Response to reviewer's comments

We thank the referee for the useful comments and suggestions which have helped us to improve the manuscript. Our point-by-point responses are below. The referee' comments are in black font and our responses are in blue font.

19-Feb-2021

Title: Highly time-resolved characterization of carbonaceous aerosols using a two-wavelength Sunset thermo/optical carbon analyzer

Author(s): Mengying Bao et al.

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Interactive comment on "Highly time-resolved characterization of carbonaceous aerosols using a two-wavelength Sunset thermo/optical carbon analyzer" by Mengying Bao et al.

Anonymous Referee #3

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As far as I understood it is for the first time real-time measurement of OC EC determination using a dual wavelength Sunset. The method is a milestone for a new technical to study EC or brown carbon. The paper is well structured and present a very new dataset which may be helpful for the scientific community. I recommend for a publication in AMT after they may address the following comments.

R: We thank the reviewer for the brief summary and positive comments on our paper.

Method: because the most important work in this study should be new instrument setup of new type of Sunset, I would suggest move this part to the very beginning part of method.

R: We thank the reviewer for the suggestion. Considering the site description was also very important where all the real-time observations and sampling were conducted, we kept the site description in the front and put the "Two-wavelength TOT measurement" after the site description.

A typical thermogram of analysis including information of temperature, NDIR values (CO2) and transmittance in two wavelengths should be added.

R: We agree with the reviewer that we need to add a figure with an example of a typical analysis. We added the sentence "The split point between OC and EC was detected by the RTCalc731 software provided by Sunset Lab. The principle was same as the traditional Sunset carbon analyzer (Birch and Cary, 1996). An example thermogram of sample analysis using the two-wavelength Sunset semi-continuous carbon analyzer was shown in Fig. 2. During the sample analysis, the laser beam at 658 nm and 405 nm were both sent through the filter and the transmitted light signal were monitored to correct the undesired formation of pyrolyzed carbon (PyrC) and then to determine the split point of OC and EC at both two wavelengths." in lines 170-176.

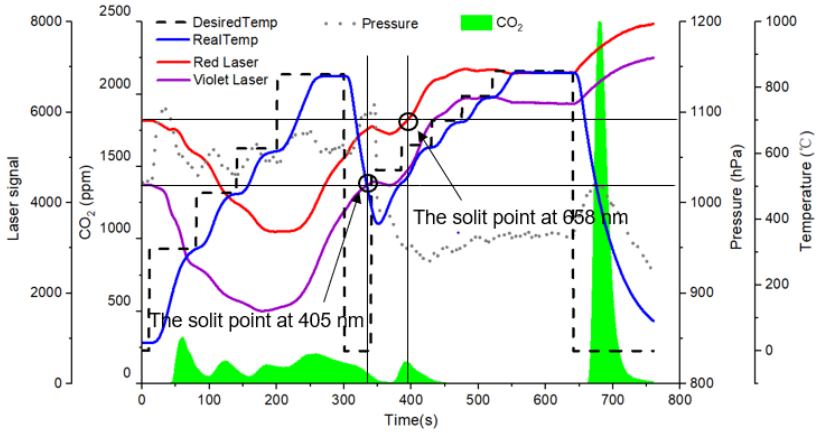

Figure 2. Example thermogram of sample analysis using the two-wavelength sunset semi-continuous carbon analyzer.

Figure 3: high dEC/OC was found in winter (Jan, Feb), whereas high OC/EC was found in late spring and summer. Such a different seasonal (and diurnal in Figure 4) trend indicate dEC/OC is not an indicator for SOC but rather an indicator of anthropogenic tracer. The seasonal variation of different carbonaceous should be discussed more carefully. Monthly and diurnal cycles of dEC may be added.

R: We thank the reviewer for the suggestions. We added the monthly and diurnal cycles of dEC in Figure 5 and Figure 6 in the revised manuscript and the sentence "High dEC/OC was found in January and February in winter, indicating strong influence of anthropogenic sources on dEC, such as coal combustion. In addition, we found strong biomass burning activities in February, which

significantly contributed to the high concentrations of dEC in February, more details could be found in section 3.3." in lines 306-309 in the revised manuscript. We added the sentence "Similar though not so obvious diurnal variations were found in dEC." in lines 339-340.

Our results did show that the sources of dEC were complicated. Anthropogenic sources could contribute to dEC. In conclusion, we pointed out that "The results showed that high (low) OC, EC and dEC concentrations were found in Winter (summer), indicating the significant impact of the increase of various emission sources in winter and wet scavenging of rain in summer." and "It should be noted that the sources of dEC were complicated and the anthropogenic emissions and secondary formations of dEC aerosols couldn't be ignored , further chemical analysis need to be conducted in the future. We also hope that the dEC data can be further applied in more researches." Using the MODIS fire information and receptor model, we proved that biomass burning significantly contributed to high dEC concentrations. However, with the limited data, the anthropogenic or secondary sources of dEC couldn't be quantified. We hope further analysis of this work can be done in the future.

The source of dEC may be linked to BrC, but this remains unclear. I suggest the authors should include study outlook to resolve this problem.

R: We added the sentence "The evaluation of SOC formation and the relationship between dEC and SOC can be conducted. In addition, More chemical analysis such as the analysis of the ion, the organic matter or the sugars in $PM_{2.5}$ can be measured, thus we can get some information of the tracers of different sources and more accurate and quantitative source apportionment can be done (Bhattaraia et al., 2019;Wu et al., 2019;Wu et al., 2018). We also hope that the dEC data can be further applied in more researches." at the end of the revised manuscript. (see lines 459-464)

Reference:

Bhattaraia, H., Saikawac, E., Wana, X., Zhue, H., Ram, K., Gao, S., Kang, S., Zhanga, Q., Zhang, Y., Wu, G., Wang, X., Kawamura, K., Fui, P., and Cong, Z.: Levoglucosan as a tracer of biomass burning recent progress and perspectives, Atmos. Res., 220, 20-33, 10.1016/j.atmosres.2019.01.004, 2019.

Birch, M. E. and Cary, R. A.: Elemental carbon-based method for occupational monitoring of particulate diesel exhaust: methodology and exposure issues, Analyst, 121, 1183-1190, 1996.

Wu, G., Wan, X., Gao, S., Fu, P., Yin, Y., Li, G., Zhang, G., Kang, S., Ram, K., and Cong, Z.: Humic-Like Substances (HULIS) in Aerosols of Central Tibetan Plateau (Nam Co, 4730 m asl): Abundance, Light Absorption Properties, and Sources, Environ. Sci. Technol., 52, 7203-7211, 10.1021/acs.est.8b01251, 2018.

Wu, G., Ram, K., Fu, P., Wang, W., Zhang, Y., Liu, X., Stone, E. A., Pradhan, B. B., Dangol, P. M., Panday, A. K., Wan, X., Bai, Z., Kang, S., Zhang, Q., and Cong, Z.: Water-Soluble Brown Carbon in Atmospheric Aerosols from Godavari (Nepal), a Regional Representative of South Asia, Environ. Sci. Technol., 53, 3471-3479, 10.1021/acs.est.9b00596, 2019.

Highly time-resolved characterization of carbonaceous aerosols using a two-wavelength Sunset thermo/optical carbon analyzer

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Abstract

Carbonaceous aerosols have great influence on the air quality, human health and climate change. Except for organic carbon (OC) and elemental carbon (EC), brown carbon (BrC), mainly originates from biomass burning, as a group of OC with strong absorption from the visible to near-ultraviolet wavelengths, makes a considerable contribution to global warming. Large amounts of studies have reported long-term observation of OC and EC concentrations throughout the word, but studies of BrC based on long-term observations are rather limited. In this study, we established a two-wavelength method (658 nm and 405 nm) applied in the Sunset thermo/optical carbon analyzer. Based on one-year observation, we firstly investigated the characteristics, meteorological impact and transport process of OC and EC. Due to BrC absorbs light at 405 nm more effectively

than 658 nm, we defined the enhanced concentrations (dEC =$EC_{405\ nm}$—$EC_{658\ nm}$) and gave the possibility to provide an indicator of BrC. The receptor model and MODIS fire information were used to identify the presence of BrC aerosols. Our results showed that the carbonaceous aerosols concentrations were highest in winter and lowest in summer. Traffic emission was an important source of carbonaceous aerosols in Nanjing. Receptor model results showed that strong local emissions were found in OC and EC aerosols, however dEC aerosols were significantly affected by regional or long-range transport. The dEC/OC and OC/EC ratios showed similar diurnal patterns and the dEC/OC increased when the OC/EC ratios increased, indicating strong secondary sources or biomass burning contributions to dEC. Two biomass burning events both in summer and winter were analyzed and the results showed that the dEC concentrations were obvious higher in biomass burning days, however, no similar levels of the OC and EC concentrations were found both in biomass burning days and normal days in summer, suggesting that biomass burning emission made a great contribution to dEC and the sources of OC and EC were more complicated. Large number of open fire counts from the northwest and southwest areas of the study site were monitored in winter, significantly contributed to OC, EC and dEC. In addition, the near-by YRD area was one of the main potential source areas of dEC, suggesting that anthropogenic emissions could also be important sources of dEC. The results proved that dEC can be an indicator of BrC in biomass burning days. Our modified two-wavelength instrument provided more information than traditional single-wavelength thermo/optical carbon analyzer and gave a new idea about the measurement of BrC, the application of dEC data need to be further investigated.

1. Introduction

Carbonaceous aerosols including organic carbon (OC) and elemental carbon (EC), which have significant influence on the global radiative transfer, human health and atmospheric visibility, have been the focus of research in the atmospheric environment field for many years (Lelieveld et al., 2015; Wu and Yu, 2016; Wang et al., 2018; Zhang et al., 2017; Liu et al., 2019; Zhang et al., 2019). EC mainly originates from fossil fuel and biomass combustion and is estimated to be the second largest warming factor behind CO_2 contributing to climate change (Liu et al., 2015; Zhang and Kang, 2019; Cao and Zhang, 2015). OC originates both from primary emissions and gas-to-particle conversion as secondary organic carbon (SOC) and can scatter the solar radiation which causes negative forcing globally (Zhou et al., 2014; Huang et al., 2014).

