Seasonal Source Apportionment of PM$_{2.5}$ in Ningbo, a Coastal City in Southeast China

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ABSTRACT

With the rapid economic development and urbanization of China, haze and photochemical smog events have been frequently observed during the last decade. To explore the temporal and spatial pollution characteristics in Ningbo, a medium-sized coastal city located in the Yangtze River Delta (YRD) in southeast China, 24-h PM$_{2.5}$ (particulate matter with aerodynamic diameter $\leq$ 2.5 µm) samples were simultaneously collected at five sites (two urban residential sites, two urban coastal sites, and one suburban site) from winter 2012 to autumn 2013. The average PM$_{2.5}$ concentration was 53.2 ± 30.4 µg m$^{-3}$. Furthermore, the concentration exhibited a seasonal variation: It was highest in winter and lowest in summer. The urban residential sites had the highest PM$_{2.5}$ concentrations, followed by the urban coastal sites, and the suburban site had the lowest concentration. OM (Organic Matters) and secondary inorganic ions (sulfate, nitrate, and ammonium) were the dominant components of the PM$_{2.5}$. As a coastal city with industrial zones, sources are more complex in Ningbo than in inland cities due to ship emissions and the interactions between land and sea, and the marine and atmospheric environments. Positive matrix factorization (PMF) was used to apportion the particle sources. Nine factors were resolved in this study: secondary nitrate, vehicle exhaust, secondary sulfate, coal combustion, industrial emission, ship emission, dust, biomass burning, and aged sea salt, with average contributions of 26%, 21%, 13%, 12%, 9%, 7%, 5%, 4%, and 3%, respectively. Secondary nitrate and vehicle exhaust were the major sources of PM$_{2.5}$ pollution in Ningbo. Coal combustion contributed significantly in winter and autumn, whereas sea salt formed a considerable contribution in summer. This study suggests that decreasing the PM$_{2.5}$ pollution in Ningbo requires not only strategies for reducing local primary sources but also joint inter-regional prevention and the control of air pollution in the YRD.

Keywords: PM$_{2.5}$; Source apportionment; Positive matrix factorization (PMF); Temporal and spatial distribution.

INTRODUCTION

Ambient particulate matter (PM) originates from natural and anthropogenic sources, including primary emissions and secondary formation through atmospheric reactions and gas-to-particle conversions (Hallquist et al., 2009). PM$_{2.5}$ (particulate matters with aerodynamic diameters $\leq$ 2.5 µm) pollution can cause serious environmental problems such as reduced visibility, acid rain, increased concentrations of cloud condensation nuclei (CCN), and climate change (Watson, 2002; Cao et al., 2004; Andreae et al., 2008). In addition, PM$_{2.5}$ has been linked to the increasing frequency of cardiovascular disease, respiratory disease, and cancer (Pope and Dockery, 2006).

A clear and complete understanding of the sources of PM$_{2.5}$ and their contributions is in urgent need for further emission reduction measures. Source apportionment of PM$_{2.5}$ has attracted increasing public attention in recent years. The scientifically reasonable results of PM$_{2.5}$ source apportionment will help the government design effective strategies for emission reduction (Viana et al., 2008). Various methods of source apportionments have been...
developed therefore. The most widely used methods are the receptor models, including positive matrix factorization (PMF), chemical mass balance (CMB), UNMIX, and principal component analysis (PCA) (Schauer et al., 1996; Song et al., 2006; Pekney et al., 2006; Zhang et al., 2009; Shi et al., 2011; Heo et al., 2013; Zhang et al., 2013). Among these models, PMF model is more favorable when sources are not well understood (Song et al., 2006; Heo et al., 2013; Zhang et al., 2013). PMF has been widely used to investigate the potential PM$_{2.5}$ sources in Europe, America, China, and some other Asian countries (Song et al., 2006; Baumann et al., 2008; Viana et al., 2008; Guo et al., 2009; Zhang et al., 2009; Deshmukh et al., 2011; Mooibroek et al., 2011; Chen et al., 2012; Heo et al., 2013; Yu et al., 2013; Zhang et al., 2013; Brines et al., 2016).

