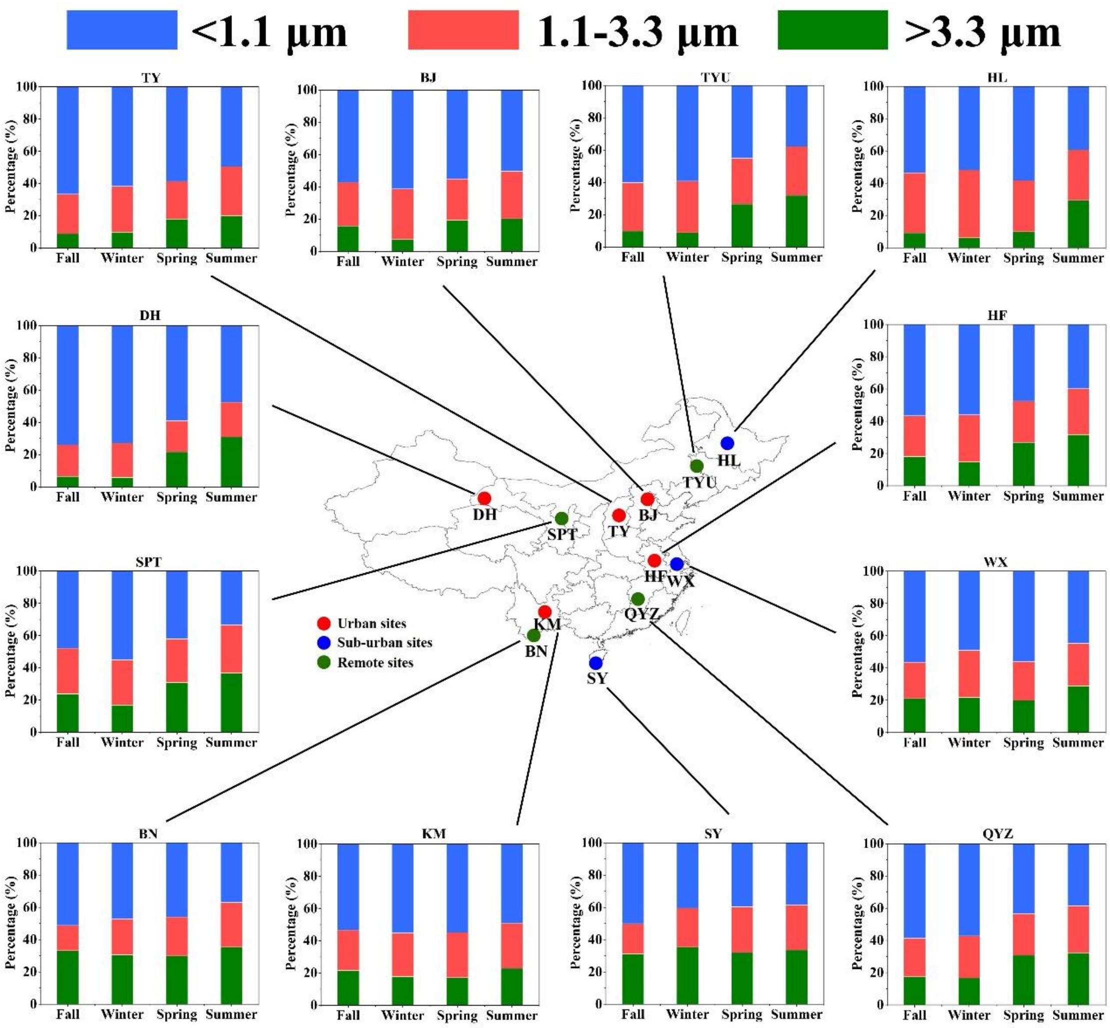

Figure S7 Size distribution of PAHs in different season over China.

