The Contribution of Black Carbon and Non-BC Absorbers to the Aerosol Absorption Coefficient in Nanjing, China

Ari Leskinen^{1,2*}, Antti Ruuskanen¹, Pekka Kolmonen¹, Yu Zhao³, Die Fang³, Qingeng Wang³, Cheng Gu³, Jorma Jokiniemi⁴, Maija-Riitta Hirvonen⁴, Kari E.J. Lehtinen^{1,2}, Sami Romakkaniemi¹, Mika Komppula¹

¹ Finnish Meteorological Institute, Yliopistonranta 1 F, FI-70210 Kuopio, Finland

² University of Eastern Finland, Department of Applied Physics, Yliopistonranta 1 F, FI-70210 Kuopio, Finland

³ Nanjing University, School of the Environment, 210023 Nanjing, China

⁴ University of Eastern Finland, Department of Environmental and Biological Sciences, Yliopistonranta 1 F, FI-70210 Kuopio, Finland

ABSTRACT

An aethalometer was employed in a 1-year campaign (November 2014–November 2015) in Nanjing, China, that aimed to estimate the contributions of black carbon (BC; annual average $\pm$ std = $4.0 \pm 3.3 \mu\text{g m}^{-3}$) and non-BC absorbers, such as brown carbon, to the aerosol absorption coefficient. We applied two methods: 1) the traditional calculation from the aethalometer data, assuming an absorption Ångström exponent (AAE) of unity for BC (“AE method”), which provided an overall average of 27.2% for the non-BC contribution, and 2) a recently developed method based on the wavelength dependence of AAE (the “WDA method”), which indicated the existence of non-BC absorbers in 25.3% of the samples on average. We utilized trajectory and source area analyses and, in agreement with the results from other studies in this region, verified biomass burning emission sources, mainly to the northwest of the measurement site.

Keywords: Aethalometer; Mie calculation; Trajectory analysis; Source area analysis.

INTRODUCTION

Particulate black carbon (BC) and non-BC absorbers have several effects on the environment and human health. For example, they affect the atmosphere’s radiation balance both directly by absorbing incoming solar radiation (Bond *et al.*, 2013) and indirectly and semi-directly by affecting the aerosols’ cloud properties (Brown *et al.*, 2018; Allen *et al.*, 2019). On snow, BC decreases the snow albedo (Hadley and Kirchstetter, 2012), leading to warming of the snowy surfaces. When inhaled, the BC and non-BC absorbers may have adverse health effects (Janssen *et al.*, 2012).

BC, also called soot, originates from incomplete combustion and consists of carbonaceous components that strongly absorb radiation across the solar spectrum. The non-BC absorbers, in turn, include 1) dust, 2) organics that are “dark” in color (humic-like substances, HULIS) and 3) light-absorbing organic matter called “brown carbon” (BrC). Dust may be, e.g., blown by wind from the soil into the atmosphere, whereas

burning of biomass and biofuel is known to produce BrC in addition to BC, and some light-absorbing organic compounds may be also generated in atmospheric transformation processes (Washenfelder *et al.*, 2015). The BrC absorbs light preferentially at the upper end of ultraviolet region, a property that can be exploited in estimating the BrC contribution to the total light absorption of aerosol particles. The optical properties of BC and non-BC absorbers may also depend on their mixing state. For example, a layer of non-BC compounds on BC particles may enhance the absorption efficiency of the BC core (Cappa *et al.*, 2012).

The contribution of non-BC absorbers has been studied traditionally by measuring the absorption coefficients at several wavelengths, ranging from the ultraviolet to near-infrared, mostly with an aethalometer (AE). However, aethalometer measurements are known to suffer from two major artifacts: the filter loading effect and multiple scattering effect. To minimize and take into account these artifacts, both the instrument and the analysis methods have been developed. The filter loading effect is known to depend on the wavelength used in the analysis (Weingartner *et al.*, 2003; Virkkula *et al.*, 2007; Virkkula *et al.*, 2015), and accurate compensation methods are needed for spectral measurements and subsequent source apportionment of BC or non-BC absorbers. Correcting the filter loading effect is quite straightforward (Virkkula *et*

* Corresponding author.

Tel.: +358 50 522 9148

E-mail address: Ari.Leskinen@fmi.fi

al., 2007), and it has already been implemented in an aethalometer using a dual-spot technology (Drinovec *et al.*, 2015). The multiple scattering correction, in turn, is more complicated, and several algorithms have been developed over time (e.g., Weingartner *et al.*, 2003; Arnott *et al.*, 2005; Schmid *et al.*, 2006; Collaud Coen *et al.*, 2010). Many of these algorithms utilize concurrent scattering coefficient measurements in the corrections. If the scattering coefficient cannot be measured concurrently, it can be estimated, for example, by using Mie calculations if particle size distribution, shape, and refractive index are measured or estimated.

A recently developed method, called the “wavelength dependence of absorption Ångström exponent” (WDA) method (Wang *et al.*, 2016), combines multi-wavelength absorption measurements and theoretical Mie calculations for BC. A great advantage of the WDA method is that it requires only multi-wavelength aethalometer measurements but no concurrent scattering or extinction measurements. However, the WDA method can be only used for finding out whether the inspected particulate sample contains other absorbers than BC or not.

In this study, the AE and WDA methods were applied in estimating the contribution of BC and non-BC absorbers to the aerosol absorption coefficient during a 1-year-long measurement campaign in Nanjing, China, in the Yangtze River Delta (YRD) region. The YRD is one of the most heavily polluted region in China with intensive industry and fossil fuel sources as well as an important area of agriculture production with occasional open biomass burning events influencing the regional and local air quality (Yang and Zhao, 2019). We evaluated the non-BC contribution to aerosol absorption coefficient for the emissions from some of these sources by using air mass back trajectories and

meteorological measurements.

METHODS

Measurement Site and Campaign Period

The measurements were carried out at the Xianlin Campus of Nanjing University on the rooftop of a five-floor building, at about 30 m above the ground level. The measurement site (32.117°N, 118.954°E) is located approximately 20 km east of the city center of Nanjing, which is an industrial megacity in the YRD (Fig. 1). The surroundings of the measurement site can be regarded as a suburban environment with the university area, residential areas, and traffic routes. A nearby highway, 300 m to the west of the measurement site, is one major source for traffic-related emissions. During the measurement campaign period of 5 November 2014–2 November 2015, construction sites at the campus area may also have caused elevated particle concentrations, particularly in the coarse fraction.

Meteorological Parameters

We measured ambient temperature, relative humidity, pressure, wind speed and direction, and precipitation intensity with a Vaisala Model WXT520 weather station that was equipped with a radiation and rain shield for the humidity and temperature sensors. The meteorological data were averaged over 1-minute time resolution for further analysis.