In the recent decades, brown carbon (BrC), as a kind of light-absorbing organic carbon which can absorb light especially from near-UV to visible wavelength, has caused global concern due to its positive climate effect (Andreae and Gelencsér, 2006; Zhang et al., 2020). BrC is mainly emitted from anthropogenic and biogenic emissions (Zhang et al., 2011). Previous studies have proved that biomass burning and biofuel combustion are the most important sources of primary BrC (Saleh et al., 2014; Wu et al., 2020; Lei et al., 2018). Recent researches reported that in developing countries such as China and India, the contribution of fossil fuel combustion to BrC can't be ignored (Satish et al., 2017; Yan et al., 2017; Kirillova et al., 2014). Secondary BrC is mainly emitted from heterogeneous photo-oxidation reactions or aqueous reactions of anthropogenic and biogenic precursors (Zhang et al., 2020; Li et al., 2020; Zhang et al., 2011). However, due to the lack of understanding of BrC at the molecular level and in situ BrC data, there are still large uncertainties in the estimates of the distribution and the magnitude of BrC climate effect in both remote sensing and modeling method (Arola et al., 2011; Feng et al., 2013).

The thermo-optical analysis (TOA) method is one of the most widely used quantitative method of OC and EC taking use of the difference between the thermo-optical properties of OC and EC (Birch and Cary, 1996; Chow et al., 2004). OC and EC will be volatilized at different heating protocol. The reflectance/transmittance of one laser source (near-infrared wavelength) through the sample filter are continuously monitored and return of the reflectance/transmittance to its initial value on the thermograph was taken as a split point between OC and EC. This way, the formation of pyrolyzed carbon which can also absorb the light and make the sample darker, is corrected. This method has been wildly used in present studies applied in the NIOSH protocol or IMPROVE_A protocol (Ji et al., 2016; Chow et al., 2007). However, the thermo-optical approach assumed that EC is the only light-absorbing species, the presence of BrC, which is part of OC but also a light-absorbing component, shifts this separation towards EC, resulting in overestimated EC values and underestimated OC values (Chen et al., 2015; Birch and Cary, 1996).

Sandradewi et al. (2008) pointed out that light absorption measurements at different wavelength by the aethalometer can be used to quantify the contributions of wood combustion and traffic emissions to aerosols since wood smoke contains organic compounds which enhance the light absorption in ultraviolet wavelength. But traffic emissions produce more BC, which dominates the light absorption in near-infrared wavelength. They took use of aethalometer data measured at 470 nm and 950 nm to quantify the BC distinction between wood burning and traffic emission. With the

similar principle, Wang et al. (2011) used a two-wavelength Aethalometer (370 and 880 nm) to identify the presence of residential wood combustion (RWC) particles which was closely associated with BrC. Organic components of wood smoke particles absorb light at 370 nm more effectively than 880 nm in two-wavelength aethalometer measurements. They believed that the enhanced absorption (Delta-C=BC_{370nm}-BC_{880nm}) can serve as an indicator of RWC particles. This method was further used by Wang et al. (2012a) and Wang et al. (2012b). Chen et al. (2015) used a modified seven-wavelength TOT/TOR instrument (Thermal Spectral Analysis – TSA) allowing the determination of the OC-EC split at different wavelengths and light absorption measurements to be made with wavelength-specific loading corrections, providing additional information including the optical properties of black carbon (BC) and BrC from the IR to UV parts of the solar spectrum and their contributions. Massabò et al. (2016) further corrected the OC/EC split point using the Multi-Wavelength Absorbance Analyzer (MWAA) which provides the aerosol absorbance values at five wavelengths from IR to UV together with a Sunset OC/EC analyzer to achieve the BrC concentration. With a set of samples collected wintertime in the Ligurian Apennines in Italy, clear correlations were found between the BrC and levoglucosan mass concentration. A further step of BrC quantification taking use of TSA was reported by Chow et al. (2018), further proving that the use of seven wavelengths in thermal–optical carbon analysis allows contributions from biomass burning and secondary organic aerosols to be estimated. Their results clearly demonstrated the role of BrC in the thermo-optical analysis. However, these techniques focus on the light absorption measurement of BrC and are still limited reported in previous researches, though they provide quartz-fiber filter samples that are currently being characterized for organic carbon (OC) and EC by thermal/optical analysis. These methods mentioned above still can't achieve the observation of long-term real-time BrC mass concentrations.

Since the establishment of the thermal–optical transmittance (TOT) method by the Sunset Laboratory, the Sunset OC/EC instrument, as part of the Chemical Speciation Network (CSN), where cover over 100 monitors across the United States over 15 years, offering long-term measurement of OC and EC concentrations, has been widely used in the United States and throughout the world providing important in-situ data of OC and EC aerosols (U.S.EPA, 2019; Birch and Cary, 1996). This instrument had been designed with a tuned diode laser (red 660 nm) to correct the formation of pyrolyzed carbon. In this study, we modified the Sunset instrument to a two-wavelength (658 nm and 405 nm) Sunset carbon analyzer by adding one more violet diode

laser at λ=405 nm. The violet diode laser together with the red diode laser, focus through the sample chamber then the laser beam passed through the filter to correct for the pyrolysis-induced error. Previous work reported by Chen et al. (2015) as mentioned above was integrating the optical instrument like the aethalometer to the traditional OC/EC analyzer, in this way, they provided the light absorption contributions of BC and BrC. The enhanced carbon analyzer provided new insight into more accurate OC and EC measurements. Their work was conducted in offline mode, based on their work, our instrument can get the real-time OC and EC mass concentrations both at 658 nm and 405 nm. BrC particles absorb light at 405 nm more effectively than 658 nm in the two-wavelength Sunset carbon measurements. We define $dEC=EC_{405\ nm}-EC_{658\ nm}$ and hope it can be an indicator of BrC aerosols so that we can divide real-time BrC mass concentration measurement from the two-wavelength measurement.

Nanjing, as one of the largest cities in the Yangzi River Delta region, represents a heavy industry area with a dense population. In addition, due to its topography, Nanjing is very sensitive to regional transport of air masses from its surrounding areas. OC, EC and dEC aerosols were observed from June 2015 to July 2016 at Nanjing University of Information Science and Technology (NUIST). Based on the abundant data, together with MODIS fire information, we can analyze the temporal variation, transport processes and sources of carbonaceous aerosols in North Nanjing and evaluate the biomass burning impact on dEC aerosols, which can be the scientific basis of pollution control policy.

2. Methods

2.1 Study site

In this study, the sampling site is located at Nanjing University of Information Science and Technology (NUIST) in the North Suburb of Nanjing (32°207′N, 118°717′E). The study site is surrounded by housing and industrial areas. Many chemical enterprises, for example, Yangzi Petrochemical, Nanjing Chemical Industry and Nanjing Iron and Steel Group are located at the northeast of the study region, which produces exhaust with large amounts of aerosol particles. The study site is adjacent to a heavily trafficked road (Ningliu Road) located near the site, approximately 600 m to the east. Therefore, this region has intense human activities, industrial emissions and heavy traffic flow.

2.2 Two-wavelength TOT measurement

Hourly concentrations of OC and EC in $PM_{2.5}$ were sampled and measured by a semi-

155 continuous carbon analyzer (Model-4, Sunset Lab, USA). Air samples were collected continuously
156 with a sample flow of ~8 L/min through a $PM_{2.5}$ cyclone. The collection time was set at 45 min
157 for each cycle. The airstream passed through a parallel plate organic denuder to reduce the effect
158 of volatile organic compounds and finally deposited on a quartz filter with a diameter of ~17mm.
159 After a sample was collected, OC and EC were analyzed using the thermal-optical
160 transmittance (TOT) method and applied a slightly modified NIOSH 5040 protocol. The details of
161 the heating setup were shown in Table S1. Figure 1 shows the structure and operational principle
162 of the instrument. Briefly, it consists of two-stages: the oven was first purged with helium and the
163 oven temperature increased in a stepped ramp to 840℃, OC was volatilized in this stage. Then the
164 oven temperature kept at 840℃ for a while and went down to 550℃. In the second stage, EC was
165 volatilized in a second temperature ramp to 850℃ while purging the oven with a mixture
166 containing 2%oxygen and 98%helium. The pyrolysis products were converted to carbon dioxide
167 (CO_2) which was quantified using a self-contained nondispersive infrared (NDIR) system.
168 Also, in this study, we used two-diode lasers (658nm and 405nm) equipped Sunset analyzer,
169 thus mass concentrations of OC and EC at different wavelengths can be measured with the 2-lasers
170 system. The split point between OC and EC was detected automatically by the RTCalc731 software
171 provided by Sunset Lab. The principle was same as the traditional Sunset carbon analyzer (Birch
172 and Cary, 1996). An example thermogram of sample analysis using the two-wavelength Sunset
173 semi-continuous carbon analyzer was shown in Fig. 2. During the sample analysis, the laser beam
174 at 658 nm and 405 nm were both sent through the filter and the transmitted light signal were
175 monitored to correct the undesired formation of pyrolyzed carbon (PyrC) and then to determine
176 the split point of OC and EC at both two wavelengths. BrC aerosols absorb light at 405nm more
177 significantly than 658nm in the 2-lasers system. Due to the strong absorption of BrC in near-
178 ultraviolet wavelength, thus this enhanced absorption at 405nm can serve as an indicator of BrC
179 aerosols (Liu et al., 2015). We define dEC data as the difference of EC concentrations at two
180 wavelengths ($dEC=EC_{405nm}-EC_{658nm}$) to identify the presence of BrC aerosols. Our study provided
181 a one-year measurement of dEC mass concentrations. Besides, OC and EC represent the OC and
182 EC concentrations at 658nm in this paper without a special explanation.
183 At the end of each analysis, a fixed volume of an internal standard containing 5%methane
184 and 95%Helium was injected and thus a known carbon mass could be derived. The external sucrose
185 standard (4.207 μg μL^{-1}) calibration was conducted every week to insure repeatable quantification.

Calibration with an instrument blank was conducted every day. Both detection limit for OC and EC of the instrument was 0.5 μg m^{-3}. We also did the measurements of OC and EC in $PM_{2.5}$ filter samples using the same method followed by the NIOSH protocol. All the data were corrected to blank measurement before comparison. Figure S1 shows the correlations between the real-time OC, EC concentrations and sampling OC, EC concentrations at the same time. The results showed that the online and offline data during the corresponding periods had good correlations with R^2 of 0.8 for OC, $R^2$of 0.4 for EC and R^2 of 0.8 for TC. In order to evaluate the impact of PyrC, we calculated the PyrC at 658 nm fraction of dEC and the average PyrC/dEC was 4.4%, indicating the little influence of PyrC.