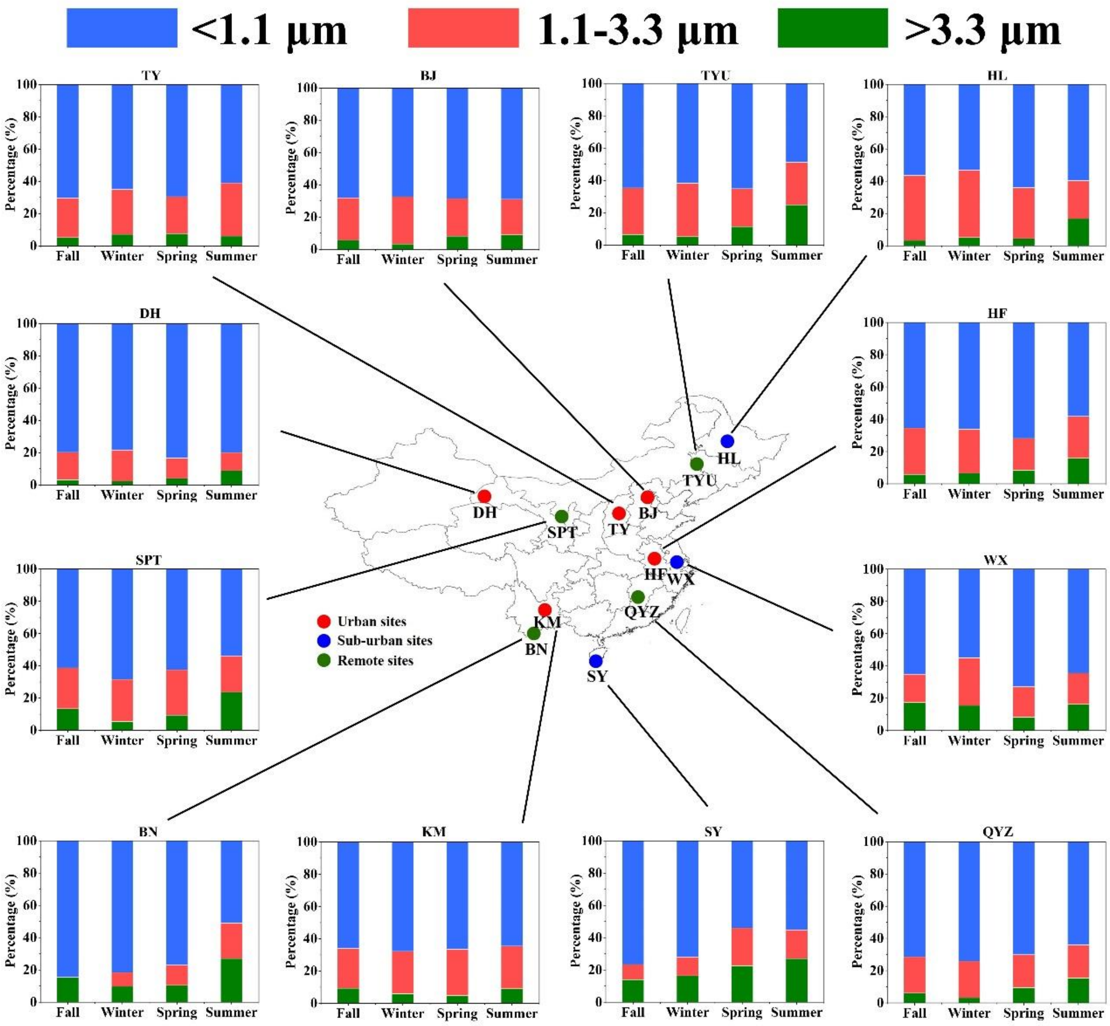

Figure S8 Size distribution of BaP_{eq} in different season over China.

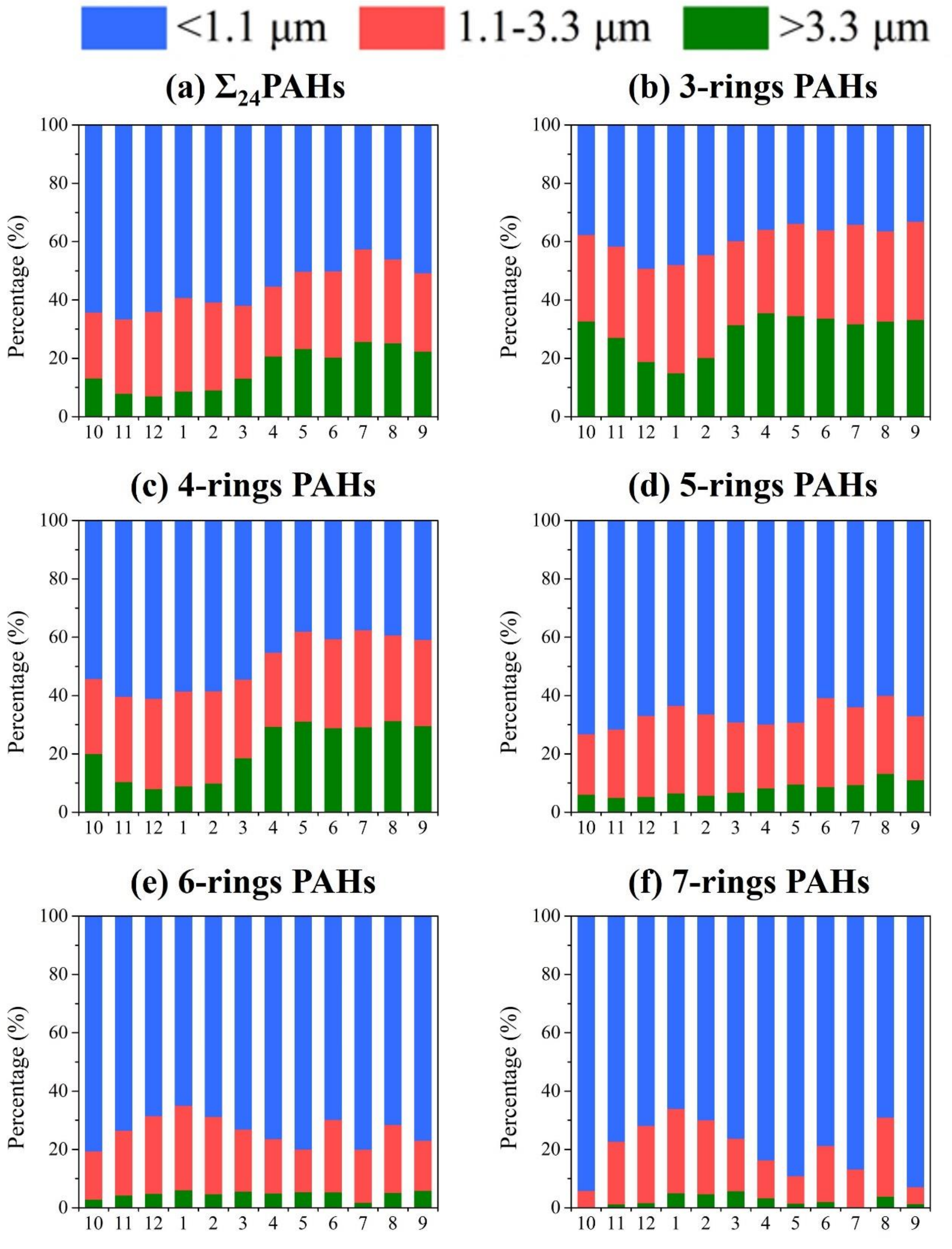

Figure S9 Monthly variations in size distribution of $\sum_{24}$PAHs (a), 3-rings PAHs (b), 4-rings PAHs (c), 5-rings PAHs (d), 6-rings PAHs (e) and 7-rings (f) PAHs over China.