Sampling and Sample Dilution

The measurement instruments (Table 1) were located in an air-conditioned container, except the weather station that was installed approximately 2 m above the container roof in order to avoid disturbances by the nearby objects. The

Fig. 1. The location of Nanjing, China (marked with the black dot), and the five selected sectors for air parcel backward trajectory analyses (separated by the black lines).

Table 1. Instruments and their inlets for the measured properties.

Measured property	Instrument	Inlet
Aerosol light absorption	Aethalometer (Model AE-31; Magee Scientific)	PM _{6.5}
Elemental carbon concentration	Thermal-optical transmittance carbon analyzer (Model-4; Sunset Lab)	PM _{2.5}
Particulate mass concentration	Tapered element oscillating microbalance (Model 1405D-BEF; Thermo-Fisher Scientific)	PM _{2.5} and PM ₁₀
Aerosol particle size distribution	NanoScan mobility particle sizer (Model 3910; TSI Inc.) and optical particle sizer (Model 3330; TSI Inc.)	PM _{6.5}

sampling line for the instruments was a vertical stainless steel tube with a 10-mm outer diameter through the roof of the container, and its inlet was covered with a rain shield. Inside the container the sampling line was thermally insulated in order to prevent condensation in the sampling line. Near the instruments, the sample line was divided for the measurement instruments. The calculated overall cut-off diameter (diameter with 50% collection efficiency for the sampling system; Fig. S1), considering diffusional, settling, and inertial losses in the inlet and sampling lines, was 6.5 μm (called “PM_{6.5} inlet”). The estimated penetration efficiency for 10- μm particles was 17%. For some instruments, standard inlets for particulate matter less than 2.5 μm in diameter (PM_{2.5}) and less than 10 μm in diameter (PM₁₀) were used (Table 1).

After 10 January 2015 the sample was diluted by using a porous tube diluter (constructed at the University of Eastern Finland) in order to lower the relative humidity in the sample and to reduce the risk of condensation in the sampling line. The dilution air flow was produced by a diaphragm pump following a high-efficiency particulate air (HEPA) filter (P/N 12144; Pall Corp.) at the flow rate of 3.0 L min⁻¹. The total sampling rate of the instruments was 4.5 L min⁻¹, so the sample flow rate from outside was 1.5 L min⁻¹, and the concentration dropped in the diluter to 1/3. The dilution air flow rate and its temperature and pressure were monitored continuously with a TSI Model 4140 mass flow meter and recorded for calculation of the exact dilution rate for every minute. The other flow rates were checked intermittently with the same mass flow meter.

Equivalent Black Carbon Concentration and Absorption Coefficient

Aerosol light absorption was measured with a Magee Scientific Model AE-31 aethalometer (“AE-31”). An aethalometer (Hansen *et al.*, 1984) collects aerosol sample on a quartz fiber filter and measures the attenuation of a light beam transmitted through the sample on a filter material, whose clean part is used as a reference. An absorption coefficient (b_{abs}), which is linearly dependent on the light attenuation, indicates how much light is absorbed per unit length (the unit of absorption coefficient is usually given as Mm⁻¹). The AE-31 reports the measurement result as equivalent black carbon (EBC; Petzold *et al.*, 2013) concentration at seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm). The EBC concentrations are obtained by dividing the absorption coefficient measured at each wavelength by a mass absorption efficiency (MAE), called also “mass absorption coefficient” (MAC). MAE is wavelength-dependent, and the

MAE values used in the AE-31, given by the instrument manufacturer, are derived from 14,625 (m² g⁻¹ nm) λ^{-1} , where λ is the wavelength in nm. By using the formula given by the manufacturer, the calculated values for the MAE are 39.5, 31.1, 28.1, 24.8, 22.2, 16.6, and 15.4 m² g⁻¹. The MAE could also be determined by dividing the measured absorption coefficient (b_{abs}) by the elemental carbon (EC) concentration ([EC]) at the same location, simply by:

$$\text{MAE} = b_{\text{abs}}/[\text{EC}] \quad (1)$$

The absorption coefficient at each of the AE-31 wavelengths was calculated by multiplying the corresponding EBC concentrations with the MAE. The aethalometer inlet flow rate was 5.9 L min⁻¹ and 2.8 L min⁻¹ before and after installing the dilution system, respectively, and the lowest possible data collection interval of 2 minutes.

In this study the EC concentration in PM_{2.5} was sampled and measured by a semi-continuous carbon analyzer (Model-4; Sunset Lab, USA) that applies the thermal-optical transmittance (TOT) method and uses a modified protocol of the National Institute of Occupational Safety and Health (NIOSH 5040) as its default protocol. The PM_{2.5} and PM₁₀ concentrations, in turn, were measured with a tapered element oscillating microbalance (TEOM; Model 1405D-BEF, Thermo-Fisher Scientific Inc., USA).

Aerosol Particle Size Distribution

The aerosol particle size distribution was measured with a TSI NanoScan scanning mobility particle sizer (SMPS) Model 3910 and a TSI optical particle sizer (OPS) Model 3330. The SMPS measures particles with their diameters in the size range of 10–420 nm in 13 size bins, while the OPS measures particles with their diameters in the size range of 0.3–10 μm in 16 size bins. The data from these two instruments were combined when applicable.

Absorption Coefficient and Absorption Ångström Exponent

The absorption coefficient for each measured wavelength, $B_{\text{aeth}}(\lambda)$, was calculated by using the equation:

$$B_{\text{aeth}}(\lambda) = (14,625 \lambda^{-1}) \times [\text{EBC}(\lambda)] \quad (2)$$

where the constant 14,625 is given by the manufacturer for “Magee BC” calibration, λ is the wavelength (nm), and [EBC(λ)] is the black carbon concentration (ng m⁻³) measured and reported by the aethalometer at that wavelength. The

term ($14,625 \lambda^{-1}$) is the MAE ($\text{m}^2 \text{g}^{-1}$). By using appropriate unit conversions, the unit of $B_{\text{aeth}}(\lambda)$ is m^{-1} , but usually, as also in this study, Mm^{-1} is used as the unit for the absorption coefficient.