2.3 Test of the new dEC data

To evaluate the new dEC data, parallel BC concentrations were measured with a seven-wavelength Aethalometer with dEC concentrations in December, 2019. Radiation attenuation of an aerosol deposition on a filter (ATN_λ) is determined by the Beer-Lambert law:

$$ATN_\lambda = ln\frac{I_{0,\lambda}}{I_\lambda} \quad \text{Equation. (1)}$$

Where $I_{0,\lambda}$ and I_λ were the measured wavelength-specific laser reflectance signals. ATN_λ is used to calculate the attenuation coefficient with Eq. (2):

$$b_{ATN} = \frac{A}{V} \quad \text{Equation. (2)}$$

Where A was the filter area and V is the sampled air volume. Then a simplified two-component model was used to calculate the contribution of light attenuation to both BC and BrC (Chow et al., 2018;Chen et al., 2015;Sandradewi et al., 2008;Hareley et al., 2008):

$$b_{ATN}(\lambda) = q_{BC} \times \lambda^{-AAE_{BC}} + q_{BrC} \times \lambda^{-AAE_{BrC}} \quad \text{Equation. (3)}$$

Where q_{BC} and q_{BrC} were fitting coefficients, AAE was the absorption Ångström exponent which represented the wavelength-dependent characteristics of light absorption capability of aerosols. The AAE of BC was assumed to be 1. Fitting coefficients in Eq. (3) were obtained for potential AAE_{BrC} between 1 and 8 by least square linear regression and the AAE_{BrC} led to the overall best fit in terms of r^2 is selected as the effective AAE_{BrC}. Using these fitting coefficients, the b_{ATN} due to BC and BrC are calculated at each wavelength. Figure S2 showed that the fitted b_{ATN} at 405 nm were within ±5 % of the measured values for $b_{ATN} > 0.01$. Figure 3 showed the relationship between the b_{ATN} due to BrC at 405 nm and the dEC. Good correlation between them were found with R square of 0.64, indicating that dEC was associated with BrC.

2.4 Sampling

2.4.1 Real-time $PM_{2.5}$ observation

The real-time $PM_{2.5}$ concentrations were measured through the Tapered Element Oscillating Microbalance (TEOM) method (TEOM1405-DF, Thermo Scientific, America) from August, 2015 to July, 2016. The resolution of the measured data was 6 min. The instrumental operation maintenance, data assurance and quality control were performed according to the Chinese Ministry of Environmental Protection Standards for PM_{10} and $PM_{2.5}$ which was named "HJ 653-2013" (Zhang and Cao, 2015b).

2.4.2 Sample collections

$PM_{2.5}$ in the atmosphere were collected on prebaked quartz fiber filters which were under 450℃ for 6 hours (QFF, PALL, America) with 8*10 inch by a high volume air sampler (KC-1000, Qingdao, China) at a flow rate of 999 L min^{-1} in four months: 4 June to 18 June, 6 October to 2 November and 10 December to 31 December in 2015, 10 May to 31 May in 2016. Sampling started and ended at around 8:00 and 20:00 every day; each sample was collected for 12 hours. A total of 148 samples were collected including four field bank filters in four seasons collected following 10 mins exposures to ambient air without active sampling.

All QFFs were pre-baked at 450 °C for 6 h before sampling to remove residual carbon. Before and after sampling, all QFFs were weighed by electronic balance (Sartorius, 0.1 mg, Germany). After weighting, the filters were wrapped in aluminum foils, packed in air-tight polyethylene bags and stored at -20˚C for further analysis. All procedures during handling of filters were strictly quality controlled to avoid any possible contamination.

2.5 Identification of potential regional sources

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSLPIT4.8) model, provided by the National Oceanic and Atmospheric Administration (NOAA) were used to investigate the air mass origins of carbonaceous aerosols. The 48-hour back trajectories at Nanjing (32.2°N, 118.7°E) were calculated every hour (Draxler and Hess, 1998;Rolph et al., 2017;Cohen et al., 2015). In order to evaluate the behavior of the air masses circulation in the planetary boundary layer (PBL), the trajectories at 500m corresponding to the upper-middle height of the PBL were calculated, representing well-mixed convective boundary layer for regional transport investigation (Xu and Akhtar, 2010). The National Center for Environmental Prediction Global Data Assimilation System (NCEP GDAS) data obtained from NOAA with a spatial resolution of 1° ×1°

and 24 levels of the vertical resolution were used as meteorological data input to the model. The Potential Source Contribution Function (PSCF) model was usually applied to localize the potential sources of pollutants. The details about the setup of the model can be seen in the research reported by Bao et al. (2017).

3. Results and discussion

3.1 Characteristics of carbonaceous aerosols

3.1.1 Concentrations of carbonaceous aerosols

The statistics for the $PM_{2.5}$, OC, EC and dEC mass concentrations at the NUIST site are summarized in Table 1. The hourly OC concentrations ranged from 0.5 to 45.8 μg m^{-3} (average of 8.9 ± 5.5 μg m^{-3}), and the EC concentrations ranged from 0.0 to 17.6 μg m^{-3} (average of 3.1 ± 2.0 μg m^{-3}). The results were comparable to those reported by Chen et al. (2017) in the Xianlin Campus of Nanjing University (5.7 μg m^{-3} for OC and 3.2 μg m^{-3} for EC), which site was located in the southeast suburb of Nanjing and close to the G25 highway and were also affected by traffic sources. The higher OC concentrations in this study were probably due to the around chemical enterprise emissions. The average contributions of OC and EC to the total measured $PM_{2.5}$ mass was 12.8% and 4.3%, respectively, suggesting that carbonaceous fraction made an important contribution to fine particulate matter. The average dEC mass concentration was 0.8 μg m^{-3} contributing 10.0% to OC, 22.3% to EC and 1.3% to $PM_{2.5}$ concentrations with max concentration of 8.1 μg m^{-3} contributing 48.2% to OC, 97.8% to EC and 17.6% to total $PM_{2.5}$ concentrations. This information can be further applied in the PMF analysis to evaluate the sources of carbonaceous aerosols (Zhu et al., 2014;Sahu et al., 2011;Yan et al., 2019).

Compared with carbonaceous aerosols levels in other cities (Table S2), the OC and EC concentrations in Nanjing were generally lower than those observed in urban sites such as Beijing and Shanghai and inland cities like Chengdu and Chongqing which was affected by the basin terrain characteristics with static wind and unfavorable diffusion conditions, but higher than those observed in the southern coastal cities such as Guangzhou, which was a megacity in China. It could be explained since the site set in Guangzhou was a rural site. In general, the level of carbonaceous aerosols concentrations in China was higher than that in developed countries in the United States and Europe and lower than that in developing countries like India, though the sampling period in India was from late autumn to winter, the much higher concentrations in India indicated the heavy pollution level. The average OC/EC ratios in this study was 3.6, which was lower than most of

those reported in other studies, indicating the important impact of vehicle emissions in our study site.

Figure 4 shows the mass fractions of hourly carbonaceous aerosols and OC/EC ratios at different $PM_{2.5}$ concentration intervals during the study periods. During the study period, 84.2% of the $PM_{2.5}$ samples exceeded the daily averaged Chinese national ambient air quality standard (NAAQS) of 35.0 μg m^{-3} for the first grade and 40.1% of the total samples exceeded the NAAQS of 75.0 μg m^{-3} for the second grade, reflecting heavy aerosol pollution in the study area. Generally, the fractions of carbonaceous components decreased with increasing $PM_{2.5}$ pollution level. Larger mass fraction (about 32.3%) of carbonaceous aerosols in $PM_{2.5}$ was found for period relatively lower $PM_{2.5}$ levels (0–20 μg m^{-3}) compared to high $PM_{2.5}$ levels (300–500 μg m^{-3}) with carbonaceous aerosols mass fraction of 5.2%. The result indicated other components like secondary inorganic aerosol (SIA) contributes more significantly to heavy haze events in Nanjing, which was also found in other cities in the Yangtze River Delta area (Yang et al., 2011; Zhang and Zhang, 2019). The contribution of dEC to OC decreased with the increase of $PM_{2.5}$ concentrations between 0-200 μg m^{-3}, and then increased with the increase of $PM_{2.5}$ concentrations between 200-500 μg m^{-3}. The dEC contributed most significantly to OC of 14.3% when $PM_{2.5}$ concentrations below 20 μg m^{-3}. Similar trend was found in OC/EC ratios which showed a sharp increase along with enhanced $PM_{2.5}$ level above 150 μg m^{-3}. Previous studies had reported that high OC/EC ratios were related to SOC formation or biomass burning emissions whereas low OC/EC ratios were related to vehicle exhaust (Wang et al., 2015). We divided the dEC/OC at different intervals of OC/EC ratios and found that the dEC/OC increased when the OC/EC ratios increased in four seasons, indicating strong secondary sources or biomass burning contributions to dEC during heavy pollution periods (Fig. S3).

3.1.2 Seasonal variations of carbonaceous aerosols

As shown in Fig. 5, the OC, EC, dEC concentrations and dEC/OC ratios showed similar variations with highest in winter and lowest in summer. The average OC and EC concentration in winter was ~1.4 times and 1.5 times higher than that in summer and the average dEC concentrations and dEC/OC in winter were approximately 1.4 and 1.6 times higher than those in summer (Table 1). High dEC/OC was found in January and February in winter, indicating strong influence of anthropogenic sources on dEC, such as coal combustion. In addition, we found strong biomass burning activities in February, which significantly contributed to the high concentrations

of dEC in February, more details could be found in section 3.3. The seasonality of carbonaceous species in $PM_{2.5}$ was strongly influenced by seasonal variations in emissions intensities and meteorological parameters. Table S3 summarizes the meteorological parameters in four seasons during the study period. The high carbonaceous aerosols concentrations in winter were mainly a result of relatively stable atmospheric conditions with low temperature, relative humidity and boundary layer on one hand, and on the other hand, increasing emissions from fossil-fuel combustion for heating from the chemical enterprises nearby. In summer, higher boundary layer resulted in the dispersion of aerosols in the atmosphere, and higher temperature promoted the partitioning of semi-volatile organic compounds (SVOCs) into gaseous phase (Yang et al., 2011). In addition, large precipitation in summer (586 mm in total) favored the wet scavenging processes of aerosols.

The OC/EC ratios in spring, summer, autumn and winter were 3.9, 4.0, 2.8 and 3.4, respectively (Table 1). The OC/EC ratio could give some information about primary and secondary organic carbon (Turpin and Huntzicker, 1995; Lim and Turpin, 2002). In summer, strong convective activities in the atmospheric boundary layer and solar radiation, high temperature and plenty of moisture in the atmosphere were favorable for the formation of SOC. On the other hand, the high OC/EC ratios in June in this study were also strongly related to biomass burning which will be discussed in the 3.3 sections. The lower ratios of OC to EC in autumn and winter indicated that strong primary sources in these two seasons. It should be noted that the OC/EC ratios were a rough indicator to estimate the primary and secondary organic carbon, further analysis of the formation of SOC need to be conducted in the future (Pio et al., 2011;Wu and Yu, 2016).

