Source apportionment of black carbon aerosols from light absorption observation and source-oriented modeling: An implication in a coastal city in China

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Abstract. Black carbon (BC) is the most important light absorbing aerosol in the atmosphere. However, sources of atmospheric BC aerosols are largely uncertain, making it difficult to assess its influence on radiative forcing and climate change. In this study, year-round light-absorption observations were conducted during 2014 using an aethalometer in Xiamen, a coastal city in southeast China. Source apportionment of BC was performed and temporal variations in BC sources were characterized based on both light absorption measurements and a source-oriented air quality model. The annual average concentrations of BC from fossil fuel (BCff) and biomass burning (BCbb) were $293 \pm 144 \text{ng m}^{-3}$ and $1340 \pm 542 \text{ng m}^{-3}$, contributing $66.7 \%$ and $33.3 \%$ to total BC, respectively. BCbb contribution exhibited clear diurnal cycle with the highest level ($37.9 \%$) in the evening rush hour and seasonal pattern with the maximum ($39.9 \%$) in winter. Conditional probability function (CPF) analysis revealed the large biomass burning contributions were accompanied by east-northeasterly and northerly winds. Backward trajectory indicated that air masses from north and east-central China were associated with larger biomass burning contributions. Potential source contribution function (PSCF) and concentration-weighted trajectory (CWT) suggested that north and east-central China and Southeast Asia were potential sources for both BCff and BCbb. The source-oriented modeling results showed that transportation, residential and open biomass burning accounting for $45.3 \%$, $30.1 \%$ and $17.6 \%$ were the major BC sources. Among the three fuel catalogs, liquid fossil fuel ($46.5 \%$) was the largest source, followed by biomass burning ($32.6 \%$) and coal combustion ($20.9 \%$). Source contributions of biomass burning and fossil fuel combustion identified by the source-oriented model and observation-based method were in good agreement. The source-oriented model also captured the majority of seasonal variations in source contributions. The findings provide solid supports for controlling fossil fuel sources to limit the impacts of BC on climate change and environmental degradation in the relatively clean region in China.
1 Introduction

Black carbon (BC) aerosol is a vital air pollutant throughout the surface earth system and it has attracted a great concern regarding its multiple impacts on human health, climate change and atmospheric visibility (Bond et al., 2013). As the most important light-absorbing component of PM$_{2.5}$ (particulate matters with aerodynamic diameter less than 2.5 µm), BC exerts a key and unique role in the climate system by absorbing solar radiation, affecting chemical/physical properties of cloud, and influencing snow and ice cover (Jacobson, 2002; Ramanathan and Carmichael, 2008; Bond et al., 2013; Qian et al., 2014; Kim et al., 2015). BC is even found as the second most important climate-warming agent after carbon dioxide, with a positive climate forcing of 1.1 W m$^{-2}$, greater than that of methane (Bond et al., 2013). BC also has impacts on urban weather conditions and may play a key role in extreme weather (Ding et al., 2013; Fan et al., 2015; Saide et al., 2015; Wang et al., 2018). Under polluted environments, BC has significant influence on pollution development (Ding et al., 2016; Peng et al., 2016; Lou et al., 2019). In addition, BC leads to visibility impairment because of its strong absorption of visible light (Watson, 2002) and has adverse impacts on human health due to its adsorption captivity (Janssen et al., 2011; Colicino et al., 2017). Nevertheless, due to lacking observational constraints and uncertainties in emission inventories, large uncertainties still exist in BC emissions including absolute fluxes and relative source contributions of fossil versus biomass combustion, which will complicate our knowledge on the multiple BC effects. It is also necessary to clarify the contributions of different sources to BC in order to determine efficient emission mitigation strategies. At global scale, BC emission sources can be attributed to fossil fuels (~ 40 %), open biomass burning (~ 40 %) and biofuels (~ 20 %) (Ramanathan and Carmichael, 2008). However, these fractions vary significantly because of the substantial spatial and temporal variations in BC emissions (Venkataraman et al., 2005; Rehman et al., 2011; Cheng et al., 2013; Andersson et al., 2015).

