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Sources and trends of black carbon aerosol in the megacity of Nanjing, eastern China, after the China Clean Action Plan and Three-Year Action Plan

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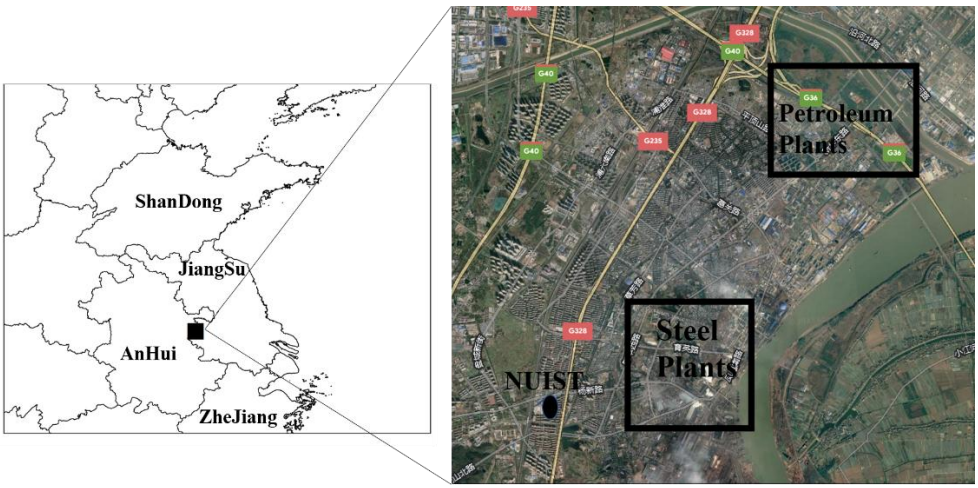

Figure S1 The location of sampling site, In the right panel, the sampling site in Nanjing is marked with a black elliptical dot (© Google Earth)

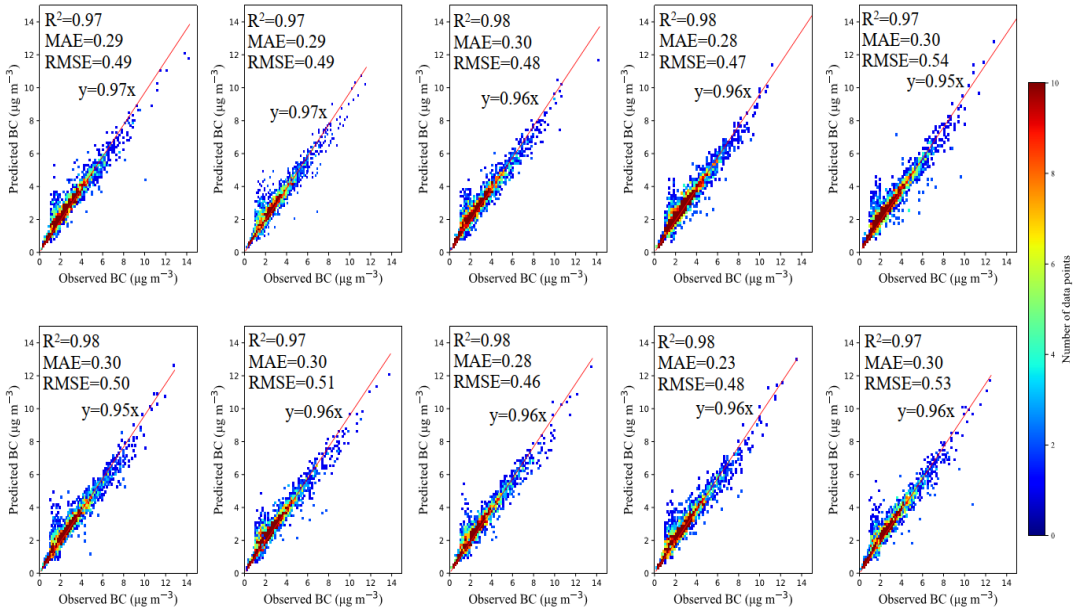

Figure S2 10-fold cross-validation results for the random forest model predicting BC concentration at 880 nm. Each panel in the figure corresponds to one of the 10 folds.

