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Supplement of

Multiyear emissions of carbonaceous aerosols from cooking, fireworks, sacrificial incense, joss paper burning, and barbecue as well as their key driving forces in China

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Sect. S1: The online carbonaceous aerosol analyzer (OCAA) was developed by the Key Laboratory of Environmental Optics & Technology, Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences (Ding et al., 2014). Repeated testing of standard samples showed a relative standard deviation of 1.5% for the analysis of OCAA. When the sampling flow is 8 L min^{-1} and the sampling time is 30 min, the OCAA can detect the lowest concentration of particulate matter containing carbon as 0.23 μgC m^{-3}. The time resolution of the OCAA is 1 or 2 h. In addition, the OCAA can be set to a single sampling time as needed. The pure substance sucrose of OC was used to configure a series of sucrose solutions with different concentrations. The sucrose solutions with the same volume and different concentrations were used for OCAA analysis. The ratio of peak area between sucrose solution and internal standard was taken as the ordinate, and the carbon content of sucrose solution was taken as the abscissa to obtain the standard curve for instrument calibration.

Sect. S2: POI data came from the Open Platform of Amap (https://lbs.amap.com/). Amap is a leading provider of digital mapping, navigation, and location service in China. We have extracted the POI data by the keywords of fireworks, barbecue, restaurants, and temples, and eliminated the points with irrelevant keywords (Wu et al., 2021). The extraction was performed on Python 3.8 platform with the Requests library and Pandas library. We got 430,343 barbecue restaurants, 1,986,674 common restaurants, 70,766 temples, and 73,523 fireworks shops. The POI data were divided among 366 cities in China. Each point of POI contains latitude, longitude, name, category, and address. Unfortunately, they do not contain more detailed information on sale volume, turnover, etc.

Sect. S3: To understand the consumption of sacrificial incenses, joss paper, and fireworks, we have organized household investigations in China. We have investigated the population during the Chinese New Year, address (in the urban or rural region), the time when local fireworks were prohibited, the date or festivals of setting off fireworks, the date or festivals of burning joss paper and sacrificial incense, the quantities of fireworks, joss paper, and sacrificial incense that per capita consumed each year. We did not design the questionnaire to ask about the gender, or nationality of the respondents, but in the process of the questionnaire, we tried to ask the older person of each family. Our questionnaire was based on provinces (27 provinces were covered), and the distribution of the questionnaire is shown in **Figure S15**. Since the burning of fireworks is concentrated during the Chinese New Year, and the population migration during the Chinese New Year is huge in China. The registration or permanent population commonly mentioned in the questionnaire was not applicable in our work. Some families did not give accurate data on the consumption of fireworks, but the approximate volume of fireworks or the number of Xiang of firecrackers (firecrackers are made of thousands of small units connected in series, each unit can be called "Xiang" in Chinese; we thought the firecrackers were also a kind of fireworks). The consumption of this family was estimated according to the local quality of unit volume or Xiang. Similar situations can be observed for the sacrificial incense and joss paper. In addition to the festivals like Chinese New Year or Lantern festivals, some respondents also gave the dates of marriage, funeral, childbirth, and housewarming, that would burn fireworks. Given that these were only relatively occasions, thus these dates were considered as other days than the festivals mentioned in **Sect. S5** and **Figure 7**. In addition, in the survey, we found that some residents were not clear about the specific quality of fireworks set off at each festival, but would flexibly be changed according to the quantity of fireworks or firecrackers purchased in the year (such as the number of fireworks boxes and the number of whole rolls of firecrackers). Therefore, we considered the proportion of the occurrence number for each festival to the number for all dates in the questionnaire as the proportion of fireworks set off during the festival in the whole year. For example, if the word "Chinese Spring Festival" appeared 100 times in the questionnaire of a province, and the word "Chinese Spring Festival", "Chinese New Year's eve", "Lantern Festival", and other possible words, have appeared 250 times, then we consider that the fireworks set off during the Spring Festival in this province account for 100/250=40% of the whole year. Finally, 2461 valid questionnaires were collected.

Sect. S4: The emissions were spatially allocated by using two datasets: a population distribution data (www.worldpop.org) and a land-use data (Gong et al., 2019, 2020). The urban region and rural region were distinguished by the land-use data. According to the distribution of rural regions and urban regions, the population distribution data was divided into the rural and the urban population. But the result was contradictory to the census. The rural population was about four times the level of the urban population. Thus, data from the statistical yearbook (366 cities) was used to correct this bias.

Sect. S5: Chinese New Year's Eve (CNE, the last day of one lunar year) and Chinese Spring Festival (CSF, the first day of the next lunar year) are the Chinese Lunar New Year, and they are also the most important traditional festivals in China. The Spring Lantern Festival (LF) falls on the 15^{th} day of the first lunar month in China. CNE, CSF, and LF are three traditional Chinese festivals when fireworks were set off for celebration. The fireworks burning has also led to explosions in the concentration of air pollutants (Yang et al., 2014; Zhao et al., 2017; Wu et al., 2018; Yao et al., 2019). The Qingming Festival (QF) and Zhongyuan Festival (ZF) are traditional sacrificial festivals. In those days, people sweep tombs and worship their ancestors by burning joss paper and sacrificial incense. These festivals also prevail in Sinosphere. The burning of joss paper and sacrificial incense has attracted the attention of the scientific community, and it's also the source of air pollutants in certain periods (Chiang & Liao, 2006; Wei et al., 2018; Chen et al., 2019; Zhang et al., 2019).

Sect. S6: The surge in ACS_{BrC} emission in 2014 might be out of the ordinary. We attempted to use the $PM_{2.5}$ concentration dataset (Wei et al., 2020, 2021) to verify the accuracy of the inventory. The ACS_{BrC} emissions mainly came from fireworks burning (**Figure S8**), and most of the fireworks were burnt in rural regions during the Chinese New Year (**Section 3.3.3**). We conducted a correlation analysis between the FMS ACS_{BrC} emissions and $PM_{2.5}$ concentration in non-urban regions for the New Year's Eve. The results were shown in **Figure S16**. There was a positive correlation (r=0.59, p<0.01) between the ACS_{BrC} emissions and $PM_{2.5}$ concentration. The correlation (r=0.85, p<0.05) was even higher if we focus only on the period after 2013. Thus, the emission surge in 2014 was possible. However, more accurate verification is still needed to be carried out by chemical transport models in the future.

Sect. S7: As shown in **Table S3**, there existed a correlation between the activity data and the emissions. Population data was the basic data of the emissions for all FMS, thus the correlations between population and emissions of each city were positive ($r > 0.47$, $p < 0.01$). The other activity data for FMS were also correlated. For example, the total emissions of SI and JP in each city were positively related to the number of temples, which lead to their spatial distribution of emissions being coincidental with that of temples to a certain extent. This phenomenon also existed for other FMS.