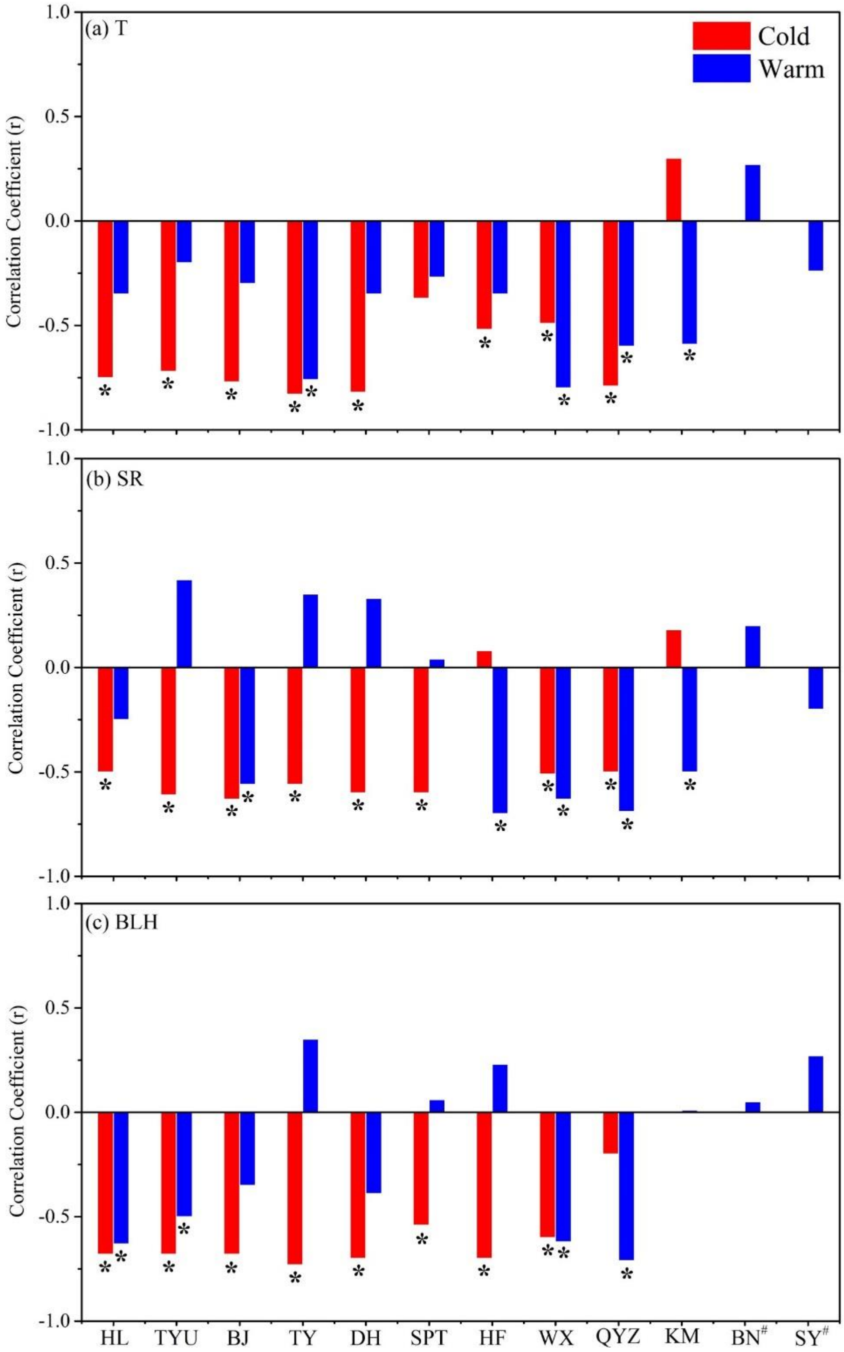

Figure S10 Correlation coefficient (r) of PAHs with T (a), SR (b) and BHL (c) at 12 sites in cold and warm season.

*: $p<0.05$

#: the ambient temperature at BN and SY are all exceed 13.9 °C, there are no cold season in these two sampling sites.

