Atmos. Chem. Phys., 17, 2839–2864, 2017
www.atmos-chem-phys.net/17/2839/2017/
doi:10.5194/acp-17-2839-2017

Atmospheric Chemistry and Physics
Open Access

A comprehensive biomass burning emission inventory with high spatial and temporal resolution in China

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Received: 1 July 2016 – Discussion started: 19 July 2016
Revised: 20 January 2017 – Accepted: 23 January 2017 – Published: 23 February 2017

Abstract. Biomass burning injects many different gases and aerosols into the atmosphere that could have a harmful effect on air quality, climate, and human health. In this study, a comprehensive biomass burning emission inventory including domestic and in-field straw burning, firewood burning, livestock excrement burning, and forest and grassland fires is presented, which was developed for mainland China in 2012 based on county-level activity data, satellite data, and updated source-specific emission factors (EFs). The emission inventory within a $1 \times 1\,km^2$ grid was generated using geographical information system (GIS) technology according to source-based spatial surrogates. A range of key information related to emission estimation (e.g. province-specific proportion of domestic and in-field straw burning, detailed firewood burning quantities, uneven temporal distribution coefficient) was obtained from field investigation, systematic combing of the latest research, and regression analysis of statistical data. The established emission inventory includes the major precursors of complex pollution, greenhouse gases, and heavy metal released from biomass burning. The results show that the emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and Hg in 2012 are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34 380.4 Gg, 369.7 Gg, 1189.5 Gg, 675 299.0 Gg, 2092.4 Gg, and 4.12 Mg, respectively. Domestic straw burning, in-field straw burning, and firewood burning are identified as the dominant biomass burning sources. The largest contributing source is different for various pollutants. Domestic straw burning is the largest source of biomass burning emissions for all the pollutants considered, except for NH_3, EC (firewood), and NO_x (in-field straw). Corn, rice, and wheat represent the major crop straws. The combined emission of these three straw types accounts for 80 % of the total straw-burned emissions for each specific pollutant mentioned in this study. As for the straw burning emission of various crops, corn straw burning has the largest contribution to all of the pollutants considered, except for CH_4; rice straw burning has highest contribution to CH_4 and the second largest contribution to other pollutants, except for SO_2, OC, and Hg; wheat straw burning is the second largest contributor to SO_2, OC, and Hg and the third largest contributor to other pollutants. Heilongjiang, Shandong, and Henan provinces located in the north-eastern and central-southern regions of China have higher emissions compared to other provinces in China. Gridded emissions, which were obtained through spatial allocation based on the gridded rural population and fire point data from emission inventories at county resolution, could better represent the actual situation. High biomass burning emissions are concentrated in the areas with more agricultural and rural activity. The months of April, May, June, and October account for 65 % of emissions from in-field crop residue burning, while, regarding EC, the emissions in January, February, October, November, and December are relatively higher than other months due to biomass

domestic burning in heating season. There are regional differences in the monthly variations of emissions due to the diversity of main planted crops and climatic conditions. Furthermore, $PM_{2.5}$ component results showed that OC, Cl^-, EC, K^+, NH_4^+, elemental K, and SO_4^{2-} are the main $PM_{2.5}$ species, accounting for 80 % of the total emissions. The species with relatively high contribution to NMVOC emission include ethylene, propylene, toluene, mp-xylene, and ethyl benzene, which are key species for the formation of secondary air pollution. The detailed biomass burning emission inventory developed by this study could provide useful information for air-quality modelling and could support the development of appropriate pollution-control strategies.

1 Introduction

Biomass burning is considered a significant source of gas and particulate matter (PM), resulting in a major impact on atmospheric chemistry, climate, and human health. Active trace gases (e.g. sulfur dioxide (SO_2), nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), ammonia (NH_3)) released from biomass burning are the major precursors of secondary inorganic/organic aerosols and tropospheric ozone (O_3) in the atmosphere (Penner et al., 1992; Kaufman and Fraser, 1997; Koppmann et al., 2005; Langmann et al., 2009). Several studies have indicated that observed local and regional air pollution could be attributed to the pollutants emitted from biomass burning (Huang et al., 2012a; Zha et al., 2013; Cheng et al., 2014; Yan et al., 2014; Zong et al., 2016). The emission factor (EF) of some biomass burning pollutants is even greater than that of coal burning, which is widely recognized as a major pollution source (Zheng et al., 2009; Fu et al., 2013). Primary particles (e.g. elemental carbon (EC) and organic carbon (OC)) discharged by biomass burning not only impact visibility, but also have an influence on climate due to the positive effects of the absorption of light and cloud condensation (IPCC, 2013). Biomass burning is also a significant source of greenhouse gases such as methane (CH_4) and carbon dioxide (CO_2) (Andreae and Merlet, 2001), which contribute to global warming (Sun et al., 2016). Moreover, several reports (Fernandez et al., 2001; Huang et al., 2012a; Shi and Yamaguchi, 2014) reveal that the long- or short-term exposure to PM (e.g. BC emitted from indoor biomass burning) can cause adverse effects to human health, such as decreased lung function, increased respiratory diseases, and increased lung cancer mortality. Furthermore, studies have identified that indoor biomass burning could bring about adverse health effects for residents (Jiang and Bell, 2008; Fullerton et al., 2008).

Prior to its rapid economic development, China was a large agricultural country and thus once consumed a large amount of biofuels (e.g. straw and firewood). With the dramatic urbanization that accompanied economic development, the pattern of energy consumption in rural areas has been gradually transformed. In particular, in some agricultural areas with relatively high income, straws were more frequently burned in the field (Sun et al., 2016). Since 1999, the Chinese government has issued a series of laws and regulations to ban the in-field burning of straw and to encourage comprehensive utilization of straw, such as returning it to the field and using it in livestock feeding, industrial raw materials manufacturing, briquette fuel processing, etc. (MEP, 1999). However, the effect of this legislation was not satisfactory because the processes of comprehensive straw utilization not only required high labour costs but also delayed sowing the next crop. Thus, the phenomenon of straw in-field burning continued to occur. The amount of in-field straw burning in China in 2009 was estimated at 0.215 billion Mg. These data were obtained from the governmental report on the investigation and evaluation of crop straw resources in various provinces in China (MA, 2011). Accordingly, a comprehensive and detailed emission inventory of biomass burning representing the current status in China is important to provide valuable information for researchers and policymakers. Examples of potential applications include research to understand the influence of biomass burning on indoor air quality and the outdoor atmospheric environment, and the development of effective management decisions to relieve the associated environmental burden and to reduce health risk.

Since the early research conducted by Crutzen et al. (1979), a series of efforts have been made to develop biomass burning emission inventories, especially in developed countries (Reddy and Venkataraman, 2002; Ito and Penner, 2004; van der Werf et al., 2006; Nelson et al., 2012; Shon, 2015). Compared with the developed countries, research by Chinese scientists on this issue started relatively late. The initial studies on biomass burning emission inventory across China (Streets et al., 2001, 2003; Tian et al., 2002; Cao et al., 2005) or in certain regions (Zheng et al., 2009; Huang et al., 2011) were developed mainly based on EFs developed for foreign nations (Turn et al., 1997; Andreae and Merlet, 2001; US EPA, 2002) because of the lack of local measurements in China. However, this approach could introduce great relative uncertainty in emission estimates because of the differences in crop types and combustion conditions between China and other counties.

In recent years, various research activities have focused on the emission characteristics of biomass burning in China, including local EF and chemical species profile tests. Li et al. (2007a, 2009) conducted field measurements to determine the EF for several of the main household biofuels in Beijing, Chongqing, Henan, and Shandong. Li et al. (2007b) determined the EF for in-field wheat and maize straw burning and Cao et al. (2008) measured EFs for the domestic burning of rice straw, wheat straw, corn straw, and cotton straw. Zhang et al. (2008) measured CO_2, carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO_2), NO_x, and PM EFs of rice, wheat, and corn straw. Wang et al. (2009) launched a study on the characteristics of gaseous pollutants from biofuel stoves in China. More recently, Y. S. Zhang et al. (2013) carried out experiments on EFs for in-field burning of sugar cane leaves and rice straw in south-eastern China. Ni et al. (2015) conducted laboratory burning tests to determine the EFs of wheat straw, rice straw, and corn straw, considering the impacts of the fuel moisture content.