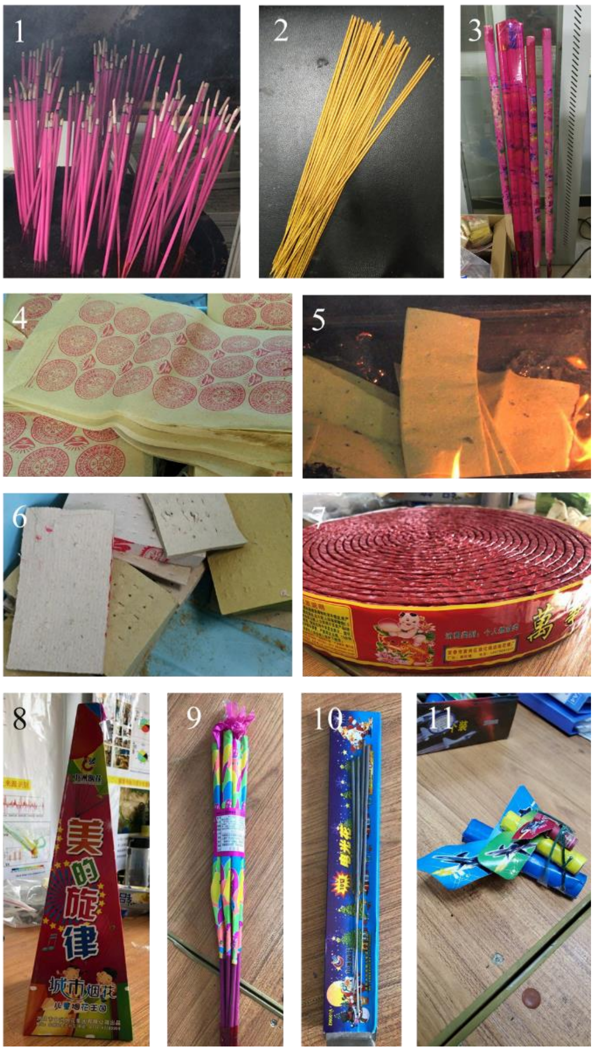

Figure S1 Pictures of materials used in this study (1: red incense (RI), 2: environmental incense (EI), 3: high incense (HI), 4: red print paper (RP), 5: large sacrificial paper (LP), 6: small sacrificial paper (SP), 7: firecrackers (FC), 8: fountain fireworks (FF), 9: handheld fireworks (HF), 10: handheld fountain (HT), 11: spin fireworks (SF). Pictures of meats used in barbecue and cooking experiments, including chicken (CK), beef (BF), lamb (LB), pork (PK), cooking of meat (MT1), cooking of meat and pepper (MT2), cooking of meat and garlic (MT3), cooking of meat, pepper, and garlic (MT4), were not present.

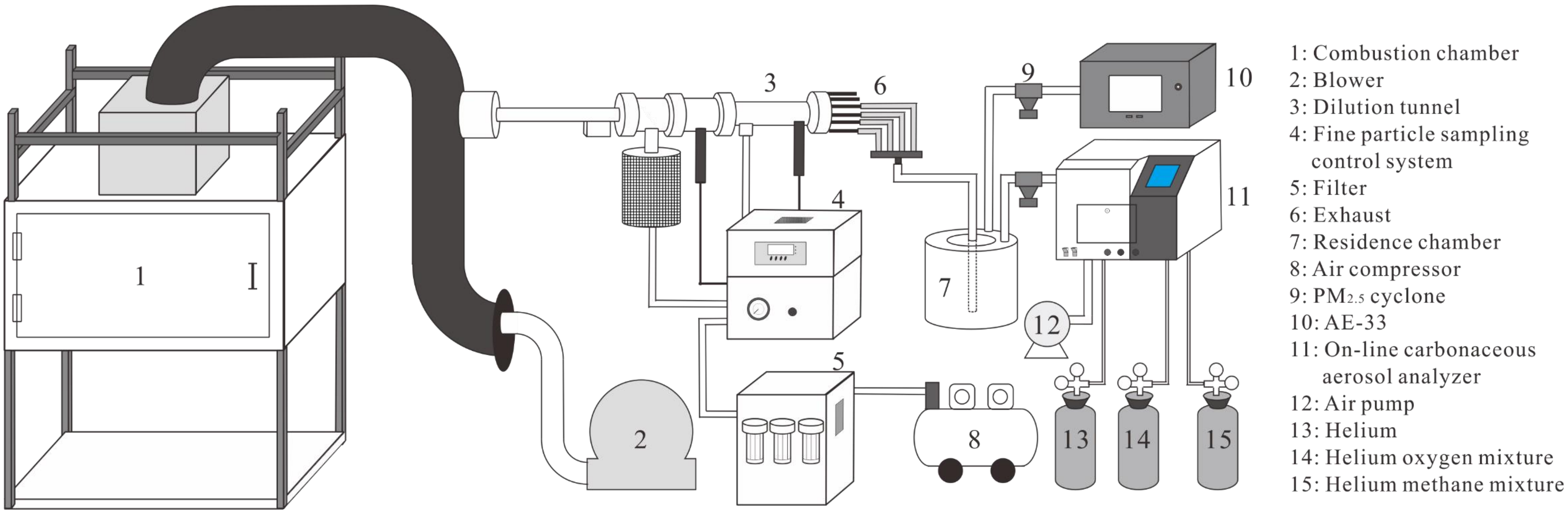

Figure S2 Structure of the combustion and dilution sampling system adopted in this study