Several source apportionments for quantitatively differentiating between biomass and fossil sources of ambient BC aerosol have been conducted using observation-based methods, such as isotope (e.g., radiocarbon) analysis technique and light-absorbing property analysis (Sandradewi et al., 2008; Gustafsson et al., 2009; Liu et al., 2014; Vaishya et al., 2017; Helin et al., 2018; Kalogridis et al., 2018; Mousavi et al., 2018; Jing et al., 2019; Kant et al., 2020). For example, Andersson et al. (2015) presented dual carbon isotope constrained BC source apportionment in three key hotspot regions in China during a severe haze event, finding that biomass burning contributed ~ 30 % to BC whereas fossil fuel sources were dramatically different between north and south. Aethalometer model was adopted to analyze light absorption at multi-wavelengths to assess the fossil fuel and biomass combustions contributions to BC in Delhi, revealing that the contribution of biomass burning was 28 % in average (Dumka et al., 2018). Mousavi et al. (2019) apportioned BC in the Milan metropolitan area to the fossil fuel and biomass burning emission using the Aethalometer model with the absorption Ångstrom exponent (AAE) values derived from the $^{14}$C radiocarbon analysis, highlighting the significant impact of residential wood burning on BC. Such observation-based source apportionment methods are powerful to understand the BC
sources at given receptor locations. However, the methods are highly dependent on accurate observations with high temporal resolution, which is unavailable for most regions without the measurement instruments. For example, the isotope method, especially the $^{14}$C analysis, is costly and lack of high temporal resolution. Therefore, although many observation-based BC source apportionments have been carried out, the source-based method can still be a strong supplement. For example, in a recent report, Winiger et al. (2019) conducted observation-based source apportionment of circum-Arctic BC with carbon isotope analysis and found that comparison of a Lagrangian atmospheric transport model (FLEXPART-ECLIPSE-GFED) predictions with the observations agreed well with each other for BC concentrations, with larger discrepancies for (fossil/biomass burning) sources, indicating the misallocations of emissions in the emission inventories.

Source-oriented modeling, which estimates pollution levels and identifies sources using chemical transport models (CTMs) with the inputs of emission inventory and meteorology, is another useful tool to study potential factors deriving BC. Such source apportionment technique has been developed and used for direct source apportionment of PM in more than a decade (Kleeman et al., 2007; Ying et al., 2008; Zhang et al., 2014). For example, Hu et al. (2015) found that residential emission was the major contributor to BC in spring and winter while industrial emission was important in summer and fall in China. Guo et al. (2017) quantified the contributions of different sources in North India and found that industry was the largest source for BC. Although the source-oriented modeling is powerful, limitations exist such as the inability to take into account unknown sources and the imprecise information on emission inventories and meteorology. The method is highly dependent on accuracy of emission inventory, which is unfortunately an enormous challenge. A pollution source not in the emission inventory will not emerge as a contributor to the CTM results. Taking into account the advantages and disadvantages of the observation- and modeling-based methods, a combination of the two methods can be a complement to each other for providing reliable and reasonable information on pollution sources and contributions.

China is the largest source of BC aerosols in the world (Wang et al., 2012; Bond et al., 2013; Huang et al., 2016), remarkable influences of BC on air quality, weather condition and climate change were revealed in China (Menon et al., 2002; Ding et al., 2016; Huang et al., 2016; Yang et al., 2017). Spatiotemporal distributions and regional transport mechanisms of BC in China as well as their affecting factors have been widely investigated with field measurements or model simulations (Cao et al., 2010; Wu et al., 2013; Wang et al., 2015; Zhang et al., 2019; Zheng et al., 2019; Deng et al., 2020). In contrast, source apportionment studies on BC aerosols in China are still limited and mostly distributed in heavily polluted areas (Chen et al., 2013; Andersson et al., 2015; Li, K. et al., 2016; Li, N. et al., 2016; Yu et al., 2018; Jing et al., 2019). In this study, the observation-based method was combined with the source-oriented modeling to quantify the contributions of different sources to BC in a relatively clean region in China. The results of the two source apportionment methods were inter-compared. Temporal variability, potential sources and transport pathways of BC from fossil fuel and biomass burning were also characterized. The findings help better understand main sources and relative contributions of BC and provide valuable information to adopt effective emission reduction measures to control BC pollution in not heavily polluted regions.
2 Methodologies

2.1 Observation site and measurements

The field campaign was performed in Institute of Urban Environment, Chinese Academy of Sciences (118°03'E, 24°36'N) in the coastal city Xiamen in China (Deng et al., 2016, 2020). Xiamen is located in the Western Taiwan Strait region, which is adjacent to the Yangtze River Delta region (YRD) and the Pearl River Delta region (PRD) (Fig. 1). Xiamen has small local emission of BC (Fig. 1) and better diffusion condition compared to some developed cities in East China, which might lead to a lower BC concentration in Xiamen (Deng et al., 2020). However, Xiamen is often affected by emissions from polluted areas by long-range transport under the influence of East Asia monsoon (Deng et al., 2020). Therefore, conducting source apportionment of BC over Xiamen is very representative to improve our understanding on the sources of BC and their transport characteristics in relatively clean region.

The observation site lies approximately 15 km away from the downtown to the southeast. None of large industrial sources was within 10 km away and there were only a few construction and traffic sources. The measurement instruments were arranged on the rooftop of a house with a height of 8 m above ground level. Real-time measurements of BC mass concentration were conducted with a seven-wavelength (370, 470, 520, 590, 660, 880 and 950 nm) aethalometer (AE31, Magee Scientific) in January–December 2014. Aethalometer worked at a flow rate of 5 L min⁻¹ and estimated light attenuation under the principle of optical transmission (Hansen et al., 1984). BC concentration was then calculated according to the light attenuation. The concentration measured at 880 nm is considered as the standard value of atmospheric BC because BC is the predominant light-absorbing species at this wavelength with little impact from other compounds (Ganguly et al., 2005). The method reported in Virkkula et al. (2007) was applied to correct BC mass concentration due to shadowing effects and multiple scattering effects.