Rapid economic development and urbanization in China has been concentrated in coastal cities, resulting in haze and photochemical smog issues. Yangtze River Delta (YRD), which is one of the largest economic regions in China, has suffered a lot from the severe air pollution events. Ningbo, with 8 million people and 1.7 million vehicles, is the 4th most developed city in YRD, according to China’s National GDP report of 2015. Ningbo is a typical city in southeast China with densely-populated urban residential areas and coastal industries. A combination of local emissions, long-range transport, and emissions from the maritime activities, leads to a complicated air pollution situation in Ningbo. Previous researches have focused more on isolated periods or separate locations (Ye, 2011; Xu et al., 2015; Xu et al., 2016), and the composition profiles need to be updated with the changes of energy consumption structure. China’s energy structure is changing from coal dominated to diversification, and the driving force of energy development is changing from traditional energy growth to new energy growth (Wei and Ma, 2015). Thus, an overall perspective of the pollution characteristics of Ningbo is required for better pollution control strategies. In this study, PM$_{2.5}$ samples were collected at five sites in Ningbo and the sampling periods covered four seasons. There were three main objectives of this work: (1) to identify the pollution level and chemical composition in Ningbo; (2) to characterize the potential sources of PM$_{2.5}$ in Ningbo by applying the PMF model; (3) to provide a comprehensive perspective of PM$_{2.5}$ pollution features and source distribution in Ningbo, with variation on both temporal and spatial scales.

**MATERIAL AND METHODS**

**Location and Sample Collection**

PM$_{2.5}$ samples were collected at five sites which were located in five different districts in Ningbo, including Shizhan (SZ, 121°31’42.65ʺE, 29°51’54.09ʺN), Cixi (CX, 121°16’20.69ʺE, 30°9’36.74ʺN), Zhenhai (ZH, 121°42’58.41ʺE, 29°57’14.61ʺN), Beilun (BL, 121°50’23.75ʺE, 29°54’27.70ʺN), and Xiangshan (XS, 121°53’19.06ʺE, 29°28’35.51ʺN), as shown in Fig. 1 (Li et al., 2017). SZ and CX were inland urban sites with heavy traffic congestion and dense populations. ZH and BL were coastal urban sites with intensive industries. XS was a suburban site without major anthropogenic sources nearby. More detailed information about the sampling sites can be found elsewhere (Li et al., 2017). Diversity in the site types and scattered distribution of the sites made the results more representative for the air pollution characteristics in Ningbo.

A total of 302 samples (including field blanks) were collected during the sampling periods at the five sites, including winter (December 2012), spring (April 2013), summer (July 2013), and autumn (October 2013). Four-channel samplers (TH-16A, Tianhong, China) were used to collect PM$_{2.5}$ from 8:30 a.m. to 7:30 a.m. the next day at a flow rate of 16.7 L min$^{-1}$, with two channels for quartz filters (Whatman Inc., USA, 47 mm) and two channels for Teflon filters (Whatman Inc., USA, 47 mm). Before sampling, the quartz filters were pre-baked at 550°C for 5.5 h in a muffle furnace to remove the absorbed carbonaceous compounds.

![Fig. 1. Locations of Ningbo and five sampling sites (Li et al., 2017).](image-url)
After sampling, all filters were stored in a refrigerator at 
-18°C until analysis.