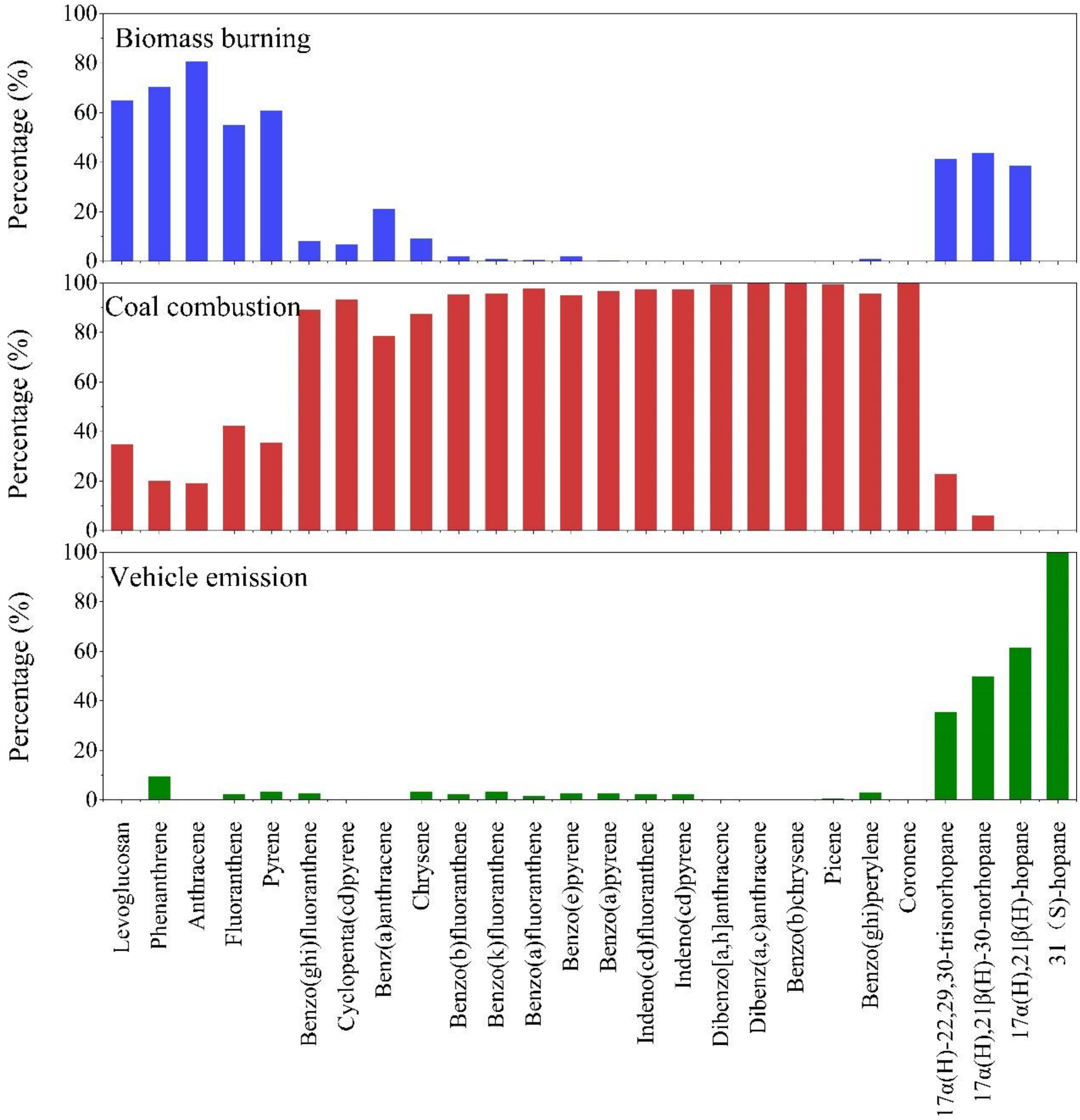

Figure S11 Source profiles (% of the species) resolved by PMF.