2.2 Observation-based source apportionment

Observation-based source apportionment of BC in Xiamen was performed with the Aethalometer method, which is based on the two-component assumption and has been widely adopted to assess the contribution from fossil fuel combustion and biomass burning (Sandra dewi et al., 2008; Favez et al., 2010; Liu et al., 2014; Rajesh and Ramachandran, 2017; Martinsson et al., 2017; Dumka et al., 2018; Helin et al., 2018; Mousavi et al., 2019; Mbengue et al., 2020). The Aethalometer method apportions the total BC to $BC_{ff}$ (BC emitted by fossil fuels) and $BC_{bb}$ (BC emitted by biomass burning) contributions. $BC_{ff}$ and $BC_{bb}$ are expressed as follows:

$$BC_{ff} = BC \times \frac{b_{abs,ff}(\lambda)}{b_{abs}(\lambda)}$$

(1)
BC_{bb} = BC \times \frac{b_{abs,bb}(\lambda)}{b_{abs}(\lambda)} \quad (2)

where $b_{abs}(\lambda)$ is light absorption at wavelength of $\lambda$, $b_{abs,ff}$ and $b_{abs,bb}$ are light absorption coefficients for fossil fuel and biomass burning, respectively. $b_{abs}$ is assumed to apportion to $b_{abs,ff}$ and $b_{abs,bb}$ contributions (Sandradewi et al., 2008). $b_{abs,ff}$ and $b_{abs,bb}$ satisfy the following equations:

$$\frac{b_{abs,ff}(\lambda_1)}{b_{abs,ff}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-AAE_{ff}} \quad (3)$$

$$\frac{b_{abs,bb}(\lambda_1)}{b_{abs,bb}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-AAE_{bb}} \quad (4)$$

where $AAE_{ff}$ and $AAE_{bb}$ are the absorption Ångström exponents (AAE) for fossil fuel and biomass burning, respectively. $AAE_{bb}$ adopted in present study is 1 since fossil fuel combustions mostly contain BC and typically no other light absorbing constituents (Kirchstetter et al., 2004; Bond et al., 2013). By contrast, in addition to BC, emissions from biomass burning contain a substantial fraction of light absorbing organic aerosols, which strongly enhances light absorption in the near-ultraviolet spectrum and has little contribution in the near-infrared spectrum. Therefore, $AAE_{bb}$ is larger than $AAE_{ff}$ due to the strong absorption in the UV regime (Sandradewi et al., 2008; Bond et al., 2013; Zotter et al., 2017). The empirical value of $AAE_{bb}$ adopted in this work is 2 according to Sandradewi et al. (2008). BC_{ff} and BC_{bb} can be obtained by combining all above equations and assumed values for $AAE_{ff}$ and $AAE_{bb}$.

In Eqs. (3)–(4), $\lambda_1$ and $\lambda_2$ should be wavelengths in the near-ultraviolet and near-infrared range, respectively. In this study, 470 nm and 950 nm were selected as near-ultraviolet and near-infrared wavelengths based on previous studies (Sandradewi et al., 2008; Favez et al., 2010; Zotter et al., 2017; Kalogridis et al., 2018).

2.3 Potential sources of BC_{ff} and BC_{bb}

The conditional probability function (CPF) was used to investigate the possible predominant directions of local sources of BC_{ff} and BC_{bb} relative to wind directions in different seasons (Ashbaugh et al., 1985). The CPF is calculated as:

$$CPF_{\Delta \theta} = m_{\Delta \theta} / n_{\Delta \theta} \quad (5)$$

where $n_{\Delta \theta}$ is the total occurrences from wind sector $\Delta \theta$ and $m_{\Delta \theta}$ is occurrences from the same wind sector with the BC_{ff} (BC_{bb}) concentration exceeding the threshold criterion. The CPF analysis was also performed for the ratio of BC_{bb} to BC (BC_{bb}/BC) to analyze the impact of local sources on the contribution from biomass burning. In this analysis, a threshold criterion of the top 25 % concentration (ratio) was chosen.

Backward trajectories were simulated with the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) from NOAA/ARL to characterize the regional sources and transport of air masses arriving in Xiamen (Stein et al., 2015).
Five-day backward trajectories ended at the height of 500 m were calculated every hour using the Global Data Assimilation System (GDAS) reanalysis meteorological dataset with a 1° × 1° latitude-longitude resolution. Hourly trajectory endpoints implying the geographical distribution and the height of the air parcel were derived from the model and trajectory clusters were then obtained from cluster analysis. The outflow regimes for air masses to the receptor site with the potential origins were traced with the trajectory clusters.