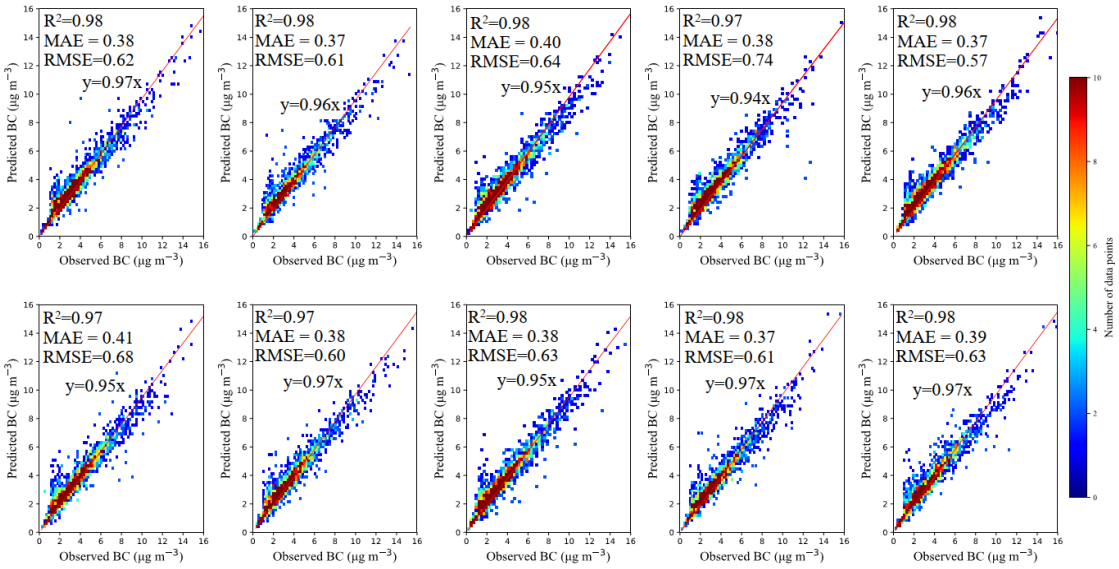

Figure S3 10-fold cross-validation results for the random forest model predicting BC concentration at 370 nm. Each panel in the figure corresponds to one of the 10 folds.

