Supplement

Size-resolved exposure risk of persistent free radicals (PFRs) in atmospheric aerosols and their potential sources

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Text S1

Factor analysis of EPFRs

As shown in Figure S5, factor analysis of summer particulates showed that Factor 4 (3.59%) and Factor 5 (0.84%) contributed little to EPFRs. Factor 4 is typically characterized by Cd, Cu and As, and also contains small amounts of Ni and Zn. These elements are commonly used in the electroplating industry and the metallurgical industry (Voutsa et al., 2002; Querol et al., 2006). This source is identified as an industrial source; The contribution of factor 5 is small and negligible. As shown in Figure S6, factor analysis of winter particulate matter indicated that Factor 4 (6.79%), Factor 5 (4.91%), Factor 6 (2.91%), and Factor 7 (1.65%) contributed little to EPFRs. The typical spectral characteristic of factor 4 is that the contribution of Ba is very high, Sr and EC also have partial contributions, Ba is related to road dust (Pan et al., 2013; Srivastava et al., 2007; Trapp et al., 2010), and Sr and EC are related to coal combustion,[3] therefore the factor is determined to be ground dust; The typical spectral characteristics of factor 5 are Ni and Cu, which represent the electroplating industry or the metallurgical industry (Voutsa et al., 2002; Querol et al., 2006); The typical spectral characteristics of Factor 6 and Factor 7 are Al, Ba, Co and Mn, all of which are associated with shell dust and resuspended road dust (Pan et al., 2013; Srivastava et al., 2007; Trapp et al., 2010), so the factor is determined to be atmospheric dust. The analysis results show that the contribution of atmospheric dust to EPFRs in winter is about 11.35%, and the contribution of industrial sources to summer and winter EPFRs is 3.44% and 4.91%, respectively.

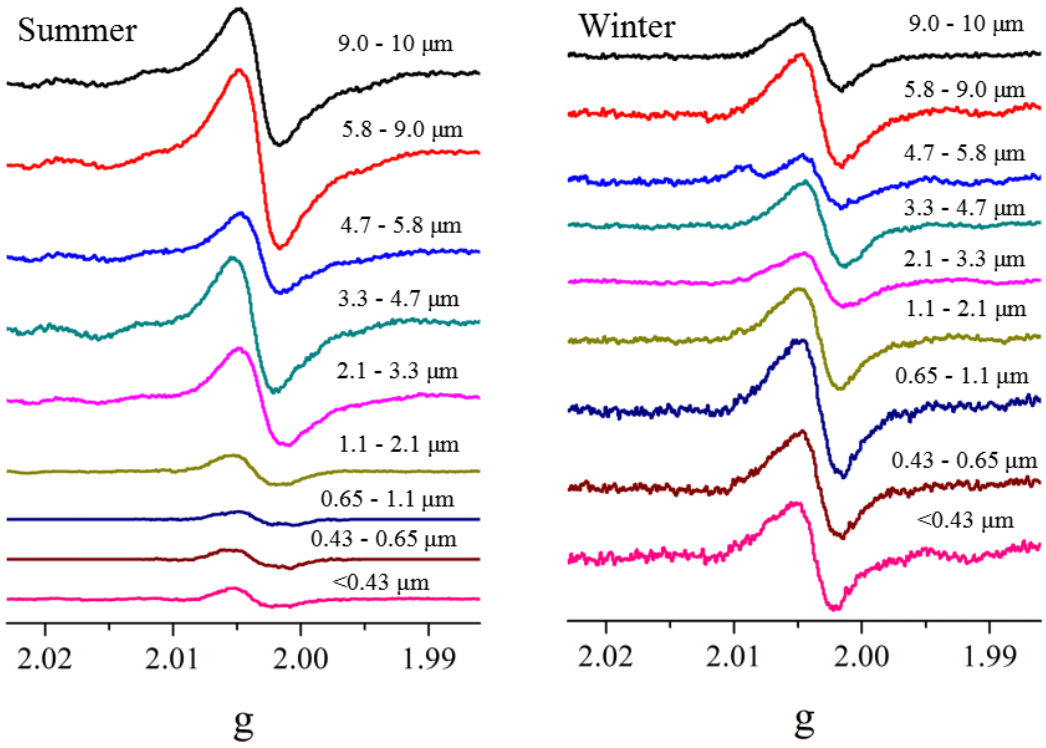

Figure S1. Average EPR spectra of EPFRs in different particle sizes in summer and winter. Summer, number of samples: 108; Winter, number of samples: 90.

