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Seasonal Study of Primary and Secondary Sources of Carbonaceous Species in PM_{10} from Five Northern Chinese Cities

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ABSTRACT

PM_{10} samples were collected from five northern Chinese cities (Tianjin, Anyang, Ji'nan, Kaifeng and Taiyuan) during the winter and summer months. Carbonaceous species concentrations in the PM_{10} samples from these cities were then measured. The concentration values show that PM_{10}, organic carbon (OC) and elemental carbon (EC) exhibited relatively high concentrations during winter months as opposed to the summer months for all five cities. The contributions from primary and secondary sources were estimated using the Chemical Mass Balance (CMB) and the CMB-Iteration models. The results of two models were comparable, and results showed that the coal combustion and crustal dust were the most important source categories for both seasons. Additionally, seasonal variations of species mass fractions in receptors were analyzed, as well as variations in the estimated source percentage contributions. Seasonal variations of species mass fractions agreed with the variations of source percentage contributions for the five cities. Finally, primary organic carbon (POC) and secondary organic carbon (SOC) concentrations, and their seasonal variations, were estimated. We found that SOC showed relatively high concentrations ($\mu g/m^3$) in the winter, while the SOC/TOC ratios in the summer were higher than those in the winter.

Keywords: CMB; CMB-Iteration; SOC; PM_{10}; Source apportionment; Carbonaceous species.

INTRODUCTION

Carbonaceous matter is one of the major components of PM_{10}, especially in urban areas (Putaud *et al*., 2004; Li *et al*., 2012). Carbonaceous matter has been found to be associated with human health and urban environmental problems (von Schneidemesser *et al*., 2010; Hagler *et al.*, 2011; Pavese *et al*., 2012; Zhou *et al*., 2012). It (total carbon: TC) consists of two major components: elemental carbon (EC) and organic carbon (OC). EC, emitted mainly during the incomplete combustion of fossil fuels and biomass, is a primary pollutant, while OC has both primary and secondary origins. Primary organic carbon (POC) is formed during combustion and is directly emitted into the atmosphere in particulate form (Hildemann *et al*., 1994; Castro *et al*., 1999). Secondary organic carbon (SOC), on the other hand, is formed within the atmosphere via atmospheric reactions involving gas-to-particle conversion processes from volatile organic compounds (VOCs) (Seinfeld and Pandis, 1998).

According to several studies (Dan *et al*., 2004; Na *et al*., 2004; Duan *et al*., 2005; Avino *et al.*, 2011), the impact of SOC cannot be ignored. With the increase in photochemical reactions, SOC attributes high contributions to carbonaceous matter. Some indirect methods have been applied for the estimation of SOC concentrations. These methods include the application of models, such as the Lagrangian trajectory model and the transport model, which describe the formation, transportation and deposition of gaseous and particulate OC (Pandis *et al*., 1992; Hildemann *et al*., 1993). The models also allow the measurement of radiocarbon (14C) (Hildemann *et al*., 1994) and the utilisation of OC/EC ratios (Turpin and Huntzicker, 1995; Raman and Hopke, 2007), especially the minimum ratio between OC and EC (Castro *et al*., 1999).

Besides the above mentioned methods, the CMB model, a useful receptor model (Srivastava *et al*., 2009), has also been applied to estimate the contributions of primary source categories and the contribution of SOC to TOC (Marmur *et al*., 2005; Lee *et al*., 2007). The CMB model needs datasets from the source profile (Lee *et al*., 2008), but there is no actual profile for SOC. Therefore, in some works, a pure

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SOC profile was used (Lee *et al*., 2007; Lee and Russell, 2007). Additionally, a new model which called CMB-Iteration model was developed in our prior study (Shi *et al*., 2011). This model can estimate the SOC concentration as well as other primary source contributions, without needing the SOC profile.

Recently, due to rapid economic development and the acceleration of urbanisation, particulate matter pollution in northern China has shown regional characteristics (Chan and Yao, 2008; Xu *et al*., 2008). In addition, it has been found that SOC is an important contributor to the formation of particulate matter in China (Cao *et al*., 2004; Chan and Yao, 2008; Feng *et al*., 2009). Therefore, understanding the source contributions and SOC concentrations in urban cities can help the government manage urban air quality. In this study, the CMB and CMB-Iteration models were applied to estimate the source contributions and SOC concentrations in five northern Chinese cities. The seasonal variations of carbonaceous matter concentrations and the source contributions were also analyzed. To our knowledge, it is the first effort to do the region-wide survey of SOC in northern China, for seasonal variation studying; and the first time to use both of the CMB and CMB-Iteration models to estimate the SOC concentrations. The findings of this work can provide theoretical underpinnings for air control strategies.

MATERIALS AND METHODS

Studying Areas

Five cities, Tianjin, Anyang, Ji'nan, Kaifeng and Taiyuan, were selected for PM_{10} sampling. The locations of the cities are shown in Fig. 1. The cities are representative of mega and medium Chinese cities located in northern China having a similar continental climate marked by well-delineated seasons with cold and dry winters and hot and humid summers. The economic development of the cities, however, varied greatly. For example, Tianjin is an open coastal city, located in the Bohai economic circle, with human activities such as traffic and industry as a result of a booming economy (Han *et al*., 2012). Anyang, on the other hand, is an important steel industry base in the Henan province. Ji'nan is surrounded by mountains on three sides. It is influenced by its topography and its atmospheric pollutants, which cannot easily become diffuse, especially in the winter. Additional, Ji'nan is an important industrial city in China, it was famous for metallurgy, machinery and building materials industry. Kaifeng is a traditional agricultural area where industry is not well-developed. Taiyuan is a famous coal-producing and coal-consuming city. In fact, the high-speed developing economy of Taiyuan is largely propelled by massive coal consumption.

Sampling and Analysis

In each city, PM_{10} samples were collected by filtration with a medium-volume air sampler situated about 5 m from the ground. The pump was set at 100 L/min and ran continuously for 24 h. Two parallel medium-volume air samplers were used to obtain PM_{10} data on polypropylene membrane filters and quartz-fibre filters (Lee and Hopke, 2006). The numbers of obtained PM_{10} samples in the five cities were Tianjin, with 35 samples from Aug. to Sep. 2001 and 35 samples from Nov. to Dec. 2000; Anyang, with 30 samples from Aug. to Sep. and 30 samples from Nov. to Dec. 2001; Ji'nan, with 30 samples from Aug. to Sep. 2000 and 30 samples from Nov. to Dec. 1999; Kaifeng, with 30 samples in Feb. and 30 samples in Sep. 2005; and Taiyuan, with 30 samples from Aug. to Sep. 2001 and 35 samples from Dec. 2001 to Jan. 2002.