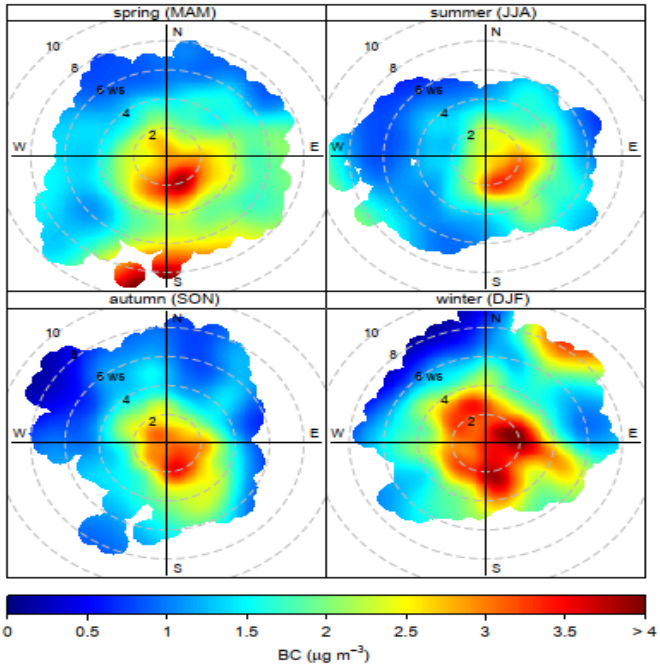

Figure S4 Bivariate polar plots of hourly BC concentration in four seasons

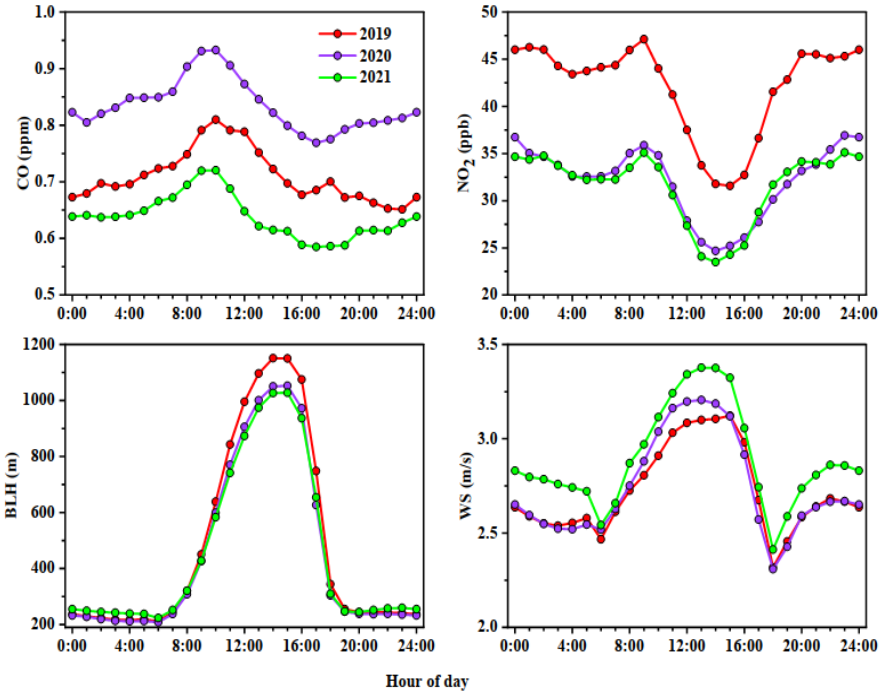

Figure S5 Diurnal variation of pollutant gases and meteorology factors

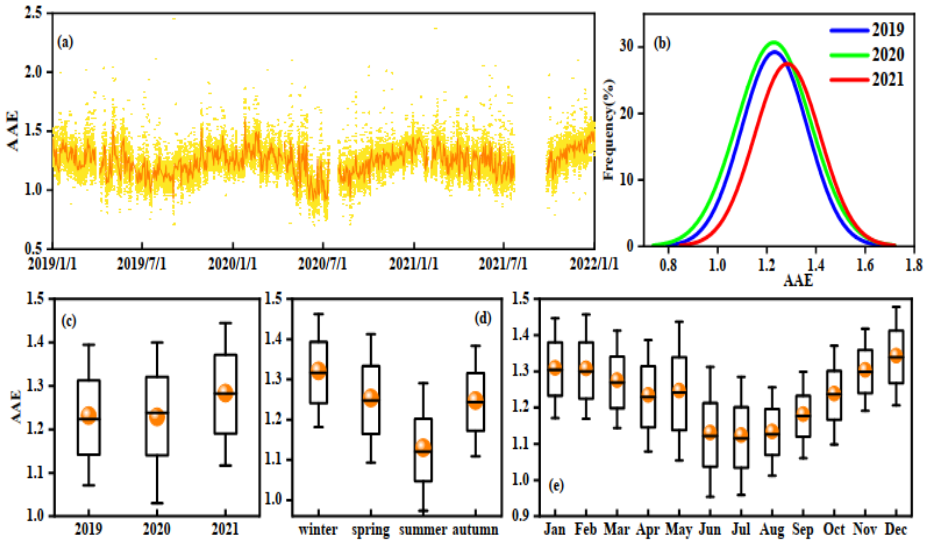

Figure S6 Hourly, annually, seasonal and monthly variation of AAE

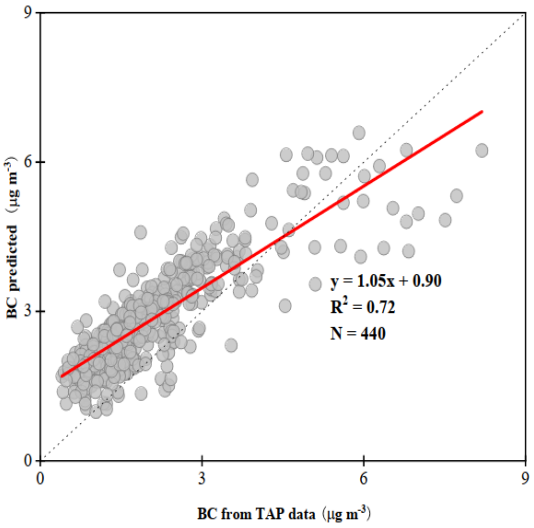

Figure S7 Comparison of BC from TAP data and predicted daily average BC by the machine learning models