3.1.3 Diurnal variation of carbonaceous aerosols

The diurnal pattern of carbonaceous aerosols can be affected by both meteorological parameters and sources (Ji et al., 2016). Figure 6 depicts the diurnal variation of OC, EC, dEC, dEC/OC and OC/EC ratios during the study period. Clear diurnal variations were observed in OC and EC aerosols. Both the OC and EC concentrations kept high levels at night and low levels in the daytime, indicating the strong influence of the atmospheric boundary layer on air quality in the northern Nanjing. The peak occurred in the morning both in OC and EC indicating the significant impact of traffic source on the OC and EC concentrations. The dEC/OC and OC/EC ratios showed similar trends in the daytime with gradually increase from morning till afternoon, indicating the importance of the contribution of secondary sources to dEC. Similar though not so obvious diurnal

variations were found in dEC. It should be noted that the vehicle emissions and the boundary layer height had no significant effect on the diurnal variation of dEC/OC, suggesting there was no significant local sources of dEC. There was a small peak in dEC/OC at 3:00 am, which might be related to the aqueous secondary organic aerosols formations during nighttime (Sullivan et al., 2016).

The relative humidity (RH) and Temperature (T) dependent distributions of OC, EC mass concentrations and dEC/OC and OC/EC throughout the study period are shown in Fig. 7. High dEC/OC (>30 %) could be found in three areas, first showed in the right area with relatively high T at 25-40 ℃ and RH at 40-60 %, which were usually found in the summer afternoon which was closely related to the strong formation of SOC. This distribution was also shown in OC/EC. The second area was displayed in the upper region with RH over 80 % and T at 10-20 ℃ and the third area appeared when RH below 30 % and T at about 10 ℃, corresponding to nighttime and winter afternoon. In general, dEC had no strong dependence on the RH and T distribution, indicating the complex formation mechanism of dEC. The OC and EC showed similar distributions with the highest mass loading (OC: > 20 μg m^{-3}; EC: > 8 μg m^{-3}) at relatively high RH at 60-80 % which usually occurred at night with relatively low boundary layer height, leading to the accumulation of aerosols. However the corresponding OC/EC ratios were low, suggesting the importance of primary sources to OC and EC in northern Nanjing, which will be verified in the wind rose of OC and EC (Fig. 8).

3.2 Air mass transport

3.2.1 Windrose of carbonaceous aerosols

To investigate the influences of air masses transport to the study site, the wind rose of OC, EC and dEC/OC using hourly data in four seasons is illustrated in Fig. 8 (Carslaw and Ropkins, 2012). Two points should be noted. First, high OC and EC mass concentrations were found near the field site (indicating by WS < 1 m s^{-1}), suggesting that local and primary emissions (e.g., industrial and vehicle emissions) were stable and important sources contributing to atmospheric OC and EC mass concentrations in northern Nanjing. The OC mass concentrations from the southwest increased with the increase of WS in summer, indicating that sources of OC are complicated in summer including secondary reaction during long-range or regional transport. Second, compared with OC and EC, dEC aerosols showed no significant local sources. The dEC/OC increased with the increasing of WS and highest dEC/OC were found when WS over 3 m s^{-1}. Long-range or regional

371 transport was highly likely the main sources contributing to dEC mass concentrations.

372 3.2.2 The potential source areas of carbonaceous aerosols

373 The possible source contributions were evaluated using the PSCF model and the PSCF map are
374 shown in Fig. 9 (Petit et al., 2017). The areas with high PSCF values were highly likely the
375 potential pollution source areas. As shown in Fig. 9, PSCF results further proved the strong
376 regional transport contribution to dEC aerosols and local contributions to OC and EC aerosols. In
377 spring, the potential source areas of OC and EC were mainly from the southwest of Nanjing,
378 however, the potential source areas of dEC aerosols were from the east of Nanjing, indicating
379 obvious different sources between OC, EC and dEC. In summer, local areas were the main sources
380 areas of EC and the near-by Yangtze River Delta City Group from southeast of Nanjing including
381 developed cities like Shanghai were the main sources areas of OC and dEC. The anthropogenic
382 emissions from these areas might be important sources of OC and dEC. Besides, both the potential
383 sources areas of dEC and EC were displayed in the northwest of Nanjing in summer, suggesting
384 strong primary sources of dEC from this area which were very likely associated to biomass burning,
385 more details were in the section 3.3. In autumn, strongest local sources from the study site of OC
386 and EC were found. However the dEC mainly originated from regional transport from the
387 northwest and southeast areas of Nanjing. Biomass burning has been proved to be an important
388 source of air pollutants in the Yangtze River Delta (YRD) area, especially in the wheat harvest
389 seasons (e.g., June and October) (Cheng et al., 2014; Zhang and Cao, 2015a). In addition, the YRD
390 area is the most economically developed region in China and has lots of industrial cities, which
391 means that industrial emissions and anthropogenic sources contributed to high carbonaceous
392 aerosols pollution levels. In winter, dEC were mainly from long-range transport from northern
393 cities and regional transport from the southwest areas of Nanjing while both long-range transport
394 and local sources were found in OC and EC concentrations.

395 **3.3 The characteristics of carbonaceous aerosols during biomass burning periods**

396 The biomass burning emission has been proved to be an important source of BrC on a global
397 scale, it is consistently observed in large-scale forest fire events (Laskin et al., 2015). Based on the
398 Fire Information for Resource Management System (FIRMS) derived from the Moderate
399 Resolution Imaging Spectroradiometer (MODIS), we found that the fire points reached to 2028,
400 1773 and 967 on 11 Jun 2015, 7 February 2016 and 2 Mar 2016 in the areas around our study site,
401 respectively, suggesting there were strong biomass burning events on these days (Fig. S4). To

402 further investigate the biomass burning impact on dEC aerosols, we analyzed the temporal trends
403 of carbonaceous aerosols from 4 June 2015 to 19 June 2015 and 7 February 2016 to 3 Mar 2016,
404 respectively. Combining the observed aerosols concentrations and fire information, we divided the
405 periods into normal days and biomass burning days. It should be noted that the biomass burning
406 days are not determined based only on fire points. We also considered the 48-h backward
407 trajectories and open biomass burning areas. For example, we did found lots of fire points from 11
408 June 2015 to 12 June 2015 and from 7 February 2016 to 10 February 2016, respectively, and the
409 48-h back trajectories went through these biomass burning areas (Fig. S5b, c). However, although
410 there were large amounts of fire points in northwest of Nanjing from 8 June 2015 to 9 June 2015,
411 the backward trajectory showed air mass during the periods came from the southeast areas where
412 no open fire points were found (Fig. S5a). In contrast, there were only a few fire points found near
413 the study site from 26 February 2016 to 27 February 2016, the 48-h backward trajectory showed
414 the air mass was exactly from the area (Fig. S5d).
415 As shown in Fig. 10 and Fig. 11, we found that dEC concentrations, dEC/OC and OC/EC
416 ratios showed peaks during each biomass burning periods which was not that obvious in OC and
417 EC concentrations, suggesting the unique biomass burning impact on dEC and the sources of OC
418 and EC were more complicated. It should be noted that there were peaks of dEC appeared on 9
419 June 2015 and 13 February 2016, which were not biomass burning days, suggesting that biomass
420 burning was not the only sources of dEC. As mentioned in the 3.1 and 3.2 section, anthropogenic
421 emissions could be the sources of dEC and the secondary sources couldn't be ignored, too.
422 Summarized in Table 2 are the average and standard deviation values of OC, EC, OC/EC, dEC
423 and dEC/OC during biomass burning and normal days. The OC/EC, dEC concentrations and
424 dEC/OC were obvious higher in biomass burning days than those in normal days, but similar levels
425 of the OC and EC concentrations were found both in biomass burning days and normal days in
426 summer, suggesting the great contribution of biomass burning emissions to dEC aerosols and there
427 were other sources of OC and EC in summer. All the carbonaceous aerosols were higher in biomass
428 burning days in winter, in addition, the location of open fire counts were mainly in the northwest
429 and southwest area of the study site (Fig. S5c, d), which were the potential source areas of OC, EC
430 and dEC in winter as discussed in the section 3.2.2, indicating strong contributions of biomass
431 burning emissions to all the carbonaceous aerosols in winter.
432 **4. Conclusion**

In this study, the characteristics and sources of carbonaceous aerosols in North Nanjing were investigated and we introduced a two-wavelength method by modifying the Sunset carbon analyzer. We incorporated a new diode laser at λ=405 nm in the instrument, making it possible to detect the laser beam passing through the filter at both wavelength at λ=658 nm and λ=405 nm, so we can obtain the dEC concentrations. Our study illustrated the feasibility of using dEC to characterize the BrC aerosols, providing a new idea about the measurement of BrC. The results showed that high (low) OC, EC and dEC concentrations were found in Winter (summer), indicating the significant impact of the increase of various emission sources in winter and wet scavenging of rain in summer. Similar diurnal cycles for OC and EC concentrations were found with high at night and low in daytime, strongly affected by the boundary layers. Traffic emissions were found to have significant influence on the concentrations of OC and EC. Similar trends were found in the diurnal cycle of dEC/OC and OC/EC and the dEC/OC increased when the OC/EC ratios increased, indicating strong secondary sources or biomass burning impact on dEC. The wind rose and receptor model results showed that strong local emissions were found in OC and EC aerosols, however dEC aerosols were significantly affected by regional or long-range transport. The near-by YRD area was one of the main potential source areas of dEC, suggesting that anthropogenic emissions could be the sources of dEC. Together with the back trajectories analysis and MODIS fire informations, we analyzed two biomass burning events both in summer and winter. The results showed that the sources of OC and EC were more complicated than those of dEC aerosols in summer. Biomass burning emission made a great contribution to dEC concentrations in summer. Large number of open fire counts from the northwest and southwest areas of the study site were monitored, significantly contributed to all the carbonaceous aerosols pollutions in winter.

Our modified two-wavelength instrument provided more information than traditional single-wavelength thermo/optical carbon analyzer. The results proved that dEC can be an indicator of BrC in biomass burning days. It should be noted that the sources of dEC were complicated and the anthropogenic emissions and secondary formations of dEC aerosols couldn't be ignored , further chemical analysis need to be conducted in the future. The evaluation of SOC formation and the relationship between dEC and SOC can be conducted. In addition, More chemical analysis such as the analysis of the ion, the organic matter or the sugars in $PM_{2.5}$ can be measured, thus we can get some information of the tracers of different sources and more accurate and quantitative source apportionment can be done (Bhattaraia et al., 2019;Wu et al., 2018;Wu et al., 2019). We also hope

that the dEC data can be further applied in more researches.