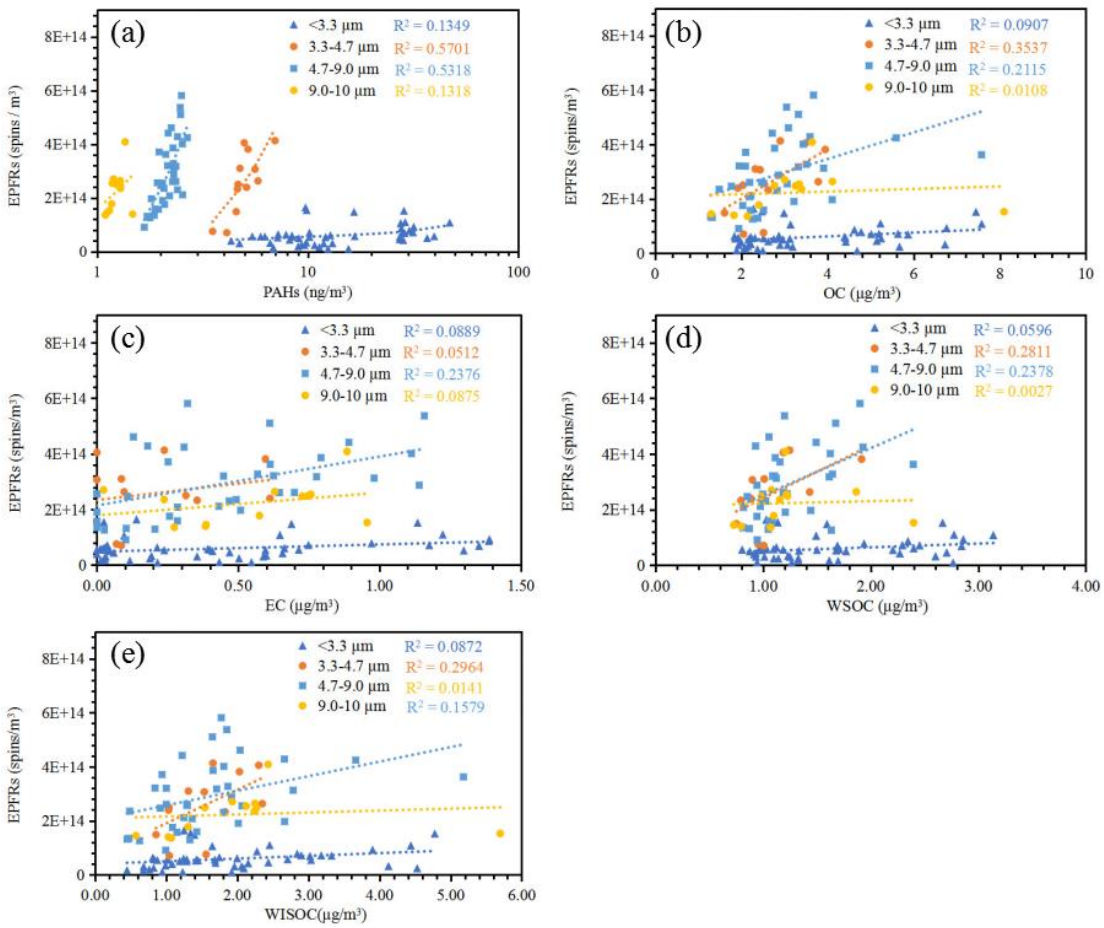

Figure S2. Correlation analysis of EPFRs with carbonaceous and PAHs in summer. a, b, c, d, and e represent polycyclic aromatic hydrocarbons (PAHs), organic carbon (OC), elemental carbon (EC), water-soluble organic carbon (WSOC), and water-insoluble organic carbon (WISOC), respectively.

