

## **Chemical characterization and source apportionment of PM<sub>2.5</sub> during spring and winter in the Yangtze River Delta, China**

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PCA/APCS model, carried out by SPSS (version 19.0, IBM Corp. 2010), is utilized to analyze the source of PM<sub>2.5</sub> in the main cities and background city in YRD during spring and winter respectively. The main steps are as follows: (1) normalization of all species concentrations; (2) analyzation of variables' factor scores from PCA; (3) regression. The principal of PCA/APCS were well introduced from previous research (Guo et al., 2004; An et al., 2014).

The source identification (Table S1 and S2) was based on the main components of each factor. Take the source identification in Shanghai in spring as an example, he PCS1 was dominated by K<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, EC, OC and OM, and it accounted as 20.76% of total variance. K<sup>+</sup> and NH<sub>4</sub><sup>+</sup> were the tracer of biomass burning, and EC as well as OC were associated with the combustion sources, such as biomass burning and fossil fuel combustion (Viana et al., 2008; Deshmukh et al., 2012; Zong et al., 2016). At the same time, OM and NO<sub>3</sub><sup>-</sup> were the tracer of traffic emission. Therefore, PCS1 could be regarded as the combination of biomass burning and vehicles. PCS2 was associated with Mg, Si, Al, Ca and Ti, which were mainly regarded as the crustal element, as a result, PCS2 was the source of dust. In PCS3, Fe and Mn were mainly emitted by steel industry and Cu was also the tracer of industry (Taiwo et al., 2014), therefore, PCS3 was defined as the source of industry activities. In PCS4, the main loadings of species were Zn, As and Ba, except EC and OM. As analyzed above, EC and OM could be the tracer of combustion source, and As was also the mainly related with coal combustion. Additionally, Zn was the major emission from fossil fuel combustion, waste incineration, and even the sectors of sinter and basic oxygen steel making (Jones and Norman, 1997; Xu et al., 2013; Taiwo et al., 2014). Therefore, PCS4 should be defined as coal combustion. The main loadings of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in PCS5 were mainly from oxidation of the gaseous precursors (SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub>) that were emitted from anthropogenic activities (Wang et al., 2016a), therefore, PCS5 was the source of secondary aerosols.

Besides, when the Kaiser-Meyer-Olkin (KMO) value was above 0.6, the PCA result was acceptable (Wang et al., 2015). However, the KMO value of the Lin'an analysis in spring was 0.544, the result of PCA was unacceptable (Table S1).

The contributions of each source to the PM<sub>2.5</sub> that in the main cities and background site during spring and winter were calculated (Table S3 and S4). It is clear that the sources could not be divided completely, the reason might be that the limitation of samples in each city and each season, for example, there were only 25 samples in Lin'an in winter. The number of samples should be more than 50 to achieve a reliable result in PCA analysis (Thurston and Spengler, 1985; Wang et al., 2016a), however only the samples of Shanghai and Ningbo during spring, as well as Nanjing during winter were more than 50. Therefore, majority of the results achieved (Table S3 and S4) were not stable and reliable. At the same time, it seems that there were differences among the sites based on our PCA/APCS results, but the entire contribution of main sources (coal combustion, secondary aerosol, vehicles and biomass burning) in each city was comparable. For example, the entire contribution of coal combustion, secondary aerosol, vehicles and biomass burning was around 60% in spring, while that was almost 55% in winter.

Table S1. PCA factor loadings for PM<sub>2.5</sub> in spring (only those with values  $\geq 0.45$  are presented)

	PCS1	PCS2	PCS3	PCS4	PCS5	PCN1	PCN2	PCN3	PCN4	PCN5
	Shanghai					Nanjing				
EC	0.763			0.514			0.920			
OC	0.814						0.913			
OM	0.748			0.520			0.925			
Mg		0.910				0.937				
Al		0.937				0.941				
Si		0.917				0.946				
Ca		0.746				0.761				
Ti		0.660				0.665				
Mn			0.968					-0.85		
Fe			0.915					0.874		
Cu			0.965					0.878		
Zn				0.746		0.581	0.498			
As				0.734			0.616			
Ba				0.615						
Cl <sup>-</sup>						0.588			0.542	
SO <sub>4</sub> <sup>2-</sup>					0.831				0.460	0.765
NO <sub>3</sub> <sup>-</sup>	0.673				0.541				0.884	
Na <sup>+</sup>	0.748									
NH <sub>4</sub> <sup>+</sup>	0.562				0.733				0.917	
K <sup>+</sup>	0.776						0.485			
% of Variance	20.76	20.15	14.81	13.06	10.52	26.77	21.57	14.53	14.11	7.52

