Multi-year emission of carbonaceous aerosols from cooking, fireworks burning, sacrificial incenses, joss paper burning, and barbecue and the key driving forces in China

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Abstract

There has been controversy about the air pollutants emitted from sources closely related to people's daily life (such as cooking, fireworks burning, sacrificial incenses and joss paper burning, and barbecue, named as five missing sources, FMS) impacting the outdoor air quality to what extent. Till now, there is no emission estimation of air pollutants from FMS, as the missing of both activity dataset and emission factors. We attempted to combine the questionnaire data, various statistical data, and data of points of interest to obtain a relatively complete set of activity data. The emission factors (EFs) of carbonaceous aerosols were tested in our lab. Then, the emission inventories of carbonaceous aerosol with a high spatial-temporal resolution for FMS were established firstly, and the spatial variation trend and driving forces were discussed. From 2000 to 2018, organic carbon (OC) emissions were in the range of 4268–4919 t. The OC emission from FMS was 1.5–2.2 ‰ of its total emission in China. The emissions of black carbon, element carbon (EC), and brown carbon absorption cross-section (ACS_{BrC}) emissions from FMS were in the ranges of 22.6–43.9 t, 213–324 t, and 14.7–35.6 Gm^2, respectively. Their emissions tended to concentrate in special periods and areas. The OC emission intensities in central urban areas were 3.85–50.5 times that of rural areas due to the high density of human activities. While the ACS_{BrC} emissions in rural regions accounted for 63.0–79.5% of the total emission result from uncontrolled fireworks burning. A mass of fireworks burning led to extremely higher ACS_{BrC} and EC emissions on Chinese New Year's eve, as 1444 and 262 times their corresponding yearly average values. Significant ($p<0.01$) correlations between human incomes and pollutant emissions were found, while they were positive ($r = 0.94$) and negative ($r = –0.94$) for urban and rural regions, indicating the necessity of regulating human lifestyle and increasing income for urban and rural peoples, respectively. This study provided the first-hand data for identifying the emissions, variation trends and impacting factors of FMS, which is helpful for modeling works on air quality, climate effect, and human health risks at specific periods or regions and for modifying their emission control policies. The data in this work could be found at https://doi.org/10.6084/m9.figshare.19999991.v2 (Cheng et al., 2022).

Keywords: Carbonaceous aerosols; Sources related to human activities; Emission inventory; Spatial-temporal variation; Driving force

1 Introduction

China has experienced a period of serious air pollution, which produces a great health impact on residents (Zheng et al., 2018; Zhang et al., 2019, 2020b; Tong et al., 2020). Carbonaceous aerosols (CA), emitted from incomplete burning, include organic carbon (OC) and black carbon (BC, or element carbon, EC), and they have attracted wide attention due to their adverse impacts on air quality, human health and climate (Venkataraman et al., 2005; Ramanathan & Carmichael, 2008; Bond et al., 2013). The optical properties of CA (especially brown carbon, BrC) are complex and mutative, which is also one of the important factors affecting the global radiation balance (Feng et al., 2013; Laskin et al., 2015).

Several sources closely related to traditional human activities were potential emission sources of CA, such as the burning of sacrificial incense and joss paper, traditional Chinese barbecue, Chinese style cooking, and fireworks burning. The estimation of air pollutants from these sources was missing in the previous emission inventory, and they were defined as five missing sources (FMS) in this study. FMS can lead to a dramatic impact on ambient quality and human health in a short period or a specific region (Chiang & Liao, 2006; Wu et al., 2015; Kong et al., 2015; Ho et al., 2016; Wang et al., 2017; Lai & Brimblecombe, 2020). For example, fireworks burning could contribute 60.1 % of $PM_{2.5}$ during the Chinese Lunar New year's eve in Nanjing, and sacrificial sources like sacrificial incense and joss paper burning could contribute 17.5% of atmospheric PAHs in Chu-Shan, and 9.6% of PCDD/F in Taipei (Lin et al., 2008; Kong et al., 2015; Ho et al., 2016). Recently, with the strengthened control of combustion-related sources, the important role of cooking emissions on affecting air quality was gradually visible in densely populated downtown areas. Cooking organic aerosols contributed to 15−34% of total OC and 6−9% of total $PM_{2.5}$ in a downtown site in Shanghai (Huang et al., 2022) and 31% of total organic aerosols in Beijing in winter (Hu et al., 2022). The existing studies on FMS were mainly based on the ambient air monitoring datasets at certain sites or certain periods (See & Balasubramanian, 2011; Wu et al., 2015; Shen et al., 2017; Lao et al., 2018; Tanda et al., 2019; Yao et al., 2019; Hu et al., 2021; Huang et al., 2021). Till now, no studies can provide their contributions quantitively on a large scale as the scare of emission inventory, which limited the identification of their contributions in various regions or periods.

There also existed extensive queries that are these conclusions tenable as they were identified through models or ambient monitoring data, but not from their real emission estimation. In China, the differences in

75 population and economy between urban and rural areas are increasing (Meng et al., 2019), and the

76 efficiencies and necessity of air quality control policies for FMS in urban and rural areas need to be assessed.

77 For instance, fireworks burning was generally banned in the central urban region, while suburbs and rural

78 regions were affected less by the policy. The cooking smokes needed to be purified in city centers, while in

79 suburban and rural areas, the policy may be not strictly executed or it is even not necessary. Such deviations

80 in policy establishment and implementation could ultimately drive the differences in the distribution of air

81 pollutant emissions, which have not been addressed yet.

82   The emission inventory is the base for the quantitative description of anthropogenic pollutant emissions

83 (Li et al., 2017). The combination of the chemical transport models and high-resolution emission inventory

84 was paramount for understanding anthropogenic perturbations' impacts on the atmosphere, and for assessing

85 corresponding air pollution control strategies (Janssens-Maenhout et al., 2019; McDuffie et al., 2020). The

86 lack of emission inventories limits the large-scale model simulation, the optimization of corresponding

87 control measures, and the settlement of related disputes. In our previous work, Wu et al. (2021) have

88 established an emission inventory of levoglucosan which included the emissions from FMS (Wu et al., 2021).

89 There were no other emission inventories of FMS reported, to the best of our knowledge.

90   To sum up, this study aimed to develop a methodological framework for establishing an emission

91 inventory of FMS, including the methods of various activity data acquisition, emission factors monitoring,

92 uncertainty assessment, and spatial-temporal allocation. The activity data were obtained based on household

93 investigation, statistic data, points of interest (POI), etc. The emission factors were monitored through a

94 unique emission monitoring test platform, especially for fireworks burning in our lab. Then a high spatial

95 (~1 km) and temporal resolution (1 day for special festivals, and 1 month for the rest) emission inventory

96 was first established. The multi-year spatiotemporal variation of CA emissions from these sources was

97 analyzed and compared with other types of sources. Optimization pollution control measures were proposed

98 for these types of sources. The study provides a methodology for establishing an emission inventory of air

99 pollutants from sources closely related to human activities. Other air pollutants emissions could also be

100 estimated in the future. The emission inventory obtained here could also provide the basic inputs for

101 corresponding modeling works.

102

2 Methodology

2.1 Combustion tests for emission factors

The combustion tests for FMS were performed with two custom-made combustion chambers. One of them had an explosion-proof function and it was used in fireworks burning experiments. Another one was used in sacrificial incense, joss paper, barbecue, and cooking emission experiments. A dilution sampling system (Dekati FPS-4000, Finland) was employed to dilute the smoke. The smokes were diluted about 16–30 times and aged for about 30 s in a residence chamber. The sampling system has been utilized in residential fuel combustion experiments (Cheng et al., 2019; Yan et al., 2020; Zhang et al., 2021b; Wu et al., 2021). Thirty-eight events were tested in this experiment, including 6 trials of sacrificial incense combustion (abbreviations of materials: red incense: RI; environmental incense: EI; high incense: HI), 6 trials of joss paper burning (red-printed paper: RP; small sacrificial paper: SP; large sacrificial paper: LP), 10 trials of fireworks burning (firecrackers: FC; fountain fireworks: FF; handheld fireworks: HF; handheld fountain: HT; spin fireworks: SF), 8 trials of barbecue (chicken: CK; beef: BF, lamb: LB; pork: PK), and 8 trials of cooking (cooking of meat: MT1; cooking of meat and pepper: MT2; cooking of meat and garlic: MT3; cooking of meat, pepper, and garlic: MT4). The experiment materials were shown in **Figure S1**.

After diluted, the OC and EC in the smokes were detected with an online carbonaceous aerosol analyzer. It was developed by the Key Laboratory of Environmental Optics & Technology (Anhui Institute of Optics and Fine Mechanics, CAS) based on the thermal-optical method (Ding et al., 2014). The analyzer showed reliable stability and repeatability. More details on the online carbonaceous aerosol analyzer can be found in **Text S1**. A dual-spot Aethalometer (Model AE33, Magee Scientific, USA) was employed to measure BC concentration and particulate optical properties (Drinovec et al., 2015). The experiment system was shown in **Figure S2**.

2.2 Calculation of emission factors and optical properties

The emission factors of OC, EC, and BC were calculated by equation (1):

$$EF_{ij} = \frac{v \times m_{ij} \times r}{v_0 \times M_j} \quad (1)$$

where i and j denoted pollutant and fuel; EF was the emission factor (mg kg^{-1}); v was the flue gas flow (L min^{-1}); v_0 was the sampling flow (L min^{-1}); m was the mass of the pollutant detected by the instruments (mg); r was the dilution ratio; M was the mass of the material used in each tail of experiments

131 (kg) (Cheng et al., 2019; Yan et al., 2020).