When there is a need to interpolate or extrapolate absorption coefficients at other wavelengths than at those with measurements, one can do this by using the absorption Ångström exponent (AAE). The AAE for the absorption coefficient, B_{aeth} , whose value is measured at wavelengths λ_1 and λ_2 , is defined as:

$$\text{AAE}_{\lambda_1-\lambda_2} = -[\log(B_{\text{aeth},\lambda_1}/B_{\text{aeth},\lambda_2})/\log(\lambda_1/\lambda_2)] \quad (3)$$

For example, if the absorption coefficient has been measured at 590 and 660 nm, the absorption coefficient at wavelength 637 nm is:

$$B_{\text{aeth},637\text{nm}} = \log(B_{\text{aeth},590\text{nm}}/B_{\text{aeth},660\text{nm}})/\log(590/660) \times \log(590/637) \quad (4)$$

Aethalometer Corrections

The aethalometer is known to suffer from artifacts due to its filter-based measurement principle. Firstly, the dependence of the absorption coefficient on the light attenuation becomes non-linear with increasing aerosol mass on the filter tape, a so-called “filter loading” or “shadowing” effect. Secondly, multiple scattering from the aerosol particles on the filter and from the filter material increases the optical path of light, and the attenuation caused by the multiple scattering is misinterpreted as an attenuation caused by absorption. The corrected absorption coefficient, B_{abs} , can be calculated from:

$$B_{\text{abs}} = B_{\text{aeth}}/(C_{\text{ref}} \times R) \quad (5)$$

where B_{aeth} is the absorption coefficient calculated from Eq. (2), C_{ref} is a correction coefficient for the multiple scattering effect and R is a correction coefficient for the filter loading effect. Both coefficients are wavelength-dependent.

In this work we used the Virkkula correction for the filter loading effect (Virkkula *et al.*, 2007; Virkkula *et al.*, 2015) with the assumption that the average of the last three values measured on a filter spot on the filter tape and the average of the first three values measured on the following filter spot are equal and that the correct value is the one measured after the filter change. It must be noted that this correction is used for filter loading effect only and it does not take the light scattering from the particles or the filter tape into account.

We performed similar validation as Drinovec *et al.* (2015) for the uncorrected and Virkkula-corrected data, averaged over the whole measurement period, and found that the correction removes the dependence of BC concentration on attenuation almost completely (Fig. S2(a)). This indicates good validity of the correction method for the BC data measured at 880-nm wavelength, particularly for the attenuation range of 5–55 with high enough observation frequency (Fig. S2(b)).

According to the recommendations by Collaud Coen *et al.* (2010), knowing or estimating the multiple scattering effect is important, especially at sites with high BC concentrations. In practice, the multiple scattering effect is estimated by

calculating the coefficient C_{ref} in Eq. (5) by conducting parallel measurements of absorption coefficient at all desired wavelengths with a reference instrument that does not suffer from such a multiple scattering artifact. An ideal reference instrument would be one that retrieves the absorption coefficient for suspended particles, either directly or indirectly, by subtracting scattering coefficient from extinction coefficient. This is, however, often quite complicated, because many instruments used as a reference have their own artifacts, especially those using a filter-based measurement technique, or they measure the reference value only at one or a couple of wavelengths.

In this work we estimated the multiple scattering effect by calculating the C_{ref} for each wavelength the aethalometer measures by using the approach of Arnott *et al.* (2005). We used the equation

$$B_{\text{abs}} = (B_{\text{aeth}} - \alpha \times B_{\text{sca}})/M \quad (6)$$

derived from Eq. (27) in Arnott *et al.* (2005) by omitting the square root term describing the filter loading effect, instead of which we used the Virkkula correction. We adopted the coefficients α and M in Eq. (6) from Arnott *et al.* (2005). The B_{sca} in Eq. (6) is the scattering coefficient measured concurrently with the aethalometer measurements.

Since we did not have concurrent scattering coefficient measurements, we calculated the scattering coefficients at each wavelength by applying Mie theory for the measured aerosol size distributions and by using the MatScat package for MATLAB (Schäfer, 2011; Schäfer *et al.*, 2012), whose implementation is based on the theory in Bohren and Huffman (1998). In the calculations we assumed spherical particles with a wavelength-independent refractive index of $1.5-0.02j$. The real part is the same as the average of the real parts of the refractive indices for different types of ambient aerosols measured in Guangzhou, China (Zhang *et al.*, 2016). The real and imaginary parts are close to the value of $1.48-0.01j$ obtained from AERONET observations in Beijing, China, and Mie calculations for fine particles at 440 nm (Zhang *et al.*, 2017). With this method we obtained long-term averages for the C_{ref} of 3.04, 3.31, 3.41, 3.51, 3.62, 3.97, and 4.04 for the wavelengths 370, 470, 520, 590, 660, 880, and 950 nm, respectively. By using Eq. (3) the C_{ref} at 637 nm would be 3.52. This is close to the reference value $C_{\text{ref}} = 3.48$ at 637 nm in Xianghe, China, approximately 900 km north of Nanjing, determined by Ran *et al.* (2016), by using a Multi-Angle Absorption Photometer (MAAP; Petzold and Schönlinner, 2004; Petzold *et al.*, 2005; Müller *et al.*, 2011) as a reference instrument. A MAAP is a recognized reference instrument because it has an internal nephelometer to correct for the artifact caused by multiple scattering.

Kim *et al.* (2018) used a linear regression method and parallel measurements of aerosol absorption coefficient with an aethalometer and a photoacoustic soot spectrometer, for estimating the multiple scattering correction factor. Their estimated C_{ref} values were 3.99, 4.48, and 5.46 at wavelengths of 405, 532, and 781 nm, respectively. The C_{ref} values obtained by Kim *et al.* (2018) are up to 44% higher than the values used in our study.

Estimating the Contribution of Non-BC Absorbers to the Absorption Coefficients

In AE-31 the absorption coefficient measured at the wavelength of 880 nm can be attributed to black carbon if there is no dust. The absorption attributed to BC at lower wavelengths can be extrapolated from 880 nm by using an AAE of unity (e.g., Kirchstetter and Thatcher, 2012). The difference between the extrapolated and measured absorption coefficients at wavelengths below 880 nm can be regarded as the fraction caused by non-BC absorbers. It must be noted that the aerosol mixing type (internal or external) may have an effect on the interpretation of the BC-derived absorption. Furthermore, the AAE for externally and internally mixed BC may not actually be unity but may vary in as wide a range as 0.55–1.7 (Lack and Langridge, 2013) because it depends on the particle size, shape, and chemical composition. Using a fixed value for AAE may lead to misinterpretations in estimating the absorption attributed to the non-BC absorbers. For example, Zhang *et al.* (2018a) used the AAE between 880 and 950 nm of 0.70, after ruling out the negligible lensing effect by possible coating, for the extrapolation to the lower wavelengths, and observed more than 20% higher non-BC contributions than those estimated by assuming an AAE of unity. Liu *et al.* (2018) estimated a relatively size-insensitive AAE of 1.05 and 0.90 for fresh and aged BC particles, respectively, with a wavelength-independent refractive index. In measurements the mixing type problem can be partly overcome by using a drier or a denuder before the analyzer. In our measurement we did not use a denuder before the aethalometer but the sample was diluted with dry air (1/3 ambient sample, 2/3 dilution air), which may have enhanced evaporation of semi-volatile compounds on the BC particles before the instrument.

