Online Density and Shape Measurement of Single Black Carbon Aerosol Particles in a Heavily Polluted Urban Area

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Abstract

Black carbon (BC) aerosol imposes adverse effects on atmospheric visibility, climate, and health. The particle density and morphology are often needed to investigate the mixing state and aging process of BC particles. A method, combining an aerodynamic aerosol classifier (AAC), a differential mobility analyzer (DMA), a single-particle soot photometer (SP2) and a single particle aerosol mass spectrometer (SPAMS), was developed to determine the density and dynamic shape factor ($\chi$) of ambient BC particles with three different aerodynamic diameters ($D_a$, 200 nm, 350 nm, and 500 nm) in Shanghai, China, a typical urban area. The BC particles were either classified as “BC-dominated particle” which is mainly made of black carbon or “BC-mixed particle” which is a mixture of both BC and non-BC substances. The results show that BC-dominated particles whose BC core mass (~2.2 fg) was almost equal to particle mass (~2.3 fg) were observed in particles with 200 nm $D_a$. The morphology of these BC-dominated particles was near-spherical ($\chi \approx 1.02$), indicating that they had undergone rapid morphology modification from the initial highly irregular morphology to near-spherical shape. Most BC particles with 350 nm or 500 nm $D_a$ were BC-mixed particles. Combining the effective densities (1.62~1.77 g cm$^{-3}$) and average single particle mass spectra of particle, the ammonium sulfate and ammonium nitrate were found to be the main secondary substances of these BC-mixed particles, indicating that condensation of inorganic species such as nitrates and sulfates could play a significant role in the aging process of fresh BC.
in Shanghai. Generally, the morphology and density information of single BC particle is crucial to identify the mixing state and aging process of BC aerosols.

**Keywords:** Black carbon aerosol, Particle mass, BC content mass, Aerodynamic diameter, Electrical mobility diameter, Dynamic shape factor, Density
1 INTRODUCTION

It has been well acknowledged that black carbon (BC) aerosol exerts significant impacts on air quality, climate and human health (Menon et al., 2002; Anenberg et al., 2012; Bond et al., 2013; Wu et al., 2018). BC aerosol can provide surfaces for many important atmospheric reactions, such as catalyzing SO$_2$ oxidation, which could play a critical role in driving the formation, and aging of regional haze (Zhang et al., 2020). In addition, it can affect climate directly by scattering and absorbing solar radiation, and indirectly by influencing cloud formation through acting as cloud condensation nuclei (CCN) (Sun and Ariya, 2006; Li et al., 2008; Li et al., 2011). Meanwhile, BC aerosol has been evaluated by epidemiological studies for their adverse health effects (Zhang et al., 2015). It is found that the dose-response relationship between mortality and BC aerosols was much higher than that between mortality and particulate matter (PM) concentration (Janssen et al., 2011).

BC particles are mostly formed from incomplete combustion of carbon-containing fuels, such as coal, diesel engines, biofuels, and biomass (Steinfeld, 1998; Moffet and Prather, 2009). Many in-situ measurements and laboratory studies show that freshly emitted BC particles exist as aggregates of small carbon spherules (Bond et al., 2013; Soewono and Rogak, 2013). The mixing state of freshly emitted BC particle begins to transform during fuel burning process and then keeps changing during atmospheric aging (Jacobson and Seinfeld, 2004; Zhang et al., 2008; Yuan et al., 2013).
Accordingly, two important properties, morphology (including the overall size and shape factor) and density, will change along with the chemical mixing state of BC, which further leads to variations in their optical properties and radiative forcing. Currently, the morphology of BC particle has been considered in climate model when investigating its direct effect due to its impact for the optical properties (China et al., 2013; Wu et al., 2016). Wang et al. (2021) investigated the optical properties of BC particles based on TEM observations and highlighted that the morphology of BC core within the BC-containing particle and coating shape can influence optical properties and should be considered as important variables in climate models.

