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Development and Application of a New $PM_{2.5}$ Source Apportionment Approach

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ABSTRACT

Due to the similarity of $PM_{2.5}$ chemical species profiles of different sources, time synchronization of source contributions and the uncertainties of source-oriented models, it is difficult to get a well-separated and relatively accurate $PM_{2.5}$ source apportionment result, especially for the secondary components, when only one method was applied. A new $PM_{2.5}$ source apportionment approach, combining the receptor models, source-oriented models and emission inventory, was developed in this study. The proposed method had following strengths: (1) it could identify the source contributions to secondary components; (2) target (or expected) sources were optional; (3) mixed sources could be avoided. The new approach was then applied in two typical cities in North China – Beijing and Tangshan, based on intensive $PM_{2.5}$ observation results from 2011 to 2013. The source apportionment result indicated that the annual average contribution to $PM_{2.5}$ in Tangshan was 7.4%, 21.5%, 7.6%, 18.0%, 14.5%, 10.9% and 20.0% for power, metallurgy, cement, coal combustion, vehicle, dust and other sources, respectively; the annual average contribution ratio for vehicle, industry and industrial coal combustion, residential coal combustion, dust and other sources in Beijing was 31.5%, 22.9%, 10.6%, 14.5% and 20.4%, respectively. Seasonal variation of the source contributions was also analyzed. The demonstration results showed that the combined method was feasible. In addition, the detailed source contribution results could also provide scientific support for making effective $PM_{2.5}$ mitigation strategy.

Keywords: Source apportionment; Receptor model; WRF-CAMx-PSAT; Beijing; Tangshan.

INTRODUCTION

$PM_{2.5}$ (i.e., the fine particles with aerodynamic diameter ≤ 2.5 μm) is the most important atmospheric environment pollution issue in China (Xu *et al.*, 2013). According to the air quality report published by Ministry of Environmental Protection of the People's Republic of China (MEPPRC), the annual average (2013) atmospheric $PM_{2.5}$ concentration in 74 important cities was 72 μg m^{-3}, approximately 2 times of the national ambient air quality standard (Level-II, 35 μg m^{-3}) (MEPPRC, 2014). In one of the three most developed regions of China - Beijing-Tianjin-Hebei (BTH), the $PM_{2.5}$ pollution is more serious, the annual average $PM_{2.5}$ concentration was 106 μg m^{-3} in 2013. $PM_{2.5}$ with such high concentration could bring significant impact on human health, atmospheric visibility, climate change and economic development in China (Boldo *et al.*, 2006; Wang *et al.*, 2006; Ramanathan and Feng, 2009; Chen *et al.*, 2013). As a result, it is urgent and of great importance to identify the $PM_{2.5}$ contribution sources, in order to provide scientific support for developing effective pollution mitigation measures.

The most commonly used method for $PM_{2.5}$ source apportionment was receptor/mathematic models, such as Chemical Mass Balance (CMB), Positive Matrix Factorization (PMF) and Principal Component Analysis (PCA) (Song *et al.*, 2006; Gugamsetty *et al.*, 2012; Singh and Sharma, 2012; Ward *et al.*, 2012). Receptor models were generally different from dispersion models. They apportioned the contributions to sources based on the $PM_{2.5}$ components in atmosphere (and of sources for CMB), without considering the complex physical diffusion and chemical reactions in the air. They simplified the actual relationship between source emission and $PM_{2.5}$ concentration as linear mathematical formulas and as a result could get the source contributions easily. However, precisely because of the simplification, the receptor models could hardly identify detailed source contributions to the secondary particles (e.g., sulfate, nitrate and ammonium), which were formed through complex chemical reactions (Cheng *et al.*, 2013a). Even for the primary $PM_{2.5}$ components (e.g., elements and EC), it was difficult to get a feasible result by a single model, because of the similarity of chemical species profiles for some emission sources (e.g., soil dust and cement), and the time synchronization of contributions for different sources due to the meteorological conditions

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(Shi *et al.*, 2009; Zhang *et al.*, 2013).

Another commonly used approach for $PM_{2.5}$ source apportionment was source-oriented model simulation, such as CMAQ and CAMx (Huang *et al.*, 2010; Burr and Zhang, 2011; Zhang *et al.*, 2012). Because the models could simulate the physical diffusion and chemical reactions of pollutants in the atmosphere, they could identify the source contributions to both primary and secondary particles (Itahashi *et al.*, 2012). However, large uncertainties generally existed in the simulation results, especially for secondary particles, because (1) the source-oriented model simplified the complex physical diffusion and chemical reactions in the real world to a great extent; (2) the emission inventory used by the air quality model had significant uncertainty due to the variety and complexity of emission sources (Zhou *et al.*, 2015); (3) the meteorological data required by the source-oriented model was obtained by meteorological model simulation, which could also bring uncertainties to the simulated air pollutant concentrations (Cheng *et al.*, 2012).

It could be found that both of the receptor and the numerical models had their strengths and weaknesses. None of them could identify the source contributions to $PM_{2.5}$ accurately. In order to solve this problem, we proposed a method combing factor analysis and MM5-CMAQ to estimate the contribution from motor vehicles to $PM_{2.5}$ in Beijing (Cheng *et al.*, 2013a), in which the MM5-CAMQ was used to calculate the contribution to sulfate, nitrate and ammonia in $PM_{2.5}$. Similar method was then used in the $PM_{2.5}$ source apportionment in Guangzhou (Cui *et al.*, 2015). In this study, a more refined approach, which combined receptor models, emission inventory and numerical simulation, was developed to identify the contribution sources for both the primary and secondary components of $PM_{2.5}$. In addition, the developed new method was then applied in the $PM_{2.5}$ source apportionment of Beijing and Tangshan, where the $PM_{2.5}$ pollution were typical in China. The main contribution sources of $PM_{2.5}$ were identified by the proposed method based on intensive monitoring results from 2011 to 2013.

METHODS

$PM_{2.5}$ Source Apportionment Method

The contributions of emission sources to the atmospheric $PM_{2.5}$ could be calculated by Eq. (1):

$$CR_{PM_{2.5},i} = CR_{primary,i} + CR_{secondary,i} \quad (1)$$

where $CR_{PM_{2.5},i}$, $CR_{primary,i}$ and $CR_{secondary,i}$ was the contribution ratio of source i to the total, primary and secondary atmospheric $PM_{2.5}$, respectively.