In this work we calculated the BC contribution at lower than 880-nm wavelengths by using the Ångström exponent law:

$$B_{\text{abs,BC}}(\lambda) = B_{\text{abs}}(\lambda = 880 \text{ nm}) \times (\lambda/880)^{\text{AAE}} \quad (7)$$

where λ (nm) is one of the lower wavelengths (370, 470, 520, 590, or 660 nm). We assumed the AAE of unity throughout the whole dataset, so the Eq. (7) reduces to:

$$B_{\text{abs,BC}}(\lambda) = B_{\text{abs}}(\lambda = 880 \text{ nm}) \times (\lambda/880). \quad (8)$$

The estimated BC contribution to the B_{abs} at each lower wavelength, $B_{\text{abs,BC}}(\lambda)$, calculated from Eq. (8) was subtracted from the measured (and corrected) absorption coefficient at the same wavelength, $B_{\text{abs}}(\lambda)$, in order to obtain the estimated contribution of non-BC absorbers to the B_{abs} at those wavelengths, $B_{\text{abs,non-BC}}(\lambda)$. The relative contribution of non-BC absorbers to the absorption coefficient at each wavelength was calculated by using:

$$f_{\text{non-BC}}(\lambda) = B_{\text{abs,non-BC}}(\lambda)/B_{\text{abs}}(\lambda). \quad (9)$$

The non-BC contribution at the ultraviolet wavelength (370 nm in this case, calculated from Eq. (9)) provides a tool to estimate, for example, the existence of biomass-burning-

originated aerosols.

We also estimated the non-BC contribution by using the WDA method (Wang *et al.*, 2016). This approach uses calculated AAE for two wavelength pairs from absorption measurements, the difference between these two AAE, and Mie calculations, for predicting the existence of non-BC absorbers in particles. In this work the AAE were calculated for the wavelength pairs of 370/880 nm, $\text{AAE}_{370/880}$, and 660/880 nm, $\text{AAE}_{660/880}$, and the WDA was calculated as:

$$\text{WDA} = \text{AAE}_{370/880} - \text{AAE}_{660/880} \quad (10)$$

The calculated WDA values for each observation by using Eq. (10) were then compared with the WDA values calculated for an atmospherically relevant population of BC particles by using Mie theory. Fig. 4 illustrates this comparison: There is an additional contribution to the absorption coefficient by non-BC absorbers in the observations (marked with crosses) which lie above the shaded region, which, in turn, refers to absorption by BC only. For our Mie calculations we assumed spherical, coated particles and included lognormal particle size distributions with geometrical mean diameter (GMD) ranging from 20 to 300 nm, geometrical standard deviation (GSD) ranging from 1.4 to 2.2, and coating thickness ranging from 10% to 90% of the core diameter. We chose these values as typical for BC particles and to perform similar analysis as Wang *et al.* (2016) did. Accordingly, we used a refractive index of 1.95–0.79j for the BC core (Bond and Bergström, 2006) and 1.55–0.001j for the coating material, as also Wang *et al.* (2016) did. It must be noted that these values are not necessarily constant over time but may vary based on varying physical properties and chemical composition of the particles. Saturno *et al.* (2018) found that changing the BC core size range from 100–275 nm to 80–275 nm and using a fixed refractive index of 1.55–0.001j for the coating material may result in a relative overestimation of 18% in BrC contribution, whereas increasing the imaginary part of the refractive index of the coating material from 0.001 to 0.2 may result in a 56% relative overestimation in BrC contribution. Liu *et al.* (2018) found that variation in the BC refractive index (the real part in the range of 1.6–2.0 and imaginary part in the range of 0.4–1.0 in their study) is the most significant challenge in determining the BC AAE as the refractive index cannot be measured directly.

The main advantages of the WDA method, compared to AE analysis, are that it does not require concurrent measurement of scattering or extinction coefficient, which reduces the number of instruments needed at the measurement site, and that it does not rely on any fixed assumption concerning the absorption Ångström exponent for BC (fixed at unity in most AE analyses) when estimating the non-BC contribution but rather relies on the atmospherically relevant range of BC properties. Furthermore, it does not require corrections due to wavelength-independent artifacts. However, aethalometer data must be corrected for the filter loading artifacts before using the WDA method in order to avoid misinterpretation due to the wavelength dependency of the filter loading effect (Virkkula *et al.*, 2007, 2015).

It must be noted that one cannot compare directly the

non-BC absorber contribution to the absorption coefficient obtained from the AE analysis to the fraction of samples containing non-BC absorbers obtained from the WDA analysis because of the different nature of these two approaches. However, these two methods can be applied in parallel for the same dataset. The WDA analysis indicates, for example, the frequency of occurrence of episodes with particles containing also non-BC absorbers in addition to BC only. The AE analysis can be applied in further analysis, reflecting the actual amount of non-BC absorbers in the particles. However, both methods can be applied in, e.g., source apportionment when the data are combined with meteorological measurements or back trajectory analysis.

Trajectory Analysis

The backward air parcel trajectories ending at the measurement site used in our source apportionment analysis were calculated by using the PC-based HYSPLIT model (Stein et al., 2015; Rolph et al., 2017). The trajectories were calculated for the preceding 5 days (120 hours) with a 1-hour time resolution. The output is the coordinates and the height above ground level of the air parcel at each hour. Other outputs used in this study were rainfall and terrain height.

The trajectories were classified according to their coordinates into five sectors, for which we assumed different types of aerosols (Fig. 1). The sectors were 1) “Yellow Sea” (0–90°), 2) “East China Sea + Shanghai” (90–135°), 3) “Southern China” (135–225°), 4) “Southwestern China” (225–270°), and 5) “Northwestern China” (270–360°). Aerosols from Sectors 1–3 are a mixture of marine and continental origin. Sector 1 includes parts of northern China, the Korean Peninsula, and Japan. Sector 2 is the most “marine” sector but the marine air masses spend also some time over the highly polluted YRD region, including the megacity of Shanghai. The air masses from Sector 3 are a mixture of marine aerosol and the southern megacities such as the Guangzhou-Shenzhen-Hong Kong region whereas the air masses from Sectors 4 and 5 are totally of continental origin. The division between southwestern and northwestern China was made by thinking of temperature differences: In the northern part houses are heated during wintertime whereas in the southern part heating is not as extensive.