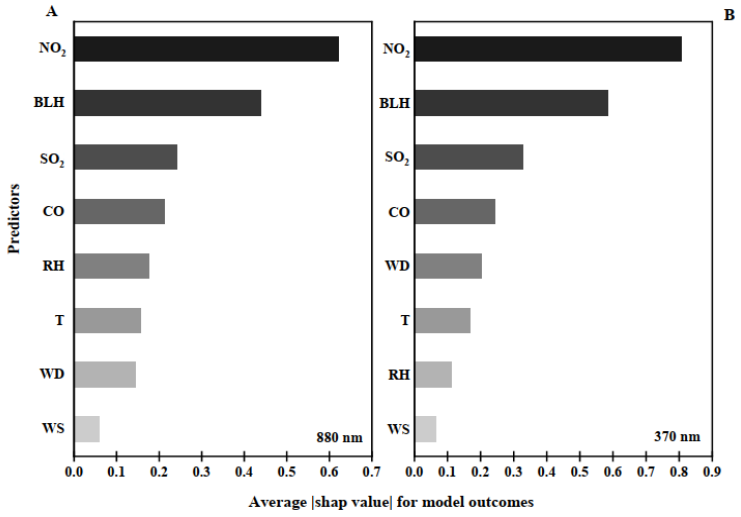

Figure S8 The predictor's importance for BC at (A) 880 nm and (B) 370 nm

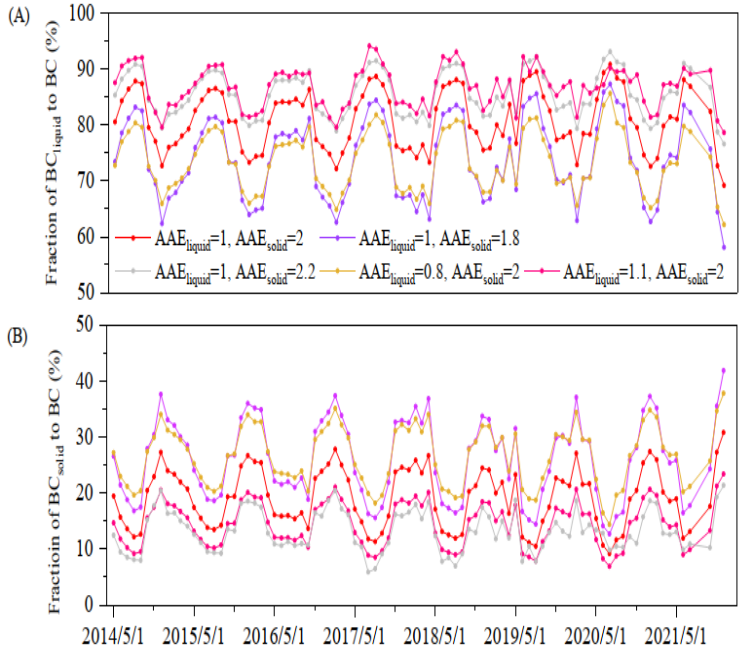

Figure S9 Uncertainty analysis of BC source apportionment using the Aethalometer model. Contributions of liquid and solid fuel combustion to BC were calculated using five different combinations of AAE_{liquid} and AAE_{solid}