Particle morphology is usually described quantitatively by dynamic shape factor ($\chi$), which is the ratio of the drag force on non-spherical particle to the force on a spherical particle with equivalent volume (Hinds, 1999). The $\chi$ depend on the flow regime and asymptotically approaches unique values in the limits of the continuum regime and free molecular regime, further termed $\chi_c$ and $\chi_v$, respectively, and the shape factor in the transition regime was usually denoted as $\chi_t$. The shape factor is usually larger than 1 for irregular particles and equal to 1 for spheres. Besides, density, another important property of particle, is related to particle’s chemical composition and mixing state. It can also influence the residence time due to the fact that density is proportional to the deposition velocity (Friedlander and Sheldon, 1977). Furthermore, the effective density can be used to convert particle number concentration to particulate mass loading (McMurry et al., 2002; Schmid
et al., 2007). A deep understanding of BC particle’s morphology and density is important for better
evaluation of its mixing state, aging process and climate impact.

Multiple aerosol instruments can be used in tandem to simultaneously obtain the information on
particle shape and effective density. For the determination for effective density, many investigations
use a differential mobility analyzer (DMA) that can measure particle’s electrical mobility diameter
\(D_m\) and the single particle mass spectrometer (SPMS) instruments that can measure particle’s
vacuum aerodynamic diameter \(D_{va}\), such as single-particle aerosol mass spectrometer (SPAMS)
or aerosol time-of-flight mass spectrometer (ATOFMS), to measure the effective density \(\rho^I_{eff}\)
of single particles via Eq. (1)-(3) (Zelenyuk et al., 2006; Spencer et al., 2007; Pratt and Prather,
2012; Zhai et al., 2017), \(\rho_0\) is unit density (1 g cm\(^{-3}\)), \(C_c\) is the Cunningham slip correction
factor, \(\rho_p\) is the true density of particle, \(D_{ve}\) is the volume equivalent diameter of particle. Only
if the particle morphology is spherical, namely the dynamic shape factor is equal to 1, the \(D_m\) is
equal to \(D_{ve}\) according to Eq. (2), the particle effective density \(\rho^I_{eff}\) is equal to particle density
\(\rho_p\) in Eq. (3).

\[
D_{va} = \frac{\rho_pD_{ve}}{\rho_0X_p} \quad (1) \\
\frac{D_m}{C_c(D_m)} = \frac{D_{ve}X_t}{C_c(D_{ve})} \quad (2) \\
\rho^I_{eff} = \frac{D_{va}}{D_m} \rho_0 = \frac{\rho_pC_c(D_{ve})}{C_c(D_m)X_vX_t} \quad (3)
\]
Besides, by combining the DMA and a mass classifier (aerosol particle mass analyzer, APM or centrifugal particle mass analyzer, CPMA) that allows for the classification of particles by mass \( m_p \), another effective density \( \rho_{eff}^{ll} \) of particles can be obtained by Eq. (4) (Rissler et al., 2013; Yin et al., 2015; Li et al., 2017).

\[
\rho_{eff}^{ll} = \frac{m_p}{V} = \frac{6m_p}{\pi D_{m}^3}
\]  

(4)

In addition, McMurry et al. (2002) proposed the effective density \( \rho_{eff}^{ll} \) of particles by calibrating the system with polystyrene latex (PSL) particles in Eq. (5), where the \( V_{APM\, test} \) and \( V_{APM\, PSL} \) were the voltages at which the transfer function is maximum for the tested particles and PSL particles. This effective density \( \rho_{eff}^{ll} \) was used by many studies (Olfert et al., 2007; Levy et al., 2013).

\[
\rho_{eff}^{ll} = \rho_{PSL} \frac{V_{APM\, test}}{V_{APM\, PSL}}
\]  

(5)