Primary components source apportionment. The contribution ratios of emission sources to the primary components of $PM_{2.5}$ could be calculated by Eq. (2):

$$CR_{primary,i} = CR_{p,i} \times P_{primary} \quad (2)$$

where $CR_{p,i}$ was the contribution ratio of source i to primary $PM_{2.5}$ components (take primary components as 100%); $P_{primary}$ was the proportions of primary components in $PM_{2.5}$. $P_{primary}$ was obtained based on $PM_{2.5}$ sampling and analysis. The source contribution ratios to primary $PM_{2.5}$ components were investigated using the combined PCA/PMF-CMB-EI (EI: emission inventory) approach through three steps: (1) the PMF or PCA model was firstly used to identify the contribution factors (sources), however, they might fail to separate out some factors (mixed sources) because of the similarity of $PM_{2.5}$ chemical species profiles for different sources and the time synchronization of contributions from various sources due to the meteorological conditions; (2) in that case, the CMB model was applied to analyze the mixed factors (sources) using the output of PMF and the separate chemical profiles of the mixed sources as inputs (Shi *et al.*, 2009); (3) after using the CMB model, mixed sources might still exist or even negative contribution could be got due to the $PM_{2.5}$ species profiles' co-linearity among some sources, under this circumstances, the mixed sources were further separated based on their primary $PM_{2.5}$ emission weight. Alternatively, the mix source contributions generated by PMF/PCA (stage 1) could be directly separated based on primary $PM_{2.5}$ emissions weight (stage 3), if the local source $PM_{2.5}$ species profiles could not be obtained. In this study, the required source chemical profiles were obtained from local measurements (Wen, 2015). The emission inventory of the demonstration regions - Beijing and Tangshan could be found in Zhou (2012).

Secondary components source apportionment. It is assumed that the source contributions to the secondary aerosols (e.g., nitrate) were proportional to those to the corresponding precursors (e.g., NO_x) in the atmosphere (Cheng *et al.*, 2013a). As a result, the contribution ratios of emission sources to the secondary components of $PM_{2.5}$ could be alternatively estimated by simulating the contribution to the precursors. The detailed calculation formula was:

$$CR_{secondary,i} = CR_{SO_2,i} \times P_{SO_4^{2-},i} \times CR_{NO_x,i} \times P_{NO_3^-,i} \times CR_{NH_3,i} \times P_{NH_4^+,i} + CR_{VOCs,i} \times P_{SOA,i} \quad (3)$$

where $CR_{SO_2,i}$, $CR_{NO_x,i}$, $CR_{NH_3,i}$ and $CR_{VOCs,i}$ was the contribution ratio of source i to atmospheric SO_2, NO_x, NH_3 and VOCs, respectively; $P_{SO_4^{2-}}$, $P_{NO_3^-}$, $P_{NH_4^+}$ and P_{SOA} was the proportions of SO_4^{2-}, NO_3^-, NH_4^+ and secondary organic aerosol (SOA) in $PM_{2.5}$, respectively. The source contributions to SO_2, NO_x, NH_3 and VOCs could be calculated by air quality model simulation. Alternatively, the source contributions to VOCs could also be obtained by receptor models, such as CMB or PMF (Liu *et al.*, 2005; Song *et al.*, 2007; Song *et al.*, 2008; Yuan *et al.*, 2009). The contribution values obtained by this new method could reflect the impacts of regional transport. For example, the contribution ratio estimated from vehicles to the $PM_{2.5}$ in target area (e.g., Beijing) was from both local and surrounding areas.

The straightforward diagram of the developed $PM_{2.5}$ source apportionment approach could be found in Fig. 1.

$PM_{2.5}$ Components in Beijing and Tangshan

Two cities – Beijing and Tangshan in the BTH region were selected as the demonstration area in this study. The

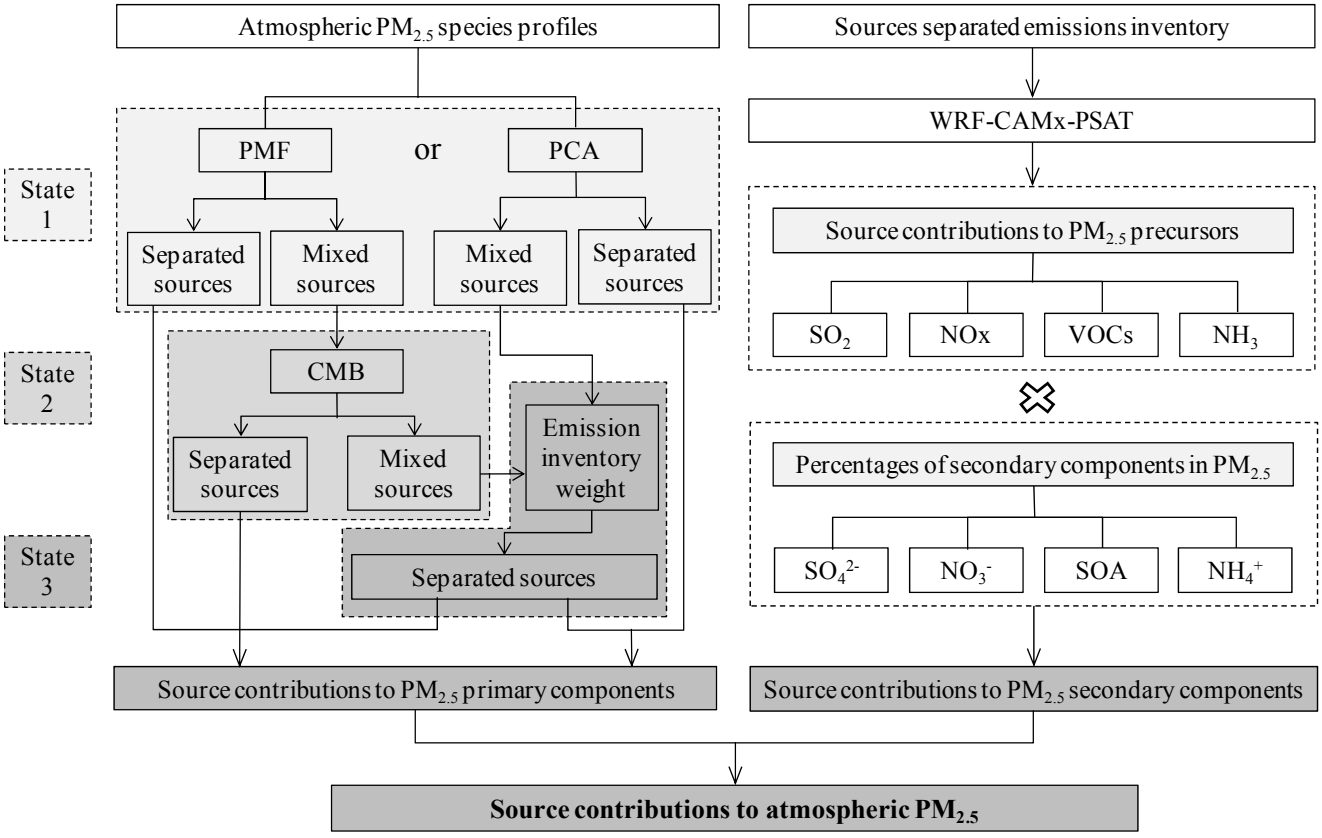

Fig. 1. Schematic of the $PM_{2.5}$ source apportionment approach.