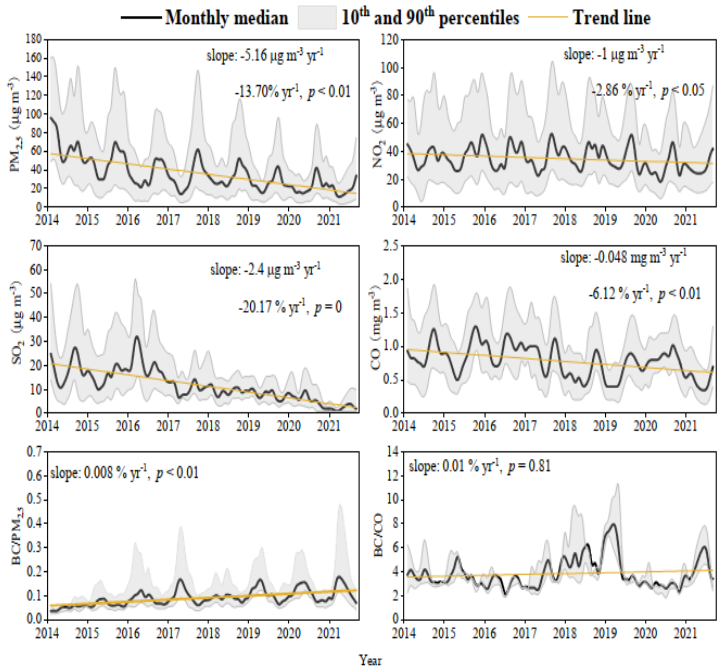

Figure S10 Trends in air pollutants at sampling site. The solid black line represents the monthly medians, the dash black lines represent the 10th and 90th monthly percentiles, and the orange line is the fitted long-term trend.

