

Supporting information

2 Nationwide increase of polycyclic aromatic hydrocarbons in ultrafine particles during 3 winter over China

4 Qingqing Yu^a, Xiang Ding^{a,d,*}, Quanfu He^a, Weiqiang Yang^e, Ming Zhu^{a,b}, Sheng Li^{a,b}, Runqi
5 Zhang^{a,b}, Ruqin Shen^a, Yanli Zhang^{a,c,d}, Xinhui Bi^{a,d}, Yuesi Wang^{c,f}, Ping'an Peng^{a,d}, Xinming
6 Wang^{a,b,c,d,*}

⁷ ^aState Key Laboratory of Organic Geochemistry and Guangdong Key Laboratory of
⁸ Environmental Protection and Resources Utilization, Guangzhou Institute of Geochemistry,
⁹ Chinese Academy of Sciences, Guangzhou 510640, China

10 ^bUniversity of Chinese Academy of Sciences, Beijing 100049, China

11 °Center for Excellence in Regional Atmospheric Environment, Institute of Urban Environment,
12 Chinese Academy of Sciences, Xiamen 361021, China

13 ^dGuangdong-Hong Kong-Macao Joint Laboratory for Environmental Pollution and Control,
14 Guangzhou Institute of Geochemistry, Chinese Academy of Science, Guangzhou 510640,
15 China

¹⁶ ^eGuangdong Provincial Academy of Environmental Science, Guangzhou 510045, China

¹⁷ State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry,
¹⁸ Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

19 *corresponding author:

20 Dr. Xinming Wang and Dr. Xiang Ding

21 State Key Laboratory of Organic Geochemistry Guangzhou Institute of Geochemistry, Chinese
22 Academy of Sciences, 511 Kehua Rd, Tianhe, Guangzhou, 510640, China.

23 Email addresses: wangxm@gig.ac.cn and xiangd@gig.ac.cn

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50 Table S1 Detail information of the sampling sites in China.

Sampling sites	Type	Region	Latitude (°N)	Longitude (°E)	Sampling duration
Hailun (HL)	Sub-urban	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)	Sub-urban	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)	Sub-urban	South China	18.23	109.48	biweekly 48-hr

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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

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

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

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	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
Σ PAHs	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

	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
Σ 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

64 Table S4 The annual level of atmospheric temperature, solar radiation and boundary layer
65 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

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68 Table S5 The monthly average temperature (°C) in each sampling site in the northern and the
 69 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

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71 Table S6 The monthly average solar radiation (w/m^2) in each sampling site in the northern and
 72 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	

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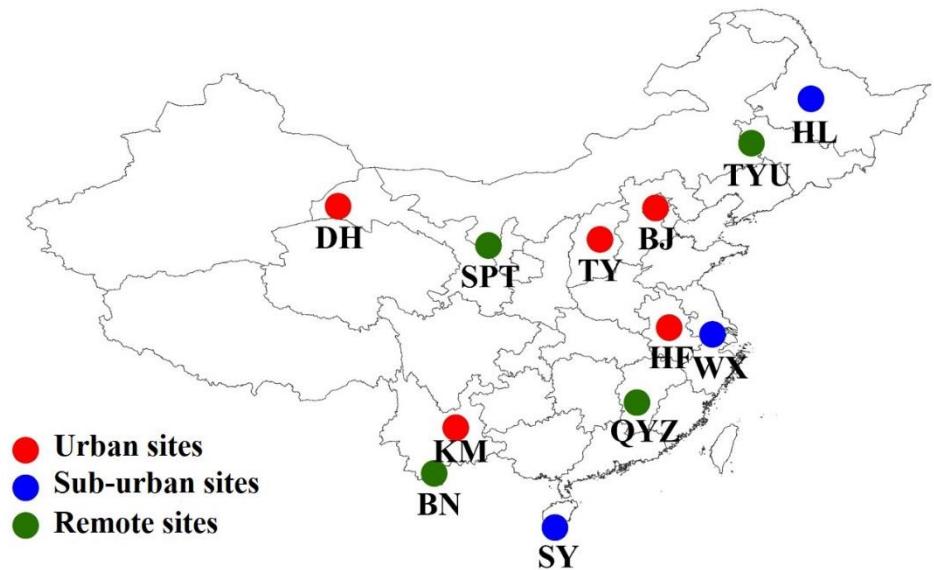
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75 Table S7 The monthly average boundary layer height (m) in each sampling site in the northern
 76 and the southern China.

