Black Carbon Particles in the Urban Atmosphere in Beijing

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ABSTRACT

A study of the concentration of black carbon particles and its variation in the urban atmosphere has been carried out since 1996 in the Beijing area. The measurements were done in the late autumn and early winter each year, the period before and after domestic heating activities begin. The results show the presence of black carbon particles at the high level that vary over a large range in the urban atmosphere in Beijing. The mean value of daily average concentration for the whole observation period of 1996–2004 is 20.0 μ g m⁻³. An evident decrease of black carbon particle concentration in the Beijing area is observed after 2000, and the daily average concentration of black carbon particles is estimated to be 16.0 μ g m⁻³ with a variation range of 2.10–50.50 μ g m⁻³ for the period of 2000–2004. The observation method and main variation behavior characteristics of black carbon particles in the urban atmosphere in the Beijing area are given and discussed.

Key words: black carbon particles, urban atmosphere, aerosols, Beijing

1. Introduction

The presence of black carbon (BC) particles in the atmosphere does not only strongly pollute the atmospheric environment or affect impair human health, but it also affects some important physical, chemical and photochemical processes in the atmosphere. This is the reason why BC in the atmosphere has become a special important issue in the study on climate changes and on air quality and human health (IPCC, 2001; Jacobson, 2001; Menon et al., 2002; Chylek et al., 1995; Zhang and Gao, 1997). The BC in the atmosphere originates mainly from incomplete combustion processes of all fuels containing carbon. In the urban atmosphere, the most significant source of BC is the combustion of fossil fuels used for industrial and traffic activities, and since in the cities, the majority of combustion activities and population are concentrated, cities are the focus of study of black carbon particles. Beijing, as one of the mega-cities in the world, is now in rapid economic development with a sharp increasing energy consumption. For example, the coal consumption in Beijing used both industrially and domestically is now near 15×10^8 t per year, and is still growing at a rate of about 3.8% per year, and the vehicle population is growing rapidly and exceeded 2.0 million in 2004. Some investigators have estimated the emission of BC in China based on energy consumption and emission factors (Streets et al., 2001; Chameides and Bergin, 2002), but due to the difference in combustion processes and emission sources, the characteristics of airborne particles in the atmosphere in Beijing should be different from that in mega-cities of Asia and of western countries (Chow et al., 2000; Shi et al., 2001; Zhang and Friedlander, 2000). Therefore, there is an urgent need for reliable data on black carbon particles and their variation behavior in the urban atmosphere in Beijing.

Some studies on BC in Beijing have been done since the late 1980s, but up to now, no BC data have been available for a long term in Beijing. Some results can be found, for example, in Su et al. (1989), Zhang and Su (1986), Dod et al. (1986), Zhang and Friedlander (2000), Shi et al. (2002), He et al. (2001), Wang et al. (2002). The common finding from these studies is the relatively high level and a wide range of variability of black carbon concentration in the atmosphere in Beijing.

The purpose of this paper is to analyze BC data obtained during the period of 1996–2004, to summarize the major findings and to discuss the variation characteristics of BC concentration in the atmosphere in Beijing.

2. Measurement and data

For BC measurement in the atmosphere, various techniques have been developed based on chem-

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ical, optical, spectroscopic and microscopic methods (Reid et al., 1998; Cachier et al., 1989; Chow et al., 1993). Among them, the optical method has been used widely. The unique property of BC in comparison with other particles in the atmosphere is its larger optical absorption, and this has been used for BC measurement. Based on the absorption property, various optical instruments have been manufactured for practical use, such as the aethalometer, which is widely used for automatic real-time measurement at the present time (Hensen et al., 1984; Hansen and McMurry, 1990; Allen et al., 1999). It should be pointed out that up to now measurement methods for BC have not been satisfactory due to the uncertainties associated with our poor knowledge of the structure, external patterns and optical properties of black carbon particles (Horvath, 1993; Ruoss et al., 1993; Reid et al., 1998). The data obtained by using an air sampling method are used in this work. In this case, the BC is collected on a fibre-quartz filter, then BC concentrations are determined by measuring the attenuation of a light beam through the filter in the visible spectral region in the laboratory. Air samples were taken by using a special air sampling collector at a flow rate of 500 ml per minute, continuously day and night, and each sample was matched for 3–6 hours depending on weather conditions. The method used is well known as the integrating plate (IP) method (Lin et al., 1973). This is an optical method, the principle of which is based on the determination of the absorption coefficient of the aerosol $\tau_{\rm a}$ by the following expression:

$$\sigma_{\rm a} = \ln(I_0/I) = \alpha_{\rm bc} \cdot C_{\rm bc} ,$$

where $C_{\rm bc}$ is the mass concentration of BC, and $\alpha_{\rm bc}$ is a factor determined by the mass absorption efficiency of BC. For this purpose, an optical analyzer was used to determine the ratio of the light intensity through the filter with BC loading I and the filter without BC loading I_0 , i.e., $\sigma_a = \ln(I_0/I)$. In our case, an analyzer with a double light beam technique was used to eliminate the influence of variations in filter substrates and the optical apparatus on the $\sigma_{\rm a}$. Due to the dependence of α_{bc} on the physical and chemical properties and the morphological structure of BC, the method was calibrated in the laboratory before use by experimental detection of $\alpha_{\rm bc}$ showing a dependence of BC concentration on the light absorption coefficient at a selected wavelength interval (Kopeikin, 1991; Kopeikin et al., 1998).

The concentration of BC in the atmosphere was measured by the above method during 1996–2004 in Beijing at the top of a building at the Institute of Atmospheric Physics (IAP), Chinese Academy Sciences, which is located in the northern part of Beijing city (39°48'N, 116°28'E). In order to evaluate the impact of human activities on the variation characteristics of BC, measurements were conducted in the period of late autumn and early winter each year, the time before and after domestic heating starts in Beijing. During the measurement period, the main meteorological variables such as air temperature, wind speed and direction, and humidity, were also measured up to 300 m in height using the IAP meteorological tower (Wang et al., 2002).

As many authors have indicated, black carbon in the atmosphere is comprised mainly of fine particles (Ruellan and Cachier, 2001; Hitzenberger and Tohno, 2001), so the relationship between BC and the presence of sub-micron aerosols determined in parallel by a nephelometer is discussed in order to evaluate the possible source of BC in the atmosphere in Beijing. The nephelometer operated at a wavelength of 0.55 μ m and was calibrated for the concentration of particles in the size range of 0.3–1.0 μ m (Wang et al., 2001).

3. Some results and discussion

3.1 General variation characteristics

Some results obtained on the concentration of BC in the atmosphere in Beijing are summarized in Table 1, where the daily average refers to the BC concentration averaged for all days in the time period of measurements in each year.

As shown in Table 1, the concentration of BC in the atmosphere in Beijing changes over quite a large range. The daily average value of BC for the whole measurement period of 1996–2004 is estimated as $20.0 \ \mu g \ m^{-3}$ with a maximum value of $29.2 \ \mu g \ m^{-3}$ in 1999. The daily average concentration varies from year to year and ranges from 2.10 $\ \mu g \ m^{-3}$ to 96.70 $\ \mu g \ m^{-3}$ during the last 9 years. It should be pointed out that the concentration of black carbon particles in the atmosphere increases rapidly in November each year as the central heating is turned on in many districts in Beijing. The analysis of all data obtained from 1996 to 2004 shows an evident decrease of BC concentration in the atmosphere in the Beijing area since 2000. Before 2000, the concentration of BC in the atmosphere is at relatively

Table 1. BC concentration in the atmosphere for 1996–2004 (units: $\mu g m^{-3}$).

Year Time period	Daily average	Range of daily average		
1996 29 Oct–20 Nov	20.4	4.90-43.00		
1997 25 Oct–24 Nov	25.2	2.80 - 45.80		
1998 22 Oct–16 Nov	24.9	4.10 - 72.50		
1999 20 Oct–29 Nov	29.2	3.40 - 96.74		
2000 20 Oct–17 Nov	15.6	4.20 - 50.50		
2001 23 Oct–19 Nov	16.2	4.50 - 36.10		
2002 4 Nov–1 Dec	15.5	2.10 - 37.90		
2003 3 Nov–30 Nov	16.7	2.40 - 37.90		
2004 23 Oct–15 Nov	15.6	3.30 - 32.0		

higher level with the highest daily average of 96.7 μ g m⁻³ in 1999. On average, the daily average is around 25 μ g m⁻³ for the whole observation period of 1996–1999, while for 2000–2004, the daily average is about 16.0 μ g m⁻³, and no clear annual variation of the daily average is found for the last 5 years.

