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Spatial distribution and seasonal variation of char-EC and soot-EC in the atmosphere over China

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ABSTRACT

A previous study on $PM_{2.5}$ carbonaceous aerosols measured with the thermal optical reflectance (TOR) method in fourteen Chinese cities is extended by subdividing total EC into char-EC and soot-EC. Average char-EC concentrations show great differences between the fourteen cities and between winter and summer periods, with concentrations of 8.67 and 2.41 μg m^{-3} in winter and summer, respectively. Meanwhile spatial and seasonal soot-EC variations are small, with average concentrations of 1.26 and 1.21 μg m^{-3} in winter and summer, respectively. Spatial and temporal distributions of char-EC, similar to EC, are mainly influenced by local fuel consumption, as well as the East Asian monsoon and some meteorological factors such as the mixing height and wet precipitation. The small spatial and seasonal variation of soot-EC is consistent with its regional-to-global dispersion, which may suggest that soot carbon is not local carbon, but regional carbon. Char-EC/soot-EC ratios show summer minimum and winter maximum in all cities, which is in good agreement with the difference in source contributions between the two periods. As OC/EC ratio is affected by the formation of the secondary organic aerosol (SOA), char-EC/soot-EC ratio is a more effective indicator for source identification of carbonaceous aerosol than previously used OC/EC ratio.

1. Introduction

Carbonaceous aerosol has been receiving increased attentions due to its link with regional climate, water resources, visibility reduction, air quality, and public health (Jacobson, 2001; Menon et al., 2002; Hansen et al., 2004; Mauderly and Chow, 2008). Aerosol carbon is commonly classified as organic carbon (OC) and elemental carbon (EC). It was thought that OC cools the atmosphere by increasing Earth's reflectivity, while EC warms it by absorbing sunlight (Ackerman et al., 2000). At present the optical effect of EC on global climate change is still under much dispute (IPCC, 2007), which may be related to the complexity of carbonaceous components. EC is a collective term that encompasses all thermally altered carbonaceous material generated from fossil fuel combustion and biomass burning (Masiello, 2004), and can be subdivided into two classes: soot and char. Char is formed from the solid residues of combustion, whereas soot is formed from volatiles within and recondensed from the high-temperature gas phase. They have different physical and chemical properties, such as production mechanism, structure, size, transport mechanism, reactivity (degradation), etc, and thus differ in their aerosol optical characteristics that influence light absorption (Reid et al., 2005). It is essential to differentiate char from soot for our better understanding of their light-absorbing effects, as well as their environmental and climatic impacts.

Recently Han et al. (2007a) evaluated the use of the thermal optical reflectance (TOR) method from the Interagency Monitoring of Protected Visual Environments (IMPROVE), the most popular method for analyzing carbonaceous aerosol (Chow et al., 2001; Han et al., 2007a, 2009a), to differentiate char from soot. It was found that the pure char materials always peaked at EC1 (evolving at 550 °C in a 2% O_2/98% He atmosphere), while pure soot samples peaked at EC2 and EC3 (evolving at 700 and 800 °C in a 2% O_2/98% He atmosphere, respectively) (Han et al., 2007a). Comparing the TOR method with the chemothermal (CTO-375) methods (Accardi-Dey, 2003) demonstrated that soot-EC determined by the CTO-375 method corresponds to EC2 and EC3 as found by the TOR method (Han et al., 2007b). It is reasonable to use the TOR method to

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differentiate between char and soot (Han et al., 2007a). This differentiation was used in previous aerosol study (Han et al., 2008), as well as in soil and road dusts to indicate local air pollution in urban areas (Han et al., 2009b).

The IMPROVE-TOR method has determined large quantities of carbonaceous aerosols (OC and EC) in the database (more than 100,000 samples before 2001, Chow et al., 2001). However, artifacts are still involved in this method (in fact, all EC methods have their own artifacts). For example, some catalysts and ions may decrease the activity energy of EC in the analysis process, leading to negative results (underestimation) (Novakov and Corrigan, 1995; Han et al., 2009a); while some water soluble organic carbon were found not to be evolved in OC oxidization steps (e.g. Yu et al., 2002), resulting in positive results (overestimation). These artifacts also exist in the differentiation between char and soot using the TOR method. Apart from them, the most important factor that complicates EC determination in the environment may come from the uncertainty of the definition of the term EC since there is no clear physical entity for previous EC quantification (Han et al., 2009c). This method for the differentiation of char-EC and soot-EC now gives relatively clear chemical and physical entities for different EC parts and provides an opportunity to re-analyze the previous EC data and to constitute the char-EC and soot-EC database. In this study, we re-analyze the carbonaceous aerosols from 14 Chinese cities reported by Cao et al. (2007) by dividing EC into char-EC and soot-EC. Also the spatial distribution and seasonal variation of char-EC and soot-EC concentrations are discussed, and the char-EC/soot-EC ratios are compared with OC/EC ratios and used for EC source identification.