132 All filter-based optical measurements will be confronted with the underestimation caused by the loading

133 effect (Drinovec et al., 2015). The loading compensation could be calculated based on dual-spot

134 measurements. The detailed calculation process was referred to Drinovec et al. (2015). There was inferior

135 dependence of BC particles on light absorption in different light wavelengths. The absorption Ångström

136 exponent (AAE) was an exceptional parameter to describe this dependence, as shown in equations (2) and

137 (3):

$$b_{abs} \sim \lambda^{-AAE} \qquad (2)$$

$$AAE = -\frac{\ln\left(\frac{b_{abs}(\lambda_1)}{b_{abs}(\lambda_2)}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)} \qquad (3)$$

140 where λ was the wavelength; b_{abs} denoted the total light absorption coefficient (Tian et al., 2019),

141 which could be calculated by equation (4) (Zotter et al., 2017):

$$BC = b_{abs}(\lambda)/MAC(\lambda) \qquad (4)$$

143 where MAC was the mass absorption cross-section, referring to the Aethalometer manufacturer.

144 As shown in equation (5), the b_{abs} of CA was aroused by BC and BrC. To calculate the $b_{abs}(\lambda, BC)$

145 at each wavelength, equation (6) was introduced. The AAE_{BC} was determined as 1.0 according to previous

146 studies (Tian et al., 2019; Liakakou et al., 2020). $f_{BrC}(\lambda)$ (equation (7) was utilized to estimate the fraction

147 of BrC light absorption ($b_{abs}(\lambda, BrC)$) in total light absorption ($b_{abs}(\lambda)$).

$$b_{abs}(\lambda) = b_{abs}(\lambda, BC) + b_{abs}(\lambda, BrC) \qquad (5)$$

$$b_{abs}(\lambda, BC) = b_{abs}(880) \times \left(\frac{\lambda}{880}\right)^{-AAE_{BC}} \qquad (6)$$

$$f_{BrC}(\lambda) = 100\% \times b_{abs}(\lambda, BrC)/b_{abs}(\lambda) \qquad (7)$$

151 Due to the complicated chemical properties of BrC, it was difficult to measure the accurate

152 concentration of BrC in flue gases. Previous studies have developed a peculiar EF called absorption emission

153 factor (AEF), as shown in equation (8) (Martinsson et al., 2015; Tian et al., 2019; Zhang et al., 2020c). Most

154 studies modeled the direct radiative forcing of BrC with its mass concentration and mass absorption

155 efficiency (MAE) as the input parameters. While the mass concentration and the total mass of BrC in the

156 atmosphere were still unclear, and the MAE values in the range of 0.08–3.8 m^2 g^{-1} were also variable (Park

157 et al., 2010; Feng et al., 2013; Wang et al., 2014b; Zhang et al., 2020a). An inventory established by AEF as

158 the following equation could avoid the deviation raised by the mass concentration and MAE of BrC (Tian et

159 al., 2019).

160
$$AEF_{ij} = \frac{\sum_{t_0}^{t_{sample}}(b_{abs_{ij}} \times r \times v)}{M_j} \quad (8)$$

2.3 Acquisition of activity data

162 Data on the activity of sources directly affected the uncertainties of emission inventory, and an accurate

163 estimate of FMS consumption was a crucial prerequisite. Statistics on direct consumption of FMS were

164 scarce. Then multiple activity data and proxy variables were adopted, including the statistical yearbooks of

165 each province in China, datasets of POI (**Text S2**, **Figure S3**), and rural household investigation data in our

166 group (**Text S3**).

167 The original consumptions of sacrificial incenses, joss paper and fireworks were from a household

168 investigation. We got the per capita consumption of sacrificial incenses, joss paper, and fireworks in each

169 province. The data were adjusted to overcome the problem of insufficient sample size. In China, sacrificial

170 activities mean honoring ancestors, and they mainly take place in temples or graveyards. China is a

171 mountainous country with rolling terrain. Most of the inhabitants of non-plain areas chose hills that might

172 cover vegetation as the site of graveyards. The data on the consumption of sacrificial incenses and joss paper

173 will be revised based on the number of temples (data from POI) and frequency of forest fires caused by

174 sacrifices to the total forest fires (data from China Forestry Statistical Yearbook), as shown in equation (9):

175
$$Cs_{adj,province} = Cs_{inv,province} \times (\frac{Nt_{province}/POP_{province}}{2 \times Nt_{nation}/POP_{nation}} + \frac{Fs_{province}/Ft_{province}}{2 \times Fs_{nation}/Ft_{nation}}) \quad (9)$$

176 where Cs was the consumption of sacrificial incenses or joss paper per capita; Nt was the number of

177 temples; POP was the population, which came from the statistical yearbook of each province; Fs was the

178 forest fires raised by sacrificial activities; Ft was the forest fires raised by all anthropogenic activities; adj

179 represented the data after adjusted; inv represented the data from the household investigation; $province$

180 represented the data of each province; $nation$ represented the data of the entire nation.

181 The fireworks consumption amounts will be revised based on the number of retail shops of fireworks

182 (data from POI) and provincial fireworks export volume (statistical data), as shown in equation (10):

183
$$Cf_{adj,province} = Cf_{inv,province} \times (\frac{Ns_{province}/POP_{province}}{2 \times Ns_{nation}/POP_{nation}} + \frac{Ve_{province}/POP_{province}}{2 \times Ve_{nation}/POP_{nation}}) \quad (10)$$

where Cf was the consumption of fireworks per capita; Ns was the number of retail shops of fireworks; Ve was the export volume of fireworks (data from China Light Industry Yearbook). In addition, the consumptions (C) of sacrificial incenses, joss paper, and fireworks at the municipal level were calculated by combining the POI data and the consumptions at the provincial level, as shown in equation (11).

$$C_{adj,city} = C_{inv,province} \times \frac{N_{city}/POP_{city}}{N_{province}/POP_{province}} \quad (11)$$

where N represented the number of temples or firework shops.

The original meats consumption per capita came from the statistical yearbook of each province. While the methods and radii of different provincial statistical yearbooks showed differences. Part of the municipal level statistics was missing. To complement the missing data, municipal per capita consumption expenditure was introduced. The logarithmic relationship between per capita consumption expenditure and provincial per capita meat consumption was adopted to complement municipal per capita meat consumption, as shown in equation (12):

$$y = a \times \ln x + b \quad (12)$$

where x represented the provincial per capita consumption expenditures in 2000–2018; y represented provincial per capita meat consumption in 2000–2018. Parameters a and b were fit-out for each province ($r = 0.60$, $p < 0.01$). The parameters a and b of the province where each city is located, and the per capita consumption expenditure of the city were substituted into equation (12) to calculate the municipal per capita meat consumption.

2.4 Calculation of the emissions

Since some cities have established policies to forbid sacrificial incense, joss paper, and fireworks burning in the main urban area, and such policies were inoperative in non-urban regions. According to our survey, the policies on forbidden sacrificial incense and joss paper were relatively vague. We assumed that if one city forbade fireworks burning, then the burning of sacrificial incense and joss paper was also banned. The total emissions E from sacrificial incense, joss paper, and fireworks were calculated by equation (13):

$$E = \sum (POP_{urban,k} \times C_{urban,k} \times FB_k + POP_{non-urban,k} \times C_{non-urban,k}) \times EF \quad (13)$$

where k denoted the different cities; $urban$ and $non-urban$ represented urban regions and non-urban regions (rural regions); POP was the population; C was the per capita consumption; EF was the emission factor. $FB = 0$ or 1 depended on whether the burning of sacrificial incense, joss paper, and

firework was forbidden in urban regions. Unfortunately, there was no such detailed consumption data that involved the fuel types of sacrificial incense, joss paper, and fireworks. Thus, for the calculation of emission from sacrificial incense, joss paper, and fireworks, mean EFs were utilized here.

Emissions from barbecue were calculated by formula (14):

$$E = \sum \left(POP_{urban,k} \times MC_{urban,k} + POP_{non-urban,k} \times MC_{non-urban,k} \right) \times \frac{T_{BBQ,k}}{T_{total,k}} \times OP \times EF \quad (14)$$

where MC was the meat consumption mass per capita; T_{BBQ} was the number of restaurants specializing in barbecue; T_{total} was the total number of restaurants. The numbers of restaurants were calculated by using POI data. OP was the percentage of meals eaten out (data from the National Institute for Nutrition and Health, Chinese Center for Disease Control and Prevention). In this study, we assumed that barbecue was a kind of eating out.

Emission from cooking was the sum of the emissions from residential cooking and from the catering industry. They were calculated by formulas (15) and (16):

$$E_{RC} = \sum \left(POP_{urban,k} \times MC_{urban,k} + POP_{non-urban,k} \times MC_{non-urban,k} \right) \times \left(1 - \frac{T_{BBQ,k}}{T_{total,k}} \right) \times (1 - OP) \times EF \times RE_{RC,k} \quad (15)$$

$$E_{CI} = \sum \left(POP_{urban,k} \times MC_{urban,k} + POP_{non-urban,k} \times MC_{non-urban,k} \right) \times \left(1 - \frac{T_{BBQ,k}}{T_{total,k}} \right) \times OP \times EF \times RE_{CI,k} \quad (16)$$

where E_{RC} was the emission from the residential cooking and E_{CI} was the emission from the catering industry; RE was the removal efficiency. The removal efficiency of the catering industry (RE_{CI}) was from the national standard (GB 18483-2001). The removal efficiency of residential cooking activity (RE_{RC}) was calculated using the popularizing rate of the range hood (data from China Statistical Yearbook) and removal efficiency of the range hoods (GB/T 17713-2011).

A Monte Carlo simulation was employed to analyze the uncertainties of the emission inventory (Wu et al., 2018). The simulation was executed 10000 times. The uncertainties of activity data were set as 0.2 or 0.5 (**Table S1**), and the uncertainties of EFs came from the actual measurements. The result of uncertainty was shown in **Table S2**.

2.5 Spatial-temporal distribution of emissions

As above-mentioned, the emissions from rural and urban FMS activity might differ greatly. During the process of spatial allocation, this difference must be emphasized. We were tempted to use GIS data for the classification of land use to divide urban and non-urban regions (Gong et al., 2019, 2020). Based on this

method, we got the data on the population distribution (data from www.worldpop.org, 10.5258/SOTON/WP00674) in urban and rural regions and constructed the emission map from FMS (**Text S4**).