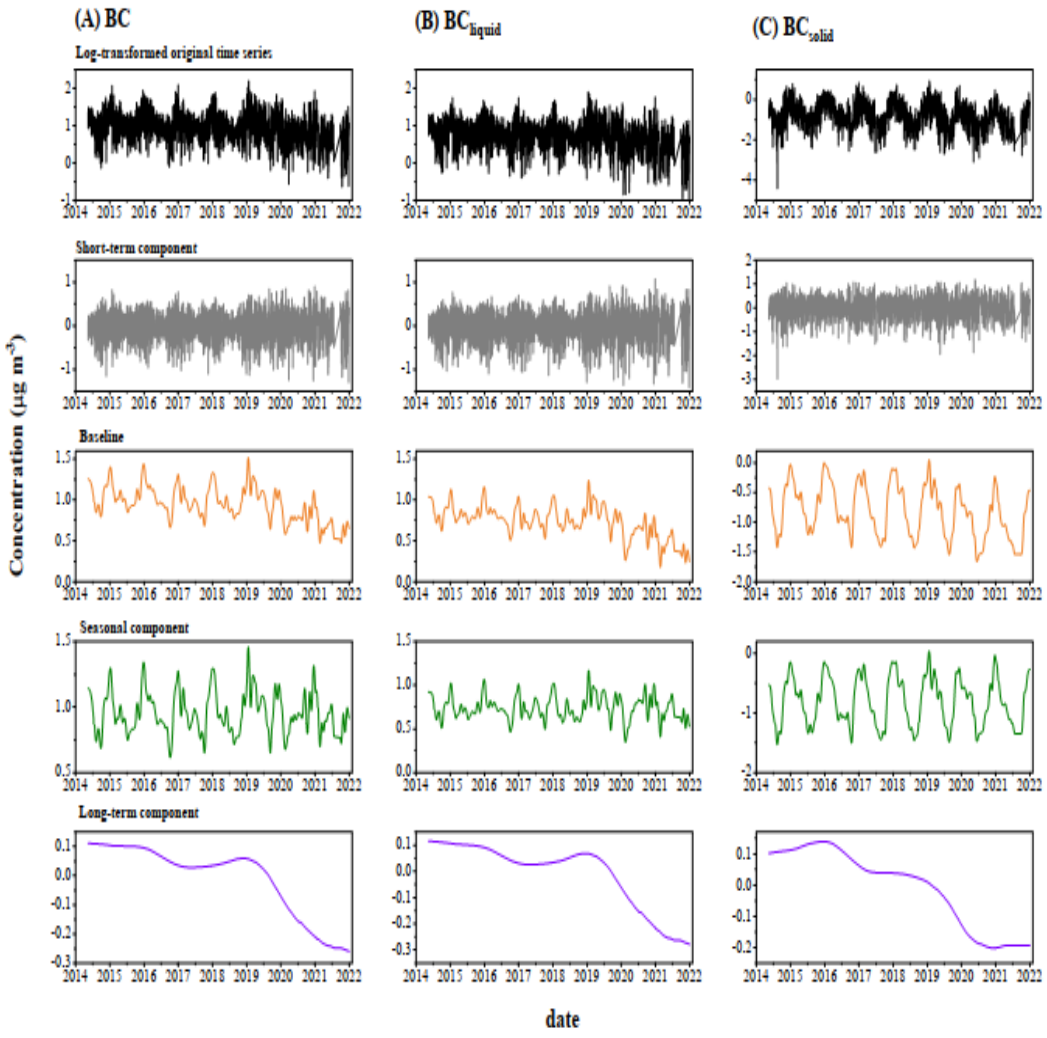

Figure S11 Different component distinguished by KZ filter for (A) BC (B) BC_{liquid} (C) BC_{solid}

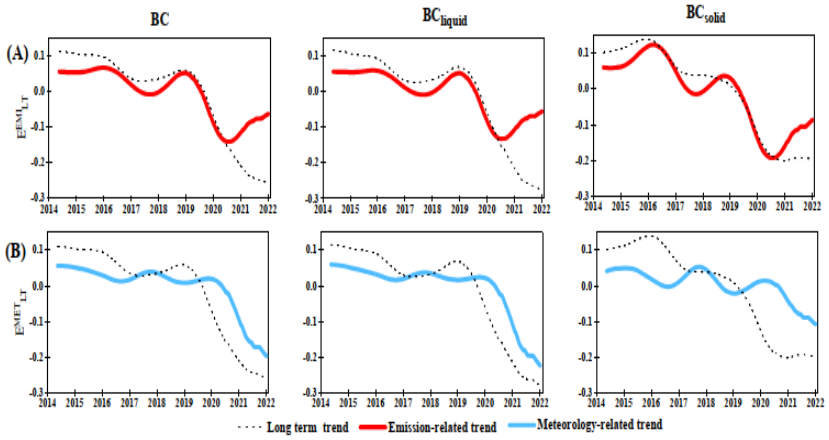

Figure S12 (A) Emission-related and (B) meteorology-related trends of BC. The left, middle and right panels represent BC, BC_{liquid} and BC_{solid}.

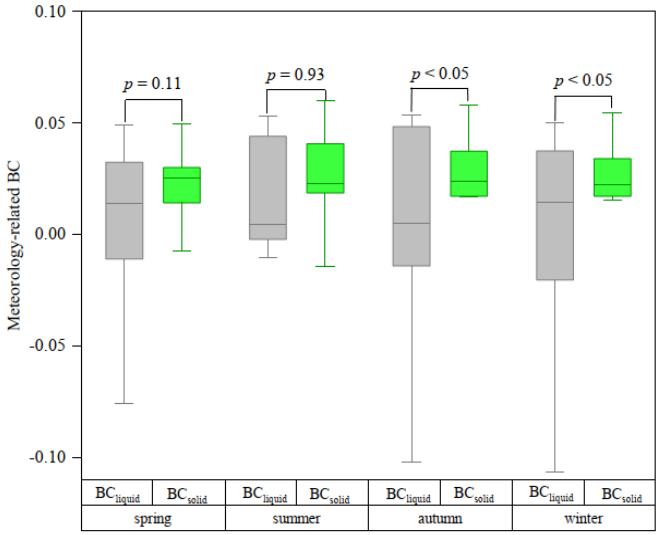

Figure S13 Seasonal variation of meteorology-related BC_{liquid} and BC_{solid}. The square in the figure represents the 25th and 75th percentiles of the data, the vertical lines represent the 10th and 90th percentiles, and the horizontal line inside the square indicates the median. The Y-axis represents the log-transformed concentrations of BC_{liquid} and BC_{solid}

Table S1 The relative contributions (%) of fossil fuel to BC estimated by Aethalometer model with the different AAE_{BB}-AAE_{FF} pairs