2. Sampling and analysis

$PM_{2.5}$ particles were sampled in fourteen cities in China: Chongqing (CQ), Guangzhou (GZ), Hong Kong (HK), Hangzhou (HZ), Shanghai (SH), Wuhan (WH), Xiamen (XM), Beijing (BJ), Changchun (CC), Jinchang (JC), Qingdao (QD), Tianjin (TJ), Xi'an (XA), and Yulin (YL). The first seven of these cities are located in southern China, and the last seven in northern China. A detailed description of the sampling location and the pretreatment of these samples have already been reported by Cao et al. (2007). The sampling was carried out simultaneously, during two periods in 2003: winter (6 January–20 January) and summer (3 June–30 July). The IMPROVE-TOR method was used to quantify carbon compositions of the samples and produced four OC fractions (OC1, OC2, OC3, and OC4 at 120, 250, 450, and 550 °C, respectively, in a helium [He] atmosphere); OP (a pyrolyzed carbon fraction determined when reflected or transmitted laser light attained its original intensity after oxygen [O_2] was added to the analysis atmosphere); and three EC fractions (EC1, EC2, and EC3 at 550, 700, and 800 °C, respectively, in a 2% O2/98% He atmosphere). OC is operationally defined as OC1 + OC2 + OC3 + OC4 + OP and EC is defined as EC1 + EC2 + EC3 − OP. Han et al. (2007a) defined EC1 − OP as char-EC and EC2 + EC3 as soot-EC. Average field blanks were 1.85, 0.53, 0.40 and 0.13 μg m^{-3} for OC, EC, char-EC, and soot-EC, respectively. The duplicate differences for char-EC and soot-EC are better than 10%.

3. Results and discussion

3.1. Concentration levels of char-EC and soot-EC

Char-EC and soot-EC concentrations in the fourteen cities are summarized in Table 1. The average char-EC concentrations for the fourteen cities are 8.67 ± 6.19 and 2.41 ± 2.28 μg m^{-3} in winter and summer, respectively, which account for 87.6% and 66.6% of the total EC, respectively. The variation in char-EC between winter and summer may be mainly attributed to the different fuel consumption in the two periods since char is mainly produced from coal and biomass combustions, which decreases in summer especially in Northern China. In addition, the East Asian monsoon (An et al., 2000) is another important factor influencing atmospheric circulation and pollutant transportation (Fig. 2). In winter, the monsoon moves southward as Northern Hemisphere insolation decreases, and carries pollutants to Southern China, while in summer, the monsoon moves northward. Clean air from the ocean related to the summer monsoon would decrease pollutants in the atmosphere in coastal cities. Furthermore, meteorological factors such as the mixing height and wet precipitation also contribute to the seasonal variations of char-EC concentrations through pollutant dispersion and deposition (Husain et al., 2007; Choi et al., 2008). High precipitation related to the summer monsoon, as well as the high mixing heights in summer would decrease char-EC concentrations. Average soot-EC concentrations show relatively small variations between winter and summer, at 1.26 and 1.21 μg m^{-3} and contributing to 2.6% and 7.0% of the total carbon (TC, Cao et al., 2007), respectively. It is interesting to note that soot-EC concentrations from the fourteen cities are comparable to total EC in urban areas of the United States (~1.0 μg m^{-3}, Malm et al., 2004; Hansen et al., 2004), where motor vehicle exhaust is likely to be the main contributor to atmospheric EC. The small variations of soot-EC between winter and summer, despite more fuel consumption in winter in northern China, may be associated with two main factors: high amount of industrial coal consumption, far exceeding the

Table 1
Winter and summer char-EC and soot-EC concentrations in fourteen cities.

Locations	Seasons	Char-EC μg m^{-3}	Soot-EC μg m^{-3}	Char-EC/soot-EC	N
Chongqing	Winter	15.46 ± 5.45	1.13 ± 0.67	13.66	15
	Summer	7.00 ± 2.69	1.04 ± 0.57	6.75	16
Guangzhou	Winter	13.09 ± 9.47	1.41 ± 0.49	9.28	14
	Summer	1.99 ± 0.56	1.19 ± 0.45	1.67	13
Hong Kong	Winter	4.28 ± 2.37	1.51 ± 0.86	2.84	16
	Summer	2.55 ± 1.02	0.98 ± 0.68	2.61	22
Hangzhou	Winter	8.01 ± 2.17	1.35 ± 0.27	5.94	14
	Summer	2.31 ± 1.24	1.31 ± 0.61	1.76	16
Shanghai	Winter	7.12 ± 4.96	1.20 ± 0.52	5.93	16
	Summer	1.81 ± 1.11	1.12 ± 0.61	1.61	14
Wuhan	Winter	7.11 ± 2.70	1.31 ± 1.00	5.41	13
	Summer	1.74 ± 0.48	1.26 ± 0.44	1.38	13
Xiamen	Winter	3.98 ± 1.34	1.01 ± 0.16	3.95	15
	Summer	0.81 ± 0.59	0.69 ± 0.76	1.17	12
Beijing	Winter	6.15 ± 3.44	0.91 ± 0.18	6.75	14
	Summer	3.42 ± 2.07	1.91 ± 1.59	1.79	14
Changchun	Winter	12.35 ± 4.20	1.15 ± 4.80	10.74	14
	Summer	1.63 ± 1.04	1.22 ± 0.44	1.34	16
Jinchang	Winter	3.97 ± 0.98	1.03 ± 0.28	3.84	15
	Summer	0.47 ± 0.57	1.11 ± 0.14	0.43	8
Qingdao	Winter	5.45 ± 2.32	0.86 ± 0.62	6.35	13
	Summer	0.67 ± 0.57	0.71 ± 0.36	0.94	9
Tianjing	Winter	7.73 ± 3.28	0.79 ± 0.57	9.74	16
	Summer	2.13 ± 0.99	1.54 ± 0.59	1.39	15
Xi'an	Winter	19.78 ± 5.22	1.84 ± 0.98	10.74	15
	Summer	5.47 ± 2.97	1.53 ± 0.25	3.58	15
Yulin	Winter	8.07 ± 3.91	1.09 ± 0.31	8.6	14
	Summer	1.86 ± 1.00	1.41 ± 0.57	1.31	12
Average	Winter	8.67 ± 6.19	1.26 ± 1.40	6.88	204
	Summer	2.41 ± 2.28	1.21 ± 0.72	1.98	195

residential coal consumption (Fig. 3), and the longer lifetime of soot than previous estimation for the total EC (Ogren and Charlson, 1983).