The temporal allocation methods for FMS were also specific. For calculating the annual trends of emissions, statistical data including annual fireworks sales (data from the statistic of the Ministry of Emergency Management of the PRC) and annual restaurant sales (data from https://data.stats.gov.cn/) were used. The monthly trends of sacrificial incenses, joss paper, and fireworks burning were calculated with data from household investigations. We believed that the activities of these sources are mainly concentrated on five traditional Chinese festivals, including Chinese New Year's Eve (CNE), Chinese Spring Festival (CSF), Spring Lantern Festival (LF), Qingming Festival (QF), and Zhongyuan Festival (ZF) (**Text S5**). We calculated the percentage of incense, joss paper, and fireworks that burned during these festivals, and spread the excess to other days. The monthly trends of barbecue and cooking emissions were calculated by using the monthly restaurant sales in each province (data from https://data.stats.gov.cn/). It should be noted that the above methods were alternatives due to the lack of direct statistical data, and the methods can be improved in the future.
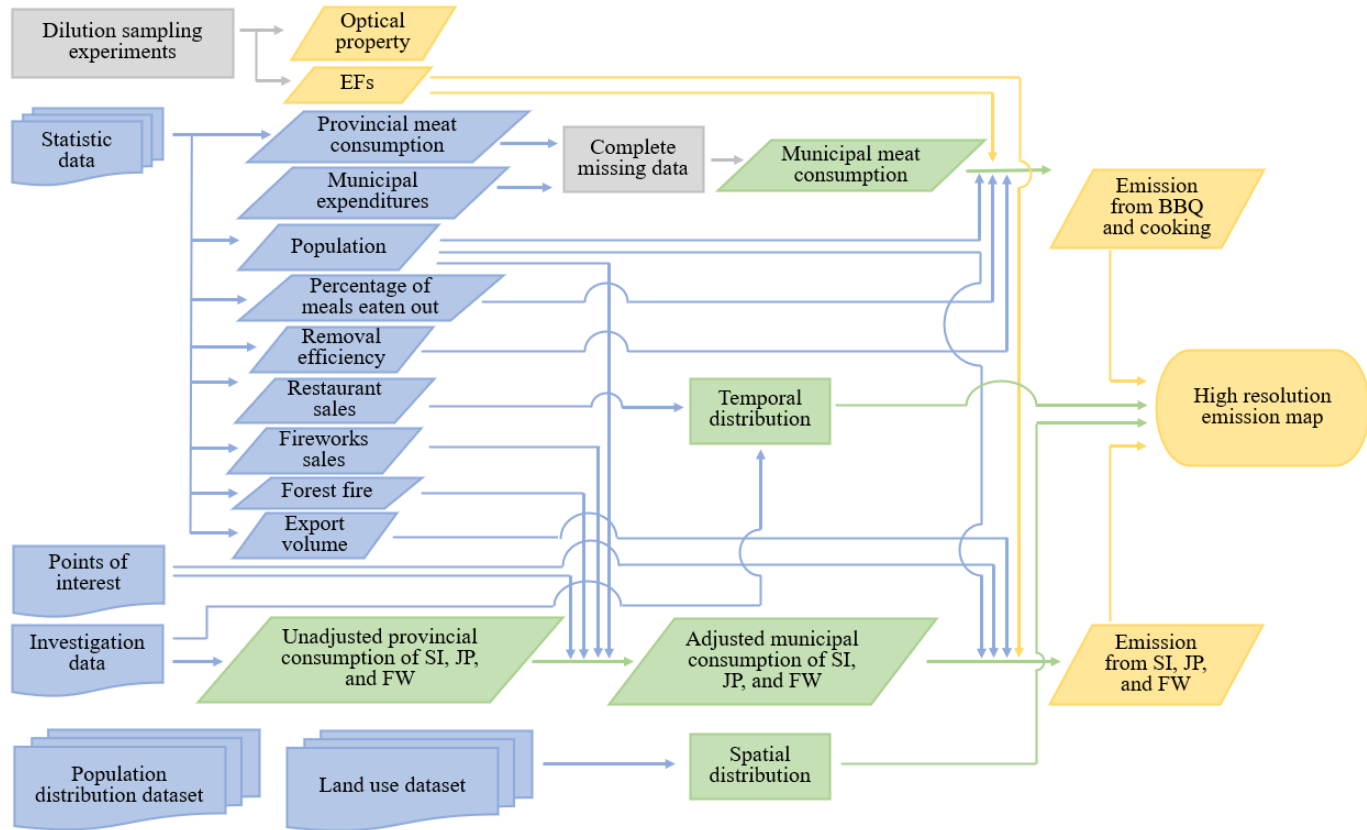
Figure 1 Methodological framework for establishing a high-resolution emission inventory for FMS.

3 Results and discussion

3.1 Emission characterization and light absorption properties

The EFs obtained from the 38 tests were shown in **Table 1**. The mean EF_{OC} of sacrificial incense, barbecue, joss paper, fireworks burning, and cooking were 32.6 ± 12.6 mg kg^{-1}, 33.2 ± 13.5 mg kg^{-1}, 41.9 ± 27.8 mg kg^{-1}, 51.9 ± 45.5 mg kg^{-1}, and 159 ± 34.0 mg kg^{-1}, respectively. While the EF_{EC} and EF_{BC} showed different tendencies. Barbecue exhibited higher EF_{EC} (5.13 ± 5.23 mg kg^{-1}) and EF_{BC} (69.6 ± 79.5 mg kg^{-1}) than those of sacrificial incense (EF_{EC}: 0.17 ± 0.07 mg kg^{-1}, EF_{BC}: 1.80 ± 0.92 mg kg^{-1}), joss paper (2.25 ± 2.47 mg kg^{-1}, 3.79 ± 2.23 mg kg^{-1}), cooking (0.005 ± 0.001 mg kg^{-1}, 1.54 ± 0.17 mg kg^{-1}), and fireworks burning (2.57 ± 5.37 mg kg^{-1}, 14.8 ± 17.3 mg kg^{-1}). Multiple factors, such as fuel properties (Chen et al., 2009; Shen et al., 2014; Cheng et al., 2019), combustion condition (Cheng et al., 2019), and stove properties (Shen et al., 2014; Chen et al., 2015), affected the emission of CA from combustion sources. Similarly, CA emissions from FMS were dominated by diverse factors. Results in previous studies were also applicable in this study. For example, the emissions from environmental or aromatic incense were lower (Lee and Wang, 2004; Lui et al., 2016), and cooking fatty pork generated higher emissions (Saito et al., 2014). In addition, the previous study showed higher EF_{OC} (0.779 g kg^{-1}) and EF_{EC} (0.339 g kg^{-1}) for sacrificial offerings (Zhang et al., 2019b). The huge differences in EFs were highly possible (Liu et al., 2015), and more detailed research is needed to expand the datasets of EFs for FMS in the future.

 Table 1 BC, OC, and EC emission factors for five missing sources (FMS) (mg kg^{-1}).

Sources	Materials[*]	BC	OC	EC
Sacrificial incense	RI	3.09±0.05	49.2±6.39	0.23±0.07
	EI	1.24±0.17	27.3±2.63	0.17±0.02
	HI	1.07±0.04	21.4±2.06	0.10±0.02
Joss paper	RP	6.27±1.59	35.5±5.59	0.97±0.10
	SP	1.65±0.41	14.6±1.64	0.64±0.50
	LP	3.45±1.18	75.5±19.2	5.12±2.28
Fireworks	FC	3.56±0.32	8.72±0.08	0.14±0.03
	FF	2.89±0.88	5.86±1.28	0.06±0.03
	HF	23.0±8.63	124±29.2	9.79±8.49
	HT	7.49±0.20	65.7±10.5	2.39±1.76
	SF	37.3±22.8	55.1±0.66	0.48±0.29
Barbecue	CK	1.66±0.30	21.5±1.11	0.15±0.02
	BF	37.2±24.4	28.6±8.85	3.78±2.28
	LB	48.5±17.7	32.2±6.35	4.21±0.58
	PK	191±59.5	50.5±12.2	12.4±4.93
Cooking	MT1	1.79±0.04	127	0.003
	MT2	1.54±0.01	124	0.004
	MT3	1.34±0.05	181	0.007
	MT4	1.48±0.07	203	0.007

274   *: Abbreviations of fuels: RI: red incense; EI: environmental incense; HI: high incense; RP: red-printed paper; SP:

275   small sacrificial paper; LP: large sacrificial paper; FC: firecrackers; FF: fountain fireworks; HF: handheld fireworks; HT:

276   handheld fountain; SF: spin fireworks; CK: chicken; BF: beef, LB: lamb; PK: pork; MT1: cooking of meat; MT2: cooking

277   of meat and pepper; MT3: cooking of meat and garlic; MT4: cooking of meat, pepper, and garlic. FMS: five missing sources.

278

279   To quantify the light absorption properties of emissions from FMS, AAEs (370-880 nm) were calculated

280   (**Figure 2**). The average AAEs of FMS were in the range of 1.26–3.15. The mean AAE of sacrificial incenses

281   (2.69 ± 0.36) was slightly higher compared to joss paper (2.22 ± 0.65), fireworks burning (2.10 ± 0.50),

282   barbecue (1.40 ± 0.14), and cooking (1.39 ± 0.08). The AAEs in 370-880 nm wavelength of woody fuel

283   burning (1.0–2.7) (Martinsson et al., 2015; Zhang et al., 2020a, 2021c), crop residues burning (1.5–3.25)

284   (Tian et al., 2019; Zhang et al., 2020c, 2021a), coal combustion (1.1–2.5) (Tian et al., 2019; Zhang et al.,

285   2021a), and engines (1.1–2.4) (Corbin et al., 2018) were comparable to our results. AAE > 1 indicates that