To apply the CMB and CMB-Iteration models, receptor and source profiles are necessary. According to the emission inventory and investigations in the monitoring areas: crustal dust, cement dust, vehicle exhaust, coal combustion and biomass burning may be important primary source categories in the cities. In Anyang and Taiyuan, the Anyang-Steel Manufactory and Taiyuan-Steel Manufactory are two of

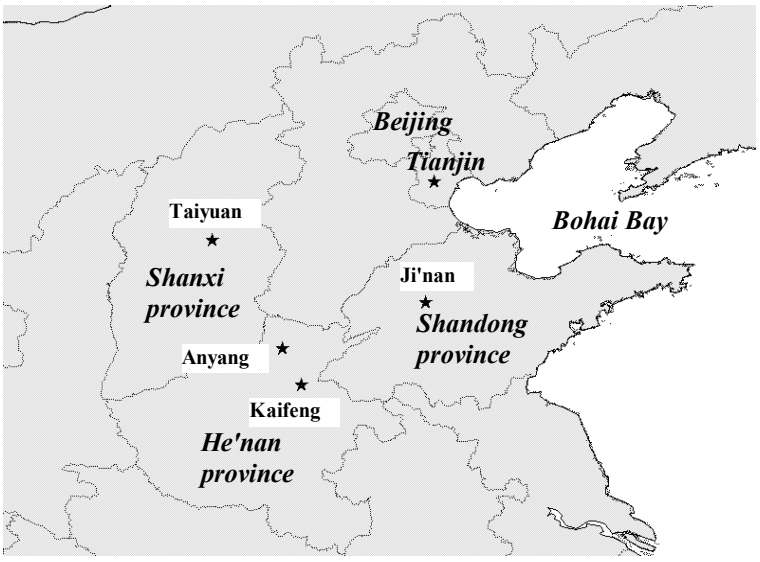

Fig. 1. Locations of the five cities in North China.

the largest factories in those cities, respectively. Therefore, steel manufacturing as a source may also be important for the two cities. Tianjin is located near the Bohai Bay, and the concentration of particulate matter was influenced by the wind from the direction of sea, so sea salt would also need to be considered. Aside from these primary sources, secondary sources such as sulfate, nitrate, and SOC may also contribute to PM_{10} in the five cities.

In the current study, source profiles reported in our prior studies (Zhao *et al*., 2006; Bi *et al*., 2007; Wu *et al.*, 2009) were used for the CMB and CMB-Iteration models to estimate source contributions. The crustal dust was collected from bare croplands, hilly countryside, and dry riverbeds in the vicinity of the cities. Coal combustion fly ash was collected from particulate pollution control devices (electrostatic precipitators, fabric filters or wet scrubbers). Steel manufacturing and vehicle exhaust source samples were taken from the steel industry and exhaust pipes, respectively. A dilution stack sampler and a vehicle exhaust sampler collected samples for PM_{10} directly on the filters. The powder samples were sieved and suspended in a re-suspension chamber or separated by a Bahco centrifugal machine. The sampling methods referred to their relative studies (Chow *et al*., 1994; Chan *et al*., 1997, 1999; Wilson *et al*., 2002). A sea salt profile and a biomass burning profile in speciate 4.2 from US EPA were used. "Pure" ammonium sulfate and ammonium nitrate were used as the secondary sulfate and nitrate source profiles (Mazzera *et al*., 2001; Park *et al*., 2005). A profile containing 100% OC was used as the SOC profile (Lee *et al*., 2007), for CMB model.

A clip in the shape of a circle (15 mm in diameter) from each quartz-fibre filter was used to measure TC and OC via an element analyser (VarioE1, Elementar Analysensysteme Gmbh, Hanau, Germany). For TC, the carbon species on the clip was oxidised into CO_2 at 980°C for 90 s, and then reduced to 600°C, with an analysis of oxygen gas. The quantity of TC was determined through the detection of CO_2 by a thermal conductivity detector (TCD). The analysis procedure of the OC fraction was similar to TC analysis, with the only difference of an oxidising temperature of 450°C. EC is derived as the difference between TC and OC (Duan *et al*., 2005).

To construct receptor and source profiles for the CMB and CMB-Iteration model, other species concentrations were determined. Elements (such as, Na, Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Pb, etc.) were analysed by Industively Coupled Plasma (IRIS Intrepid II, Thermo Electron) (Baldwin, 1994; Watson *et al*., 1999). Water soluble NH_4^+, Cl^-, NO_3^- and SO_4^{2-} were extracted by an ultrasonic extraction system (AS3120, AutoScience) and analysed by ion chromatography (DX-120, DIONEX) (Carvalho *et al*., 1995; Chow and Watson, 1999).

CMB-Iteration Model

The principle of CMB model had been described in lots of literature (Hopke, 2003; Shi *et al*., 2009). In this work, only the principle of CMB-Iteration model is presented, considering the length of the paper. CMB-Iteration model is a new method to estimate SOC and POC, based on the iterative method of CMB model (Shi *et al*., 2011). The model could estimate SOC concentrations as well as source contributions directly without using SOC profile. The detailed algorithm of the CMB-Iteration model is described in our prior study (Shi *et al*., 2011). CMB-Iteration is based on the traditional CMB model, and if the contribution of SOC is accounted for separately, it can be described as follows:

$$C_{(n\times1)} = F_{(n\times m)} \times S_{(m\times1)} + SOC_{(n\times1)} \quad (1)$$

where C ($n \times 1$) is the receptor species concentration vector ($\mu g/m^3$); F ($n \times m$) is the source profile matrix ($\mu g/\mu g$); S ($m \times 1$) is the source contribution vector ($\mu g/m^3$); n is the number of species measured; m is the number of source categories (expect for SOC); and 1 is the number of receptor samples. SOC ($n \times 1$) is the profile of SOC which can be expressed by

$$SOC_{(n\times1)} = [OC\ 0\ 0\ \ldots\ 0]^T_{(n\times1)} \quad (2)$$

The SOC profile isn't attempted in CMB-Iteration method, thus:

$$C_{(n\times1)} - SOC_{(n\times1)} = F_{(n\times m)} \times S_{(m\times1)} \quad (3)$$

where $C_{(n\times1)}$ is the original receptor, $SOC_{(n\times1)}$ is the SOC receptor.