The disadvantage of DMA, APM and CPMA is that these classifications rely on the use of charged particles which presents complications in the analysis (Radney and Zangmeister, 2016). In 2013 the aerodynamic aerosol classifier (AAC) that can determine the aerodynamic diameter \( D_a \) was developed (Tavakoli and Olfert, 2013), \( D_a \) is defined as the diameter of a sphere with 1.0 g cm\(^{-3}\) density that settles at the same terminal velocity as an irregular particle. The advantage of AAC is that it does not rely on particle charging so it produces a monodisperse aerosol without mixing multiple-charged particles like a DMA, CPMA, or APM. Tavakoli and Olfert, (2014) firstly used a
tandem AAC-DMA configuration to measure the effective density of liquid dioctyl sebacate
droplets via Eq. (4), where the $D_m$ was determined by DMA, the particle mass ($m_p$) was
determined by combing Eq. (6)-(8), $\mu$ is the viscosity of air, $\tau$ is the particle relaxation time, and
$B$ is the particle mobility.

\[ B = \frac{C_c(D_m)}{3\pi\mu D_m} \quad (6) \]

\[ \tau = \frac{\rho_0 C_c(D_a) D_a^2}{18\mu} \quad (7) \]

\[ m_p = \frac{\tau}{B} \quad (8) \]

Recently, Peng et al. (2021) firstly proposed AAC-SPAMS system to achieve the measurement of
effective density ($\rho_{\text{eff}}^{IV}$) of aspherical particle. In short, based on the approximation that $\chi_v=\chi_t$, the
volume equivalent diameter ($D_{ve}$) of particle can be calculated via Eq. (1) and Eq. (9). Then the
effective density of particle can be obtained via Eq. (10).

\[ D_a = D_{ve} \sqrt[3]{\frac{\rho_p C_c(D_{ve})}{\chi_t \rho_0 C_c(D_a)}} \quad (9) \]

\[ \rho_{\text{eff}}^{IV} = \left( \frac{D_{ve}}{D_{ve} \rho_0} \right) \quad (10) \]

Su et al. (2021) also used this AAC-SPAMS system to determine the effective density of primary
particles emitted from a light-duty diesel vehicle (LDDV) under the launching and idling engine.

Therefore, most existing tandem systems usually determine the effective density instead of particle
density ($\rho_p$). Besides, the measurement for particle’s dynamic shape factor required additional
information about particle density. For example, for the AAC-DMA system proposed by Tavakoli
and Olfert (2014), measurements of $D_a$ and $D_m$ alone are not sufficient to determine the shape factor, only if the particle density is known (or can be assumed), it is possible to determine $D_{ve}$ via Eq. (11) based on the measured particle mass ($m_p$), then the dynamic shape factor in transition regime ($\chi_t$) can be measured via Eq. (2).

$$D_{ve} = \sqrt[3]{\frac{6m_p}{\pi \rho_p}}$$  \hspace{1cm} (11)

In addition, for the differential mobility analyzer (DMA) and the single particle mass spectrometer (SPMS) tandem system, like the work of Zelenyuk et al. (2006), they also measure the dynamic shape factor of polystyrene latex (PSL) spheres agglomerates with known density (1.05 g cm$^{-3}$) via Eq. (3) based on the approximation that $\chi_v = \chi_t$. Zhang et al. (2016) combined a single-particle soot photometer (SP2) and a volatility tandem differential mobility analyzer (VTDMA) to measure the dynamic shape factor of the BC cores (density:1.8 g cm$^{-3}$) within the BC-containing particle via Eq. (2) and Eq. (11), where the BC core mass was measured by SP2. Thus, it can be known that for the ambient particle whose density is unknown, using existing tandem online system to measure their dynamic shape factor without assumption is challenging.

The goals of this study were to investigate the density, shape and the chemical composition of ambient BC-containing particle in Shanghai (a typical urban environment in China) by adopting an online method consisting of an aerodynamic aerosol classifier (AAC), a differential mobility...
analyzer (DMA), a single-particle soot photometer (SP2) and a single particle aerosol mass spectrometer (SPAMS). It has been shown that BC particles play a significant role in the PM pollution in Shanghai (Wei et al., 2020). The method adapted in this study can easily identify the “BC-dominated particle” mainly made of BC from all the BC-containing particles by comparing the particle mass ($m_p$) and mass of BC core in particle ($m_{BC}$). This identification method for BC-dominated particle has not been reported before. Then the BC-dominated particle density instead of effective density can be known, thus the measurement for their dynamic shape factor can be achieved further. Meanwhile, for “BC-mixed particle” made of BC and non-BC substances, their effective densities can also be investigated. The results of this study can provide helpful information for estimating the mixing state and aging process of BC aerosols in Shanghai.