3.2. Spatial distribution and seasonal variation of char-EC

The spatial and seasonal distributions of char-EC are presented in Fig. 1. In winter, the highest char-EC concentration is observed in Xi'an, followed by Chongqing, Changchun, and Guangzhou, which all have concentrations higher than (or close to) 10 μg m^{-3}. The reason for the high EC concentration in Xi'an was discussed by Cao et al. (2007), and was associated with the combination of coal combustion during the "heating season" from November to March and the local unfavorable meteorological conditions. EC comes mainly from three sources: biomass burning, coal combustion, and vehicle exhaust. As char-EC is produced mainly from coal combustion and biomass burning, the explanations for high winter EC concentrations in Xi'an (Cao et al., 2007) are also applicable to char-EC. High char-EC in winter in Changchun can also be attributed to the "heating season" as it is among one of the coldest mega-cities. Chongqing has no formal "heating season", but it is surrounded by Sichuan, a developing province, which produces the highest EC emissions from biomass burning and coal combustion in China (Streets et al., 2001). It was estimated about 21.7 Gg (Gigagrams) EC emitted from residential combustion in Chongqing in 2000 (Cao et al., 2006a). Because of the sparse attention paid to combustion processes and emission controls, heavy emissions were expected around Chongqing (Cao et al., 2006a). Meanwhile, the low wind speeds, high relative humidity, and stable atmospheric conditions suggested by Cao et al. (2007) may enhance the levels.

There is no clear reason for high char-EC concentrations during winter in Guangzhou. As a developed city, its fuel structure has switched to natural gases and central stream (Fig. 3). However, biomass burning in both suburban and urban Guangzhou is still ubiquitous as suggested by Wang et al. (2007). Using acetonitrile as a biomass tracer, Wang et al. (2007) estimated that the contribution of biomass burning to ambient $PM_{2.5}$ in Guangzhou was about 4.0–19.0%. In addition, the winter prevailing wind related to the East Asian monsoon may bring pollutants to Guangzhou from inland regions (Fig. 2), where there are high EC emissions from biomass and coal combustion. Furthermore, of the most importance is the short-term sampling period in this study, which may introduce a bias to our understanding of char-EC distribution in Guangzhou. For example, Duan et al. (2007) measured EC concentrations in Guangzhou in winter and summer based on short-term sampling periods, and showed lower concentrations with no distinct difference between the two seasons (5.1 and 4.0 μg m^{-3} in Liwan, a downtown area of Guangzhou, in summer and winter, respectively). Compared with the 2002 winter EC concentration (8.1 μg m^{-3}) in Guangzhou reported by Cao et al. (2003), the char-EC concentrations in this study (13.1 μg m^{-3}) are still too high. We suggest this unexpected high char-EC concentration as "unknown events".

Low winter char-EC concentrations occur in Qingdao, Hong Kong, Xiamen, and Jinchang. The first three of these cities are in coastal environments where the local air mass is mixed with maritime air. Jinchang is in a desert region and has the lowest industrial and residential coal consumption among the fourteen cities (Fig. 3). The average char-EC concentration in Jinchang (3.97 μg m^{-3}) is still much higher than that in Daihai (2.43 μg m^{-3}), a rural mountain area in Inner Mongolia (Han et al., 2008). This may suggest that in extensive remote rural areas in China, the winter char-EC concentrations may be much lower than those reported in these cities due to the relatively lower fuel consumption.

In summer the corresponding char-EC concentrations are all lower than those in winter, which may be associated with changes in energy use, as well as the prevailing wind variation related to the East Asian monsoon in the different seasons (Fig. 2). The highest summer char-EC concentrations occur in Chongqing, reaching 7.0 μg m^{-3}, a value even higher than the winter char-EC concentrations in Beijing (6.15 μg m^{-3}). This is in good agreement with the biomass burning around the city (Streets et al., 2001; Cao et al., 2006a), since biomass