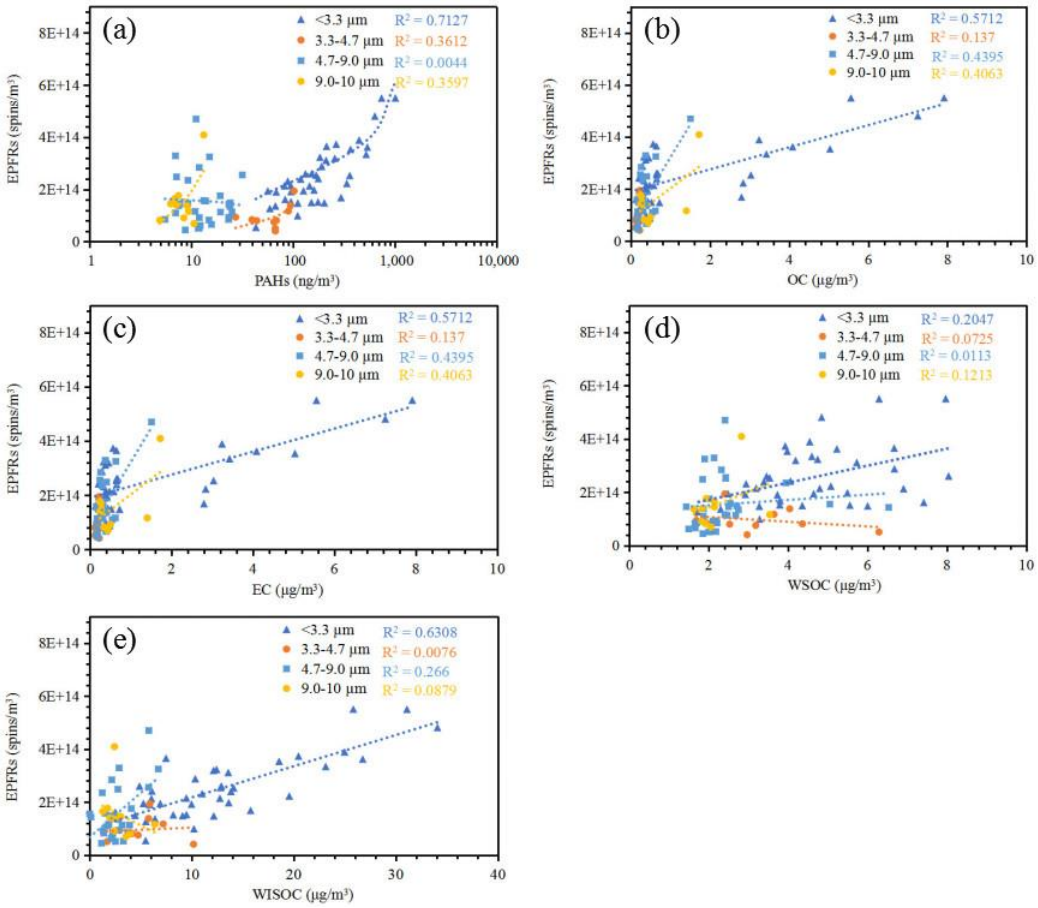

Figure S3. Correlation analysis of EPFRs with carbonaceous and PAHs in winter. a, b, c, d, and e represent polycyclic aromatic hydrocarbons (PAHs), organic carbon (OC), elemental carbon (EC), water-soluble organic carbon (WSOC), and water-insoluble organic carbon (WISOC), respectively.

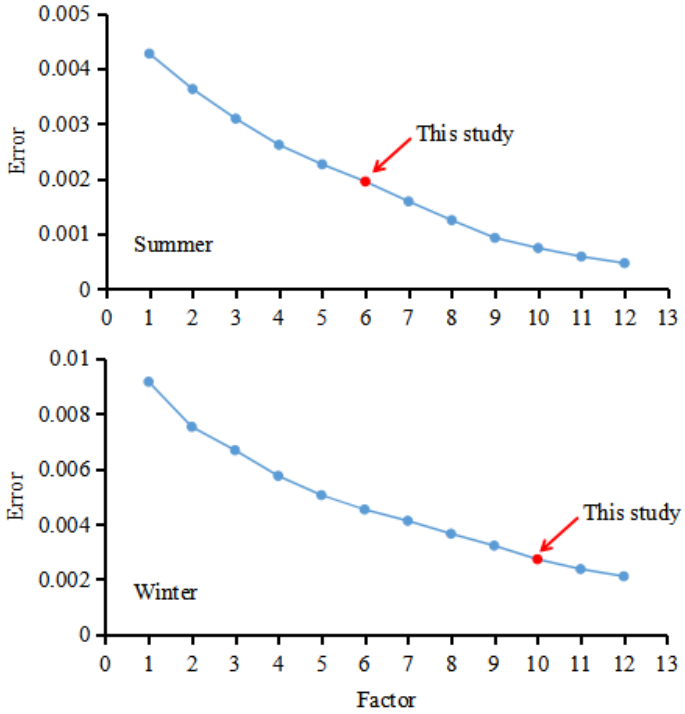

Figure S4. Factor analysis error.