  

	PCNB1	PCNB2	PCNB3	PCNB4	PCLA1	PCLA2	PCLA3	PCLA4
	Ningbo				Lin'an			
EC	0.890				-	-	-	-
OC	0.885				-	-	-	-
OM	0.703			0.464	-	-	-	-
Mg		0.911			-	-	-	-
Al		0.957			-	-	-	-
Si		0.956			-	-	-	-
Ca		0.767			-	-	-	-
Ti		0.677			-	-	-	-
Mn			-0.947		-	-	-	-

Fe			0.857		-	-	-	-
Cu			0.849		-	-	-	-
Zn				0.849	-	-	-	-
As				0.755	-	-	-	-
Cl				0.677	-	-	-	-
SO <sub>4</sub> <sup>2-</sup>	0.851				-	-	-	-
NO <sub>3</sub> <sup>-</sup>	0.930				-	-	-	-
NH <sub>4</sub> <sup>+</sup>	0.870				-	-	-	-
K	0.848				-	-	-	-
% of Variance	30.07	22.41	13.56	12.93	-	-	-	-

Table S2. PCA factor loadings for PM<sub>2.5</sub> in winter (only those with values  $\geq 0.45$  are presented)

	PCSH1	PCSH2	PCSH3	PCSH4	PCNJ1	PCNJ2	PCNJ3	PCNJ4
	Shanghai				Nanjing			
OC			0.848		0.835			
EC			0.893		0.781			
Mg		0.858				0.930		
Al		0.888				0.975		
Si		0.853				0.944		
Ca		0.872				0.862		
Ti		0.710				0.861		
Mn				-0.944				-0.957
Fe				0.867				0.731
Cu				0.827				0.846
Zn	0.730	0.475			0.821			
As	0.669				0.813			
Cl <sup>-</sup>	0.832						0.454	
SO <sub>4</sub> <sup>2-</sup>	0.813						0.832	
NO <sub>3</sub> <sup>-</sup>	0.848						0.803	
Na <sup>+</sup>							0.745	
NH <sub>4</sub> <sup>+</sup>	0.882				0.808			
K <sup>+</sup>	0.870				0.869			
% of Variance	29.79	22.29	15.21	14.07	25.82	25.49	15.38	12.74

	PCNB1	PCNB2	PCNB3	PCNB4	PCLA1	PCLA2	PCLA3
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	Ningbo				Lin'an		
OC	0.465	0.797			0.662		
EC	0.531	0.743			0.737		0.462
Mg	0.861				0.892		
Al	0.924				0.938		
Si	0.932				0.920		
Ca	0.576				0.866		
Ti	0.777					0.830	
Mn				0.856			0.810
Fe				-0.844			
Cu			-0.500	-0.669			-0.784
Zn	0.552	0.751			0.512		0.603
As	0.604	0.635			0.620		0.473
Cl		0.879					
SO42			0.822			0.910	
NO3			0.723			0.829	
Na		0.660					
NH4		0.565	0.678			0.856	
K		0.693					0.767
% of Variance	28.78	25.79	15.22	12.76	35.53	24.37	21.81

Table S3. The source contributions (%) of PM<sub>2.5</sub> in spring

	Coal combustion	Secondary aerosols	Vehicles	Biomass burning	Dust	Industry activities
SH	9.00	21.00		25.97	18.29	5.03
NJ		47.37		19.05	9.31	4.70
NB	24.91		45.87		0.17	8.01
LA	-	-	-	-	-	-

Table S4. The source contribution (%) of PM<sub>2.5</sub> in winter

	Secondary aerosols	Coal combustion	Biomass burning	Vehicles	Dust	Industry activities
SH		43.41		11.57	16.12	10.26
NJ	18.00	-		54.20	5.00	2.21
NB	20.75		30.95		18.89	11.96
LA	41.89	-	-	19.02		20.79

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