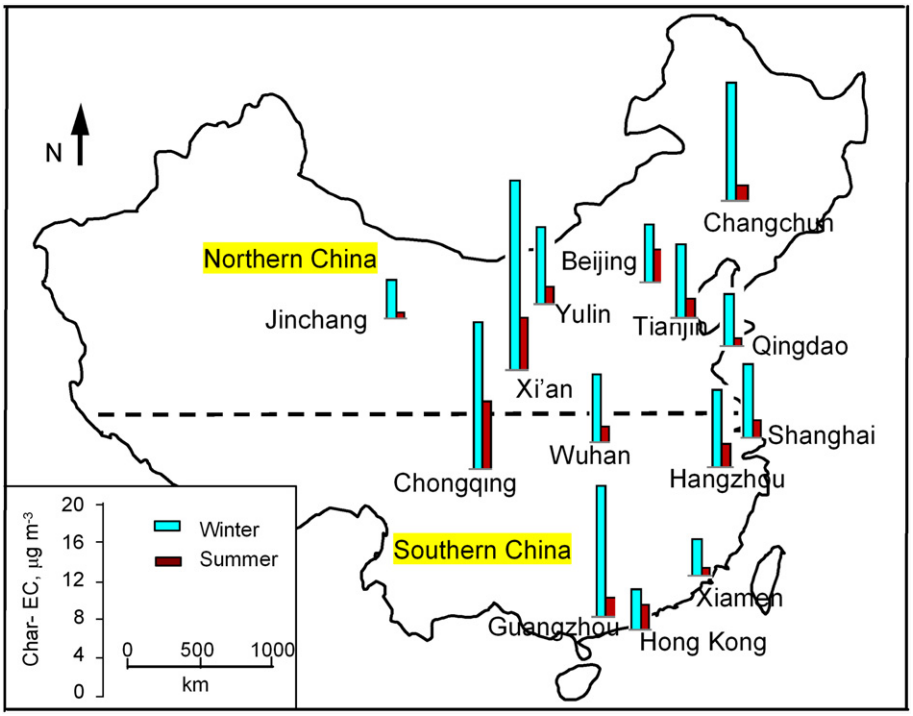

Fig. 1. Spatial distribution of char-EC concentrations in winter and summer.

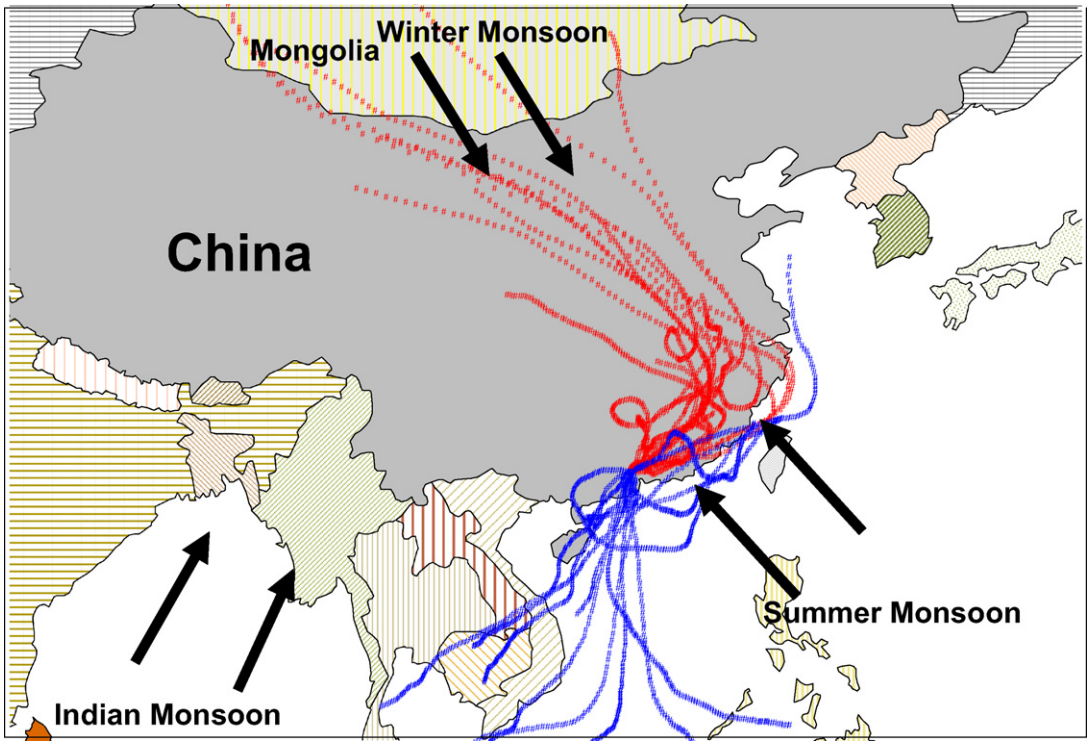

Fig. 2. 120-h backward air mass trajectories passing over Guangzhou in winter (red lines) and summer (blue lines) during sampling time (Produced using NOAA ARL Website: www.arl.noaa.gov/ready). The climatic system of China, including the summer and winter East Asian monsoon and the Indian monsoon, is also presented. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

burning for cooking and land clearance occurs all year in the Sichuang Basin. Following Chongqing, Xi'an is another city with high summer char-EC. This has been discussed before in terms of its unfavorable meteorological conditions and high industrial emissions as a developing city (Cao et al., 2007). Low summer char-EC concentrations occur in Xiamen, Qingdao, and Jinchang, with concentrations all lower than 1 μg m^{-3}, which is consistent with the low industrial coal consumption in these cities (Fig. 3).

The ratios of winter to summer average char-EC concentrations are in the decreasing order of Jinchang (8.5), Qingdao (8.1), Changchun (7.6), Guangzhou (6.5), Xiamen (4.9), Yulin (4.3), Wuhan (4.1), Shanghai (4.0), Tianjin (3.6), Xi'an (3.6), Hangzhou (3.5), Chongqing (2.2), Beijing (1.8), Hong Kong (1.7). The high ratios generally occur in the northern and some coastal cities. Cao et al. (2007) linked total carbon (TC) variations to both the fuel structure difference and the meteorological conditions such as mixing height and the monsoon system, and these factors may also be important for char-EC variations in this study. High ratios occurring in Jinchang, Qingdao and Changchun in northern China are obviously associated with the different fuel structure, since these cities have a long-term 'heating season' in which coal is the main winter fuel. Chongqing and Hong Kong show the lowest winter to summer char-EC ratios among these cities, probably due to a small difference in fuel structure between summer and winter in these cities. Throughout the year Chongqing is affected by biomass burning, while Hong Kong is affected by heavy vehicle exhaust.