Potential regional source contributions of BC_{df} and BC_{bf} were further identified with the potential source contribution function (PSCF) method on basis of the backward trajectories. PSCF is a widely adopted tool to identify regional source distributions of air pollutants at a receptor site (Hopke et al., 1995; Bari et al., 2015; Zhang et al., 2017). The study domain is divided into \(i \times j\) grid cells and PSCF values can be calculated as follows:

\[
PSCF_{i,j} = \frac{m_{i,j}}{n_{i,j}}
\]  

(6)

where \(n_{i,j}\) is the number of endpoints that in the \(i/j\)th grid cell and \(m_{i,j}\) is the number of endpoints for the same grid cell that have BC_{df} (BC_{bf}) concentration higher than a criterion. These grid cells with high PSCF values are the maximum probability potential source areas contributing to high BC_{df} (BC_{bf}) mass concentrations at the receptor location. In this work, the top 25 % concentrations were set as the threshold. The study domain covered 10°–55° N and 80°–140° E, which comprises 10800 grid cells with the size of 0.5° × 0.5° latitude and longitude.

To minimize the uncertainty in grid cells with low \(n_{i,j}\), an empirical weight function \(w_{i,j}\) was multiplied with the PSCF values. \(w_{i,j}\) was defined as follows:

\[
w_{i,j} = \begin{cases} 
1.00 & n_{i,j} > 3n_{ave} \\
0.70 & 1.5n_{ave} < n_{i,j} \leq 3n_{ave} \\
0.42 & n_{ave} < n_{i,j} \leq 1.5n_{ave} \\
0.05 & n_{i,j} \leq n_{ave}
\end{cases}
\]  

(7)

It is difficult for PSCF method to identify the source intensity and separate strong sources and weak sources. Therefore, concentration-weighted trajectory (CWT) model was also performed to overcome this limitation. In this method, each grid cell is assigned a weighted concentration by averaging the sample concentrations that have associated trajectories crossing the grid cell (Hsu et al., 2003). The average weighted concentration \(C_{i,j}\) in the \(i/j\)th grid cell was calculated as follows:

\[
C_{i,j} = \frac{1}{\sum_{l=1}^{M} \tau_{i,j,l}} \sum_{l=1}^{M} C_{i} \tau_{i,j,l}
\]  

(8)

where \(M\) is the total number of trajectories, \(C_{i}\) is the observed BC_{df} (BC_{bf}) concentration at receptor site on arrival of trajectory \(l\) and \(\tau_{i,j,l}\) is the number of endpoints in the \(i/j\)th grid cell of trajectory \(l\). In general, the grid cells with high CWT values are high strength sources. The weighting function \(w_{i,j}\) was also adopted in the CWT analysis to reduce the effect of the small values of \(n_{i,j}\).
2.4 Source-oriented modeling

In this analysis, source apportionment of BC over Xiamen using an updated source-oriented CMAQ model for primary particulate matter (CMAQ-PPM) (Hu et al., 2015; Guo et al., 2017) was also implemented in addition to the observation-based source apportionment. The CMAQ-PPM model was updated on basis of CMAQ v5.0.1, which was developed by the U.S. EPA Atmospheric Science Modeling Division. The photochemical mechanism and aerosol chemistry mechanism adopted in this study were SAPRC-11 and AERO6, respectively. In the source-oriented model, tagged non-reactive PM tracers are used to estimate the source contributions of PPM and its chemical components. The PM tracers are set to undergo the same atmospheric processes as other species. The emissions of the tracers are set to 0.001 % of the PPM emissions from each corresponding source sector and region. It ensures that the tracers will not significantly change the particle mass and size. After scaling up by $10^4$, the simulated tracer concentration represents the PPM concentrations from a specific source type/region. The concentrations of the inert chemical components in PPM can be estimated with source specific emission profiles as follows:

$$C_{i,j} = A_{i,j} \times PPM_i$$

where $C_{i,j}$ is the concentration of the $j$th component from the $i$th source, $A_{i,j}$ is the ratio of the $j$th component in PPM mass from the $i$th source and $PPM_i$ is the simulated concentration for the $i$th source. Detailed descriptions of the model can be found in Hu et al. (2015).

Regional distributions of BC from different categories (sectors) as well as the source category (sector) contributions to BC at the receptor site were determined with the source-oriented CMAQ-PPM model. A 36 km horizontal resolution domain that covers China and surrounding countries in East Asia (Fig. 1) was applied. There are 18 vertical layers with surface layer thickness of 35 m and the overall model height of 20 km. The Weather Research & Forecasting model (WRF) v3.9.1 was utilized to generate meteorology inputs with initial and lateral boundary conditions from NCEP FNL reanalysis data from NCAR, which is available on $1^\circ \times 1^\circ$ grids continuously for every 6 h (http://dss.ucar.edu/datasets/ds083.2/). There are 29 vertical layers in the WRF domain. The first eight layers of the WRF and CMAQ domains are identical. The outputs of WRF were post-processed by Meteorology-Chemistry Interface Processor (MCIP) v4.2 to the format CMAQ requires. Anthropogenic emissions in China were generated according to the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University (http://www.meicmodel.org). Emissions from other countries and regions outside China were generated with the Regional emission inventory in Asia version 2 (REAS2) (Kurokawa et al., 2013). The fire emissions were derived from the Fire Inventory from NCAR (FINN) based on satellite observations (Wiedinmyer et al., 2011). Anthropogenic emissions were grouped into four sectors including industrial, power, transportation and residential. Open burning emissions are considered as the fifth emission sector. Sources from five sectors were further classified into three categories such as solid fossil fuel (i.e. coal) combustion, liquid fossil fuel combustion and biomass burning on basis of the
energy consumption data provided by Wang et al. (2012). Spatial distribution of BC concentration over China are depicted in Fig. S1, suggesting that BC concentration in the Western Taiwan Strait region was much lower than that in other urban agglomerations in north China, east-central China and Sichuan Basin. However, Xiamen was with relatively higher abundance compared to surrounding areas.

