Supplementary Information

Size Distribution of Airborne Particle-Bound PAHs and o-PAHs and Their Implications for Dry Deposition

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Lists of PAHs and o-PAHs cited in the main text

List S1: (Beijing-Tianjin region)

acenaphthene (ACE), acenaphthylene (ACY), fluorene (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz[a] anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DahA), indeno [1,2,3-cd]pyrene (IcdP), and benzo[ghi]perylene (BghiP).

List S2: (Guangzhou)

benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthracene (DahA) and benzo[ghi]perylene (BghiP)

List S3: (Turkey)

acenaphthylene (ACT), fluorene (FLN), phenanthrene (PHE), anthracene (ANT), fluoranthene (FL), pyrene (PY), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthracene (DahA), benzo[ghi]perylene (BghiP)

Table S1 Physiochemical properties of the target 5 PAHs and 6 o-PAHs

Compounds	Abbreviation	CAS No.	MW	P^{o}_{L}(Pa)[1]
Naphthalene	NAP	91-20-3	128.18	39.8
Fluorene	FLU	86-73-7	166.22	0.618
Phenanthrene	PHE	85-01-8	178.24	8.74×10^{-2}
Anthracene	ANT	120-12-7	178.24	6.59×10^{-2}
Benz[a]anthracene	BaA	56-55-3	228.30	1.07×10^{-4}
1,4-Naphthoquinone	1,4NQ	130-15-4	158.16	0.244
9-Fluorenone	9FLO	486-25-9	180.21	2.80×10^{-2}
9,10-Anthracenequinone	9,10AQ	84-65-1	208.22	5.90×10^{-3}
9,10-Phenanthraquinone	9,10PQ	84-11-7	208.22	1.02×10^{-2}
Benzanthrone	Bzone	82-05-3	230.27	9.28×10^{-4}
Benz(a)anthracene-7,12-quinone	BAQ	2498-66-0	258.28	1.59×10^{-4}

[1] The subcooled vapor pressure, under 298K, was estimated by the modified Grain method embedded in Estimation Programs Interface (EPI) Suite software (version 4.1) developed by the US Environmental Protection Agency.

Table S2 GC-MS/MS method parameters applied in analyzing parent PAHs and o-PAHs.

Compounds	RT(min)	Precursor	Product	Collision
NAP	6.00	128	128.1	5
		128	102.1	25
FLU	10.20	166	166.1	5
		166	165.1	20
PHE	11.85	178	178.1	5
		178	152.1	25
ANT	11.85	178	178.1	5
		178	152.1	25
BaA	16.00	228	228.1	5
		228	226.0	35
1,4NQ	8.00	158	130.1	5
		158	102.0	20
9FLO	11.00	180	180.1	5
		152	151.1	15
9,10AQ	13.00	208	180.1	10
		152	152.0	5
9,10PQ	14.15	180	152.1	20
		152	151.1	15
Bzone	16.00	230	230.1	5
		202	202.1	5
BAQ	16.90	230	202.1	20
		202	202.1	5

Table S3 The recovery and relative standard deviation of the target compounds.

Compounds	Recovery	Relative standard deviation
NAP	93	9
FLU	80	15
PHE	89	7
ANT	92	17
BaA	93	10
1,4NQ	115	16
9FLO	96	5
9,10AQ	105	13
9,10PQ	95	8
Bzone	97	12
BAQ	95	8

Table S4 Correlation matrix between o-PAH and corresponding parent PAH

	NAP	1,4NQ	FLU	9FLO	ANT	9,10AQ	PHE	9,10PQ	BaA	Bzone	BAQ	p-PAHs	o-PAHs
NAP	1.00	-0.32	**-0.47***	-0.14	-0.33	-0.40	-0.33	-0.39	-0.42	**-0.49***	-0.25	-0.29	-0.28
1,4NQ		1.00	**0.68****	**0.70****	**0.72****	**0.88****	**0.75****	**0.90****	**0.79****	**0.77****	**0.74****	**0.77****	**0.89****
FLU			1.00	**0.53***	**0.91****	**0.57****	**0.86***	**0.67****	**0.87****	**0.64****	**0.69****	**0.89****	**0.66****
9FLO				1.00	**0.55****	**0.64****	**0.63****	**0.74****	**0.66****	**0.78****	**0.85****	**0.66****	**0.88****
ANT					1.00	**0.57****	**0.95****	**0.74****	**0.91****	**0.65****	**0.63****	**0.94****	**0.67****
9,10AQ						1.00	**0.62****	**0.73****	**0.70****	**0.70****	**0.70****	**0.63****	**0.86****
PHE							1.00	**0.81****	**0.93****	**0.69****	**0.71****	**0.96****	**0.71****
9,10PQ								1.00	**0.80****	**0.80****	**0.75***	**0.77****	**0.88****
BaA									1.00	**0.81****	**0.74****	**0.94****	**0.77****
Bzone										1.00	**0.86****	**0.71****	**0.86****
BAQ											1.00	**0.73****	**0.87****
p-PAHs												1.00	**0.75****
o-PAHs													1.00

Significant values are marked in bold. *Correlation is significant at 0.05 level (two-tailed). **Correlation is significant at 0.01 level (two-tailed).