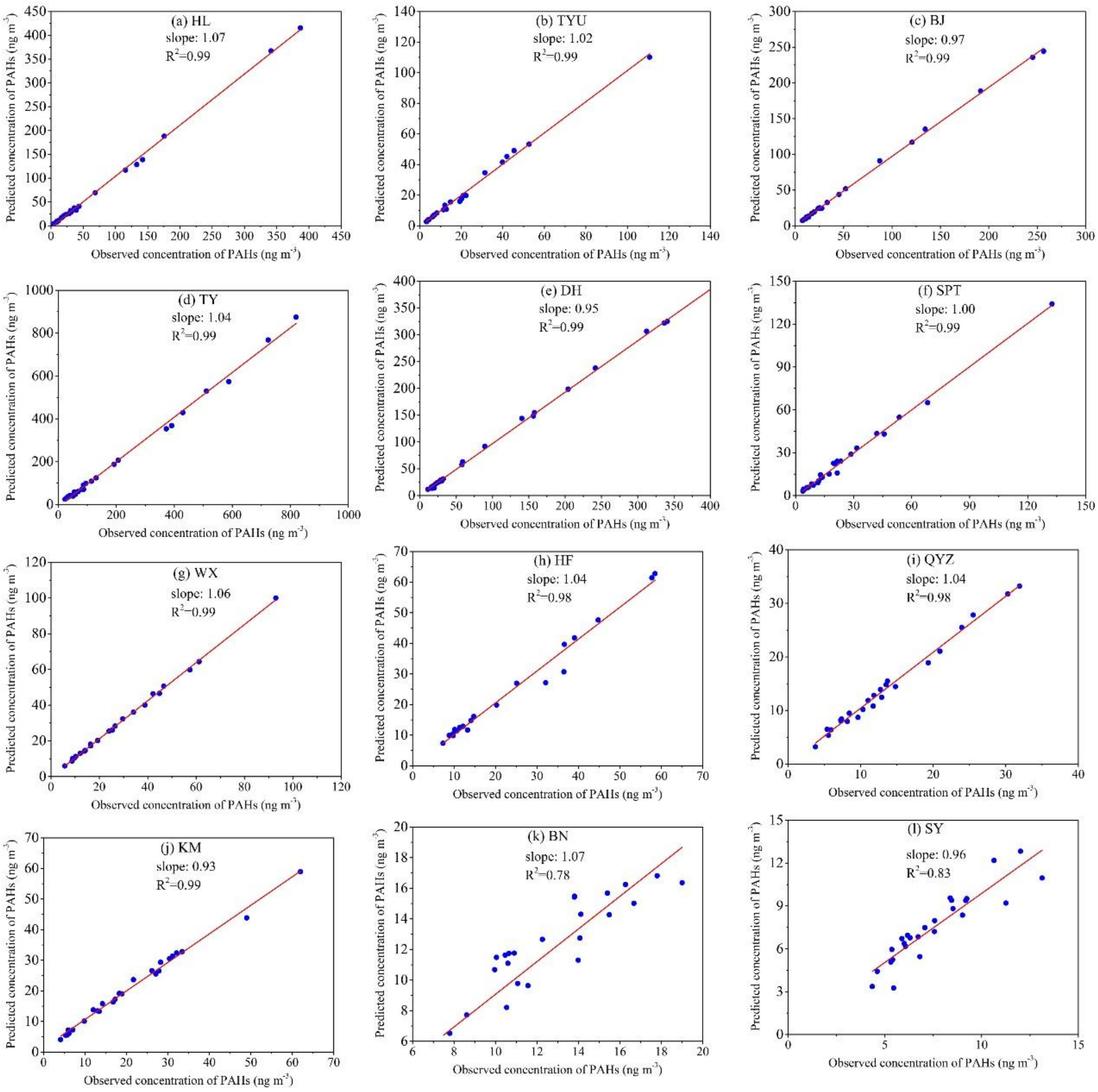

Figure S12 Correlations of the predicted PAHs by PMF with the observed PAHs.

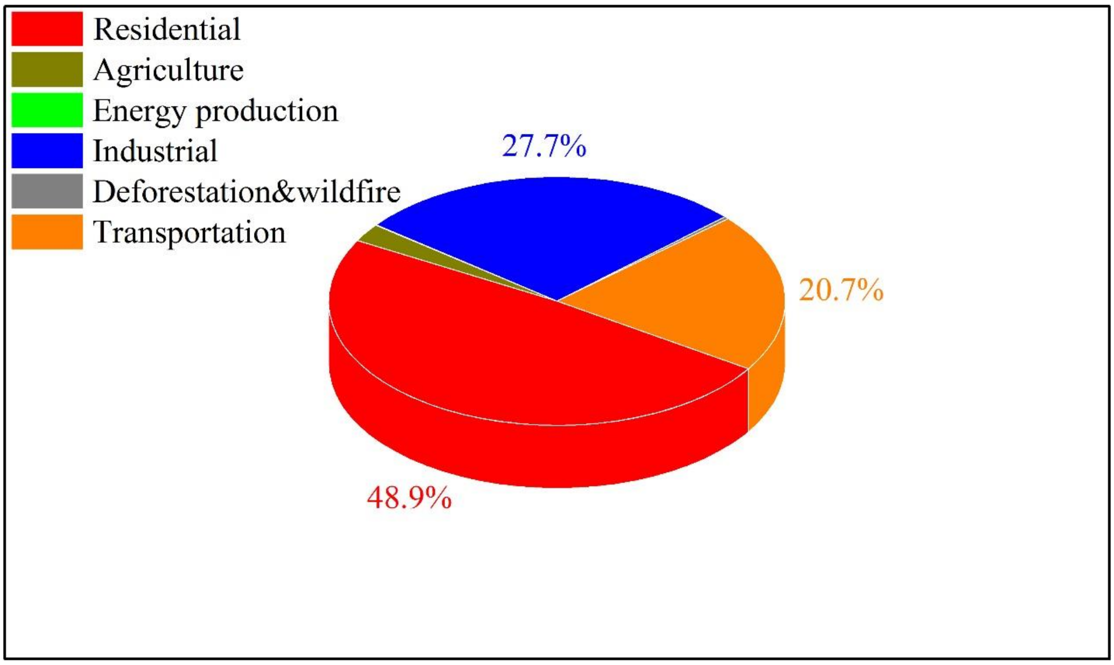

Figure S13 Atmospheric emissions of polycyclic aromatic hydrocarbons in China in 2013. (http://inventory.pku.edu.cn).