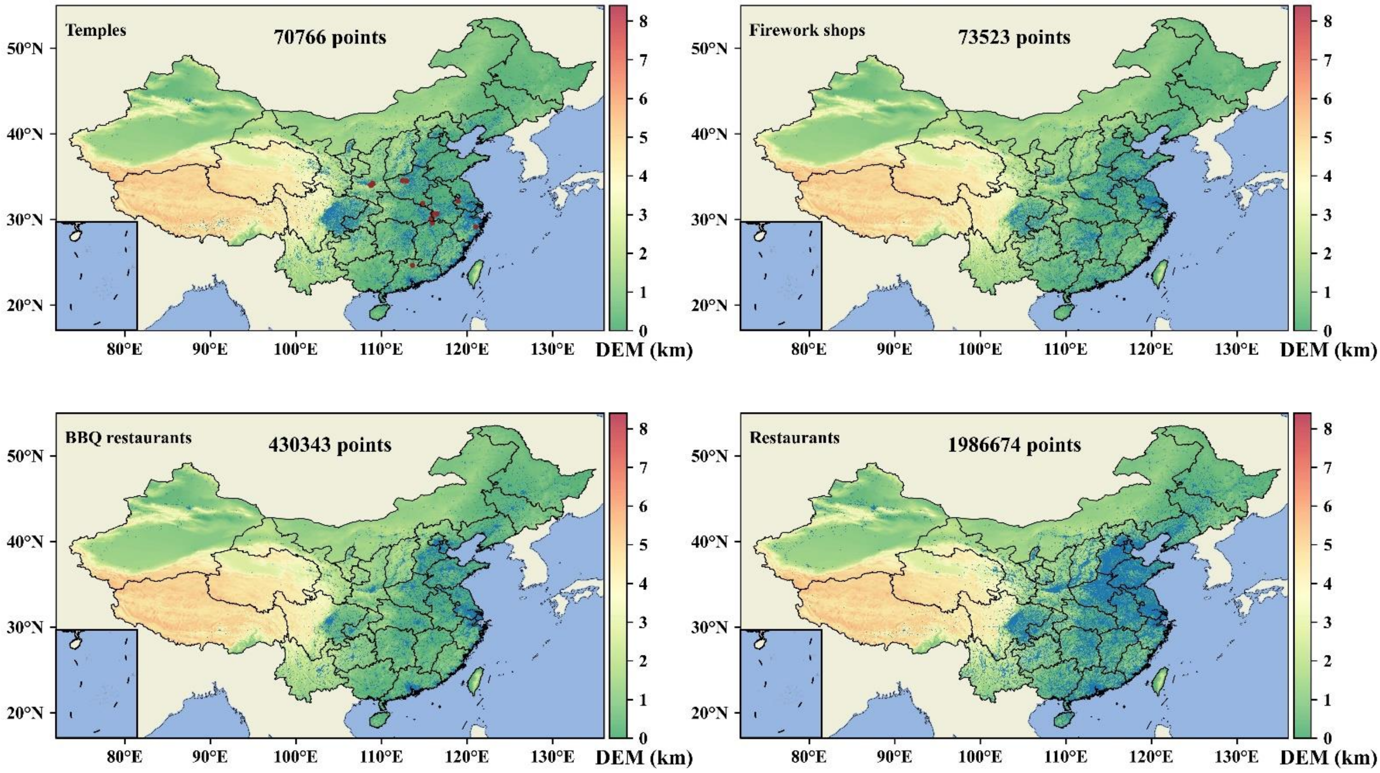

Figure S3 The distribution of temples, firework shops, barbecue restaurants, and common restaurants in China. The blue dots represent points of interest, and the red dots in the first picture are cradles of Chinese Buddhism.

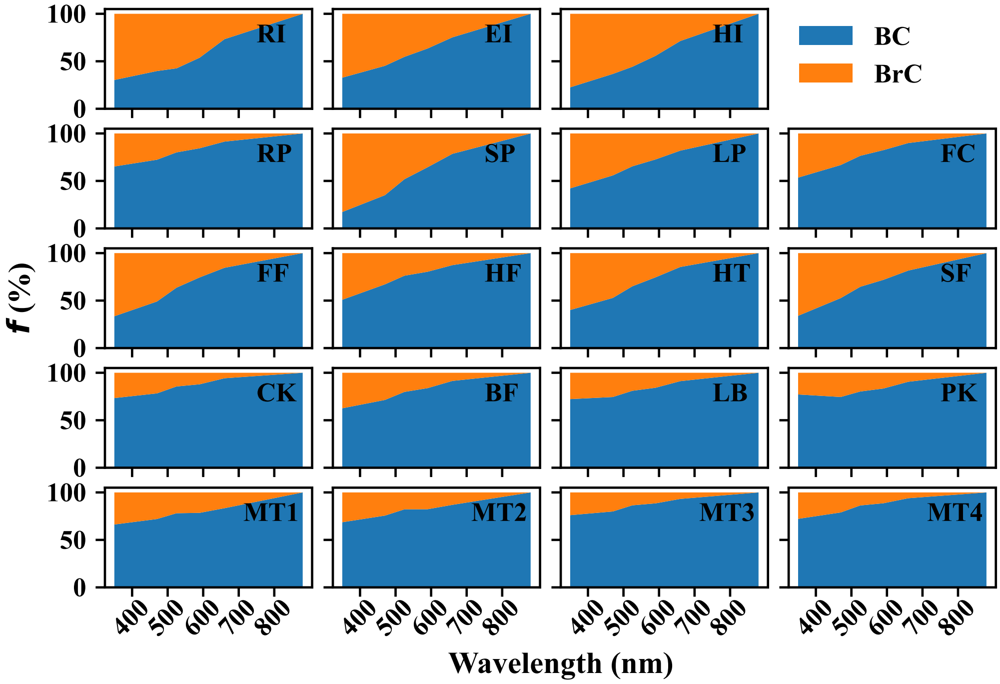

Figure S4 Contributions (%) of BC and BrC to the total light absorption in different wavelengths.

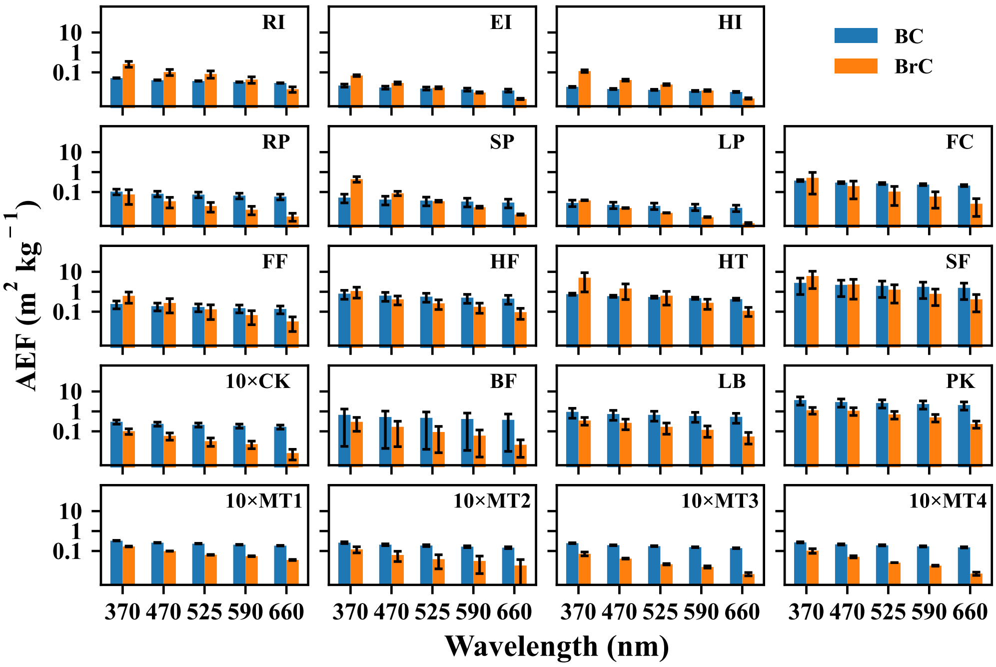

Figure S5 AEF_{BrC} and AEF_{BC} of FM in different wavelengths.

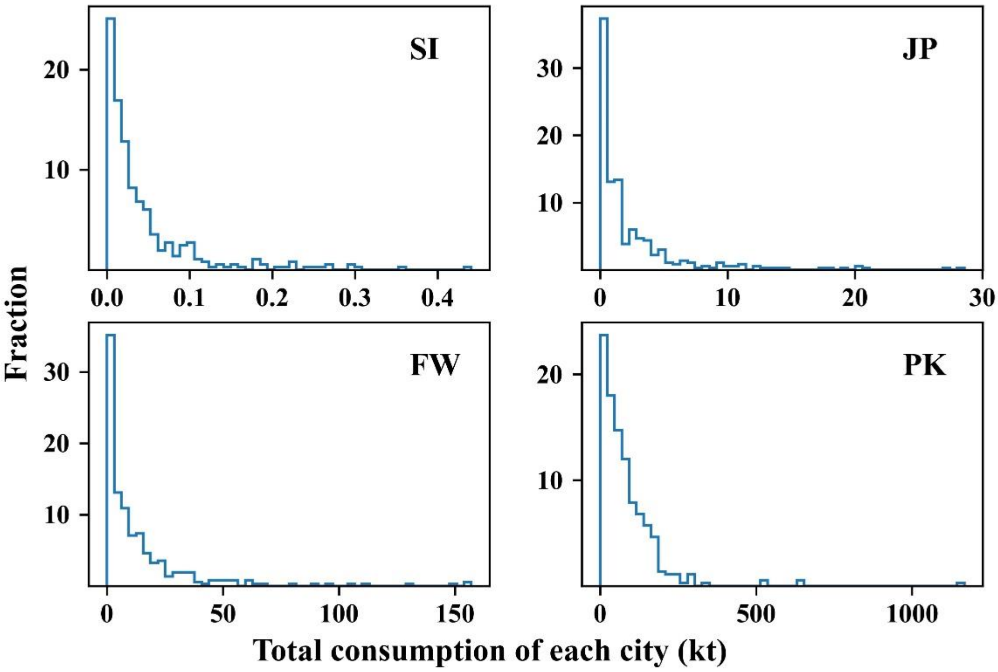

Figure S6 The distribution of total consumption amounts of sacrificial incense, joss paper, fireworks, and pork in 366 cities of China