Chemical Analysis
Before and after sampling, the Teflon filters were equilibrated for 24 h in a controlled environment (relative humidity = 40 ± 5% and temperature = 20 ± 1°C) prior to the gravimetric measurement using an AXI05DR microbalance (Mettler-Toledo Ltd., Switzerland) with a precision of ± 0.001 mg. Teflon filters were used to analyze the water-soluble ions (Na\(^+\), K\(^+\), NH\(_4\)+, Ca\(^2+\), Mg\(^2+\), SO\(_4\)\(^2-\), NO\(_3\)\(^-\), Cl\(^-\), F\(^-\)) with the Dionex ICS-2000 and Dionex ICS-2500 Ion Chromatography systems (Guo et al., 2010), and to quantify the metal elements (Na, Mg, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Mo, Ag, Cd, Ba, Ti, Pb) with an ICP-MS (inductively coupled plasma mass spectrometry) 7500ce (Agilent, Japan). The analysis of the water-soluble ions was conducted with the following steps: 1) Each Teflon filter was extracted with 10 mL deionized water (Milli-Q Gradient, 18.2 MΩ·cm, USA) in an ultrasonic bath for 30 min; 2) the extraction solution was then filtered with a 0.45 µm PTFE filter (Gelman Sciences Inc., USA); 3) the anions (SO\(_4\)\(^2-\), NO\(_3\)\(^-\), Cl\(^-\), F\(^-\)) and cations (Na\(^+\), K\(^+\), NH\(_4\)+, Ca\(^2+\), Mg\(^2+\)) were analyzed with the Dionex ICS-2000 and Dionex ICS-2500 Ion Chromatography systems, respectively (Li et al., 2017). The detection limits for the water-soluble ions were 0.03 mg L\(^-1\), 0.10 mg L\(^-1\), 0.06 mg L\(^-1\), 0.05 mg L\(^-1\), 0.10 mg L\(^-1\), 0.01 mg L\(^-1\), 0.01 mg L\(^-1\), 0.03 mg L\(^-1\), and 0.03 mg L\(^-1\) in aqueous solution, specifically for Na\(^+\), K\(^+\), NH\(_4\)+, Ca\(^2+\), Mg\(^2+\), SO\(_4\)\(^2-\), NO\(_3\)\(^-\), Cl\(^-\), F\(^-\), respectively. The ICP-MS-based metal elements analysis including ultrasonic digestion (digest with 10 mL HNO\(_3\)-HCl solution at 200°C for 15 min) and electric plate digestion (digest with 10 mL HNO\(_3\)-HCl solution at 100°C for 2 h). The detection limits for the metal elements were 0.03 µg L\(^-1\), 0.007 µg L\(^-1\), 1 µg L\(^-1\), 0.05 µg L\(^-1\), 0.002 µg L\(^-1\), 0.02 µg L\(^-1\), 0.002 µg L\(^-1\), 0.4 µg L\(^-1\), 0.0009 µg L\(^-1\), 0.005 µg L\(^-1\), 0.003 µg L\(^-1\), 0.003 µg L\(^-1\), 0.006 µg L\(^-1\), 0.003 µg L\(^-1\), 0.003 µg L\(^-1\), 0.0002 µg L\(^-1\), 0.006 µg L\(^-1\), and 0.001 µg L\(^-1\) for Na, Mg, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Mo, Ag, Cd, Ba, Ti, and Pb, respectively. The quartz filters were analyzed for organic carbon (OC) and elemental carbon (EC) with a DRI thermal/optical reflectance (TOR) carbon analyzer, following the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance (TOR) protocol (Chow et al., 2007). The detection limits for EC and OC were 0.20 µgC cm\(^-2\) and 0.82 µgC cm\(^-2\). Field blanks were collected once or twice at every site in each season during the sampling period. Laboratory blanks were taken every 20 samples. Laboratory and field blanks were analyzed following the same procedures applied to the real samples. The final sample concentrations were calculated based on the differences in concentrations between the samples and the blanks.

Positive Matrix Factorization (PMF) Source Apportionment Model
A multivariate factor analysis tool, PMF, was used for source apportionment in this study. PMF is a weighted, non-negative least-squares variant of factor analysis that decomposes a matrix of assorted sample data into two-matrix factor contributions and factor profiles (Paatero and Tapper, 1994; Paatero, 1997). The newest version of the EPA PMF 5.0 develops new tools to evaluate the uncertainties in the extracted profiles (Paatero et al., 2014). The detailed description and algorithms of the PMF model can be found in previous studies (Song et al., 2006; Brines et al., 2016).

