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Black carbon relationships with emissions and meteorology in Xi'an, China

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ABSTRACT

Aerosol black carbon (BC) was measured every 5 min at Xi'an, China from September 2003 to August 2005. Daily BC concentrations ranged from 2 to 65 μg m^{-3}, averaging 14.7 ± 9.5 μg m^{-3} and displayed clear summer minima and winter maxima. BC typically peaked between 0800 and 1000 LST and again between 2000 and 2200 LST, corresponding with morning and evening traffic combined with nighttime residential cooking and heating. The nocturnal peak was especially evident in winter, when more domestic heating is used and pollutant-trapping surface-inversions form earlier than in summer. BC frequency distributions the most commonly occurring concentrations occurred between 5 and 10 μg m^{-3} in all four seasons. BC ranged from 1.6% and 15.6%, and averaged 8.3% of $PM_{2.5}$. A clear inverse relationship between BC and wind speed (WS) was found when WS was below 2.5 to 3.0 m s^{-1}, implying a local origin for BC. Mixed layer depths (MLDs) were shallower during BC episodes compared to cleaner conditions.
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1. Introduction

Black carbon (BC) is an important constituent of airborne particulate matter (PM) that is often emitted as product of incomplete combustion (Koelmans et al., 2006). The two most important BC sources are fossil fuel combustion and biomass burning (Penner et al., 1993; Cooke and Wilson, 1996). Interest in BC has grown because it strongly absorbs light, thereby degrading visibility (Watson, 2002) and altering the Earth's radiation balance (Penner et al., 1993; Jacobson, 2001). BC can heat the air, alter atmospheric stability, influence large-scale circulations, and affect cloud albedo by changing the hygroscopicity of cloud condensation nuclei (Liousse et al., 1996). BC-containing particles are also associated with adverse health effects (Pope and Dockery, 2006; Badarinath et al., 2007; Mauderly and Chow, 2008), may lower crop yields (Chameides et al., 1999), contaminate building materials (Ghedini et al., 2000), and adversely impact terrestrial and aquatic ecosystems (Forbes et al., 2006).

BC research has special importance in China because it is the world's largest BC emitter (Cooke et al., 1999), and BC regional climate effects have been observed (Menon et al., 2002). Even though megacity air pollution is an important environmental issue (Molina and Molina, 2004), there are only limited urban BC measurements from China's large population centers (e.g., He et al., 2001; Cao et al., 2003, 2005, 2006, 2007; Ye et al., 2003; Cheng et al., 2006; Guinot et al., 2006). In this study, two years of aethalometer BC measurements from an urban-scale (Chow et al., 2002) Xi'an site in northwest China are examined for daily, monthly, and seasonal variations that relate to source emissions and meteorology.

2. Experimental methods

Fig. 1 shows the sampling site location relative to downtown Xi'an, a city of >7 million and the largest in northwest China. Samples were taken from 13 September 2003 to 31 August 2005. $PM_{2.5}$ ($PM \leq 2.5$ μm in aerodynamic diameter) BC was measured continuously as 5-min averages by quartz-fiber filter tape transmission at an 880 nm wavelength with an aethalometer (Model AE-16, Magee Scientific Company, Berkley, CA, USA) (Hansen et al., 1984). The $PM_{2.5}$ cut-point

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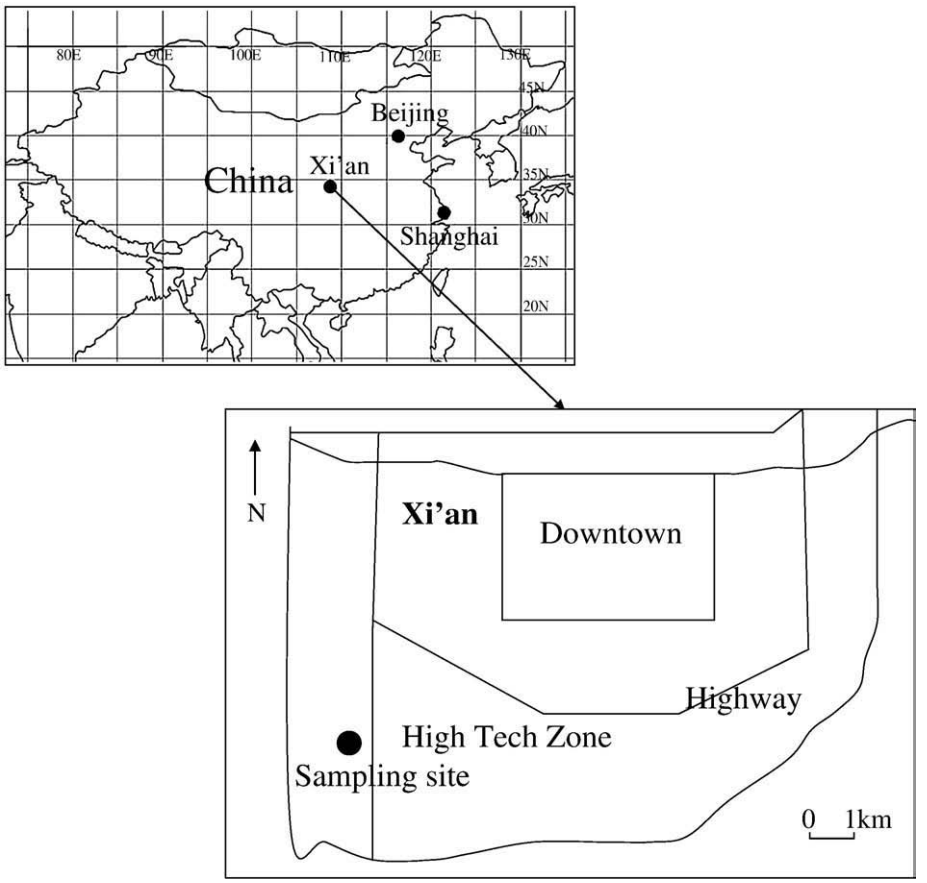