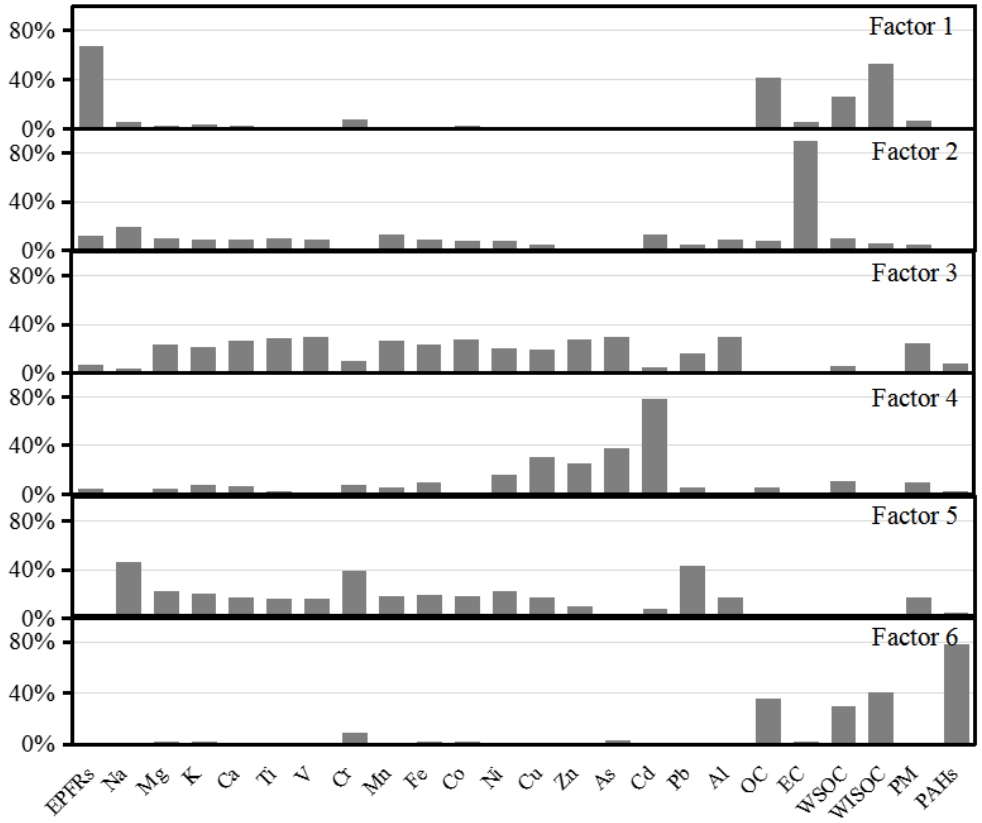

Figure S5. Factor analysis of atmospheric particulate matter in summer.