3 Results and discussion

3.1 Light absorption-based source apportionment of BC

Fig. 2 demonstrates the temporal variations in daily mean concentrations of BC$_{ff}$ and BC$_{bb}$ with the BC$_{bb}$/BC fraction in Xiamen during the field campaign. The missing data were due to the instrument maintenance. Daily concentrations of BC$_{ff}$ and BC$_{bb}$ were 445 – 9545 ng m$^{-3}$ (average: 2932 ± 1444 ng m$^{-3}$) and 334–4031 ng m$^{-3}$ (1340 ± 542 ng m$^{-3}$), respectively. Daily contribution of BC$_{bb}$ to total BC varied significantly in the range of 18.4% – 58.3%. The annual average contribution of BC$_{bb}$ to BC was 33.3%, much smaller than that of BC$_{ff}$ (66.7%), indicating the predominant contribution of fossil fuel combustion in Xiamen. Source apportionment results under different pollution levels of BC and PM$_{2.5}$ in each season were further investigated to understand BC sources on pollution and clean days (Fig. S2). The data of PM$_{2.5}$ concentration were from Xiamen Environmental Monitoring Central Station. High (low)-BC periods were the days with the average concentration of BC higher (lower) than the seasonal average concentration plus (minus) one standard deviation, and so did PM$_{2.5}$. Generally, source contributions of BC show obvious variability among different pollution levels in all seasons, and the BC$_{bb}$ percentage decreases with the increasing concentrations of BC and PM$_{2.5}$. Biomass burning contributed more during low-BC (30.8% – 43.1%) and low-PM$_{2.5}$ days (31.5% – 40.7%) compared to high-BC (24.8% – 34.4%) and high-PM$_{2.5}$ episodes (26.6% – 36.1%), implying that emissions from coal combustion and vehicle exhausts are major contributors of particulate pollution in Xiamen. The fractional contribution of fossil fuel to BC in Xiamen derived by the Aethalometer method in this work suggests a slighter larger role of fossil fuel compared to that (61%) estimated according to the “bottom-up” emission inventories (Chen et al., 2013). However, it was similar to the contribution (~70%) in YRD and PRD, which was estimated based on dual carbon isotope constrained source apportionment (Andersson et al., 2015). BC$_{ff}$ and BC$_{bb}$ percentages in different regions calculated with the Aethalometer method were summarized in Table S1 for comparison. BC$_{bb}$ fraction in Xiamen were overall larger than that in Nanjing in China and other sites in India, suggesting that contribution of biomass burning increases over the relatively clean region due to the weak emissions of traffic and coal combustion.

Fig. 3 illustrates the diurnal and monthly cycles of BC$_{ff}$ and BC$_{bb}$ concentrations as well as the relative contribution of biomass burning (BC$_{bb}$/BC) during the measurements. BC$_{ff}$ exhibited a pronounced diurnal variation, increasing steadily before dawn with the major morning peak (4427 ng m$^{-3}$) observed around 6:00 in the morning. The high BC$_{ff}$ concentrations
at the observation site from late night to the early morning (~21:00 to 8:00) may ascribe to enhanced traffic emissions from diesel trucks during nighttime and cars during rush hours. The heavy diesel trucks, which are major emission sources of BC_{dt}, were allowed to enter the city from 22:00 to 7:00. Therefore, BC_{dt} decreased during daytime and reached the diurnal minimum of 1950 ng m^{-3} at 13:00 in the afternoon. BC_{hb} exhibited a diurnal trend that was different with BC_{dt}. The morning peak (1755 ng m^{-3}) at 6:00 was also found for BC_{hb}. However, BC_{hb} concentration kept a steady state rather than increase after 20:00 since BC_{hb} was less influenced by traffic-related emission. Clear diurnal variation in contribution of BC_{hb} to total BC was found. The BC_{hb} fraction reached its valley of 30.4 % at 8:00, increased due to the decrease in traffic emission and maximized with the ratio of 37.9 % at 19:00 during the rush hour in the evening. The diurnal cycles of BC_{dt} and BC_{hb} were affected by not only the BC emission trend but also the evolution of atmospheric boundary layer. According to our previous study on atmospheric boundary layer height in Xiamen (Deng et al., 2020), the boundary layer height was ~three times larger in the afternoon than that in the early morning, leading to the better diffusion conditions in the afternoon.