286   there existed BrC in aerosols (Saleh et al., 2013; Sun et al., 2017). Thus, it is necessary to investigate BrC

emission characteristics and the contribution of BrC to the total light absorption from various sources.
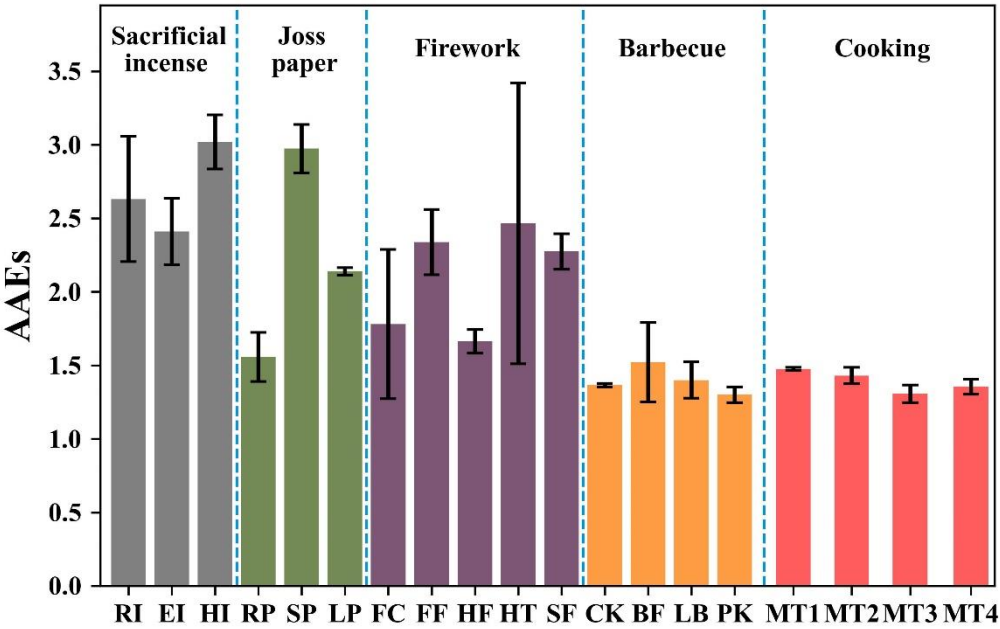
Figure 2 The absorption Ångström exponents (370-880 nm) of FMS.

We have calculated f_{BrC} to estimate the light absorption ability of BrC in aerosols (**Figure S4**). f_{BrC} showed a decreasing tendency toward the long wavelengths, which proved the understanding that the light absorption ability of BrC had stronger spectral dependence than that of BC (Sun et al., 2017). At 370 nm wavelength, f_{BrC} of sacrificial incense, joss paper, fireworks, barbecue, and cooking were 71.5 ± 5.32%, 58.4 ± 24.0%, 57.6 ± 9.36%, 28.7 ± 6.19%, and 29.2 ± 4.30%, respectively. f_{BrC} of cooking sources (22.9–37.4% with an average of 29.0) like barbecue and cooking seemed to be much lower than other combustion sources (34.8–82.7% with an average of 61.6%). f_{BrC} were 47% for coal combustion at 355 nm wavelength (Sun et al., 2017), and 68–85% for biomass burning at 370 nm (Tian et al., 2019). As some emission sources were neglected, the particulate absorption and warming effect contributed by BrC may be underestimated in former modeling works (Laskin et al., 2015).

Furthermore, the AEF of BrC and BC have been calculated, as shown in **Figure S5**. As the wavelength increased, the AEFs showed a decreasing trend. When λ=370 nm, AEF_{BrC} from fireworks burning was the

304   highest, as 2.65 ± 3.23 m^2 kg^{-1}, followed by barbecue (0.45 ± 0.49 m^2 kg^{-1}), joss paper (0.19 ± 0.21 m^2 kg^{-1}),

305   sacrificial incenses (0.15 ± 0.10 m^2 kg^{-1}), and cooking (0.012 ± 0.004 m^2 kg^{-1}). At 370 nm, the AEF$_{BrC}$ of

306   coal combustion and biomass burning have been reported as 14.3–46.6 m^2 kg^{-1} and 2.01–24 m^2 kg^{-1}

307   (Martinsson et al., 2015; Tian et al., 2019), which were 1–3 order of magnitude higher than those of FMS.

3.2 Characterization of activity

309   The total consumption of FMS was shown in **Figure 3**. In 2018, 16.5 kt, 919 kt, and 5139 kt of sacrificial

310   incenses, joss paper, and fireworks were consumed in China. 30996 kt, 2872 kt, 1920 kt, and 12057 kt of

311   pork, beef, lamb, and chicken were consumed. The total consumption amount of FMS was about 26.4% of

312   the residential coal consumption in China (Peng et al., 2019). The consumptions of sacrificial incense, joss

313   paper, and fireworks were highest in Shandong (394 kt) and Sichuan (470 kt). Apart from lamb, Guangdong

314   province has the largest consumption of three kinds of meats (6197 kt), and the province with the largest

315   consumption of lamb was Xinjiang (361 kt). The consumption of FMS can be a reflection of the local

316   customs. For example, lamb consumption in Xinjiang was the highest in China. The reason may be that

317   Xinjiang is the main producing area of lamb, one of the five pastoral areas in China, and Xinjiang's ethnic

318   structure makes mutton a dominant part of the daily diet (Xu et al., 2018).

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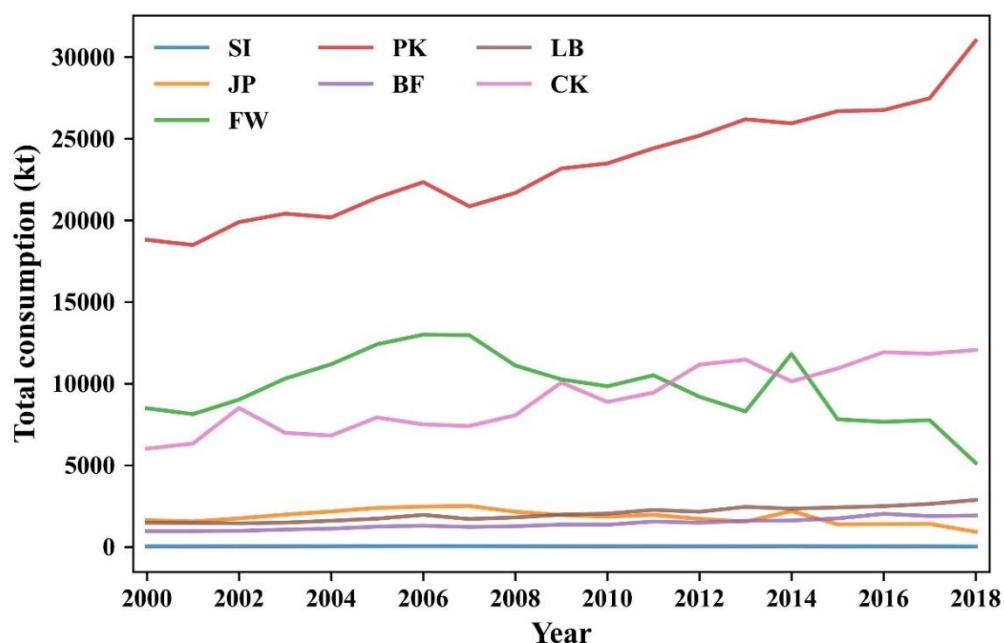
320   **Figure 3** The total consumption of FMS during 2000–2018 in China (Abbreviation: SI: sacrificial incense; JP: joss paper;

321   FW: fireworks; PK: pork; BF: beef; LB: lamb; CK: chicken).

322   The FMS consumptions in most cities were at low levels. The top 30 cities (about 8% of the total

323   number of cities) with the largest fireworks consumption contributed 41.8% of the national consumption

324   (**Figure S6**). These cities have higher population densities, and the control measures for fireworks were not

325   yet in place. Per capita fireworks consumption in 52% of the cities was less than 5 kg yr^{-1}, and the mass of

326   a common firecracker can exceeds 5 kg. As shown in **Figure S7**, the distribution pattern of per capita

327   consumption of sacrificial incenses and joss paper was similar to that of fireworks. The differences in meat

328   consumption between cities were relatively smaller. The top 30 cities with the largest pork consumption only

329   contributed 30.8% of the national consumption. The lowest per capita pork consumption was only 2.31 kg

330   yr^{-1} and the highest was 45.6 kg yr^{-1} (**Figure S7**). While 71.9% of the cities had per capita pork consumption

331   of 10–30 kg yr^{-1}. From 2000 to 2018, the consumption of four types of meat increased by 49.3%, and the

332   trends of increased meat consumption were similar to a previous study (Batis et al., 2014). While the

333   consumption of sacrificial incense, joss paper, and fireworks showed a trend of increasing first and then

334   decreasing.

335   **3.3 CA emission from FMS in China**

336   **3.3.1 Multi-year variation**

337   In 2000–2018, OC, EC, BC, and BrC absorption cross-section (ACS$_{BrC}$, in 370 nm wavelength)

338   emissions from FMS were 4268–4919 t, 22.6–43.9 t, 213–324 t, and 14.7–35.6 Gm^2, respectively (**Figure**

339   **4**). Severe air pollution over the past decade has led China to enact a series of policies to limit emissions

340   from various sources. Thus, the total CA emission in China presented approximately monotonically

341   decreased tendencies. During 2010–2017, the total OC and BC emissions in China decreased from 3.2 Tg

342   and 1.7 Tg to 2.1 Tg and 1.3 Tg, mostly contributed by residential sources (76.9–80.3% of OC, 41.8–51.5%

343   of BC) (http://meicmodel.org, Li et al., 2017; Zheng et al., 2018). The emission tendencies of FMS showed

344   different variation tendencies compared with these above sources. 82.7–92.3% of OC emissions came from

345   cooking (**Figure S8**). Due to the increased meat consumption (increased by 49%) and the popularizing rates

346   of range hoods (increased by 43.0%), the OC emissions from FMS increased by 14.4% before 2013 and then

347   decreased by 10.6%. The EC, BC, and ACS$_{BrC}$ emissions showed similar tendencies. From 2000 to 2006,

348   EC, BC, and ACS$_{BrC}$ emissions from FMS increased by 52.3%, 45.4%, and 51.2%, respectively. Then they

349   decreased by 48.7%, 27.8%, and 58.4% in 2006–2018. Fireworks burning was one of the main contributors

350   to CA emissions from FMS, which contributed 58.6–76.0%, 33.7–61.9%, and 88.5–96.6% of the EC, BC,

351 and ACS$_{BrC}$ emissions. The consumption of fireworks showed a trend of inverted U shape. It dominated the

352 tendencies of EC, BC, and ACS$_{BrC}$ emissions from FMS. Moreover, there was a surge in emissions due to

353 the high consumption of fireworks in 2014, which is consistent with the temporal distribution of PM$_{2.5}$ (Wei

354 et al., 2020, 2021) (**Text S6**). The surge in sales might have been caused by destocking after the Air Pollution

355 Prevention and Control Action Plan (APPCP) was implemented. From 2000 to 2006, the resident's income

356 raised by 76.5% due to the booming economy. The residents have more money to purchase fireworks. And

357 only another 12 cities have forbidden fireworks burning in 2000–2006. It can be the reason for the increase

358 in fireworks consumption amounts. From 2006 to 2018, although people's incomes continued to rise, while

359 the urbanization rate increased by 16.0% and additional 201 cities have forbidden the fireworks burning,

360 which lead to the decrease of fireworks consumption amount at this period.