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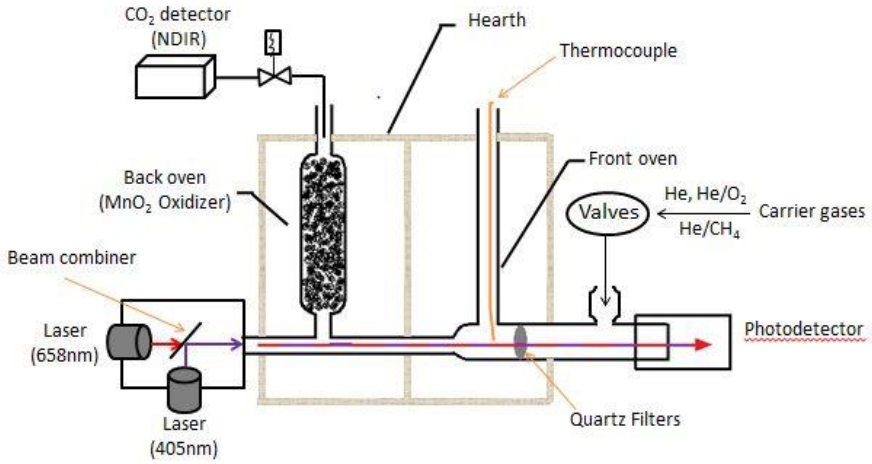

674

675 **Figure 1**. Principle and structure of the Sunset semi-continuous carbon analyzer.

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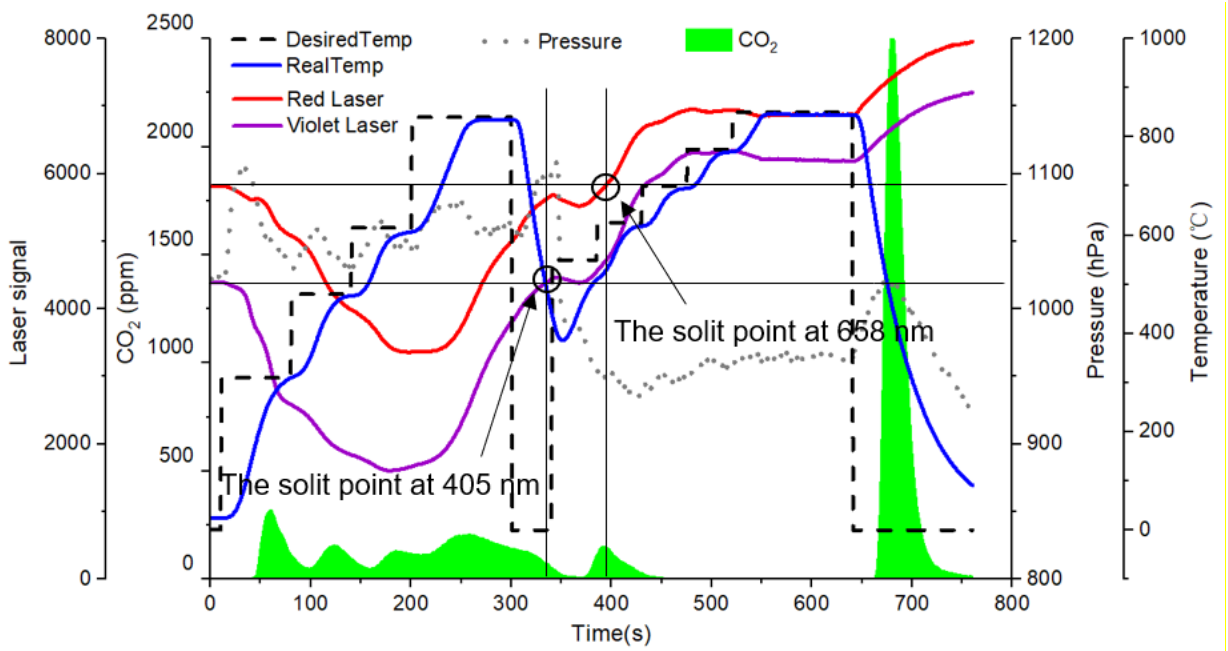
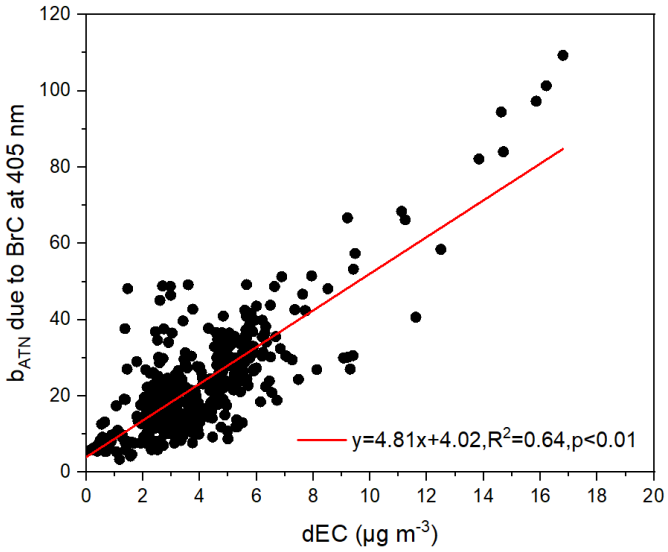


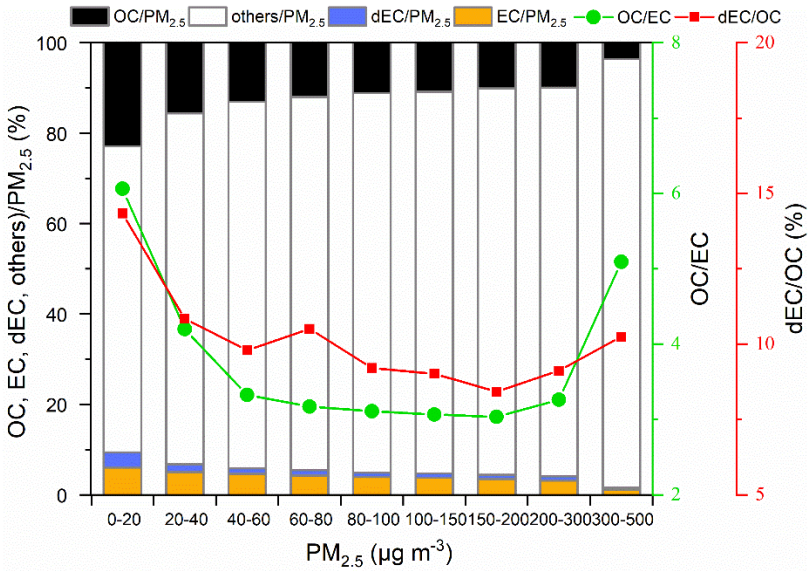
677

678 **Figure 2.** Example thermogram of sample analysis using the two-wavelength Sunset semi-
679 continuous carbon analyzer.

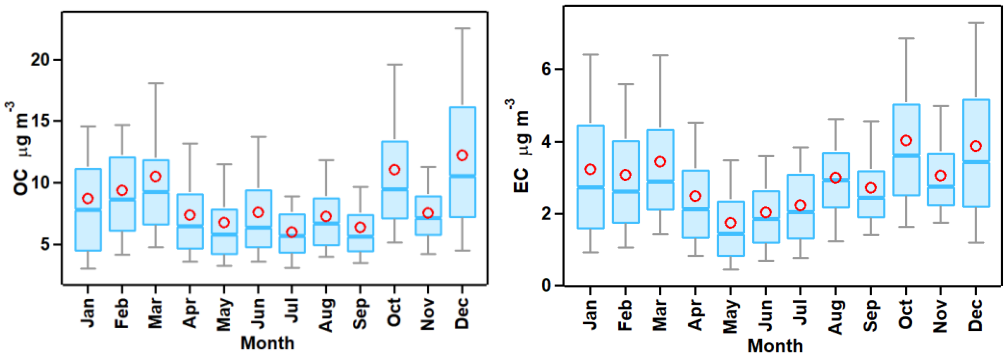


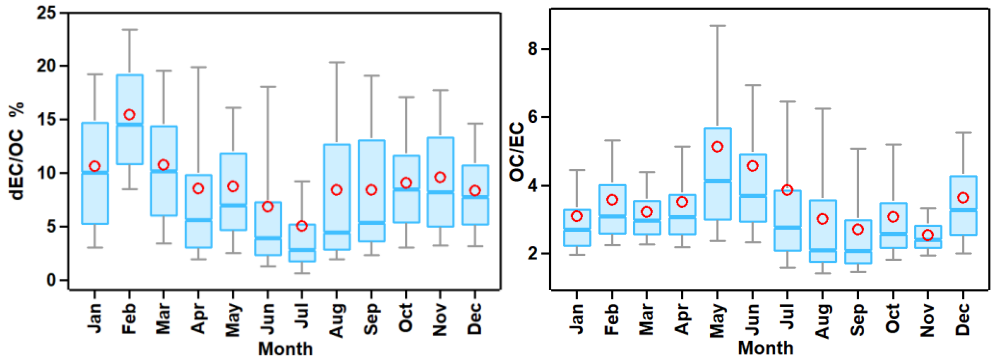
680

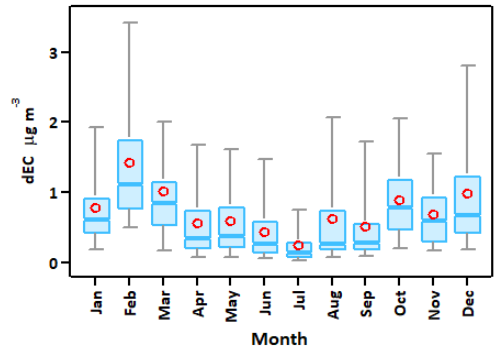
681 **Figure 3.** Relationship between the b_{ATN} due to BrC at 405 nm and the dEC concentrations.



682
683 **Figure 4**. Carbonaceous species fractions of $PM_{2.5}$ and OC/EC ratios at different $PM_{2.5}$
684 concentration intervals at NUIST from June 2015 to August 2016.

685 

686 

687 

688 **Figure 5.** Monthly variations of OC, EC, dEC, dEC/OC and OC/EC ratios at NUIST from June
689 2015 to August 2016. The boundary of the box indicates the 25% and 75% percentile,
690 respectively. The lower and upper whiskers indicate the 10% and 90% percentile, respectively.
691 The red circle within the box marks the average while the line within the box marks the median.

