- 1 Multi-year emission of carbonaceous aerosols from cooking, fireworks burning,
- 2 sacrificial incenses, joss paper burning, and barbecue and the key driving forces in
- 3 China
- 4 Yi Cheng<sup>1,2</sup>, Shaofei Kong<sup>1,2\*</sup>, Liquan Yao<sup>1,2</sup>, Huang Zheng<sup>1,2</sup>, Jian Wu<sup>1,2</sup>, Qin Yan<sup>1,2</sup>, Shurui Zheng
- <sup>5</sup> <sup>1, 2</sup>, Yao Hu <sup>1, 2</sup>, Zhenzhen Niu <sup>1, 2</sup>, Yingying Yan <sup>1</sup>, Zhenxing Shen <sup>3</sup>, Guofeng Shen <sup>4</sup>, Dantong Liu <sup>5</sup>,
- 6 Shuxiao Wang<sup>6</sup>, and Shihua Qi<sup>2</sup>
- 7 <sup>1</sup>Department of Environment Science and Engineering, School of Environmental Studies, China University
- 8 of Geoscience, Wuhan 430074
- 9 <sup>12</sup> Department of Atmospheric Science, School of Environmental Studies, China University of Geoscience,
- 10 Wuhan 430074
- <sup>1</sup> <sup>2</sup> Department of Environment Science and Engineering, School of Environmental Studies, China University
- 12 of Geoscience, Wuhan 430074
- <sup>3</sup> Department of Environmental Science and Engineering, School of Energy and Power Engineering, Xi'an
- 14 Jiaotong University, Xi'an 710049
- <sup>4</sup> Laboratory for Earth Surface Process, College of Urban and Environmental Sciences, Peking University,
- 16 Beijing 100871
- <sup>5</sup> Department of Atmospheric Science, School of Earth Science, Zhejiang University, Hangzhou 310027
- <sup>6</sup> State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment,
- 19 Tsinghua University, Beijing 100084
- 20 Corresponding <u>toauthor</u>: Shaofei Kong (kongshaofei@cug.edu.cn)
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#### 22 Abstract

There has been controversy about the air pollutants emitted from sources closely related to people's 23 24 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 25 emission estimation of air pollutants from FMS, as the missing of both activity dataset and emission factors. 26 We attempted to combine the questionnaire data, various statistical data, and data of points of interest to 27 28 obtain a relatively complete set of activity data. The emission factors (EFs) of carbonaceous aerosols were 29 tested in our lab. Then, the emission inventories of carbonaceous aerosol with the a high spatial-temporal 30 resolution for FMS were established firstly, and the spatial variation trend and driving forces were discussed. 31 From 2000 to 2018, organic carbon (OC) emissions were in the range of increased from 4268 to 4919 t. 32 The OC emission from FMS was 1.485–2.218 ‰ of its total emission in China. The emissions of black 33 carbon, element carbon (EC), and brown carbon absorption cross-section (ACS<sub>BrC</sub>) emissions from FMS were in the ranges of 22.6–43.9 t, 213–324 t, and 14.7–35.6 Gm<sup>2</sup>, respectively. Their emissions tended to 34 concentrate in special periods and areas. The OC emission intensities in central urban areas were 3.85-50.5 35 times that of rural areas due to the high density of human activities. While the ACS<sub>BrC</sub> emissions in rural 36 regions accounted for 63.0–79.5% of the total emission result from uncontrolled fireworks burning. A mass 37 of fireworks burning led to extremely higher ACS<sub>BrC</sub> and EC emissions on Chinese New Year's eve, as 1444 38 39 and 262 times their corresponding yearly average values. Interestingly, sS ignificant (p < 0.01) correlations 40 between human incomes and pollutant emissions were found, but while they were positive (r = 0.94) and 41 negative (r = -0.94) for urban and rural regions, indicating the necessity of regulating human lifestyle and 42 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.v24 (Cheng et al., 2022).

Keywords: Carbonaceous aerosols; Sources related to human activities; Emission inventory; Spatialtemporal variation; Driving force

#### 50 1 Introduction

51 China has experienced a period of serious air pollution, which produces a great health impact on 52 residents (Zheng et al., 2018; Zhang et al., 2019, 2020b; Tong et al., 2020). Carbonaceous aerosols (CA), 53 emitted from incomplete burning, include organic carbon (OC) and black carbon (BC, or element carbon, 54 EC), and they have attracted wide attention due to their adverse impacts on air quality, human health and 55 climate (Venkataraman et al., 2005; Ramanathan & Carmichael, 2008; Bond et al., 2013). The optical 56 properties of CA (especially brown carbon, BrC) are complex and mutative, which is also one of the 57 important factors affecting the global radiation balance (Feng et al., 2013; Laskin et al., 2015).

58 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, 59 and fireworks burning. The estimation of air pollutants from these sources was missing in the previous 60 emission inventory, and they were defined as five missing sources (FMS) in this study. FMS can lead to a 61 62 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). 63 64 For example, fireworks burning could contribute 60.1 % of PM<sub>2.5</sub> 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 65 atmospheric PAHs in Chu-Shan, and 9.6% of PCDD/F in Taipei (Lin et al., 2008; Kong et al., 2015; Ho et 66 67 al., 2016). Recently, with the strengthened control of combustion-related sources, the important role of 68 cooking emissions onin affecting air quality was gradually visible in densely populated downtown areas. 69 Cooking organic aerosols contributed to 15–34% of total OC and 6–9% of total PM<sub>2.5</sub> in a downtown site in 70 Shanghai (Huang et al., 2022) and 31% of total organic aerosols in Beijing in winter (Hu et al., 2022). The 71 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., 72 73 2019; Yao et al., 2019; Hu et al., 2021; Huang et al., 2021). Till now, no studies can provide their 74 contributions quantitively on a large scale as the scare of emission inventory, which limited the identification 75 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 population and economy between urban and rural areas are increasing (Meng et al., 2019), and the efficiencies and necessity of air quality control policies for FMS in urban and rural areas need to be assessed. For instance, fireworks burning was generally banned in the central urban region, while suburbs and rural regions were affected less by the <u>same</u> policyies. The cooking smokes needed to be purified in city centers, while in suburban and rural areas, the policy may be not strictly executed or it is even not necessary. Such deviations in policy establishment and implementation could ultimately drive the differences in the distribution of air pollutant emissions, which have not been addressed yet.