Fig. 1. Location of the IEE (Institute of Earth Environment, Chinese Academy of Sciences) sampling site (34.23°N, 108.88°E, 410 m above sea level) in the suburban High Tech Zone ~15 km southwest of downtown Xi'an (Cao et al., 2005). Samples were taken on the roof of a two-story building ~10 m above ground level, ~50 m west of a moderately travelled 4-lane round and ~25 m north of a lightly travelled 2-lane road.

was achieved with a 4 L min^{-1}, sharp-cut cyclone inlet (Kenny et al., 2000). The aethalometer was factory calibrated with ±2% accuracy (Hansen et al., 1984; Allen et al., 1999). Filter transmittance in inverse megameters (Mm^{-1}) was converted to concentration in μg m^{-3} using 16425 / λ or 16.6 m^2 g^{-1} when λ = 880 nm, which is the manufacturer's default derived from comparison with thermal elemental carbon (EC) measurements. This default varies by aerosol mixture (Arnott et al., 2003; Watson et al., 2005; Park et al., 2006), but the default was used in this study to retain consistency over the modeling period and with other comparison studies. No data were acquired from May to June 2004 due to instrument failure. The minimum detection limit of the aethalometer, defined as twice the standard deviation the baseline, is 5 ng m^{-3}. Only the BC values > 5 ng m^{-3} were used for this study.

Twenty-four hour average $PM_{2.5}$ filter samples were obtained from 1000 to 1000 LST at 5 L min^{-1} with mini-volume samplers (Airmetrics, Oregon, USA) (Cao et al., 2003) on quartz micro-fiber filters (QM/A™ 47 mm diameter, Whatman Ltd, Maidstone, UK). Filters were weighed before and after sampling using an electronic microbalance with a sensitivity of ±1 μg (MC5, Sartorius, Göttingen, Germany) to determine $PM_{2.5}$ mass concentrations. Filters were conditioned for at least 24 h in an enclosure at a temperature between 20 °C and 23 °C and relative humidity (RH) between 35% and 45%. These filters were later submitted for thermal/optical reflectance carbon analysis of organic and elemental carbon (OC and EC) and thermal carbon fractions (Chow et al., 1993), and comparability between aethalometer BC and filter-based EC will be addressed in a subsequent report.

Continuous hourly temperature and precipitation were obtained from a local weather station (15 km north of the sampling site). Wind speed (WS) was monitored within 20 m of the sampling site, 10 m above the ground level using an HFY-IA wind speed/wind direction instrument (Changchun Institute of Metrological Instruments, Changchun, Jilin Province, China). Mixed layer depths (MLDs) during the study period were calculated from the upper-air meteorological data based on the average value of UTC (Universal Time Coordinated) 16 and UTC 6 soundings archived by the U.S. National Oceanic and Atmospheric Administration (NOAA, http://www.arl.noaa.gov/ready/hysplit4.html).