Source Area Analysis

In order to determine BC and non-BC absorber sources outside of the Nanjing area, air mass back trajectories were utilized together with the BC measurements. The basic idea for this source area analysis has been introduced by Hyvärinen et al. (2011) and is described here briefly. The region of eastern China around Nanjing was divided into a $0.5 \times 0.5^\circ$ regular grid. 72-hour air mass back trajectories were computed using the HYSPLIT model. The trajectories were computed every 3 hours for the period of the BC/non-BC measurements, leading to 7,614 trajectory-BC/non-BC measurement pairs. For each grid cell the number of “hits” from a trajectory corresponding to a BC measurement was recorded. This number represents the time that the air mass following a trajectory spends in a grid cell. When this procedure was repeated for each trajectory-BC/non-BC pair, an observation matrix, A , was constructed. The vector of measurements was denoted by b and the source BC/non-BC concentration vector was denoted by x . Now the source determination problems can be formulated as the equation $b = Ax$, and the unknown source vectors can be solved simply as an inverse parameter estimation problem by $x = A^{-1}b$. It must be noted that Matrix A of the source area analysis is very rank-deficient implying that some sort of regularization method must be used. The chosen method in our analysis was the minimum norm Tikhonov regularization (Tarantola, 1987).

RESULTS AND DISCUSSION

Meteorological Conditions

The monthly averages of selected meteorological parameters and the cumulative rainfalls are given in Table 2. The lowest and highest temperatures, -3.1°C and 37.3°C , were observed on 9 February 2015 and 4 August 2015, respectively. The humid summer with high relative humidity and rainfall and dry winter are typical near an east coast in a subtropic climate. This can also be seen in the monthly trajectory distribution (Fig. S3) with continental air masses in winter and maritime air masses in summer. The prevailing winds at the measurement site were from north and south (Fig. S4). The winds from the west at and in the vicinity of the

Table 2. Monthly averages $\pm$ std of meteorological parameters. T = temperature, RH = relative humidity, p = pressure, ws = wind speed.

Month	T ($^\circ\text{C}$)	RH (%)	p (mbar)	ws (m s^{-1})	Rainfall (mm)
November 2014	12.9 ± 2.9	65.0 ± 16.2	1018.5 ± 4.3	2.0 ± 1.1	48.2
December 2014	5.4 ± 3.1	49.1 ± 15.8	1024.0 ± 4.1	2.1 ± 1.2	2.3
January 2015	5.8 ± 3.8	57.6 ± 17.8	1022.1 ± 5.8	2.0 ± 1.0	11.3
February 2015	5.0 ± 3.9	50.6 ± 18.1	1023.1 ± 6.5	1.8 ± 1.0	0.2
March 2015	11.3 ± 5.7	64.5 ± 16.1	1016.0 ± 6.3	2.2 ± 0.9	47.4
April 2015	16.2 ± 5.9	56.7 ± 19.2	1011.2 ± 7.1	2.4 ± 1.4	101.6
May 2015	22.0 ± 3.5	58.4 ± 17.4	1005.0 ± 3.6	2.3 ± 1.1	57.8
June 2015	24.8 ± 3.1	70.8 ± 16.3	1000.3 ± 2.9	2.2 ± 0.9	561.0
July 2015	26.3 ± 4.0	69.0 ± 13.3	1000.6 ± 4.0	2.2 ± 1.2	79.5
August 2015	27.7 ± 3.4	66.8 ± 13.7	1002.5 ± 2.1	2.1 ± 1.1	267.6
September 2015	23.5 ± 2.5	65.8 ± 13.6	1009.3 ± 3.5	1.9 ± 1.0	76.3
October 2015	19.1 ± 3.5	60.1 ± 16.0	1015.6 ± 3.9	1.9 ± 1.1	19.2

measurement site have been sparse (Ding *et al.*, 2013; Chen *et al.*, 2017).

Equivalent Black Carbon Concentration

The annual average $\pm$ std of the measured EBC concentration at 880 nm was $4.0 \pm 3.3 \mu\text{g m}^{-3}$ with a maximum of $50.9 \mu\text{g m}^{-3}$ (Fig. S5(a)). The annual average of the EBC concentration is 2–5% lower than the values of $4.1 \pm 2.8 \mu\text{g m}^{-3}$ and $4.2 \pm 2.6 \mu\text{g m}^{-3}$ measured at the nearby SORPES station during July 2013–May 2015 (Shen *et al.*, 2018; re-calculated from the 2-year average of absorption coefficient of 16 Mm^{-1} , by using Eqs. (2) and (5) and the reported C_{ref} value of 4.26) and in Nanjing city center during the year 2012 (Zhuang *et al.*, 2014), respectively. Zhuang *et al.* (2017) reported an average absorption coefficient of 29.615 Mm^{-1} at 550 nm in the Nanjing city center for the time period March 2014–February 2016. By using the given statistics and Eqs. (2), (3) and (5), the average EBC concentrations at 660 nm and 880 nm are estimated to be $4.0 \mu\text{g m}^{-3}$ and $3.8 \mu\text{g m}^{-3}$, respectively. Our values, $4.0 \mu\text{g m}^{-3}$ at both 660 nm and 880 nm, are within 5% of those calculated from the Zhuang *et al.* (2017) data. The differences in the EBC concentrations measured at our site, the SORPES station, and Nanjing city center may arise from bigger BC emissions in the city center, or a decreasing trend in the annual average of BC concentrations, which can be deduced from the EBC concentrations of $4.2 \mu\text{g m}^{-3}$ for 2012 (measured at 880 nm by Zhuang *et al.*, 2014) and $3.8 \mu\text{g m}^{-3}$ for 2014–2016 (estimated at 880 nm from Zhuang *et al.*, 2017) in Nanjing city center. Furthermore, differences in the averaging periods, experimental setups, and measurement errors, as well as data processing procedures may complicate the comparison.

The seasonal averages $\pm$ std of EBC concentrations at 880 nm were $3.5 \pm 3.6 \mu\text{g m}^{-3}$ in the spring (March–May), $4.0 \pm 3.1 \mu\text{g m}^{-3}$ in the summer (June–August), $3.3 \pm 2.3 \mu\text{g m}^{-3}$ in the autumn (September–November), and $5.5 \pm 3.8 \mu\text{g m}^{-3}$ in the winter (December–February). It must be noted that the gaps in the aethalometer data before mid-January 2015 may have an influence on the results. We estimated this influence by calculating the averages of PM_{10} and $\text{PM}_{2.5}$ that had far less gaps. During this period the averages for PM_{10} were 101.8, 103.6, and $100.6 \mu\text{g m}^{-3}$ for the whole dataset, for the hours when AE data was available, and for the hours when AE data was unavailable, respectively. For $\text{PM}_{2.5}$ the corresponding values were 58.2, 59.0, and $57.7 \mu\text{g m}^{-3}$, respectively. Based on these calculations, we may have overestimated the monthly averages of EBC concentrations but by no more than 3%.