The emission inventory is the base for the quantitative description of anthropogenic pollutant emissions 85 86 (Li et al., 2017). The combination of the chemical transport models and high-resolution emission inventory 87 was paramount for understanding anthropogenic perturbations' impacts on the atmosphere, and for assessing 88 corresponding air pollution control strategies (Janssens-Maenhout et al., 2019; McDuffie et al., 2020). The 89 lack of emission inventories limits the large-scale model simulation, the optimization of corresponding 90 control measures, and the settlement of related disputes. In our previous work, Wu et al. (2021) have 91 established an emission inventory of levoglucosan which has-included the emissions from FMS (Wu et al., 92 2021). There were no other emission inventories of FMS reported, to the best of our knowledge.

93 To sum up, this study aimed to develop a methodological framework for establishing an emission 94 inventory of FMS, including the methods of various activity data acquisition, emission factors monitoring, 95 uncertainty assessment, and spatial-temporal allocation. The activity data were obtained based on household investigation, statistic data, points of interest (POI), etc. The emission factors were monitored through a 96 97 unique emission monitoring test platform, especially for fireworks burning in our lab. Then a high spatial (4)  $km \times 1 km \sim 1 km$ ) and temporal resolution (1 day for special festivals, and 1 month for the rest) emission 98 99 inventory was first established. The multi-year spatiotemporal variation of CA emissions from these sources 100 was analyzed and compared with other types of sources. Optimization pollution control measures were 101 proposed for these types of sources. The study provides a methodology for establishing an emission inventory of air pollutants from sources closely related to human activities. Other air pollutants emissions 102 103 could also be estimated in the future. The emission inventory obtained here could also provide the basic 104 inputs for corresponding modeling works.

105

### 106 2 Methodology

## 107 **2.1 Combustion tests for emission factors**

108 The combustion tests for FMS were performed with two custom-made combustion chambers. One of 109 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 110 system (Dekati FPS-4000, Finland) was employed to dilute the smoke. The smokes were diluted about 16-111 30 times and aged for about 30 s in a residence chamber. The sampling system has been utilized in residential 112 fuel combustion experiments (Cheng et al., 2019; Yan et al., 2020; Zhang et al., 2021b; Wu et al., 2021). 113 114 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 115 paper burning (red-printed paper: RP; small sacrificial paper: SP; large sacrificial paper: LP), 10 trials of 116 fireworks burning (firecrackers: FC; fountain fireworks: FF; handheld fireworks: HF; handheld fountain: 117 118 HT; spin fireworks: SF), 8 trials of barbecue (chicken: CK; beef: BF, lamb: LB; pork: PK), and 8 trials of 119 the cooking (cooking of meat: MT1; cooking of meat and pepper: MT2; cooking of meat and garlic: MT3; 120 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 drawn into-detected with an online carbonaceous aerosol analyzer. This analyzer<u>It</u> 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.

#### 128 **2.2** Calculation of emission factors and optical properties

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

130 
$$EF_{ij} = \frac{v \times m_{ij} \times r}{v_0 \times M_i} \quad (1)$$

131 where *i* and *j* denoted pollutant and fuel; *EF* was the emission factor (mg kg<sup>-1</sup>); *v* was the flue 132 gas flow (L min<sup>-1</sup>);  $v_0$  was the sampling flow (L min<sup>-1</sup>); *m* was the mass of the pollutant detected by the 133 instruments (mg); *r* was the dilution ratio; *M* was the mass of the material used in each tail of 134 <u>experiments</u> the fuel consumption qulity (kg) (Cheng et al., 2019; Yan et al., 2020).

All filter-based optical measurements will be confronted with the underestimation caused by the loading effect (Drinovec et al., 2015). The loading compensation could be calculated based on dual-spot measurements. The detailed calculation process was referred to Drinovec et al. (2015). There was inferior dependence of BC particles on light absorption in different light wavelengths. The absorption Ångström exponent (AAE) was an exceptional parameter to describe this dependence, as shown in equations (2) and (3):

$$b_{abs} \sim \lambda^{-AAE} \tag{2}$$

142 
$$AAE = -\frac{\ln\left(\frac{b_{abs}(\lambda_1)}{b_{abs}(\lambda_2)}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)} \quad (3)$$

143 where  $\lambda$  was the wavelength;  $b_{abs}$  denoted the total light absorption coefficient (Tian et al., 2019), 144 which could be calculated by equation (4) (Zotter et al., 2017):

145 
$$BC = b_{abs}(\lambda) / MAC(\lambda) \quad (4)$$

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

147 As shown in equation (5), the  $b_{abs}$  of CA was aroused by BC and BrC. To calculate the  $b_{abs}(\lambda, BC)$ 148 at each wavelength, equation (6) was introduced. The  $AAE_{BC}$  was determined as a value of 1.0 according 149 to previous studies (Tian et al., 2019; Liakakou et al., 2020).  $f_{BrC}(\lambda)$  (equation (7) was utilized to estimate 150 the fraction of BrC light absorption ( $b_{abs}(\lambda, BrC)$ ) in total light absorption ( $b_{abs}(\lambda)$ ).

151 
$$b_{abs}(\lambda) = b_{abs}(\lambda, BC) + b_{abs}(\lambda, BrC)$$
(5)

152 
$$b_{abs}(\lambda, BC) = b_{abs}(880) \times \left(\frac{\lambda}{880}\right)$$
(6)

153 
$$f_{BrC}(\lambda) = 100\% \times b_{abs}(\lambda, BrC)/b_{abs}(\lambda) \quad (7)$$

Due to the complicated chemical properties of BrC, it was difficult to measure the accurate concentration of BrC in-the flue gases. Thus, pPrevious studies have developed a peculiar EF called absorption emission factor (AEF), as shown in equation (8) (Martinsson et al., 2015; Tian et al., 2019; Zhang et al., 2020c). Most studies modeled the direct radiative forcing of BrC with its mass concentration and mass absorption efficiency (MAE) as the input parameters. While the mass concentration and the total mass of BrC in the atmosphere were still unclear, and the MAE values in the range of 0.08–3.8 m<sup>2</sup> g<sup>-1</sup> were also variable (Park et al., 2010; Feng et al., 2013; Wang et al., 2014b; Zhang et al., 2020a). Thus, a<u>A</u>n inventory
established by AEF as the following equation could avoid the deviation raised by the mass concentration and
MAE of BrC (Tian et al., 2019).