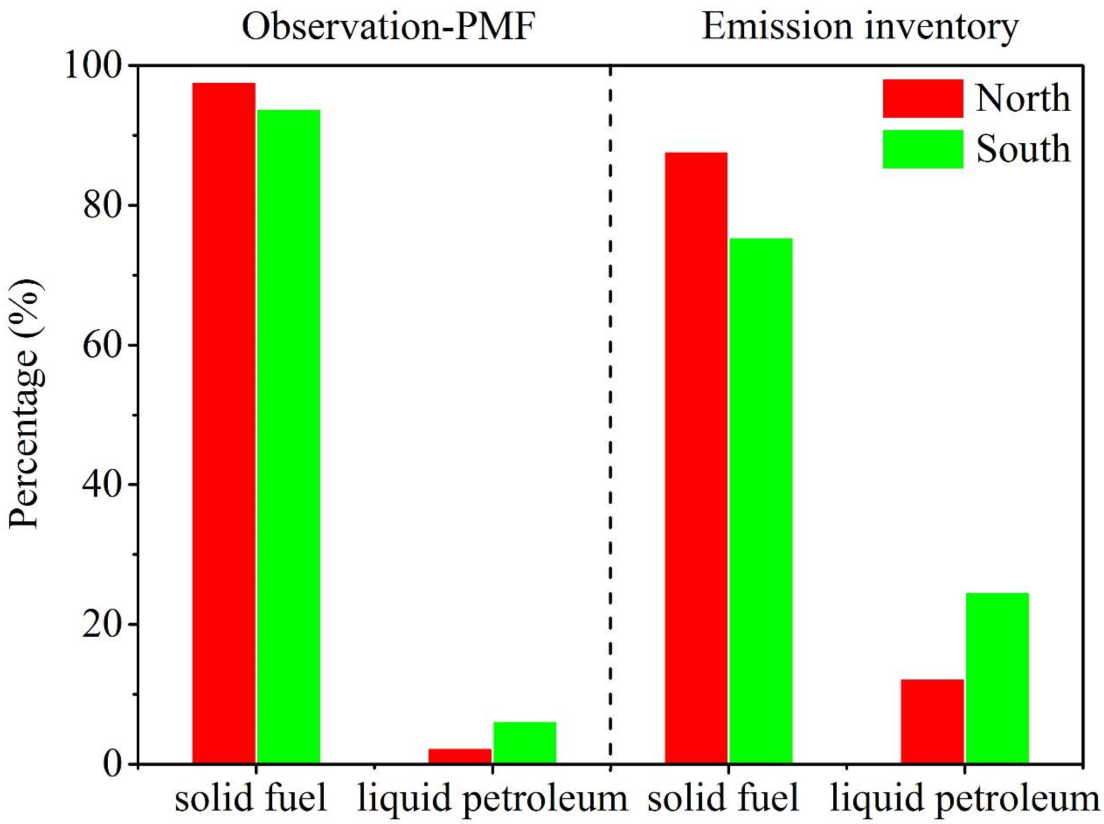

Figure S14 Comparison of source contribution from PMF and emission inventory.

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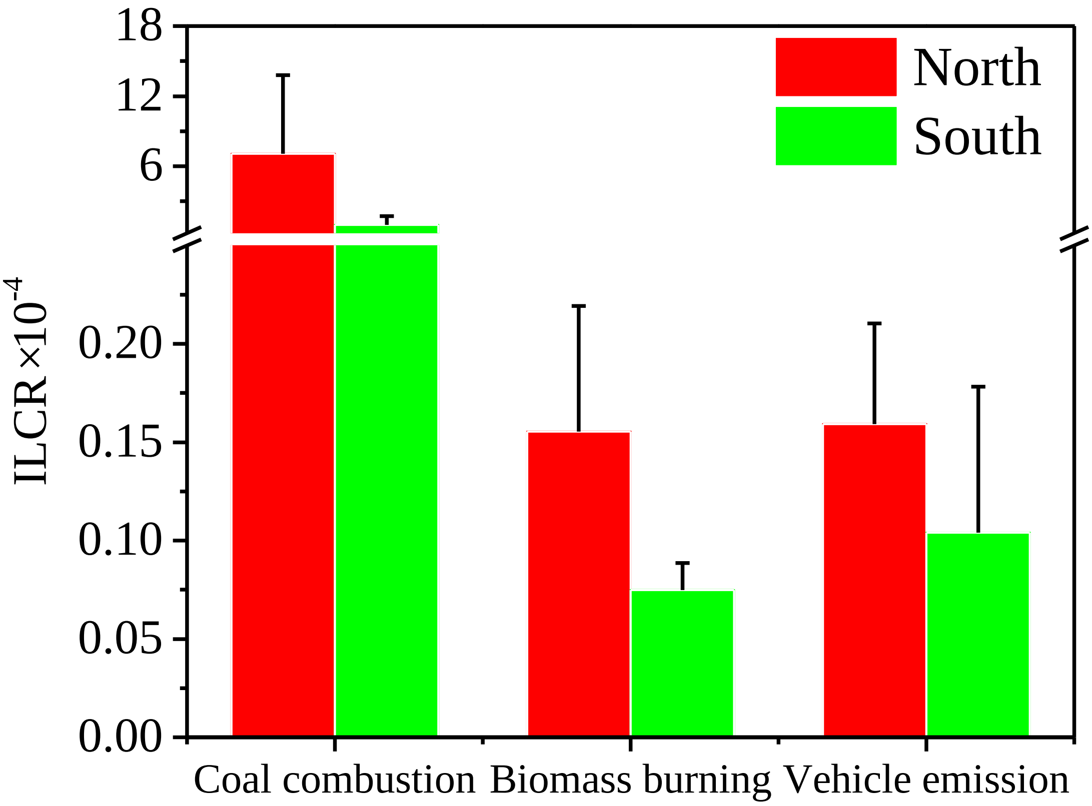

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123 Figure S15 Source contributions to ILCR in north and south China.

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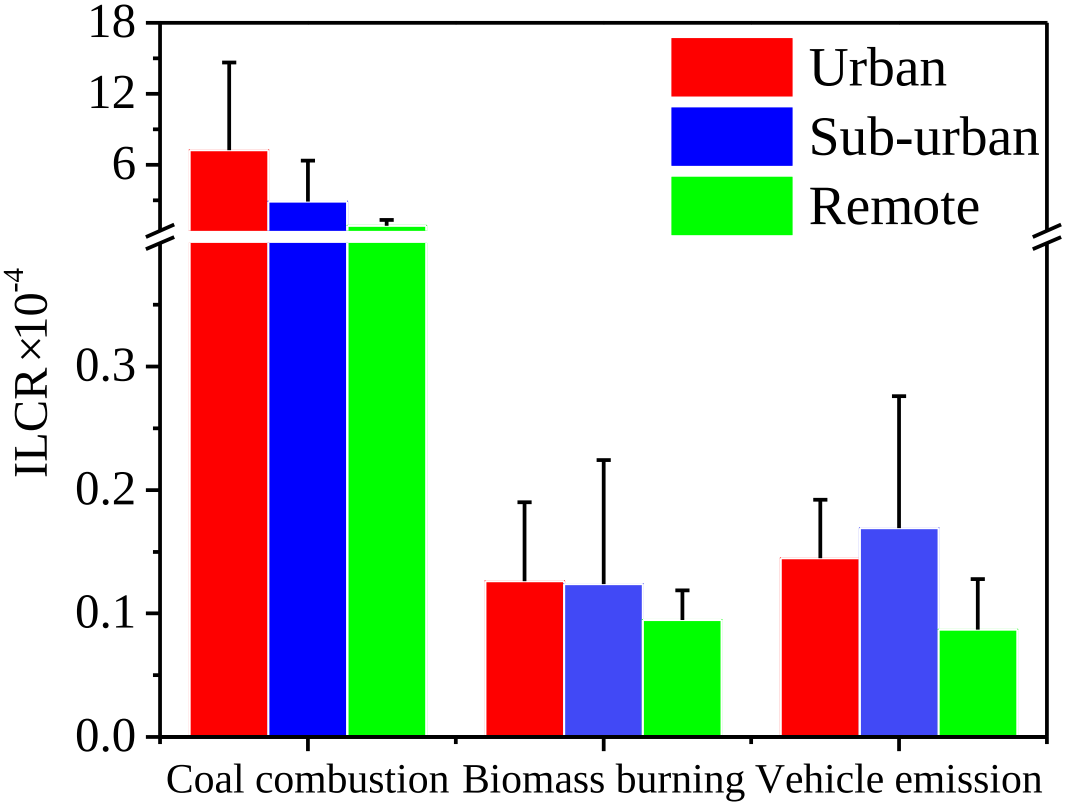