	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	

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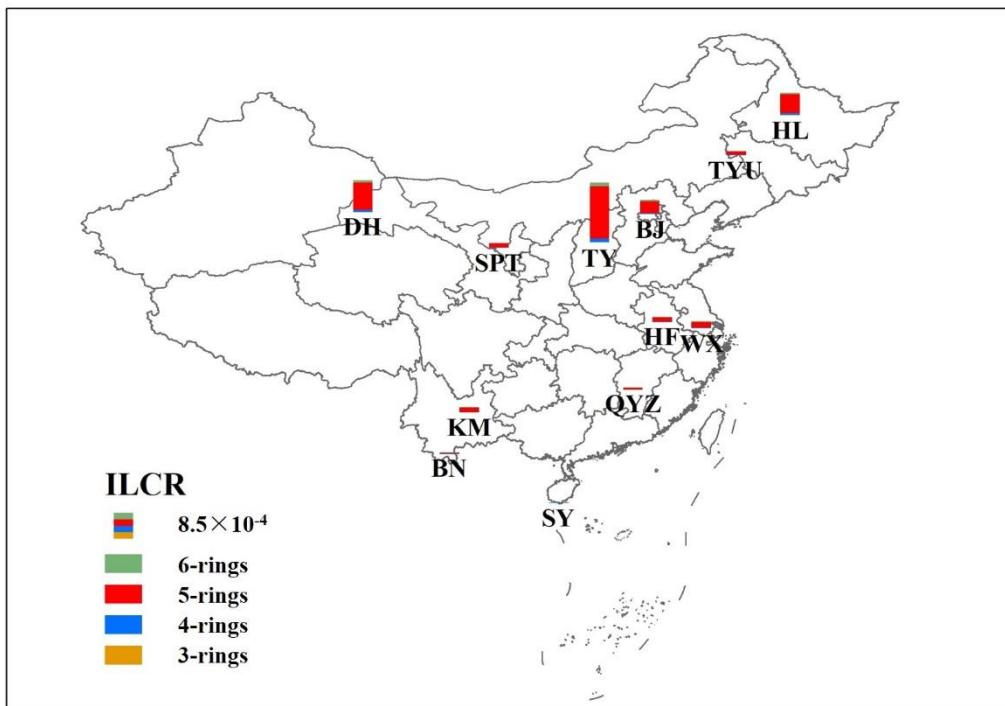
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80 Figure S1 Sampling sites in China, including five urban sites: Beijing (BJ), Taiyuan (TY), Hefei
81 (HF), Kunming (KM), and Dunhuang (DH), three sub-urban sites: Hailun (HL), Wuxi (WX),
82 and Sanya (SY), four remote sites: Tongyu (TYU), Shapotou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

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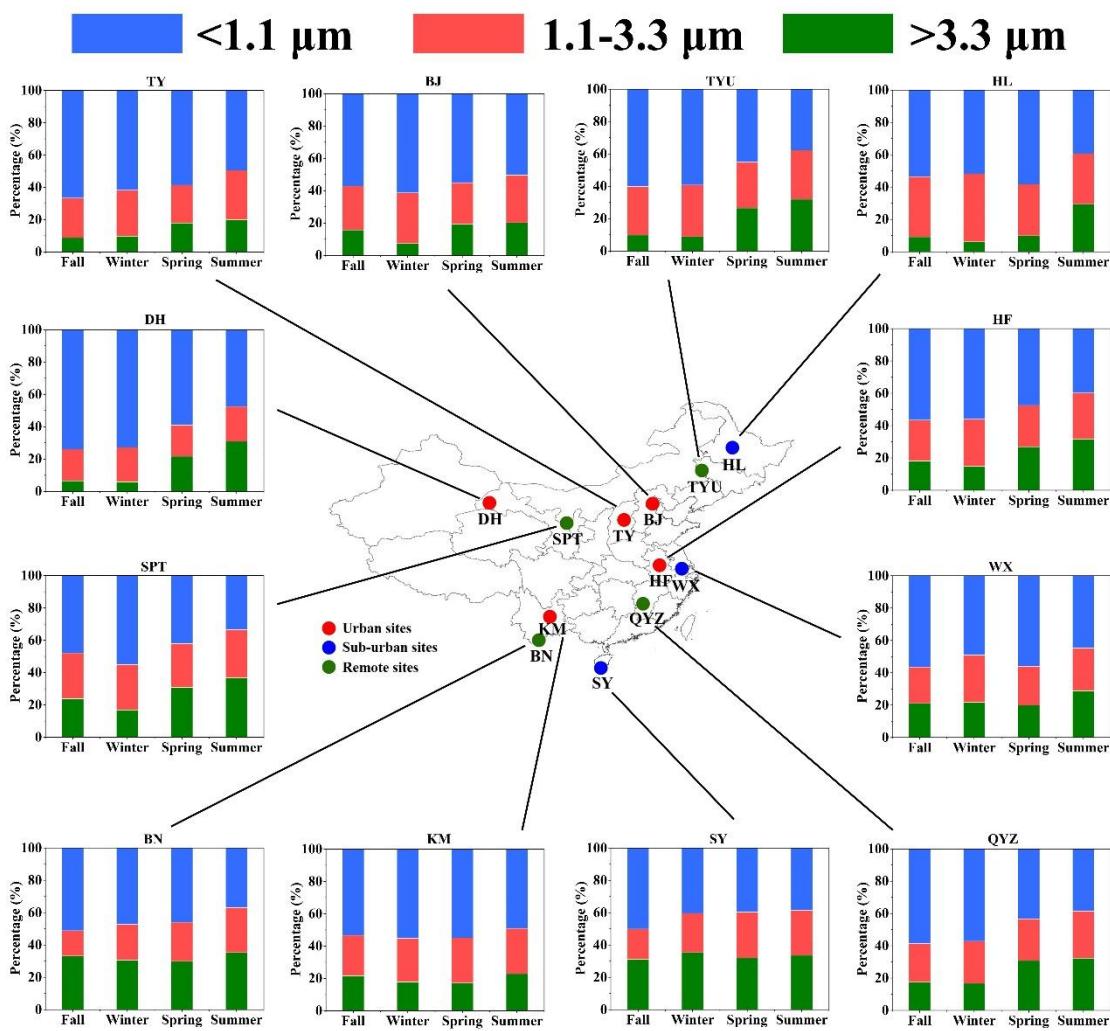
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Figure S2 Annual averages of ILCR at 12 sites over China.