A total of 25 fitting species are used as input variables, including EC, OC, 6 water-soluble ions (SO\(_4\)\(^2-\), NO\(_3\)\(^-\), Cl\(^-\), Na\(^+\), NH\(_4\)+, K\(^+\)), and 17 metal elements (Mg, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Mo, Ag, Cd, Ba, Ti, Pb). Because of the better detection limits, elements Ca and Mg measured by ICP-MS were used as PMF inputs, rather than the ionic Ca\(^2+\) and Mg\(^2+\) measured with IC. F\(^-\) was excluded from the PMF inputs due to the high uncertainties associated with its concentrations. Data inputs included the concentrations and uncertainties. The uncertainties (UNC) were calculated (Polissar et al., 1998) by:

\[
\text{UNC} = \frac{5}{6} \times \text{MDL} \tag{1}
\]

and the concentrations are lower than the MDL (minimum detection limit of the instrument). In this case, the concentrations were replaced by half of the MDL. The uncertainties of missing data were calculated as the concentrations lower than the MDL:

\[
\text{UNC} = \sqrt{\text{Error Fraction} \times \text{Concentration}} + \text{MDL}^2 \tag{2}
\]

when the concentrations are greater than the MDL. The error fraction was 0.10 in this study.

The signal to noise ratios (S/N) were greater than 2.0 for all the 25 species, therefore they were defined to be “strong” (Paatero and Hopke, 2003).

RESULTS AND DISCUSSION

PM\(_{2.5}\) Mass Concentration and Chemical Composition
The temporal variations of PM\(_{2.5}\) concentrations at the five sites in four seasons is shown in Fig. 2. Similar pollution patterns occurred at the five sites during the sampling period. The average PM\(_{2.5}\) concentration was lower than the Chinese National Ambient Air Quality Standard (NAAQS) limit 75 µg m\(^{-3}\). Among the total 302 daily samples, 74.5% of the daily concentrations were lower than this limit.

Fig. 3 shows the chemical composition of PM\(_{2.5}\) concentration. The OC concentrations were multiplied by a conversion factor (the OM/OC ratio) to calculate the OM concentrations, by considering the unmeasured elements H, O, N, and S in the organic compounds (Turpin and Lim, 2002; Chow et al., 2002). In this study, the OM/OC ratio was 1.4 for winter and 1.6 for the other three seasons. The concentrations of salt spray were determined by summing Na\(^+\) and Cl\(^-\) ions (Rogula-Kozłowska et al., 2012). Crustal elements were estimated by assuming the oxidation state.
of mineral elements (Chow et al., 2015), and calculated by summing Ca concentration (multiplied by 1.4) and Fe concentration (multiplied by 1.4), considering the soil elemental composition in Ningbo. Trace elements were the sum of the metal elements measured, excluding the crustal species (Solomon et al., 1989; Maenhaut et al., 2002). Other constituents (labeled “others” in Fig. 3) comprised the remaining portion of the PM$_{2.5}$ mass, which were attributed to measurement errors and uncertainties resulting from the conversion factors.

The average concentration of PM$_{2.5}$ in Ningbo was $53.2 \pm 30.4 \mu g m^{-3}$, which was lower than the average concentrations measured for the period of January, May, and October 2010 ($70.97 \pm 47.40 \mu g m^{-3}$; Xu et al., 2015) and other

![Fig. 2. PM$_{2.5}$ concentrations variation at the five sites, by season.](image-url)
Fig. 3. Composition of PM$_{2.5}$ concentration measured during the sampling period, overall, by season, and by site.

The seasonal average PM$_{2.5}$ concentrations decreased in the order of 67.5 ± 32.2 µg m$^{-3}$ in winter, 62.4 ± 33.1 µg m$^{-3}$ in spring, 47.9 ± 16.7 µg m$^{-3}$ in autumn, and 26.8 ± 11.6 µg m$^{-3}$ in summer. OM was the dominant component in all the four seasons, accounting for 23% to 38% of PM$_{2.5}$ mass. OM comes from both primary sources, such as fossil fuel combustion and biomass burning, and from secondary transformation by photochemical reactions (Zhao et al., 2013). SO$_4^{2-}$ showed a relatively stable contribution to PM$_{2.5}$ mass in the four seasons, varying from 20% to 24%. SO$_4^{2-}$ was mostly from secondary formation and long-range transport. The formation of secondary sulfate might be dominated by the heterogeneous reactions resulting from high relative humidity in the coastal cities (Wang et al., 2006; Wu et al., 2017). NO$_3^-$ had an outstanding seasonal variation feature. It accounted for only 2% of PM$_{2.5}$ mass in summer, which was much lower than in other three seasons (12% to 18%). NH$_4^+$ also showed a low proportion in summer (8%), compared to other three seasons (11%–15%). Ammonium nitrate (NH$_4$NO$_3$) was formed of nitric acid (HNO$_3$) and ammonia (NH$_3$) in the aerosol by the reaction (NH$_3$(g) + HNO$_3$(g) ⇌ NH$_4$NO$_3$(s)) (Kuang et al., 2015; Zhao et al., 2015; Park et al., 2018), of which the gas-particle equilibrium was mainly controlled by the temperature. The average temperature of Ningbo during the sampling period was 7.1°C, 17.0°C, 31.4°C, and 18.5°C in winter, spring, summer, and autumn, while the RH (relative humidity) was 73.9%, 60.0%, 67.0%, and 70.3% in the four seasons. Higher temperature would help enhance the reverse equilibrium. Therefore, the formation of particulate NH$_4$NO$_3$ was favored at low temperature (Pathak et al., 2009), which might also explain the higher contribution of nitrates in the colder season (winter) and lower proportion in the hotter season (summer).