2 METHODS

2.1 Instruments

2.1.1 AAC

In this study, the particles with known aerodynamic diameter ($D_a$) were selected by aerodynamic aerosol classifier (AAC, Cambustion Ltd., UK). AAC consists of two concentric cylinders rotating at the same rotational speed and the same direction. The classifier speed controls the centrifugal force applied to particles, the total classifier axial flow controls the residence time ($\tau$) of particle. Once the aerosols enter the sheath flow through a slit in the inner cylinder wall, the particles experience both a centrifugal force and drag force in the radial direction, particles with $D_a$ smaller
than the setpoint of AAC remain entrained in the sheath flow due to insufficient radial trajectory, and particles with $D_a$ larger than the setpoint of AAC will impact and adhere to the outer surface of the classifier due to excessive radial trajectory. Hence, only particles within a narrow $D_a$ range follow the correct trajectory and pass through the AAC classifier with the sample flow. More detailed description for AAC is in the previous study (Tavakoli and Olfert, 2013).

2.1.2 SP2

Single-particle soot photometer (SP2, Droplet Measurement Technologies, Boulder, CO, USA) was utilized to quantify the mass of BC ($m_{BC}$) inside individual particle down to ~0.3 fg by means of laser-induced incandescence (LII) (Stephens et al., 2003; Moteki and Kondo, 2007, 2010). Briefly, the SP2 uses a continuous Nd:YAG intra-cavity laser beam to heat a BC particle to its vaporization temperature of ~4000K, at which point, detectable amounts of incandescent light are emitted, the intensity of the incandescent light is linearly proportional to the BC mass (Moteki et al., 2010). Therefore, using black carbon particle standards to calibrate the incandescence signal of SP2 is required. In this study, Aquadag® black carbon particle standard (Acheson Inc., USA, Ultra-fine graphite), one of a commonly used BC reference material was used to calibrate SP2 (Moteki and Kondo, 2010; Gysel et al., 2011). The details about the calibration experiment of SP2 were shown in the Text S1 of Supporting Information.

2.1.3 SPAMS
Here, a single-particle aerosol mass spectrometer (SPAMS, Hexin Ltd. China) was adopted to obtain the chemical information of single BC particle. More detailed information on the SPAMS has been described elsewhere (Li et al., 2011). The particles in the size range of 0.2–2.0 μm are drawn into the vacuum through an aerodynamic focusing lens, each particle is accelerated to a size-dependent aerodynamic velocity. Subsequently, particles with the specific velocity pass through two lasers that are fixed at a 6 cm distance, the scatter light is collected by two photomultiplier tubes (PMTs). Afterwards, each single particle arrives at the ion source region, a pulsed desorption–ionization laser (Qswitched Nd :YAG; 266 nm) is triggered. Both positive and negative mass spectra for each single particle were recorded by a bipolar time-of-flight spectrometer. The single-particle mass spectra were converted to a list of peaks at each m/z by setting a minimum signal threshold of 30 arbitrary units above the baseline. The resulting peak lists together with other SPAMS data were imported into YAADA (version 2.11; www.yaada.org), a software toolkit for single-particle data analysis written in MATLAB (version R2014b). According to the similarities of the mass-to-charge ratio and peak intensity, the BC particles were identified from the ambient particles by utilizing a clustering method called “adaptive resonance theory neural network” (ART-2a) (Phares et al., 2001; Wang et al., 2019). BC particles were defined as the aggregation of two types of particles classified by ART-2a: EC particle with strong black carbon fragment ions (C_n^+ and C_n^-) and ECOC particle with both strong black carbon fragment ions (C_n^+ and C_n^-) and some
organic carbon peaks (e.g., m/z 27[C₂H₃⁺], 37[C₃H⁺], 43[C₃H₇⁺]). The vigilance factor, learning rate and iterations for ART-2a were set as 0.85, 0.05 and 20, respectively.