163 
$$AEF_{ij} = \frac{\sum_{t_0}^{t_{sample}} (b_{abs_{ij}} \times r \times v)}{M_j} \quad (8)$$

## 164 **2.3 Acquisition of activity data**

Data on the activity of sources directly affected the uncertainties of emission inventory, and an accurate estimate of FMS consumption was a crucial prerequisite. Statistics on direct consumption of FMS were scarce. Thus,Then multiple activity data and proxy variables were adopted, including the statistical yearbooks of each province in China, datasets of POI (Text S21, Figure S3), and rural household investigation data in our group (Text S32).

170 The original consumptions of sacrificial incenses, joss paper and fireworks were from a household 171 investigation. We got the per capita consumption of sacrificial incenses, joss paper, and fireworks in each province. The data were adjusted to overcome the problem of insufficient sample size. In China, sacrificial 172 173 activities mean honoring ancestors, and they mainly take place in temples or graveyards. China is a mountainous country with rolling terrain. Most of the inhabitants of non-plain areas chose hills that might 174 cover vegetation as the site of graveyards. Most traditional graveyards would be placed in hills that might 175 176 be covered with vegetation. The data on the consumption of sacrificial incenses and joss paper will be revised 177 based on the number of temples (data from POI) and frequency of forest fires caused by sacrifices to the total forest fires (data from China Forestry Statistical Yearbook), as shown in equation (9): 178

179 
$$Cs_{adj,province} = Cs_{inv,province} \times \left(\frac{Nt_{province}/POP_{province}}{2 \times Nt_{nation}/POP_{nation}} + \frac{Fs_{province}/Ft_{province}}{2 \times Fs_{nation}/Ft_{nation}}\right)$$
(9)

where Cs was the consumption of sacrificial incenses or joss paper per capita; Nt was the number of temples; POP was the population, which came from the statistical yearbook of each province; Fs was the forest fires raised by sacrificial activities; Ft was the forest fires raised by all anthropogenic activities; adjrepresented the data after adjusted; *inv* represented the data from the household investigation; *province* represented the data of each province; *nation* represented the data of the entire nation.

Consumption of The fireworks consumption amounts will be revised based on the number of retail shops
 of fireworks (data from POI) and provincial fireworks export volume (statistical data), as shown in equation

187 (10):

188 
$$Cf_{adj,province} = Cf_{inv,province} \times \left(\frac{Ns_{province}/POP_{province}}{2 \times Ns_{nation}/POP_{nation}} + \frac{Ve_{province}/POP_{province}}{2 \times Ve_{nation}/POP_{nation}}\right)$$
(10)

189 where *Cf* was the consumption of fireworks per capita; *Ns* was the number of retail shops of 190 fireworks; *Ve* was the export volume of fireworks (data from China Light Industry Yearbook). In addition, 191 the consumptions (*C*) of sacrificial incenses, joss paper, and fireworks at the municipal level were calculated 192 by combining the POI data and the consumptions at the provincial level, as shown in equation (11).

193 
$$C_{adj,city} = C_{inv,province} \times \frac{N_{city}/POP_{city}}{N_{province}/POP_{province}}$$
(11)

194 where *N* represented the number of temples or firework shops.

The original meats consumption per capita came from the statistical yearbook of each province. WhileBut, 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. Using tThe logarithmic relationship between per capita consumption expenditure and provincial per capita meat consumption was adopted to complement municipal per capita meat consumption<sub>s</sub>- <u>a</u>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). Then, tThe 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.

## 207 **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. Thus, wWe 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):

214 
$$E = \sum (POP_{urban,k} \times C_{urban,k} \times FB_k + POP_{non-urban,k} \times C_{non-urban,k}) \times EF \quad (13)$$

where k denoted the different cities; *urban* and *non – urban* represented urban regions and nonurban 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.

## 221 Emissions from barbecue were calculated by formula (14):

222 
$$E = \sum \left( POP_{urban,k} \times MC_{urban,k} + POP_{non-urban,k} \times MC_{non-urban,k} \right) \times \frac{T_{BBQ,k}}{T_{total,k}} \times OP \times EF \quad (14)$$

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 the emission from the catering industry. And tThey were calculated by formulas (15) and (16):

$$E_{RC} = \sum \left( POP_{urban,k} \times MC_{urban,k} + POP_{non-urban,k} \times MC_{non-urban,k} \right) \times \left( 1 - \frac{T_{BBQ,k}}{T_{total,k}} \right) \times (1 - OP) \times EF \times RE_{RC,k}$$
(15)  
$$E_{CI} = \sum \left( POP_{urban,k} \times MC_{urban,k} + POP_{non-urban,k} \times MC_{non-urban,k} \right) \times \left( 1 - \frac{T_{BBQ,k}}{T_{total,k}} \right) \times OP \times EF \times RE_{CI,k}$$
(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<sub>cI</sub>*) was from the national standard (GB 18483-2001). The removal efficiency of residential cooking activity (*RE<sub>RC</sub>*) 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**.