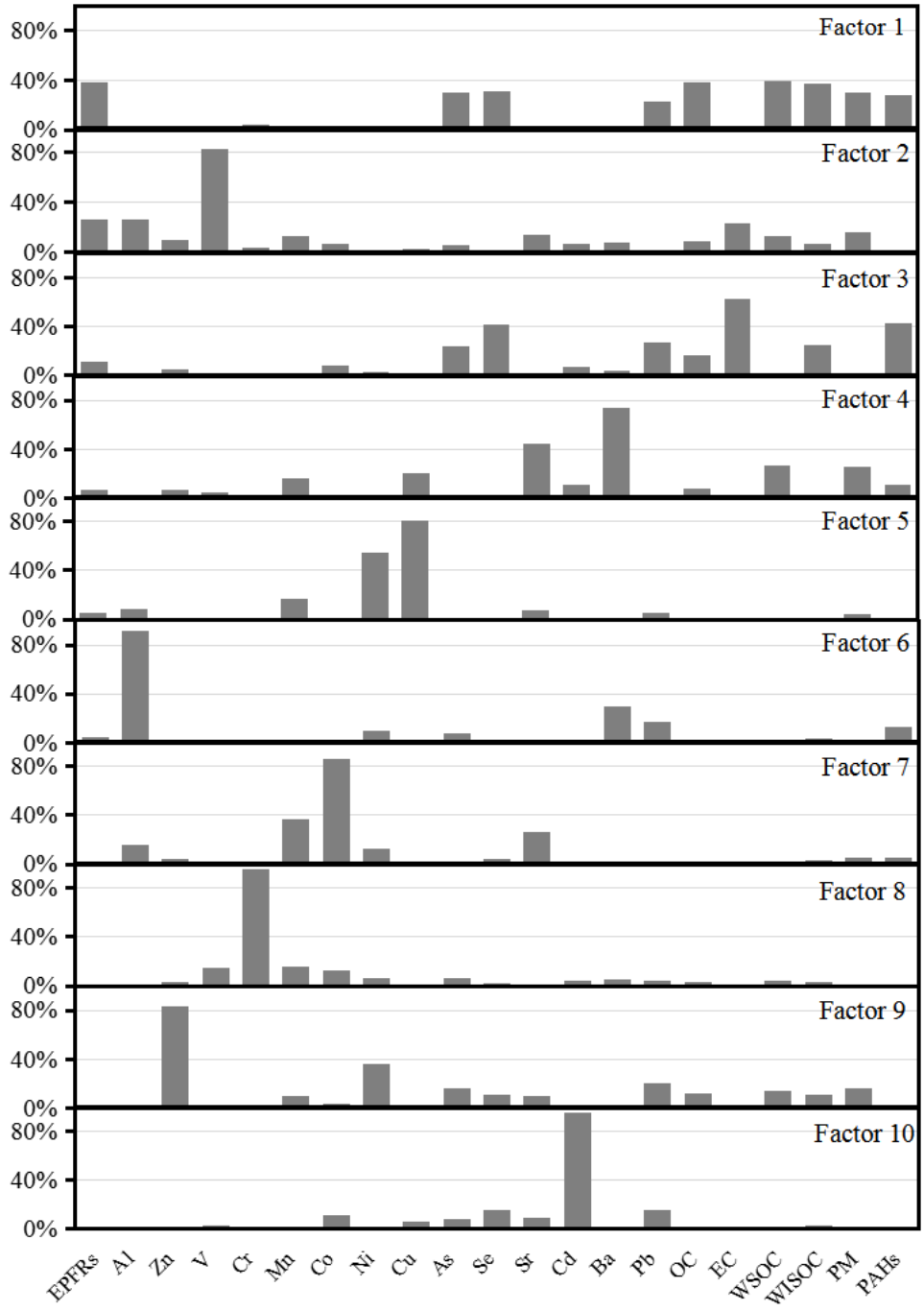

Figure S6. Factor analysis of atmospheric particulate matter in winter.

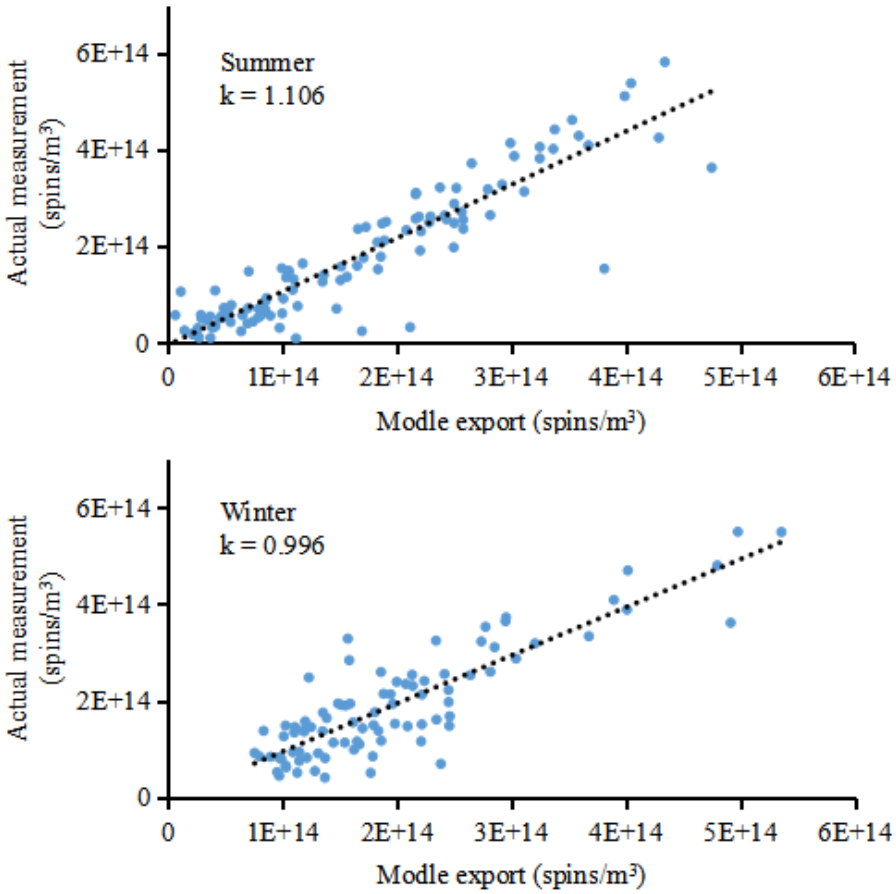

Figure S7. Comparison of the concentration of modle export and actual measurement.