$PM_{2.5}$ sampling was conducted in typical months of different seasons (i.e., spring, summer, autumn and winter) from 2011 to 2013. The sampling sites could be found in Fig. 2. Table 1 listed the atmospheric $PM_{2.5}$ components in Beijing and Tangshan. Details about $PM_{2.5}$ sampling and chemical components analysis could be found in the supplementary material file.

Source-Oriented Model Simulation

The coupled WRF-CAMx-PSAT model was used to simulate the source contributions to the secondary $PM_{2.5}$ components precursors (SO_2, NO_x, NH_3 and VOCs) for Beijing and Tangshan. The performance of the coupled model was approved as acceptable for simulating the air quality in Beijing and Tangshan (supplementary material). The meteorological model WRF was employed to provide meteorological field data required by CAMx; the air quality model CAMx was run to simulate the diffusion and reactions of air pollutants; while the PSAT was applied to identify the source contributions to target pollutants.

A three-level nested-grid architecture was designed for the implementation of WRF-CAMx-PSAT (as shown in Fig. 2). The spatial resolution of modelling domain 1, domain 2 and domain 3 was 27 km × 27 km, 9 km × 9 km and 3 km × 3 km, respectively. Domain 3 covered Beijing and Tangshan and their surrounding areas. The receptors were designed as the grids in the PSAT where the sampling sites located. Vertically, 35 sigma levels were designed in the WRF simulation. The WRF outputs were collapsed into 13 layers with format required by CAMx through WRFCAMx (version 4.0). The emission inventories in Beijing-Tianjin-Hebei region required by CAMx were obtained from Zhou (2012), while those in other regions covered by the modelling domain were obtained from the Multi-resolution Emission Inventory for China developed by Tsinghua University (http://www.meicmodel.org). More detailed description of WRF and CAMx could be found in our previous papers (Huang *et al.*, 2010; Cheng *et al.*, 2012; Li *et al.*, 2013).

CASE STUDY AREAS

Beijing (115.7°E–117.4°E, 39.4°N–41.6°N) is the capital, and also the politics, culture, science and education center of China, covering a territorial area of 16.4 thousand km^2. Tangshan (117.5°E–119.3°E, 38.9°N–40.5°N) is a typical industry city of China, covering a territorial area of 13.5 thousand km^2. The population of Beijing and Tangshan in 2014 was 21.5 and 7.6 million, respectively. The highest wind speed of Beijing and Tangshan occurred in spring and the highest temperature occurred in summer. The Gross Domestic Product (GDP) of Beijing and Tangshan in 2013 was 2133 and 622 billion Yuan, respectively. Service sector was the dominant contributor to the GDP in Beijing, with percentage of 76.9%. Industry especially the heavy industry, such as metallurgy and cement, was the main contributor to the GDP in Tangshan, with percentage of 55.3%. The $PM_{2.5}$ pollution problem in Beijing and Tangshan was

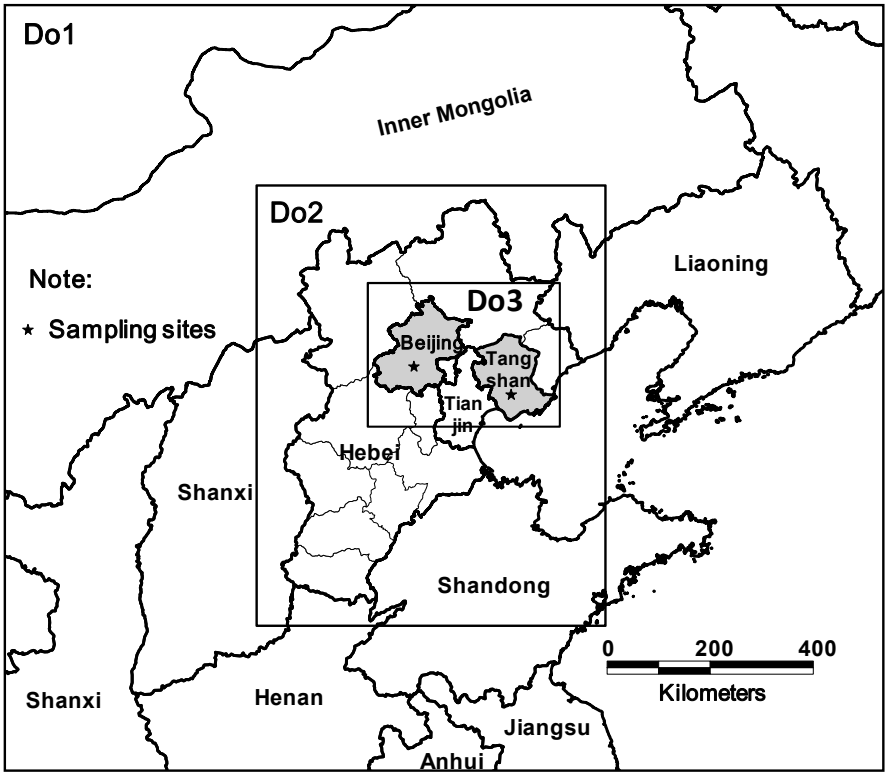

Fig. 2. Location of case study areas, $PM_{2.5}$ sampling sites and model simulation domains.

Table 1. Proportions of $PM_{2.5}$ components in different seasons of Beijing and Tangshan (%).

City	Season	POA	EC	Elements	SOA	SO_4^{2-}	NO_3^-	NH_4^+	Unidentified
Beijing	Spring	10.4	3.9	24.1	7.2	8.4	11.8	7.4	26.8
	Summer	10.5	5.1	16.0	11.5	15.2	15.7	11.8	14.2
	Autumn	14.6	4.7	18.1	9.8	12.1	18.1	10.6	12.0
	Winter	15.6	4.0	15.2	11.2	15.3	15.1	10.2	13.4
Tangshan	Spring	16.9	6.4	28.1	7.4	7.1	8.8	4.8	20.6
	Summer	10.8	5.5	15.7	9.8	20.8	11.3	11.7	14.4
	Autumn	13.3	7.0	21.7	14.3	8.4	10.4	8.0	16.9
	Winter	13.6	4.0	13.4	12.6	15.0	8.5	8.0	24.9

serious. The annual average $PM_{2.5}$ concentration in 2013 was 90 μg m^{-3} and 114 μg m^{-3}, about 2.57 and 3.26 times of the national class II standard limit (i.e., 35 μg m^{-3}) in Beijing and Tangshan, respectively.