The SOC should be set to zero at the beginning of the iteration. Then the revised receptor $C^*_{(n\times1)} = C - SOC$ and sources profiles can be introduced into the EPA CMB8.2. The revised receptor and sources are established at the k^{th} iteration as:

$$(C^*)^k_{(n\times1)} = F_{(n\times m)} \times S^k_{(m\times1)} \quad (4)$$

At the k^{th} iteration, the POC* concentration is estimated from source apportionment results based on the ratio of POC to total particle mass from source tests:

$$OC_j = S_j \times (OC/TOT)_j \quad (5)$$

$$\text{and} \quad POC^{*k} = \sum_{j=1}^{m} OC_j \quad (6)$$

where OC_j is the contribution ($\mu g/m^3$) of the jth primary emission source to total OC in the receptor; S_j is the estimated contribution ($\mu g/m^3$) of the jth primary emission source; $(OC/TOT)_j$ is the ratio of OC to total particle mass in the jth source profile ($\mu g/\mu g$).

$$POC^{*k} = \sum_{j=1}^{m} OC_j \quad (7)$$

Then, the k^{th} iterated concentration of SOC can be estimated as follows:

$$SOC^k = TOC - POC^{*k} \quad (8)$$

So, SOC^k is the final results of the CMB-Iteration.

RESULTS AND DISCUSSION

OC and EC Concentration in PM_{10}

The average values of PM_{10} and carbonaceous species concentrations ($\mu g/m^3$) for the five studied cities are shown in Table 1. For each city, the average concentration of PM_{10} in the winter is higher than that in the summer. According to several studies, PM mass usually showed high concentrations in the winter (Duan *et al*., 2005; Bi *et al*., 2007). During the summer, the PM_{10} concentrations ($\mu g/m^3$) were ranked in the following order: Taiyuan (146.36) > Anyang (126.17) > Tianjin (102.22) > Kaifeng (93.02) > Ji'nan (82.05). In the winter, however, the order was: Taiyuan (214.62) > Ji'nan (186.75) > Tianjin (177.78) > Anyang (170.36) > Kaifeng (169.24). Table 1 also lists some reported PM_{10} concentrations for other cities in China. It can be seen that the PM_{10} concentrations of the five studied cities are comparable to those of Beijing (Duan *et al*., 2005) and Guangzhou (Tan *et al*., 2009).

Table 1 further shows the OC and EC concentrations ($\mu g/m^3$) in the five cities. Similar to PM_{10}, Taiyuan got the highest OC and EC concentrations among the five cities, for both seasons. Coal combustion is one of important sources of OC and EC emissions (Duan *et al*., 2005). Taiyuan is the famous "coal city" in China. Therefore, it is reasonable that Taiyuan showed the highest concentrations of OC and EC. In the summer, the OC concentration showed the following ranking order: Taiyuan (25.89) > Anyang (17.20) > Tianjin (14.24) > Ji'nan (13.46) > Kaifeng (12.91), whereas the ranking order in the winter was: Taiyuan (40.98) > Tianjin (30.88) > Anyang (26.21) > Ji'nan (19.47) > Kaifeng (17.72). Some reported OC and EC concentrations from other cities in China and from around the world are also shown in Table 1. The concentrations of OC and EC for the five studied cites are comparable to concentrations found in Beijing and Guangzhou, and they are much higher than those of cities in other countries shown in Table 1 (Viidanoja *et al*., 2002; Harrison and Yin, 2008; Terzi *et al*., 2010).

Analysis of Seasonal Variation

The values in Table 1 indicate that the OC and EC concentrations during the winter were higher than those during the summer for the five cities. This is due to the contribution of coal combustion for residential heat supplies, which occurs during the winter in northern Chinese cities (Duan *et al*., 2005). The ratios of winter concentrations to summer concentrations for OC and EC are listed in Table 1. For each city, the EC-ratio was higher than the OC-ratio. Such results are similar to those in Duan *et al*. (2005).

In addition to the analysis of seasonal variation, similarities between summer and winter for the five cities were analysed using the coefficient of divergence (CD). The CD is self-normalising and can be calculated from short-term measurements or long-term averages. The CD is determined as follows (Lagudu *et al*., 2011):

$$CD_{fj} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}\left(\frac{x_{if} - x_{ij}}{x_{if} + x_{ij}}\right)^2} \quad (9)$$

Table 1. Average concentrations of PM_{10}, carbonaceous species and the ration of OC, EC in winter to summer for five cities ($\mu g/m^3$).

Cities	Season	PM_{10}	OC	EC	$OC_{winter/summer}$*	$EC_{winter/summer}$*	Reference
Tianjin, China	Summer	102.22	14.24	4.89	2.17	2.66	This study
	Winter	177.78	30.88	13.01			
Anyang, China	Summer	126.17	17.20	4.93	1.52	1.68	
	Winter	170.36	26.21	8.30			
Ji'nan, China	Summer	82.25	13.46	4.25	1.45	2.44	
	Winter	186.75	19.47	10.36			
Kaifeng, China	Summer	93.02	12.91	7.82	1.37	3.54	
	Winter	169.24	17.72	27.68			
Taiyuan, China	Summer	146.36	25.89	6.82	1.58	1.77	
	Winter	214.62	40.98	12.07			
Beijing, China	Summer	153.5	16.4	4.8			Duan *et al*., 2005
	Winter	171.2	25.6	10.5			
Helsinki, Finland	Summer		3.2	0.6			Viidanoja *et al*., 2002
	Winter		3.8	1.2			
Thessaloniki, Greece	Summer	58	6.40	2.91			Terzi *et al*., 2010
	Winter	69	8.73	2.93			
Birmingham, U.K.	Summer		4.7	1.1			Harrison and Yin, 2008
	Winter		3.0	2.3			
Guangzhou, China	Summer	116.25	21.75	5.95			Tan *et al*., 2009
	Winter	162.85	27.40	8.15			

$OC_{winter/summer}$: ration of OC in winter to summer
$EC_{winter/summer}$: ration of EC in winter to summer

where x_{if} is the average concentration of the *i*th species measured for the *f*th sample. *f* and *j* represent two ambient receptors, and *p* is the number of species.

In this work, we studied the similarity or dissimilarity of species mass fractions in different seasons for each city. Thus, in the Eq. (9), x_{ij} is the species mass fraction of the *i*th species measured at the *j*th city (Zhang *et al*., 2000). Species mass fraction was calculated as follows:

$$x_{ij} = \frac{c_{ij}}{TOT_j} \times 100\% \quad (10)$$

where x_{ij} is the species mass fraction (%) of the *i*th species in the *j*th source category; c_{ij} is the species concentration ($\mu g/m^3$) in *j*th source category; and, TOT_j is the PM_{10} mass concentration ($\mu g/m^3$) of *j*th source category.

In addition, we studied mass fraction diagrams, or scatter plots of species mass fractions comparing the sampling sites. The diagonal line with unit slope represents a hypothetical case in which the species mass fractions at the two sampling sites are equal. The diagonal line divides the diagram into two different regions, showing enhancement (above the line) and depletion (below the line) for individual species with respect to the reference site shown on the abscissa (Wongphatarakul *et al*., 1998; Zhang *et al*., 2000).