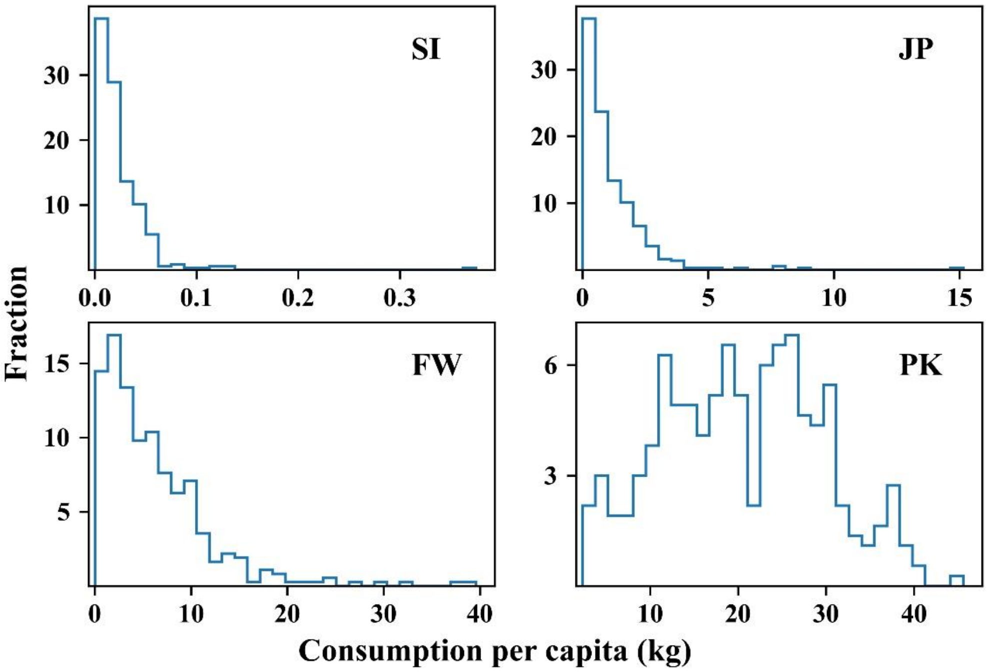

Figure S7 The distribution of per capita consumption amounts of sacrificial incense, joss paper, fireworks, and pork in 366 cities of China

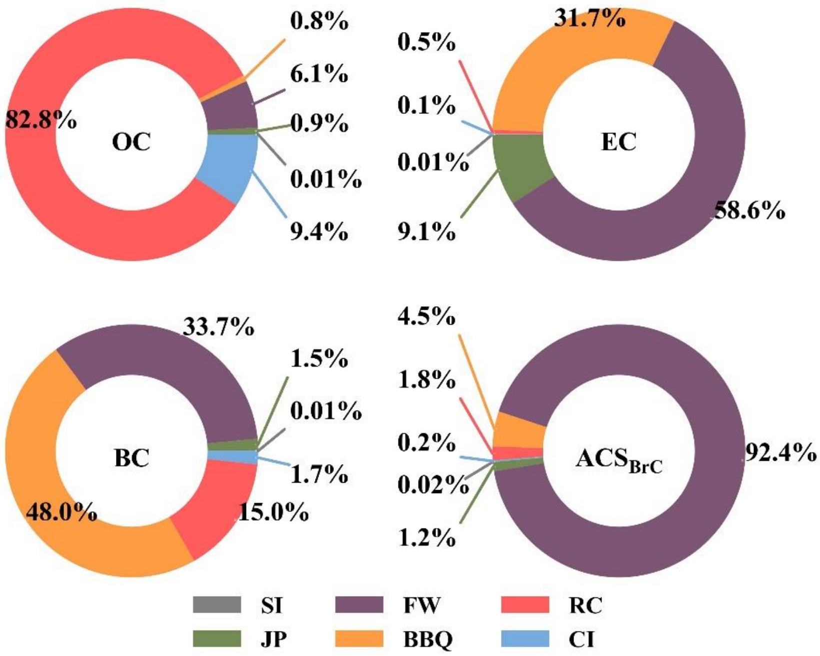

Figure S8 Contribution of different sources to the total CA emissions from FMS of China in 2018 (SI: sacrificial incense; JP: joss paper; FW: fireworks; BBQ: barbecue; RC: residential cooking; CI: catering industry).