#### 241 **2.5 Spatial-temporal distribution of emissions**

As above-mentioned, the emissions from rural and urban FMS activity might differ greatly. During the 242 243 process of spatial allocation, this difference must be emphasized. Thus, wWe were tempted to use GIS data 244 for the classification of land use to divide urban and non-urban regions (Gong et al., 2019, 2020). Based on 245 this method, we got the data on the population distribution of population (data from www.worldpop.org, 10.5258/SOTON/WP00674) in urban and rural regions and constructed the emission map from FMS (Text S43). 246 The temporal allocation methods for FMS were also specific. For calculating the annual trends of 247 emissions, statistical data including annual fireworks sales (data from the statistic of the Ministry of 248 249 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 250 251 from household investigations. We believed that the activities of these sources are mainly concentrated on 252 five traditional Chinese festivals, including Chinese New Year's Eve (CNE), Chinese Spring Festival (CSF), 253 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 254 255 the excess to other days. The monthly trends of barbecue and cooking emissions were calculated by using 256 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 <u>a high-resolution emission inventory</u> 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 EFoc of sacrificial incense, 265 barbecue, joss paper, fireworks burning, and cooking were  $32.6 \pm 12.6$  mg kg<sup>-1</sup>,  $33.2 \pm 13.5$  mg kg<sup>-1</sup>,  $41.9 \pm$ 266 27.8 mg kg<sup>-1</sup>, 51.9  $\pm$  45.5 mg kg<sup>-1</sup>, and 159  $\pm$  34.0 mg kg<sup>-1</sup>, respectively. While the EF<sub>EC</sub> and EF<sub>BC</sub> showed 267 different tendencies. Barbecue exhibited higher  $EF_{EC}$  (5.13 ± 5.23 mg kg<sup>-1</sup>) and  $EF_{BC}$  (69.6 ± 79.5 mg kg<sup>-1</sup>) 268 than those of sacrificial incense (EF<sub>EC</sub>:  $0.17 \pm 0.07$  mg kg<sup>-1</sup>, EF<sub>BC</sub>:  $1.80 \pm 0.92$  mg kg<sup>-1</sup>), joss paper (2.25 ± 269 2.47 mg kg<sup>-1</sup>,  $3.79 \pm 2.23$  mg kg<sup>-1</sup>), cooking (0.005 ± 0.001 mg kg<sup>-1</sup>,  $1.54 \pm 0.17$  mg kg<sup>-1</sup>), and fireworks 270 271 burning  $(2.57 \pm 5.37 \text{ mg kg}^{-1}, 14.8 \pm 17.3 \text{ mg kg}^{-1})$ . Multiple factors, such as fuel properties (Chen et al., 272 2009; Shen et al., 2014; Cheng et al., 2019), combustion condition (Cheng et al., 2019), and stove properties 273 (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 274 275 this study. For example, the emissions from environmental or aromatic incense were lower (Lee and Wang,

- 276 <u>2004; Lui et al., 2016), and cooking fatty pork generated higher emissions (Saito et al., 2014). In addition,</u>
- 277 the previous study showed higher  $EF_{OC}$  (0.779 g kg<sup>-1</sup>) and  $EF_{EC}$  (0.339 g kg<sup>-1</sup>) for sacrificial offerings
- 278 (Zhang et al., 2019b). The huge differences in EFs were highly possible (Liu et al., 2015), and more detailed
- 279 research is needed to expand the datasets of EFs for FMS in the future. The diversities can result from the
- 280 fuel properties and burning conditions.
- 281

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

Table 1 BC, OC, and EC emission factors for five missing sources (FMS) (mg kg<sup>-1</sup>).

\*: Abbreviations of fuels: RI: red incense; EI: environmental incense; HI: high incense; RP: red-printed paper; SP:
small sacrificial paper; LP: large sacrificial paper; FC: firecrackers; FF: fountain fireworks; HF: handheld fireworks; HT:
handheld fountain; SF: spin fireworks; CK: chicken; BF: beef, LB: lamb; PK: pork; MT1: cooking of meat; MT2: cooking
of meat and pepper; MT3: cooking of meat and garlic; MT4: cooking of meat, pepper, and garlic. FMS: five missing sources.

To quantify the light absorption properties of emissions from FMS, AAEs (370-880 nm) were calculated 288 289 (Figure 2). The average AAEs of FMS were in the range of 1.26–3.15. The mean AAE of sacrificial incenses  $(2.69 \pm 0.36)$  was slightly higher compared to joss paper  $(2.22 \pm 0.65)$ , fireworks burning  $(2.10 \pm 0.50)$ , 290 barbecue (1.40  $\pm$  0.14), and cooking (1.39  $\pm$  0.08). The AAEs in 370-880 nm wavelength of woody fuel 291 burning (1.0–2.7) (Martinsson et al., 2015; Zhang et al., 2020a, 2021c), crop residues burning (1.5–3.25) 292 293 (Tian et al., 2019; Zhang et al., 2020c, 2021a), coal combustion (1.1–2.5) (Tian et al., 2019; Zhang et al., 2021a), and engines (1.1-2.4) (Corbin et al., 2018) were comparable to our results. AAE > 1 indicates that 294 295 there existed was 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.

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299 Figure 2 The absorption Ångström exponents (370-880 nm) of FMS.

300

We have calculated  $f_{BrC}$  to estimate the light absorption ability of BrC in aerosols (Figure S4).  $f_{BrC}$ 301 302 showed a decreasing tendency toward the long wavelengths, which proved the understanding that the light 303 absorption ability of BrC had stronger spectral dependence than that of BC (Sun et al., 2017). At 370 nm 304 wavelength,  $f_{BrC}$  of sacrificial incense, joss paper, fireworks, barbecue, and the cooking were 71.5 ± 5.32%,  $58.4 \pm 24.0\%$ ,  $57.6 \pm 9.36\%$ ,  $28.7 \pm 6.19\%$ , and  $29.2 \pm 4.30\%$ , respectively. f<sub>BrC</sub> of cooking sources (22.9– 305 306 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 307 (Sun et al., 2017), and 68–85% for biomass burning at 370 nm (Tian et al., 2019). As some emission sources 308 309 were neglected, the particulate absorption and warming effect contributed by BrC may be underestimated in 310 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  $\lambda$ =370 nm, AEF<sub>BrC</sub> from fireworks burning was the

highest, as  $2.65 \pm 3.23 \text{ m}^2 \text{ kg}^{-1}$ , followed by barbecue  $(0.45 \pm 0.49 \text{ m}^2 \text{ kg}^{-1})$ , joss paper  $(0.19 \pm 0.21 \text{ m}^2 \text{ kg}^{-1})$ , sacrificial incenses  $(0.15 \pm 0.10 \text{ m}^2 \text{ kg}^{-1})$ , and cooking  $(0.012 \pm 0.004 \text{ m}^2 \text{ kg}^{-1})$ . At 370 nm, the AEF<sub>BrC</sub> of coal combustion and biomass burning have been reported as  $14.3-46.6 \text{ m}^2 \text{ kg}^{-1}$  and  $2.01-24 \text{ m}^2 \text{ kg}^{-1}$ (Martinsson et al., 2015; Tian et al., 2019), which were 1–3 order of magnitude higher than those of FMS.