3.3. Spatial distribution and seasonal variations of soot-EC

Unlike the high variations in char-EC among the different cities, the soot-EC levels have relatively small variations in both spatial and seasonal scales (Fig. 4). The average winter soot-EC concentrations in different cities vary between 1.84 and 0.79 μg m^{-3}. The summer soot-EC ranges from 1.91 to 0.71 μg m^{-3}, with high concentrations in inland cities such as Beijing, Tianjin, Xi'an, Yulin, Hangzhou, Wuhan and Changchun, and low concentrations in coastal cities such as Hong Kong, Qingdao and Xiamen; this pattern appears to be associated with the summer monsoon climate as clean ocean air would decrease soot-EC concentrations in coastal cities. In both winter and summer the soot-EC concentrations in inland cities are generally higher than 1 μg m^{-3}, which is a little higher than in Daihai (0.81 μm^{-3} in winter), a rural mountain area in Inner Mongolia (Han et al., 2008).

Also, the average soot-EC concentrations from the fourteen cities vary very little between winter and summer. The winter

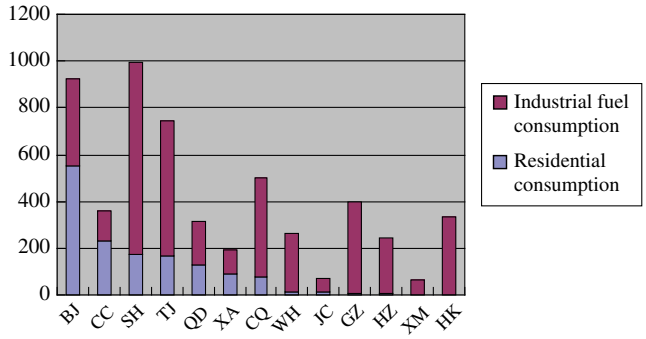

Fig. 3. Winter coal consumption (10^4 ton) in 2004 in different cities (Data from http://air.ipe.org.cn/qyInfo.do and reference therein). Yulin's data are not available and are therefore not presented. Industrial fuel consumption in winter are the total consumption divided by 4 since there are four seasons; residential consumption from cities in southern China are also the corresponding total consumption divided by 4, while those from cities in northern China are the total consumption as it is assumed that they are mainly derived from the winter heating. Industrial and residential coal combustions have different EC emission factors (EFs), and industrial coal combustion was estimated to have lower EFs (Streets et al., 2001).

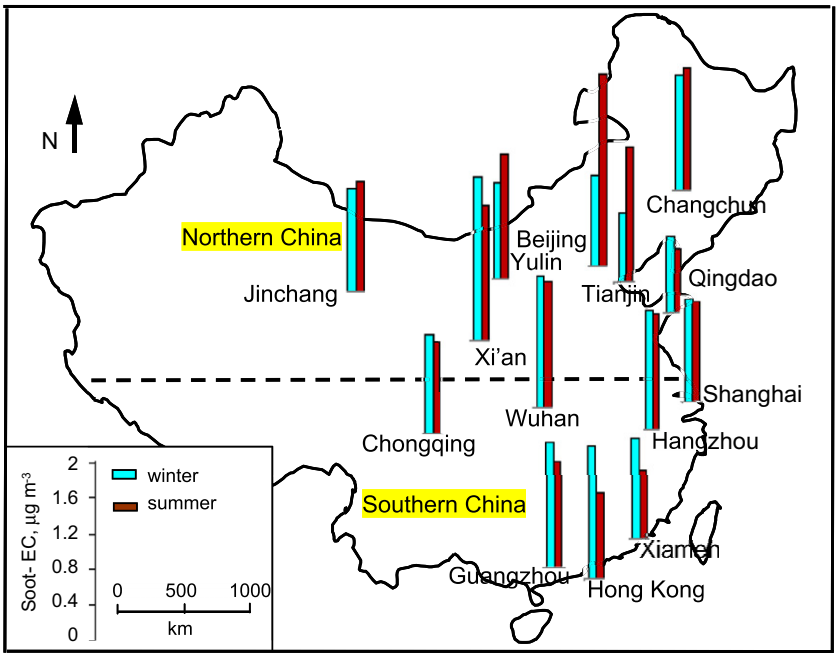

Fig. 4. Spatial distribution of soot-EC concentrations in winter and summer.

soot-EC concentrations for these cities are generally comparable with, and sometimes even lower than, the corresponding summer concentrations (Table 1), with the ratios of winter to summer soot-EC ranging from 1.5 to 0.5. This pattern is consistent with the submicron particle size of soot, which allows it to be regionally and globally dispersed. The spatial and temporal small variations in soot-EC concentrations suggest that soot may have a longer lifetime in the atmosphere than previously estimated for the total EC (Ogren and Charlson, 1983), and thus has a greater contribution to the global warming.