Monthly mean BC_{hb} concentration peaked with value of 1979 ng m^{-3} in December and reached its valley of 923 ng m^{-3} in June. The monthly pattern of BC_{dt} was similar but a bit different with that of BC_{hb}. The maximum monthly mean BC_{dt} concentration was 3636 ng m^{-3} in March, while the minimum was 1881 ng m^{-3} in February. The valley of BC_{dt} concentration occurring in February was maybe because of the lack of vehicle (e.g., diesel trucks) emissions around the Spring Festival holiday, which again prove the conjecture in the diurnal pattern of BC_{dt}. Similar to the seasonal pattern of absorption Ångstrom exponent (Qiu et al., 2019), noticeable seasonal variation in the BC_{hb}/BC fraction was found. Winter (December–February) had the largest BC_{hb} contribution (39.9 %), followed by fall (September–November) (32.1 %), spring (March–May) (31.1 %) and summer (June–August) (29.6 %). The much larger contribution in winter are possibly due to the enhanced source from open-field biomass and domestic burning in China. The higher BC_{hb} concentration and contribution lasted from fall to early winter, consistent with previous emission inventory of biomass burning, which found higher BC emissions from November to February than other months (He et al., 2011). Unlike BC_{dt}, BC_{hb} exhibited an increase trend in July, leading to a relatively large contribution of BC_{hb}. It might be affected by the long-range transport of air pollutants emitted from biomass burning in Southeast Asia under the control of summer monsoon (Qiu et al., 2019). The monthly variation in boundary layer height, which was larger in warm season and smaller in cold season, also affected the monthly patterns of BC_{dt} and BC_{hb} (Deng et al., 2020).

3.2 Sources and transport pathways of BC_{dt} and BC_{hb}

The CPF results for the top 25 % thresholds of concentrations of BC_{dt} (3797 ng m^{-3}) and BC_{hb} (1813 ng m^{-3}) as well as BC_{hb} contribution (45 %) over different periods are shown in Fig. 4. In the whole year, high BC_{dt} concentrations were mainly associated with winds from west-southwest to north-northeast with wind speed (ws) < 2 m s^{-1} (Fig. 4a). Particularly, high
BC_{ff} concentrations were most remarkably distributed in winds from the northwest at low ws (< ~1 m s^{-1}) and to a lesser extent from the west and north-northeast at moderate ws (< 3 m s^{-1}). It implies the impacts of local sources such as the traffic emissions to the northwest of the site within a short distance. The CPF pattern for BC_{fb} was similar to but not the same with that of BC_{ff} (Fig. 4b). In addition to northwesterly wind with low ws, northeasterly and easterly winds with ws < 5 m s^{-1} were also accompanied by high BC_{fb} concentration. Correspondingly, the CPF plot for BC_{fb}/BC fraction implies the significant influence of east-northeasterly wind with ws > 2 m s^{-1} on large contribution of biomass burning (Fig. 4c). In addition, northerly wind with wind speed > ~4 m s^{-1} were also frequently associated with large BC_{fb} fraction. CPF patterns presented obvious seasonality. For BC_{ff} and BC_{fb}, the CPF distributions over spring, summer and fall were similar and the high concentrations were mainly associated with northwesterly wind with ws < 2 m s^{-1}. However, in winter, additionally with wind from northwest, high BC_{ff} and BC_{fb} concentrations were also frequently associated with wind from southwest and west with ws < 3 m s^{-1}. For BC_{fb}/BC fraction, large fractions were mainly distributed in northeasterly wind with ws > 4 m s^{-1} and most remarkably distributed in winds from the northeast at high ws (> 6 m s^{-1}) in spring and summer. In fall, east-northeasterly wind with ws > 3 m s^{-1} was more frequently associated with large contribution of biomass burning. However, in winter, northerly wind with ws > 4 m s^{-1} and northeasterly wind with ws > 2 m s^{-1} were most remarkably associated with large BC_{fb} percentage.

Seasonal clusters of backward trajectories obtained by the HYSPLIT model with the average BC_{fb} contributions are illustrated in Fig. 5. Mean concentrations of BC_{fb}, BC_{ff} and BC of each cluster in different seasons are summarized in Table S2. It is clearly shown that origins and transport pathways of air masses arriving in Xiamen exhibited distinct seasonal variations. In summer, air masses were characterized by a predominance of southerly origination. In contrast, in other seasons, air masses from the north had a dominant position, which was particularly the case in winter. Generally, air masses from the northern inland region such as north China and east-central China had larger biomass burning contributions comparing to those from the seas such as East China Sea and South China Sea, since there are dense emissions of biomass burning in northern and eastern China including Hebei, Henan, Shandong and Jiangsu (Huang et al., 2012; Wu et al., 2018).