3. Results and discussion

3.1. Temporal variations of BC concentrations

Daily median BC concentrations varied by >30-fold from 2 μg m^{-3} to 65 μg m^{-3} (Fig. 2), with much day-to-day

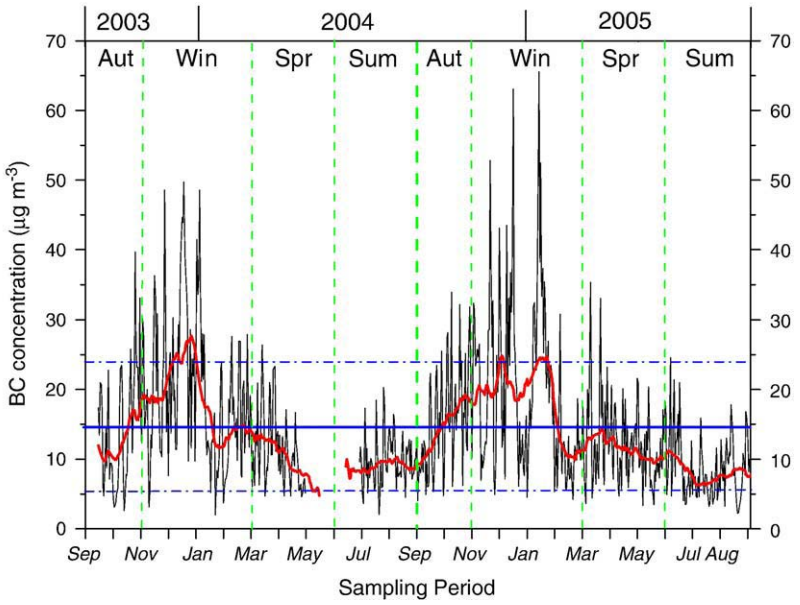

Fig. 2. Daily average 5-min BC concentration (thin line) and 30-day running average (thick line). Seasons are defined as: Winter — November to the following February; Spring — March to May, Summer — June to August; and Autumn — September to October. The 2-year BC average of 14.7 μg/m^3 is indicated by the solid horizontal line, with standard deviation of the 5-min averages indicated by the horizontal dotted lines.

variation. Average BC concentrations in winter (20.1 μg m^{-3} for 2003/2004 and 19.9 μg m^{-3} for 2004/2005) were twice those for summer (9.8 μg m^{-3} for 2003/2004 and 8.6 μg m^{-3} for 2004/2005). Average winter BC concentrations were 30% higher than the two-year average of 14.7 ± 9.5 μg m^{-3} while summer and spring averages were 40% and 18% lower than the two-year average, respectively.

Table 1 shows that monthly average BC values varied by nearly fourfold, from a low of 7 ± 3 μg m^{-3} (July 2005) to 27 ± 11 μg m^{-3} (December 2003). Monthly BC concentrations increased during autumn (12 ± 6 μg m^{-3}) and peaked during winter (27 ± 11 μg m^{-3} in December 2005 and 26 ± 13 μg m^{-3} in January 2005). Monthly relative standard deviations (RSD, standard deviation divided by the mean) were 3–10% higher during autumn and winter (0.41 to 0.64) compared to spring and summer (0.38 to 0.53).

Table 1
Monthly average and standard deviations for Xi'an BC concentrations, September 2003–August 2005 in Xi'an.

Monthly average	2003 BC, μg m^{-3}	2004 BC, μg m^{-3}	2005 BC, μg m^{-3}
Jan		18.2 ± 11.5	26.3 ± 13.0
Feb		15.3 ± 7.0	11.1 ± 5.3
Mar		14.0 ± 6.2	14.5 ± 7.7
Apr		9.0 ± 3.4	12.6 ± 5.0
May		N.A.[a]	10.1 ± 4.4
Jun		N.A.	9.8 ± 5.0
Jul		9.3 ± 4.4	7.0 ± 3.1
Aug		10.3 ± 4.8	9.1 ± 4.3
Sep	11.8 ± 6.1	12.1 ± 6.1	
Oct	16.1 ± 9.4	18.6 ± 7.6	
Nov	19.8 ± 10	21.8 ± 11.4	
Dec	27.1 ± 11.2	20.3 ± 12.9	

[a] Missing data due to instrument failure.

Coal-burning is China's major energy source. During winter (November to February), Xi'an's nighttime temperature lows range from ~−5 °C to ~3 °C, and indoor heating is accomplished with small coal-burning boilers and stoves (Streets et al., 2001; Cao et al., 2005). Annual energy consumption is ~7.2 × 10^6 MT of standard coal equivalent (TCE), of which 34.8% derives from coal burning, 13.7% from oil burning, 43.9% from electricity (often from centralized coal-fired power stations), and 7.6% from natural gas and other fuels (Xi'an Clean Energy Office, 2002). Winter heating (1.5 × 10^6 TCE coal) accounts for 58% of annual coal burned in Xi'an, with the remaining coal used for electrical generation. Biomass burning is also a large carbon emitter on the agriculturally-productive Guangzhou Plain. Crops are harvested during early summer and mid-autumn when crop residues are cleared by open burning (Cao et al., 2005). China's gasoline and diesel engine exhaust contains a large BC content (Cao et al., 2006) and is emitted all year. Asian and other fugitive dust, some of which also absorbs light (Chow et al., 2000; Shen et al., 2006), may contribute during the spring and early summer dry seasons.