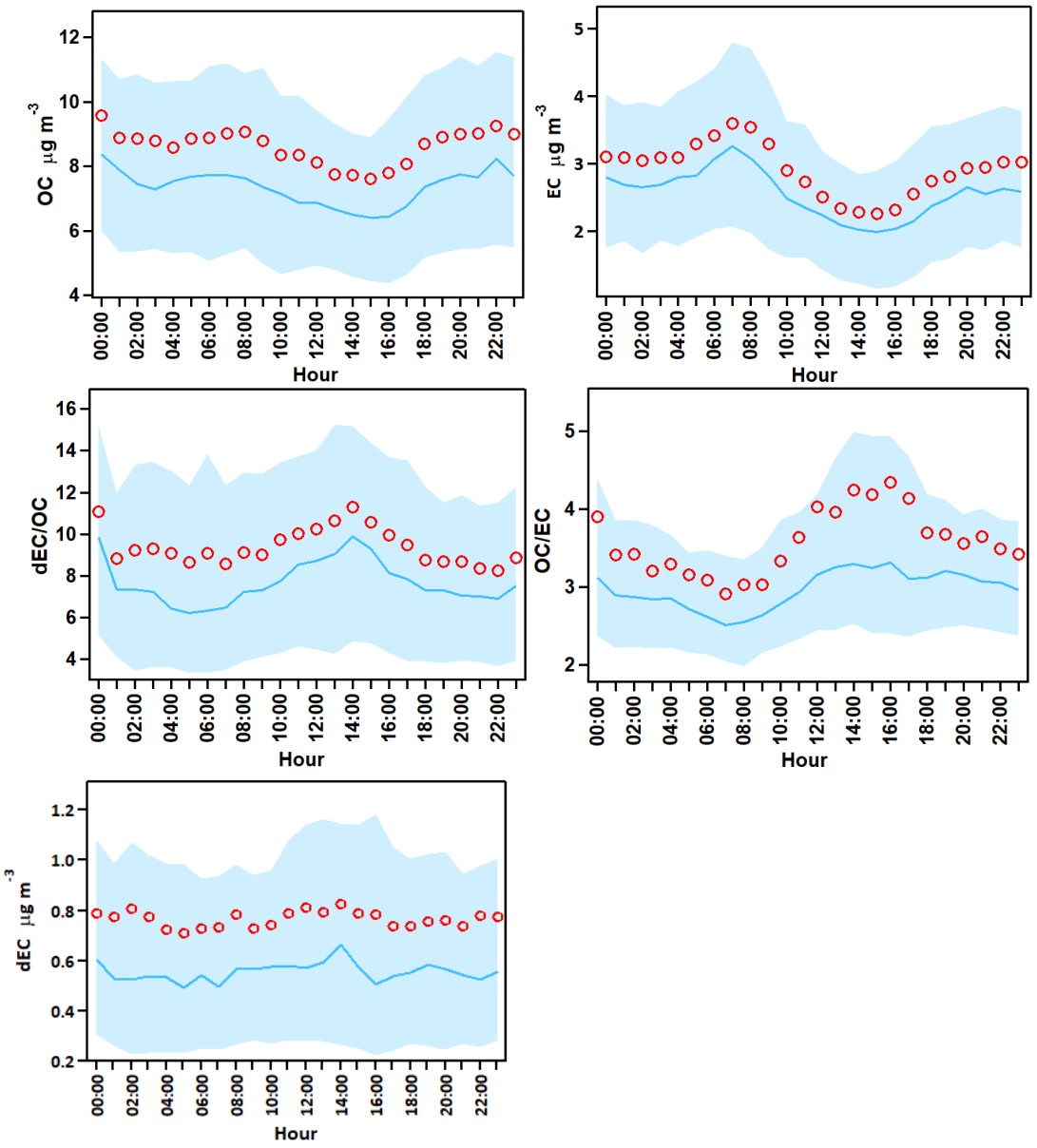


Figure 6. Diurnal variations of OC, EC, dEC concentrations, dEC/OC and OC/EC ratios during the study period. The boundary of the shaded area indicates the 25% and 75% percentile, respectively. The red circle marks the average while the blue line marks the median.

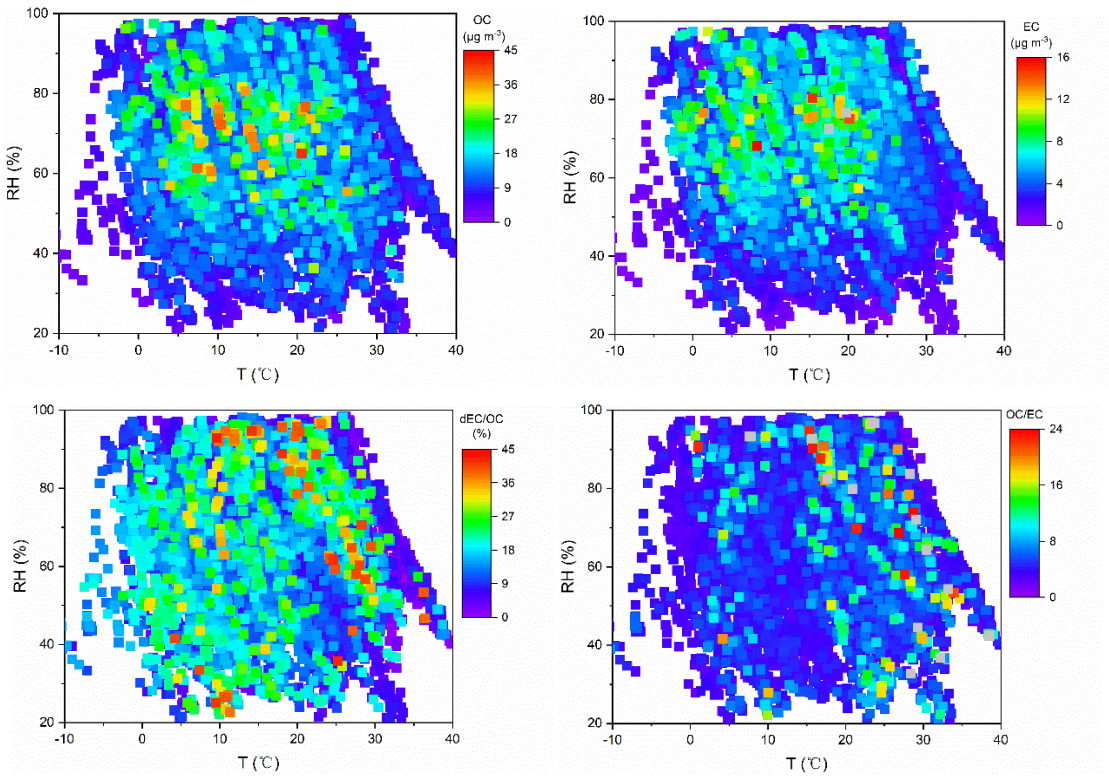


Figure 7. RH/T dependence of OC, EC, dEC/OC and OC/EC ratios during the study periods.

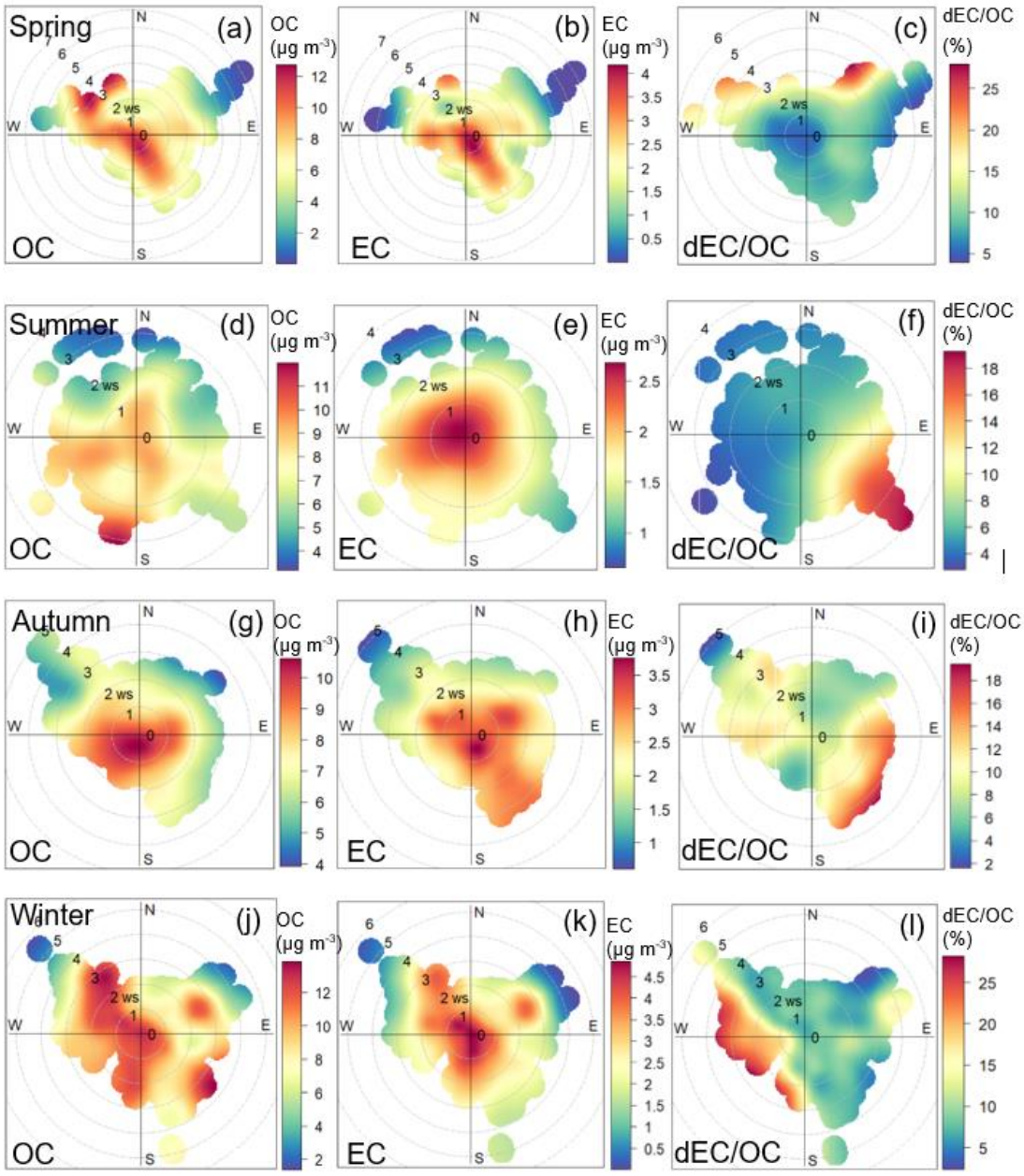


Figure 8. Wind rose of OC, EC and dEC/OC in spring ((a), (b), (c)), summer ((d), (e), (f)), autumn (g), (h), (i)) and winter ((j), (k), (l)).