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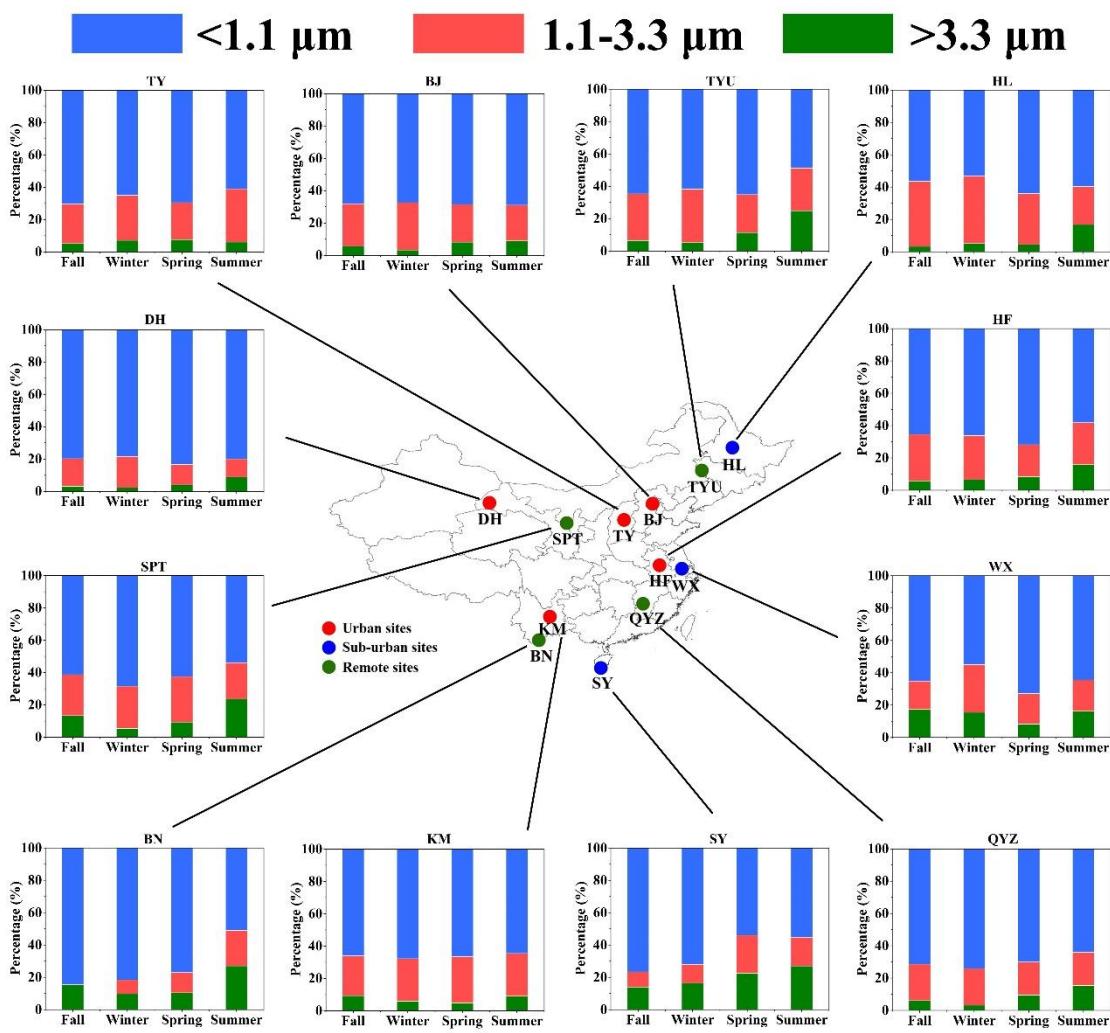


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Figure S3 Size distribution of PAHs in different season over China.