In this section, the CD values were combined with species mass fraction diagrams to study the species seasonal variations for each city. The results are shown in Fig. 2. CD values can show the similarity or dissimilarity of species mass fractions in receptors for different seasons. The species mass fractions in receptors can reflect contributions from possible categories. According to Fig. 2, Tianjin showed the highest CD values, which indicate that source contributions may produce large differences between the summer and winter months for the city. Anyang exhibited the lowest CD value, indicating that the difference was not very significant. The order of the CD values of the species fractions was: Tianjin (0.29) > Ji'nan (0.19) > Kaifeng (0.14) > Taiyuan (0.08) > Anyang (0.07).

Descriptions of the species mass fraction diagrams follow:

(a) For Tianjin, OC and EC are above the diagonal line. OC and EC mainly originate from coal combustion and vehicle exhaust emissions. OC may also originate from SOC. Thus, these three source categories may show higher contributions in the winter than in the summer. Ca is below the diagonal line and is the source marker of cement. Thus, cement may show higher fractions in the summer as compared to the winter. Al and Si are above the diagonal line. These species mainly come from crustal sources and coal combustion. Crustal sources and coal combustion may be more significant in the winter than in the summer. K is under the diagonal line and is the marker for the biomass burning profile (Lee *et al*., 2008). Biomass burning may exhibit higher contributions (%) in the summer than in the winter. Na and Cl^- showed much higher fractions in the summer than in the winter, indicating that sea salt was much more important in the summer than it was in the winter. SO_4^{2-} and NO_3^- are below the diagonal line. They are the source markers of secondary sulfate and nitrate. These two source categories may show relatively higher contributions (%) in the summer than in the winter.

(b) For Anyang, OC and EC are above the diagonal line, indicating that coal combustion, vehicle exhaust and SOC may show relatively higher contributions in the winter than in the summer. SO_4^{2-} is also above the diagonal line, indicating that secondary sulfate might be higher in winter. K, Fe and NO_3^- are close to the diagonal line. This indicates that there may be no significant differences in contributions (%) in the winter and summer for biomass burning, steel manufacture and secondary nitrate source categories. Ca was below the line, meaning that cement got higher contribution in summer.

(c) For Ji'nan, OC is under the diagonal line, while EC is close to the line. EC is emitted from primary sources (such as coal combustion and vehicles), while OC originates from both primary and secondary sources. Thus, there may be no significant differences in the contributions (%) during the winter and the summer for coal combustion and vehicles. This explains why SOC shows higher contributions (%) in the summer than in the winter. In addition, Al and Si show higher fractions in the winter than in the summer, while Ca and K exhibit higher fractions in the summer than in the winter. This phenomenon indicates that crustal dust may be more important in the winter as opposed to the summer while cement dust and biomass burning may be more important during the summer.

(d) For Kaifeng, OC is below the diagonal line while EC is above it, which means that coal combustion may show relatively higher contributions (%) in the winter than in the summer. Thus, vehicle exhaust and SOC may exhibit higher contributions (%) in the summer rather than in the winter. Al, Si and NO_3^- are above the line, and Ca is under the line, indicating that crustal dust and secondary nitrate may be more important in the winter than in the summer. Cement dust may also be more important in the summer than in the winter. No significant differences were seen for secondary sulfate and biomass burning.

(e) For Taiyuan, OC, EC, and NO_3^- showed higher fractions in the winter than in the summer, indicating that coal combustion, vehicle exhaust, SOC and secondary nitrate may show higher contributions (%) in the winter months. Ca and SO_4^{2-} however showed higher fractions in the summer. This means that cement dust and secondary sulfate may exhibit higher contributions (%) in the summer months. The locations of Fe and K on the plot show that there may be no significant differences in the winter and summer for steel manufacturing and biomass burning. Al and Si are close to the diagonal line. However, considering that coal combustion may be more significant during the winter, crustal dust sources may show more contributions (%) in the summer.

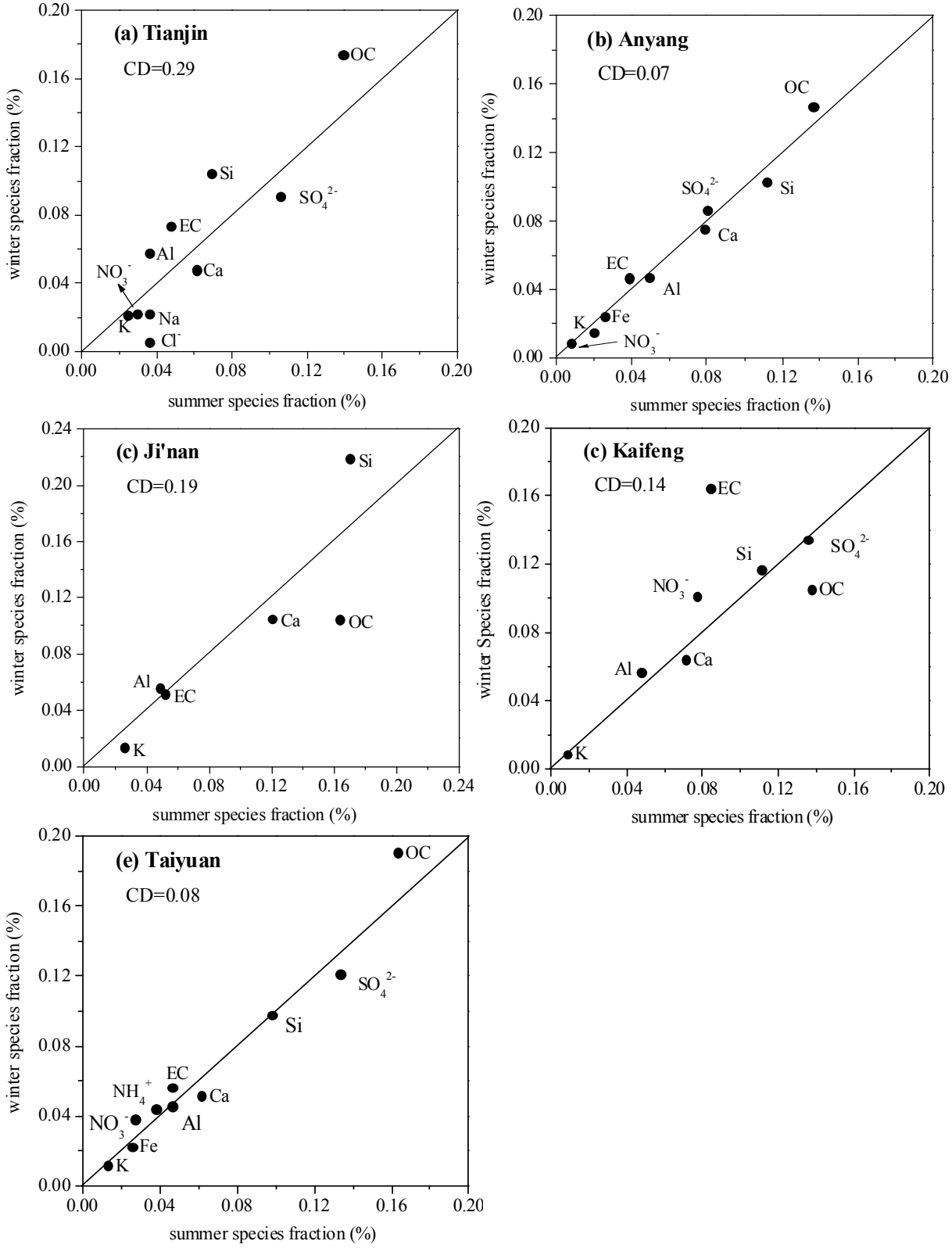

Fig. 2. Species fraction diagrams and CD values of species fractions for five cities.