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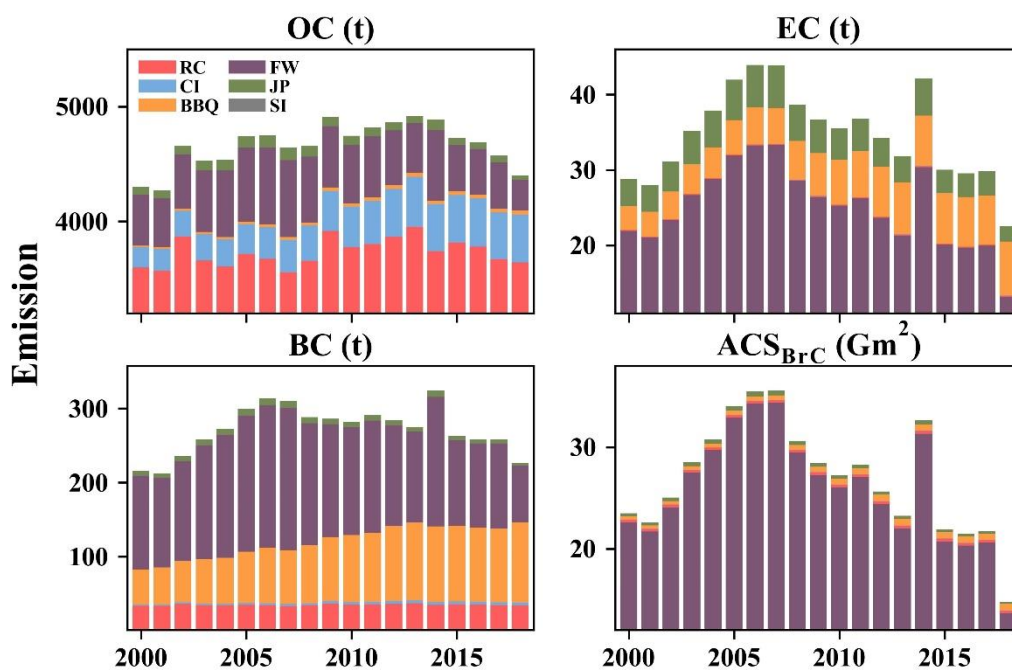
362 **Figure 4** Total CA emission from FMS in China from 2000 to 2018 (SI: sacrificial incense; JP: joss paper; FW: fireworks;

363 BBQ: barbecue; RC: residential cooking; CI: catering industry).

364 **3.3.2 Spatial variation**

365 There are seven geographical regions in China (**Figure S9**), of which East China was the largest CA

366 emission region. East China contributed 24.2–27.7%, 23.9–29.6%, 24.2–29.0%, and 23.5–29.9% of the OC,

367 EC, BC, and ACS$_{BrC}$ total emissions from FMS, respectively (**Figure 5**). The dense distribution of the

368 population (28.2–30.3% of the population) was responsible for the high emissions in East China. The OC,

EC, and BC emission from FMS in Southernwest China was second to that of East China. OC, EC, and BC emissions in Central China accounted for 21.5–27.2%, 18.0–21.3%, and 19.2–22.5% of their national total emissions. Eating habits in Southernwest China led to its high emission. Southernwest China has a higher density of barbecue restaurants (11.9% higher than the national average) and per capita meat consumption (33.4% higher than the national average), as well as a large population (14.9% of the national total). The ACS_{BrC} emission from Central China was second to that of East China, accounting for 14.3–21.6% of the national total. 90.9–96.4% of ACS_{BrC} emission was from fireworks burning. Hunan province in Central China is one of the main fireworks-producing regions. The density of fireworks stores in Hunan province was 2.3 times that of the national average. What's more, Central China was also one of the densely populated regions, accounting for 16.0–17.6% of the national population. Due to the heating needs in winter, the OC and BC emissions from other sources in North China contributed 14.8–17.2% and 17.6–21.1% of the national total (Li et al., 2017). However, the contributions of OC and BC emissions from FMS in North China were only 7.8–8.5% and 9.5–10.5%. The lower contribution was due to the lower per capita meat consumption (25.4% lower than the national average) and fewer restaurants (5.5% lower than the national average).

The emission distributions from different sources showed great differences, which came from the regional culture and economic diversity (**Figure S10** and **Figure S11**). High emission regions of sacrificial incense and joss paper overlapped with the areas with large numbers of temples and cradles of Chinese Buddhism (**Figure S3**, **Text S7**), where people in those areas may be more devout about sacrifice. The distributions of cooking emissions (both residential cooking emissions and catering industry emissions) and barbecue emissions were highly similar to the population distribution, especially in urban regions. This is consistent with previous studies. For example, the cooking-related organic aerosol (COA) concentrations at urban sites (6.46–6.97 μg m^{-3} in Beijing and 14.2 μg m^{-3} in Shijiazhuang) were much higher than that at the rural site (2.96–3.74 μg m^{-3} in Gucheng, a rural site near Beijing), and COA concentration was 0 at the background site (Sun et al., 2013, 2020; Wang et al., 2015b, 2020; Huang et al., 2019). Areas with higher economic consumption have more demands for repast styles and varieties, leading to more emissions. Emissions of fireworks showed an obvious difference in urban and rural regions. Emissions from urban regions were near zero, while emissions from suburbs and rural regions were much higher (more details can be found in **Section 3.3.3**, **Figure 8**).

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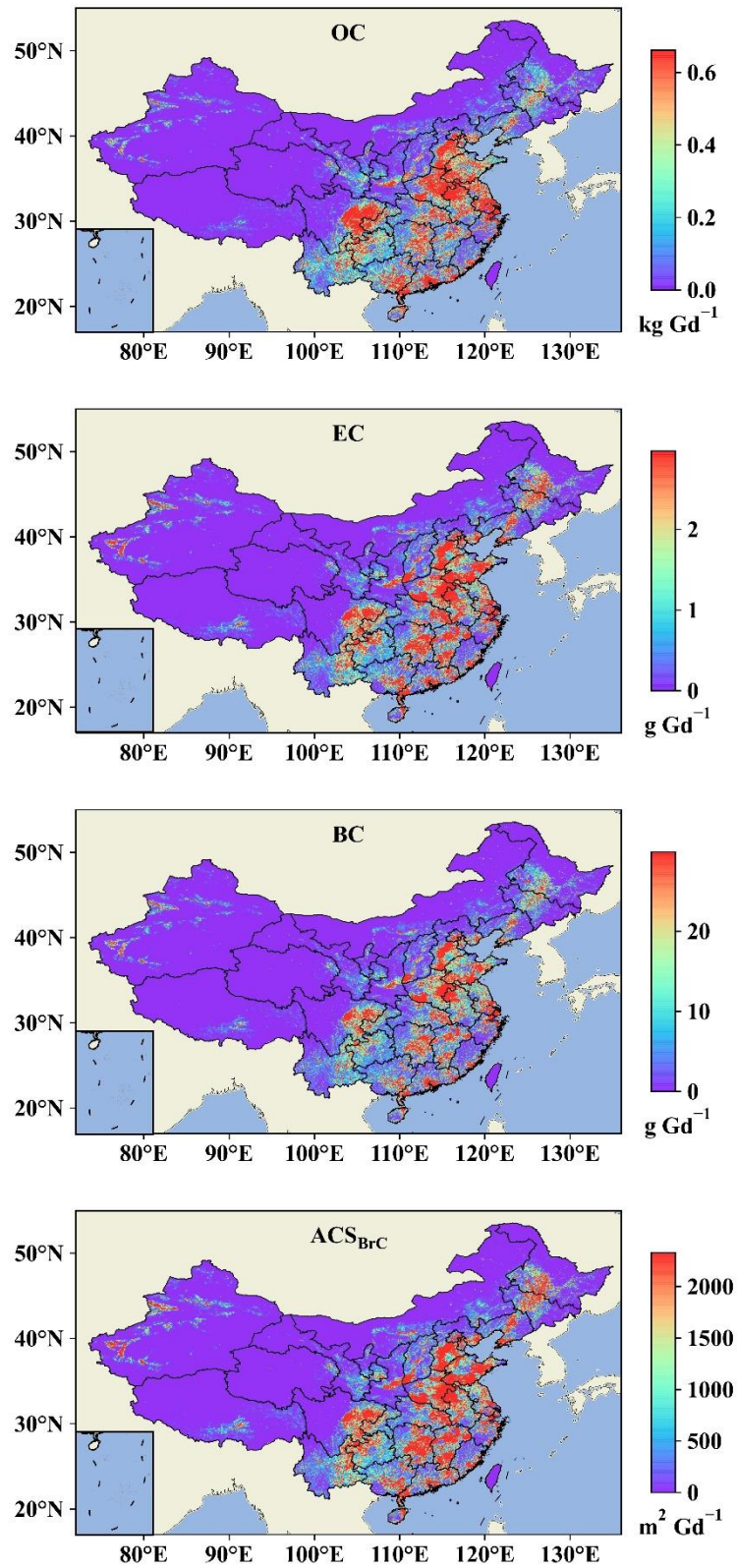
Figure 5 Spatial distribution of CA emission from FMS in China in 2018. The colorbar showed the emission in each grid.