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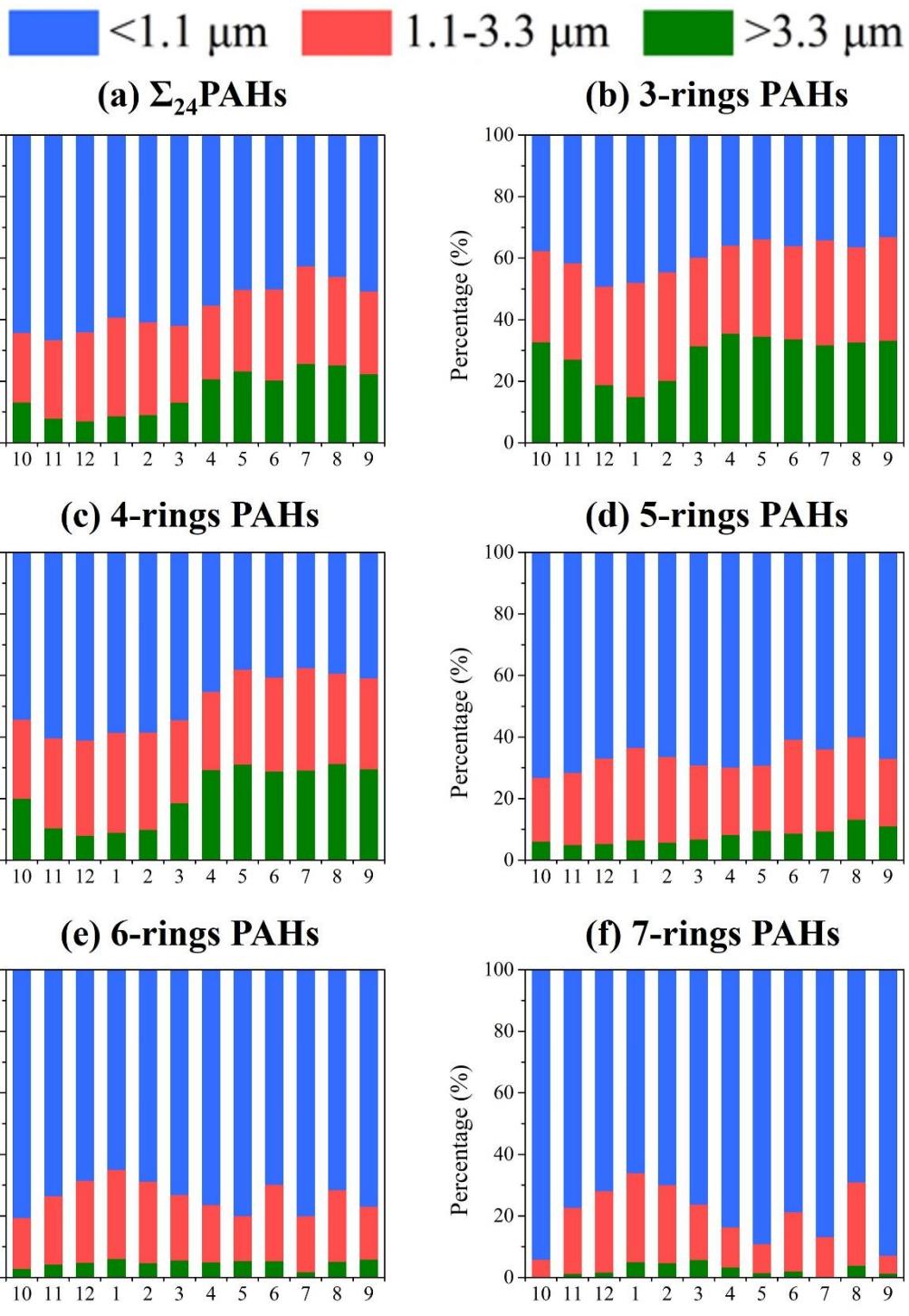