For the spatial variation, PM$_{2.5}$ concentration ranked in the order of urban residential sites (64.5 ± 33.4 µg m$^{-3}$ at SZ, 60.0 ± 28.8 µg m$^{-3}$ at CX) > urban coastal industrial sites (53.4 ± 31.8 µg m$^{-3}$ at ZH, 46.1 ± 27.5 µg m$^{-3}$ at BL) > suburban site (42.4 ± 25.4 µg m$^{-3}$). The inland sites had dense population as well as heavy traffic. On the other hand, the industrial sites were usually located near the coast and enjoyed better dispersion conditions. In contrast to both types of the urban sites, the suburban site did not have any primary source in the surrounding areas. However, the chemical composition of PM$_{2.5}$ in the five sites exhibited similar proportions for different species. OM took the highest proportion (25% to 32%), followed by sulfate (19% to 26%), nitrate (14% to 19%), and ammonium (13% to 15%). The secondary inorganic ions (SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$) accounted for half of the total PM$_{2.5}$ mass, indicating intensive secondary formation in Ningbo.
**PM$_{2.5}$ Source Apportionment by PMF Model**

The PMF model was used to evaluate the potential sources of PM$_{2.5}$ in Ningbo during the sampling period, based on the data set of 302 samples at the five sites. Six to ten factors were tested, the nine-factor solution was found to be optimum source profiles with a reasonable physical meaning. The uncertainties on estimated factor profiles and contributions were evaluated using the bootstrap method (Paatero et al., 2014). Rotations were explored by adjusting the FPEAK value, and the values of –1, –0.5, 0.5, 1, and 1.5 were tested in this study. All the five FPEAK values converged. The FPEAK value of –0.5, with the lowest increased Q values (% dQ (Robust) = 0.48), was chosen as the optimal solution.