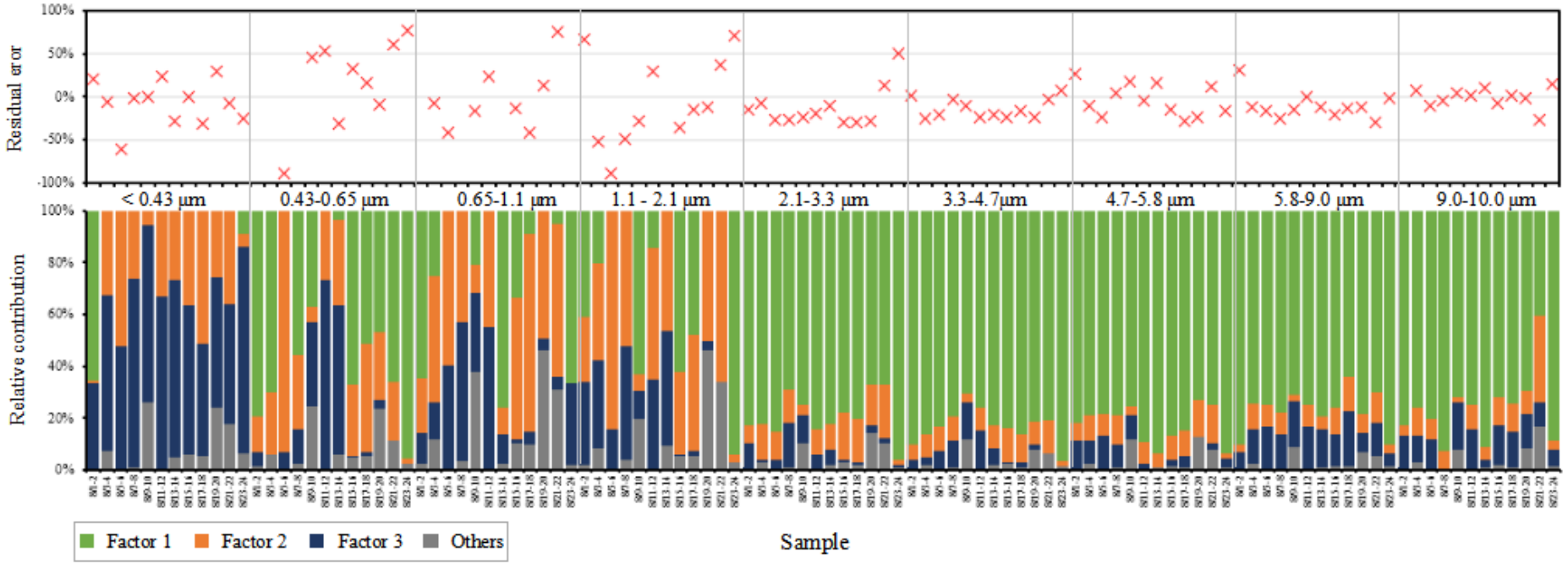

Figure S8. Relative contribution of EPFRs and residual of model in summer.