Based on the local EFs, emission inventories that focused on certain provinces (Li et al., 2015; He et al., 2015) or city group regions (He et al., 2011; Fu et al., 2013) were developed. In our previous study, we reported an emission inventory with high resolution in the Beijing–Tianjin–Hebei region of China (Zhou et al., 2015). To produce a national emission inventory, several studies of biomass burning have been carried out without distinguishing the detailed crop straws (Lu et al., 2011; Yan et al., 2006; Tian et al., 2011). Moreover, there are several studies that have focused on certain pollutants (Huang et al., 2012c; Chen et al., 2013; W. Zhang et al., 2013; Kang et al., 2016; Li et al., 2016) and on certain crop straws (Zhang et al., 2008; Hong et al., 2016; Sun et al., 2016). In recent years, the comprehensive biomass emission inventory has been limited. Most recent studies have concentrated on biomass open burning, including the multi-year trend analysis on certain or multiple pollutants (Wang and Zhang, 2008; Song et al., 2009; Shi and Yamaguchi, 2014; Shon, 2015; Xu et al., 2016; Zhang et al., 2016). Few studies have covered recent firewood burning (see next paragraph for details regarding the reason for this). In addition to the EF, detailed activity data are also important for a reliable emission inventory, such as domestic and in-field straw burning percentages, which are not currently publicly available. Gao et al. (2002) produced a study on the percentage of straw used as fuel and for direct incineration in 2000. Wang et al. (2008) investigated the percentage of in-field straw burning in six regions in China in 2006, which were divided according to their similarities in agriculture, climate, economy, and region. Tian et al. (2011) estimated the proportion of domestic and in-field straw burning in 2007 for seven and three regions of China, respectively. Thus, there is limited information about the percentage of straw used as fuel or waste in the field that reflects the status of China in recent years for different provinces. Moreover, because of the lack of a firewood consumption report in the *China Energy Statistical Yearbook* (NBSC, 2009–2015), few studies have developed a comprehensive biomass burning emission inventory in China in recent years. The *China Energy Statistical Yearbook* provides official information on energy construction, production, and consumption, including detailed firewood consumption in various regions. However, the firewood consumption data have not been included in the NBSC (2009–2015) since 2008; as a result, there is little literature containing a comprehensive biomass burning emission inventory for China.

Consequently, we have identified several weaknesses in the current biomass burning emission inventories. First, not all biomass burning sources have been included in recent years, especially since 2008, because of the lack of firewood consumption data in the various statistical yearbooks (e.g. the *China Energy Statistical Yearbook*, *China Statistical Yearbook*, and *China Rural Statistical Yearbook*). Second, the source-specific EFs used in emission estimation need to be updated based on the systematic combing of local tests in the latest research. Third, the proportion of domestic and in-field straw burning, which could reflect the recent conditions of different provinces in China, needs to be investigated. Fourth, the current biomass burning emission inventory for China is generally at province resolution because detailed activity data cannot be directly obtained from the various statistical yearbooks in China. Activity data at coarse resolution are likely to be associated with great uncertainty in grid emissions generated according to source-based gridded spatial surrogates (e.g. population) using GIS technology (Zheng et al., 2014). As a result, it is of great importance to develop an integrated and model-ready biomass burning emission inventory with high spatial and temporal resolution.

In this study, a comprehensive biomass burning emission inventory including domestic and in-field straw burning, firewood and livestock excrement burning, and forest and grassland fires was developed for the Chinese mainland (excluding Hong Kong, Macao, and Taiwan) for 2012, based on detailed activity data and satellite burned-area data. In addition, we attempted to take the source-specific EFs measured in China into full account. A range of important information for estimating emissions (e.g. province-specific domestic/in-field straw burning percentage, detailed firewood burning quantities, and uneven temporal distribution coefficient) were obtained from a field investigation, systematic combing of latest research, and regression analysis of statistical data. A 1 km-resolution emission inventory was generated using GIS software. The gaseous and particulate pollutants examined in this research included SO_2, NO_x, particulate matter with a diameter below 10 µm (PM_{10}), particulate matter with a diameter below 2.5 µm ($PM_{2.5}$), NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and mercury (Hg), covering the major precursors of complex pollution, greenhouse gases, and heavy metals released from biomass burning. The detailed emission inventory given by this paper could provide valuable information to support further biomass burning pollution research and the development of a targeted control strategy of all regions across the Chinese mainland.

The remainder of this paper is structured as follows. In Sect. 2, we describe the methodology, including the emission estimation method, the selection and handling of activity data and corresponding parameters, determination of EFs, spatial and temporal allocation, and speciation of $PM_{2.5}$ and NMVOC. In Sect. 3.1, we describe the total emission in China, and the contribution of various biomass burning sources and crop straws. In Sect. 3.2, we describe the emission from different regions and the contributions of different biomass sources and crop straws of each province. Spatial

Table 1. Classification of biomass burning emission sources.

I	II	III
Domestic burning	Firewood	Firewood
	Livestock excrement	Cattle Horses Donkeys Mules Camels
	Straw	Corn, wheat, cotton, sugar cane, potato, peanut, rapeseed, sesame, sugar beet, hemp, rice, soya bean
Open burning	In-field straw	Corn, wheat, cotton, sugar cane, potato, peanut, rapeseed, sesame, sugar beet, hemp, rice, soya bean
	Forest	Evergreen needleleaf forest Evergreen broadleaf forest Deciduous needleleaf forest Deciduous broadleaf forest Mixed forest Closed shrublands Open shrublands Evergreen needleleaf forest
	Grassland	Woody savannas
		Savannas
		Grasslands

and temporal distribution of biomass burning emissions is discussed in Sect. 3.3 and 3.4, respectively. In Sect. 3.5, we present the emissions of $PM_{2.5}$ and NMVOC species. Uncertainty in biomass burning emission estimates is described in Sect. 3.6. The comparison between this study and other studies appears in Sect. 3.7. In Sect. 4, we summarize our conclusions.

2 Methodology

2.1 General description

The biomass burning considered in this study is mainly divided into two categories, domestic burning and open burning. Domestic burning mainly involves domestic straw (straw burned as fuel indoors), firewood, and livestock excrement (mainly used in pastoral and semi-pastoral areas) burning. Open burning includes in-field straw burning (straw burned as waste outdoors, including crop stalk and residue) and forest and grassland fires. Straw burning without a specific description in this paper refers to the total straw burning, including in-field and domestic straw burning. Details of the source classifications are shown in Table 1.

A bottom-up approach was used to develop the biomass burning emission inventory for all districts or counties. The annual biomass burning emissions (E_i) were calculated using Eq. (1) as follows:

$$E_i = \sum \left(A_j \times \mathrm{EF}_{i,j}\right)/1000, \quad (1)$$

where subscripts i and j represent the type of pollutant and biomass burning source, respectively; E is the annual typical pollutant emission ($\mathrm{Mg\,year^{-1}}$); A is the annual amount of dry biomass burned ($\mathrm{Mg\,year^{-1}}$), for which the detailed calculation method is shown in Sect. 2.2; and EF is the emission factor ($\mathrm{g\,kg^{-1}}$), for which a detailed description is presented in Sect. 2.3.

2.2 Activity data

2.2.1 Straw burning

The burning mass of domestic and in-field straw burning can be calculated using Eq. (2) as follows:

$$A_{i,k} = P_{i,k} \times N_k \times R_{i,k} \times D_k \times \mathrm{CE}_k, \quad (2)$$

where subscripts i and k represent the region (district or county) and crop type, respectively; $A_{i,k}$ is the annual burning mass of each crop straw in each region ($\mathrm{Mg\,year^{-1}}$); $P_{i,k}$ is the amount of crop-specific yield per year in each region

(Mg year^{-1}); N_k is the straw-to-product ratio of each straw type; $R_{i,k}$ is the domestic or in-field straw burning percentage; D_k is the dry matter fraction of each straw type; and CE_k is the combustion efficiency of each straw type.

There are currently no statistics on the amount of each crop yield at the county resolution ($P_{i,k}$) in various yearbooks in China. Therefore, in this study, we conducted a correlation analysis between grain yield and crop yield at prefecture resolution, and found a good correlation ($R = 0.747$; a detailed analysis is provided in the Supplement, Fig. S1). The grain yield at prefecture resolution was summarized from the *China Statistical Yearbook* for 2012 (NBSC, 2013b). The crop yield at prefecture resolution was summarized from statistical yearbooks edited by the National Bureau of Statistics in 2012 for each province. Next, the $P_{i,k}$ at county level was calculated based on the various types of crop yield at prefecture resolution and of grain yield at county resolution. Grain yield at county resolution was summarized from a range of statistical yearbooks edited by the National Bureau of Statistics in 2012 for each province and city (NBSC, 2013a, b). The total amount of straw in China in 2012 calculated in this study is 832.5 Tg, which is similar to the data from Chinese governmental annual statistical reports on straw utilization and burning (NDRC, 2014; the amount of straw that can be collected is 817.4 Tg). The maps at prefecture and county resolution are shown in Fig. S2 in the Supplement.

The variable $R_{i,k}$ is important for biomass burning emission estimation, and the information that can represent the recent status in China needs to be updated because of continued economic development and gradual implementation of national control policies for in-field straw burning. In this study, we conducted a detailed review of recent literature to derive the percentage of straw burned as domestic fuel and that burned as waste for each province. For some provinces in which the current reporting is limited (e.g. Heilongjiang, Zhejiang, Guangdong, Inner Mongolia, and Hebei), a questionnaire-based survey was carried out. Details of the questionnaire survey are presented in the Supplement (Sect. S3). $R_{i,k}$ is summarized in Table 2. According to our estimation, the amount of domestic and in-field straw burning in China in 2012 were 0.26 billion and 0.19 billion Mg, respectively, which is similar to other recently published results for 2012 (0.26 billion Mg domestic straw burning, Tian, 2014) and 2009 (0.215 billion Mg in-field straw burning, MA, 2011).