3.4. Correlations in char-EC and soot-EC

Correlations between mass, OC, EC, char-EC, and soot-EC in winter and summer are summarized in Table 2. Strong correlations between OC and EC have been observed in urban areas (e.g. Cao et al., 2003, 2007), and have been attributed to the common sources of OC and EC. Moderate correlations between OC and EC have also been found in rural and remote areas (e.g. Han et al., 2008), where long range transport of OC and EC mixes with local sources. Char-EC correlates well with mass, OC, and in particular EC, in both seasons. Among these components, the strongest correlations are between EC and char-EC, in both winter and summer (Table 2). Considering that char-EC dominates EC and that soot-EC varies only slightly at spatial and seasonal scales, it can be inferred that EC concentration is mainly determined by char-EC concentration. Thus EC concentrations reported in the literature can be used to calculate char-EC concentrations. The corresponding formulae are illustrated in Fig. 5. The slopes in winter and summer are very similar, and the intercepts of the lines of the best fits, which may represent the background soot-EC concentrations in summer and winter, also show close values. This is consistent with the seasonal small variations of soot-EC.

Soot-EC, compared with char-EC, shows very weak (especially in winter) correlations (such as in winter) with mass, OC, and EC, which may be associated with the different formation modes of char and soot. Generally soot is formed from volatiles recondensed from the high-temperature gas phase, while char forms from the solid residues of combustion, and tends to keep its parent characteristics. Correlation between soot-EC and EC is better in summer than in winter, consistent with the main contributions to EC from vehicle exhaust in summer. Correlation between char-EC and soot-EC is similar to the findings of Han et al. (2008), with very poor correlation in winter and moderate positive correlation in summer, probably due to the motor vehicle emissions dominating in summer. However, this pattern is very different from OC/EC correlations, which are generally strong in winter, but more moderate in summer (e.g. Cao et al., 2007; Han et al., 2008).

3.5. Char-EC/soot-EC ratio as an indicator of source characterization

Previously, OC/EC ratio has been used to identify the origin of carbonaceous aerosol (i.e. Novakov et al., 2000; Cao et al., 2005, 2007; Malm et al., 2004; Zhang et al., 2007). It is thought that the primary emission has distinct OC/EC ratios, with fossil fuel emissions tending to have low OC/EC ratios, while emissions from

Table 2
Correlations of mass, OC, EC, char-EC, and soot-EC in winter and summer.

Winter correlations					
$N = 204$	Mass	OC	EC	Char-EC	Soot-EC
Mass	1	0.939[a]	0.827[a]	0.814[a]	0.141
OC		1	0.904[a]	0.892[a]	0.143
EC			1	0.975[a]	0.211[a]
Char-EC				1	−0.011
Soot-EC					1
Summer correlations					
$N = 195$	Mass	OC	EC	Char-EC	Soot-EC
Mass	1	0.848[a]	0.697[a]	0.674[a]	0.282[a]
OC		1	0.830[a]	0.814[a]	0.298[a]
EC			1	0.958[a]	0.430[a]
Char-EC				1	0.154
Soot-EC					1

[a] Correlation is significant at the 0.01 level (2-tailed).

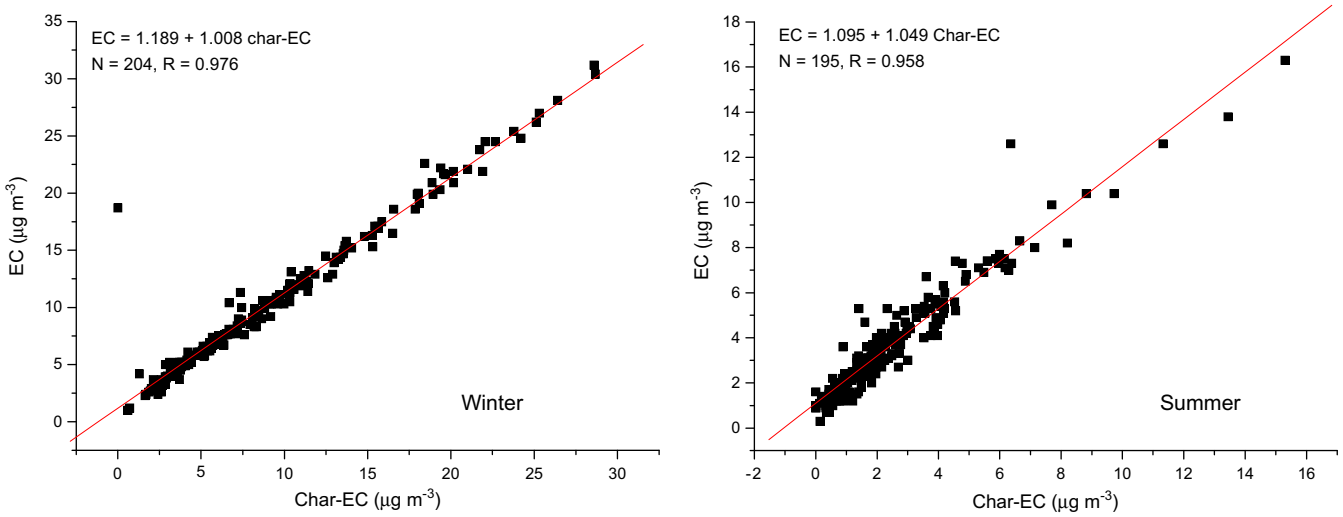

Fig. 5. Strong correlations of char-EC and EC, which indicate that EC concentrations can be used to calculate char-EC concentrations.