RESULTS AND DISCUSSION

$PM_{2.5}$ Source Apportionment in Tangshan

Sources contribution to primary components. At stage 1, the PMF model was applied to the source apportionment of primary $PM_{2.5}$ components in Tangshan. All chosen parameters were retained in order to fulfill the conditions of minimizing model uncertainty. The sampling and analysis data of $PM_{2.5}$ in four seasons were separately selected as the PMF inputs in order to obtain the seasonal source contributions. More details about uncertainty, optimum value and choosing of feasible number referred to Reff *et al.* (2007). Table 2 lists the output results of PMF for different seasons. In spring, factor 1 accounted for 17.8% of the primary $PM_{2.5}$ concentration. The main species included Zn, Cd, Pb, As, Na and Cu. These species were typically associated with metallurgy industry. As a result, factor 1 could be identified as metallurgy sources. Factor 2 contributed 31.6% of the primary $PM_{2.5}$ concentration. The dominant species were S and As, which were typical elements for coal combustion, consequently, this factor could be identified as a mix source of power (the fuel for most of the power plants was coal) and coal combustion. The contribution ratio of factor 3 was 33.9%. The main species in this factor were Ti, Ca, Al, Na and Mg, in which Ti and Al were the typical elements of dust, Ca and Mg were the typical elements of cement production. As a result, this factor may be attributed to a mix source of dust and cement. The contribution ratio of factor 4 was 16.7%. The main species were Ni, Cr and Pb, which were the typical elements of motor vehicles, as a result, this factor could be identified as vehicle source. Based on the same method, the contribution sources could be identified as mix source

Table 2. PMF output results in different seasons of Tangshan.

Season	Factor	Main typical species	Contributions
Spring	Factor 1	Zn, Cd, Pb, As, Na, Cu	17.8%
	Factor 2	S, As	31.6%
	Factor 3	Ti, Ca, Al, Na, Mg	33.9%
	Factor 4	Ni, Cr, Pb	16.7%
Summer	Factor 1	Ni, Al, Na, As	31.6%
	Factor 2	Zn, Cd, Pb, Mn, Fe	22.3%
	Factor 3	S, Ti, Pb, Cu	23.1%
	Factor 4	Mg, Ca, Ti, Fe, Al	23.0%
Autumn	Factor 1	Ni	15.2%
	Factor 2	Ca, Pb, Mg, Cd	50.7%
	Factor 3	Zn, As, Pb, Cd	11.8%
	Factor 4	S, Mn, Cu, Na	22.3%
Winter	Factor 1	Ca, Mg, Al, Ni	29.1%
	Factor 2	S, As, Al	31.0%
	Factor 3	Zn, Cd, Pb, Mn	17.2%
	Factor 4	Ti, Al, S, Cu, Na	22.5%

of vehicle and dust, metallurgy, mix source of power and coal combustion, mix source of cement and other in summer; vehicle, mix source of dust, cement and other, metallurgy, and mix source of power and coal combustion in autumn; mix source of vehicle and cement, mix source of power and coal combustion, metallurgy, mix source of dust and other in winter, respectively. The corresponding contributions could be found in Table 2.

At stage 2, the CMB model was used to separate the mix sources generated in stage 1. The potential source profiles which contributed to the mix sources obtained from PMF output were provided as input for CMB. The CMB output result was accepted only when the statistics' results (e.g., chi-square value) were lower than the limits. The details could be found in Shi *et al.* (2009). At this stage, the contribution of the mix sources of power and coal combustion in spring and the mix sources of vehicle and dust in summer could be separated by CMB. The required source $PM_{2.5}$ profiles of the above four sources were obtained by field test in Tangshan (Wen, 2015). The contribution to the primary $PM_{2.5}$ components of power and coal combustion in spring was 8.5% and 21.7%, respectively, while that of vehicle and dust in summer was 14.5% and 17.1%, respectively.

At stage 3, the contribution of the mix sources could not be separated by CMB was further identified based on the $PM_{2.5}$ emission weight. For example, the mixed sources including power and coal combustion in summer could not be separated by CMB, however, based on the primary $PM_{2.5}$ emission inventory, the percentage of power and coal combustion in their total emission was 37% and 53%; then the total contribution ratio of the mix sources (23.1%) was further separated as 8.5% and 14.6% for power and coal combustion, respectively. The mix source contributions in other seasons were also separated using the above method. The detailed contribution for various sources in different seasons estimated based on stage 1, stage 2 and stage 3 was listed in Fig. 3.

Sources contribution to secondary component precursors. Based on the WRF-CAMx-PSAT simulation, the seasonal source contributions to the precursors (SO_2, NO_x, NH_3 and VOCs) of secondary $PM_{2.5}$ in Tangshan were calculated. Fig. 4 showed that metallurgy and coal combustion were the main contributors to the atmospheric SO_2 in Tangshan. The annual average contribution ratio was 37.1% and 26.5%, respectively. Metallurgy and vehicle were the main contributors to the atmospheric NO_X, with annual average contribution ratio of 25.9% and 31.2%, respectively. The main VOCs contribution sources were complex, including metallurgy, coal combustion, vehicle and others (such as biogenic sources, biomass and solvent use), which accounted for 27.4%, 26.3%, 16.2% and 23.3% of the atmospheric VOCs on an annual average, respectively. As for NH_3, other sources, mainly farmland, livestock and human excrement, were the dominant contributors. They accounted for more than 90% of the atmospheric NH_3 in Tangshan. Among all the sources, coal combustion had obvious seasonal variation because of the residential heating in winter. Its contribution ratio in winter was 38.3%, 15.7% and 39.5% for SO_2, NO_x and VOCs, respectively, about 1.6–1.7, 1.9–2.2 and 2.2–3.4 times of that in other seasons, respectively.

$PM_{2.5}$ source apportionment result. Taking the total percentage of primary and secondary components as 100%, the source contribution (Fig. 5) to the atmospheric $PM_{2.5}$ of Tangshan was calculated based on formulas (1)–(3). The annual average contribution to $PM_{2.5}$ in Tangshan was 7.4%, 21.5%, 7.6%, 18.0%, 14.5%, 10.9% and 20.0% for power, metallurgy, cement, coal combustion, vehicle, dust and other sources (such as biomass, solvent use, biogenic, farmland and livestock sources), respectively. It can be found that metallurgy had the highest contribution ratio. This was mainly caused by the high steel output. The steel output in Tangshan in 2011 was 120 million ton, accounting for about 50.0% of that in Hebei province and 14.6% of that in the whole China (NBSC, 2012; TSB, 2013). Tangshan had a high coal consumption, and as a result the contribution of coal combustion to the atmospheric $PM_{2.5}$ was also high. Vehicle was the third highest contribution source. The vehicle population had reached more than 1.6 million by