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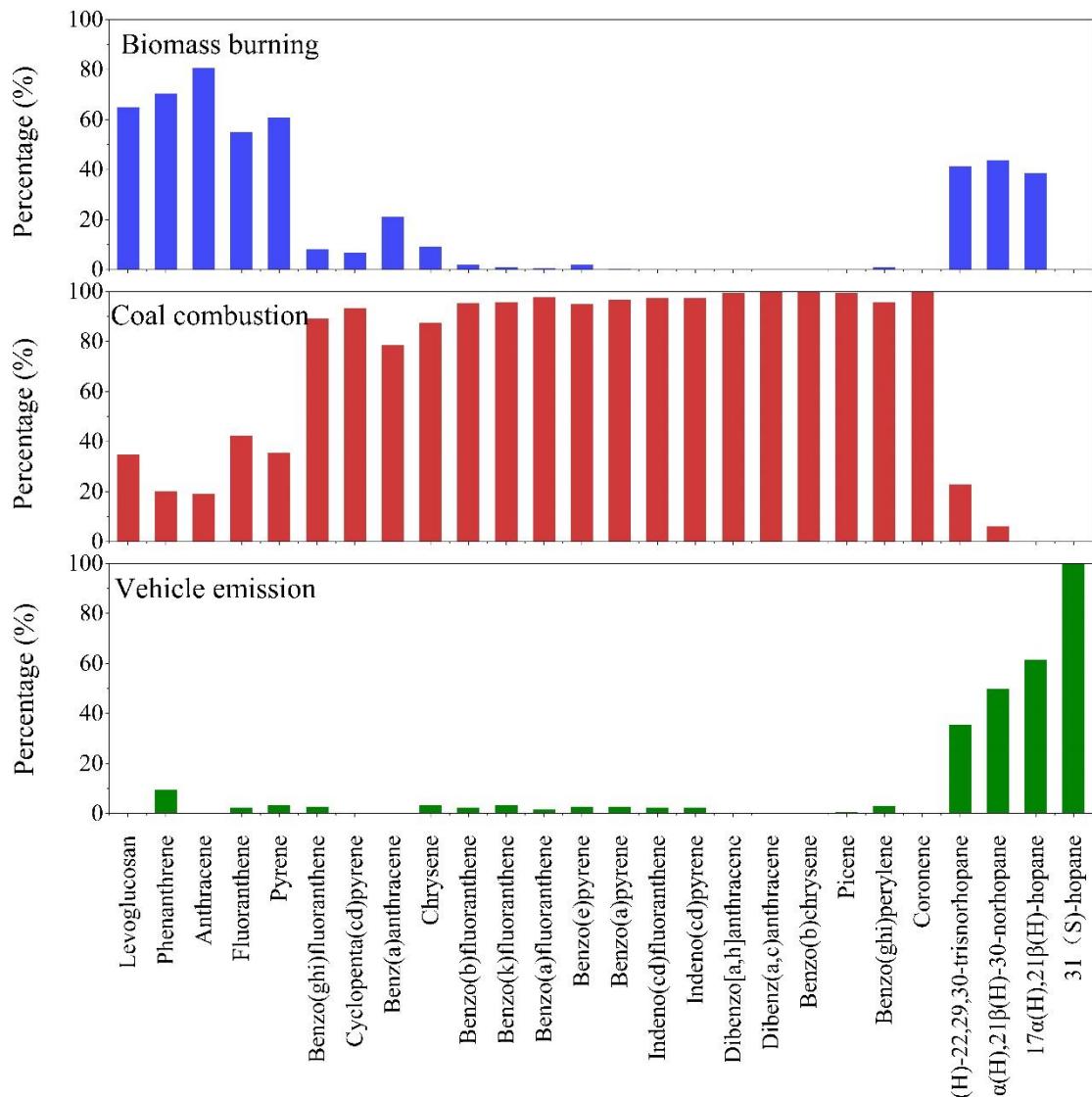
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Figure S4 Size distribution of BaP_{eq} in different seasons over China.



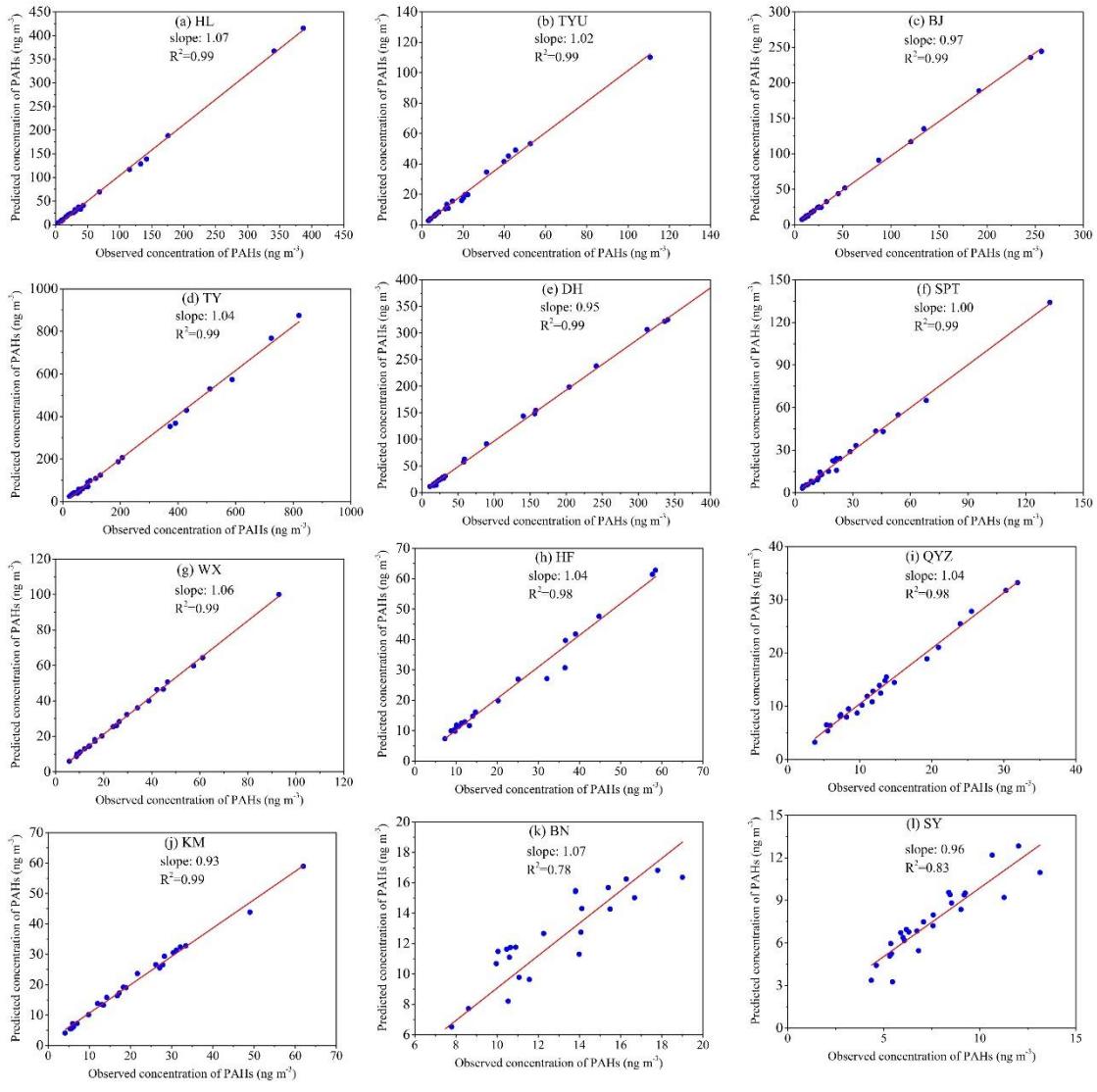
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96 Figure S5 Monthly variations in size distribution of Σ_{24} PAHs (a), 3-rings PAHs (b), 4-rings
97 PAHs (c), 5-rings PAHs (d), 6-rings PAHs (e) and 7-rings (f) PAHs over China.
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100 Figure S6 Source profiles (% of the species) resolved by PMF.