2.2 Sampling information

The field sampling was carried out on the sixth floor of the building of the Department of Environmental Science and Engineering, Fudan University (31°20’N, 121°30’E), located in Shanghai (Figure. 1). The sampling site is surrounded by residential and commercial zones, thus it is mainly influenced by a mixture of pollutant emissions from surrounding human activities and traffic. Ambient aerosols were drawn to the measurement system through a 6 m long stainless-steel pipe (1/4 inch). Figure. 2 shows the schematic flow chart of the instrument arrangements. Firstly, the ambient aerosols were classified by AAC with a specific setpoint ($D_a$) and then selected by a DMA (Model 3081, TSI Inc., USA) with a specific electrical mobility diameter ($D_m$). Then, these particles were parallelly measured by a condensation particle counter (CPC, Model 3775, TSI Inc., USA), SP2 (Droplet Measurement Technologies, Boulder, CO, USA) and SPAMS (Hexin Ltd. China). Considering that the transmission efficiency of AAC is higher than 80% for particles with $D_a$ between 200 nm to 1000 nm (Johnson et al., 2018), and the particle whose vacuum aerodynamic diameter ($D_{va}$) smaller than 200 nm is hard to be detected by SPAMS (Li et al., 2011), thus, three groups of ambient aerosols ($D_a = 200$ nm, $D_a = 350$ nm and $D_a = 500$ nm) were extensively studied here. The detailed sampling schedule is shown in Table S1. During the whole experimental process, the aerosol flow rates of SP2 and SPAMS were respectively 0.03 L min⁻¹ and 0.1 L min⁻¹, the aerosol flow rate of DMA was 0.33 L min⁻¹ (0.3 L min⁻¹ from CPC and 0.03 L min⁻¹ from SP2) and the sheath flow rate of DMA was set as 3.3 L min⁻¹, namely, the aerosol to sheath flow ratio of DMA was 1/10. For AAC, the resolution, defined as the relaxation time corresponding to the maximum value of the transfer function divided by the full width of the transfer function evaluated at one-half of the maximum value of the transmission efficiency.
(Tavakoli and Olfert, 2013), was maintained at 16.5, corresponding to a 1/10 aerosol to sheath flow ratio during the whole study. The power of the desorption–ionization laser of SPAMS was set to ~0.5 mJ per pulse. Before conducting any measurements, aerosols were dried (RH < 20%) by silica diffusion dryers.

**Figure 1.** The location of the sampling site.

**Figure 2.** Schematic diagram of the experimental set-up
2.3 Data analysis

2.3.1 Estimation for particle mass and BC mass

For particle selected by AAC with a fixed set value \(D_a\) and DMA with a fixed set value \(D_m\), the particle mass \(m_p\) can be obtained by Eq. (6)-(8), like the previous study (Tavakoli and Olfert, 2014). Meanwhile, for a group of AAC-DMA selected BC particles, the mass of BC content \(m_{BC}\) within each individual particle was further measured by the SP2. Thus, the BC mass distribution, namely the number distribution of these AAC-DMA selected particles based on the BC mass, can be obtained. If there existing a peak in the BC mass distribution, then the Gauss fitting was found to be an appropriate function to do peak-fitting \((R^2 > 0.90)\). The peak center was exactly the mass of BC content of particles under this peak.