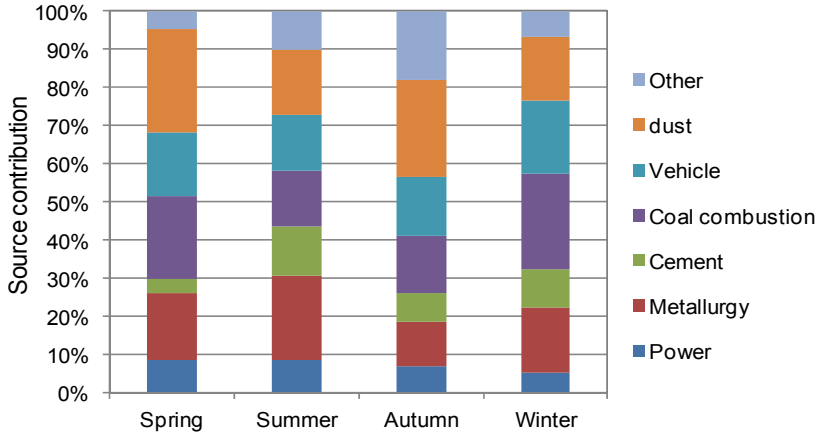

Fig. 3. Source contributions to primary $PM_{2.5}$ components in Tangshan.

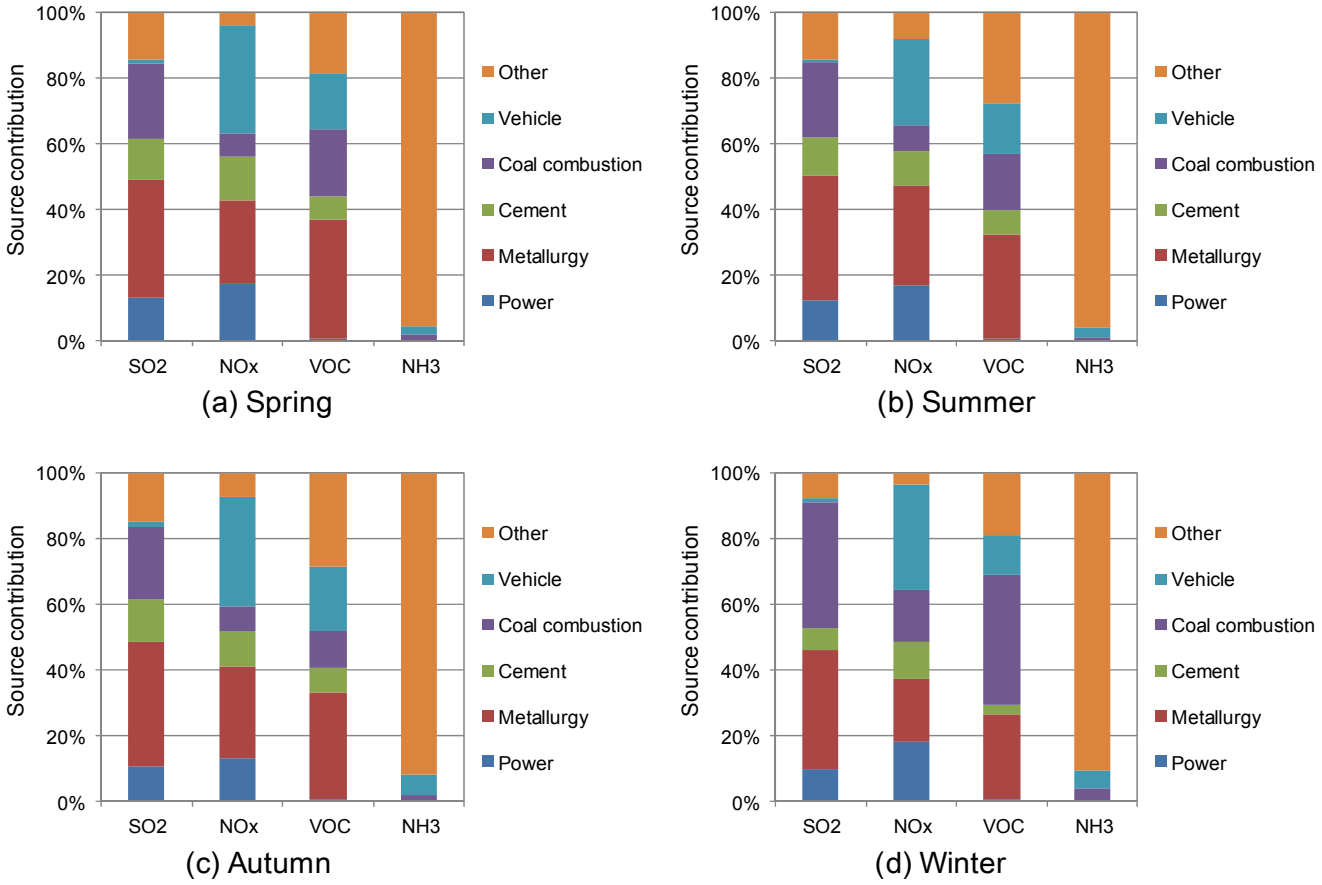

Fig. 4. Seasonal source contributions to precursors of secondary $PM_{2.5}$ in Tangshan.

the end of 2012 (TSB, 2013). Fig. 5 illustrated the seasonal variation of the source contributions to the atmospheric $PM_{2.5}$ of Tangshan. Dust had the highest contribution ratio in spring, accounting for approximately 17.5% of the total $PM_{2.5}$. This ratio was 1.4–2.7 times of that in other seasons. Similar with the explanation in section 3.1, large amount of dust emission caused by the higher wind speed resulted in the larger contribution. The contribution of coal combustion also had obvious seasonal variation, which could be attributed to the residential heating in winter. The contribution ratio was approximately 26.8% in winter, about 1.4–2.0 times of that in other seasons. The source apportionment results could provide scientific support for the $PM_{2.5}$ pollution control strategy making.

$PM_{2.5}$ Source Apportionment in Beijing

Sources contribution to primary components. At stage 1, the PCA model was used to apportion the source contributions to primary $PM_{2.5}$ components in Beijing. The PCA output results for different seasons of Beijing were listed in Table 3.