Table 2 compares average aethalometer BC concentrations from Xi'an with levels from other cities. Xi'an's two-year average BC concentrations were twice those from Sao Paulo, Brazil, more than three times those at Pune and Bangalore, India and Mexico City, Mexico, ten times those at Helsinki, Finland and Fresno, USA, and >20 times those at remote Réunion Island in the South Indian Ocean. Kanpur, India had average BC levels similar to those in Xi'an. The roadside site in Paris, France also had comparable BC levels. Only Lahore, Pakistan seems to have higher average BC levels.

Fig. 3 shows that hourly BC concentrations follow a diurnal cycle that has been seen elsewhere (Babu and Moorthy, 2002; Bhugwant et al., 2000; Chen et al., 2001; Madhavi Latha et al.,

Table 2
BC concentrations by 880 nm aethalometer light transmission for different urban locations.

Location	Observation period	BC, μg m^{-3}	Reference
Xi'an, China	Sept. 2003–Aug. 2005	14.7 ± 9.5	This study
Pune, India	Jan. –Dec. 2005	4.1	Safai et al. (2007)
Kanpur, India	Dec. 2004	6.0–20.0	Tripathi et al. (2005)
Bangalore, India	Nov. 2001	4.2	Babu and Moorthy, 2002
Helsinki, Finland	Nov. 1996–Jun. 1997	1.38	Pakkanen et al. (2000)
Lahore, Pakistan	Nov. 2005–Jan. 2006	21.7	Husain et al. (2007)
Fresno, USA	Jan. –Dec. 2000	1.28 ± 1.06	Watson and Chow (2002)
Paris, France	Aug. –Oct. 1997	14 ± 7	Ruellan and Cachier (2001)
Réunion Island, South Indian Ocean	1996–1998	0.27–0.65	Bhugwant et al. (2000)
Sao Paulo, Brazil	Jun.–Sept., 1997	7.6	Castanho and Artaxo (2001)
Mexico City, Mexico	Feb. 2–Mar. 3, 1997	4.0	Watson and Chow (2001)

2004; Watson and Chow, 2002), with a gradual build-up starting at 0600 LST and peaking around 0800 to 0900 LST, an afternoon minimum and an evening increase with maxima occurring between 2100 and 2300 LST. Maximum hourly BC concentrations during the evening were ~17, 11, 21, and 26 μg m^{-3}, and during the morning were ~15, 12, 17, and 24 μg m^{-3} for spring, summer, autumn and winter, respectively. These levels are more than twice those measured during the afternoon (Fig. 3).

These patterns can be explained by a combination of emissions and meteorology. The gradual buildup of morning BC corresponds with increases of morning traffic during work commutes. The surface inversion couples to layers aloft a few hours after sunrise, resulting in vertical mixing of primary pollutants with a subsequent decrease. Wind speeds also increase with this mixing, thereby facilitating dispersion and dilution. After 1600, the evening rush hour commences as does cooking and residential heating during winter. Soon after sunset, the surface inversion begins to form, trapping more primary pollutants related to BC. The earlier onset of the morning buildup and the later onset of the evening accumulation in summer than in winter correspond with the earlier sunrises and later sunsets in summer.

Fig. 4 shows that hourly BC distributions are skewed toward lower values during all seasons, with most BC levels at 5 to 10 μg m^{-3}. During summer, hourly BC levels were <10 μg m^{-3} for more than 67% of samples, but during winter 67% of the BC levels were <20 μg m^{-3}, and nearly 17% of the values exceeded 30 μg m^{-3}. During autumn, BC concentrations occurred in all of the concentration segments from <5 to >60 μg m^{-3}, but there were fewer hourly BC concentrations with >60 μg m^{-3} in winter. A similar pattern is seen for spring, but all BC levels are <55 μg m^{-3}.

3.2. Relationship of BC between $PM_{2.5}$

BC fractions of 24-hour (1000 to 1000 LST) $PM_{2.5}$ mass are shown in Fig. 5. The average of BC/$PM_{2.5}$ ratios is 8.3%, ranging from 1.6% and 15.6% for individual $PM_{2.5}$ samples. Autumn had the highest average ratio of 10%, with the minimum of the $PM_{2.5}$ mass, whereas the lowest BC/$PM_{2.5}$ fraction (6.9%) was found in winter.