Such an abrupt change in daily average BC concentration before and after 2000 reflects the evident improvement of air quality in Beijing after 2000. This conclusion is consistent with that drawn from other sources. For example, according to the air quality data reported by the Beijing Environmental Protection Agency (BJEPA), the daily average concentration of PM₁₀ was 180 μ g m⁻³ in 1999 and 162 μ g m⁻³ in 2000, whereas, the daily average concentration of SO₂ was 80 μ g m⁻³ and 71 μ g m⁻³ for 1999 and 2000 respectively.

Concentrations of some pollution species showing the annual variation of air quality in the atmosphere in Beijing are given in Table 2, where the data for PM_{10} and SO_2 are taken from the annual report of BJEPA, and data for $PM_{1.0}$ are the results of the present work (for time period of measurement each year). All data in Table 1 and Table 2 show a clear abrupt change in the daily averages of PM_{10} , $PM_{1.0}$, SO_2 and BC in Beijing between 1999 and 2000.

Although the clean air days (days with daily averaged PM_{10} mass concentration less than 150 μ g m⁻³, i.e., having an Air Pollution Index less than 200, the Class II National Air Quality Standard of China (GB3059–1996)) increase in number from year to year (from 100 days in 1998 to 229 days in 2004), no distinct changes in the concentrations of PM_{10} , SO₂ and BC are observed after 2000. As for $PM_{1.0}$, even a slight increase of its concentration is observed in the last two years (see Table 2).

Another important point regarding the general variation characteristics of BC is that concentration of BC in the atmosphere is closely related to that of $PM_{1.0}$. To verify such a relation, the correlation between $PM_{1.0}$ and BC is given in Fig. 1. The data for PM_{10} and BC are given for the time period of 1996–1999 and for the time period of 2000–2004 separately in consideration that the former time period is a period with relative heavy air pollution. As can be seen, the correlation coefficient between $PM_{1.0}$ and BC is as high as 0.92 for both relatively heavy and light air pollution situations. The only difference is that the ratio

of BC to $PM_{1.0}$ decreases evidently for the relatively light air pollution situation. Such a difference implies a decrease of BC content in the fine particles due to the strong measures taken by the Beijing government to control air pollution since 1999. The decrease of BC in $PM_{1.0}$ might have resulted from the gradual replacement of coal by gas for domestic heating and the use of low-sulphur and low-ash coal in Beijing after 2000.

According to the BJEPA data, the most heavy air pollution cases in Beijing occurred in 1997–1999. For this reason, urgent control measures to counteract air pollution were taken after 1998 by the Beijing government, and as a result, the concentrations of the main pollution species, such as SO₂, NO_x, and PM₁₀, have dropped, and the air quality in Beijing has evidently improved since 2000 (see Table 2).

The BC concentration value of 16.0 $\mu g m^{-3}$ in Beijing for late autumn and early winter in this study is comparable with the value 16.4 $\mu g m^{-3}$ (Chen et al., 1994), but higher than those measured in Beijing by others, such as the value of 10.7 $\mu g m^{-3}$ for autumn and winter (He et al., 2001), 11.4 μ g m⁻³ in winter (Lou et al., 2005), and higher than those measured in other cities, for example, 14.8 $\mu g m^{-3}$ for autumn in Paris, France (Ruellan and Cachier, 2001), 12.8 μ g m^{-3} for winter in Vienna, Austria (Hitzenberger and Tohno, 2001), 8.4 $\mu g m^{-3}$ for winter in Seoul, Korea (Park et al., 2001), $8.5 \,\mu \text{g m}^{-3}$ for autumn in Los Angeles, USA (Chow et al., 1994), $10.4 \,\mu \text{g m}^{-3}$ for winter in Guangzhou, China (Cao et al., 2003), and 5.1 $\mu g m^{-3}$ for winter in Hong Kong, China (Cao et al., 2003). It should be noted out that only a few long-term (at least one year) datasets on BC in the atmosphere are available. In addition, there is a large temporal and spatial variability in BC in the atmosphere. So comparing BC data measured by different authors at various places should be made very carefully, since a large difference in the BC concentrations might result from measurements made in different locations even for a given city. For example, the BC concentrations were 4.7 $\mu g m^{-3}$ and 8.0 $\mu g m^{-3}$ for a territory-wide and for a roadside station in Hong Kong, respectively (Yu et al., 2004), and were 8.7 $\mu g m^{-3}$ and 10.1 $\mu g m^{-3}$ for downtown Beijing and Tsinghua University in Northwest in Beijing respectively (He et al., 2001).