#### 317 **3.2 Characterization of activity**

318 The total consumption of FMS has been was shown in Figure 3. In 2018, 16.5 kt, 919 kt, and 5139 kt of sacrificial incenses, joss paper, and fireworks were consumed in China. 30996 kt, 2872 kt, 1920 kt, and 319 12057 kt of pork, beef, lamb, and chicken were consumed. The total consumption amount of FMS was about 320 321 26.4% of the residential coal consumption in China (Peng et al., 2019). The consumptions of sacrificial incense, joss paper, and fireworks were highest in Shandong (394 kt) and Sichuan (470 kt). Apart from lamb, 322 323 Guangdong province has the largest consumption of three kinds of meats (6197 kt), and the province with 324 the largest consumption of lamb was Xinjiang (361 kt). The consumption of FMS can be a reflection of the 325 local customs. For example, lamb consumption in Xinjiang was the highest in China. The reason was-may 326 be that Xinjiang is the main producing area of lamb, one of the five pastoral areas in China, and Xinjiang's 327 ethnic structure makes mutton a dominant part of the daily diet (Xu et al., 2018).



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

<sup>330</sup> FW: fireworks; PK: pork; BF: beef; LB: lamb; CK: chicken).

332 The FMS consumptions in most cities were at low levels. The top 30 cities (about 8% of the total number of cities) with the largest fireworks consumption contributed 41.8% of the national consumption 333 (Figure S6). These cities have higher population densities, and the control measures for fireworks were not 334 yet in place. Per capita fireworks consumption in 52% of the cities was less than 5 kg yr<sup>-1</sup>, and the mass of 335 a common firecracker can exceed 5 kg. As shown in Figure S7, the distribution pattern of per capita 336 consumption of sacrificial incenses and joss paper was similar to that of fireworks. The differences in meat 337 consumption between cities were relatively smaller. The top 30 cities with the largest pork consumption only 338 339 contributed 30.8% of the national consumption. The lowest per capita pork consumption was only 2.31 kg  $yr^{-1}$  and the highest was 45.6 kg  $yr^{-1}$  (Figure S7). While 71.9% of the cities had per capita pork consumption 340 of 10–30 kg yr<sup>-1</sup>. From 2000 to 2018, the consumption of four types of meat increased by 49.3%, and the 341 trends of increased meat consumption were similar to a previous study (Batis et al., 2014). While the 342 consumption of sacrificial incense, joss paper, and fireworks showed a trend of increasing first and then 343 decreasing. 344

345 **3.3 CA emission from FMS in China** 

### 346 3.3.1 Multi-year variation

In 2000–2018, OC, EC, BC, and BrC absorption cross-section (ACS<sub>BrC</sub>, in 370 nm wavelength) 347 emissions from FMS were 4268–4919 t, 22.6–43.9 t, 213–324 t, and 14.7–35.6 Gm<sup>2</sup>, respectively (Figure 348 349 4). Severe air pollution over the past decade has led China to enact a series of policies to limit emissions from various sources. Thus, the total CA emission in China presented approximately monotonically 350 351 decreased tendencies. During 2010–2017, the total OC and BC emissions in China decreased from 3.2 Tg 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% 352 353 of BC) (http://meicmodel.org, Li et al., 2017; Zheng et al., 2018). The emission tendencies of FMS showed 354 different variation tendencies compared with these above sources. 82.7–92.3% of OC emissions came from cooking (Figure S8)., and the OC emissions from FMS increased first and then decreased. From 2000 to 355 2018, meat consumption per capita increased from 22.7 kg to 33.9 kg with the rapid development of the 356 economy and living standards in China. However, the popularizing rates of range hoods in China have 357 increased by 44.9% in the urban regions and 23.3% in rural regions, and it led to a decrease in OC emissions. 358 The combined effect of these two factors made the emission of FMS OC raised by 14.4% from 2000 to 2013 359







EC (t)

Figure 4 Total CA emission from FMS in China from 2000 to 2018 (Abbreviations: SI: sacrificial incense; JP: joss paper;
FW: fireworks; BBQ: barbecue; RC: residential cooking; CI: catering industry).

379

### 380 3.3.2 Spatial variation

381 There are seven geographical regions in China (Figure S9), of which East China was the largest CA 382 emission region, and. East China contributed 24.2–27.7%, 23.9–29.6%, 24.2–29.0%, and 23.5–29.9% of the 383 OC, EC, BC, and ACS<sub>BrC</sub> total emissions from FMS, respectively (Figure 5). The dense distribution of the 384 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 385 emissions in Central China accounted for 21.5-27.2%, 18.0-21.3%, and 19.2-22.5% of their national total 386 emissions. Eating habits in Southernwest China led to its high emission. Southernwest China has a higher 387 density of barbecue restaurants (11.9% higher than the national average) and per capita meat consumption 388 389 (33.4% higher than the national average), as well as a large population (14.9% of the national total). The ACS<sub>BrC</sub> emission from Central China was second to that of East China, accounting for 14.3–21.6% of the 390 391 national total. 90.9–96.4% of ACS<sub>BrC</sub> emission was from fireworks burning. The Hunan province in Central 392 China is one of the main fireworks-producing regions. The density of fireworks stores in Hunan province 393 was 2.329 times that of the national average. What's more, Central China was also one of the densely 394 populated regions, accounting for 16.0–17.6% of the national population. Due to the heating needs in winter, 395 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 396 397 China were only 7.78–8.52% and 9.53–10.5%. The lower contribution was due to the lower per capita meat 398 consumption (25.4% lower than the national average) and fewer restaurants (5.5% lower than the national 399 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**, <u>Text S7</u>), 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 406 urban sites (6.46–6.97  $\mu$ g m<sup>-3</sup> in Beijing and 14.2  $\mu$ g m<sup>-3</sup> in Shijiazhuang) were much higher than that at the 407 rural site (2.96–3.74 µg m<sup>-3</sup> in Gucheng, a rural site near Beijing), and COA concentration was 0 at the 408 background site (Sun et al., 2013, 2020; Wang et al., 2015b, 2020; Huang et al., 2019). Areas with higher 409 economic consumption have more demands for repast styles and varieties, leading to more emissions. 410 411 Emissions of fireworks showed an obvious difference in urban and rural regions. Emissions from urban 412 regions were near zero, while emissions from suburbs and rural regions were much higher (more details can 413 be found in Section 3.3.3, Figure 8).