Tripathi et al. (2005) reported BC as 7 to 15% of total suspended particulate (TSP) at Kanpur, India, Madhavi Latha and Badarinath (2005) reported BC as 7% of TSP at Hyderabad, India, and Safai et al. (2007) reported BC as 2.3% of TSP for Pune, India. In suburban regions of Europe and North America, BC contributes about 5% of TSP (Ramanathan and Crutzen, 2003). TSP contains more geological material than $PM_{2.5}$, so these lower BC fractions are expected. Venkatachari et al. (2006) reported higher BC fractions, 13% and 11% of $PM_{2.5}$ mass at two sites in New York City.

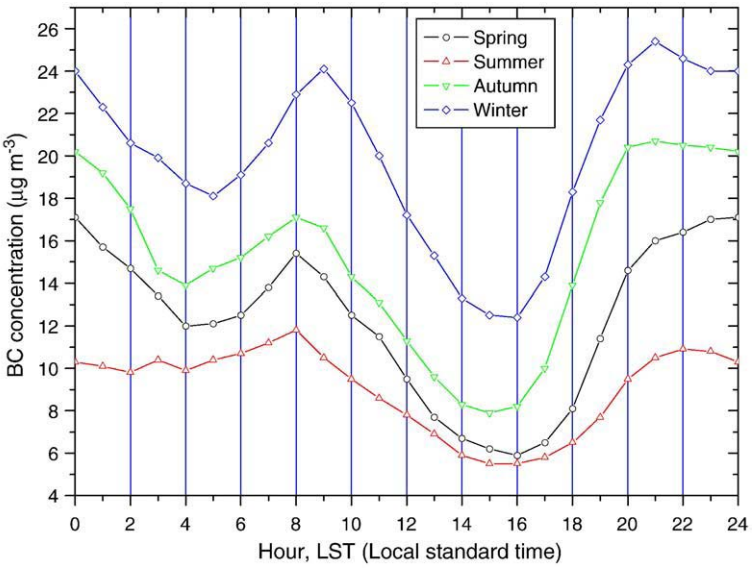

Fig. 3. Hourly average BC changes throughout the day for each season, with seasons defined in Fig. 2.

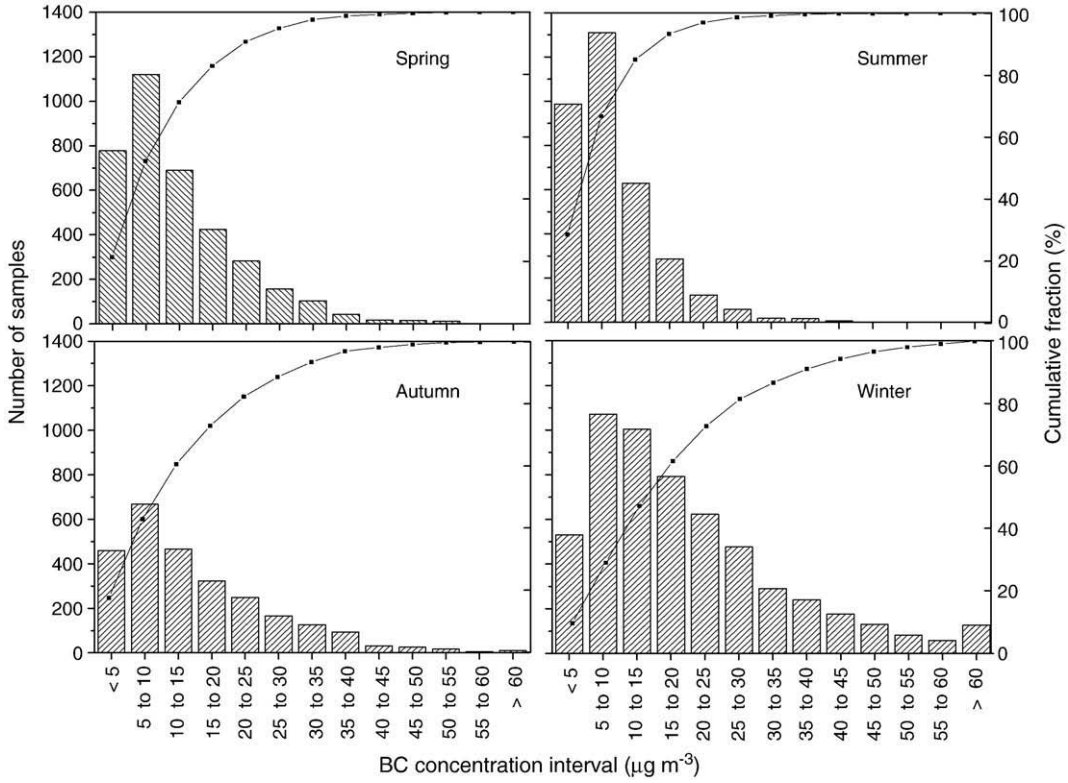

Fig. 4. Frequency distributions of 1-hour average BC from September 2003 to August 2005. The curves refer to the cumulative fractions of BC numbers in each concentration segment.