Figure S16 Source contributions to ILCR at urban, suburban and remote sites.

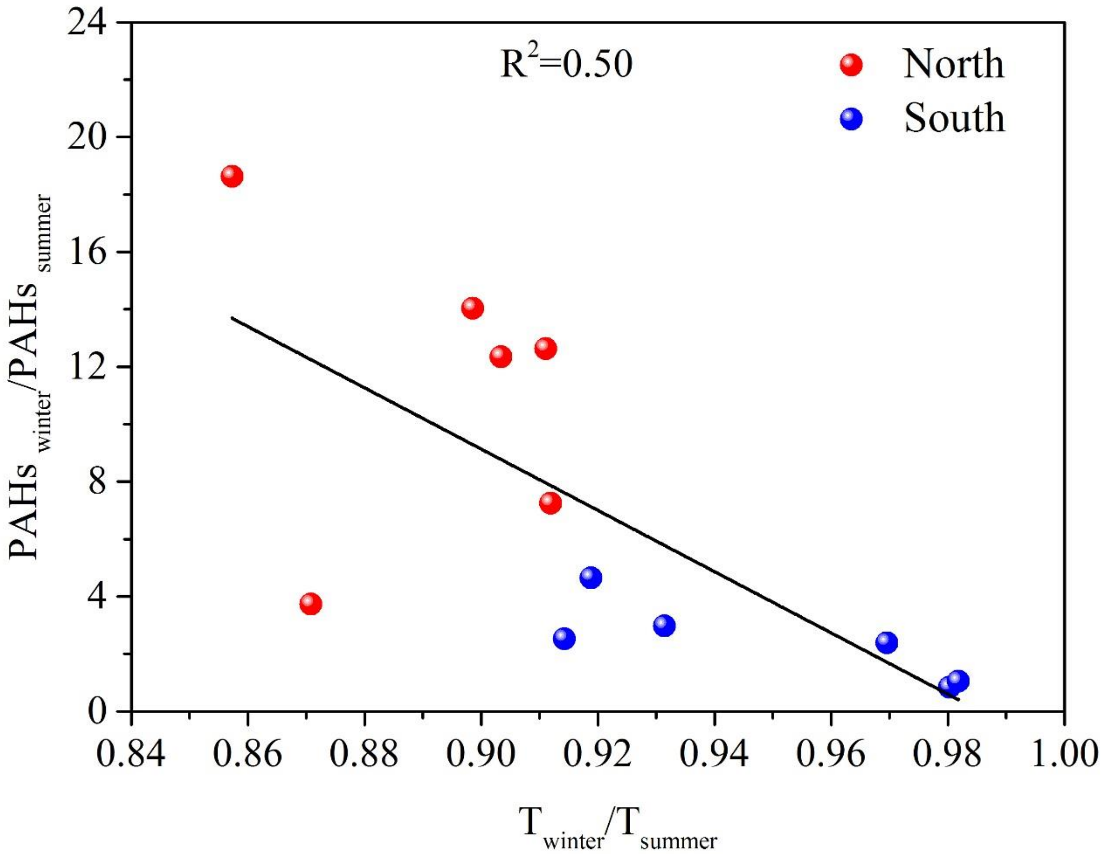


Figure S17 Ratios of $PAHs_{winter}/PAH_{summer}$ versus T_{winter}/T_{summer} in north and south China.

