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A size-segregation method for monitoring the diurnal characteristics of atmospheric black carbon size distribution at urban traffic sites

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HIGHLIGHTS

- A novel method for determining the mass size distribution of BC is developed.
- 90% of BC aerosols are smaller than 0.5 μm at urban traffic sites.
- Most BC aerosols are in the size range of 0.1–0.25 μm.
- Size distribution of BC exhibits a single mode at 0.16 μm at this sampling site.

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ABSTRACT

Understanding the characteristics of the size distribution of ambient black carbon (BC) in distinct environments is critical because the influence of BC aerosols on climate, visibility, and human health depends strongly on the distribution of BC aerosols over the particle size spectrum. In this study, a novel method for determining the mass size distribution of BC in atmospheric aerosols was developed. This size-segregation method relies on measuring BC in parallel using two aethalometers, one of which is used to measure the total BC (BC_T) mass concentration as a reference level and the other is used to measure the BC (BC_i) mass concentration for BC sizes below specific particle sizes that are selected using a size cut-off inlet. In this study, this method was applied to measure BC in atmospheric samples at an urban traffic site. The aethalometers were operated continually from December 15, 2012 to January 31, 2013, and from February 15, 2013 to March 31, 2013. The measurement results presented in this paper are for the diurnal variation patterns, average concentrations, mass fractions, and size distributions of BC aerosols. The results indicate that BC_T mass concentration is approximately 2.8 μg m^{-3} in the Taipei urban area. The levels of BC at this sampling site were affected markedly by traffic emission levels and local wind speed. At the sampling site, the average $BC_{2.5}/BC_T$, $BC_{1.0}/BC_T$, $BC_{0.5}/BC_T$, $BC_{0.25}/BC_T$, and $BC_{0.1}/BC_T$ were 0.96 ± 0.04, 0.92 ± 0.07, 0.89 ± 0.04, 0.73 ± 0.10, and 0.18 ± 0.08, respectively. The results indicate that approximately 90% of the BC aerosols were smaller than 0.5 μm, that most of the BC aerosols (55%) were in the size range of 0.1–0.25 μm, and that approximately 18% of the BC aerosols were ultrafine. Moreover, the daily average mass size distribution of BC exhibited a single accumulation mode at 0.16 μm at this sampling site. The mode of the BC mass size distribution at rush hour (9 AM) was only 0.14 μm, which is smaller than the daily average. Moreover, the mode of the BC mass size distribution at an early morning hour (3 AM) was 0.18 μm, and a minor coarse mode was also observed during this period. The results further revealed that the geometric diameter (D_{pg}) of the BC aerosols varied between 0.14 and 0.22 μm and the geometric standard deviation (σ_g) of these BC aerosols ranged between 1.4 and 2.3 during the sampling period. Because BC exhibits extremely low chemical reactivity; the size distribution of BC in the atmosphere does not change substantially except through coagulation.

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1. Introduction

Black carbon (BC) is a major component of aerosols produced by the incomplete combustion of carbonaceous fuels. BC plays a key role in aerosol climatic forcing because it exhibits light-absorption properties (Bond and Bergstrom, 2006; Jacobson, 2010). Ramanathan and Carmichael (2008) noted that the direct radiative forcing of BC can be high as +0.9 W m^{-2} and can be higher than the radiative forcing of other greenhouse gases such as CH_4, CFCs, N_2O, and tropospheric ozone. Furthermore, BC can cause numerous respiratory diseases and adversely affect the cardiovascular system because submicron BC particles can penetrate deep into the lungs and be deposited on the pulmonary alveolus (Suglia et al., 2008; Power et al., 2011; Cornell et al., 2012). Therefore, BC emissions are both a regional and global concern. Bond et al. (2013) reported that the global emissions of BC in 2000 were as high as 7.6 Tg, with approximately 27% of the BC being derived from diesel engines, 25% from residential solid fuel, 40% from open burning, and 8% from other sources such as ship emissions. However, large uncertainties exist regarding BC emissions because of insufficient field data, particularly in Asia. Ramanathan and Carmichael (2008) reported that BC emissions from China and India accounted for roughly 30% of global emissions. Zhang et al. (2009) suggested that the total BC emissions in Asia in 2006 were approximately 3.0 Tg, and that the BC emissions from China accounted for 1.8 Tg. Despite these discrepancies in the measurements of BC emissions, lowering BC emission represents a potential mitigation strategy that could reduce the global climate forcing caused by anthropogenic activities in the short term, and slow the associated rate of climate change and diminish the effect of BC on air quality and human health.