2.3.2 Measurement for “BC-dominated particle” and “BC-mixed particle”

Comparing the BC mass \(m_{BC}\) measured by SP2 and the particle mass \(m_p\) measured by AAC-DMA system, if \(m_{BC}\) in a single particle was nearly equal to \(m_p\), such a BC particle was referred to “BC-dominated particle”. If \(m_{BC}\) in a single particle was far less than \(m_p\), such a BC particle was referred to “BC-mixed particle”. The shape and density of these two types of BC particles were investigated separately. For BC-dominated particle, their density should be very close to the material density of black carbon as we assume that there are no internal voids within BC particle. The value of 1.8 g cm\(^{-3}\) had been commonly used as the material density of BC particle (Moteki and Kondo, 2010; Moteki et al., 2010; Zhang et al., 2016). Then, the dynamic shape factor (\(\chi\)) of
these BC-dominated particles can be obtained by Eq. (2) and Eq. (11) based on the known density \( \rho_p \), mass \( m_p \) and mobility diameter \( D_m \). For BC-mixed particles, their effective density can be derived by Eq. (4). The uncertainty estimation for all the measured and derived parameters in this study was shown in the Text S2 in Supporting information.

3 RESULTS AND DISCUSSION

3.1 Aerosol with 200 nm \( D_a \)

In this study, the electrical mobility size distribution of aerosol selected by AAC at 200 nm setpoint was measured by a scanning mobility particle sizer (SMPS, combining DMA and CPC). Given that both in winter sampling time of 2018 and summer sampling time of 2019 the largest peak’s location of the electrical mobility size distribution of these AAC-selected particles was constant at \( \sim 135 \) nm (See Figure 3), thus, the setpoints of AAC and DMA were respectively set as 200 nm and 135 nm to select the particles in Mode \( D_a = 200 \) nm, \( D_m = 135 \) nm, which were further measured by SP2 and SPAMS.
(a) winter of 2018

Average result of 12h (2h per day)
$R^2 = 0.997$ (Gauss Fit) $d_1 = 135$ nm

(b) summer of 2019

Average result of 24h (15 Aug 18:00-16 Aug 18:00)
$R^2 = 0.985$ (Gauss Fit) $d_1 = 135$ nm
Figure 3. Average electrical mobility size distribution of particle with 200 nm $D_a$ in (a) winter sampling time of 2018 and (b) summer sampling time of 2019. The insert panel shows the 2 hours average size distribution of these particles.

Based on the known $D_a$ and $D_m$, the particle mass ($m_p$) in this Mode was calculated as ~2.3 fg via Eq. (6)-(8). Meanwhile, SP2 measured the mass of BC content ($m_{BC}$) within each individual particle. Fig. 4 presents the BC mass distribution, namely the number distribution of the particles in Mode ($D_a$=200 nm, $D_m$=135 nm) based on their $m_{BC}$. Two peaks (Peak1, Peak2) were found. The location of Peak 1 was fitted to be ~2.2 fg, which was close to the particle mass (~2.3 fg), thus, it can be known that the particles in Peak 1 were almost made of BC and named as BC-dominated particles. Here, $m_{BC}$ was not strictly equal to $m_p$, which could be partly due to the existence of non-BC components or the accuracy of $m_{BC}$ measured by AAC-DMA-SP2 (~5%). For particles in Peak 2, the mass of BC content ($m_{BC}$) within BC particle was apparently less than the particle mass, $m_p$ (~2.3 fg), thus these particles with a fair amount of non-BC components was BC-mixed particle. Their effective density was derived as 1.75 g cm$^{-3}$ via Eq. (4), suggesting that ammonium nitrate (1.73 g cm$^{-3}$) and/or ammonium sulfate (1.77 g cm$^{-3}$) were probably the main coating substances (Levy et al., 2013).
In terms of the density of BC-dominated particle, as introduced in “METHODS”, their density can be estimated as 1.8 g cm\(^{-3}\), along with the known particle mass, their dynamic shape factor (\(\chi\)) was calculated as \(~1.02\) via Eq. (2) and Eq. (11). The value of \(\chi\) indicated that the morphology of BC-dominated particle almost made up by BC was near-spherical. It is acknowledged that the shape of freshly emitted BC particles are usually chains or some other irregular shapes such as aggregates of small carbon spherules (Bond et al., 2013; Soewono and Rogak, 2013), thus, this results potentially indicated the morphology modification of freshly emitted BC from irregular to near-spherical shape may occur quickly during the sampling time. Meanwhile, Peng et al. (2016) also demonstrated that in Beijing the fresh BC particles underwent rapid morphology modification from a highly fractal structure to a near-spherical shape within 2.3 hours by quantifying the dynamic shape factor of BC particles under particle-free ambient gases at a steady flow rate in a chamber. Moffet and Prather (2009) also found that within 3 h after sunrise photochemical activity results in the rapid conversion of fresh non-spherical soot to aged spherical coated soot particles. The collapse in structure of BC particle may be related to the condensation of water and other gas-phase species upon the BC aggregates (Huang et al., 1994; Weingartner et al., 1997; Ma et al., 2013).
Figure 4. BC mass distribution for the particles collected in summer sampling time of 2019 in Mode ($D_a = 200 \text{ nm}, D_m = 135 \text{ nm}$). The insert panels show the daily BC mass distribution.