399

3.3.3 Intense short-term and regionally concentrated emissions

As shown in **Figure S12** and **Figure S13**, CA emissions from residential sources in winter were extremely higher than in summer resulting from the heating demand (Wang et al., 2012; Huang et al., 2015; Li et al., 2017), while emissions from FMS showed a similar seasonal trend due to the fireworks burning. During the Chinese Spring Festival, fireworks burning results in massive pollutant emissions and severe air pollution (Kong et al., 2015; Yao et al., 2019; Ding et al., 2019; Lai & Brimblecombe, 2020). We have investigated the CA emissions from FMS in the month and at several related Chinese festivals (CNE, CSF, LF, QF, and ZF). As shown in **Figure 6**. The emissions were mostly concentrated in January and February (all CNE and CSF are in the same month in 2000–2018, after the calculation of multi-year data, the results for January and February in **Figure 6** seemed to be lower than those in **Figure S13**). 75.8% of fireworks were set off on CNE and CSF, and 20.4% were set off on LF (**Figure 7**). Thus, the ACS$_{BrC}$ emission on CNE was 1444 times the yearly average, and the OC, EC, and BC emissions were 10.9, 262, and 74.6 times the average, respectively. The highly short-term emission of fireworks led to a sharp increase in the concentration of air pollutants (Vecchi et al., 2008; Shi et al., 2011; Cao et al., 2018; Lai & Brimblecombe, 2020).
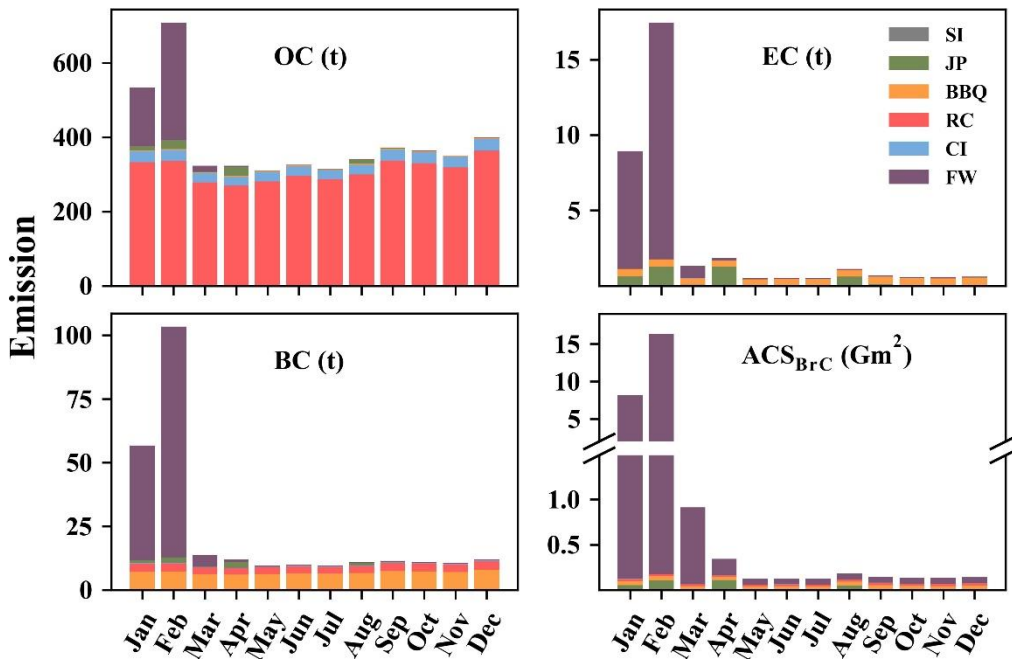
Figure 6 Averaged monthly CA emissions from FMS in China for the year of 2000–2018.

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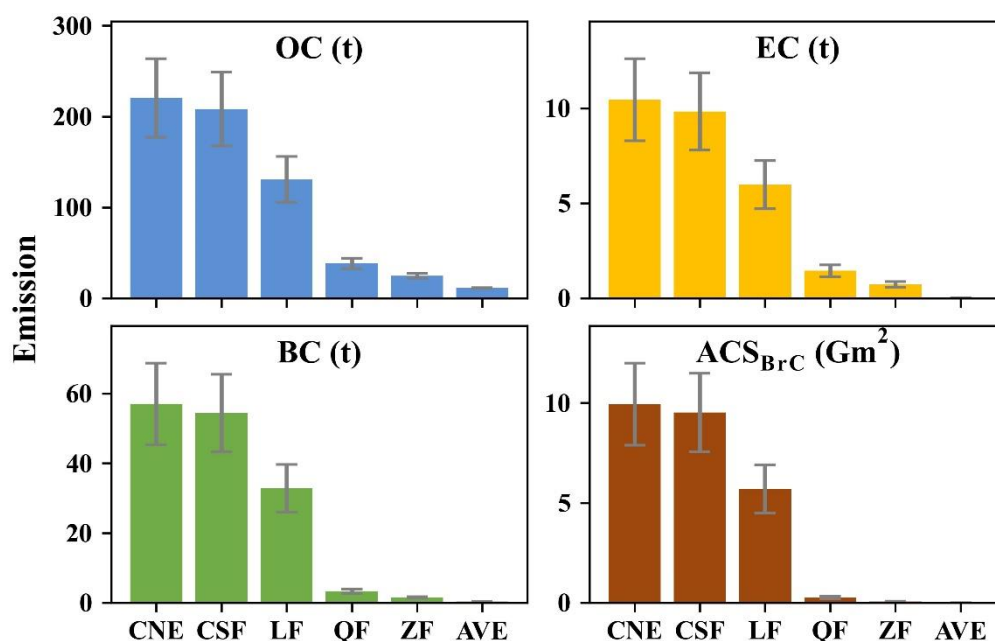
Figure 7 Average CA emissions on Chinese New Year's Eve (CNE), Chinese Spring Festival (CSF), Spring Lantern Festival (LF), Qingming Festival (QF), and Zhongyuan Festival (ZF) for the year of 2000–2018. The AVE showed the average daily emissions except for the festivals mentioned above.

For a short-term period, emissions from FMS also showed obvious spatial distribution. 83.2–93.1% of OC emissions came from barbecue and cooking. The higher population density and living quality led to higher OC emissions in urban regions. As shown in **Figure 8**, the OC emission intensities (average emission per grid) in the urban regions of Chengdu, Xi'an, Beijing, and Tianjin were 62.6, 63.1, 27.0, and 14.6 times of those for rural regions in 2018. This situation was common in China. China set up 13 prevention and control regions (3 key regions and 10 city clusters, 3-10R) in 2013 to improve air quality, and they are relatively developed regions (https://www.mee.gov.cn/gkml/hbb/bgg/201303/t20130305_248787.htm). The OC emission intensities in the urban regions of 3-10R were 3.9–50.5 times those in the surrounding rural regions. Thus, OC emissions from FMS were concentrated in urban regions overall. Since fireworks burning was concentrated in CNE or CSF and in rural regions, the feature that OC emission concentrated in urban regions would be attenuated at CNE. In contrast, ACS_{BrC} emissions tended to concentrate in rural regions, especially during the periods of CNE and CSF (**Figure 8**). Fireworks burning was the main contributor (>88.5%) to

436   ACS$_{BrC}$ emissions, and the fireworks burning was concentrated in CNE or CSF. The ACS$_{BrC}$ emission

437   intensities in rural regions of Chengdu, Xi'an, Beijing, and Tianjin were 18.8, 20.0, 107, and 150 times those

438   for urban regions at the 2018 CNE. Many cities have introduced policies to control firework burning, and

439   civilized sacrifice is encouraged. But these policies tend to be implemented effectively only in central urban

440   regions. Suburbs and surrounding rural regions, which are densely populated, are areas that policies do not

441   consider or be executed efficiently. The contribution of rural ACS$_{BrC}$ emissions in 3-10R was ~79.0% and

442   even as high as 96.7% at the 2018 CNE. However, the rural population in these regions only accounted for

443   14.1–41.9%. In fact, 63.0–79.5% of ACS$_{BrC}$ emissions at CNE came from the rural regions in China. During

444   the period of CNE and CSF, pollutants emitted from rural residents' activities were likely to be transmitted