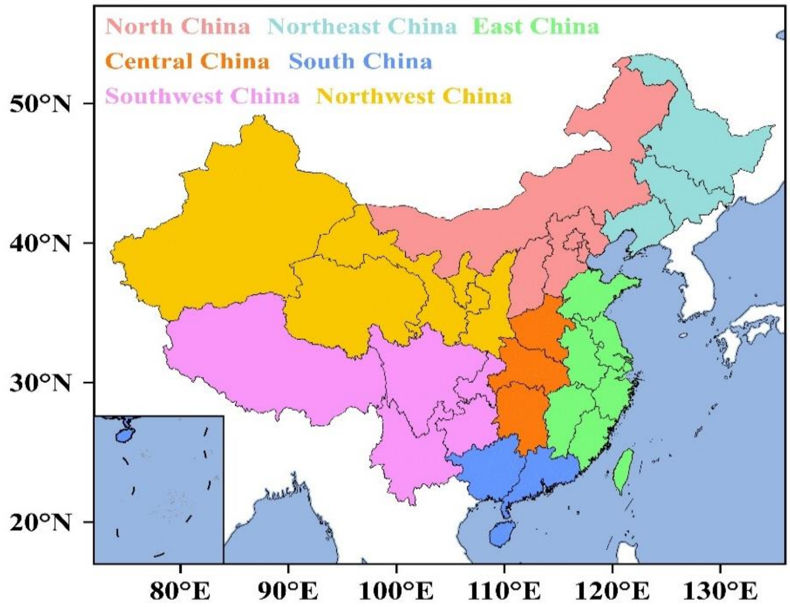

Figure S9 The seven geographical regions in China

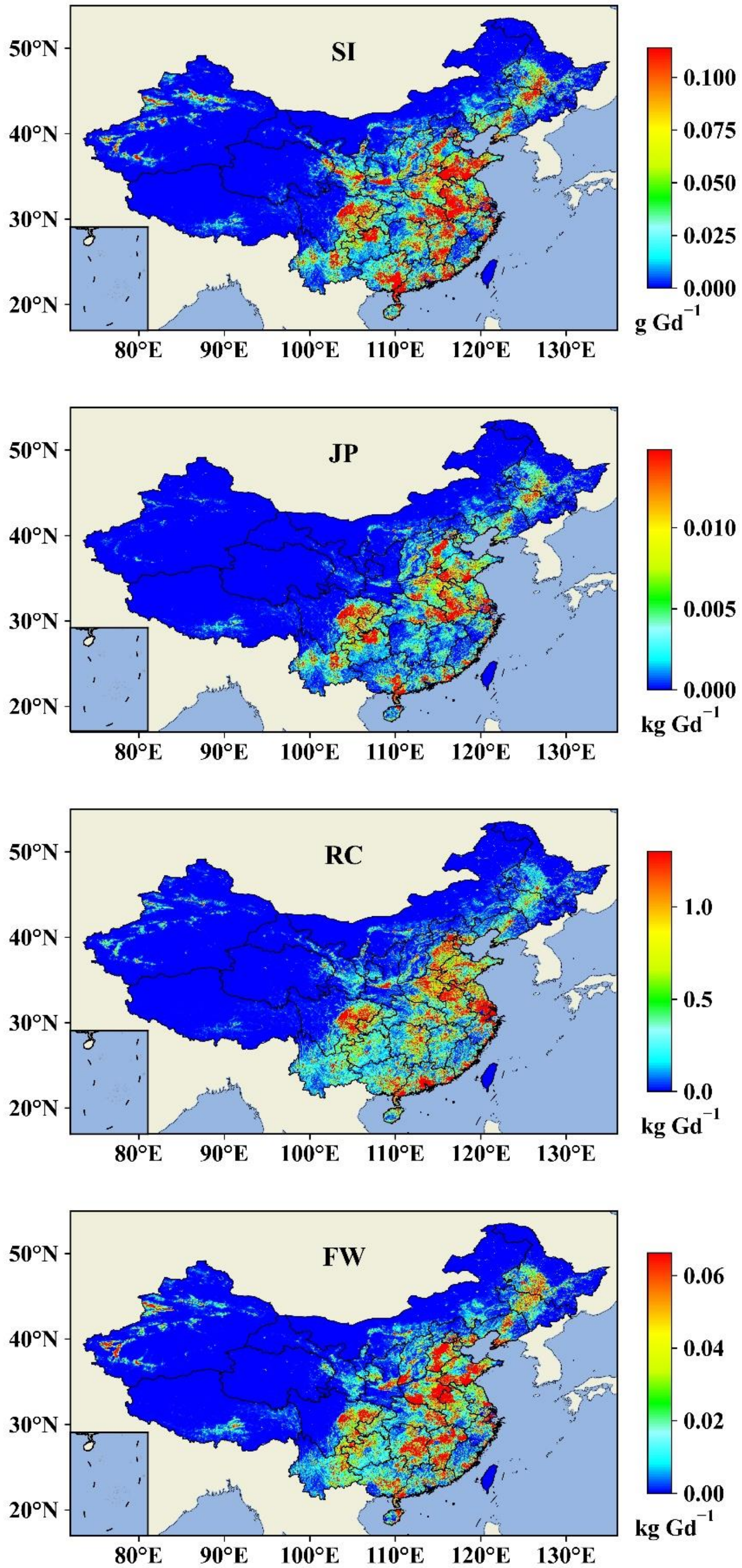

Figure S10 Distribution of OC emission intensities from different sources in 2018. Colorbar shows the emissions in each grid.

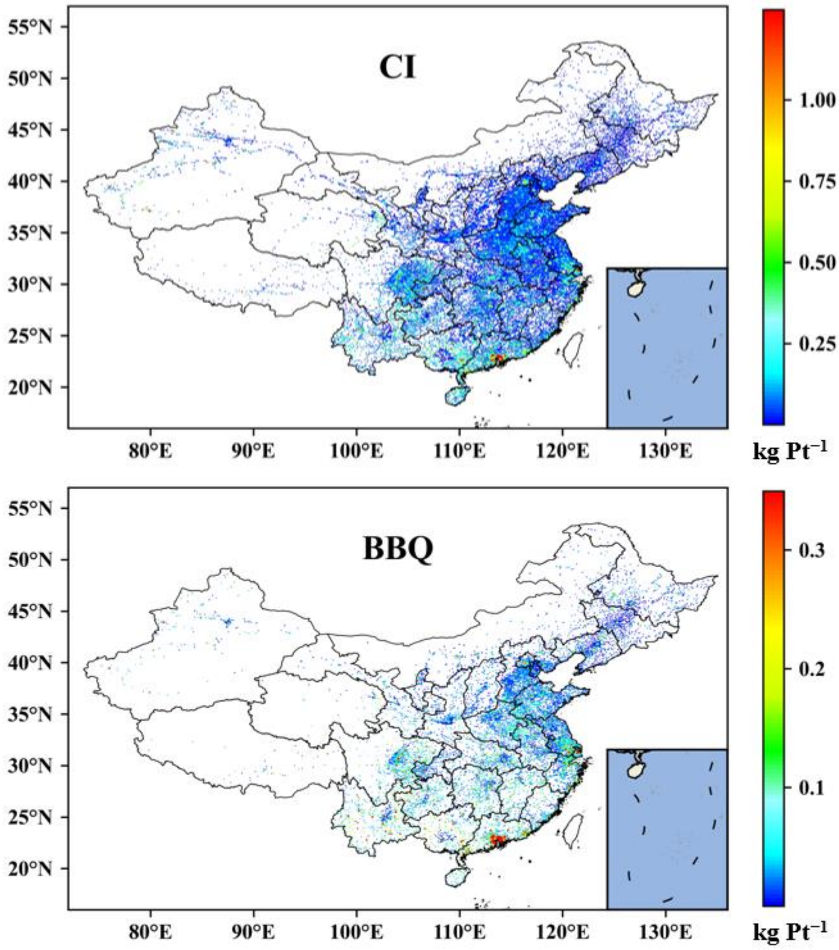

Figure S11 Distribution of OC emission intensities from different sources in 2018. Each point represents a restaurant that deals in barbecue or cooking. Colorbar shows the emission from each point.