Source Apportionment

In this section, ambient datasets from the five cities were introduced into the EPACMB 8.2 and CMB-Iteration models, contributions of potential source categories to PM_{10} and carbonaceous matter were estimated. The potential source categories in these five cities were also investigated in our prior studies, and the data of source profiles were also reported in these studies (Zhao *et al*., 2006; Bi *et al*.,

2007; Wu *et al*., 2009).

(1) Source contributions to PM_{10}:

The estimated source contributions to PM_{10} from the five cities during summer and winter by CMB and CMB-Iteration models are shown in Table 2 and 3, respectively. Results of two models were comparable. For the performance indices of CMB model: PM ranged from 83% to 109%; the χ^2 ranged from 0.50 to 2.92; and the R^2 ranged from 0.82 to 0.99. For the performance indices of CMB-Iteration model: PM ranged from 82% to 108%; the χ^2 ranged from 0.33 to 2.54; and the R^2 ranged from 0.82 to 0.99. These values met the requirement of the CMB model, meaning that these results can be acceptable.

According to Table 2 and 3, in Tianjin, the highest contributor during the summer was crustal dust, accounting for 25.62 μg/m^3 (25%) for CMB model and 25.17 μg/m^3 (25%) for CMB-Iteration model; in winter, the coal combustion got highest contribution for CMB model (61.87 μg/m^3, 35%), while for CMB-Iteration model, it was crustal dust, accounting for 54.37 μg/m^3 (31%). In Anyang, crustal dust was the highest contributor during summer from the both models, 25.42 μg/m^3 (20%) for CMB model and 25.38 μg/m^3 (20%) for CMB-Iteration model; in winter, the highest contributor for CMB model was crustal dust (47.32 μg/m^3, 28%), while for CMB-Iteration model, the highest contributor was coal combustion (46.07 μg/m^3, 27%). In Ji'nan, cement dust was the highest contributor in summer from both of the models, 16.88 μg/m^3 (21%) for CMB model and 16.86 μg/m^3 (20%) for CMB-Iteration model; whereas in the winter, it was crust dust for both models 72.11 μg/m^3 (39%) for CMB model and 71.96 μg/m^3 (39%) for CMB-Iteration model. In Kaifeng, crustal dust was the highest contributor during the summer and winter for both models: for CMB model, they were 28.36 μg/m^3 (30%) and 61.42 μg/m^3 (36%) in summer and winter, respectively; while for CMB-Iteration model, they were 27.79 μg/m^3 (30%) and 59.97 μg/m^3 (35%) in summer and winter, respectively. Lastly, in Taiyuan, crustal dust was an important source during the summer from both models, 35.55μg/m^3 (24%) for CMB model and 34.90 μg/m^3 (24%) for CMB-Iteration model; in the winter, it was coal combustion from both models 50.38 μg/m^3 (23%) for CMB model and 50.62 μg/m^3 (24%) for CMB-Iteration model.

According to the discussion above, coal combustion and crustal sources were the most important sources during summer and winter. The contributions from coal combustion during the winter were much higher than those during the summer because of the coal combustion used in residential heating during the winter.

The seasonal variations of source contributions were also analysed by the CD and source percentage contribution (%) diagrams. For the CD values of source contribution, the x_{ij} in Eq. (9) is the percentage contribution (%) of the *i*th source category in the summer, while x_{if} is the percentage contribution (%) of the *i*th source category in the winter. The results of the CD and source percentage contribution diagrams by CMB model and CMB-Iteration model are presented in Figs. 3 and 4 respectively, and results of two

Table 2. Source contribution (μg/m^3) and source percentage contribution (%) to PM_{10} by CMB model.

Source	Tianjin		Anyang		Jinan		Kaifeng		Taiyuan	
	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter
coal	10.33 (10%)	61.87(35%)	24.25 (19%)	43.43 (25%)	14.41 (18%)	31.95 (17%)	17.83 (19%)	38.91 (23%)	25.44 (17%)	50.38 (23%)
crust dust	25.62 (25%)	48.60(27%)	25.42 (20%)	47.32 (28%)	12.49 (15%)	72.11 (39%)	28.36 (30%)	61.42 (36%)	35.55 (24%)	39.77 (19%)
cement	10.53 (10%)	6.77(4%)	23.99 (19%)	28.87 (17%)	16.88 (21%)	28.48 (15%)	9.34 (10%)	11.58 (7%)	13.80 (9%)	16.12 (8%)
vehicle	7.21 (7%)	15.61(9%)	12.34 (10%)	21.54 (13%)	9.00 (11%)	23.73 (13%)	14.74 (16%)	14.49 (9%)	11.32 (8%)	20.78 (10%)
salt	6.94 (7%)	0.31(0%)	-	-	-	-	-	-	-	-
sulfate	11.74 (11%)	16.15 (9%)	11.88 (9%)	17.72 (10%)	-	3.25 (2%)	14.14 (15%)	26.38 (16%)	24.22 (17%)	26.81 (12%)
nitrate	3.62 (4%)	4.57 (3%)	1.01 (1%)	1.26 (1%)	-	0.96 (1%)	8.84 (10%)	21.45 (13%)	4.74 (3%)	9.45 (4%)
biomass	11.35 (11%)	14.53 (8%)	0.76 (1%)	1.82 (1%)	9.05 (11%)	6.59 (4%)	2.84 (3%)	5.90 (3%)	14.87 (10%)	31.30 (15%)
OC	5.78 (6%)	11.50 (6%)	8.39 (7%)	10.57 (6%)	5.65 (7%)	4.68 (3%)	3.28 (4%)	3.63 (2%)	6.30 (4%)	7.87 (4%)
Steel manufacture	-	-	1.73 (1%)	3.51 (2%)	-	-	-	-	8.26 (6%)	9.73 (5%)
PM (%)	91	101	87	103	83	94	107	109	98	99
χ^2	2.24	2.41	2.50	2.92	0.92	1.98	2.68	0.64	0.84	0.50
R^2	0.87	0.98	0.93	0.82	0.95	0.98	0.96	0.98	0.99	0.99

Table 3. Source contribution (μg/m^3) and source percentage contribution (%) to PM_{10} by CMB-Iteration model.