The N_k, D_k, and CE_k values were obtained from the collection of literature reviewed. Detailed parameters used in this study are summarized in Table 3.

2.2.2 Firewood

Firewood consumption is recorded as non-commodity energy in the *China Energy Statistical Yearbook*. However, detailed firewood consumption data have not been publicly available since 2008. For more recent years, we obtained the total firewood consumption for China in 2012 and for each province in 2010 (Tian et al., 2014; OECD, 2012). However, these data could not support the development of an emission inventory at high resolution. There are several detailed statistics available in the yearbook, such as the rural population, gross agricultural output, and timber yield, which are likely to have a relationship with the firewood consumption. Therefore, we produced a correlation analysis between the three statistics and the firewood consumption of each province for different years in which the firewood consumption data were available at province resolution, as shown in Fig. 1. The best correlation relationship was found between rural population and firewood consumption. The correlation coefficient for the different years ranged from 0.66 to 0.82, and, therefore, we chose rural population as the surrogate to calculate the detailed firewood consumption. The firewood consumption at county resolution was obtained based on the rural population at county resolution and on the total firewood consumption reported by Tian et al. (2014) and OECD (2012). China's rural population, gross agriculture output, and timber yield of each province were from NBSC (1999–2008a). Firewood consumption data were from NBSC (1999–2008b).

2.2.3 Forest and grassland burning

The burning mass of forest/grassland can be calculated from the annual mass of forest/grassland burned (Mg year^{-1}) using Eq. (3) as follows:

$$A = \left(\sum_{j=1}^{10} \mathrm{BA}_{x,j} \times \mathrm{FL}_{x,j} \times \mathrm{CF}_j \right) \times 10^{-6}, \quad (3)$$

where subscripts j and x represent the land cover type and location, respectively; A is the annual burning mass of forest and grassland fires (Mg year^{-1}); $\mathrm{BA}_{x,j}$ is the burned area (m^2 year^{-1}) of land cover type j at x; $\mathrm{FL}_{x,j}$ is the biomass fuel loading (the above-ground biomass density in this study; g m^{-2}) of land cover type j at x; and CF_j is the combustion factor (the fraction of above-ground biomass burned) of land cover type j.

Burned-area data for 2012 were derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) direct-broadcast burned-area product (MCD64A1; http://modis-fire.umd.edu/). This product employs an automated algorithm for mapping MODIS post-fire burned areas and deriving the approximate burn date within each burn cell combined with surface reflectance, land cover products, and daily active fires. The MCD64A1 product has a primary spatial resolution of 500 m. Daily burned areas could be obtained from the product.

Earlier research on the estimation of fuel loading (FL) values for forest and grassland typically employed an averaged value of above-ground biomass density. However, these values do not reflect the spatial variations of FL for each vegetation type well. In this study, numerous local FL values

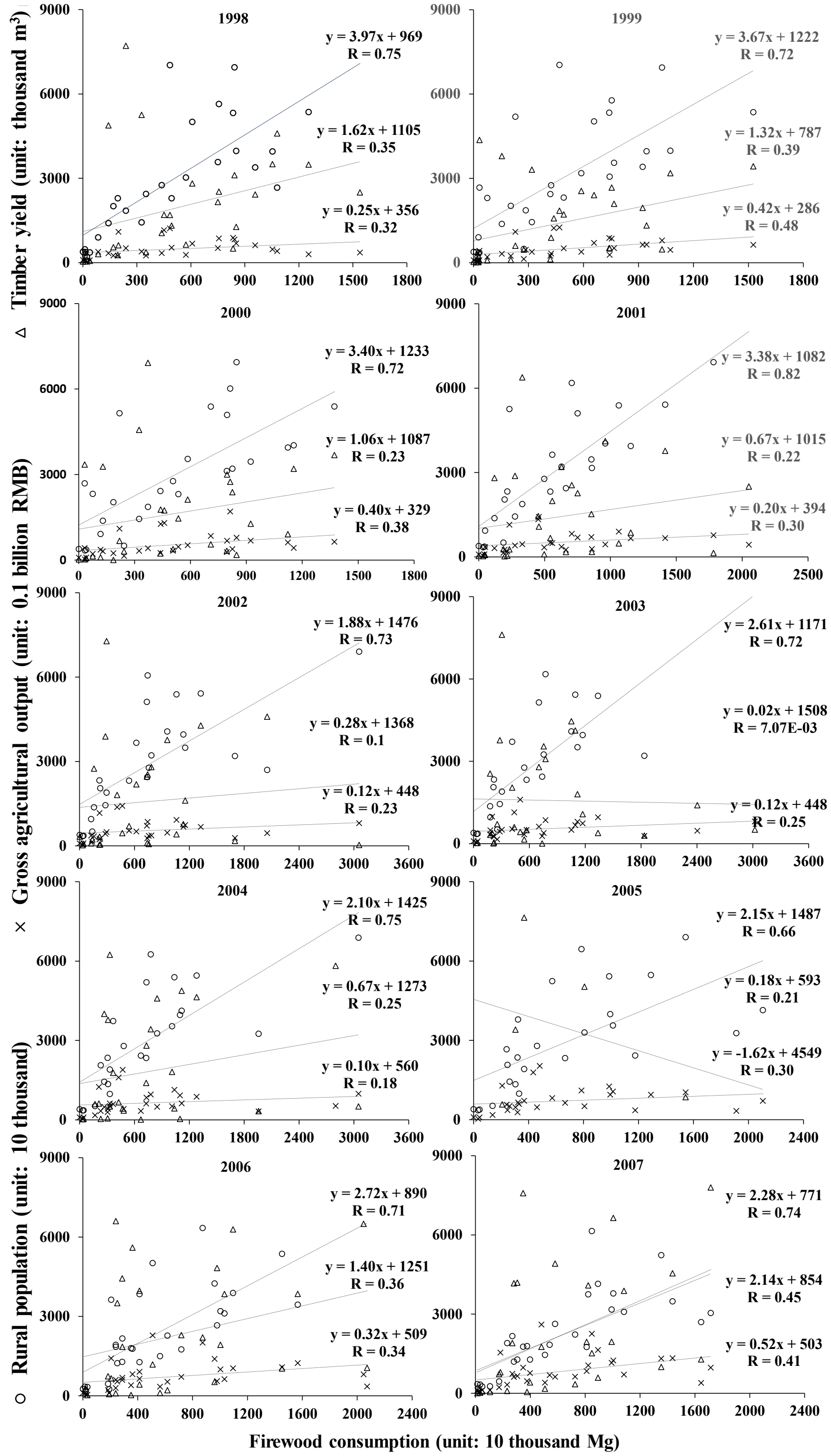

Figure 1. Regression analysis between firewood consumption at province resolution and (1) rural population, (2) gross agricultural output, and (3) timber yield denoted by circles, crosses, and triangles, respectively. Regression equations for each figure are provided in the top, middle, and bottom panels.

Table 2. Domestic and in-field straw burning percentage of each province.

Province	Domestic straw burning percentage	In-field straw burning percentage	Province	Domestic straw burning percentage	In-field straw burning percentage
Beijing	0.0923[a]	0.096[b]	Hubei	0.283[j]	0.197[o]
Tianjin	0.42[a]	0.165*	Hunan	0.4[c]	0.2[c]
Hebei	0.35*	0.165*	Guangdong	0.17*	0.1976*
Shanxi	0.45[c]	0.2[c]	Guangxi	0.2226[k]	0.2273[k]
Inner Mongolia	0.338*	0.246*	Hainan	0.45[c]	0.2[c]
Liaoning	0.396[e]	0.2[c]	Chongqing	0.4922[l]	0.1211[l]
Jilin	0.3[c]	0.259[f]	Sichuan	0.45[c]	0.2[c]
Heilongjiang	0.26*	0.5*	Guizhou	0.35[m]	0.2[c]
Shanghai	0.2[c]	0.148*	Yunnan	0.2[c]	0.1*
Jiangsu	0.3[g]	0.225[g]	Xizang	0.338[d]	0.148[d]
Zhejiang	0.3*	0.3*	Shaanxi	0.338[d]	0.159[o]
Anhui	0.29[h]	0.319*	Gansu	0.338[d]	0.159[o]
Fujian	0.3[c]	0.188[i]	Qinghai	0.338[d]	0.159[o]
Jiangxi	0.23*	0.2[c]	Ningxia	0.338[d]	0.159[o]
Shandong	0.45[c]	0.2[c]	Xinjiang	0.143[n]	0.137[n]
Henan	0.3[c]	0.2[c]			

[a] Fang et al. (2015). [b] Zhao (2015). [c] Tian et al. (2011). [d] Bao et al. (2014). [e] Chang et al. (2012). [f] Liu et al. (2010). [g] Wang and Zhao (2011). [h] Qin and Ge (2012). [i] Huang (2014). [j] Liu et al. (2014). [k] Li (2013). [l] Li et al. (2013). [m] Zhang et al. (2015). [n] Hou et al. (2013). [o] EPD (2014). * Result from our questionnaire.