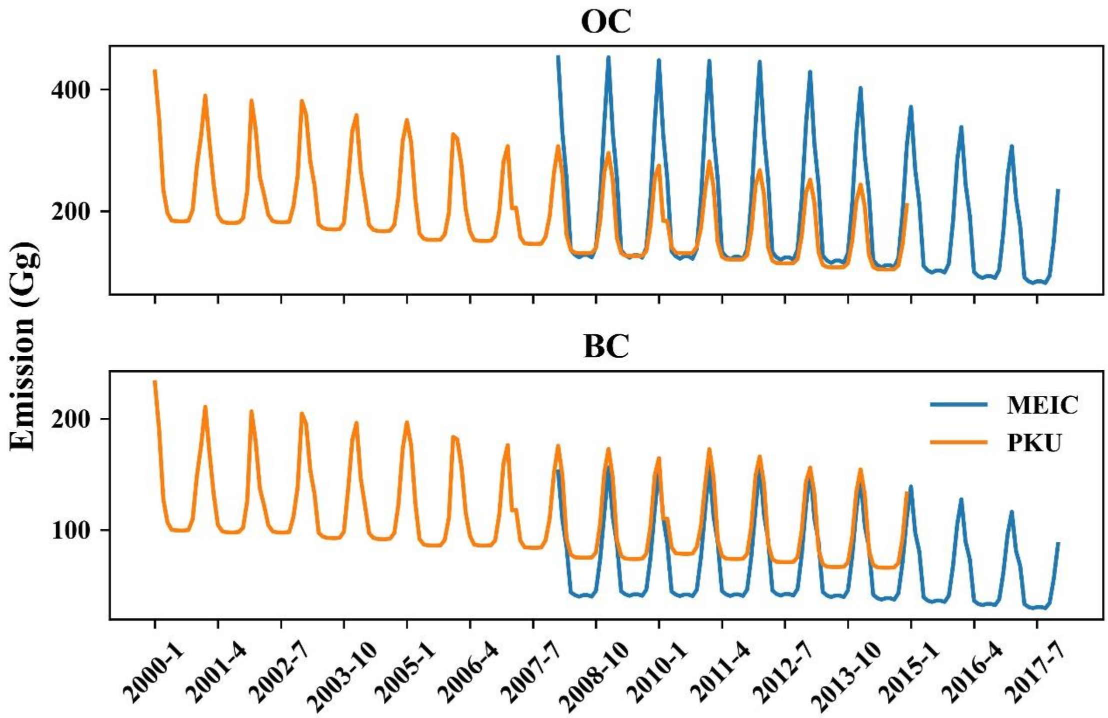

Figure S12 Monthly OC and EC emissions from residential sources (residential and commercial sources) in MEIC inventory (Li et al., 2017) and PKU inventory (Wang et al., 2014; Huang et al., 2015).

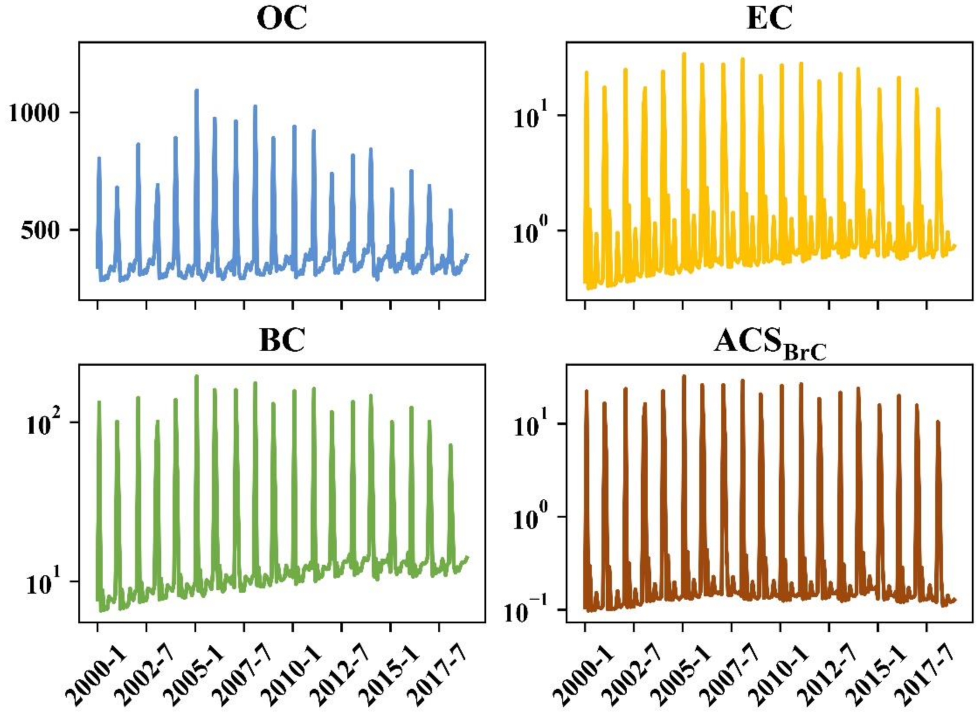

Figure S13 Monthly variation of CA emission from FMS from 2000 to 2018 in this study (OC, EC, BC: t, ACS_{BrC}: Gm^2).

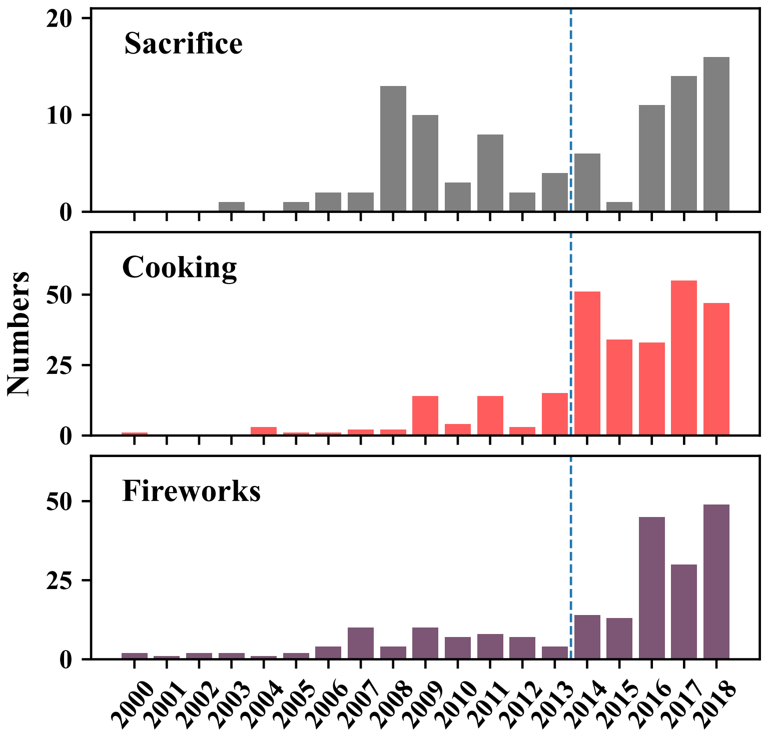

Figure S14 Number of cities that implemented emission control policies of FMS in 2000–2018. The blue dash line represents the pronouncement time of the Air Pollution Prevention and Control Action Plan on September 10, 2013.