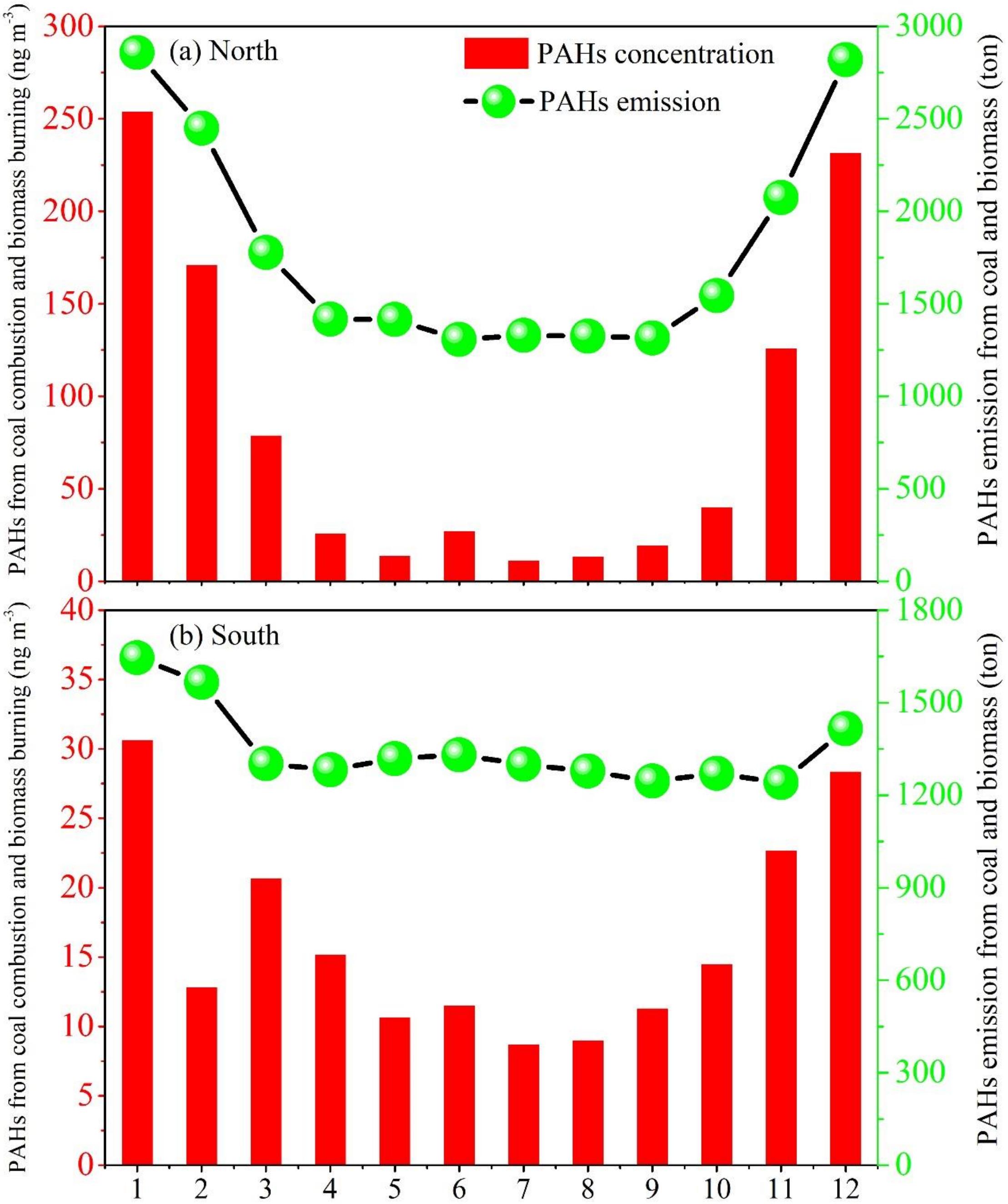
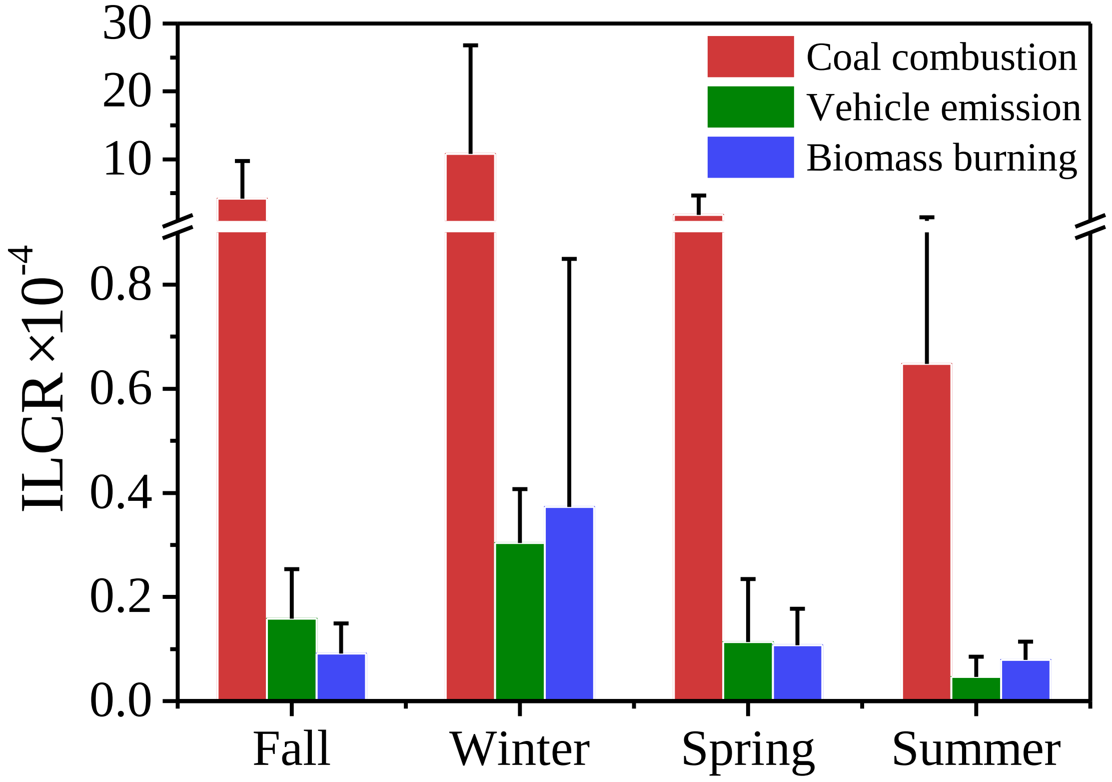


Figure S18 Monthly variations of source contribution from PMF (con) and emission inventory (ton) from solid fuel (coal and biomass) burning in the northern (a) and the southern (b) China.



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135 Figure S19 Seasonal variations of ILCR source contributions in China.