The EBC concentration varied also diurnally (Fig. S6).

The diurnal variation pattern was similar for each season, showing the lowest concentrations at 1–2 p.m. when the mixing depth is at its highest, and highest concentrations during the night. At around 7 a.m., a small increase in the EBC concentration was seen, probably due to morning traffic and its emissions. The pattern was similar to those measured at the same site (Chen *et al.*, 2017), at the nearby SORPES station (Shen *et al.*, 2018), and in the Nanjing city center (Zhuang *et al.*, 2014).

With the measured $\text{PM}_{2.5}$ and PM_{10} hourly averages (Fig. S5(b)), the annual average $\pm$ std of the EBC/ $\text{PM}_{2.5}$ and EBC/ PM_{10} were 0.14 ± 1.36 and 0.14 ± 2.9 , respectively. The high standard deviation arises from the fact that the calculated fractions become inaccurate and large with low PM concentrations. The 1st, 50th (median), and 99th percentiles for the EBC/ $\text{PM}_{2.5}$ and EBC/ PM_{10} fractions were 0.015, 0.073, and 0.49, and 0.009, 0.046, and 0.45, respectively.

The median EBC concentration in the air masses from Sectors 1–5 ranged from 2.7 to $4.6 \mu\text{g m}^{-3}$ (Table 3). Overall, the highest EBC concentrations were observed in the air masses from the southern sector (Sector 3) and with southerly winds, except during the autumn and winter (Fig. S7). The average EBC concentration in Sector 3 may be, however, biased by the fact that it was the main sector more seldom than the northern sectors, Sectors 1 and 5 (number of observations = 3727, 693, 713, 354, and 3153 for Sectors 1–5, respectively), and mainly in summer (Figs. S3 and S15). Furthermore, in most cases there had been rain relatively close to the measurement site (Fig. S3), which means that the higher EBC concentration in the air masses from Sector 3 (Table 3) can be attributed to emission sources in the vicinity of the measurement site rather than to long-range transported aerosol.

Aethalometer Equivalent Black Carbon and Thermal-optical Elemental Carbon Comparison

The hourly average EC concentration had an annual average $\pm$ std of $3.3 \pm 2.0 \mu\text{g m}^{-3}$ and a maximum of $37.6 \mu\text{g m}^{-3}$ (Fig. S8(a)). This is continuation for the earlier EC measurements at the same site by Chen *et al.* (2017), who observed an annual average EC concentration of $3.2 \mu\text{g m}^{-3}$ for the year 2014. By calculating the monthly averages for our EC data and combining them with those calculated by Chen *et al.* (2017) we can see that the EC concentration decreased at an average rate of $0.078 \mu\text{g m}^{-3}$ per month between June 2013 and October 2015 (Fig. S9). This may be due to an overall improvement of regional air quality thanks to reduction of emissions, or local actions taken in order to improve air quality. For example, actions were taken to reduce pollutant emissions before and during the Nanjing

Table 3. Medians of several parameters from different trajectory analysis sectors.

	Sector 1	Sector 2	Sector 3	Sector 4	Sector 5
EBC conc ($\mu\text{g m}^{-3}$)	2.68	2.69	4.58	3.92	3.25
EC conc ($\mu\text{g m}^{-3}$)	2.56	2.67	3.17	3.51	3.11
MAE	4.41	4.71	6.16	4.98	4.35
non-BC fraction AE (%)	26.3	24.5	25.8	28.1	29.5
non-BC contribution WDA (%)	22.2	30.6	21.2	30.5	21.3

2014 Youth Olympic Games (YOG) that took place between 16–28 August 2014. The effect of the emission reductions can be seen as a lower EC concentration during the summer of 2014 compared to that during the summer of 2015 (Fig. S9). Also reduction in other air pollution components during the YOG have been reported (Miettinen *et al.*, 2019; Zhao *et al.*, 2017a; Zhou *et al.*, 2017).

The seasonal averages of the EC concentration were $2.8 \pm 1.3 \mu\text{g m}^{-3}$ (spring), $2.8 \pm 1.9 \mu\text{g m}^{-3}$ (summer), $3.2 \pm 1.8 \mu\text{g m}^{-3}$ (autumn), and $4.5 \pm 2.4 \mu\text{g m}^{-3}$ (winter). Chen *et al.* (2017) reported seasonal average EC concentrations of 3.41, 2.51, 2.54, and $4.37 \mu\text{g m}^{-3}$ for spring 2014, summer 2014, autumn 2014, and winter 2014–2015, respectively. The EBC and EC concentrations in the winter were 1.4–1.6 times that during the other seasons, which can be at least partly explained by an increased need of energy for heating of housings during the cold seasons, which increases emissions from power plants, and partly by the 1.2-fold average of mixing depth over the other seasons (462 m) than over winter (391 m), which causes the emissions to accumulate in a lesser volume in winter. It must be noted that there were gaps in our aethalometer data due to instrument or data acquisition system malfunction, which may cause bias to the calculated averages, particularly for the summer values when a longer break in the valid data was experienced between 19–29 July 2015. However, we expect the calculated summertime average for the aethalometer data to represent well the summer average, because during this 10-day time period the average $\pm$ std of EC concentration was $3.0 \pm 1.6 \mu\text{g m}^{-3}$ which is close to the average $\pm$ std of the EC concentration of $2.8 \pm 1.9 \mu\text{g m}^{-3}$ for the whole summer. The EC concentrations were highest during winter and in the air masses from the southern sector (Fig. S10).

The hourly ratio of EBC to EC concentration had high variability (Figs. S8(b) and S11). The linear relationship is $[\text{EBC}] = 1.05[\text{EC}]$ (Fig. S11). Zhao *et al.* (2017b) reported a linear relationship of $[\text{BC}] = 0.6812[\text{EC}] + 0.9683$ to the values measured in Nanjing city center in 2015. Zhi *et al.* (2014) observed a BC/EC ratio of 0.92 in Shanghai on clear weather, while during heavy haze days the ratio increased up to 1.9. Haze may also play a role in our observations in Nanjing. The differences in the EBC/EC ratios may arise from differences in the mixing state of particles, i.e., internal mixing of EC with other species. It is known that an organic coating may enhance light absorption into an aerosol particle (e.g., Bond *et al.*, 2013), which increases the EBC/EC ratio.