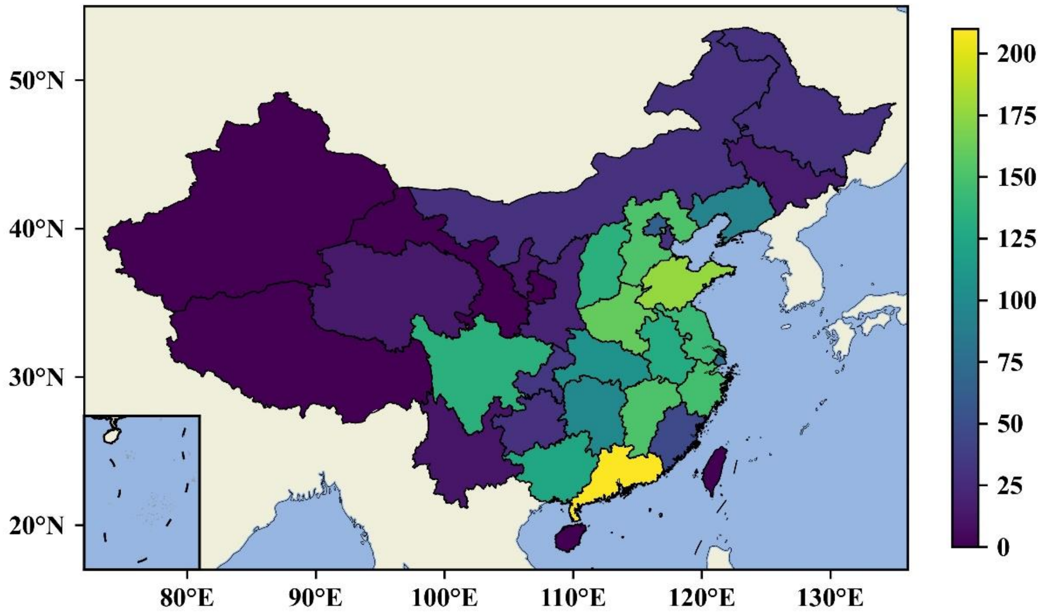

Figure S15 The distribution of questionnaire numbers obtained in each province.

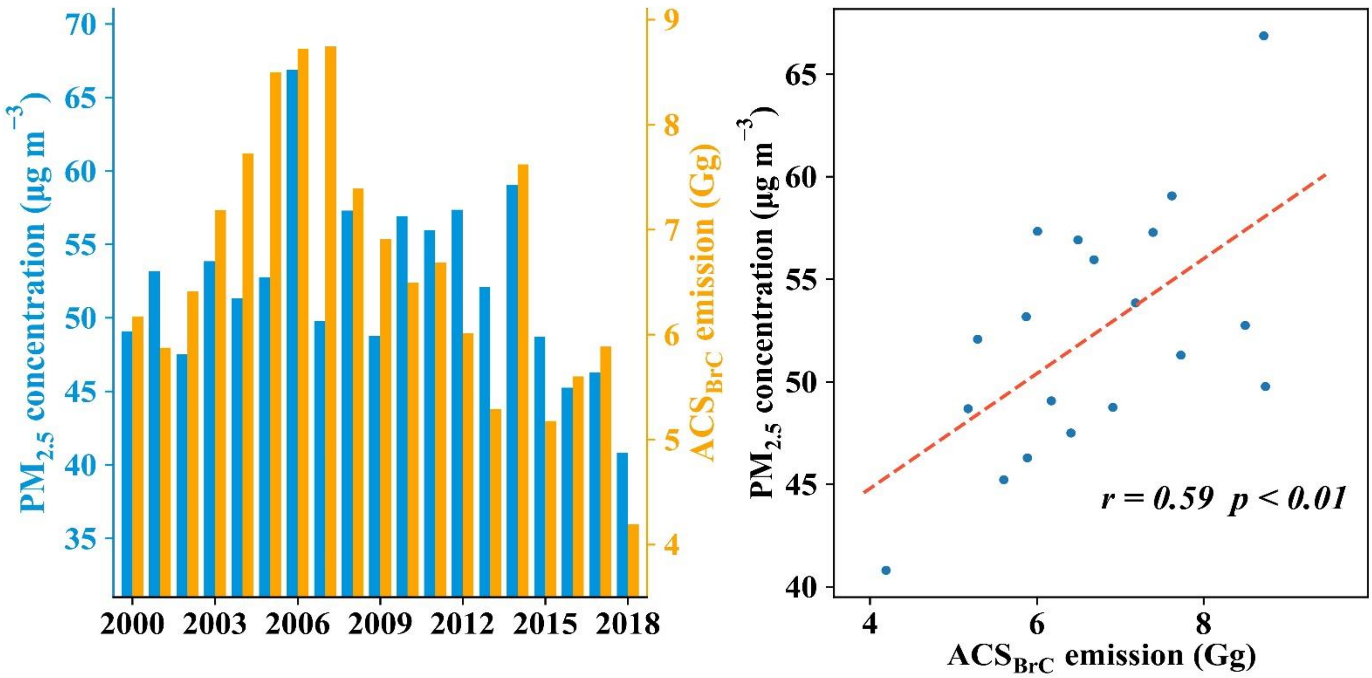

Figure S16 $PM_{2.5}$ concentration and ACS_{BrC} emission from FMS in rural China in 2000–2018 and the correlation between them.

Table S1 The coefficients of activity data for FMS.

Sources	CV	References
Sacrificial incense	−50–50%	Subject judgment
Joss paper	−50–50%	Subject judgment
Barbecue	−20–20%	(Wu et al., 2021)
Cooking	−20–20%	(Wu et al., 2021)
Fireworks	−50–50%	Subject judgment

Table S2 Uncertainties of CA emission inventories of FMS.

	OC	EC	BC	ACS_{BrC}
2000	−16.7–17.2%	−109–112%	−40.4–42.4%	−110–117%
2001	−16.4–16.8%	−110–112%	−40.0–43.1%	−112–113%
2002	−16.6–17.0%	−110–108%	−40.3–41.7%	−113–112%
2003	−16.0–16.8%	−110–109%	−42.3–43.9%	−114–116%
2004	−16.1–17.1%	−109–112%	−42.9–44.4%	−113–116%
2005	−16.0–16.9%	−108–111%	−42.9–43.1%	−113–115%
2006	−16.2–17.2%	−109–112%	−42.7–44.3%	−113–114%
2007	−16.2–16.5%	−108–114%	−42.9–44.6%	−111–116%
2008	−16.3–17.0%	−109–111%	−41.1–42.0%	−111–116%
2009	−16.5–17.3%	−103–108%	−39.9–40.8%	−110–115%
2010	−16.1–17.2%	−103–108%	−39.6–40.4%	−112–111%
2011	−16.4–17.0%	−102–106%	−38.5–40.7%	−110–114%
2012	−16.7–17.1%	−100–104%	−39.2–39.1%	−109–115%
2013	−16.6–17.4%	−99.6–102%	−37.9–39.8%	−109–112%
2014	−16.2–16.9%	−105–107%	−39.1–42.2%	−110–117%
2015	−16.7–17.4%	−96.6–101%	−37.0–37.9%	−111–112%
2016	−16.8–17.5%	−95.7–101%	−37.0–38.0%	−109–112%
2017	−17.0–17.7%	−97.8–99.3%	−36.5–37.9%	−109–115%
2018	−17.6–17.8%	−85.9–91.4%	−35.7–35.1%	−105–107%

Table S3 Correlations of the OC emissions and activity data at a city level in 2018

Source	Activity data	r^{**}
SI	POI of temples	0.36
JP	POI of temples	0.44
FW	POI of firework shops	0.53
BBQ	POI of BBQ restaurants	0.68
CI	POI of restaurants	0.67
BBQ	meat consumptions	0.85
CI	meat consumptions	0.87
RC	meat consumptions	0.78

**: $p<0.01$

Tabel S4 Emission factors of FMS in the literatures.