Table S5 The statistical significance using non-parametric Mann-Whitney *U* test for testing the seasonal variation of PAH and o-PAH species

	Abbreviation	*p*
Naphthalene	NAP	0.32
Fluorene	FLU	0.004
Anthracene	ANT	0.001
Phenanthrene	PHE	0.000
Benz(a)anthracene	BaA	0.004
∑5PAHs	PAHs	0.001
1,4-Naphthoquinone	1,4NQ	0.155
9-Fluorenon	9FLO	0.055
9,10-Anthracenequinone	9,10AQ	0.32
9,10-Phenanthraquinone	9,10PQ	0.023
Benzoanthrone	Bzone	0.286
Benz(a)anthracene-7,12-quinone	BAQ	0.055
∑6o-PAHs	o-PAHs	0.102

[1] *p* values are for warm season (summer and spring) and cold season (winter and autumn).

Table S6 Size distributions of particle-bound PAHs and o-PAHs (ng m^{-3})

Size	<0.4	0.4~0.7	0.7~1.1	1.1~2.1	2.1~3.3	3.3~4.7	4.7~5.8	5.8~9.0	9.0~10.0
NAP	0.15±0.02	0.15±0.02	0.16±0.03	0.15±0.05	0.15±0.03	0.15±0.02	0.15±0.03	0.17±0.03	0.17±0.03
1,4NQ	0.21±0.06	0.23±0.09	0.23±0.08	0.22±0.06	0.21±0.07	0.21±0.06	0.21±0.07	0.22±0.07	0.21±0.07
FLU	0.13±0.03	0.13±0.03	0.14±0.03	0.15±0.04	0.12±0.02	0.14±0.03	0.14±0.03	0.17±0.05	0.15±0.04
9FLO	0.25±0.08	0.29±0.12	0.25±0.10	0.26±0.10	0.26±0.09	0.25±0.09	0.26±0.09	0.31±0.11	0.27±0.08
ANT	0.13±0.02	0.13±0.02	0.13±0.03	0.13±0.02	0.13±0.02	0.13±0.02	0.12±0.02	0.13±0.02	0.13±0.02
9,10AQ	0.67±0.31	0.61±0.24	0.57±0.30	0.57±0.29	0.50±0.22	0.53±0.29	0.53±0.26	0.51±0.24	0.60±0.31
PHE	0.31±0.09	0.31±0.10	0.33±0.12	0.34±0.16	0.30±0.10	0.29±0.09	0.28±0.08	0.30±0.09	0.31±0.09
9,10PQ	0.38±0.15	0.38±0.19	0.47±0.34	0.49±0.40	0.32±0.10	0.29±0.07	0.28±0.06	0.30±0.07	0.32±0.07
BaA	0.48±0.09	0.54±0.14	0.56±0.17	0.52±0.13	0.22±0.06	0.18±0.04	0.13±0.03	0.17±0.04	0.22±0.06
Bzone	0.41±0.17	0.45±0.24	0.45±0.26	0.35±0.14	0.26±0.03	0.25±0.02	0.24±0.02	0.24±0.02	0.25±0.02
BAQ	0.45±0.07	0.45±0.11	0.47±0.16	0.43±0.12	0.35±0.03	0.34±0.02	0.34±0.03	0.35±0.03	0.36±0.03

Table S7 Calculated dry deposition fluxes (F_D, ng m^{-2} d^{-1}) and dry deposition velocities (V_d, cm s^{-1}) of size-fractionated PAHs and o-PAHs in Shanghai.

Compound	F_D	V_d
NAP	241±54	0.20±0.03
1,4NQ	327±127	0.19±0.03
FLU	204±63	0.21±0.03
9FLO	408±112	0.20±0.03
ANT	194±45	0.20±0.03
9,10AQ	834±254	0.20±0.03
PHE	457±146	0.19±0.03
9,10PQ	498±135	0.19±0.03
BaA	434±103	0.17±0.03
Bzone	445±72	0.18±0.03
BAQ	574±74	0.19±0.03

Table S8 Correlation matrix between dry deposition fluxes of PAHs, o-PAHs, TPAHs and meteorological parameters (temperature, relative humidity and wind speed)

	PAHs	o-PAHs	TPAHs[1]
Temperature	**-0.65****	**-0.65****	**-0.71****
Relative humidity	0.38	-0.08	0.18
Wind speed	0.30	-0.17	0.24

[1] TPAHs is the sum of PAHs and o-PAHs.

Significant values are marked in bold. **Correlation is significant at 0.01 level (two-tailed).