The BC abundance in $PM_{2.5}$ is affected by the mixture of pollution sources and meteorological conditions. At Xi'an, Asian and other fugitive dust during spring (Zhang et al., 2002) increases $PM_{2.5}$ with non-carbonaceous material, thereby decreasing the $BC/PM_{2.5}$ ratio. Higher $BC/PM_{2.5}$ ratios in autumn may be influenced by biomass burning (Cao et al., 2005).

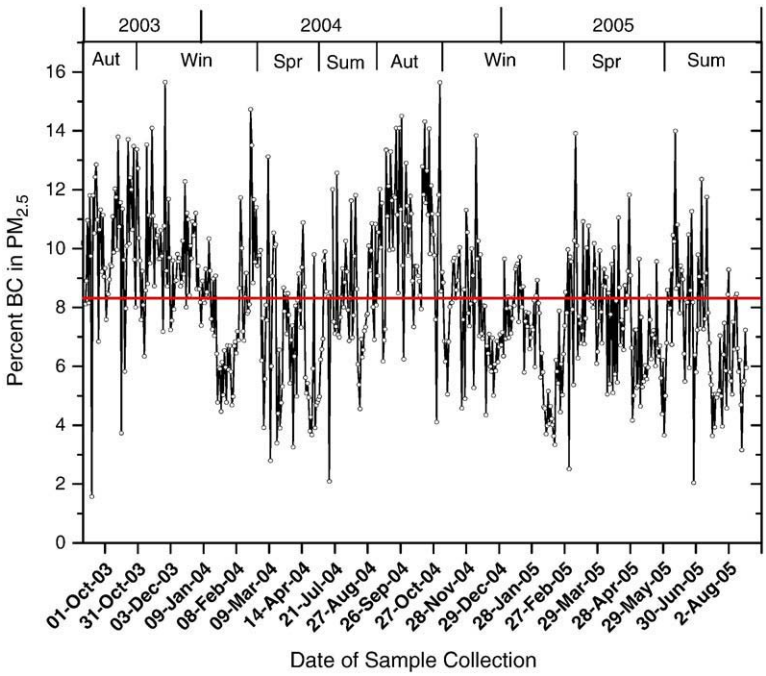

Fig. 5. Fraction of 24-hour average BC in 24-hour average $PM_{2.5}$ mass (1000–1000 LST). The horizontal line represents the average BC for the entire monitoring period, excluding May and June 2004 when the aethalometer malfunctioned.

3.3. Relationships to meteorological conditions

Fig. 6 compares time-series of BC with meteorological measurements. Low wind speeds in autumn and winter (0.5 to 1.5 m s^{-1}) and higher wind speeds during spring and summer (~2 m s^{-1}) are inversely related with BC ($r=-0.67$). A suburban and an urban site in Canada showed negative BC correlations with wind speeds, but this was not found at a rural location (Sharma et al., 2002). Ramachandran and Rajesh (2007) also reported a negative correlation ($r=-0.54$) between BC and wind speed at urban sites in Ahmedabad, India. This is consistent with important contributions from locally-generated BC which can accumulate under low wind speed conditions. This is examined in Fig. 7 for the morning rush-hour period which shows high BC traffic contributions at wind speeds less than 2.5–3.0 m s^{-1} (Fig. 7), i.e., a clear inverse trend between BC concentrations and WS can be found only when WS was less than 2.5 to 3.0 m s^{-1}.

Reddy and Venkataraman, (1999) suggest that BC residence times vary as a function of precipitation, from roughly 7 to 10 days during dry conditions to about 5 days or less during wet periods. As shown in Fig. 6, monthly rainfall in Xi'an mainly occurs in summer and autumn, but monthly BC in Xi'an is not strongly related to precipitation levels ($r=-0.35$), comparable to similar correlation ($r=-0.35$) at Ahmedabad, in western India (Ramachandran and Rajesh, 2007). However, monthly averaged BC at Trivandrum, India was negatively related to rainfall ($r=-0.74$) (Babu and Moorthy, 2002).

The number of days with precipitation during a given month may be a better determinant BC scavenging than total monthly precipitation. During September 2004, from Sept. 2 to Sept. 5, with a mean precipitation of 5.4 mm, the daily average BC concentration was 6.9 μg m^{-3}; from Sept. 13 to Sept. 14, as the mean precipitation increased to 6.6 mm, the daily average BC concentration decreased to 6.1 μg m^{-3}; on Sept. 19, precipitation of 19.5 mm compared to average BC concentration of 5.3 μg m^{-3}; on Sept. 30 precipitation reached 34.5 mm and the daily average BC concentration was 4.7 μg m^{-3}. The 24 h average BC concentration on rainless days was 14.9 μg m^{-3}, 2.6 times higher than for days with measurable precipitation (5.8 μg m^{-3}).