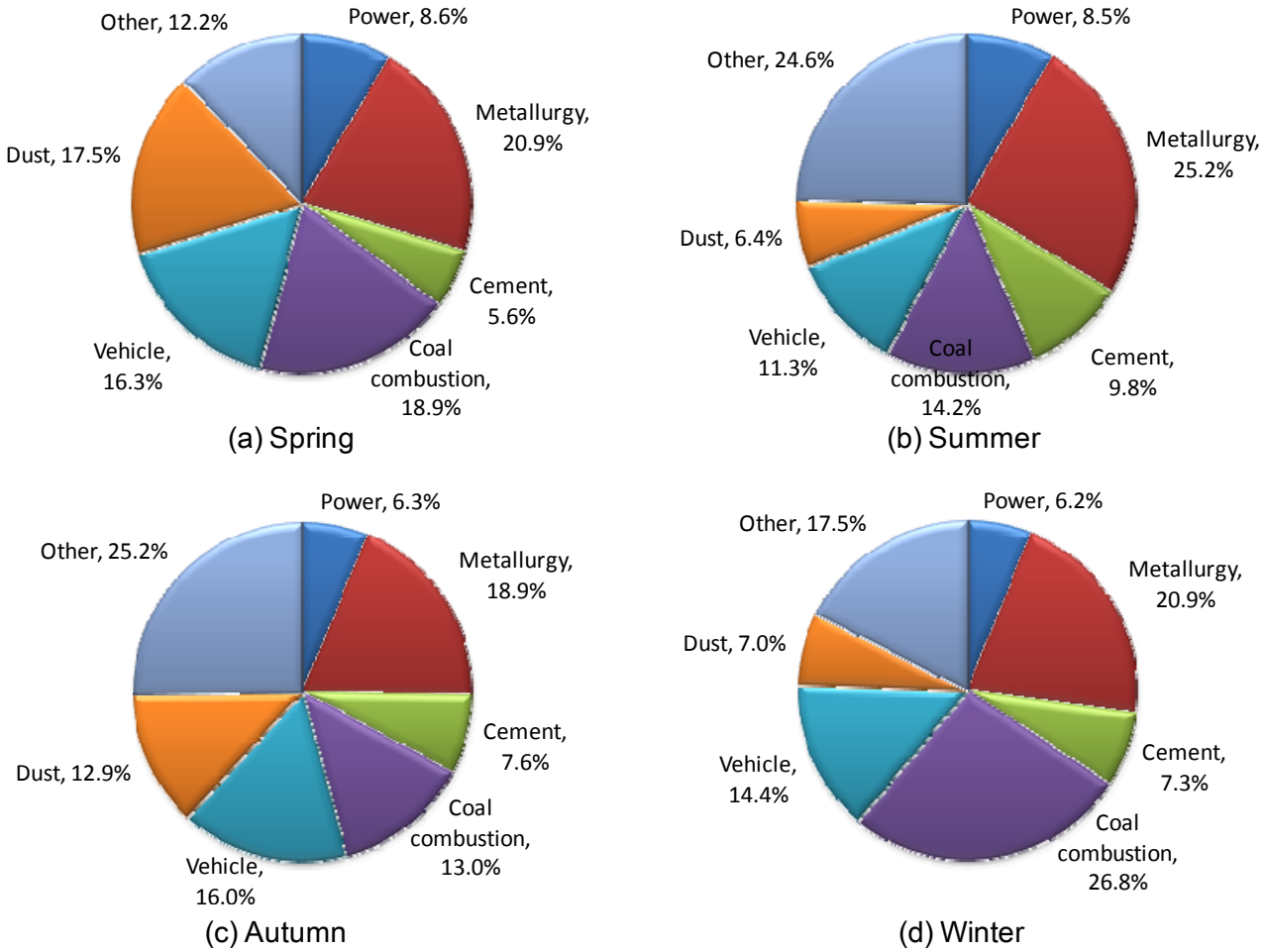

Fig. 5. Source contributions to $PM_{2.5}$ in Tangshan.

Table 3. PCA output results in different seasons of Beijing.

Season	Factor	Main species	Contributions
Spring	Factor 1	Mg, Al, Ca, Ti, Cu, Mn	36.6%
	Factor 2	Co, Ni, Zn	18.8%
	Factor 3	S, As, Cd, Pb	16.1%
Summer	Factor 1	Ni, Co, Fe	37.6%
	Factor 2	Mg, Al, Na, Na, Ti, Ca	34.2%
	Factor 3	S, V, Pb, Cu	15.2%
Autumn	Factor 1	Na, Mg, Ca, Ti, Fe, Al	36.1%
	Factor 2	Co, Ni, Cd	32.7%
	Factor 3	S, Cu, Zn, As, Pb, Mn	19.9%
Winter	Factor 1	S, Zn, As, Cd, Pb, Mn	39.8%
	Factor 2	Mg, Al, Ti, Ca, Fe	30.0%
	Factor 3	Cr, Co, Ni	19.9%

As shown in Table 3, in spring, factor 1 accounted for 36.6% of the total variance. It had high loadings for Mg, Al, Ca, Ti, Cu and Mn. These elements were typical associated with dust sources, such as road dust and resuspended dust. Factor 2 was high loaded with Co, Ni and Zn, which could be attributed to motor vehicles. As a result, factor 2 could be identified as vehicle source. The contribution to the primary $PM_{2.5}$ was 18.8%. Factor 3 contributed 16.1% of the total variance. It had high loadings for S, As, Cd and Pb, which could be attributed to coal combustion and industry plants in Beijing. As a result, factor 3 could be identified as a mix source of coal combustion and industry. Based on the same method, the contribution sources could be identified as vehicle, dust, mix source of coal combustion and industry in summer; dust, vehicle, mix source of coal combustion and industry in autumn; mix source of coal combustion and industry, dust and vehicle in winter, respectively. The corresponding source contributions to

primary $PM_{2.5}$ in Beijing were listed in Table 3. It could be found that there was the same mix source - coal combustion and industry in different seasons. The contributions other than the identified three factors were attributed to other sources, with percentages of 28.5%, 13.0%, 11.3% and 10.3% in spring, summer, autumn and winter, respectively.

At stage 2, because the local source emission profile could not be obtained, the mix source of coal combustion and industry was separated based on their primary $PM_{2.5}$ emission weight, rather than CMB. Because the residential coal combustion was an important contribution source in Beijing, the mix source was further separated as two sources: (1) residential coal combustion (RCC) and (2) industry and industrial coal combustion (ICC). The industry and ICC contributed approximately 54%, 91%, 56% and 55% of the total primary $PM_{2.5}$ emissions of (1) and (2) in spring, summer, autumn and winter, respectively. As a result, based on the mix source contributions obtained by PCA, the source contribution of industry and ICC to primary components was 8.7%, 13.8%, 11.1% and 21.9% in spring, summer, autumn and winter, respectively, while the corresponding contribution of RCC was 7.4%, 1.4%, 8.8% and 17.9%, respectively.