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

# 418 **3.3.3 Intense short-term and regionally concentrated emissions**

419 As shown in Figure S12 and Figure S13, CA emissions from residential sources in winter were 420 extremely higher than in summer resulting from the heating demand (Wang et al., 2012; Huang et al., 2015; 421 Li et al., 2017), while emissions from FMS showed a similar seasonal trend due to the fireworks burning. 422 During the Chinese Spring Festival, the fireworks burning results in massive pollutant emissions and severe 423 air pollution (Kong et al., 2015; Yao et al., 2019; Ding et al., 2019; Lai & Brimblecombe, 2020). We have 424 investigated the CA emissions from FMS in the month and at several related Chinese festivals (CNE, CSF, LF, QF, and ZF)(Chinese New Year's Eve (CNE), Chinese Spring Festival (CSF), Spring Lantern Festival 425 (LF), Qingming Festival (QF), and Zhongyuan Festival (ZF) (Text S4)). As shown in Figure 6. The 426 427 emissions were mostly concentrated in January and February (all CNE and CSF are in the same month in 428 2000–2018, after the calculation of multi-year data, the results for January and February in Figure 6 seemed 429 to be lower than those in Figure S13). 75.8% of fireworks were set off on CNE and CSF, and 20.4% were 430 set off on LF (Figure 7). Thus, the ACS<sub>BrC</sub> emission on CNE was 1444 times the yearly average, and the 431 OC, EC, and BC emissions were also-10.9, 262, and 74.6 times the average, respectively. The highly short-432 term emission of fireworks led to a sharp increase in the concentration of air pollutants (Vecchi et al., 2008; 433 Shi et al., 2011; Cao et al., 2018; Lai & Brimblecombe, 2020).





436 Figure 6 Averaged monthly CA emissions from FMS in China for the year of 2000–2018.



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437

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.

444 In addition to the feature of For a short-term period, emissions from FMS also showeds obvious spatial 445 distributions. 83.2-93.1% of the OC emissions came from barbecue and cooking. The higher population 446 density and living quality led to higher OC emissions in urban regions. As shown in Figure 8, the OC 447 emission intensities (average emission per grid) in the urban regions of Chengdu, Xi'an, Beijing, and Tianjin 448 were 62.6, 63.1, 27.0, and 14.6 times of those for rural regions in 2018. This situation was common in China. 449 China set up 13 prevention and control regions (3 key regions and 10 city clusters, 3-10R) in 2013 to improve 450 quality, relatively developed regions air and they are 451 (https://www.mee.gov.cn/gkml/hbb/bgg/201303/t20130305 248787.htm). The OC emission intensities in 452 the urban regions of 3-10R were 3.859–50.5 times of those for in the surrounding rural regions. Thus, OC emissions from FMS were concentrated in urban regions overall. Since fireworks burning was concentrated 453

454 in CNE or CSF, due to the emission from fireworks burning and in rural regions, the feature that OC emission 455 concentrated in urban regions would be attenuated at CNE. In contrast, ACSBrc emissions tended to 456 concentrate in rural regions, especially during the periods of CNE and CSF (Figure 8). Fireworks burning was the main contributor (>88.5%) to ACSBrC emissions, and the fireworks burning was concentrated in CNE 457 or CSF. The ACS<sub>BrC</sub> emission intensities in rural regions of Chengdu, Xi'an, Beijing, and Tianjin were 18.8, 458 459 20.0, 107, and 150 times-of those for urban regions at the 2018 CNE. Many cities have introduced policies 460 to control firework burning, and civilized sacrifice is encouraged. But these policies tend to be implemented 461 effectively only in central urban regions. Urban sSuburbs and surrounding rural regions, that which are 462 densely populated, are areas that policies do not consider or be executed efficiently. The contribution of rural ACS<sub>BrC</sub> emissions in 3-10R was ~79.0% and even as high as 96.7% at the 2018 CNE. However, the rural 463 population in these regions only accounted for 14.1-41.9%. In fact, 63.0-79.5% of ACS<sub>BrC</sub> emissions at 464 CNE came from the rural regions in China. During the period of CNE and CSF, pollutants emitted from rural 465 residents' activities were likely to be transmitted to urban areas, leading to serious air pollution in urban 466 regions (Yao et al., 2019; Pang et al., 2021). 467



468

Figure 8 Differences in OC (sixthree figures above) and  $ACS_{BrC}$  (three 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 citiesy of North China Plain). The first and second lines showed the total OC emissions in the total year of 2018, and inon the day of CNE, respectively. The third and fourth lines showed the  $ACS_{BrC}$  emissions in the total year of 2018, and

 $\frac{1}{2}$   $\frac{1}$ 

## 475 **3.3.4 Emissions impacted by economical development**

Barbecue and cooking contributed a significant portion of OC emissions from FMS, which led to a 476 477 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 478 city. The relationship between the disposable income and OC emission per capita has been assessed. As 479 shown in Figure 9, like other emission sources, OC emissions from FMS and disposable income showed an 480 481 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<sub>BrC</sub> emissions dominated by fireworks burning, while the 482 correlation was weaker ( $r^2 = 0.59$ , p < 0.01) than that of OC emissions dominated by cooking sources. 483 484 However, ilf we separated the emission-economical relationship in urban regions from rural regions, the 485 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 486 487 (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 488 per capita have increased by 36.9% in rural regions. In urban regions, meat consumption increased by 22.0%, 489 while OC emissions per capita decreased by 39.1%. The reason for this phenomenon was the higher 490 491 popularizing rate of range hoods in urban regions, which was 5.2<sup>+</sup> 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 492 result, OC emissions that would have been raised were eliminated by a large number of range hoods in urban 493 494 regions.