Table 3. Straw-to-product ratio (N_k), dry matter fraction (D_k), and combustion efficiency (CE_k) of crop straw used in this study.

Crops	N_k	D_k^f	CE_k^f
Corn	1.269[a]	0.87	0.92
Wheat	1.3[b]	0.89	0.92
Cotton	3[b]	0.83	0.9
Sugar cane	0.3[c]	0.45	0.68
Potato	0.5[d]	0.45	0.68
Peanut	1.5[b]	0.94	0.82
Rapeseed	1.5[d]	0.83	0.9
Sesame	2.2[d]	0.83	0.9
Sugar beet	0.1[b]	0.45	0.9
Hemp	1.7[e]	0.83	0.9
Rice	1.323[a]	0.89	0.93
Soya bean	1.6[d]	0.91	0.68

[a] Zhang and Zhu (1990). [b] Bi (2010). [c] Han et al. (2002). [d] NATESC (1999). [e] Gao et al. (2009). [f] He et al. (2015).

were collected for each province and vegetation type. The type of vegetation burned in each pixel was determined by the 1 km-resolution MODIS Land Cover product produced by Ran et al. (2010). We considered 10 vegetation types as forest and grassland (i.e. evergreen needleleaf forest, evergreen broadleaf forest, deciduous needleleaf forest, deciduous broadleaf forest, mixed forest, closed shrublands, open shrublands, woody savannas, savannas, and grassland). The FL values employed in this study are listed in Table 4. As for the combustion factor (CF), it has usually been set as a constant in previous literature. In our paper, CF values were collected for each vegetation type, and the CF in each pixel was determined by the MODIS Land Cover product and the CF values of specific vegetation. The CFs of forest, closed shrublands, open shrublands, woody savannas, and grassland were set as 0.25, 0.5, 0.85, 0.4, and 0.95, respectively (Michel et al., 2005; Kasischke et al., 2000; Hurst et al., 1994).

2.2.4 Livestock manure

The mass of biomass burned by animal waste was calculated using Eq. (4) as follows:

$$A = S \times Y \times C \times R, \quad (4)$$

where A is the annual burning mass of livestock manure ($\mathrm{Mg\,year^{-1}}$), S represents the number of each livestock type in pastoral and semi-pastoral areas at the end of the year ($\mathrm{head\,year^{-1}}$), Y is a single livestock annual fecal output per year ($\mathrm{Mg\,head^{-1}}$), C represents livestock manure dry matter fraction, and R is the proportion of total livestock manure directly combusted.

The S values were taken from Chinese governmental annual statistical reports, including EOCAIY (2013) and NBSC (2013c). The Y values were related to the large animals only. Among these large animals, single cattle annual manure output was 10 Mg and single horse annual manure output was 7.3 Mg (Li and Zhao, 2008). The livestock annual manure output of other animals was set at 8 Mg, according to Tian et al. (2011). The C value was set as 18 %

Table 4. Forest and grassland biomass fuel loadings in each province.

Province	Biomass fuel loadings ($g\,m^{-2}$)				
	Needleleaf Forest[a,d]	Broadleaf Forest[a,e]	Mixed Forest[a,f]	Shrublands[b,g]	Grassland[c,h]
Heilongjiang	8140	7610	7875	1387	180
Jilin	9340	10 710	10 025	1387	140
Liaoning	2620	8250	5435	1387	160
Inner Mongolia	8140	4470	6305	1387	90
Gansu	8900	6630	7765	1500	90
Ningxia	6910	6280	6595	1386	50
Qinghai	8800	5430	7115	1545	110
Shaanxi	3730	7550	5640	1442	100
Xinjiang	14 410	3060	8735	1387	70
Tibet	13 990	6490	10 240	2007	60
Beijing	1560	6750	4155	1387	170
Hebei	2480	5150	3815	1388	150
Henan	1550	5560	3555	1388	140
Shandong	1280	5660	3470	1387	130
Shanxi	3640	4790	4215	1387	130
Tianjin	0	6760	3380	1378	160
Anhui	1690	10 360	6025	2447	140
Hubei	1680	8060	4870	1573	160
Hunan	2080	10 650	6365	3471	150
Jiangxi	1820	9370	5595	3699	140
Fujian	2910	9700	6305	3773	160
Guangdong	2060	8970	5515	3702	140
Hainan	4810	9220	7015	3739	150
Jiangsu	2630	5530	4080	1371	120
Shanghai	3060	9250	6155	1371	110
Zhejiang	1710	10 500	6105	3682	160
Chongqing	8090	9900	8995	3010	170
Guangxi	2110	9280	5695	3142	150
Guizhou	2210	11 410	6810	3431	150
Sichuan	8090	9900	8995	3006	170
Yunnan	5760	14 510	10 135	3534	150

[a] Fang et al. (1996, 1998). [b] Hu et al. (2006). [c] Pu et al. (2004); all the biomasses listed here are calculated using the above-ground biomass density. [d] Needleleaf forest includes needleleaf deciduous forest and needleleaf evergreen forest. [e] Broadleaf forest includes broadleaf deciduous forest and broadleaf evergreen forest. [f] Biomass of mixed forest is the mean of needleleaf forest and broadleaf forest. [g] Shrublands include closed shrublands and open shrublands. [h] Grassland includes woody savannas, savannas, and grasslands.

(Tian et al., 2011) and R as 20 % (Li, 2007; Liu and Shen, 2007). Since not all regions use livestock manure in biomass burning, we considered only pastoral and semi-pastoral areas, including Tibet, Inner Mongolia, and Gansu, Xinjiang, and Qinghai provinces in this study (Tian et al., 2011).

2.3 Determination of EFs

In order to ensure the accuracy of the emission inventory as much as possible, it is important to choose the appropriate EF. The EFs used in this study were mainly based on localized measurements. When selecting the EFs, we applied the following principles: first, for a certain type of biomass source or crop type, we prioritized the use of EFs from localized measurements in the literature. Second, for the biomass sources or crop types which lacked localized measurements, we prioritized results from developing foreign countries similar to China above those of developed countries. Third, when localized measurement data of a certain crop type were missing, the average value of the mainstream literature in the foreign country was used as an estimate. After an extensive literature review, the resultant EFs of domestic and open burning for each pollutant and source were summarized and are presented in Tables 5 and 6, respectively.

2.4 Spatial distribution

In order to obtain the detailed spatial distribution characteristics of biomass burning emissions, and to provide grid-based data for the air-quality model simulation, the biomass burning inventory in this study was assigned into $1 \times 1\,km^2$ grid cells based on the source-specific surrogate. We applied

Table 5. Emission factors used in the estimation of domestic biomass burning emissions.

	Material	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
		g kg$^{-1,*}$											ng g$^{-1,*}$
Domestic burning	Corn	1.33[h]	1.86[a,b,h]	7.39[h]	6.87[h]	7.34[h]	0.68[h]	82.37[a,b,h,l,m]	0.95[a]	2.25[a]	1491[b,l]	3.91[b,m]	7.94[n,o]
	Wheat	1.2[a,h]	1.19[a,b,h,l]	8.86[h]	8.24[h]	9.37[h]	0.37[h]	136.46[a,b,h,l,m]	0.42[a]	3.46[f]	1246.7[b,l]	8.3[b]	11.09[n,o]
	Cotton	0.53[d,k,e,g,h]	2.49[a]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	121.7[b,h]	0.82[a]	1.83[a]	963.42[b]	6.08[b]	3.12[n,o]
	Sugar cane	0.53[d,k,e,g,h]	1.12[d,e,f,g]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	121.7[b,h]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	6.5[n,o]
	Potato	0.53[d,k,e,g,h]	1.12[d,e,f,g]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	121.7[b,h]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	6.5[n,o]
	Peanut	0.53[d,k,e,g,h]	1.12[d,e,f,g]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	121.7[b,h]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	4.82[n,o]
	Rapeseed	1.36[h]	1.65[f]	13.73[h]	12.77[h]	7.97[h]	0.52[h]	133.5[f]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	6.5[n,o]
	Sesame	0.53[d,k,e,g,h]	1.12[d,e,f,g]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	121.7[b,h]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	6.5[n,o]
	Sugar beet	0.53[d,k,e,g,h]	1.12[d,e,f,g]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	121.7[b,h]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	6.5[n,o]
	Hemp	0.53[d,k,e,g,h]	1.12[d,e,f,g]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	121.7[b,h]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	6.5[n,o]
	Rice	0.48[h]	1.92[a,b,f,l]	6.88[h]	6.4[h]	8.4[h]	0.52[h]	79.7[a,b,f,h]	0.49[a]	2.01[a]	1147.4[a,b,l]	4.8[b]	5.56[n,o]
	Soya bean	0.53[d,k,e,g,h]	1.12[d,e,f,g]	7.69[h]	7.15[h]	8.82[b,j]	1.3[b,e]	80.7[f]	0.51[d,e,g,c]	2.21[d,e,c]	963.42[b]	6.08[b]	4.48[n,o]
	Feces	0.28[h]	0.58[h]	8.84[h]	7.15[h]	3.13[h]	1.3[h]	19.8[h]	0.53[g]	2.2*	1060[g]	4.14[g]	–
	Firewood	0.4[e,g]	1.49[b,f,h]	5.66[h,i]	5.22[h,d]	3.13[j]	1.3[e]	48.25[b,f,h]	1.49[c]	1.14[c]	1445.2[b,m]	2.48[b,m]	7.2[n,o]

Note that lowercase letters indicate the data source. Sources are from the following:[a] Cao et al. (2008). [b] Wang et al. (2009). [c] Li et al. (2009). [d] Reddy and Venkataraman (2002). [e] Andreae and Merlet (2001). [f] Tang et al. (2014). [g] Tian et al. (2011). [h] EPD (2014). [i] Cao et al. (2004). [j] Wei et al. (2008). [k] Turn et al. (1997). [l] Zhang et al. (2008). [m] Zhang et al. (2000). [n] Chen et al. (2013). [o] W. Zhang et al. (2013). * Unit of emission factor.