Table 2. Daily averages of PM_{10} , SO_2 and $PM_{1.0}$ in Beijing (1996–2004), units: $\mu g m^{-3}$.

Species	1996	1997	1998	1999	2000	2001	2002	2003	2004
PM_{10}^*	_	_	_	180	162	165	166	141	149
SO_2	100	125	120	80	71	64	67	61	65
$PM_{1.0}$	235	290	292	349	192	222	189	234	288

* There is no data for PM_{10} before 1999.

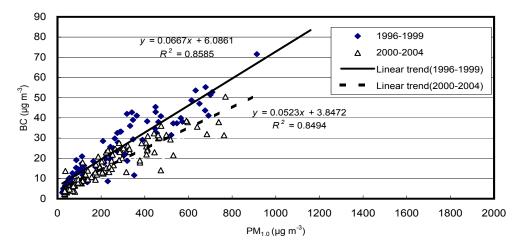


Fig. 1. Scatter plot showing the relationship between $PM_{1.0}$ and BC concentration for 1996–1999 and 2000–2004.

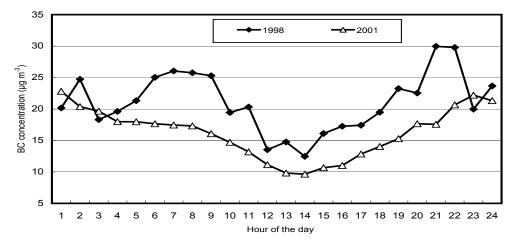


Fig. 2. Diurnal variation of hourly average BC concentration in the atmosphere for 1998 and 2001.

3.2 Diurnal variation

In most cases, the concentration of BC in the atmosphere exhibits an evident variation during the daytime. A detailed analysis shows that the diurnal variation is more distinct in days with a high concentration of BC. In that case, the absolute amplitude of the diurnal variations is as much as $23-30 \ \mu \text{g m}^{-3}$ and about 70 $\ \mu \text{g m}^{-3}$ in the extreme cases. For example, the diurnal variations of the hourly average concentrations of BC in the atmosphere averaged for all observation days in 1998 and 2001 are shown in Fig. 2,. These two years are presented as typical cases for a relatively heavy air pollution year and a relatively light air pollution year, respectively.

Figure 2 shows that a diurnal variation with a clear double-peak behavior is observed in 1998. The higher concentrations of BC are observed typically during 0700–1000 LST and 2000–2400 LST, while the lower concentrations appear at 1300–1600 LST during the day. The main factors affecting the diurnal variation of BC in the atmosphere are meteorological conditions and human activities. The high concentrations at 700–1000 LST are mainly caused by human activities (e.g., the increase of vehicular traffic), while the high concentrations at 2000–2400 LST are mainly from the presence of an inversion structure in the boundary layer. The increased convection and raised mixing layer height can be considered as the main reasons for the lower concentration at 1300–1600 LST during the day. In contrast with 1998, a more typical diurnal variation of hourly average BC concentration with a maximum value at night and with a minimum value at midday is observed in 2001. It should be pointed out that due to the dependence on meteorological conditions, the concentration of BC in the atmosphere could vary over a large range even during the daytime. For

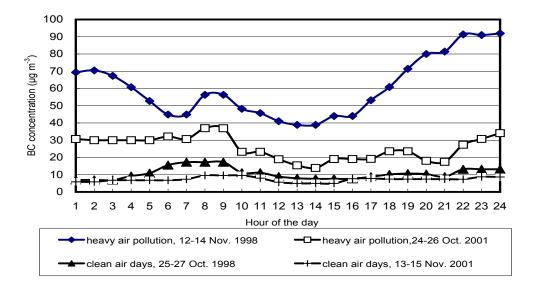


Fig. 3. Diurnal variations of BC under different weather conditions.