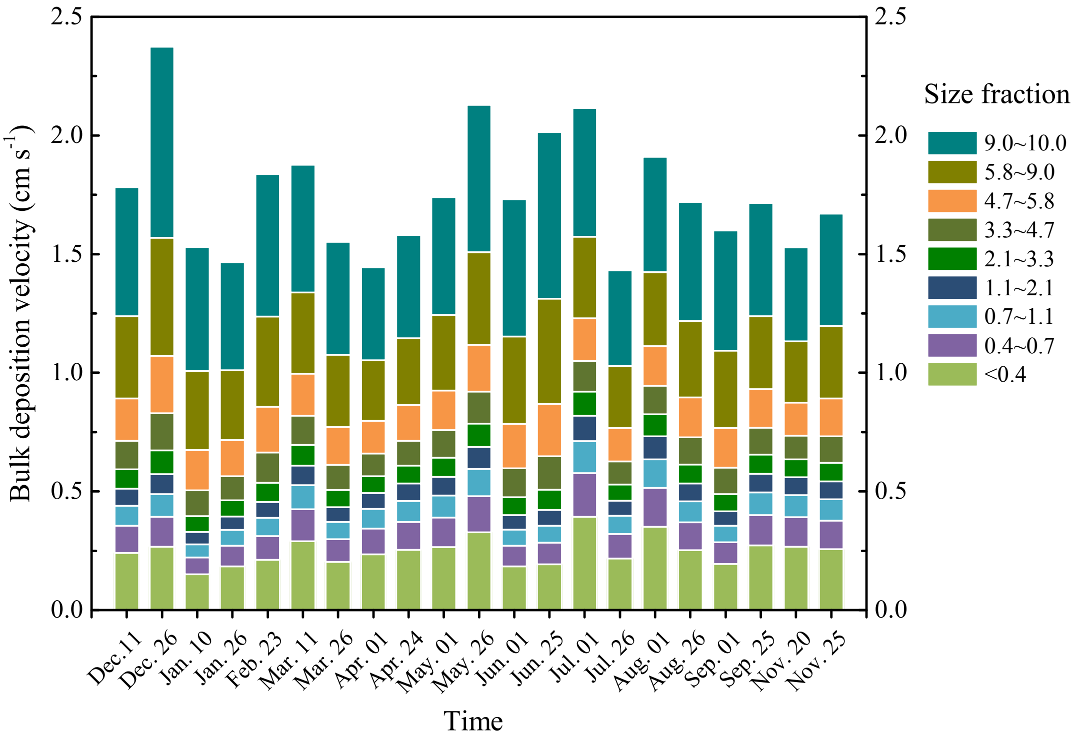

Fig. S1 Estimated bulk dry deposition velocity (cm s^{-1}) for size-resolved particles during one-year sampling period in Shanghai, China.

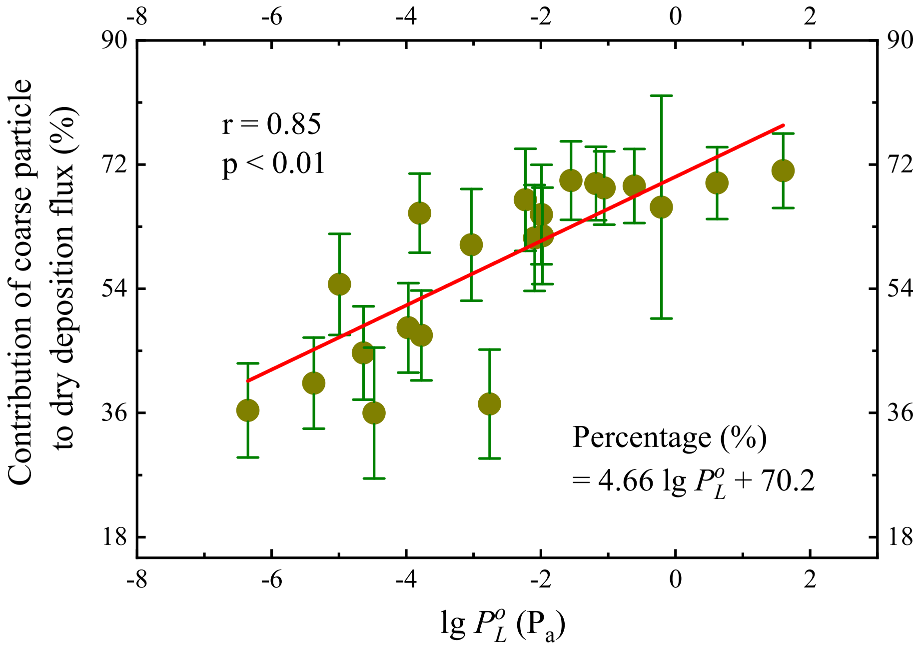

Fig. S2 Correlation between the contributions of coarse particle (> 2.1 μm) bound individual PAHs and o-PAHs and their respective lg P_L^o(Pa).