It must be noted that our estimation for the EBC in the $\text{PM}_{2.5}$ fraction relies solely on the particulate masses in $\text{PM}_{2.5}$ and in the fraction of larger than 2.5- μm particles. BC, however, resides in the smaller submicron size fraction, whereas the supermicrometer size fraction is dominated by dust (Clarke *et al.*, 2004). Therefore, we conclude that our EBC mass concentrations can be safely used in comparison with the PM_{10} and $\text{PM}_{2.5}$ mass concentrations, as well as in comparison with the EC concentration measured from the $\text{PM}_{2.5}$ fraction, even though our sampling system had an estimated cut-off diameter of 6.5 μm . It must also be noted that the larger dust particles that are not removed by any pre-cut instrument in our case, may scatter light effectively, which

would cause enhanced scattering by the sample collected on the aethalometer filter, leading to an overestimation of absorption. In order to take this into account, we calculated the scattering by applying Mie theory to the measured size distributions.

By using Eq. (1) we obtained an average MAE of $5.3 \text{ m}^2 \text{ g}^{-1}$ at 880 nm. It must be noted that MAE is not constant over time (Fig. S8(c)). During our campaign the MAE varied between $0.98 \text{ m}^2 \text{ g}^{-1}$ (1st percentile) and $19.9 \text{ m}^2 \text{ g}^{-1}$ (99th percentile). Elevated MAE values were observed in winter with westerly winds and in summer with southerly winds (Fig. S12). The average MAE values for Sectors 1–5 were 5.28, 5.82, 6.98, 5.49, and $4.84 \text{ m}^2 \text{ g}^{-1}$, respectively. The MAE values are inversely proportional to the non-BC fraction obtained from the AE analysis (Table 3). Cheng *et al.* (2011) attributed a lower MAE value of EC to an increased impact of biomass burning aerosols. Correspondingly, the highest MAE values in Sector 3 in our study may be attributed to aerosols from nearby sources and a relatively fresh aerosol with a lower non-BC fraction.

Our observations are in conjunction with the fact that the MAE bound to BC only, MAE_{BC} , has been observed to vary between a range of 2–39 (Quinn and Bates, 2005; Cheng *et al.*, 2011; Bond *et al.*, 2013). The variation is at least partly connected to abundant organic carbon and organic-to-elemental carbon ratio (OC/EC), particularly those in the secondary organic aerosol after atmospheric transformation, as was found, for example, by Cheng *et al.* (2011) in Beijing, China, and Zhang *et al.* (2018b) in Xi'an, China. The highest MAE values have been assigned to the internally mixed core-shell particle type or to elevated ambient relative humidity, as a coating on BC particles acts as a lens that may enhance absorption (Quinn and Bates, 2005; Bond *et al.*, 2013; Ammerlaan *et al.*, 2017).

Contribution of Non-BC Absorbers to the Absorption Coefficients

The annual averages $\pm$ std of the absorption coefficients at 370, 520, and 880 nm, for example, were 54 ± 44 , 32 ± 25 , and $17 \pm 14 \text{ Mm}^{-1}$, respectively, with a large variation (Fig. 2(a)). The averages are 38%, 23%, and 6% higher than the values of 39 ± 30 , 26 ± 19 , and $16 \pm 11 \text{ Mm}^{-1}$ observed at the SORPES station during July 2013–May 2015 (Shen *et al.*, 2018). The differences may arise from the different time spans of the two datasets, although there is an overlap of seven months. The annual average $\pm$ std of the absorption Angström exponent between 370 nm and 880 nm, $\text{AAE}_{370/880}$, was 1.05 ± 0.32 . The $\text{AAE}_{370/880}$ has maxima in the winter and in the summer (Fig. 2(b)). Ran *et al.* (2016) found a similar pattern (except for June) for the AAE in the North China Plain during April 2013–March 2015. Bukowiecki *et al.* (2019) linked an elevated AAE to high biomass burning periods in northern Southeast Asia.

The annual averages of the absorption coefficient and the non-BC attributed absorption at 370 nm, calculated by using the AE method, were 54.0 Mm^{-1} and 14.4 Mm^{-1} , respectively (Fig. 3), and the median and 99th percentile of the relative contribution of the non-BC absorbers at 370 nm were 27.2% and 64.8%, respectively. The relative non-BC contribution

Fig. 2. A time series of (a) absorption coefficient at 370 (light gray) and 880 nm (black) and (b) the absorption Ångström exponent (AAE) between 370 nm and 880 nm.

Fig. 3. Long-term average of absorption coefficient and the contributions of all, BC, and non-BC absorbers at each measured wavelength.

was highest in December and in June, and lowest in March (Fig. 5). The elevated non-BC contribution in December and June may also be attributed to open biomass burning during post-harvest seasons, typically taking place in May–June and October–November (Cheng *et al.*, 2014). Ran *et al.* (2016) found a 33–40% BrC contribution during non-heating seasons, and up to 59% contribution during the heating season.

The elevated contribution of non-BC absorbers during the heating season may arise from higher organic emissions (Chen *et al.*, 2017).

The WDA analysis showed that there were non-BC absorbers, on average, in 25.3% of the samples during the measurement period (Fig. 4). Also here, monthly variation was observed (Figs. 6 and S13), ranging from 8% (January

Fig. 4. WDA as a function of $AAE_{660/880}$ for the Mie-calculated BC and aethalometer measurements. In the measurements above the shaded area, non-BC contribution to absorption coefficient is expected.

Fig. 5. The relative contribution of non-BC absorbers to the absorption coefficient at 370 nm, obtained from the AE analysis, as a function of time, as hourly and monthly averages.

2015) to 43% (May 2015). The fraction of samples with non-BC contribution was lower in winter and higher in spring, which reflect the higher and lower BC concentration and absorption coefficient at 880 nm in winter and spring, respectively (Figs. S5(a) and 2(a)). It must be noted that the number of valid samples for the WDA analysis was lower during the winter due to the gaps in the aethalometer data

before mid-January 2015 (Fig. S13), which resulted in an overestimation of 3% in BC concentration as discussed earlier. This may also have an influence on the estimated non-BC contribution in winter. Unfortunately, due to missing AE data, a more detailed estimate for this influence is not possible.

In Fig. S14 the median non-BC contribution is given as a function of wind direction for different seasons and for the

whole measurement period. This figure shows that there could be sources of non-BC absorbers, such as biomass burning, in the directions of northwest, where the median of the non-BC contribution was 34.6% during the whole measurement period and 41.1% in winter, and in the direction of southeast with a 38.6% median non-BC contribution in winter.