Table 6. Emission factors used in the estimation of open biomass burning emissions.

	Material	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
		g kg$^{-1,*}$											ng g$^{-1,*}$
Open burning	Corn	0.44[a,b,c]	4.3[a,b,c]	11.95[c]	11.7[b,c]	10[b]	0.68[b,c]	53[a,c,h]	0.3[b,h]	4.35[b,h]	1350[b,h]	4.4[b]	7.94[k,l]
	Wheat	0.85[a,b,c]	3.3[a,b,c]	7.73[c]	7.58[c]	7.5[b,c]	0.37[b]	55.8[a,b,c,d]	0.37[b,h]	3.9[b,h]	1390[b,h]	3.4[b]	11.09[k,l]
	Cotton	0.53[e,f,b,g]	3.16[e,f,b,g]	6.93[c]	6.79[c]	9.5[c,f,i]	1.3[j]	66.1[b,c,i]	0.42[b]	3.3[b]	1410[b]	3.9[b]	3.12[k,l]
	Cane	0.53[e,f,b,g]	3.16[e,f,b,g]	6.93[c]	6.79[c]	11.02[d]	1[j]	40.08[d]	0.42[b]	3.3[b]	1410[b]	3.9[b]	6.5[k,l]
	Potato	0.53[e,f,b,g]	3.16[e,f,b,g]	6.93[c]	6.79[c]	9.5[c,f,i]	0.53[b,c]	66.1[b,c,i]	0.42[b]	3.3[b]	1410[b]	3.9[b]	6.5[k,l]
	Peanut	0.53[e,f,b,g]	3.16[e,f,b,g]	6.93[c]	6.79[c]	9.5[c,f,i]	0.53[b,c]	66.1[b,c,i]	0.42[b]	3.3[b]	1410[b]	3.9[b]	4.82[k,l]
	Rapeseed	0.53[e,f,b,g]	1.12[g]	6.93[c]	6.79[c]	9.5[c,f,i]	0.53[b,c]	34.3[g]	0.23[g]	1.08[g]	1410[b]	3.9[b]	6.5[k,l]
	Sesame	0.53[e,f,b,g]	3.16[e,f,b,g]	6.93[c]	6.79[c]	9.5[c,f,i]	0.53[b,c]	66.1[b,c,i]	0.42[b]	3.3[b]	1410[b]	3.9[b]	6.5[k,l]
	Beet	0.53[e,f,b,g]	3.16[e,f,b,g]	6.93[c]	6.79[c]	9.5[c,f,i]	0.53[b,c]	66.1[b,c,i]	0.42[b]	3.3[b]	1410[b]	3.9[b]	6.5[k,l]
	Hemp	0.53[e,f,b,g]	3.16[e,f,b,g]	6.93[c]	6.79[c]	9.5[c,f,i]	1.3[j]	66.1[b,c,i]	0.42[b]	3.3[b]	1410[b]	3.9[b]	6.5[k,l]
	Rice	0.53[c]	1.42[c,g]	5.78[c]	5.73[c,g,h]	7.25[c,d]	0.53[b,c]	46.03[d,g,h]	0.16[g,h]	2.03[g,h]	1393[h]	3.9[b]	5.56[k,l]
	Soya bean	0.53[e,f,b,g]	1.08[g]	6.93[c]	6.79[c]	9.5[c,f,i]	0.53[b,c]	32.3[g]	0.13[g]	1.05[g]	1410[b]	3.9[b]	4.48[k,l]
	Evergreen Needleleaf Forest	1[m]	1.8[m]	13.1[o]	12.7[o]	28[n]	3.5[n]	118[n]	0.2[p]	7.8[p]	1514[n]	6[n]	113[q,r]
	Evergreen Broadleaf Forest	0.45[n]	2.6[n]	12.8[o]	10.2[o]	24[n]	0.76[n]	92[n]	0.5[n]	4.7[n]	1643[n]	5.1[n]	113[q,r]
	Deciduous Needleleaf Forest	1[m]	3[m]	13.1[o]	12.7[o]	28[n]	3.5[n]	118[n]	0.2[p]	7.8[p]	1514[n]	6[n]	113[q,r]
	Deciduous Broadleaf Forest	1[m]	1.3[n]	12.8[o]	12.3[o]	11[n]	1.5[n]	102[n]	0.6[n]	9.2[n]	1630[n]	5[n]	113[q,r]
	Mixed Forest	1[m]	1.3[n]	12.8[o]	12.3[o]	14[n]	1.5[n]	102[n]	0.6[n]	9.2[n]	1630[n]	5[n]	113[q,r]
	Closed Shrublands	0.68[n]	3.9[n]	8.5[o]	7.9[o]	4.8[n]	1.2[n]	68[n]	0.5[p]	6.6[p]	1716[n]	2.6[n]	80[q,r]
	Open Shrublands	0.68[n]	3.9[n]	8.5[o]	7.9[o]	4.8[n]	1.2[n]	68[n]	0.5[p]	6.6[p]	1716[n]	2.6[n]	80[q,r]
	Woody Savannas	0.68[n]	3.9[n]	8.5[o]	7.9[o]	4.8[n]	1.2[n]	68[n]	0.5[p]	6.6[p]	1716[n]	2.6[n]	80[q,r]
	Savannas	0.68[n]	2.8[n]	9.9[o]	6.3[o]	9.3[n]	0.5[n]	59[n]	0.4[n]	2.6[n]	1692[n]	1.5[n]	80[q,r]
	Grasslands	0.68[n]	2.8[n]	9.9[o]	6.3[o]	9.3[n]	0.5[n]	59[n]	0.4[n]	2.6[n]	1692[n]	1.5[n]	80[q,r]

Note that lowercase letters indicate the data source. Sources are from the following: [a] Li et al. (2015). [b] Li et al. (2007b). [c] EPD (2014). [d] Y. S. Zhang et al. (2013). [e] Tian et al. (2011). [f] Wang and Zhang (2008). [g] Tang et al. (2014). [h] Ni et al. (2015). [i] Streets et al. (2003). [j] Kanabkaew and Nguyen (2011). [k] Chen et al. (2013). [l] W. Zhang et al. (2013). [m] Andreae and Rosenfeld (2008). [n] Akagi et al. (2011). [o] Song et al. (2009). [p] McMeekin et al. (2008). [q] Friedli et al. (2003). [r] Streets et al. (2005). * Unit of emission factor.

GIS software as the main tool to produce the spatial distribution. In this paper, the approaches used to determine spatial distribution varied between biomass sources; thus, we selected different methods of spatial allocation according to the homologous source characteristics. The regions in which in-field straw burning occurred can be located according to the MODIS fire count data (MOD14/MYD14) (van der Werf et al., 2006; Huang et al., 2012d). Farmland fire point is the spatial surrogate of in-field straw burning. Land use data (MODIS Land cover) were provided by Ran et al. (2010). Detailed descriptions of the MODIS fire count data (MOD14/MYD14) are shown in the Supplement (Sect. S4). As for forest and grassland fires, the emissions of forest and grassland fires were estimated at 500 m resolution, which can be reshaped into a 1 km grid using GIS software. The emissions of straw, firewood, and livestock excrement burning were treated as area sources and the spatial surrogates used to distribute these biomass sources were the population densities of different land use types (e.g. rural population density, grassland population density) (Zheng et al., 2009; Huang et al., 2012b). The population density of different land use types is based on the land use data provided by Ran et al. (2010) and on 1 km-grid population distribution data provided by Fu et al. (2014). Details of the calculation