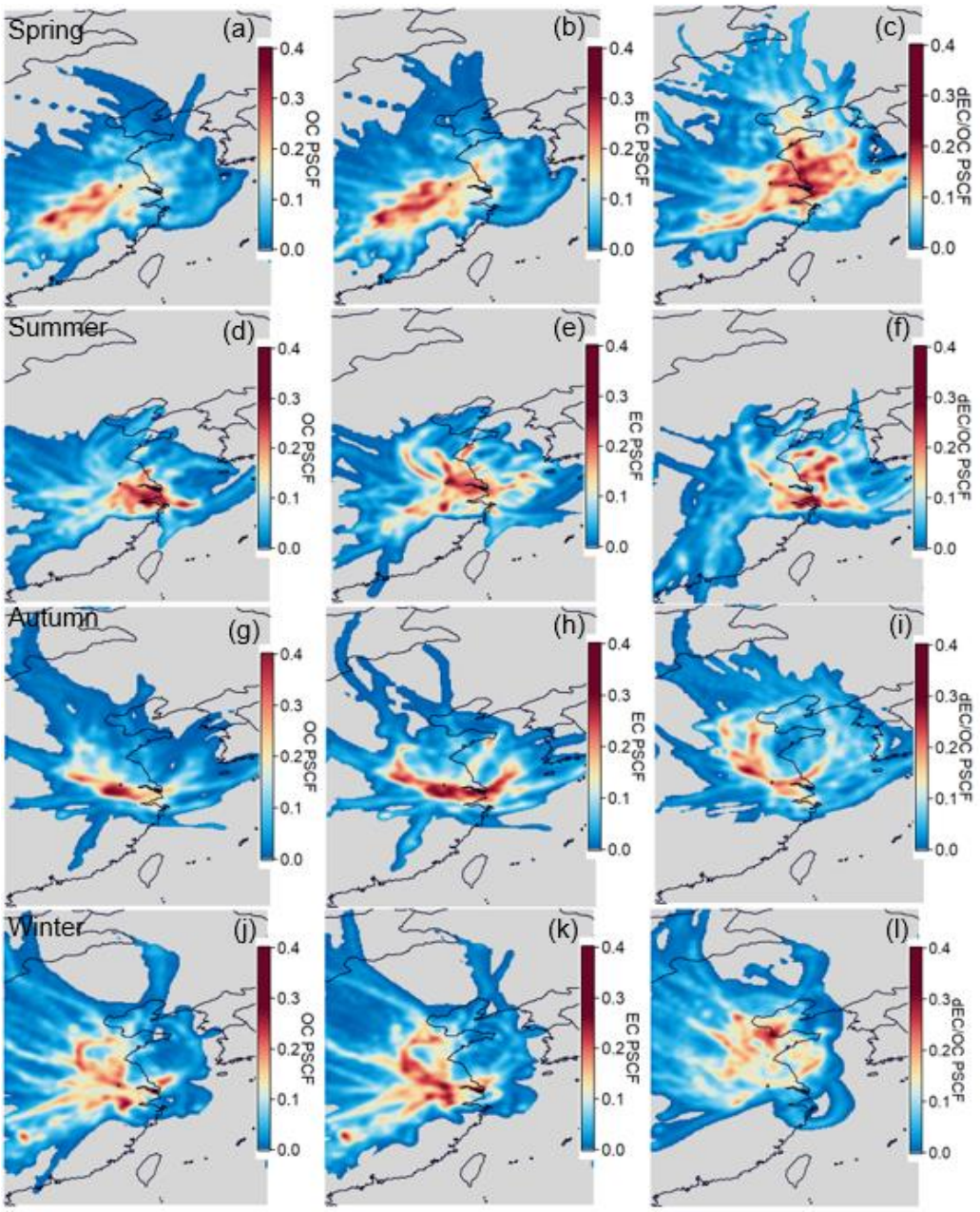


Figure 9. PSCF map for OC, EC and dEC/OC in spring ((a), (b), (c)), summer ((d), (e), (f)), autumn (g), (h), (i)) and winter ((j), (k), (l)).

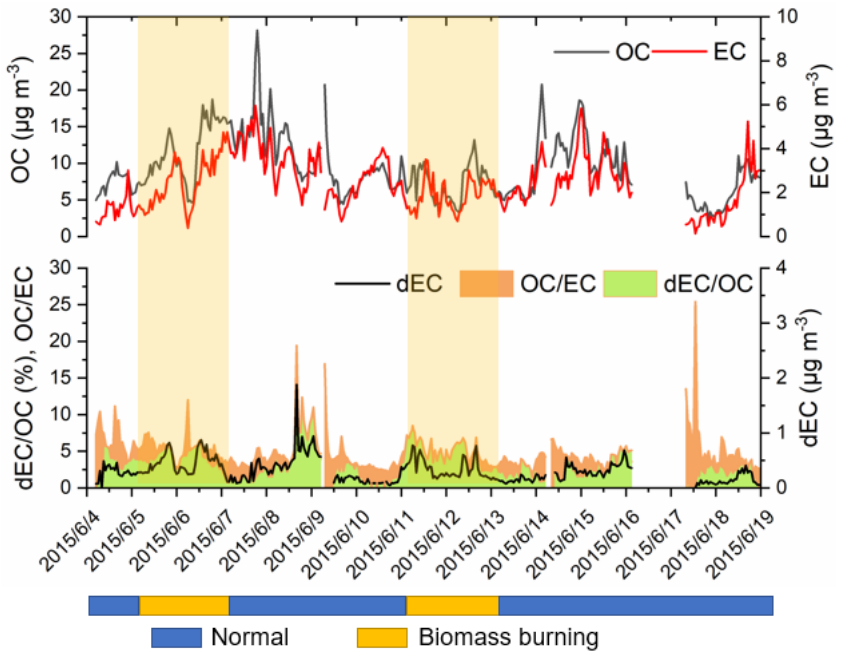


Figure 10. Time series of OC, EC, dEC/OC, dEC and OC/ EC from 4 June 2015 to 19 June 2015. The period was divided into normal days (blue bar) and biomass burning days (yellow bar). The yellow shadow represents the biomass burning periods.

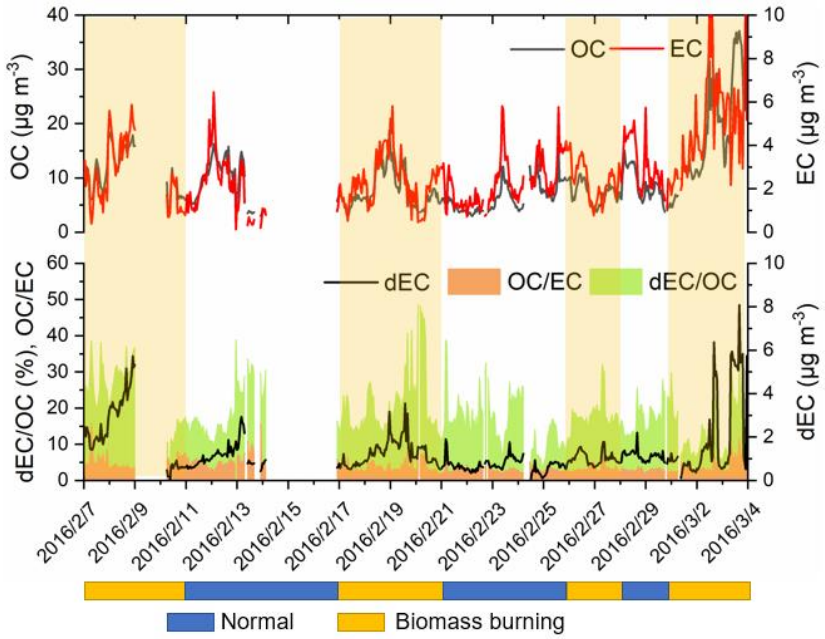


Figure 11. Time series of OC, EC, dEC/OC, dEC and OC/ EC from 7 February 2016 to 3 Mar 2016. The period was divided into normal days (blue bar) and biomass burning days (yellow bar). The yellow shadow represents the biomass burning periods.

715 **Table 1.** Statistical summary on the $PM_{2.5}$ and carbon species concentrations.

N=5113	Annual					Spring	Summer	Autumn	winter
	Average	Standard Deviation	Median	Min	Max	Average	Average	Average	Average
$PM_{2.5}$ (μg m^{-3})	77.2	48.6	65.0	2.5	458.1	72.1	47.9	70.5	91.8
OC (μg m^{-3})	8.9	5.5	7.5	0.5	45.8	8.4	7.2	8.4	10.2
EC (μg m^{-3})	3.1	2.0	2.6	0.0	17.6	2.6	2.3	3.3	3.4
OC/EC	3.5	2.4	2.9	1.0	29.3	3.9	4.0	2.8	3.4
dEC (μg m^{-3})	0.8	0.8	0.6	0.0	8.1	0.8	0.5	0.7	1.1
dEC/OC (%)	10.0	7.2	8.6	0.0	48.2	9.5	6.9	9.0	11.3
dEC/EC (%)	22.3	16.7	18.5	0.1	97.8	24.5	18.2	18.7	25.9
OC/$PM_{2.5}$ (%)	12.8	5.6	11.6	0.7	66.2	13.2	14.4	14.1	11.1
EC/$PM_{2.5}$ (%)	4.3	2.3	3.9	0.0	33.2	3.9	4.7	5.8	3.7
dEC/$PM_{2.5}$ (%)	1.3	1.2	0.9	0.0	17.6	1.4	1.3	1.2	1.3

716

717 **Table 2.** Statistics of OC, EC, OC/EC, dEC and dEC/OC during biomass burning days and normal
718 days. The values represent average±standard deviation.

		OC ($\mu g\ m^{-3}$)	EC ($\mu g\ m^{-3}$)	OC/EC	dEC ($\mu g\ m^{-3}$)	dEC/OC (**%**)
June 4th to 19th	Normal days	9.5±4.5	2.6±1.3	4.3±2.3	0.2±0.1	2.5±1.3
	Biomass burning days	9.0±3.6	2.0±0.9	4.8±1.6	0.4±0.2	4.6±1.4
February 7th to Mar 3rd	Normal days	7.5±3.3	2.5±1.2	3.3±1.3	0.8±0.3	12.7±5.6
	Biomass burning days	11.2±7.2	3.1±1.9	4.0±1.8	1.7±1.4	15.4±7.8

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Supplement

Content of this file

Reference for the supplement

748 **Table S1.** Temperature protocol of the modified NIOSH 5040 method used in this study.

Gas	Temperature(°C)	Time(s)
He-1	310	70
He-2	480	60
He-3	615	60
He-4	840	100
He/O_2-1	550	45
He/O_2-2	625	45
He/O_2-3	700	45
He/O_2-4	775	45
He/O_2-5	850	120
CH_4/He	0	120

749

Table S2. Comparisons of the concentrations of OC and EC in $PM_{2.5}$ between different cities in China and around the world using the TOT method applied in the NIOSH 5040 protocol.