496 497

Figure 9 The relationship between per capita disposable income and OC emissions from FMS for the years of 2000–2018 498 in China. The left figure showed the invert-U shape relationship between the income and the national average emissions. 499 The right figure showed the correlation between the income and emissions in-the urban regions and the rural regions. The 500 shaded areas represent 95% confidence intervals.

In contrast to the relatively developed 3-10R, there are some contiguous poor regions (CPR) in China, 502 where located in the borderland or mountains (http://www.gov.cn/gzdt/2012-06/14/content 2161045.htm). 503 The other regions (OR) excluding 3-10R and CPR, were at a moderate level of development in China. The 504 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– 505 506 2018. OC emissions per capita in 3-10R, OR, and CPR have all crossed the inflection point of the emission-507 economical correlation. Thus the relatively developed 3-10R have lower per capita emissions. It also verified 508 the economic impact of OC emissions. However, 3-10R has 76.2–77.4% of the population, thus the emission 509 intensities in 3-10R were still 3.41-3.439 times that of the national average.

#### 510 3.3.5 The implication for modifying related air pollution control policies

511 To combat air pollution, China introduced its toughest air pollution control plan (Air Pollution 512 Prevention and Control Action Plan, APPCP) ever in 2013 (Zhang et al., 2019a). The implementation of the

513 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 514 to restrict the emissions from the catering industry and fireworks burning before 2018. The removal 515 efficiencies of pollutants for small, medium, and large catering industries were higher than 60%, 75%, and 516 517 85% (GB 18483-2001). The Local governments have the right to designate the areas where fireworks were 518 forbidden, usually urban areas, along with hospitals, factories, power plants, schools, and transportation hubs 519 (http://www.gov.cn/zwgk/2006-01/25/content 170906.htm). In addition, the government had proposed 520 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 521 522 recommendations mentioned above have been implemented at various times in different cities, and they all 523 have positive significance for emission reduction. The timing of implementation varies in local governments, and the implementation of these policies has produced positive significance. As a result, OC, EC, BC, and 524 ACS<sub>BrC</sub> emissions from FMS have declined by 14.3-47.1%, 9.8-45.4%, 9.2-42.2%, and 10.4-48.2% in 525 526 2000–2018, respectively (Figure 10). OC, EC, BC, and ACSBIC emissions from have declined by 14.3–47.1%, 45.4%, 9.23 42.2%, and 10.4 48.2% in 2000 2018 (Figure 10). 527

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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.

534 If we assume that there was also a quadratic fitting relationship between rural per capita OC emissions 535 and income, then the rural per capita OC emissions would start to decline when rural per capita income reaches 16.8 k Yuan. The control of CA emissions from FMS like cooking should start from the perspective 536 of increasing the income of rural residents. With enough income, residents will tend to a more 537 538 environmentally friendly and green lifestyle. The green lifestyle is embodied by the installation of a range hood in this work. In 2017, the impervious surfaces of urban regions only accounted for 1.52% of the national 539 540 area (Gong et al., 2019), and rural regions are vast by contrast. Thus, the cost of controlling the activities of fireworks burning, sacrificial incenses, and joss paper burning in rural regions will be much higher than in 541 urban regions. For these sources, policies and standards should be set to limit their emissions from the 542 burning processes. In addition, it is questionable whether the environmentally friendly fireworks currently 543 544 on the market have a lower impact on the environment (Fan et al., 2021). Thus, manufacturers should be guided to develop environmentally friendly fireworks, joss paper, and sacrificial incense to reduce emissions. 545

## 546 **3.4 Comparison with other studies**

As an emission source with less attention, most of the relevant studies focused on the EFs of PM (Jetter 547 et al., 2002; Lee & Wang, 2004; Wang et al., 2015a; Kuo et al., 2016; Jilla & Kura, 2017; Amouei 548 Torkmahalleh et al., 2018; Wang et al., 2018b; Zhao et al., 2018; Lin et al., 2019), PAHs (Yang et al., 2005, 549 2013; Zhao et al., 2019), and VOCs (Cheng et al., 2016; Wang et al., 2018b). Several metallic elements 550 551 (Croteau et al., 2010; Shen et al., 2017) and organic matters (Xiang et al., 2017; Que et al., 2019) have also been tested. Few studies have tested OC and EC EFs of FMS (See & Balasubramanian, 2011; Zhang et al., 552 2019b; Lin et al., 2021). Fireworks burning was the least studied emission source, while fireworks can emit 553 large amounts of particles. EFs of PM<sub>10</sub> from fireworks burning as 54–429 g kg<sup>-1</sup> have been reported 554 555 (Camilleri & Vella, 2016; Keller & Schragen, 2021), which were much higher than the CA EFs in this study. 556 The EFs in the literatures have been shown in Table S43.

557 Several studies have calculated the emission amount of the catering industry (**Table S<u>5</u>4**). For example,

558 Wang et al. (2018a) have calculated the VOCs emission (66245 t) from restaurants in China based on samplings of 9 types of restaurants. Jin et al. (2021) have calculated the OC from the catering industry in 559 China by investigations in two cities in Shandong and Shanxi provinces. The results showed that OC 560 emissions from the catering industry were 26.8 Gg, which was 66.0 times that of our results. The EFs used 561 in Jin et al. (2021) were the generation rates of pollutants, which were 0.48 mg m<sup>-3</sup> for OC in oil fumes. 562 Different EFs and calculation methods may be the main reason for the discrepancy. Emissions from cooking 563 have been reported as the main driver of OC in urban regions, as it contributed large portions of organic 564 aerosols in Shanghai (20-35%) and Beijing (10-19%) (Liu et al., 2021; Zhu et al., 2021b). The effects of 565 566 cooking emissions on the urban atmosphere should not be neglected when other sources like residential or industry sources were efficiently controlled (Zhang et al., 2021c; Zhu et al., 2021a). 567