In spring, the eastern cluster (C4) originating from the East China Sea had the lowest BC_{fb} fraction (31%). However, the northern cluster (C3) originating from Siberia and passing through Mongolia and north and east China had much larger biomass burning contribution (42%) in comparison to the other clusters. In summer, northeastern coastal cluster (C4) originating from the East China Sea and passing along with East China Coast region had larger biomass burning contribution (38%). The north cluster passing through Jiangsu, Zhejiang and north Fujian province also had relatively higher BC_{fb} fraction (35%). In fall, the northern inland cluster (C3) originating from Siberia and passing through the heavily polluted areas such as North China Plain and YRD were associated the largest biomass burning contribution (40%), followed by the other long-range inland cluster (C2) with the BC_{fb} fraction of 36%. In winter, similar to spring and fall, the northern cluster from Siberia (C2) had the largest biomass burning contribution (48%). Contrarily, the northeastern marine air masses
passing along with the coastal region had the lowest BC\textsubscript{bb} fraction (35 %).

Potential sources of BC\textsubscript{ff} and BC\textsubscript{bb} in Xiamen with their contributions were characterized with PSCF and CWT analyses and the results are presented in Fig. 6. According to the PSCF and CWT maps of BC\textsubscript{ff} (Fig. 6a, c), the strong potential sources probabilities for BC\textsubscript{ff} distributed to southwest of Xiamen, including southwest Fujian province as well as Guangdong province. Significant potential sources were also located in Hebei, Anhui, Jiangxi and Henan provinces in east-central China and Southeast Asia with strong regional BC emissions (Permadi et al., 2018) was also indicated as the potential source regions. For BC\textsubscript{bb}, similar to BC\textsubscript{ff}, the PSCF and CWT distributions (Fig. 6b, d) show that the exogenous potential sources were mainly distributed in east-central China, which belonged to the major areas of biomass burning in China (Yan et al., 2006; Huang et al., 2012; Wu et al., 2018). Guangdong province in south China was also suggested as the source of BC\textsubscript{bb}. Unlike BC\textsubscript{ff}, the strong potential sources probabilities from Southeast Asia to BC\textsubscript{bb} were less significant.

Fig. 7 and Fig. 8 depict the seasonal PSCF and CWT distributions for BC\textsubscript{ff} and BC\textsubscript{bb}. Source distributions of both BC\textsubscript{ff} and BC\textsubscript{bb} in different seasons significantly varied due to the variability in the airflows. In spring, the terrestrial contributions from Guangdong province and north China as well as the potential source from the East China Sea for BC\textsubscript{ff} and BC\textsubscript{bb} were significant. The South China Sea were the main potential source for BC\textsubscript{ff} and BC\textsubscript{bb} during summer. In addition, high PSCF and CWT values for summer BC\textsubscript{bb} were also found in north China. In fall season, similar to spring, high PSCF and CWT values for BC\textsubscript{ff} and BC\textsubscript{bb} were distributed in east-central China and the East China Sea. In winter, the main potential sources for BC\textsubscript{ff} and BC\textsubscript{bb} were also located in east-central China, while the contributions from the seas were small. The potential sources for BC\textsubscript{bb} in central China were much stronger in winter than that in other seasons.

### 3.3 Source-oriented modeling-based source apportionment of BC

Relative source contributions to BC in Xiamen from different source sectors and fuel catalogs were assessed with the source-oriented CMAQ-PPM model. Fig. 9 illustrates the seasonal and annual average contributions of each source sector in Xiamen. Overall, transportation, residential and open burning sectors were the major sources of BC, with the annual contributions of 45.3 %, 30.1 % and 17.6 %, respectively. By comparison, power plants and industrial sectors made insignificant contributions to BC. The transportation sector was the dominant source in all seasons, especially in summer, contributing 36.5 % – 56.6 % to total BC. The residential sector contributing 20.5 % – 37.2 % was the second largest source in all seasons except spring. On contrary, power plants and industrial sectors were minor sources in all seasons, with the seasonal contributions of 2.2 % – 6.2 % and 2.8 % – 4.6 %, respectively. Obvious seasonal pattern of contribution of open burning was found. In spring and summer, open burning played a vital role by contributing 35.5 % and 17.8 %, respectively. However, its relative contributions dramatically decreased to 7.6 % in fall and 7.5 % in winter. Source contributions of the
The remarkable seasonal and spatial variations from open burning are consistent with those derived in previous study (Hu et al., 2015). In spring, strong open burning in south China might significantly influence BC concentrations in the surrounding regions near the sources, which would contribute to larger biomass burning contribution in Xiamen. Intensive open burning in South Asia and Southeast Asia countries in spring also affected biomass burning contribution in Xiamen through long-range transport.

Seasonal variations in simulated relative contributions of three fuel catalogs, including coal, liquid fossil fuel and biomass, to BC in Xiamen were also characterized and compared with relative contributions of biomass burning estimated according to light-absorption properties (Fig. 10). The source-oriented model almost captured the seasonal trends in contributions of fossil fuel combustion and biomass burning, although its largest seasonal contribution of biomass burning occurred in spring rather than winter. For the source-oriented modeling results, liquid fossil fuel combustion had the largest contribution (46.5%), followed by biomass burning (32.6%) and coal combustion (20.9%). Contributions of different fuel catalogs exhibited distinct seasonality. Seasonal contributions of coal combustion were in the order of winter (27.8%) > fall (23.4%) > spring (15.6%) > summer (14.5%). Different from coal combustion and biomass burning, liquid fossil fuel combustion had the largest contribution in summer (57.5%) while smallest contribution (37.4%) in spring.