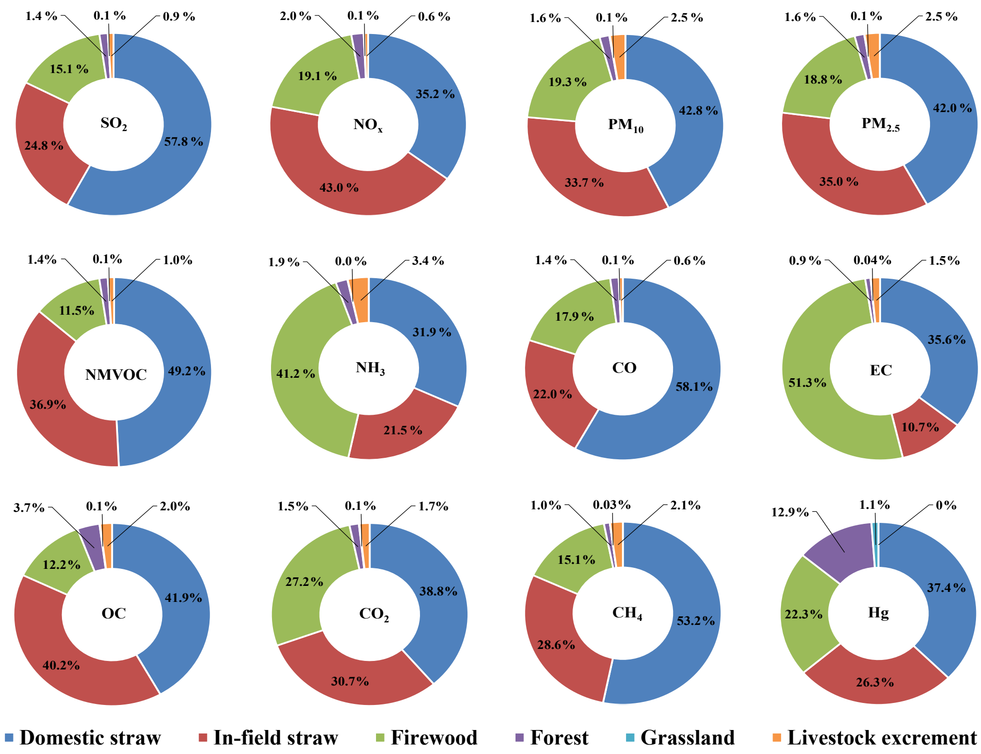

Figure 2. Contributions of different sources to total biomass burning emissions in China in 2012.

method and the equation for the gridded emission are presented in the Supplement (Sect. S4).

2.5 Temporal distribution

According to the temporal resolution of MODIS fire count data (MOD14/MYD14), the monthly/daily emission of in-field straw burning can be estimated based on the number of specific fire points, and the monthly/daily emission of forest and grassland fires can be calculated by the Julian day emission of forest and grassland fires. For domestic biomass source, the monthly emission of each source can be estimated based on the monthly uneven coefficient derived from our survey questionnaire. Details of the survey questionnaire are presented in the Supplement (Sect. S3). The daily domestic emission is equally allocated from the monthly emission.

2.6 Speciation of NMVOCs and $PM_{2.5}$

Detailed speciation of NMVOC and $PM_{2.5}$ emissions is necessary to model gas and aerosol chemistry and to simulate the impact of biomass burning on atmospheric composition, and it has received extensive attention by domestic scholars in recent years (Song et al., 2007; Li et al., 2007b; Liu et al., 2008).

In this study, the species emission was mainly estimated based on the total emission, and the NMVOC and $PM_{2.5}$ source profiles (mass fraction) of biomass sources were collected from the literature. In terms of the data selection, we prioritized domestic measurement for the species as much as possible. Therefore, the NMVOC source profile mainly refers to data from Liu et al. (2008) and from Akagi et al. (2011), including species covering alkane, alkene, alkyne, aromatic, etc. The $PM_{2.5}$ source profile data are cited from the work of Li et al. (2007b) and from Watson et al. (2001), including 36 species, such as element, ion, etc.

3 Results and discussion

3.1 Total emissions in China

3.1.1 Contributions by biomass burning sources

The annual emissions of biomass burning in mainland China are presented in Table 7. The total annual emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and Hg for the Chinese mainland in 2012 are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34 380.4 Gg, 369.7 Gg, 1189.5 Gg, 675 299.0 Gg, 2092.4 Gg, and 4.12 Mg, respectively. The contribution of different sources to the total emissions of various pollutants is shown in Fig. 2, which shows that domestic straw burning, in-field straw burning, and firewood burning are the dominant biomass burning sources, with the total contribution ranging from 86.02 to 97.58 % for various pollutants.

Table 7. Biomass burning emission inventory in the 31 provinces or municipalities of China in 2012.

Province	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
	unit: Gg											unit: Mg
Beijing	0.5	1.9	6.9	6.5	4.8	1.2	58	1.3	1.9	1507	3.1	0.01
Tianjin	1.2	3.1	11.6	10.9	10.4	1.3	116	1.4	3.7	2136	6.6	0.01
Hebei	21.4	52.8	200.2	188.6	178.7	21.4	2023	22.7	65.9	36 308	115.4	0.22
Shanxi	9.4	22.9	83.5	78.8	74.9	8.1	777	8.7	27.5	14 668	43.9	0.09
Inner-Mongolia	16.1	45.2	217.5	204.9	154.5	24.1	1309	16.8	65.3	32 278	103.6	0.16
Liaoning	13.8	40.7	144.3	136.1	128.2	17.4	1277	18.0	42.5	27 369	72.4	0.14
Jilin	16.5	54.3	179.6	171.5	165.6	15.7	1395	14.7	58.3	29 529	84.7	0.16
Heilongjiang	30.0	117.5	397.4	383.3	395.4	32.5	2878	22.8	132.1	65 619	200.7	0.36
Shanghai	0.3	0.8	3.1	3.0	3.6	0.2	33	0.2	1.1	566	2.1	0.00
Jiangsu	14.7	39.7	154.5	146.8	167.0	12.4	1614	10.2	52.6	27 527	102.2	0.16
Zhejiang	3.8	12.4	48.2	45.7	48.2	6.1	451	5.5	13.6	9986	28.6	0.05
Anhui	19.7	56.5	210.1	199.9	209.9	19.7	2046	17.6	71.4	38 539	127.7	0.23
Fujian	3.0	10.7	40.6	38.1	36.4	6.3	387	6.4	10.5	8905	22.9	0.04
Jiangxi	8.0	27.4	105.0	99.0	102.7	14.3	998	13.4	28.6	22 445	62.0	0.10
Shandong	34.7	77.9	304.3	287.4	296.6	25.2	3318	24.9	108.9	50 493	192.2	0.33
Henan	33.1	82.1	313.0	296.5	301.3	26.6	3294	26.0	112.7	52 896	194.5	0.35
Hubei	13.2	40.7	158.2	149.0	147.6	19.9	1530	19.1	44.5	31 167	91.5	0.16
Hunan	15.6	51.6	199.2	187.4	198.0	24.1	1949	22.8	54.0	39 478	118.5	0.19
Guangdong	5.9	21.1	78.9	74.2	70.0	12.8	726	12.5	21.0	17 619	43.4	0.08
Guangxi	8.1	29.1	105.6	100.0	108.0	15.2	1001	12.6	31.8	21 300	61.5	0.11
Hainan	1.4	5.0	19.1	17.9	17.7	3.0	191	2.9	5.1	4032	11.0	0.02
Chongqing	5.5	15.6	61.3	57.4	58.7	7.4	619	7.3	17.0	11 564	35.6	0.06
Sichuan	19.3	53.0	212.1	199.6	206.4	22.1	2115	21.0	63.2	38 192	125.1	0.23
Guizhou	6.4	19.5	74.7	70.1	62.9	10.4	679	10.8	19.8	14 944	38.9	0.08
Yunnan	8.8	27.6	108.7	101.4	90.7	17.2	972	17.4	31.8	22 370	54.0	0.20
Tibet	3.0	15.4	40.6	37.6	24.8	5.4	305	2.4	26.5	7554	14.8	0.30
Shaanxi	8.5	22.2	86.5	81.1	71.6	11.1	832	12.0	25.9	16 701	47.0	0.10
Gansu	6.2	15.9	66.6	62.5	52.6	8.1	579	7.8	20.1	11 814	35.2	0.07
Qinghai	0.9	2.3	10.5	9.8	7.2	1.2	84	0.9	3.6	1683	5.2	0.02
Ningxia	1.6	4.1	14.9	14.1	14.8	1.2	144	1.2	5.1	2515	8.5	0.02
Xinjiang	6.2	21.8	71.5	67.5	64.9	9.8	682	8.5	23.7	13 596	39.5	0.08
Total	336.8	990.7	3728.3	3526.7	3474.2	401.2	34380	369.7	1189.5	675 299	2092.4	4.12

However, the largest contributing sources to different pollutants are not similar. Domestic straw burning is the largest source of biomass burning emissions for SO_2 (57.8 %), PM_{10} (42.8 %), $PM_{2.5}$ (42.0 %), NMVOC (49.2 %), CO (58.1 %), OC (41.9 %), CO_2 (38.8 %), CH_4 (53.2 %), and Hg (37.4 %). This has a direct impact on residents. Moreover, prolonged exposure under high domestic straw burning emission (e.g. SO_2, CO, CH_4, and Hg) can cause many adverse health effects (e.g. acute respiratory infections and chronic bronchitis) (Donohoe and Garner, 2008). The contribution of firewood to each pollutant cannot be neglected, especially for EC (51.3 %) and NH_3 (41.2 %). According to the localized measurement of EF by Li et al. (2009), the EF_{EC} for firewood (1.49 g kg^{-1}) is 3.5 times that of the average for in-field straw (0.43 g kg^{-1}). The EF_{NH_3} of firewood is larger than the average of various straws. This results in a large contribution by firewood for these two pollutants. The contribution of domestic and in-field straw burning to NO_x, PM_{10}, $PM_{2.5}$, NMVOC, Hg, OC, and CO_2 is nearly equal. Straw burning has an important influence on indoor air quality and outdoor atmospheric environment.