Source	Tianjin		Anyang		Jinan		Kaifeng		Taiyuan	
	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter
coal	10.80 (11%)	31.37 (18%)	24.88 (19%)	46.07 (27)	14.59 (18%)	32.14 (17%)	17.49 (19%)	38.65 (23%)	26.30 (18%)	50.62 (24%)
crust dust	25.17 (25%)	54.37 (31%)	25.38(20%)	45.60 (26)	12.40 (15%)	71.96 (39%)	27.79 (30%)	59.97 (35%)	34.90 (24%)	39.59 (18%)
cement	10.59 (10%)	7.78 (4%)	23.99(19%)	28.84 (17)	16.86 (20%)	28.47 (15%)	9.47 (10%)	11.89 (7%)	13.90 (9%)	16.14 (8%)
vehicle	6.92 (7%)	24.33 (14%)	12.35(10%)	21.59 (13)	8.89 (11%)	23.75 (13%)	16.84 (18%)	18.94 (11%)	11.29 (8%)	20.77 (10%)
salt	6.84 (7%)	3.02 (2%)	-	-	-	-	-	-	-	-
sulfate	11.75 (11%)	16.11 (9%)	11.88 (9%)	17.69 (10)	-	3.24 (2%)	13.92 (15%)	25.91 (15%)	24.21 (17%)	26.81 (12%)
nitrate	3.62 (4%)	4.35 (2%)	1.01(1%)	1.25 (1)	-	0.96 (1%)	8.80 (9%)	21.36 (13%)	4.74 (3%)	9.45 (4%)
biomass	11.89 (12%)	16.60 (9%)	0.76(1%)	1.78 (1%)	9.37 (11%)	6.60 (4%)	2.66 (3%)	5.57 (3%)	14.94 (10%)	31.38 (15%)
OC	5.65 (6%)	10.96 (6%)	8.38(7%)	10.18 (6%)	5.52 (7%)	4.63 (2%)	2.62 (3%)	2.14 (1%)	6.03 (4%)	7.74 (4%)
Steel manufacture	-	-	1.73 (1%)	3.44 (2%)	-	-	-	-	8.31 (6%)	9.74 (5%)
PM (%)	93	95	87	103	82	93	107	108	99	100
χ^2	1.81	1.70	1.67	2.54	0.69	0.99	1.34	0.43	0.42	0.33
R^2	0.87	0.96	0.93	0.82	0.94	0.98	0.96	0.98	0.99	0.99

models were similar. The order of the CD values from Figs. 3 and 4 is: Tianjin > Ji'nan > Kaifeng > Anyang > Taiyuan. This is the same as the order of the CD values shown in Fig. 2. Results shown in Figs. 3 and 4, indicate the following: (a) For Tianjin, coal combustion, crustal dust, SOC and vehicle emissions showed relatively higher contributions (%) in the winter than in the summer, while cement dust, sea salt, biomass burning, secondary sulfate and nitrate showed relatively higher contributions (%) in the summer than in the winter. (b) For Anyang, crustal dust, coal combustion, sulfate and vehicle exhaust exhibited relatively higher contributions (%) in the winter than in the summer, while cement, SOC, showed relatively higher contributions (%) in the summer than in the winter, biomass burning, steel manufacturing and nitrate showed little seasonal variations. (c) For Ji'nan, crustal dust, vehicle exhibited relatively higher contributions (%) in the winter than in the summer. However, SOC, biomass burning and cement showed the opposite trend, coal combustion, showed little seasonal variations. (d) For Kaifeng, coal combustion, crustal dust and secondary nitrate revealed higher contributions (%) in the winter than in the summer, whereas SOC, cement and vehicle emissions demonstrated the opposite trend, biomass burning and secondary sulfate showed little seasonal variations. (e) Lastly, for Taiyuan, coal combustion, vehicle exhaust secondary nitrate and biomass burning demonstrated higher contributions (%) in the winter than in the summer, however, cement, crustal dust, secondary sulfate, steel manufacture and SOC demonstrated the opposite trend. Among all of the source categories, coal combustion exhibited the highest seasonal variation for the five cities, due to heating supply in the winter. In conclusion, the seasonal variation results for estimated source contribution (%) (Figs. 3 and 4) agree with the seasonal variation results for the species fraction (%) (Fig. 2).

(2) POC and SOC concentrations:

The SOC concentrations were estimated using the CMB and CMB-Iteration models directly. POC concentrations can be calculated according to the primary source contributions. In the studies of Zheng *et al*. (2002, 2007), PM mass apportionment was calculated from the OC source apportionment results based on the ratios of OC to the total particle mass from source tests (Zheng *et al*., 2002, 2007). In our study, the contributions of primary emission sources to TOC in receptors are calculated as follows:

$$OC_j = s_j \times f_{OCj} \quad (11)$$

where OC_j is the contribution (μg/m^3) of the *j*th primary emission source to TOC in the receptor; s_j is the estimated contribution (μg/m^3) of the *j*th primary emission source; and f_{OCj} is the fraction (μg/μg) of OC in *j*th source profile. Therefore, POC is calculated as follows:

$$POC = \sum_{j=1}^{m} OC_j \quad (12)$$

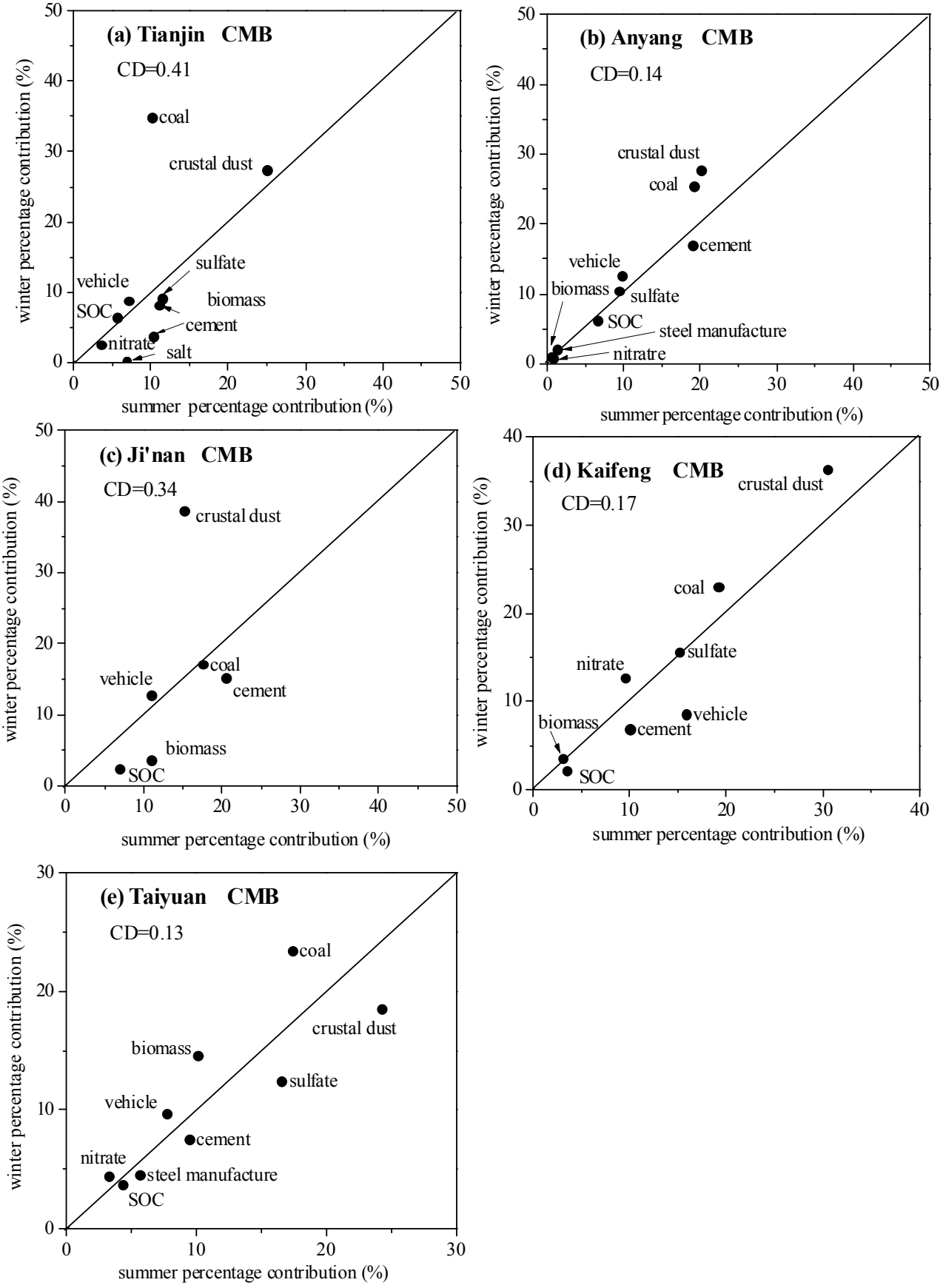

Fig. 3. Source percentage contribution diagrams and CD values of source percentage contributions for five cities by CMB model.

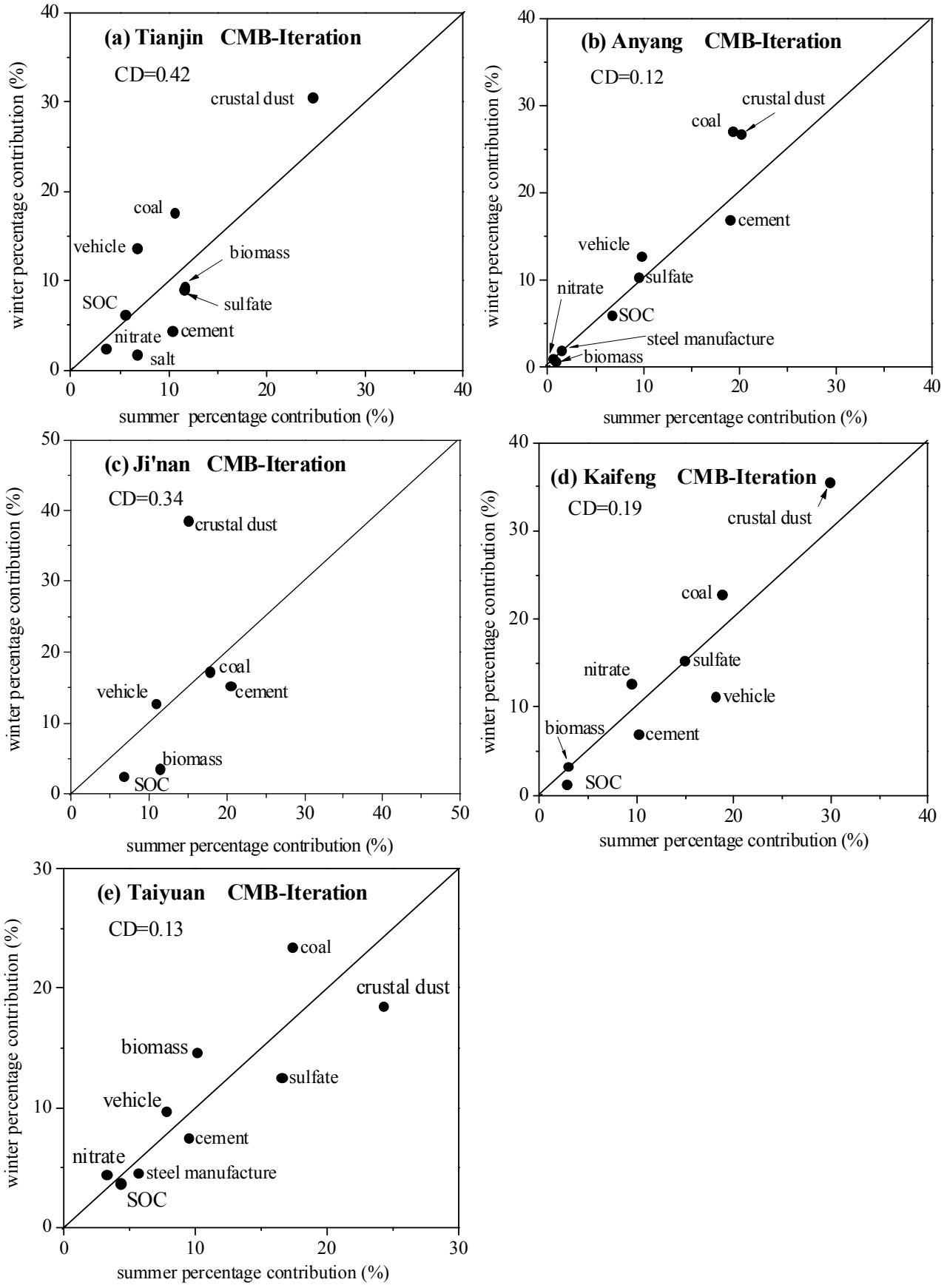

Fig. 4. Source percentage contribution diagrams and CD values of source percentage contributions for five cities by CMB-Iteration model.