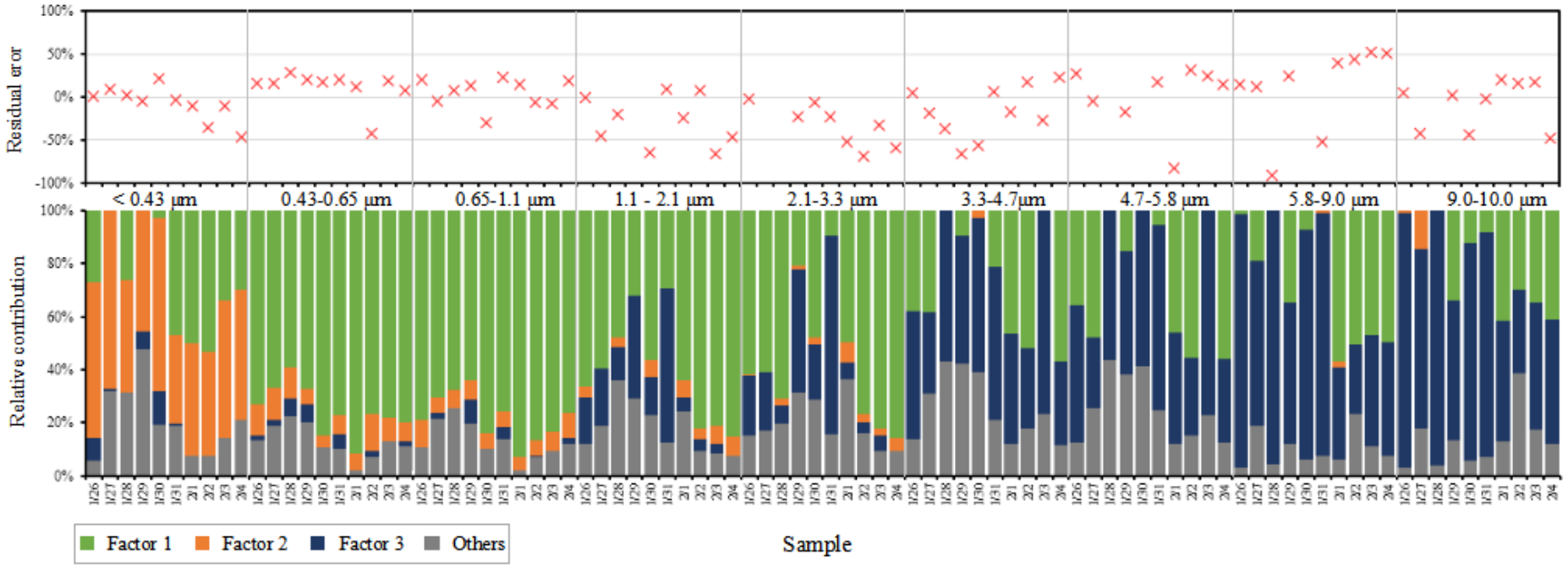

Figure S9. Relative contribution of EPFRs and residual of model in winter.

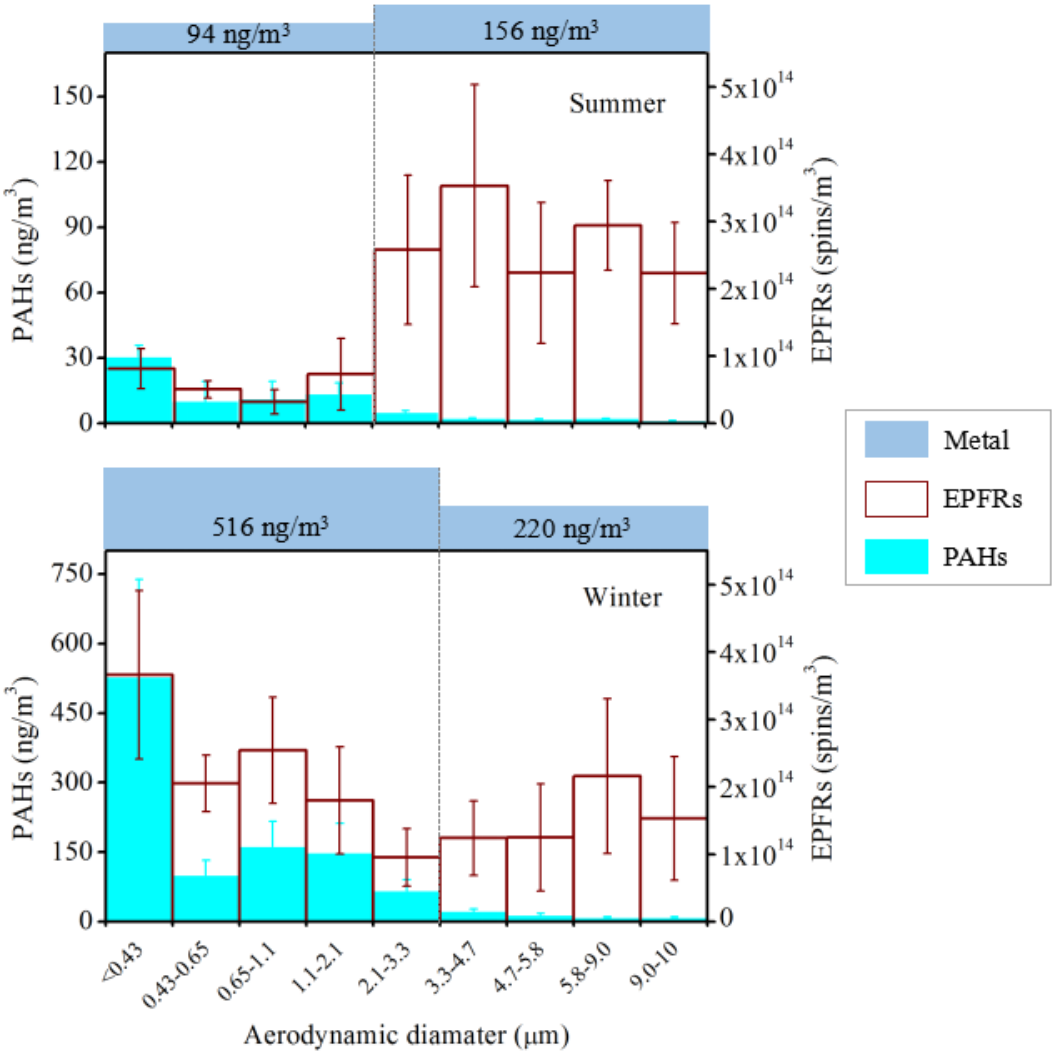

Figure S10. Correlation between EPFR and PAH concentrations in different particle sizes.

Table. S1.

Correlation analysis between EPFRs and metal element concentrations in particulate matter in summer.