Monthly average temperatures ranged from 0 to 28.0 °C, and an inverse relationship between temperature and BC can

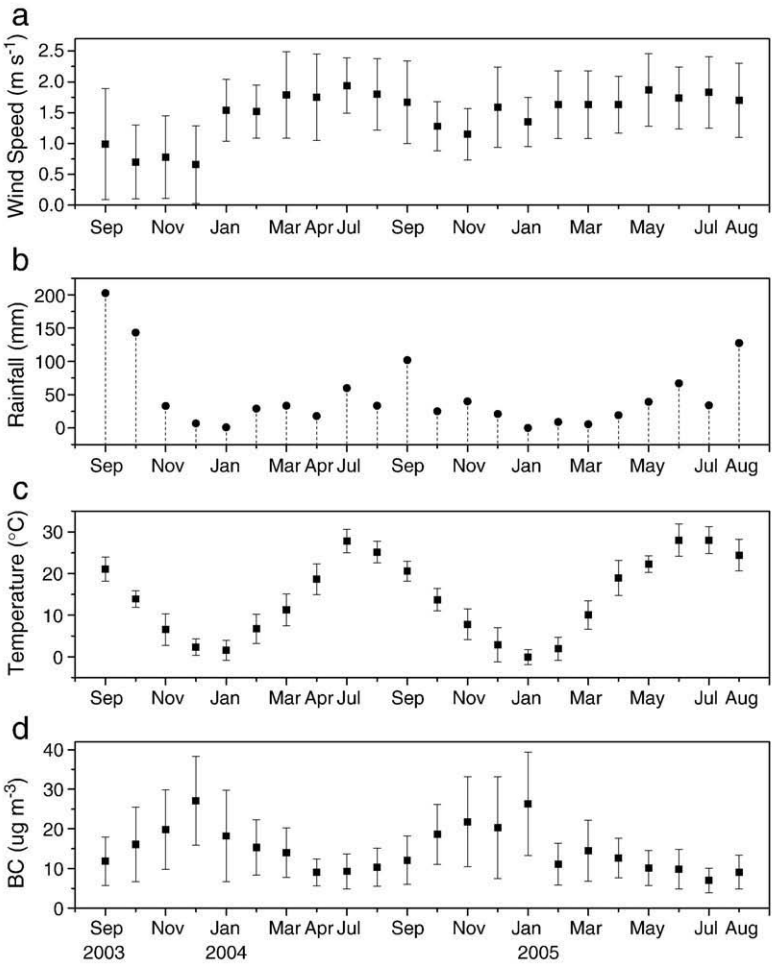

Fig. 6. Monthly average (a) wind speed (WS), (b) rainfall, (c) ambient temperature, and (d) BC concentrations in Xi'an. Vertical bars indicate ±1 standard deviation of the average. Aethalometer data for May and June 2004 are not available.

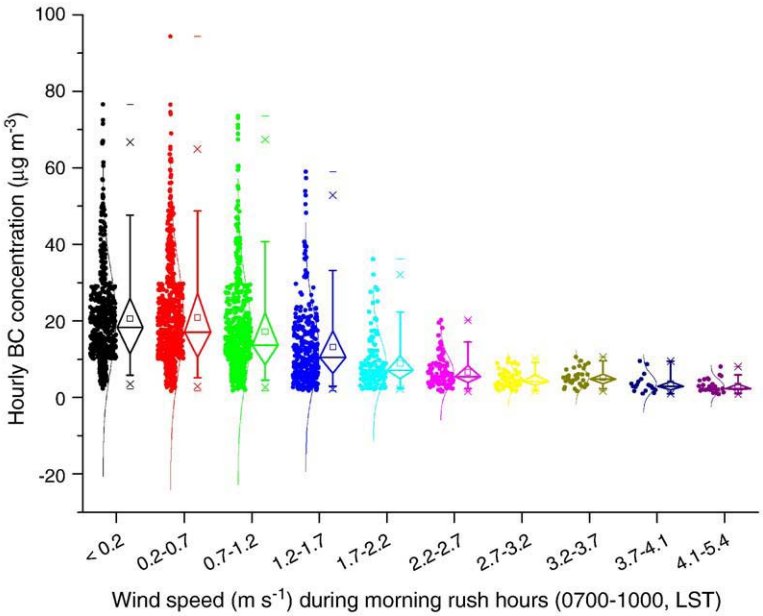

Fig. 7. Three-hour average (0700 to 1000 LST) BC concentrations (μg m^{-3} grouped by wind speed (m s-1) intervals. The time-period corresponds to Xi'an's morning rush hour. A normal distribution is fit to the measurements within each bin and 1st, 25th, 50th, 75th, 99th and maximum percentiles.

be seen in Fig. 6 ($r=-0.80$). This is consistent with the expected increase in residential coal burning during low temperatures, as well as with the more stable atmosphere. Cao et al. (2005) showed that residential coal-combustion contributed ~44% of the total carbon in Xi'an during winter.