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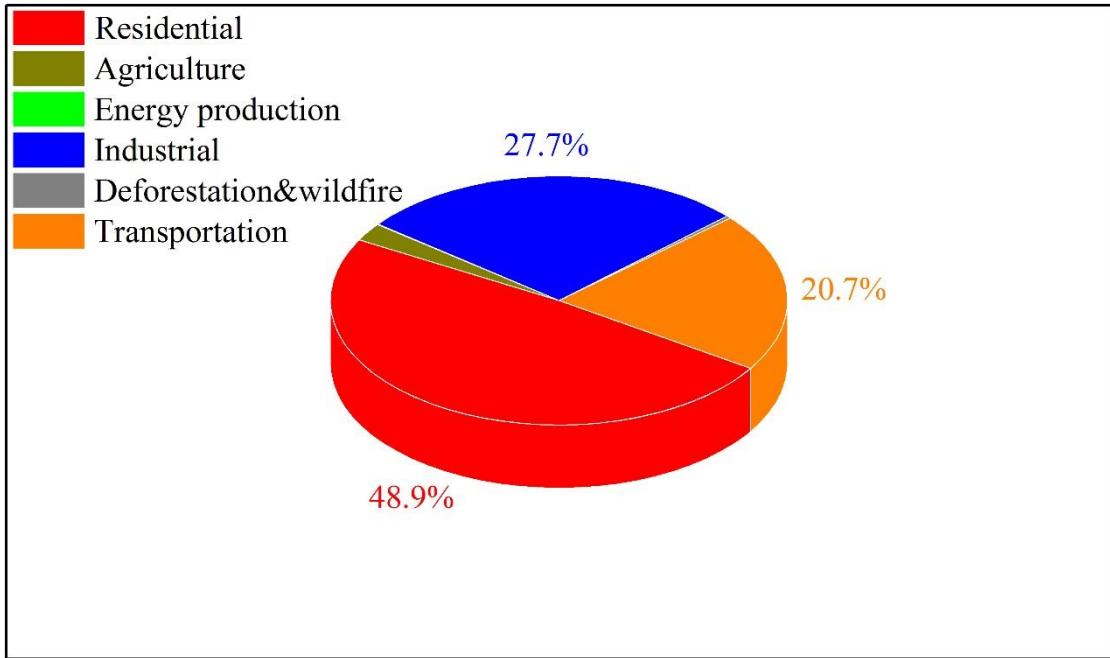


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Figure S7 Correlations of the predicted PAHs by PMF with the observed PAHs.