Particles in the air masses originating from the continent (Sectors 4 and 5) contain more non-BC absorbers than those in the air masses originating from the marine sectors (Fig. 7). The highest non-BC contribution is in the particles originating from the northwestern sector (Sector 5). The probabilities of existence of non-BC absorbers in the samples from Sectors

Fig. 6. Monthly distribution of the fraction of samples containing non-BC absorbers in addition to BC.

Fig. 7. Frequency distribution of relative non-BC absorber contribution to absorption coefficient for (a) all trajectories and (b–f) Sectors 1–5.

1–5 are 22.2%, 30.6%, 21.2%, 30.5%, and 21.3%, respectively. This may be linked to the observations that the northwestern sector dominated during the wintertime (Fig. S3) and that elevated BrC concentrations in Xi'an, to the northwest of Nanjing, have been observed in winter (Huang *et al.*, 2018). During the spring and summer months the air masses were mainly of maritime origin in the east and south directions (Fig. S15), which was also observed in Nanjing in 2013–2016 by Cheng *et al.* (2017).

It must be noted that our sector classification is based strictly on the highest fraction of the time spent in that sector (called “main sector”) and that the air masses may also have spent almost as much time in another sector, as can be seen in the monthly distribution of the trajectories into different sectors (Fig. S15). This is true, in particular, for the trajectories which are near the sector borders. On average, the residence time in the main sector during the last 120 hours was 74–92 hours, i.e., 61.5–76.5% of the time. The shortest residence time, 74 hours, was for Sector 4, whose occurrence was also lowest, 4.1%. For the other sectors, the occurrences and residence times were, respectively, 43.1% and 89 hours (Sector 1), 8.0% and 76 hours (Sector 2), 8.3% and 92 hours (Sector 3), and 36.5% and 82 hours (Sector 5). Our analysis relies on straightforward application of latitude and longitude of the backward trajectories, and some bias may arise from ignoring the air mass height and meteorological conditions along the trajectory. For example, a heavy rain can reduce

the air pollutant concentrations remarkably, or intense solar irradiation may alter the properties of the aerosols in the air mass. During the local rain events during our campaign the PM₁₀ and PM_{2.5} concentrations were observed to decrease. After the rain event the concentrations either increased back to the pre-rain level, or remained low. For the BC concentration and the contribution of non-BC absorbers to the absorption coefficient, the rain events had only a minor effect.

As each sector covers relatively big areas, it may be insufficient to apply local emission inventory to evaluate the source. In particular, the biomass burning from the northwestern sector (Sector 5) could be verified by a recent study (Yang and Zhao, 2019), which suggested intensive biomass burning emissions in northern provinces of Jiangsu and Anhui. Sector 5 was also the main sector in May–June and October–November (Fig. S3), the typical times for post-harvest open biomass burning in eastern China (Cheng *et al.*, 2014; Li *et al.*, 2014).

Source Area Analysis: Black Carbon and Non-BC Absorbers

The minimum norm Tikhonov regularization method may mean that the actual absolute values for the source concentrations are not to be taken as “the truth,” at least not before some sort of a validation is done. For this reason, only relative source concentrations are shown in Fig. 8 by normalizing the resulting source vectors x . However, the

Fig. 8. The results of the source area analysis presented as source distribution maps: (a) high-BC sources, (b) high non-BC contribution, (c) low-BC sources, (d) low non-BC contribution. All maps have been normalized by maximum values for each case. Source concentration is indicated by darkness (darker color means a higher value).

spatial distribution of the solution should be quite reliable. It must be noted that here the setting of the source area analysis differs considerably from the one presented in Hyvärinen *et al.* (2011) who exploited measurements from several measurement sites located around Finland instead of only one measurement site in this study. The lateral information in the back trajectories, i.e., the trajectory coordinates away from the strictly radial direction, becomes crucial when only one measurement site is available; otherwise only the direction of a source could be estimated, but not the distance of the source from the site.

To study the effect of BC and non-BC contribution, we divided the normalized values into two categories: “low” when the value was below the median ($3.0 \mu\text{g m}^{-3}$ for BC concentration, 0.272 for non-BC contribution), and “high” when the value was equal or greater than the median. Fig. 8(a) shows that the highest-BC sources reside in the vicinity of the measurement site and mostly in the northeast direction. Fig. 8(c), in turn, shows that the low-BC sources reside mainly in the sector between north and east of the measurement site, and thus the average BC concentration in these directions is lower than, e.g., in the south (Fig. S7). Shen *et al.* (2018) made a similar observation of lower BC concentration in the northeast direction from the nearby SORPES station, and they attributed this to a low photochemical age and thus a relatively close BC source, which supports our finding of the highest-BC sources in the same direction (Fig. 8(a)). There are also both high-BC sources and the highest end of low-BC sources in the south, which can also be seen as an elevated average BC concentration in this direction (Fig. S7). This may be attributed, at least partly, to emissions from the Nanjing city center, as was also observed by Shen *et al.* (2018).

The higher non-BC contribution (Fig. 8(b)) shows a similar pattern to the low-BC sources (Fig. 8(c)), indicating the abundance of non-BC absorbers in particles with less BC. The low-non-BC contribution pattern (Fig. 8(d)) is more scattered than the high-non-BC-contribution pattern (Fig. 8(c)), and it can be seen that the highest non-BC sources reside in the vicinity of the measurement site and in the southwest and northwest directions. The chemical analysis by Shen *et al.* (2018) revealed elevated scattering in west-northwest from the nearby SORPES station, and they suggested that aged biomass burning aerosol could be one factor for the elevated scattering. The non-BC contribution is, in turn, lower in the particles coming from a longer distance.

CONCLUSIONS

Based on both previous studies (e.g., Yang and Zhao, 2019) and the trajectory and source area analyses conducted in this study, we identify biomass burning emissions to the northwest of Nanjing as a source candidate for non-BC absorbers. The source area analysis also reveals that local emissions play a major role in both BC concentrations and non-BC absorber contributions, although particles arriving from farther away were also observed to exert an influence.

Additionally, we agree with the conclusion drawn by

Miettinen *et al.* (2019), Zhao *et al.* (2017a), and Zhou *et al.* (2017) that the emission control measures implemented during the Nanjing Youth Olympic Games (YOG) in August 2014 improved the air quality in Nanjing. Combining our observations with those of Chen *et al.* (2017), we found that the elemental carbon (EC) concentrations were lower on average during August 2014 than during the same month in the preceding or the following year. Thus, although the improvement may have been temporary, the control measures produced a noticeable effect. Furthermore, the EC concentration trendline illustrated in this study suggests that the overall air quality in Nanjing has potentially improved in recent years.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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