biomass fires are substantially higher; these ratios can thus be used to identify pollutant sources. Similar to OC/EC ratio, char-EC/soot-EC ratios also vary with different source emissions. Char-EC/soot-EC ratios from biomass burning and coal combustion are generally higher than those from motor vehicle exhaust (Han et al., 2008), though they may be influenced by combustion mode (flaming versus smoldering), fuel type, and moisture content. Calculations using carbon fractions reported by Chow et al. (2004), show char-EC/soot-EC ratios of 22.6 for biomass burning and 0.60 for motor vehicle exhaust. The average char-EC/soot-EC ratios from Xi'an city are 11.6 for biomass burning and 1.9 for coal combustion (Cao et al., 2005). Hong Kong roadside samples show char-EC/soot-EC ratios of 0.3 for diesel exhaust and 0.7 for gasoline emissions (Cao et al., 2006b). Emissions from laboratory combustion of wildland fuels reveal char-/soot-EC ratio of up to 31 from sagebrush burning (Chen et al., 2007). So char-/soot-EC ratio can be used for source identification (Han et al., 2008, 2009b).

OC/EC ratios are generally influenced by three factors including primary emission sources, SOA formation, and different OC and EC removal rates by deposition (Cachier et al., 1996; Cao et al., 2005). Great bias may exist in source identification using OC/EC ratios because of the influence of the SOA formation (e.g. Castro et al., 1999; Cao et al., 2003; Yuan et al., 2006; Zhang et al., 2007; Schichtel et al., 2008). SOA in the atmosphere can exist for several days, affecting the OC/EC ratio significantly. Numerous studies have shown that OC/EC ratios vary little between winter and summer (e.g. Novakov et al., 2005), and this is also the case for the fourteen Chinese cities (Cao et al., 2007). Obviously, this is not consistent with the changes in fuel structure between winter and summer. Thus there may be great biases for using OC/EC ratio for carbonaceous aerosol source apportionment. As char and soot are predominately produced from combustion processes, their ratios are generally influenced by only two factors: primary emission sources and removal rate by deposition. For close-source particles, such as in urban areas, the removal rate factor can be neglected. This means that char-EC/soot-EC ratio is mainly influenced by emission sources and thus is a more effective source indicator than OC/EC ratio. The poor correlations between OC/EC ratios and char-EC/soot-EC ratios (Fig. 6) indicate their differences in source identification.

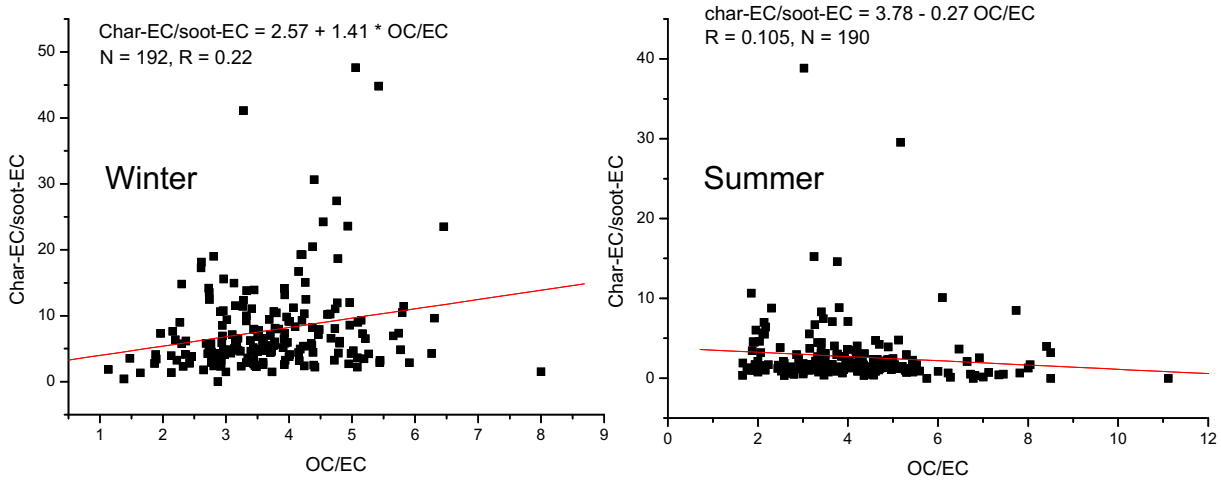

Fig. 6. Poor relationship of char-EC/soot-EC ratio and OC/EC ratio in winter and summer. Some samples have soot-EC concentrations to zero or close to zero, which would create too large and unavailable char-EC/soot-EC values, influencing the assessment of correlation between char-EC/soot-EC ratio and OC/EC ratio. Thus these samples are deleted in this analysis.