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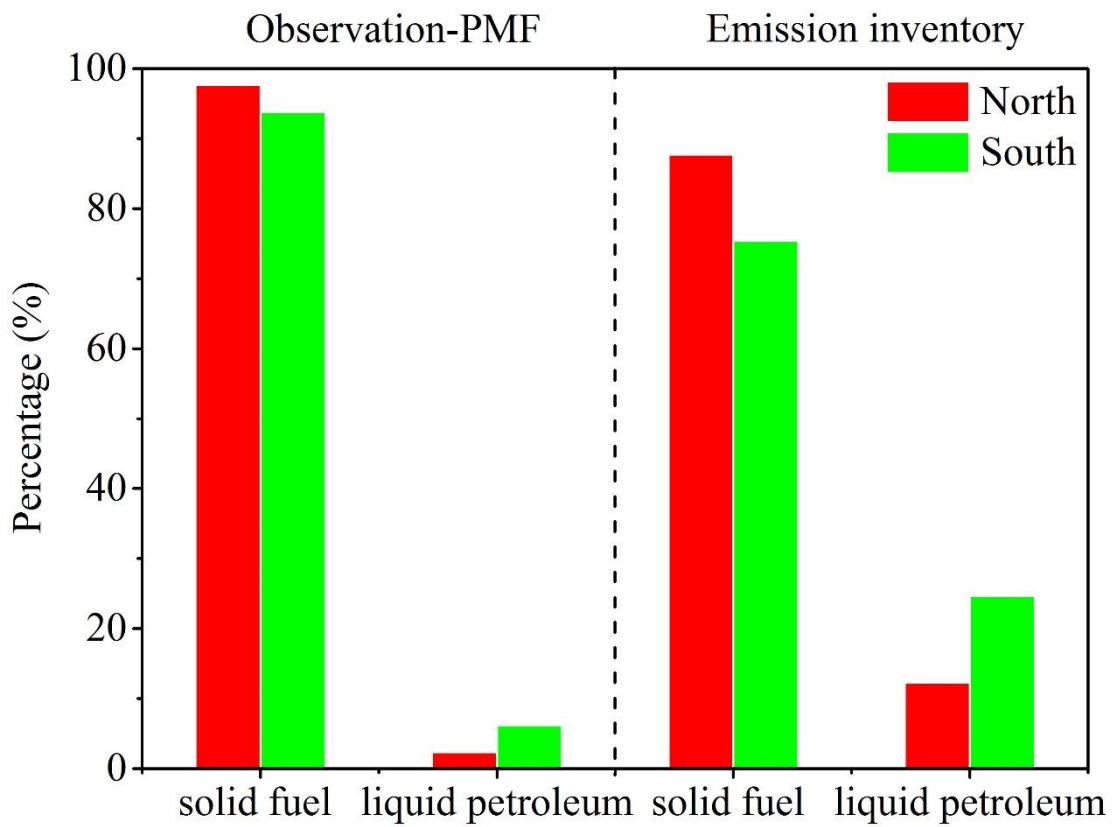


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106 Figure S8 Atmospheric emissions of polycyclic aromatic hydrocarbons in China in 2013.

107 (<http://inventory.pku.edu.cn>).

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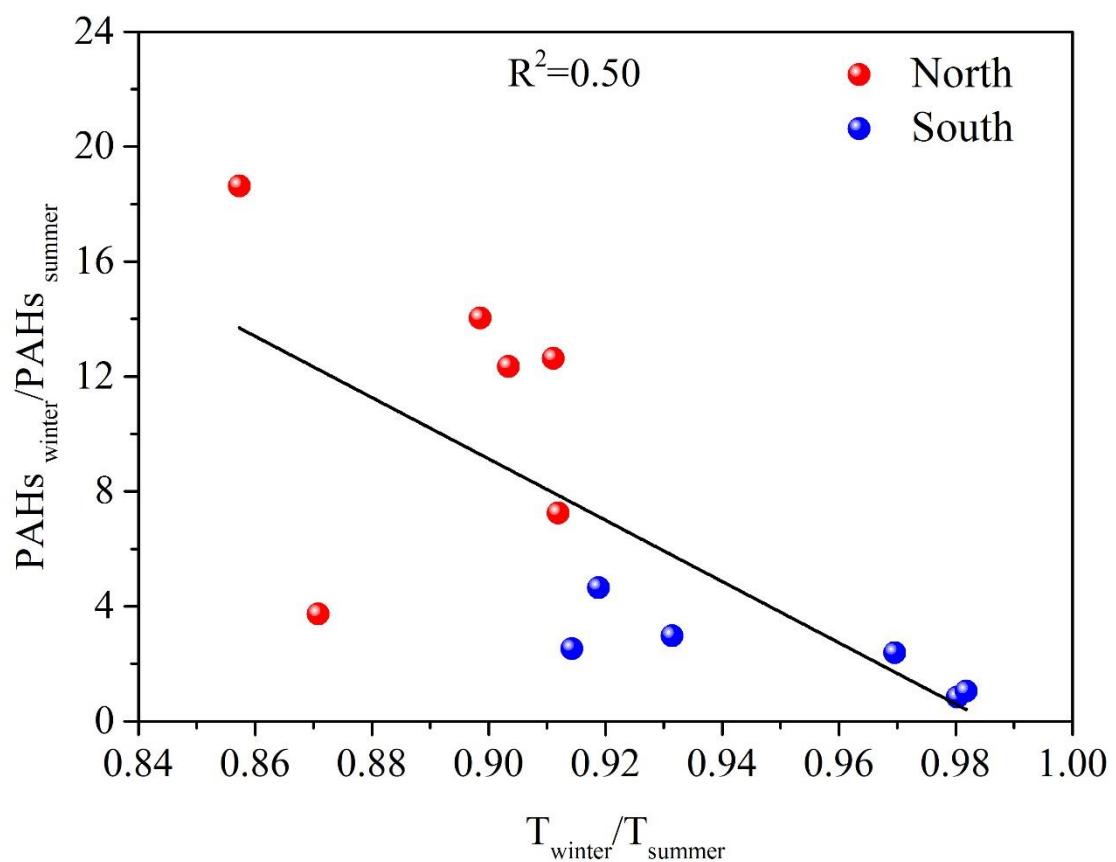