568 Previous research has calculated the total OC and BC emissions in China, such as the widely used MEIC 569 (OC: 2080-3190 Gg, BC: 1253-1728 Gg) and PKU emission inventory (OC: 2345-3587 Gg, BC: 1455-570 1624 Gg) (Wang et al., 2014a; Huang et al., 2015; Li et al., 2017). Residential sources or residential & commercial source contributed most of OC (80.3% in MEIC, and 71.4% in PKU) and BC (51.5% in MEIC, 571 572 and 51.0% in PKU) emissions (Peng et al., 2019). The OC and BC emissions from FMS accounted for only 573 1.548-2.218‰ and 0.16-0.20‰ of their national total emission. Thus, the OC and BC emissions from FMS 574 were generally meager. During key periods like the CNE, the contributions of FMS to the total OC and BC 575 emissions can rise to 2.325-3.546% and 1.105-1.658%. In key areas, the contribution rates would be 576 relatively higher. For instance, in CNE of 2014 CNE, FMS contributed 6.31% of OC emission in the Sichuan Basin and 2.94% of BC emission in the Jiangxi-Hunan area. However, it should be noted that, the fireworks 577 were always set off from about 20:00 to 00:00 in CNE, so the intensive emission amounts could be 578 579 considered at these times. Therefore, the contribution of fireworks burning to CA in the atmosphere during CNE and CSF is still open to debate. 580

It has caused widespread controversy that why the governments do not control the emissions from industries and vehicles in CNE but emphasize the control of emissions from fireworks burning. The public can not accept or believe that the emissions from fireworks can lead to serious air pollution, which <u>could</u> <u>beis</u> the key reason why they can not be completely eradicated in cities. From this study, the CA emissions are limited compared with those from residential sources. An interesting question that the atmospheric

- 586 scientists needed to be solved in the future is that if the fireworks burning were not controlled, how many air 587 pollutants from other main sources of cities should be controlled alternatively.
- 588 Summary and conclusions

The absence of anthropogenic sources in the existing inventory prevents people from drawing accurate 589 590 conclusions about the control of short-term pollution. To calculate the emissions from these sources which 591 are difficult to estimate, we construct an emission inventory establishment framework including a series of 592 equations and methods method. We use the multiple proxy data, such as the questionnaire, various statistics, 593 and points of interest, to build a dataset of the activity of five missing sources (FMS, including cooking, 594 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 595 EFs varied in 5.86–203 mg kg<sup>-1</sup>, 0.003–12.4 mg kg<sup>-1</sup>, and 1.07–191 mg kg<sup>-1</sup>, respectively. BrC absorption 596 EFs were in the range of  $0.01-6.05 \text{ m}^2 \text{ kg}^{-1}$  (370 nm). From 2000 to 2018, the activity of FMS emitted 4268– 597 4919 t, 22.6-43.9 t, 213-324 t, and 14.7-35.6 Gm<sup>2</sup> of OC, EC, BC, and BrC absorption cross-section 598 (ACS<sub>BrC</sub>, in 370 nm wavelength). Emissions from FMS would concentrate on special festivals. For example, 599 600 CA emission in Chinses New Year'eve was more than 10.8 times of itsthe yearly average value. The 601 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) 602 between individual emissions and disposable income in rural areas and a positive correlation (r = 0.94, p < 0.94) 603 604 0.01) in urban areas. The policy implications led to a reduction of over 42.2% of CA emissions from FMS. 605 This study complements the lack of emission inventory research of such missing sources and provides the 606 prerequisite for modeling studies. Meanwhile, we suggest that raising residents' income can be a feasible 607 solution when reducing FMS emissions sources that are difficult to control., Thesuch as fireworks burning, can be controlled from the manufacturer's side by guiding them to develop more environmentally friendly 608 products. We also suppose that whether it is possible to control other emission sources for providing the 609 environmental capacity for the emissions of fireworks burning. 610

## 611 Data availability

612 The dataset generated in this work is available at https://doi.org/10.6084/m9.figshare.19999991.v24
613 (Cheng et al., 2022).

614 The POI data (points of barbecue restaurants, temples, common restaurants, and firework shops) wereas from the Open Platform of Amap (https://lbs.amap.com/). The China Forestry Statistical Yearbook (forest 615 fires), China Light Industry Yearbook (fireworks export volume), and statistical yearbook of each province 616 (urban and rural population, meat consumption, consumption expenditure, and disposable income) came 617 618 from https://data.cnki.net. The percentage of meals eaten out came from https://www.chinanutri.cn/. The China Statistical Yearbook (popularizing rate of the range hood) came from https://data.cnki.net. The 619 distribution of population was from www.worldpop.org. The land use data was from 620 http://data.ess.tsinghua.edu.cn/. The annual fireworks sales came from https://www.mem.gov.cn/gk/tjsj/. The 621 622 annual and monthly restaurant sales came from https://data.stats.gov.cn/.

## 623 Author contribution

Yi Cheng: Experiments, Visualization, Writing - original draft. Shaofei Kong: Conceptualization,
Methodology, Supervision, Writing - review & editing. Liquan Yao: Experiments, Formal analysis. Huang
Zheng: Experiments, Formal analysis. Jian Wu: Formal analysis. Qin Yan: Experiments. Shurui Zheng:
Experiments. Yao Hu: Validation, Zhenzhen Niu: Experiments. Yingying Yan: Supervision. Zhenxing
Shen: Supervision. Guofeng Shen: Supervision. Dantong Liu: Supervision. Shuxiao Wang: Supervision.
Shihua Qi: Supervision.

#### 630 **Competing interests**

631 The authors declare no competing financial interest.

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### 636 <u>Changes in Supporting Information:</u>

- 637 <u>Three Texts (Text S1, Text S6, and Text S7), two figures (Figure S15, Figure S16), and one Table (Table S3) have been added. The content of Text S3 has been supplemented. Serial numbers of part of Texts and Table have been changed: Text S1 (original) → Text S2 (revised), Text S2 → Text S3, Text S3 → Text S4,
  640 <u>Text S4 → Text S5, Table S3 → Table S4, Table S4 → Table S5.</u>
  </u>
- 641

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