445   to urban areas, leading to serious air pollution in urban regions (Yao et al., 2019; Pang et al., 2021).
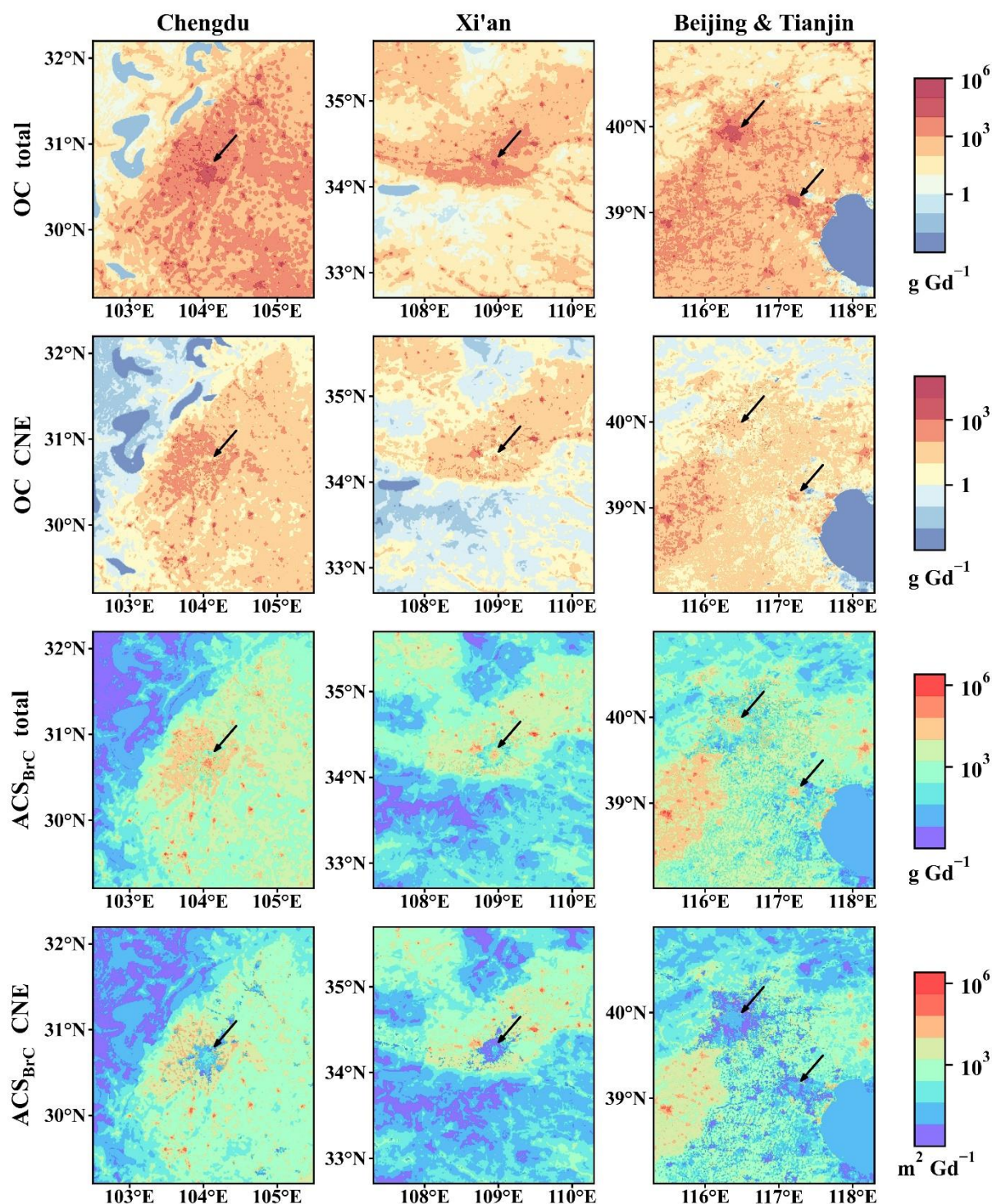
Figure 8 Differences in OC (six figures above) and ACS$_{BrC}$ (six figures below) emissions distributions in urban and rural regions in Chengdu (core city of Sichuan Basin), Xi'an (core city of Guanzhong Plain), and Beijing & Tianjin (Core cities of North China Plain). The first and second lines showed the total OC emissions in 2018, and in the day of CNE, respectively. The third and fourth lines showed the ACS$_{BrC}$ emissions in 2018, and in the day of CNE. The arrows pointed to the city centers. The colorbars showed the emission in each gird for OC and ACS$_{BrC}$.

3.3.4 Emissions impacted by economical development

Barbecue and cooking contributed a significant portion of OC emissions from FMS, which led to a distinctive feature of emissions from FMS. There was a certain correlation between OC emissions and local economic development. We have gathered disposable income per capita data from 2000 to 2018 for each city. The relationship between the disposable income and OC emission per capita has been assessed. As shown in **Figure 9**, like other emission sources, OC emissions from FMS and disposable income showed an inverted U-shape relationship ($r^2 = 0.73$, $p < 0.01$) (Environmental Kuznets Curves) (Wu et al., 2020; Zhong et al., 2020). This correlation existed for ACS_{BrC} emissions dominated by fireworks burning, while the correlation was weaker ($r^2 = 0.59$, $p < 0.01$) than that of OC emissions dominated by cooking sources. If we separated the emission-economical relationship in urban regions from rural regions, the results would be different. The relationships were linear in both urban and rural regions (**Figure 9**). However, the correlation was significantly negative ($r = -0.97$, $p < 0.01$) in urban regions compared to the positive one ($r = 0.94$, $p < 0.01$) in rural regions. As discussed above, cooking sources dominated the OC emissions from FMS in China. From 2000 to 2018, meat consumption per capita increased by 83.4%, and the OC emissions per capita have increased by 36.9% in rural regions. In urban regions, meat consumption increased by 22.0%, while OC emissions per capita decreased by 39.1%. The reason for this phenomenon was the higher popularizing rate of range hoods in urban regions, which was 5.2 times that of rural regions. What's more, the popularizing rate of range hoods in urban regions has also increased by 132% in urban regions. As a result, OC emissions that would have been raised were eliminated by a large number of range hoods in urban regions.

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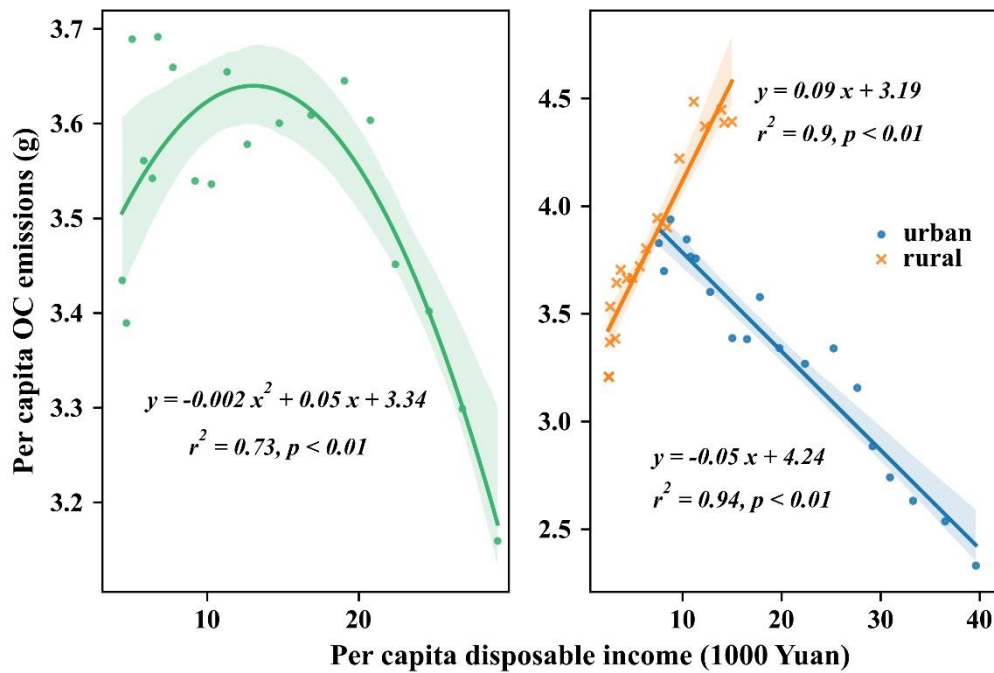
Figure 9 The relationship between per capita disposable income and OC emissions from FMS for the years of 2000–2018 in China. The left figure showed the invert-U shape relationship between the income and the national average emissions. The right figure showed the correlation between the income and emissions in urban regions and rural regions. The shaded areas represent 95% confidence intervals.

In contrast to the relatively developed 3-10R, there are some contiguous poor regions (CPR) in China, where located in the borderland or mountains (http://www.gov.cn/gzdt/2012-06/14/content_2161045.htm). The other regions (OR) excluding 3-10R and CPR, were at a moderate level of development in China. The OC emissions per capita in 3-10R, OR, and CPR, were 3.04–3.77 g, 3.49–4.00 g, and 3.54–4.11 g in 2000–2018. OC emissions per capita in 3-10R, OR, and CPR have all crossed the inflection point of the emission-economical correlation. Thus the relatively developed 3-10R have lower per capita emissions. It also verified the economic impact of OC emissions. However, 3-10R has 76.2–77.4% of the population, thus the emission intensities in 3-10R were still 3.1–3.4 times that of the national average.

3.3.5 The implication for modifying related air pollution control policies

To combat air pollution, China introduced its toughest air pollution control plan (APPCP) ever in 2013 (Zhang et al., 2019a). The implementation of the APPCP had led to significant improvements in China's air

quality. The control measures of FMS have also begun to be widely promoted (**Figure S14**). There were

76.3% and 66.5% of cities have introduced policies to restrict the emissions from the catering industry and

fireworks burning before 2018. The removal efficiencies of pollutants for small, medium, and large catering

industries were higher than 60%, 75%, and 85% (GB 18483-2001). Local governments have the right to

designate the areas where fireworks were forbidden, usually urban areas, along with hospitals, factories,

power plants, schools, and transportation hubs (http://www.gov.cn/zwgk/2006-01/25/content_170906.htm).

In addition, the government had proposed residents install range hoods to control the emissions from cooking

in APPCP, and the national popularizing rate of range hoods increased by 43.6% from 2000 to 2018. The

control policies and recommendations mentioned above have been implemented at various times in different

cities, and they all have positive significance for emission reduction. As a result, OC, EC, BC, and ACS_{BrC}

emissions from FMS have declined by 14.3–47.1%, 9.8–45.4%, 9.2–42.2%, and 10.4–48.2% in 2000–2018,

respectively (**Figure 10**).
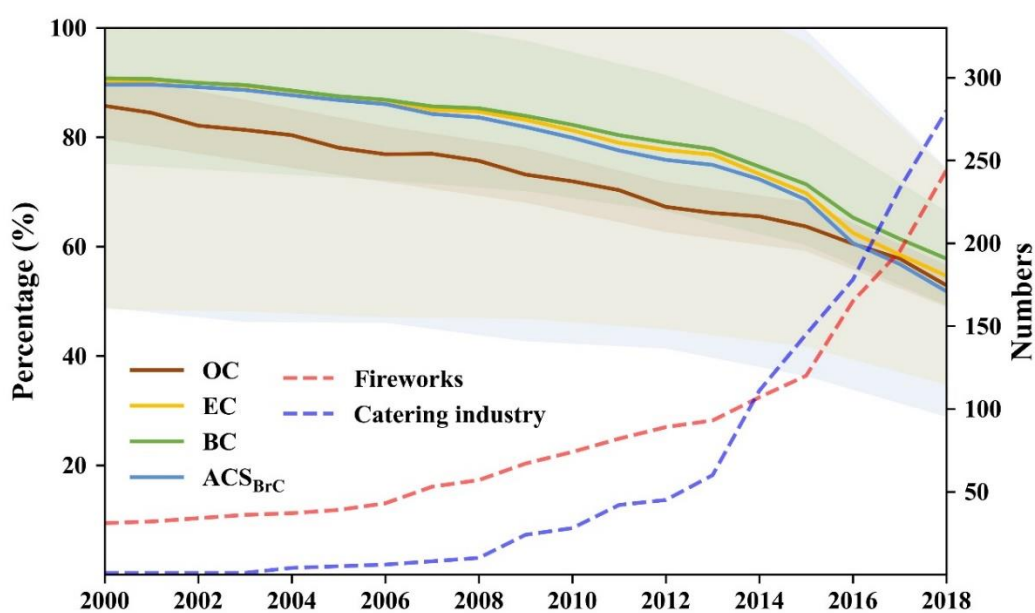
Figure 10 The impact of policies on the reductions of CA emissions. The solid lines (left y-axis) represented the actual CA

emissions compared to the emissions without the policy impact (100% on the left y-axis). The shaded part of the solid line

represents uncertainties. The dotted line (right y-axis) represented the number of cities that issued policies to control FMS

emissions.