Particle size(μm)	Correlation coefficient	Na	Mg	K	Ca	Ti	V	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb	Al
< 2.1	Pearson	0.374	0.527	0.093	0.448	0.369	0.101	-0.142	-0.043	0.012	0.032	0.518	0.504	-0.277	-0.284	0.549
	Significance two - tailed	0.231	0.078	0.774	0.144	0.237	0.756	0.66	0.894	0.97	0.923	0.085	0.095	0.383	0.371	0.065
	N								12							
2.1 - 10	Pearson	0.054	.772**	.786**	.862**	.917**	.928**	.730**	.784**	.874**	0.41	-0.032	-0.009	-0.288	-0.452	.901**
	Significance two - tailed	0.868	0.003	0.002	0	0	0	0.007	0.003	0	0.185	0.923	0.978	0.364	0.14	0
	N								12							

*Indicates a significant correlation at the 0.05 level. **Indicates a significant correlation at the 0.01 level; indicates $p < 0.001$.

Table. S2.

Correlation analysis between EPFRs and metal element concentrations in particulate matter in winter.

Particle size(μm)	Correlation coefficient	Al	Zn	V	Cr	Mn	Co	Ni	Cu	As	Se	Sr	Cd	Ba	Pb
< 3.4	Pearson	0.561	-0.15	-0.206	-0.129	-0.158	0.680*	-0.754*	-0.521	0.546	0.580	0.475	0.403	0.063	0.290
	Significance two - tailed	0.092	0.679	0.569	0.722	0.663	0.031	0.012	0.123	0.102	0.079	0.165	0.248	0.862	0.416
	N	10													
3.4 - 10	Pearson	.629*	.702*	0.143	-0.130	0.117	.682*	-0.568	-0.081	.737**	.723*	.621*	0.508	0.142	.633*
	Significance two - tailed	0.038	0.016	0.674	0.703	0.731	0.021	0.068	0.813	0.01	0.012	0.041	0.111	0.677	0.036
	N	10													

*Indicates a significant correlation at the 0.05 level. **Indicates a significant correlation at the 0.01 level; indicates $p < 0.001$

Table. S3.

Particulate matter deposition rate and EPFRs concentration in different areas of the respiratory tract in summer.

Particle size (μm)	Area of the respiratory system					
	ET		TB		P	
	Deposition rate	EPFRs (spins/m^3)	Deposition rate	EPFRs (spins/m^3)	Deposition rate	EPFRs (spins/m^3)
<0.43	0.08	6.78E+12	0.12	9.50E+12	0.30	2.41E+13
0.43-0.65	0.05	2.53E+12	0.01	2.53E+11	0.23	1.14E+13
0.65-1.1	0.12	3.84E+12	0.01	1.60E+11	0.19	5.92E+12
1.1-2.1	0.17	1.24E+13	0.01	7.30E+11	0.20	1.46E+13
2.1-3.3	0.36	9.16E+13	0.05	1.29E+13	0.23	5.80E+13
3.3-4.7	0.63	2.22E+14	0.22	7.76E+13	0.14	4.94E+13
4.7-5.8	0.59	1.31E+14	0.16	3.47E+13	0.20	4.36E+13
5.8-9.0	0.63	1.85E+14	0.30	8.67E+13	0.07	2.06E+13
9.0-10	0.63	1.41E+14	0.37	8.26E+13	0.00	0.00E+00

Table. S4.

Particulate matter deposition rate and EPFRs concentration in different areas of the respiratory tract in winter.

Particle size (μm)	Area of the respiratory system					
	ET		TB		P	
	Deposition rate	EPFRs (spins/m^3)	Deposition rate	EPFRs (spins/m^3)	Deposition rate	EPFRs (spins/m^3)
<0.43	0.08	3.05E+13	0.12	4.27E+13	0.30	1.09E+14
0.43-0.65	0.05	1.02E+13	0.01	1.02E+12	0.23	4.60E+13
0.65-1.1	0.12	3.05E+13	0.01	1.27E+12	0.19	4.71E+13
1.1-2.1	0.17	3.06E+13	0.01	1.80E+12	0.20	3.60E+13
2.1-3.3	0.36	3.38E+13	0.05	4.77E+12	0.23	2.14E+13
3.3-4.7	0.63	7.81E+13	0.22	2.73E+13	0.14	1.74E+13
4.7-5.8	0.59	7.33E+13	0.16	1.94E+13	0.20	2.44E+13
5.8-9.0	0.63	1.36E+14	0.30	6.37E+13	0.07	1.51E+13
9.0-10	0.63	9.63E+13	0.37	5.66E+13	0.00	0.00E+00

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