Table 4 lists the concentrations of TOC, POC and SOC for the five cities during the different seasons studied. From the results of the CMB model, POC concentrations in Tianjin were 8.46 (μg/m^3) during the summer and 19.38 (μg/m^3) during the winter. In Anyang, the concentrations were 8.81 (μg/m^3) during the summer and 15.64 (μg/m^3) during the winter. In Ji'nan, they were 7.81 (μg/m^3) for the summer and 14.80 (μg/m^3) for the winter. Kaifeng showed POC concentrations of 8.87 (μg/m^3) during the summer and 12.65 (μg/m^3) during the winter. Lastly, in Taiyuan the concentrations were 17.60 (μg/m^3) during the summer and 33.10 (μg/m^3) during the winter. From the results of the CMB-Iteration model, POC concentrations in Tianjin were 8.59 (μg/m^3) during the summer and 19.92 (μg/m^3) during the winter. In Anyang, the concentrations were 8.82 (μg/m^3) during the summer and 16.02 (μg/m^3) during the winter. In Ji'nan, they were 7.95 (μg/m^3) for the summer and 14.85 (μg/m^3) for the winter. Kaifeng showed POC concentrations of 10.18 (μg/m^3) during the summer and 15.56 (μg/m^3) during the winter. In Taiyuan, the concentrations were 17.86 (μg/m^3) during the summer and 33.24 (μg/m^3) during the winter. The result of CMB model and CMB-Iteration model were comparable, and POC showed higher concentrations (μg/m^3) in the winter than those in the summer, for all five cites.

From the result of the CMB model, SOC concentrations in Tianjin were 5.78 (μg/m^3) during the summer and 11.50 (μg/m^3) during the winter. Anyang exhibited SOC concentrations of 8.39 (μg/m^3) in the summer and 10.57 (μg/m^3) in the winter. In Ji'nan, these concentrations were 5.65 (μg/m^3) in the summer and 4.68 (μg/m^3) in the winter. In Kaifeng, they were 3.28 (μg/m^3) during the summer and 3.63 (μg/m^3) during the winter. Finally, in Taiyuan, the SOC concentrations were 6.30 (μg/m^3) in the summer and 7.87 (μg/m^3) in the winter. From the result of the CMB-Iteration model, SOC concentrations in Tianjin were 5.65 (μg/m^3) during the summer and 10.96(μg/m^3) during the winter. Anyang exhibited SOC concentrations of 8.38 (μg/m^3) in the summer and 10.18 (μg/m^3) in the winter. In Ji'nan, these concentrations were 5.52 (μg/m^3) in the summer and 4.63 (μg/m^3) in the winter. In Kaifeng, they were 2.62 (μg/m^3) during the summer and 2.14 (μg/m^3) during the winter. Lastly, in Taiyuan, the SOC concentrations were 6.03 (μg/m^3) in the summer and 7.74 (μg/m^3) in the winter. Also, result of CMB model and CMB-Iteration model were comparable.

The ratios of SOC/TOC are shown in Table 4. Compared with primary sources, SOC was an important contributor to the TOC in each city. In addition, SOC/TOC ratios in the summer were higher than SOC/TOC ratios in the winter from both models. SOC formation typically occurs during the summer when photochemical activities are intensive under favourable meteorological conditions (Duan *et al*., 2005). Table 4 also presents ratios of SOC/TOC during

Table 4. Concentration of TOC, OC and EC for five cities in different season (μg/m^3).

Cities	Model	Season	TOC	POC	SOC	SOC/TOC	Reference
Tianjin	CMB	Summer	14.24	8.46	5.78	0.41	This study
		Winter	30.88	19.38	11.5	0.37	
	CMB-Iteration	Summer	14.24	8.59	5.65	0.40	
		Winter	30.88	19.92	10.96	0.35	
Anyang	CMB	Summer	17.2	8.81	8.39	0.49	
		Winter	26.21	15.64	10.57	0.40	
	CMB-Iteration	Summer	17.2	8.82	8.38	0.49	
		Winter	26.21	16.02	10.18	0.39	
Ji'nan	CMB	Summer	13.46	7.81	5.65	0.42	
		Winter	19.47	14.8	4.68	0.24	
	CMB-Iteration	Summer	13.46	7.95	5.52	0.41	
		Winter	19.47	14.85	4.63	0.24	
Kaifeng	CMB	Summer	12.91	8.87	3.28	0.25	
		Winter	17.72	12.65	3.63	0.20	
	CMB-Iteration	Summer	12.91	10.18	2.62	0.20	
		Winter	17.72	15.56	2.14	0.12	
Taiyuan	CMB	Summer	25.89	17.6	6.3	0.24	
		Winter	40.98	33.1	7.87	0.19	
	CMB-Iteration	Summer	25.89	17.86	6.03	0.23	
		Winter	40.98	33.24	7.74	0.19	
Beijing		Summer	16.4		9.2	0.57	Duan *et al*., 2005
		Winter	25.6		10.7	0.4	
Birmingham		Summer	4.77	1.67	3.1	0.65	Castro *et al*., 1999
		Winter	3.71	3.08	0.63	0.17	
Coimbra		Summer	5.16	2.58	2.58	0.5	
		Winter	9.76	6.15	3.61	0.37	
Birmingham		Summer	3	0.9	2.1	0.72	Harrison and Yin, 2008
		Winter	4.7	1.8	2.9	0.61	

different seasons in other cities (Castro *et al*., 1999; Duan *et al*., 2005; Harrison and Yin, 2008), which show similar patterns.

CONCLUSIONS

In this study, PM_{10} samples were collected during different seasons (summer and winter) for five northern cities in China. PM_{10}, OC and EC concentrations ($\mu g/m^3$) were then measured. For each city, PM_{10}, OC and EC showed higher concentrations in winter than in summer.

The potential source contributions to PM_{10} and TOC in each city were calculated with the CMB and CMB-Iteration models, results of two models were comparable. The results of source apportionment for the five cities showed that coal combustion and crustal sources were the most important sources category during summer and winter.

The coefficient of divergence (CD) was used to study the species mass fractions and source percentage contributions for the five cities to discuss the seasonal variations. The results of the CD values for source percentage contributions agreed with the results of the species mass fractions: seasonal variation in Tianjin was higher than it was for other cities, while Taiyuan showed the lowest variation between summer and winter. Ji'nan, Kaifeng and Anyang were at medium levels as compared to Tianjin and Taiyuan.

The results of the POC and SOC concentrations estimated during different seasons show that SOC was an important contributor to TOC in northern Chinese cities. The SOC/TOC ratios all showed higher values in summer as compared with winter, indicating that SOC formation occurs more actively in the summer than the winter.

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