In addition to the sources mentioned above, the contribution of livestock excrement burning and of forest and grassland fires is relatively small. It is mainly due to the small amount of biomass consumption. The biomass fuel consumptions of these three biomass sources are 10 614, 6647, and 505 Gg, respectively, which are significantly lower than that of domestic straw burning (201 582 Gg), in-field straw burning (147 178 Gg), and firewood burning (127 250 Gg). The contributions of livestock excrement burning to PM_{10}, $PM_{2.5}$, NH_3, EC, OC, CO_2, and CH_4 are 2.52, 2.47, 3.44, 1.52, 1.96, 1.67, and 2.10 %, respectively. The contribution of forest and grassland fires to biomass burning emissions for most pollutants in China is small (0.9–3.7 %), except for the contribution of forest fires to Hg emissions (14.0 %).

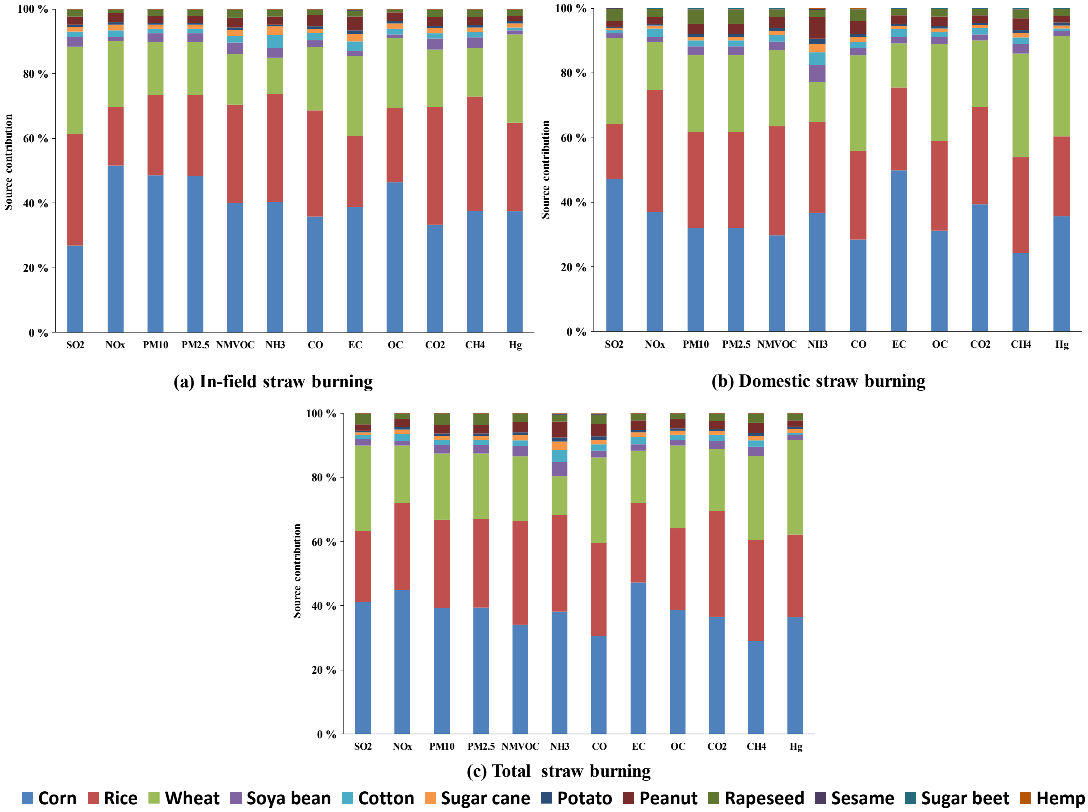

Figure 3. Contributions of 12 crop straw types for various pollutants in China in 2012.

3.1.2 Contributions by various crop straw

As mentioned in Sect. 3.1.1, straw burning is the most important biomass burning source with considerable influence on the pollutants that most strongly impact air quality, climate, and human health. Therefore, the contribution of major crop straw types was analysed. Figure 3 shows the contributions of 12 different crop straw types for various pollutants in 2012 for mainland China. Figure 3c indicates that corn, rice, and wheat straw are the major crop straws burned as fuel and as waste in China. The contribution is more than 80 % of the total straw burning emissions of all pollutants studied in this paper. Corn, rice, and wheat are the major three food crops in China with a large planting area (the output of these three kinds of grain accounts for 70 % of the total grain output in China; NBSC, 2013c), resulting in a large amount of straw production. Among the various crops, corn straw burning has the largest contribution to all of the pollutants except for CH_4. Rice straw burning is the largest contributor to CH_4 and the second largest contributor to other pollutants, except for SO_2, OC, and Hg. Wheat straw burning is the second largest contributor to SO_2, OC, and Hg and the third largest contributor to other pollutants. Compared to the three kinds of crops mentioned above, the total contribution of soya bean, cotton, sugar cane, potato, peanut, and rapeseed straw burning to the various pollutants is relatively small, accounting for 8.1–19.2 % of the total emissions for all pollutants considered; the contributions of sesame, sugar beet, and hemp straw burning to various pollutants are negligible, never exceeding 0.5 %. In addition, Fig. 3a and b show the contribution of each type of straw burning emission to the in-field and domestic straw burning emission, respectively. Similar to what is shown in Fig. 3c, corn, rice, and wheat straw are the main contributors whether for in-field or domestic burning emission. However, the dominant contributors of certain pollutants are different in in-field and domestic straw burning: for SO_2 and CO_2, rice straw is the largest contributor to in-field straw burning emission, while corn straw is the largest contributor to domestic straw burning emission; for NO_x and VOC, corn straw contributes most to the in-field straw burning emission, while rice straw contributes most to the domestic straw burning emission; for CO and CH_4, corn straw has the largest contribution to in-field straw burning emission, while wheat straw has the largest contribution to domestic straw burning emission.