506   If we assume that there was also a quadratic fitting relationship between rural per capita OC emissions

507   and income, then the rural per capita OC emissions would start to decline when rural per capita income

508   reaches 16.8 k Yuan. The control of CA emissions from FMS like cooking should start from the perspective

509   of increasing the income of rural residents. With enough income, residents will tend to a more

510   environmentally friendly and green lifestyle. The green lifestyle is embodied by the installation of a range

511   hood in this work. In 2017, the impervious surfaces of urban regions only accounted for 1.52% of the national

512   area (Gong et al., 2019), and rural regions are vast by contrast. Thus, the cost of controlling the activities of

513   fireworks burning, sacrificial incenses, and joss paper burning in rural regions will be much higher than in

514   urban regions. For these sources, policies and standards should be set to limit their emissions from the

515   burning processes. In addition, it is questionable whether the environmentally friendly fireworks currently

516   on the market have a lower impact on the environment (Fan et al., 2021). Thus, manufacturers should be

517   guided to develop environmentally friendly fireworks, joss paper, and sacrificial incense to reduce emissions.

518   **3.4 Comparison with other studies**

519   As an emission source with less attention, most of the relevant studies focused on the EFs of PM (Jetter

520   et al., 2002; Lee & Wang, 2004; Wang et al., 2015a; Kuo et al., 2016; Jilla & Kura, 2017; Amouei

521   Torkmahalleh et al., 2018; Wang et al., 2018b; Zhao et al., 2018; Lin et al., 2019), PAHs (Yang et al., 2005,

522   2013; Zhao et al., 2019), and VOCs (Cheng et al., 2016; Wang et al., 2018b). Several metallic elements

523   (Croteau et al., 2010; Shen et al., 2017) and organic matters (Xiang et al., 2017; Que et al., 2019) have also

524   been tested. Few studies have tested OC and EC EFs of FMS (See & Balasubramanian, 2011; Zhang et al.,

525   2019b; Lin et al., 2021). Fireworks burning was the least studied emission source, while fireworks can emit

526   large amounts of particles. EFs of PM_{10} from fireworks burning as 54–429 g kg^{-1} have been reported

527   (Camilleri & Vella, 2016; Keller & Schragen, 2021), which were much higher than the CA EFs in this study.

528   The EFs in the literatures have been shown in **Table S4**.

529   Several studies have calculated the emission amount of the catering industry (**Table S5**). For example,

530   Wang et al. (2018a) have calculated the VOCs emission (66245 t) from restaurants in China based on

531   samplings of 9 types of restaurants. Jin et al. (2021) have calculated the OC from the catering industry in

532   China by investigations in two cities in Shandong and Shanxi provinces. The results showed that OC

533   emissions from the catering industry were 26.8 Gg, which was 66.0 times that of our results. The EFs used

534   in Jin et al. (2021) were the generation rates of pollutants, which were 0.48 mg m^{-3} for OC in oil fumes.

535   Different EFs and calculation methods may be the main reason for the discrepancy. Emissions from cooking

536   have been reported as the main driver of OC in urban regions, as it contributed large portions of organic

537   aerosols in Shanghai (20–35%) and Beijing (10–19%) (Liu et al., 2021; Zhu et al., 2021b). The effects of

538   cooking emissions on the urban atmosphere should not be neglected when other sources like residential or

539   industry sources were efficiently controlled (Zhang et al., 2021c; Zhu et al., 2021a).

540   Previous research has calculated the total OC and BC emissions in China, such as the widely used MEIC

541   (OC: 2080–3190 Gg, BC: 1253–1728 Gg) and PKU emission inventory (OC: 2345–3587 Gg, BC: 1455–

542   1624 Gg) (Wang et al., 2014a; Huang et al., 2015; Li et al., 2017). Residential sources or residential &

543   commercial source contributed most of OC (80.3% in MEIC, and 71.4% in PKU) and BC (51.5% in MEIC,

544   and 51.0% in PKU) emissions (Peng et al., 2019). The OC and BC emissions from FMS accounted for only

545   1.5–2.2‰ and 0.16–0.20‰ of their national total emission. Thus, the OC and BC emissions from FMS were

546   generally meager. During key periods like the CNE, the contributions of FMS to the total OC and BC

547   emissions can rise to 2.3–3.5% and 1.1–1.6%. In key areas, the contribution rates would be relatively higher.

548   For instance, in CNE of 2014, FMS contributed 6.3% of OC emission in the Sichuan Basin and 2.9% of BC

549   emission in the Jiangxi-Hunan area. However, it should be noted that, the fireworks were always set off from

550   about 20:00 to 00:00 in CNE, so the intensive emission amounts could be considered at these times.

551   Therefore, the contribution of fireworks burning to CA in the atmosphere during CNE and CSF is still open

552   to debate.

553   It has caused widespread controversy that why the governments do not control the emissions from

554   industries and vehicles in CNE but emphasize the control of emissions from fireworks burning. The public

555   can not accept or believe that the emissions from fireworks can lead to serious air pollution, which could be

556   the key reason why they can not be completely eradicated in cities. From this study, the CA emissions are

557   limited compared with those from residential sources. An interesting question that atmospheric scientists

558   needed to be solved in the future is that if the fireworks burning were not controlled, how many air pollutants

559   from other main sources of cities should be controlled alternatively.

560   **Summary and conclusions**

561   The absence of anthropogenic sources in the existing inventory prevents people from drawing accurate

conclusions about the control of short-term pollution. To calculate the emissions from these sources which are difficult to estimate, we construct an emission inventory establishment framework including a series of equations and methods. We use multiple proxy data, such as the questionnaire, various statistics, and points of interest, to build a dataset of the activity of five missing sources (FMS, including cooking, fireworks burning, sacrificial incenses, and joss paper burning, and barbecue). The carbonaceous aerosols (CA) emission factors were tested in our lab using a self-designed sampling platform. The OC, EC, and BC EFs varied in 5.86–203 mg kg^{-1}, 0.003–12.4 mg kg^{-1}, and 1.07–191 mg kg^{-1}, respectively. BrC absorption EFs were in the range of 0.01–6.05 m^2 kg^{-1} (370 nm). From 2000 to 2018, the activity of FMS emitted 4268– 4919 t, 22.6–43.9 t, 213–324 t, and 14.7–35.6 Gm^2 of OC, EC, BC, and BrC absorption cross-section (ACS$_{BrC}$, in 370 nm wavelength). Emissions from FMS would concentrate on special festivals. For example, CA emission in Chinses New Year'eve was more than 10.8 times of its yearly average value. The distribution of pollutants also showed great differences between urban and rural regions due to the demographic, economic, and policy implications. There was a negative correlation ($r = -0.97$, $p < 0.01$) between individual emissions and disposable income in rural areas and a positive correlation ($r = 0.94$, $p < 0.01$) in urban areas. The policy implications led to a reduction of over 42.2% of CA emissions from FMS. This study complements the lack of emission inventory research of such missing sources and provides the prerequisite for modeling studies. Meanwhile, we suggest that raising residents' income can be a feasible solution when reducing FMS emissions sources that are difficult to control. The fireworks burning can be controlled from the manufacturer's side by guiding them to develop more environmentally friendly products. We also suppose that whether it is possible to control other emission sources for providing the environmental capacity for the emissions of fireworks burning.

Data availability

The dataset generated in this work is available at https://doi.org/10.6084/m9.figshare.19999991.v2 (Cheng et al., 2022). The POI data (points of barbecue restaurants, temples, common restaurants, and firework shops) were from the Open Platform of Amap (https://lbs.amap.com/). The China Forestry Statistical Yearbook (forest fires), China Light Industry Yearbook (fireworks export volume), and statistical yearbook of each province (urban and rural population, meat consumption, consumption expenditure, and disposable income) came from https://data.cnki.net. The percentage of meals eaten out came from

590 https://www.chinanutri.cn/. The China Statistical Yearbook (popularizing rate of the range hood) came from

591 https://data.cnki.net. The distribution of population was from www.worldpop.org. The land use data was

592 from http://data.ess.tsinghua.edu.cn/. The annual fireworks sales came from

593 https://www.mem.gov.cn/gk/tjsj/. The annual and monthly restaurant sales came from

594 https://data.stats.gov.cn/.

595 **Author contribution**

596 **Yi Cheng**: Experiments, Visualization, Writing - original draft. **Shaofei Kong**: Conceptualization,

597 Methodology, Supervision, Writing - review & editing. **Liquan Yao**: Experiments, Formal analysis. **Huang**

598 **Zheng**: Experiments, Formal analysis. **Jian Wu**: Formal analysis. **Qin Yan**: Experiments. **Shurui Zheng**:

599 Experiments. **Yao Hu**: Validation, **Zhenzhen Niu**: Experiments. **Yingying Yan**: Supervision. **Zhenxing**

600 **Shen**: Supervision. **Guofeng Shen**: Supervision. **Dantong Liu**: Supervision. **Shuxiao Wang**: Supervision.

601 **Shihua Qi**: Supervision.

602 **Competing interests**

603 The authors declare no competing financial interest.

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607 China (2016YFA0602002; 2017YFC0212602).

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