Country	City or region	Site type	Sampling period	OC	EC	OC/EC	References
China	Beijing	Urban	Mar 2013-Feb 2014	14.0	4.1	3.4	(Ji et al., 2016)
China	Shanghai	Urban	Oct 2005-Jul 2006	14.7	2.8	5.0	(Feng et al., 2009)
China	Chengdu	Urban	May 2012-Apr 2013	19.0	4.6	4.3	(Chen et al., 2014)
China	Chongqing	Urban	May 2012-Apr 2013	15.2	4.0	3.8	(Chen et al., 2014)
China	Nanjing	Suburban	Annual 2014	5.7	3.2	1.8	(Chen et al., 2017)
China	Guangzhou	Rural	Mar 2012–Feb 2013	6.1	0.8		(Lai et al., 2016)
China	Mount Heng	Set at 1269 m asl	Mar-May 2009	3.0	0.5	5.2	(Zhou et al., 2012)
Mexico	Mexico City	Suburban	Mar 2006	6.4	2.1	4.5	(Yu et al., 2009)
India	Delhi	Suburban	Nov 2010-Feb 2011	54.1	10.4	5.2	(Tiwari et al., 2012)
America	Philadelphia	Suburban	Jul 2002-Aug 2002	4.8	0.4	18.7	(Jeong et al., 2004)
America	Rochester	Suburban	Jun 2002	9.2	0.3	23.6	(Jeong et al., 2004)
Spain	Aragón	Urban	Dec 2011	3.6	1.1	4.7	(Escudero et al., 2015)
China	Nanjing	Suburban	Jun 2015-Aug 2016	8.6	2.9	3.6	This study

753 **Table S3.** Statistics on the meteorological factors in four seasons at NUIST site during the study
754 period.

	Atmospheric Pressure (hPa)	Relative Humidity (%)	Temperature (°C)	Wind Speed ($m\ s^{-1}$)	Total Precipitation (mm)
Spring	1009.9	66.0	16.8	1.9	256.3
Summer	1000.7	72.6	26.7	1.4	586.0
Autumn	1014.6	71.0	19.5	1.7	218.5
Winter	1027.0	63.9	5.7	1.7	82.1

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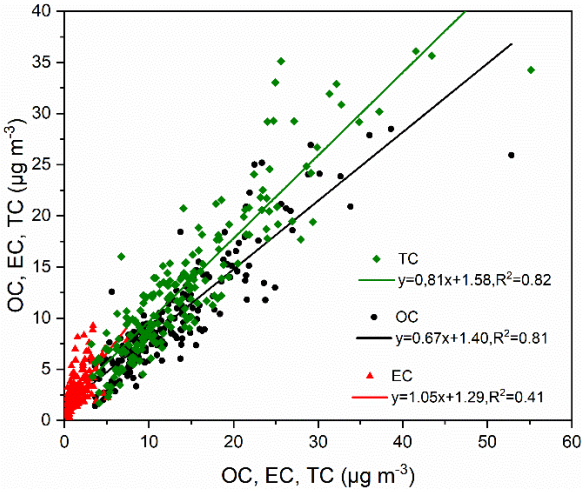


Figure S1. Correlations between the real-time OC, EC and TC concentrations (y-axis) and sampling OC, EC and TC concentrations (x-axis) during the corresponding periods.

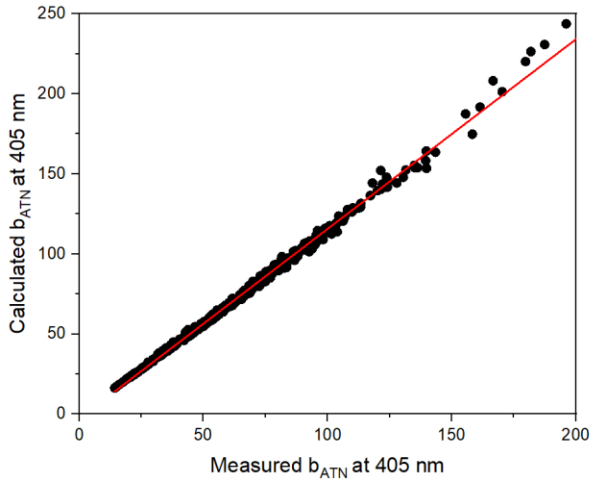
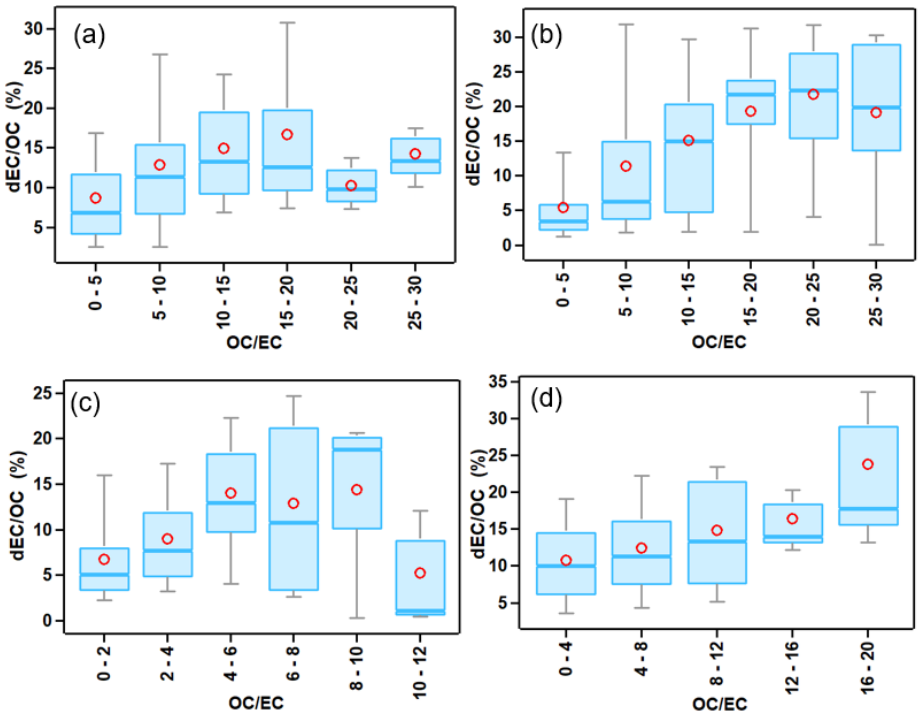


Figure S2. Meaured b_{ATN} at 405 nm compared with b_{ATN} fitted from Eq. (3) using a two-component model.



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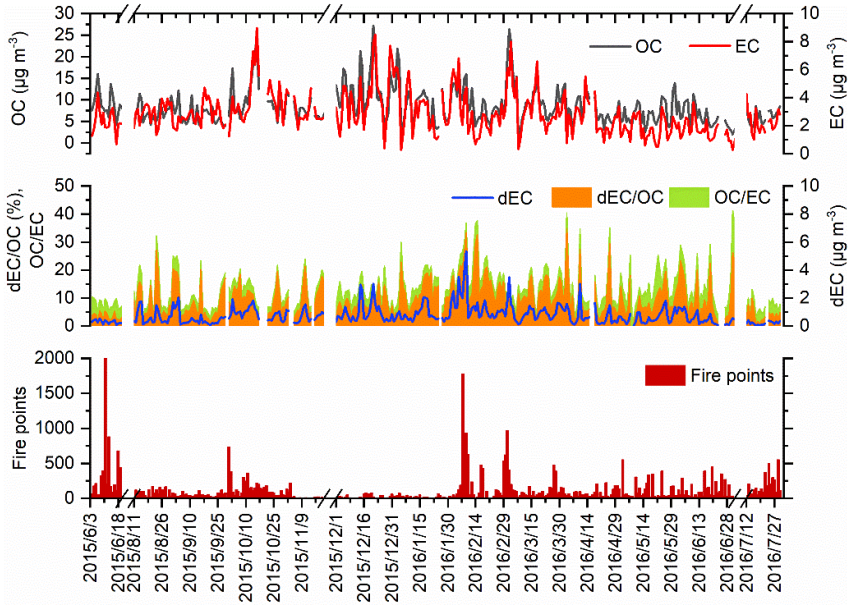
764 **Figure S3.** dEC/OC variation at different intervals of OC/EC ratios in spring (a), summer (b),
765 autumn (c) and winter (d).



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767 **Figure S4.** Time variations of OC, EC, dEC, dEC/OC, OC/EC and fire points obtained from the
768 Fire Information for Resource Management System (FIRMS) derived from the Moderate
769 Resolution Imaging Spectroradiometer (MODIS).

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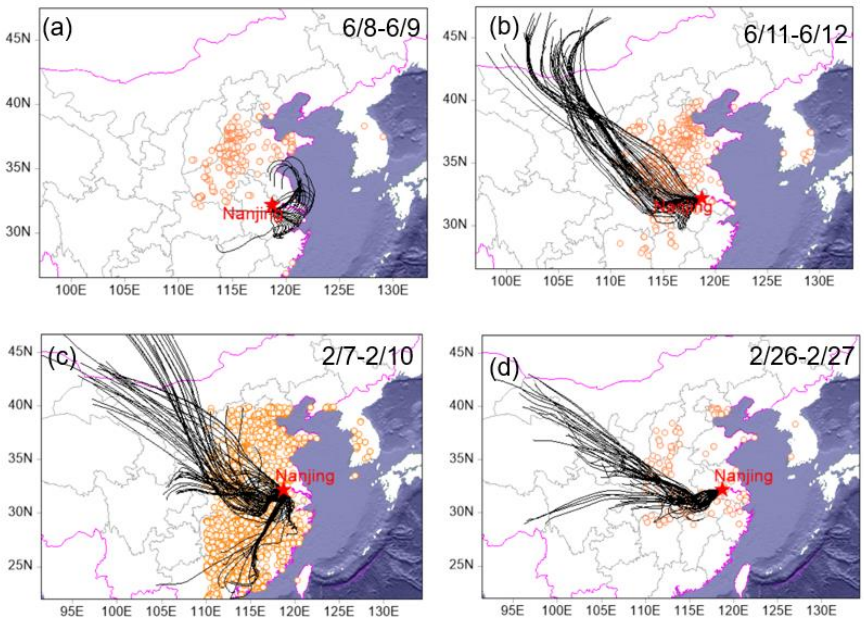

Figure S5. 48-h back trajectories at 500 m from the study site from 8 June 2015 to 9 June 2015(a), 11 June 2015 to 12 June 2015 (b), respectively and from 7 February 2016 to 10 February 2016 (c) and 26 February 2016 to 27 February 2016 (d), respectively.

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