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110 Figure S9 Comparison of source contribution from PMF and emission inventory.

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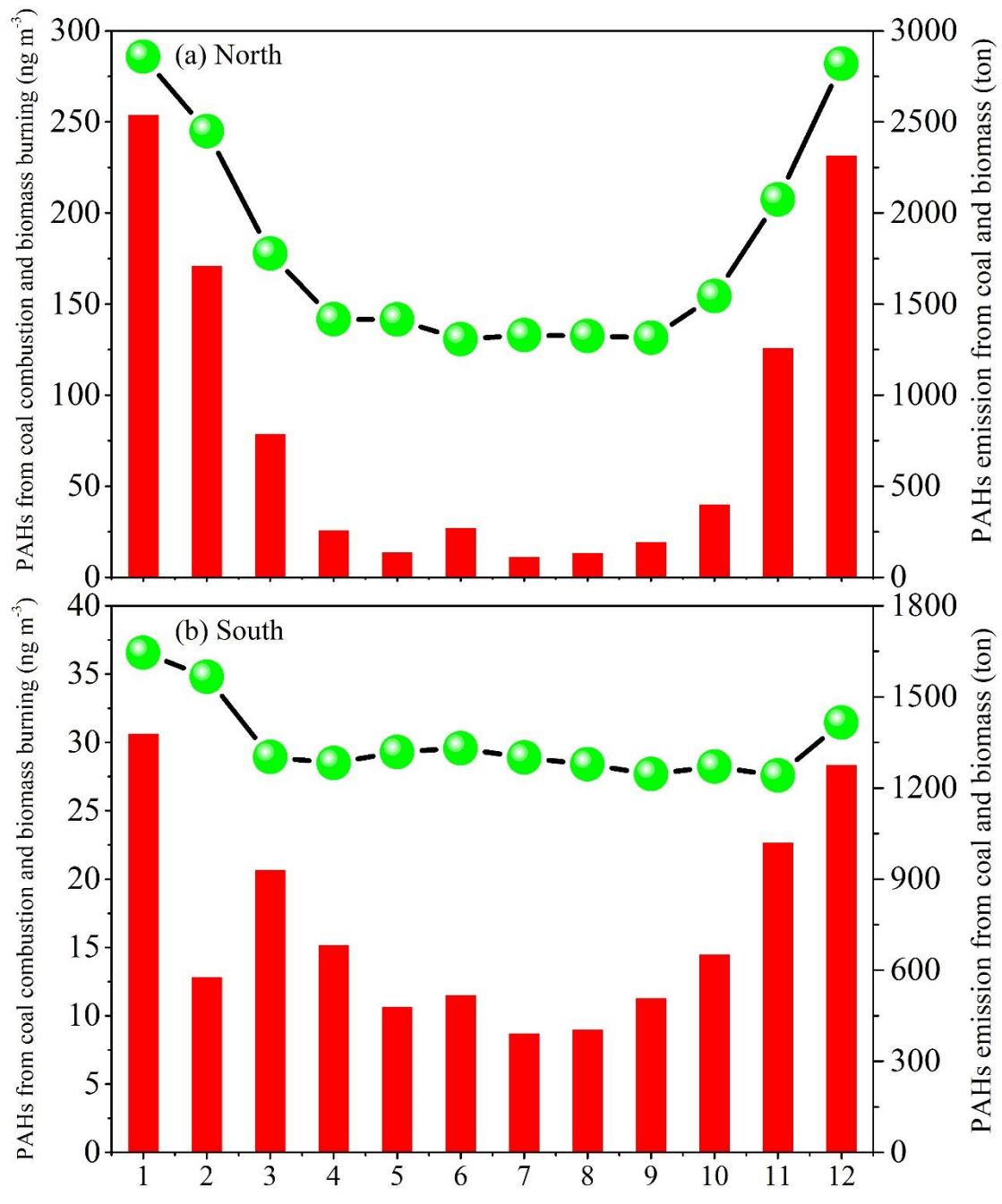


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Figure S10 The winter to summer ratios of PAHs to T.



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117 Figure S11 Monthly variations of source contribution from PMF (con) and emission inventory

118 (ton) from solid fuel (coal and biomass) burning in the northern (a) and the southern (b)

119 China.