To develop strategies to control atmospheric BC, understanding the characteristics of the ambient BC size distributions in distinct environments is critical; this is because the influence of BC aerosols on climate, visibility, and human health strongly depends on the distribution of the BC aerosols over the particle size spectrum. However, the database on ambient BC size distributions is more limited than the database on total BC mass concentrations. Impactors have been used to collect size-segregated aerosol samples for BC analysis (Hitzenberger and Tohno, 2001; Viidanoja et al., 2002a; Mallet et al., 2003; Cuccia et al., 2013), but BC particle size distributions measured using the impactor have not provided results that exhibit adequate time resolution. Recently, a single particle soot photometer (SP2; Droplet Measurement Technologies) has been used to measure BC size distributions in real time by using the laser-induced incandescence technique (Schwarz et al., 2006; Shiraiwa et al., 2008; Schwarz et al., 2008; Kondo et al., 2011). However, SP2 is not widely used for studying BC in atmospheric aerosols because of the cost and performance limitations of SP2 (Gysel et al., 2012). Lately, a differential mobility analyzer (DMA) combined with an aethalometer had been applied to determine BC mass size distributions by using the different electrical mobility and light absorption methods (Stabile et al., 2012; Ning et al., 2013). This DMA-aethalometer system can be used to determine the BC modal characteristics directly in the size range of 20–600 nm.

The most common method used to measure BC involves collecting aerosols on a filter and measuring the reduction in light transmission through the filter (Hansen et al., 1984). The aethalometer (AE; Magee Scientific), multiangle absorption photometer (MAAP; Thermo Scientific), and particle soot absorption photometer (PSAP; Radiance Research) are among the currently available devices used for measuring BC by using filter-based optical techniques. These devices have been used extensively to monitor environmental BC mass concentrations because they can be operated easily and they provide favorable time resolution (Watson et al., 2005; Park et al., 2006; Chow et al., 2009). In this study, a method for determining the mass size distribution of BC in atmospheric aerosols was developed. This size-segregation method relies on measuring BC in parallel using two aethalometers; one of the two devices is used to measure the total BC mass concentration as a reference level, whereas the other is used to measure the BC mass concentration for BC sizes below specific particle sizes selected using a size cut-off inlet. The mass fractions of BC at various cut-off sizes in total BC can be determined using this size-segregated method, and the BC size distribution can be subsequently evaluated using these mass fraction data under various size ranges. One valuable feature of this method is that it offers time resolution for assessing BC mass size distribution. In this study, this method was applied to measure BC in atmospheric samples at an urban traffic site. The measurement results reveal the diurnal variation patterns, average concentrations, mass fractions and size distributions of BC aerosols.

2. Methods

2.1. Sampling equipment and data collection

In this study, two aethalometers (AE31; Magee Scientific) were used concurrently to measure BC mass concentrations at a traffic site. The sampling site (25° 04′ 20″ N; 121° 53′ 75″ E) used in this study is located on the campus of National Taipei University of Technology at Daan, Taipei, Taiwan, which is near the intersection of Jianguo Expressway and Zhongxiao E. Rd. (Fig. 1). Jianguo Expressway and Zhongxiao E. Rd. are north-south and east-west arterial roads, respectively, in Taipei City. Jianguo Expressway is linked to Taipei's road network and Zhongxiao E. Rd. connects two major commercial areas of Taipei City, the east and west commercial districts. Moreover, the east-west Civic Expressway is located 300 m north of the sampling site. Consequently, traffic is the primary local source of atmospheric particulate matter at this sampling site.

BC was measured in parallel using two aethalometers in this study; one aethalometer (Aethalometer A) was used to measure the total BC (BC_T) mass concentration as a reference level, whereas the other (Aethalometer B) was used to measure the BC mass concentrations for BC sizes below selected particle sizes (Fig. 2). The five cut-off sizes chosen in this study were 2.5, 1.0, 0.5, 0.25, and 0.1 μm, and the measured BC mass concentrations at these sizes were represented as $BC_{2.5}$, $BC_{1.0}$, $BC_{0.5}$, $BC_{0.25}$, and $BC_{0.1}$, respectively. The cut-off inlets of the various sizes used in this study for Aethalometer B are listed in Table 1. The $PM_{2.5}$ and $PM_{1.0}$ cyclones (BGI Inc.) had cut-off diameters of 2.5 and 1.0 μm, respectively, at a sampling flow rate of 5 L min^{-1}. These two cyclones were used to

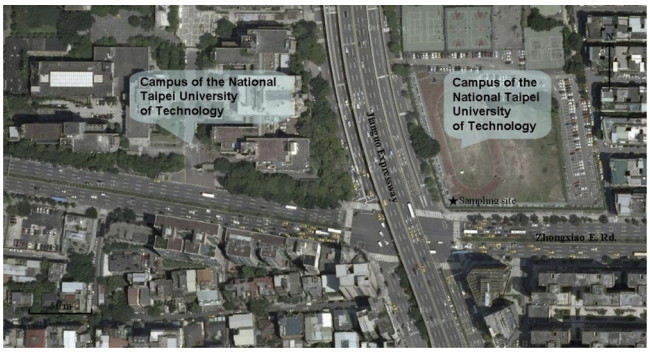

Fig. 1. Location of the sampling site on the campus of the National Taipei University of Technology.