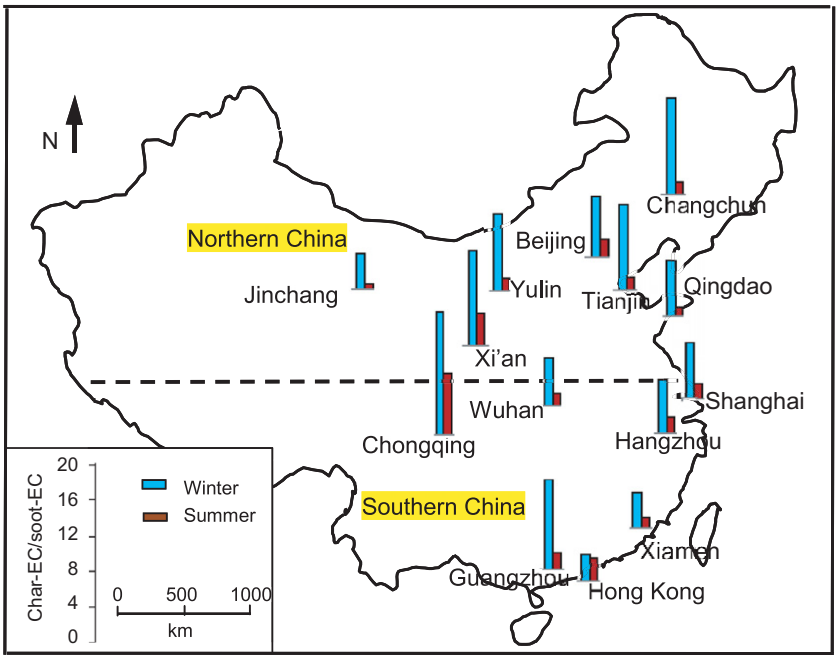

Fig. 7. Spatial distribution of Char-EC/soot-EC ratio in winter and summer.

The temporal and spatial distributions of char-EC/soot-EC ratio (Fig. 7) support its source indicator. The winter average char-EC/soot-EC ratio is 6.88, indicating significant contributions of EC from coal combustion and biomass burning in China (Streets et al., 2001; Cao et al., 2006a; Fig. 3). The winter ratios among the fourteen cities vary in the decreasing order of Chongqing, Changchun, Xi'an, Tianjing, Guangzhou, Yulin, Beijing, Qingdao, Hangzhou, Shanghai, Wuhan, Xiamen, Jinchang and Hong Kong. Generally high char-EC/soot-EC ratios occur in the northern Chinese cities, while low char-EC/soot-EC ratios are observed in the southern Chinese cities. This pattern is consistent with the fact that coal and some biofuels are generally used for heating in northern China (Fig. 3). However, in southern China the winter char-EC/soot-EC ratios are also higher than their corresponding values in summer (Table 1). This may be mainly associated with the long range transport of EC entrained by the winter Asian monsoon from northern to southern China. Chongqing is located in southern China and has no distinct "heating season", but is surrounded by biomass burning all year, so the high char-EC/soot-EC ratios in winter there can be attributed to the emissions from biomass combustion. The high ratio of char-EC/soot-EC during winter in Guangzhou is an exception because of the "unknown events" as the short-term sampling may bias the data.

The lowest winter char-EC/soot-EC ratios occur in Hong Kong, Xiamen and Jinchang. The first two cities located in southern China have the lowest residential coal consumption (Fig. 3). Jinchang, located in a desert area, has a char-EC/soot-EC ratio of 3.84, which is close to the winter char-EC/soot-EC ratio of 3.2 in Daihai, a rural area in Inner Mongolia with an even lower human population. Since soot is composed of submicron particles that can experience regional-to-global dispersion, and this study has shown that soot-EC concentrations have very small variation within the fourteen cities, the low char-EC/soot-EC in Jinchang can be attributed to the lower residential and industrial coal consumption from local sources (Fig. 3). Thus, even lower char-EC/soot-EC ratios can be expected in the extensive rural areas in China.

Compared with the char-EC/soot-EC ratio from primary motor vehicle exhaust, which is generally lower than 1.0 (Chow et al., 2004; Cao et al., 2006b), the summer char-EC/soot-EC ratio is still higher. This indicates that even in summer the industrial and residential EC emissions are still high (Fig. 3). The highest summer char-EC/soot-EC ratio occurs in Chongqing, reaching 6.75, which is consistent with the biomass burning occurring there all the year round. The other cities have char-EC/soot-EC ratios far lower than that in Chongqing, reflecting the decreased contributions from residential biomass and coal combustion. The lowest char-EC/soot-EC ratio occurs in Jinchang, with a value of 0.43, similar to the values from motor vehicle exhaust emissions (Chow et al., 2004; Cao et al., 2006b). This is consistent with the lowest industrial coal consumption among these cities (Fig. 3). In addition, Jinchang is beyond the impact of the summer Asian monsoon (An et al., 2000; Han et al., 2008), which limits pollutants from southeastern China to there.

4. Conclusion

This study is to re-analyze existing data on carbonaceous aerosols by further differentiating EC into char-EC and soot-EC. The spatial distribution and seasonal variation of char-EC and soot-EC from fourteen cities in China show good correlations with fuel consumption, as well as the influence of the Asian monsoon system and meteorological conditions. The char-EC/soot-EC ratio distribution indicates their association with source profiles. The differentiation between char-EC and soot-EC provides an opportunity for our better understanding of carbonaceous aerosols. The high differences in spatial and seasonal char-EC concentrations and the small variation in soot-EC concentrations from fourteen cities in Chinese may indicate that different from char, soot carbon may not be local carbon, but regional carbon. Controls on local coal and biomass burning emissions may be the most effective means of reducing char-EC and total EC concentrations in the atmosphere for local environmental management, while for soot-EC, control may need regional and worldwide cooperation due to its regional-to-global dispersion. Because of the limited sampling periods in the fourteen cities, bias may exist in the data. Extensive long-term observation data, extracted from previous carbonaceous aerosol studies measured with the TOR method, along with future

observations, would improve our understanding of char and soot behaviors.

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