Sources contribution to secondary component precursors. Fig. 6 showed the source contributions to the precursors (SO_2, NO_x, NH_3 and VOCs) of secondary $PM_{2.5}$ components in different seasons of Beijing. The contributions of SO_2, NO_x and NH_3 were simulated by the integrated WRF-CAMx-PSAT model, while those of VOCs were obtained from the VOCs source apportionment results in previous studies (Huang *et al.*, 2010; Cheng *et al.*, 2012; Li *et al.*, 2013). It could be found that industry and ICC, RCC were the main contributor to atmospheric SO_2. The annual average contribution ratio was 60.0% and 34.3%, respectively. Vehicle, industry and ICC had the largest contribution to NO_x, with annual average percentages of 64.1% and 26.6%, respectively. Other sources, including farmland, livestock and human excrement, were the dominant contributor to the atmospheric NH_3. The annual average contribution ratio was approximately 87.3%. The contribution of vehicle to NH_3 was 12.5%. The annual average contributions of vehicle, industry and ICC, other sources (e.g., biogenic source, solvent use and biomass burning) were 56.6%, 26.5% and 10.4%, respectively. Seasonal variation analysis indicated that industry and ICC had significant high contribution to SO_2 in summer (81.9%). This may be caused by the reasons that (1) the emission of power plants was much higher in summer due to the high electric demand for cooling; (2) the emission of other sources (e.g., residential coal combustion) except for industry and ICC was much lower in summer. The seasonal variation of the source contributions to NO_x, NH_3 and VOCs was not so obvious.

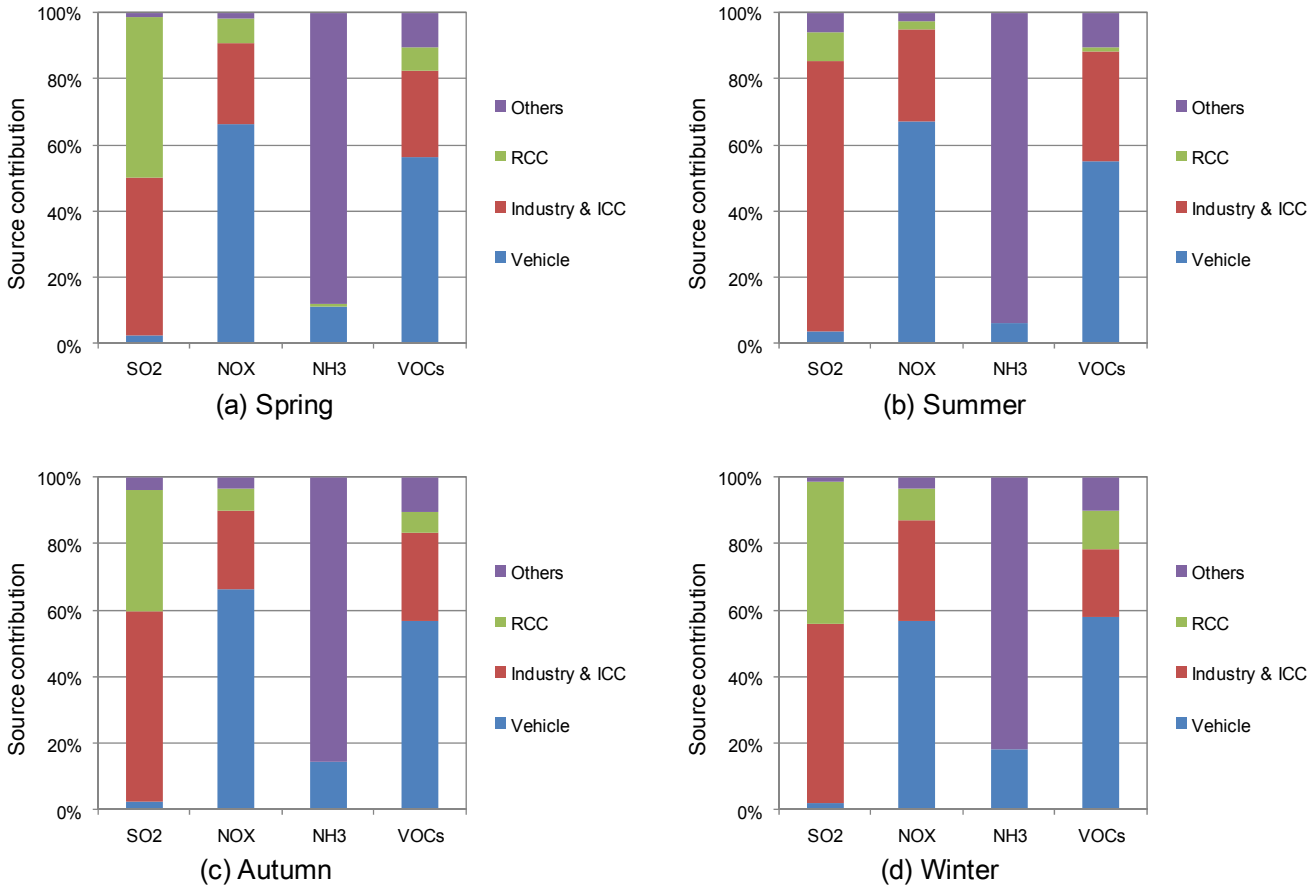

Fig. 6. Seasonal source contributions to precursors of secondary $PM_{2.5}$ in Beijing (ICC: industrial coal combustion; RCC: residential coal combustion).

$PM_{2.5}$ source apportionment result. By taking the total percentage of primary and secondary components as 100%, the $PM_{2.5}$ concentration was apportioned to different sources by formula (1)–(3), based on the observed $PM_{2.5}$ components results and the estimated source contributions to primary and secondary components. The annual average contribution ratio for vehicle, industry and ICC, RCC, dust, and other sources (e.g., biomass burning, biogenic source, farmland, livestock and solvent use) was 31.5%, 22.9%, 10.6%, 14.5%, and 20.4%, respectively. Unlike Tangshan, the source with the highest contribution ratio was vehicle, rather than industry sources. The reasons for this phenomenon may be explained as follows: (1) the vehicle population has reached more than 5.4 million by the end of 2013, resulting in higher vehicular emissions; (2) there were much less industrial plants in Beijing than in Tangshan, especially the enterprises with high pollutants emissions, such as iron and steel, coking and cement plants. Industry and ICC had the second largest contribution (22.9%), although its percentage was lower than that in Tangshan (36.6%, sum contribution of power, metallurgy and cement). In addition, the contribution of residential coal combustion (10.6%) was also identified as a separate source, considering the large population (more than 21 million by the end of 2013) and the relative high RCC emission. The seasonal variation was shown in Fig. 7. Dust had higher contribution (17.6%) in spring than in other seasons. This was similar with Tangshan. The highest contribution ratio (17.0%) of residential coal combustion (RCC) occurred in winter due to the residential heating. The contribution of vehicle was higher in summer (35.4%) and autumn (34.6%) with high temperature and solar radiation intensity, which could facilitate the transformation from vehicular NO_x and VOCs to nitrate and SOA. In addition, the industry and ICC had highest contribution in summer (28.7%). In summer, the predominant wind direction was southwest and south. Such wind could bring the high industry and ICC emissions from Hebei and Shandong to Beijing.