Mixed layer depths determine the volume through which surface-emitted pollutants can be diluted and reflect boundary layer turbulence (Stull, 1988). The surface boundary layer is shallow over Xi'an during winter, and this results in pollutant trapping. With increases in surface temperatures and convective activity during summer, pollutants are dispersed as the boundary layer deepens, thus lowering BC concentrations.

"BC events" are defined as periods when the daily BC concentrations exceed the seasonal geometric mean concentration plus one geometric standard deviation of the mean. These days were also characterized by high-levels of other pollutants. "Clean days" were defined as those for which BC daily concentrations are less than the seasonal geometric mean concentration minus one standard deviation. Fig. 8 shows that MLDs were lower (773 m) during BC events (28.3 μg m^{-3}) than for clean days (5.4 μg m^{-3}) with MLDs of

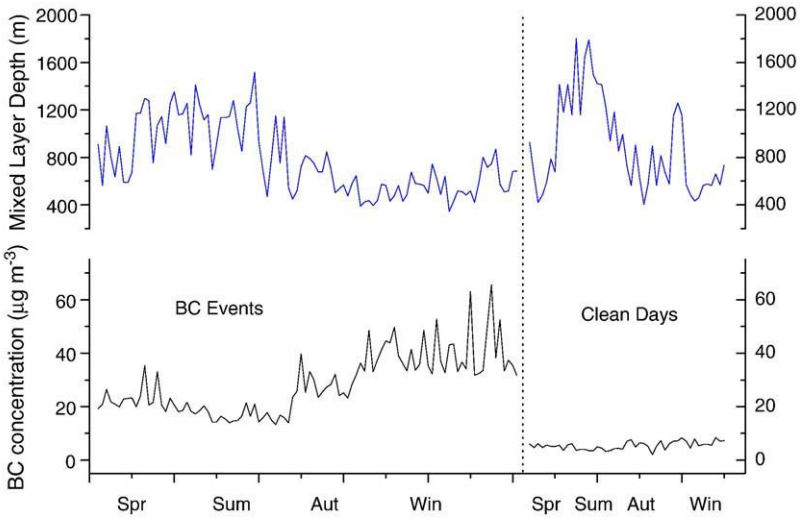

Fig. 8. BC concentrations and mixed layer depth (MLD) for BC events and clean days. "BC events" are defined as periods when the daily BC concentrations exceed the seasonal geometric mean concentration plus one geometric standard deviation of the mean. "Clean days" were defined as those for which BC daily concentrations are less than the seasonal geometric mean concentration minus one standard deviation. All the events and clean days were plotted together at each season.

897 m. This was the case for all seasons, although the MLDs were shallower in winter than during other seasons. For the BC events versus clean days, the respective seasonal mean MLDs were 980 m versus 1026 m in spring; 1026 m versus 1327 m in summer; 642 m versus 705 m in autumn; and 551 m versus 700 m in winter. Pollutant dispersion is weaker during BC events; in agreement with observations made in a coastal area of south China (Cheng et al., 2006).

4. Conclusions

Over two years of continuous $PM_{2.5}$ BC monitoring in Xi'an, daily average BC concentrations averaged 14.7 ± 9.5 μg m^{-3} and ranged from 2 μg m^{-3} to 65 μg m^{-3}. Summer minima and winter maxima can be explained by changes in emission sources and variability in meteorological conditions, especially increased domestic heating with coal, lower wind speeds, and shallower mixed layer depths in winter. Diurnal distributions showed BC concentration peaks occurring from 0800 to 1000 LST and 2000 to 2200 LST. The first peak corresponds to morning traffic while the second peak results from evening traffic combined with nighttime residential cooking and heating. On average, daily BC concentrations accounted for 8.3% of the $PM_{2.5}$ mass, with a range of 1.6% and 15.6%. A clear inverse relationship between BC concentrations and wind speeds was found for winds below 2.5 to 3.0 m s^{-1}, consistent with much of the BC originating from accumulation of local emissions. Mixed layer depths were shallower during BC events and deeper on clean days.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.atmosres.2009.05.009.

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