Parameters	AAE_{solid}=1.8	AAE_{solid}=2.0	AAE_{solid}=2.2
AAE_{liquid}=0.8	62%	71%	77%
AAE_{liquid}=1	73%	80%	85%
AAE_{liquid}=1.1	81%	87%	90%

Table S2 $BC/PM_{2.5}$ and BC/CO ratios from different sources

Sources	$BC/PM_{2.5}$
Biomass burning & coal	
Agricultural burning	0.058-0.131[a]
Forest fire	0.032[a]
Residential wood combustion	0.042-0.33[a]
Residential coal combustion	0.26[a]
Fossil fuel	
Light-duty gasline	0.059-0.37[a]
Heavy-duty diesel	0.33-0.74[a]
Light-duty diesel	0.62-0.64[a]

Sources	BC/CO
Industry	0.0072[b]
Power plant	0.0177[b]
Residential	0.0371[b]
Traffic	0.0052[b]

a:(Chow et al., 2011)

b:(Zhang et al., 2009)

Table S3 The change rates of BC and other air pollutants across different seasons

season	pollutants	absolute slope[a]	relative slope[b]	p
spring	BC	-0.19	-7.16%	0.003
	BC_{liquid}	-0.16	-7.60%	0.003
	BC_{solid}	-0.05	-8.74%	0.013
	$PM_{2.5}$	-6.00	-15.90%	0.001
	NO_2	-3.00	-7.82%	0.003
	SO_2	-3.38	-28.15%	0.001
	CO	-0.09	-11.67%	0.008
summer	BC	-0.13	-5.16%	0.013
	BC_{liquid}	-0.09	-4.16%	0.011
	BC_{solid}	-0.04	-12.65%	0.000
	$PM_{2.5}$	-6.67	-25.44%	0.001
	NO_2	-1.50	-5.50%	0.054
	SO_2	-2.20	-20.17%	0.000
	CO	-0.04	-6.06%	0.090
autumn	BC	-0.13	-4.87%	0.030
	BC_{liquid}	-0.11	-5.08%	0.008
	BC_{solid}	-0.01	-1.64%	0.790
	$PM_{2.5}$	-6.00	-20.02%	0.001
	NO_2	-0.36	-3.28%	0.790
	SO_2	-2.29	-20.61%	0.001
	CO	-0.03	-3.55%	0.001
winter	BC	-0.33	-9.95%	0.001
	BC_{liquid}	-0.26	-10.50%	0.001
	BC_{solid}	-0.06	-8.55%	0.006
	$PM_{2.5}$	-7.06	-14.20%	0.000
	NO_2	-1.33	-3.35%	0.413
	SO_2	-4.67	-36.15%	0.001
	CO	-0.09	-8.63%	0.001

[a]: $\mu g\ m^{-3}\ yr^{-1}$

[b]: $\%\ yr^{-1}$

Table S4 Total variance of log-transformed time series of BC, BC_{liquid} and BC_{solid} and relative contributions of variances of and covariances among each component to total variance

Component	BC	Bc_{liquid}	BC_{solid}
Var(X)	0.16	0.16	0.40
$Var(X_{ST})$	68.87%	72.80%	51.71%
$Var(X_{SN})$	15.90%	11.37%	39.71%
$Var(X_{LT})$	8.00%	8.10%	0.35%
Cov (X_{SN}, X_{ST})	2.73%	2.76%	0.23%
Cov (X_{SN}, X_{LT})	0.90%	1.15%	0.00%
Cov (X_{ST}, X_{LT})	0.00%	0.00%	2.44%
$Var(X_{LT}^{emi})$	2.76%	2.31%	2.31%
$Var(X_{LT}^{met})$	2.16%	2.68%	0.35%
Cov (X_{ST}, X_{LT})	1.50%	1.54%	0.41%

Chow, J. C., Watson, J. G., Lowenthal, D. H., Antony Chen, L. W., and Motallebi, N.: PM2.5 source profiles for black and organic carbon emission inventories, Atmospheric Environment, 45, 5407-5414, https://doi.org/10.1016/j.atmosenv.2011.07.011, 2011.

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