DISCUSSION

The results were also compared with previous studies. Take the dust (only contributed to the primary $PM_{2.5}$) in Beijing as example, the contribution ratio (14.5%) obtained by the proposed approach has no obvious difference with that in Zhang *et al.* (2013) (15.0%) and Zheng *et al.*'s (2005) (19.7%) study. However, because the developed method could identify the precursors' contribution (e.g., SO_2 to sulfate, NO_x to nitrate) to the secondary $PM_{2.5}$, the contribution from motor vehicle obtained in this study (31.5%) was much higher than that in previous studies (4%–11.5%) (Zheng *et al.*, 2005; Song *et al.*, 2006; Zhang *et al.*, 2013).

Based on the application in Beijing and Tangshan, it could be found that the proposed combined $PM_{2.5}$ source apportionment method had the following strengths: (1) source contributions to the secondary $PM_{2.5}$ components

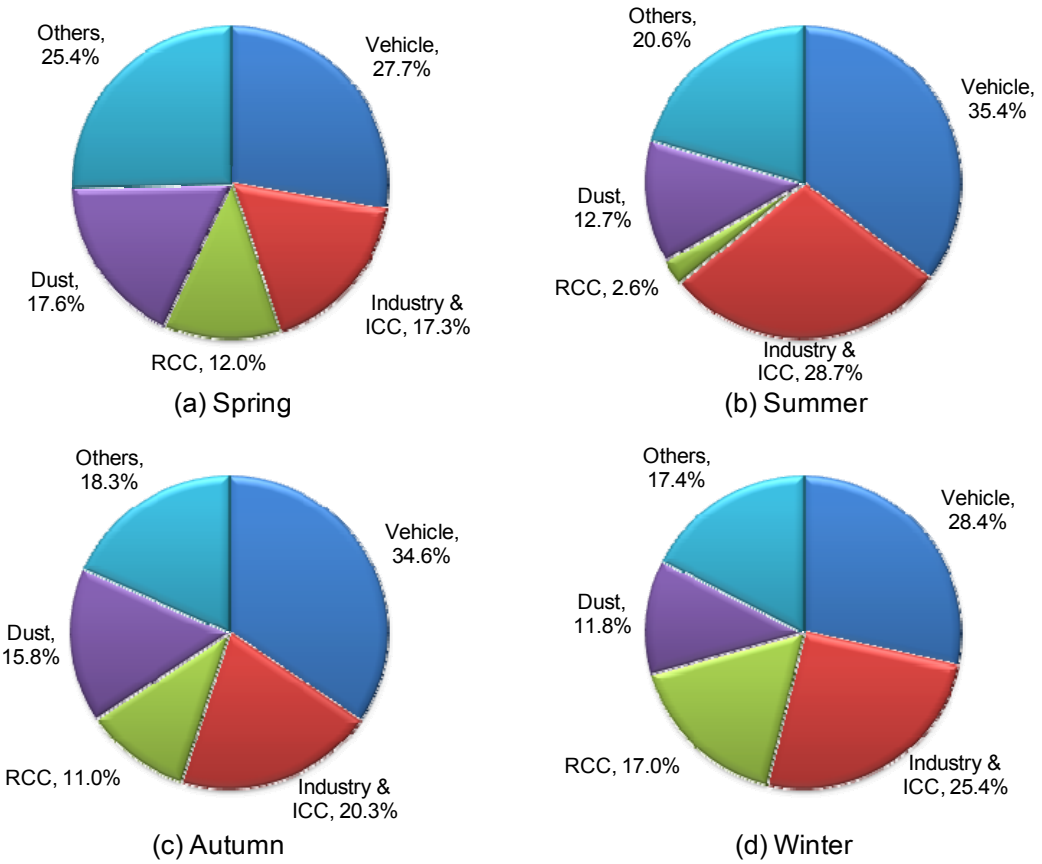

Fig. 7. Source contributions to $PM_{2.5}$ in Beijing (ICC: industrial coal combustion; RCC: residential coal combustion).

could be identified; (2) target (or expected) sources were optional and mixed sources could be avoided because the inventory and source-oriented air quality model were introduced. As compared with the method in Cheng *et al.* (2013), (1) the PCA-EI or PMF-CMB-EI method was developed to estimate the source contribution to primary $PM_{2.5}$; (2) the on-line source apportionment technology (PSAT) replaced the zero-out method to identify the source contribution to the $PM_{2.5}$ precursors (e.g., SO_2 and NO_x), this could eliminate the nonlinear influence on the contribution result when using the zero-out method. However, the proposed $PM_{2.5}$ source apportionment approach also has its disadvantages. Firstly, the transport and formation of $PM_{2.5}$ was complex courses. The proposed approach simplifies these courses and consequently could bring uncertainties to the $PM_{2.5}$ source apportionment results. Secondly, in order to obtain comprehensive source apportionment result, several methods (e.g., receptor model and numerical simulation) were used. Every method could bring uncertainty to the final results. It was quite difficult to assess these uncertainties; however, this could be further investigated in following studies. Thirdly, some primary sulfate, nitrate and ammonium exist in the primary source emissions (Watson *et al.*, 2001). However, in this paper, all of these components were assumed to be secondary aerosols. This will result in differences between the real contribution and the estimation result. But the error is small (Zhang *et al*., 2012). In addition, a detailed source specific emission inventory was also needed. This made the application of the proposed method was more complex than the existed methods, such as PMF and PCA.

CONCLUSIONS

In this study, a new approach combining receptor models, source-oriented air quality models and emission inventory was developed to identify the $PM_{2.5}$ source contributions. The PMF/PCA-CMB-EI (emission inventory) was used for the primary aerosols source apportionment. The source-oriented model WRF-CAMx-PSAT was used for simulating the source contributions to the secondary components precursors. Then the source contributions to $PM_{2.5}$ could be calculated by multiplying the percentages of primary/secondary components to the corresponding source contributions to the primary components/precursors. The proposed method had the following strengths: (1) it could identify the source contributions to secondary components; (2) target (or expected) sources were optional; (3) mixed sources could be avoided.

The proposed approach was then applied in the $PM_{2.5}$ source apportionment in two typical cities - Beijing and Tangshan. Source apportionment results estimated by the proposed combined approach indicated that the annual average contribution to $PM_{2.5}$ in Tangshan was 7.4%, 21.5%, 7.6%, 18.0%, 14.5%, 10.9% and 20.0% for power, metallurgy, cement, coal combustion, vehicle, dust and other sources (such as biomass, solvent use, biogenic, farmland and livestock sources), respectively; the annual average contribution ratio for vehicle, industry and ICC, RCC, dust and other sources (e.g., biomass burning, biogenic source, farmland, livestock and solvent use) in Beijing was 31.5%, 22.9%, 10.6%, 14.5% and 20.4%, respectively. Dust had the highest contribution in spring and coal combustion/residential coal combustion had the highest contribution in winter. The application results showed that the combined method was feasible. The source apportionment results could provide scientific support for the $PM_{2.5}$ pollution control strategy making.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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