The factor profiles with clear physical meanings are shown in Fig. 4. Factor 1 was dominated by EC, OM, Zn, Cu, and Pb. EC is often emitted from the engines, formed during the incomplete combustion in primary emission (Song et al., 2006; Liu et al., 2009). OM is an important constituent of the road dust (Chow et al., 2003). Cu mainly originates from the wearing of tires, brakes, and exhaust emission (Amato et al., 2009; Yang et al., 2016). Zn is normally emitted from the lubricant oil, used in tire manufacturing and brake linings (Zhou et al., 2004; Song et al., 2006; Zikovska et al., 2016; Du et al., 2017). Therefore, Factor 1 was distinguished as the vehicle exhaust. Factor 2 showed significant high proportion of Mg, followed by Ca and Na$^+$. Mg and Ca are associated with both natural soil-derived dust and anthropogenic construction dust (Xiu et al., 2004; Sun et al., 2005; Wang et al., 2006; Deshmukh et al., 2011; Zhang et al., 2013). Therefore, Factor 2 was identified as dust. Factor 3 was characterized as shipping emission with high loads of V and Ni, which can be attributed to fuel oil combustion from shipping emissions (Guo et al., 2009; Mooibroek et al., 2011; Kuang et al., 2015; Brines et al., 2016). Factor 4 was featured as high loading of NO$_3^-$, NH$_4^+$, and SO$_4^{2-}$, thus it represented secondary nitrate (Song et al., 2006). Factor 5 showed a high proportion of K$^+$, with some other species of metals. K$^+$ is recognized as the tracer of biomass burning (Andreae, 1998; Nolte et al., 2001; Hueglin et al., 2005; Yu et al., 2013; Zhang et al., 2013; Qin, 2015; Chuang et al., 2016). Therefore, Factor 5 was characterized as biomass burning. With high loading of SO$_4^{2-}$ and NH$_4^+$, Factor 6 was identified as secondary sulfate (Song et al., 2006; Kuang et al., 2015). Factor 7 was dominated by Na$^+$ and Ca, with contributions of some other metal species. Park et al. (2013) reported that high Na$^+$ and Ca$^{2+}$ but low Cl$^-$ indicated the aged sea salt. According to this, Factor 7 represented the aged sea salt contribution. Ti, Ag, Cd, Mo, Cu, Co, and Pb were the main featured species in Factor 8, which were related to glass processing, dye industry, metal industry, and smelting (Amato et al., 2009; Zhang et al., 2013; Chuang et al., 2016). Therefore, Factor 8 was identified as industrial emission. Factor 9 showed high loadings of Cl$^-$, with EC, OM, Cd, Pb, Zn, Mo, and Cu also contributing to this factor. Abundant Cl$^-$ in the atmosphere is primarily emitted from coal combustion (Song et al., 2006; Baumann et al., 2008; Tao et al., 2013; Yu et al., 2013), which was also an important source for Cd, Pb, and Zn (Mukai et al., 2001; Yang et al., 2016). Accordingly, Factor 9 was distinguished as coal combustion.

The contribution of each factor to the PM$_{2.5}$ mass is shown in Fig. 5. The nine factors, secondary nitrate, vehicle exhaust, secondary sulfate, coal combustion, industrial emission, ship emission, dust, biomass burning, and aged sea salt, had the average PM$_{2.5}$ contribution of 26%, 21%, 13%, 12%, 9%, 7%, 5%, 4%, and 3%, respectively. Secondary nitrate, vehicle exhaust, secondary sulfate, and coal combustion contributed the most to PM$_{2.5}$ pollution in Ningbo.

Secondary nitrate was one of the largest contributors in winter, spring, and autumn which disappeared in summer. High temperature in summer facilitated particulate nitrate to evaporate (May et al., 2014). The absence of secondary nitrate factor in summer made it quite different from other three seasons. Vehicle exhaust had a stable contribution (18% to 19%) in winter, spring, and autumn. Ship emission was reduced significantly in winter, relating to the slack season of this freight industry. Biomass burning showed a stable contribution (4%) in spring, summer, and autumn, while a little higher in winter (6%). Frequent biomass burning events in winter might be the cause (Chen et al., 2016). Secondary sulfate was lower in winter and summer. The aged sea salt was quite low in winter, spring, and autumn (2% to 3%). In contrast, it made a considerable contribution to PM$_{2.5}$ in summer (10%). According to the backward trajectory (Fig. 6) performed with the NOAA/ARL HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998), the air masses all originated from or passed through the marine area, carrying marine aerosols to Ningbo. Industrial emission was higher in summer and autumn, which may be associated to the busy season of industry, as reported by Ningbo EPB (Environmental Protection Bureau). Coal combustion was significant higher in winter than the other three seasons. Air masses originated in the north (Fig. 6) brought the emissions of coal combustion to Ningbo.

The source contributions showed distinguished spatial variations. Vehicle exhaust had higher contribution in the urban sites SZ and CX, which had busy traffic surroundings. Ship emission was higher in the coastal sites BL and ZH than other three sites, and significant high in BL (15%). There is a large port located in BL, leading to the busy business in freight industry. Secondary nitrate had the highest contribution in XS, which was a suburban site without point source nearby, and was more influenced by regional transport and secondary formation. Aged sea salt was higher in the coastal sites ZH (6%) and BL (4%) than the other three sites (1% to 2%). Coastal sites ZH and BL are more close to the sea and most likely influenced by the marine air masses. Industrial emission was also higher in ZH (9%) and BL (11%) than the other three sites (8%), linking to the intensive industries in these cities.