In addition, traffic was found to be a possible contributor to these BC-dominated particles based on Fig. 5, which shows that in many sampling days the diurnal variation of the number ratio of BC-dominated particles to BC-containing particles (the aggregation of BC-dominated particles and BC-mixed particles) presents an ascending tendency during the morning (red shaded area) and evening rush hours (grey shaded area). The calculation method for the number of three types of particles (Non-BC Particle, BC-dominated particle, and BC-mixed particle) are shown in Text S3. Gong et al. (2015) also found that traffic was a main source of BC-mixed particles that accounted
for almost all of the particles observed in the size ranges of 100–300 nm in Shanghai, which was consistent with our results that these BC-dominated particles in Mode ($D_a = 200$ nm, $D_m = 135$ nm) were mainly emitted from traffic. Meanwhile, Zhou et al. (2009) found that traffic should be the dominant source of BC particles in Shanghai based on the significant correlation between BC, CO and NOx and the poor correlation between BC and SO2. According to the diurnal variation of the concentration of black carbon (BC) in PM$_{2.5}$, Feng et al. (2014) also demonstrated that traffic emission was the main source of BC in urban Shanghai. Recently, Wei et al. (2020) found that a bimodal distribution, with two peaks in the morning and evening rush hours, appeared in the diurnal BC concentration variation, indicating the influences of local traffic emissions. too.
3.2 Aerosol with 350 nm $D_a$

In addition to the study for ambient BC particle with 200 nm $D_a$, the morphology and density of particles selected by AAC at 350 nm $D_a$ were measured, too. Figure 6 shows that the location of the largest peak (~75%) in the average electrical mobility distribution of the particles selected by AAC at 350 nm setpoint was ~260 nm, thus the particles in Mode ($D_a =$ 350 nm, $D_m =$ 260 nm) were studied by SP2 and SPAMS. The mass of particles in this mode was ~14.9 fg. Fig. S1A shows the BC mass distribution of the particles in this mode, indicating that the BC mass was smaller than the particle’s mass, thus most BC particles in this mode were BC-mixed particles. The effective density can be calculated as ~1.62 g cm$^{-3}$ via Eq. (4). Meanwhile, the average mass spectra of these BC-mixed particle were shown in Fig. 7, indicating the presence of high intensity of carbon fragment ions ($C_n^+$ and $C_n^-$), sulfate-related ions (HSO$_4^-$, HSO$_3^-$), nitrate-related ions (NO$_3^-$, NO$_2^-$) and some organic carbon peaks (e.g., m/z 37[C$_3$H$_7^+$/43[C$_3$H$_7^-$]), thus, combining the effective density and the mass spectra information, it can be deduced that ammonium sulfate (density: 1.77 g cm$^{-3}$) or ammonium nitrate (density: 1.73 g cm$^{-3}$) internally mixed with organic materials (average density: ~1.3 g cm$^{-3}$) (Dinar et al., 2006; Malloy et al., 2009; Levy et al., 2013) probably were the main compositions of these BC-mixed particles.
Figure 6. The electrical mobility size distribution of particles with a $D_a$ of 350 nm, the insert panel shows the 1-hour average size distribution of these particles sampled each day.