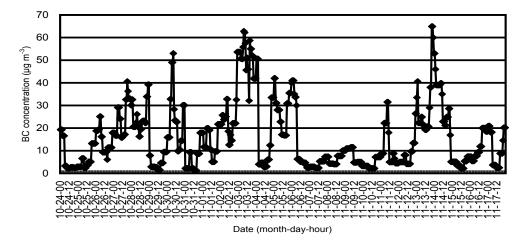


Fig. 4. Example of daily variation of BC concentration in the atmosphere (24 October–17 November 2000).

example, diurnal variations of the hourly average BC concentrations for different weather conditions (favorable and unfavorable for diffusion of air pollution species) are given in Fig. 3. The two top curves in Fig.3 represent heavy air pollution days (days with an Air Pollution Index more than 300) during 12-14 November 1998 and 24 October-26 November 2001, respectively, and the two bottom curves represent clean air days during 25-27 October 1998 and 13-15 November 2001, respectively. Each curve in Fig. 3 is averaged for three successive days. The following conclusions may be drawn from Fig. 3, namely unfavorable meteorological conditions cause heavy air pollution and higher concentrations of BC in Beijing, where the distinct diurnal variation of BC with the maximum at night and the minimum in the daytime is observed, while under favorable meteorological conditions, the pollution

species spread much easier, thus the BC concentration was lower and varies more smoothly during the day and night. In comparison to 1998, the lower average BC concentration in the atmosphere in Beijing and smaller diurnal variation amplitudes are observed in all weather conditions in 2001.

3.3 Daily variation

Due to changes in meteorological conditions and human activities, the concentration of BC might vary strongly from day to day. As can be seen from Table 1, for all observation periods of 1996–2004, the daily average BC concentration ranges from 2.10 μ g m⁻³ to 4.9 μ g m⁻³ for clean air days and from 32.0 μ g m⁻³ to 96.7 μ g m⁻³ for heavy air pollution days. Moreover, an obvious decrease in the highest daily BC concentration is observed after 2000. The highest daily average

BC concentration for 2000–2004 is $38.9 \ \mu g m^{-3}$, which is about 41% of that for 1996–1999. The daily variation of hourly average BC concentration for each date during the period of observation in 2000 is given in Fig. 4 as an example. For this period, the daily average BC varies in the range of $4.20-50.50 \ \mu g m^{-3}$, while the hourly average ranges from $1.30 \ \mu g m^{-3}$ to $64.9 \ \mu g m^{-3}$. Table 1 and Fig.4 indicate that the variation of the BC daily average from day to day is as high as 8 times and greater according to the change in meteorological conditions.

Example of daily variation of BC concentration in the atmosphere (daily averages of every hour for each date during 24 October-17 November 2000)

Beside the meteorological conditions, another factor influencing the daily variation of BC in the atmosphere in Beijing might be the use of central heating and domestic heating in the winter. Generally speaking, the particulate pollution during the heating period in Beijing increases about by 30% compared to that in the non-heating period. So it is reasonable that an early winter might show a strong daily variation of BC concentration in the atmosphere during the early winter.

3.4 Background level

In general, favorable meteorological conditions usually exist in the Beijing area during late autumn and early winter, so lower concentrations of BC are often observed then. There are many methods to determine the background level of pollution species in the atmosphere. For simplicity, here, we assume that the minimum value of BC concentrations observed on clean air days can be considered as the BC concentration background level. Based on the analysis of all data obtained during 1996–2004, the background level of BC in the atmosphere in Beijing is estimated as 1.31 $\mu g m^{-3}$. In the period of 1996–1999, when the air is more polluted, the background value is as high as 1.70 $\mu g m^{-3}$, while for the last five years, this background value is about $1.00 \,\mu \text{g m}^{-3}$. This BC background value for Beijing is as high as 20 times that of the continental background value in the Northern Hemisphere.

4. Conclusions

(1) The daily average concentrations of black carbon particles in the atmosphere in Beijing vary over quite a large range for the period of 1996–2004, namely $2-5 \ \mu \text{g m}^{-3}$ on clean air days, and $32-97 \ \mu \text{g m}^{-3}$ on heavily polluted days.

(2) The daily average BC concentration for whole period of 1996–2004 is estimated as $20.0 \ \mu g \ m^{-3}$, while for the last five years, this value is about $16.0 \ \mu g \ m^{-3}$ with a variation range of $2.10{-}50.5 \ \mu g \ m^{-3}$. An abrupt

decrease of BC concentration is observed in the atmosphere in Beijing after 2000.

(3) In most cases, the hourly average concentration of BC in the atmosphere has an evident double-peak of diurnal variation behavior with the maximum occurring at 0700–1000 LST and 2000–2400 LST, and the minimum occurring at 1300–1600 LST.

(4) A concentration value of black carbon particles of 1.0 μ g m⁻³ might be considered as the background level in the atmosphere in the Beijing area for late autumn and early winter at present.

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