Source	Pollutant	Emission factors	Experiments	References
Sacrificial incenses	$PM_{2.5}$	2.5–3 g kg^{-1}	Sampling at a chamber using US EPA method 1.	Jilla & Kura. (2017)
	CO	110–120 g kg^{-1}		
	$PM_{2.5}$	7.7–205.4 g kg^{-1}	Chamber experiments with 10 incenses.	Lee & Wang. (2004)
	$PM_{2.5}$	11.09–23.38 g kg^{-1}	Chamber experiments with 4 incenses.	Kuo et al. (2016)
	$PM_{2.5}$	5.0–55.7 g kg^{-1}	23 tests were conducted in a specifically designed chamber, and particles were collected on filters.	Jetter et al. (2002)
	PM_{10}	5.4–49.4 g kg^{-1}		
	$PM_{2.5}$	0.4–44.5 g kg^{-1}	6 brands of Singaporean incenses were burned. Filter samples were collected in a combustion chamber.	See & Balasubramanian. (2011)
	OC	0.01–1.79 g kg^{-1}		
	EC	0.26–29.5 g kg^{-1}		
	mercury	9.78–13.82 ng g^{-1}	Sampling at a traditional temple in China.	Shen et al. (2017)
	PAHs	8.81–9.14 mg kg^{-1}	Considering the impact of additions on PAHs emissions from incenses.	Yang et al. (2013)
	OC	21.4–49.2 mg kg^{-1}	-	This study
	BC	1.07–3.09 mg kg^{-1}		
Joss paper	PAHs	67.3–74.6 mg kg^{-1}	Sampling at two paper furnaces at temples.	Yang et al. (2005)
	mercury	4.67–13.16 ng g^{-1}	Sampling at a traditional temple in China.	Shen et al. (2017)
	$PM_{2.5}$	4.23±0.71 g kg^{-1}	Four types of sacrificial offerings were burned in an incinerator. Particles were sampled using a dilution tunnel.	Zhang et al. (2019)
	OC	1.26±0.42 g kg^{-1}		
	EC	0.63±0.28 g kg^{-1}		

	OC	14.6–75.5 mg kg^{-1}	-	This study
	BC	1.65–6.27 mg kg^{-1}		
Fireworks	TSP	67–140 g kg^{-1}	Fireworks were burned in a block room (41.2 m^3). Fireworks were put on preweighed aluminum foil and then ignited.	Croteau et al. (2010)
	K	23–45 g kg^{-1}		
	Mg	1.3–6.8 g kg^{-1}		
	PM_{10}	54–429 g kg^{-1}	Fireworks were burned in a sampling chamber with four ventilators.	Camilleri & Vella. (2016)
	$PM_{2.5}$	200–325 g kg^{-1}	7 types of fireworks were tested in a measurement chamber.	Keller & Schragen. (2021)
	PM_{10}	134–281 g kg^{-1}		
	OC	5.86–124 mg kg^{-1}	-	This study
	EC	2.89–37.3 mg kg^{-1}		
Cooking	$PM_{2.5}$	2.06±3.03 mg min^{-1}	15 Chinese dishes, and considering influences of oil temperature, meat type, cooking method, and so on.	Zhao et al. (2018)
	$PM_{2.5}$	4.88±3.43 g h^{-1}	Sampling particle and VOCs in 18 traditional cuisine restaurants in Beijing.	Lin et al. (2021)
	OC	0.42–7.28 g h^{-1}		
	Carbonyl compounds	0.70–1.53 μg kg^{-1}	Laboratory tests, and considering cooking styles.	Xiang et al. (2017)
	VOCs	12–38 mg kg^{-1}	Laboratory sampling with a dilution system.	Cheng et al. (2016)
	VOCs	11.15–189.8 g h^{-1}	Sampling at restaurants of seven cuisine types.	Wang et al. (2018a)
	Carbonyl compounds	90.4–274 μg kg^{-1}	Test by US EPA Test Method 5G.	Que et al. (2019)
	PM	16.8–22.3 mg min^{-1}	Chamber samplings of PM.	Wang et al. (2018b)
	PAHs	79.9–270.6 ng min^{-1}	Sampling at a residential kitchen in Beijing.	Zhao et al. (2019)

	$PM_{2.5}$	19–39 mg kg^{-1}	Laboratory tests, direct sampling of particles without cooling and dilution process.	Wang et al. (2015)
	$PM_{2.5}$	0.1–9.2 g kg^{-1}	Laboratory tests, and the sampling system were devised according to the restaurant facility.	Lin et al. (2019)
	OC	124–203 mg kg^{-1}	-	This study
	BC	1.34–1.79 mg kg^{-1}		
Barbecue	$PM_{2.5}$	15.48±7.22 g h^{-1}	Sampling particle and VOCs in 18 traditional cuisine restaurants in Beijing.	Lin et al. (2021)
	OC	4.31–32.1 g h^{-1}		
	Carbonyl compounds	1.60 µg kg^{-1}	Laboratory tests, and considering cooking styles.	Xiang et al., 2017)
	PM_1	8.5–270 mg min^{-1}	Laboratory tests, grilling meat 12 min on 550–600 ℃. Considering the influences of the burner, pan, meat, and salt.	Amouei Torkmahalleh et al. (2018)
	Carbonyl compounds	191µg kg^{-1}	Test by US EPA Test Method 5G	Que et al. (2019)
	VOCs	41 mg kg^{-1}	Laboratory sampling with a dilution system.	Cheng et al. (2016)
	$PM_{2.5}$	21 mg kg^{-1}	Laboratory tests, direct sampling of particles without cooling and dilution process.	Wang et al. (2015)
	OC	21.5–50.5 mg kg^{-1}	-	This study
	BC	1.66–191 mg kg^{-1}		

Table S5 Emission amounts of air pollutants from FMS or other sources.

Sources	Region	Pollutant	Time	Emission amount	References
Total	China	OC	2000–2014	2345–3587 Gg	Huang et al. (2015)
Residential & commercial				1675–2931 Gg	
Total	China	BC	2000–2014	1455–1624 Gg	Wang et al. (2014)
Residential & commercial				1061–1589 Gg	
Total	China	OC	2008–2017	2080–3190 Gg	Li et al. (2017)
		BC		1253–1728 Gg	
Residential		OC		1689–2512 Gg	
		BC		626–828 Gg	
Cooking in restaurants	China	VOCs	-	66245 t	Wang et al. (2018a)
Household cooking	Individual household	$PM_{2.5}$	-	12.4 kg	Wang et al. (2015)
Household cooking	Beijing	$PM_{2.5}$	2015	3.99 Gg	Cai et al. (2018)
		NMVOCs		4.02 Gg	
Catering industry	China	$PM_{2.5}$	2017	38.2 Gg	Jin et al. (2021)
		OC		26.8 Gg	
		BC		0.8 Gg	
Barbecue	Individual household	$PM_{2.5}$	-	3.8 kg	Wang et al. (2015)
Sacrificial incense	China	OC	2000–2018	0.54–1.43 t	This study
Joss paper				38.5–105 t	
Fireworks				267–674 t	
Barbecue				14.8–35.3 t	
Cooking				4418–7443 t	
Sacrificial incense		EC		0.03–0.08 t	
Joss paper				3.49–9.50 t	
Fireworks				76.3–193 t	
Barbecue				47.3–109 t	
Cooking				41.3–69.6 t	

Reference

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