The source contributions of Ningbo showed features as a typical coastal city with dense population and developed industry in China, with the impact from secondary formation (nitrate and sulfate), vehicle emission, coal combustion, biomass burning, dust, industrial emission, sea salt, and...
Fig. 4. PMF sources profiles for PM$_{2.5}$ samples obtained during the sampling period.
Fig. 5. Source contributions to PM$_{2.5}$ during the sampling period, overall, by season, and by site.

Fig. 6. Backward trajectory clusters in four seasons. Four backward trajectories were calculated each day, with the starting times of 4:00, 8:00, 14:00, and 20:00 (local time), at 100m height, located at 29.88°E, 121.53°N. Numbers (1–4) identify individual air masses.
ship emission. The source profiles were quite different from the northern cities in China, which had higher contribution from dust and coal combustion (Song et al., 2006; Cao et al., 2012; Yang et al., 2016). However, Ningbo showed a similarity with Shanghai (Zhao et al., 2015; Qiao et al., 2016) and Nanjing (Li et al., 2016) with the contribution from sea salt and ship emission. Previous studies on the source apportionment in Ningbo showed a dominant contribution from dust and coal combustion, accounting for approximately 20% and 14% of the total PM$_{2.5}$ mass (Ye, 2011; Xiao et al., 2012). The transformation of energy structure promoted the atmospheric pollution types changing from solely coal-smoke pollution to the multi-pollutant complex pollution.

CONCLUSION

With the aim of investigating and assessing the PM$_{2.5}$ pollution characteristics on both temporal and spatial scales and identifying the potential sources of ambient PM$_{2.5}$ in Ningbo, 302 PM$_{2.5}$ samples from four seasons were collected at five scattered sites in Ningbo. The average concentration of PM$_{2.5}$ in Ningbo was 53.2 ± 30.4 µg m$^{-3}$. 25.5% of the daily PM$_{2.5}$ concentrations were found to exceed the NAAQS limit. OM and secondary inorganic ions (sulfate, nitrate, and ammonium) were the dominant pollutants in the PM$_{2.5}$. The PM$_{2.5}$ pollution level decreased in the following order: urban residential sites > urban coastal sites > suburban sites, with the highest level in winter and the lowest in summer.

Nine types of PM$_{2.5}$ sources were identified with the PMF model: (1) secondary nitrate, (2) vehicle exhaust, (3) secondary sulfate, (4) coal combustion, (5) industrial emission, (6) ship emission, (7) dust, (8) biomass burning, and (9) aged sea salt, with contributions of 26%, 21%, 13%, 12%, 9%, 7%, 5%, 4%, and 3%, respectively, over the entire sampling period. Secondary nitrate, vehicle exhaust, secondary sulfate, and coal combustion were the largest contributors on average to the PM$_{2.5}$ pollution in Ningbo. Secondary nitrate, vehicle exhaust, and coal combustion made the largest contributions in winter, whereas secondary nitrate, vehicle exhaust, and secondary sulfate were the main sources in spring. Vehicle exhaust was dominant in summer, while vehicle exhaust, secondary sulfate, secondary nitrate, industrial emission, and coal combustion accounted for a higher proportion in autumn. Besides the large contribution of secondary nitrate and secondary sulfate, the inland urban sites were impacted more by vehicle exhaust, while the coastal urban sites had larger contributions from ship emission and industrial emission.

The ambient PM$_{2.5}$ pollution in Ningbo is decreasing, indicating a cleaner atmospheric environment in the whole YRD region. However, efforts are still needed to further reduce the emissions. The large contribution of vehicle exhaust and coal combustion calls for more public attention and policy constraints. A preliminary analysis was made to distinguish the local and regional contributions to Ningbo based on the concentrations at the suburban receptor site XS. Ningbo’s PM$_{2.5}$ pollution was mainly contributed by regional transport, not local emission—a characteristic verified by the Models-3/CMAQ modeling system (Zheng et al., 2015), which demonstrated that regional transport contributed 55% to Ningbo’s PM$_{2.5}$ mass concentration. This result indicates that both reduction strategies targeting local primary sources (vehicle exhaust, coal combustion, industrial emission, ship emission, dust, and biomass burning), and joint inter-regional prevention and the control of air pollution in the YRD region and on an even larger scale are required.

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