Figure 7. Average single particle mass spectra of BC particles in Mode ($D_a=350$ nm, $D_m=260$ nm)
3.3 Aerosol with 500 nm $D_a$

For particles selected by AAC at 500 nm setpoint, the location of the largest peak (~82%) in the electrical mobility size distribution of these particles in Fig. 8 was ~359 nm, thus, the particles in Mode ($D_a = 500 \text{ nm}, D_m = 359 \text{ nm}$) were mainly studied. The mass of particles in this mode was ~42.7 fg. According to the BC mass distribution of particles in this mode in Fig. S1B, most BC particles were found to be BC-mixed particles. The effective density was calculated to be ~1.77 g cm$^{-3}$. Meanwhile, Figure 9 showed that the average mass spectra of these BC-mixed particles measured by SPAMS presented high intensity peaks of HSO$_4^-$, NO$_3^-$, NO$_2^-$ with weaker negative carbon ion signals (C$_n^-$) than the BC-mixed particles in Mode ($D_a = 350 \text{ nm}, D_m = 260 \text{ nm}$), indicating the existence of ammonium sulfate (density: 1.77 g cm$^{-3}$) or ammonium nitrate (density: 1.73 g cm$^{-3}$), and the effective density was within the density range of these coating materials.
Figure 8. The electrical mobility size distribution of particles with 500 nm $D_a$, the insert panel shows the 2 hours average size distribution of these particles sampled each day.

![Figure 8](image)

Figure 9. Average single particle mass spectra of BC particles in Mode ($D_a=500$ nm, $D_m=359$ nm)

4 CONCLUSIONS

In this study, the density and morphology of ambient BC particles in Shanghai, China were investigated by measuring the BC particles in three typical modes: Mode ($D_a=200$ nm, $D_m=135$ nm), Mode ($D_a=350$ nm, $D_m=260$ nm) and Mode ($D_a=500$ nm, $D_m=359$ nm). The mass of BC particles ($m_p$) can be computed directly from measured aerodynamic diameter ($D_a$) and electrical diameter ($D_m$), which were readily determined by an AAC and a DMA, respectively. Then, the mass of black carbon content ($m_{BC}$) of these AAC-DMA selected particles can be measured by
By comparing the particle mass \( (m_p) \) and the mass of BC content \( (m_{BC}) \) in particle, the BC particles were classified into “BC-dominated particle” mainly made of BC and “BC-mixed particle” made of both BC and non-BC substances.

The results showed that a fraction of BC-dominated particles whose dynamic shape factor \( (\chi) \) was \( \sim 1.02 \) were detected in Mode \( (D_a=200 \text{ nm}, D_m=135 \text{ nm}) \), which means that their shape was near-spherical, potentially indicating that morphology modification from the initial highly irregular morphology to near-spherical shape was rapid. The increasing number ratio of BC-dominated particles to BC particles during commuting hours suggests traffic emission are the primary source of these BC-dominated particles. Meanwhile, for larger particles in Mode \( (D_a=350 \text{ nm}, D_m=260 \text{ nm}) \) and Mode \( (D_a=500 \text{ nm}, D_m=359 \text{ nm}) \), most BC particles were found to be BC-mixed particles.

The effective densities of particles in Mode \( (D_a=350 \text{ nm}, D_m=260 \text{ nm}) \) and in Mode \( (D_a=500 \text{ nm}, D_m=359 \text{ nm}) \) were respectively \( \sim 1.62 \text{ gcm}^{-3} \) and \( \sim 1.77 \text{ gcm}^{-3} \), along with their average single particle mass spectra information, it can be deduced that during the sampling periods condensation of inorganic species such as nitrates and sulfates could play a significant role in the aging process of fresh BC in Shanghai.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Nos. 21906024, 91544224, 41775150, 41827804), and Shanghai Natural Science Foundation (No. 19ZR1404000).
DISCLAIMER

The authors declare that they have no conflict of interest.

DATA AVAILABILITY

All the data from this study is available at http://dx.doi.org/10.17632/jfyfs9nvtn.1.

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