Compared to observation-based results, relative contribution of biomass burning derived by the source-oriented modeling was 14.3% smaller in winter, while 15.9% larger in spring. The large gap in spring may be resulted from the uncertainties in satellite-based inventory of biomass-burning emissions in South Asia and south China (Wiedinmyer et al., 2011), while the disagreement in winter may ascribe to the underestimation of biomass-burning emissions in north and central China (Huang et al., 2012; Zhou et al., 2016). In China, some open burning activities such as local/small-scale open burning and smoldering are important sources of biomass-burning BC, which was particularly the case in winter. However, these burning activities are difficult to be detected by satellite, leading to underestimation of biomass-burning emissions. The discrepancies between the results from the two source apportionment methods were much smaller in summer (1.7%) and fall (6.0%). Seasonal relative contributions of biomass burning estimated by the source-oriented model were in the range of 25.7% – 47.0%, with an annual average of 32.6%, which was very close to the observed results derived from the Aethalometer method. The consistence of the two different source apportionment methods confirms that the source apportionment results in Xiamen from this study are reasonable and benefiting future emission-control strategies.

4 Conclusions

In this study, the observation-based light absorption and source-oriented modeling were combined to reveal the contributions of biomass burning and fossil fuel combustion to ambient BC aerosol as well as their temporal variations in a relatively clean region in China. The annual average concentration of BC from fossil fuel (BC$_{ff}$) and biomass burning (BC$_{bb}$) identified by...
the Aethalometer method were $2932 \pm 1444$ ng m$^{-3}$ and $1340 \pm 542$ ng m$^{-3}$, accounting for 66.7% and 33.3% of total BC, respectively. For biomass burning contribution, its highest level occurred in the evening rush hour, while the maximum seasonal value was in winter. East-northeasterly and northerly wind was more likely to result in large biomass burning contribution. Air masses from the northern inland region including north China and east-central China had larger biomass burning contributions. Potential sources for BC$_{ff}$ and BC$_{bb}$ indicate the impact of long-range transport from north and east-central China and Southeast Asia. Overall, the source-oriented model presented a good agreement with the Aethalometer method and it was able to reproduce the observed seasonal variability of biomass burning based on light absorption. Based on the source-oriented model, the transportation, residential and open burning sectors were the larger contributors to BC compared to the power and industrial sectors. The largest contribution of liquid fossil fuel combustion to BC was identified by the source-oriented model, followed by biomass burning and coal combustion. The discrepancies between the two source apportionment methods suggest emission inventory with higher spatiotemporal resolution is required in future studies to provide more accurate source apportionment results of BC in China.

Data availability. The data is available upon request from Junjun Deng (dengjunjun@tju.edu.cn).

Author contribution. JD and HZ designed the experiments and carried them out. JD and WZ performed the analysis of observation. HG and HZ performed the source-oriented modeling. JZ, WH, LW, XW and PF provided suggestions for data analysis. JD prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interests.

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Figure 1. Location of Xiamen, China, with spatial distribution of annual average BC emission rate (g s\(^{-1}\)). BC emission data in China are from the MEIC inventory developed by Tsinghua University.

Figure 2. Daily BC\(_{ff}\) and BC\(_{bb}\) concentrations and BC\(_{ff}\)/BC fraction in Xiamen in 2014.
Figure 3. Diurnal and monthly variations in BC\textsubscript{ff} and BC\textsubscript{bb} concentrations with the BC\textsubscript{bb}/BC fraction.
Figure 4. CPF plots for (a) BC\textsubscript{ff}, (b) BC\textsubscript{bb} and (c) BC\textsubscript{bb} contribution in Xiamen in 2014. ws represents wind speed (m s\(^{-1}\)).
Figure 5. Seasonal cluster-mean of five-day backward trajectories at 500 m with the corresponding trajectory percentages and BC$_{10}$ contributions in Xiamen.

Figure 6. Potential source contribution function (PSCF) maps for (a) BC$_{ff}$ and (b) BC$_{bb}$ and concentration-weighted trajectory (CWT) maps (ng m$^{-3}$) for (c) BC$_{ff}$ and (d) BC$_{bb}$ in Xiamen in 2014.
Figure 7. Potential source contribution function (PSCF) maps for BC_{ff} and BC_{bb} in Xiamen for different seasons in 2014.
Figure 8. Concentration weighted trajectory (CWT) maps for BC_{ff} and BC_{bb} in Xiamen for different seasons in 2014.
Figure 9. Source contributions to BC of five source sectors in each period.

Figure 10. (a) Seasonal and (b) diurnal variations in relative source contribution of three fuel catalogs to BC in Xiamen.