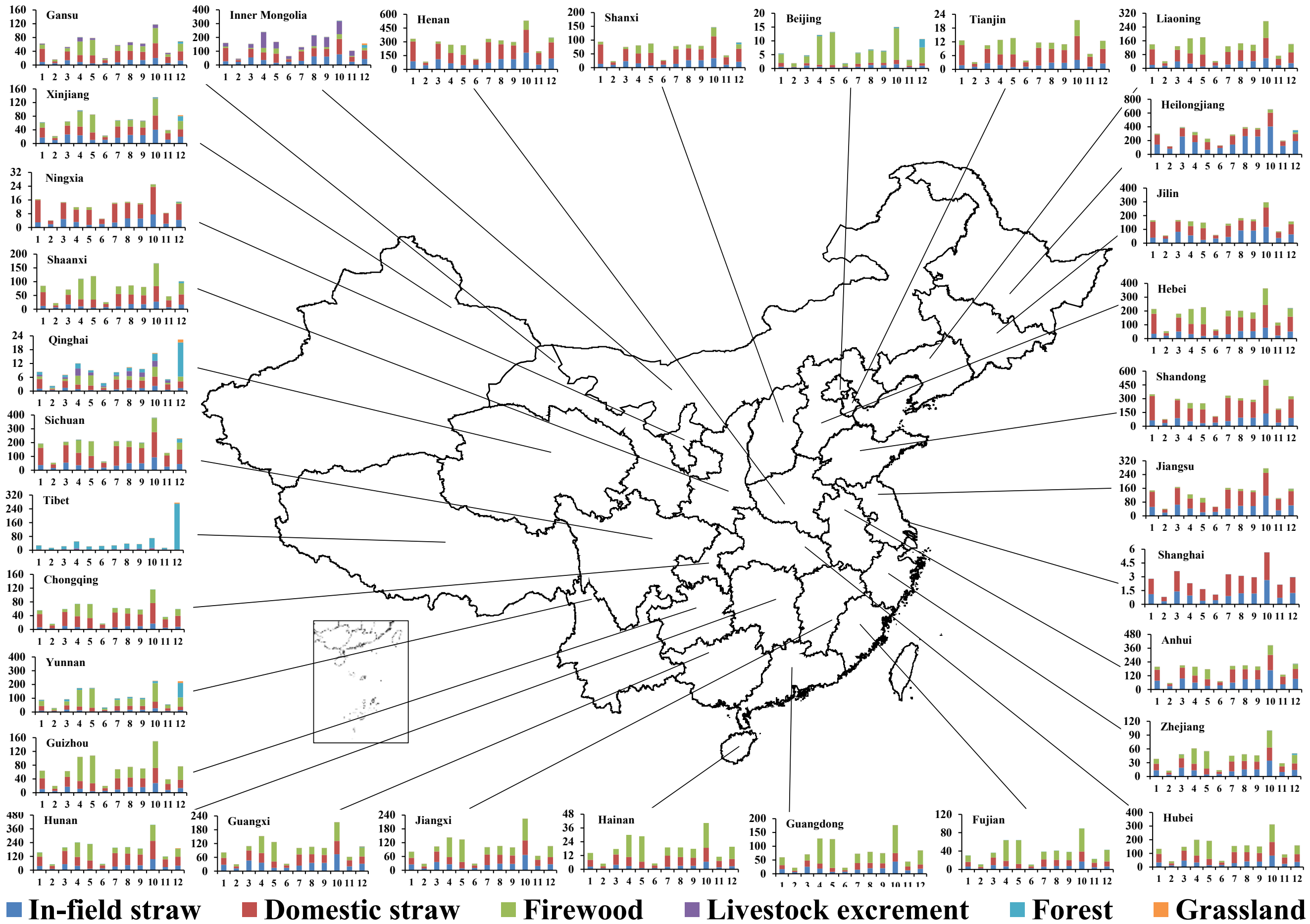

Figure 4. Contributions of different biomass sources to the emission in each province (Gg). Note that numbers 1–12 represent the pollutants of $SO_2 \times 10$, NO_x, NMVOC, $NH_3 \times 10$, EC × 10, OC, CO/10, PM_{10}, $PM_{2.5}$, $CO_2/100$, CH_4, and Hg × 1 000 000, respectively.

3.2 Emissions from different regions

3.2.1 Total emissions for different provinces

The total biomass burning emissions in 31 provinces in 2012 are presented in Table 7. These results indicate that Heilongjiang, Shandong, Henan, Hubei, Anhui, Sichuan, Jilin, Inner Mongolia, Hunan, and Jiangsu provinces are the major contributors, with the total emission contributions ranging from 53 to 65 % for various pollutants. The province with the highest contribution to total emissions of NO_x, PM_{10}, $PM_{2.5}$, NMVOC, NH_3, OC, CH_4, Hg, and CO_2 is Heilongjiang, while Shandong province has the highest emissions of SO_2, CO, and EC. This could be attributed to different types of biomass consumption in each province due to geographical location, climatic conditions, and population density. Detailed discussion of the contribution by biomass source and crop straw type of different regions is provided below.

3.2.2 Contributions by biomass sources of each province

The emission of detailed biomass sources of each province is presented in Fig. 4. The provinces with major contributions to total pollutant emissions for each biomass source are various. Straw burning emissions are mainly distributed in Shandong, Henan, Heilongjiang, Hebei, Anhui, Sichuan, Jilin, and Hunan provinces. The total contribution of these provinces to various pollutants is more than 58 %. This is due to the large amount of cultivated land in the northern plain region, as cultivated land in this region prioritizes economic crops that produce rich straw resources. Several regions in which firewood produces large emissions are Hunan, Yunnan, Hubei, Hebei, Sichuan, Guangdong, Shaanxi, Liaoning, and Jiangxi provinces. More than 54 % of firewood burning emissions is contributed by these provinces. These areas are mainly distributed in southern China, a mountainous region in which the forest cover is greater than 30 % (NBSC, 2013c). Livestock excrement burning emissions are mainly distributed in Tibet, Inner Mongolia, Gansu, Xinjiang, and

Qinghai provinces, since livestock manure is burned as fuel only in pastoral and semi-pastoral areas in China. Emissions from forest and grassland fires are mainly distributed in Tibet, Yunnan, Heilongjiang, Xinjiang, Inner Mongolia, and Sichuan provinces, owing to the high vegetation cover and climatic conditions in these areas.

The contribution of biomass sources to total emissions in each province is also distinct. Straw burning has a large contribution to various pollutant emissions in Heilongjiang (79–97 %), Ningxia (87–98 %), Shandong (74–95 %), Jilin (74–95 %), Henan (61–93 %), Anhui (51–91 %), and Shanxi (61–90 %) provinces. The economic income of the rural areas in these provinces is relatively low. A large amount of straw is consumed as the main non-commodity source of energy. In addition, firewood resources are scarce in these areas and, as a result, the usage of straw is very high. Figure 4 also indicates that, for most provinces (e.g. Beijing, Tianjin, and Hebei), the contribution of domestic straw burning is greater than that of in-field straw burning. This is mainly attributable to the gradual response to the prohibition of burning straw and to the introduction of straw resource utilization measures. The emission contribution of in-field straw burning is higher than that of domestic straw burning in Hebei, Heilongjiang, and Anhui provinces. This suggests that measures undertaken to prohibit the burning of straw in these provinces still need to be strengthened. Several regions in which firewood burning produces a large component of total emissions of various pollutants are Beijing (47–90 %), Guangdong (31–83 %), Yunan (31–79 %), Fujian (30–81 %), Hainan (26–77 %), and Guizhou (27–74 %) provinces. The quantity of straw in the rural areas of these provinces is relatively low. Firewood is the main non-commodity energy used by rural people. It is worth noting that although the biomass fuel consumption in Beijing is small, compared to the straw burning emission contribution (9–41 %), firewood burning emission (47–90 %) represents a large proportion of the total biomass burning in Beijing. This is mainly due to the severe restriction of in-field straw burning. Firewood has gradually replaced straw as the main non-commodity biomass energy source in suburban Beijing in recent years (Wang, 2010; Liu, 2012). In addition, Tibet and Inner Mongolia are the major provinces where livestock excrement produces a large component of total pollutant emissions. Less crop straw and little firewood is used as a fuel source and thus livestock excrement makes a large contribution to total biomass emissions in these provinces. Forest and grassland fires have a small contribution to pollutant emissions in each province. The contribution of Hg emission by forest fires in Inner Mongolia, Sichuan, Yunnan, Qinghai, Tibet, and Xinjiang provinces is considerable (exceeding 10 %) and mainly due to the high EF of Hg for forest fires.

3.2.3 Contributions from different crop straws of each province

As the largest biomass source, crop straw burning represents a major contribution to the total emissions from biomass burning. The 12 different types of straw burning emission of each province were further analysed and the results are presented in Fig. 5. The corn straw burning emission is concentrated in Heilongjiang, Shandong, Inner Mongolia, Hebei, Henan, Shanxi, and Sichuan provinces, with the total contribution reaching more than 72 %. Wheat crop straw emissions are mainly distributed in Henan, Shandong, Anhui, Hebei, Jiangsu, Sichuan, Shaanxi, Hubei, and Shanxi provinces. More than 89 % of the wheat crop straw burning emission is contributed by these provinces. Rice crop straw burning emissions are mainly distributed in Heilongjiang, Hunan, Jiangsu, Sichuan, Anhui, Hubei, Guangxi, Guangdong, and Zhejiang provinces, with the total contribution amounting to more than 71 %. Water conditions, light, and heat are better for the cultivation of rice in southern China, while low temperature, long sunshine duration, and the large temperature difference between day and night are suitable for wheat growing in northern China. In addition, soya bean, cotton, sugar cane, potato, peanut, and rape straw have a small contribution to the various pollutants, and these straws are mainly distributed in Heilongjiang, Xinjiang, Guangxi, Sichuan, Henan, and Sichuan provinces, respectively.

3.2.4 Emissions intensity at county resolution

At county resolution, we found that the spatial distributions of emissions for various pollutants are similar, and we thus took $PM_{2.5}$ as an example to analyse the emission intensity (e.g. per unit area, per capita) at county resolution. Figure 6a shows the county-level geographic distribution of $PM_{2.5}$ emissions in 2836 counties or districts. The distribution of county-level annual $PM_{2.5}$ emissions is shown in Fig. 6d. The spatial diversity of various counties emission is obvious. There are 406 districts without biomass burning, because they are mainly distributed in the urban areas of developed cities, such as the Dongcheng and Xicheng districts in Beijing and the Jing'an district in Shanghai. The total emissions of 32.3 % of districts and counties (917) in China were less than 0.25 Gg. The cumulative frequency analysis result indicates that the emissions in most of the counties (i.e. more than 90 %) were less than 4.0 Gg, including the regions with low crop yields or scarce population. The emissions of 30.9 % of the total districts and counties (875) were more than the average emission across all counties (1.245 Gg). The two largest emissions (approximately 16 Gg) appeared in Longjiang and Wuchang, which are major grain-producing counties in Heilongjiang province.

Figure 6b shows the $PM_{2.5}$ emission intensities per unit area. Most of the high values (more than 3 Mg km^{-2} $year^{-1}$) mainly appeared in the northern and central regions of China

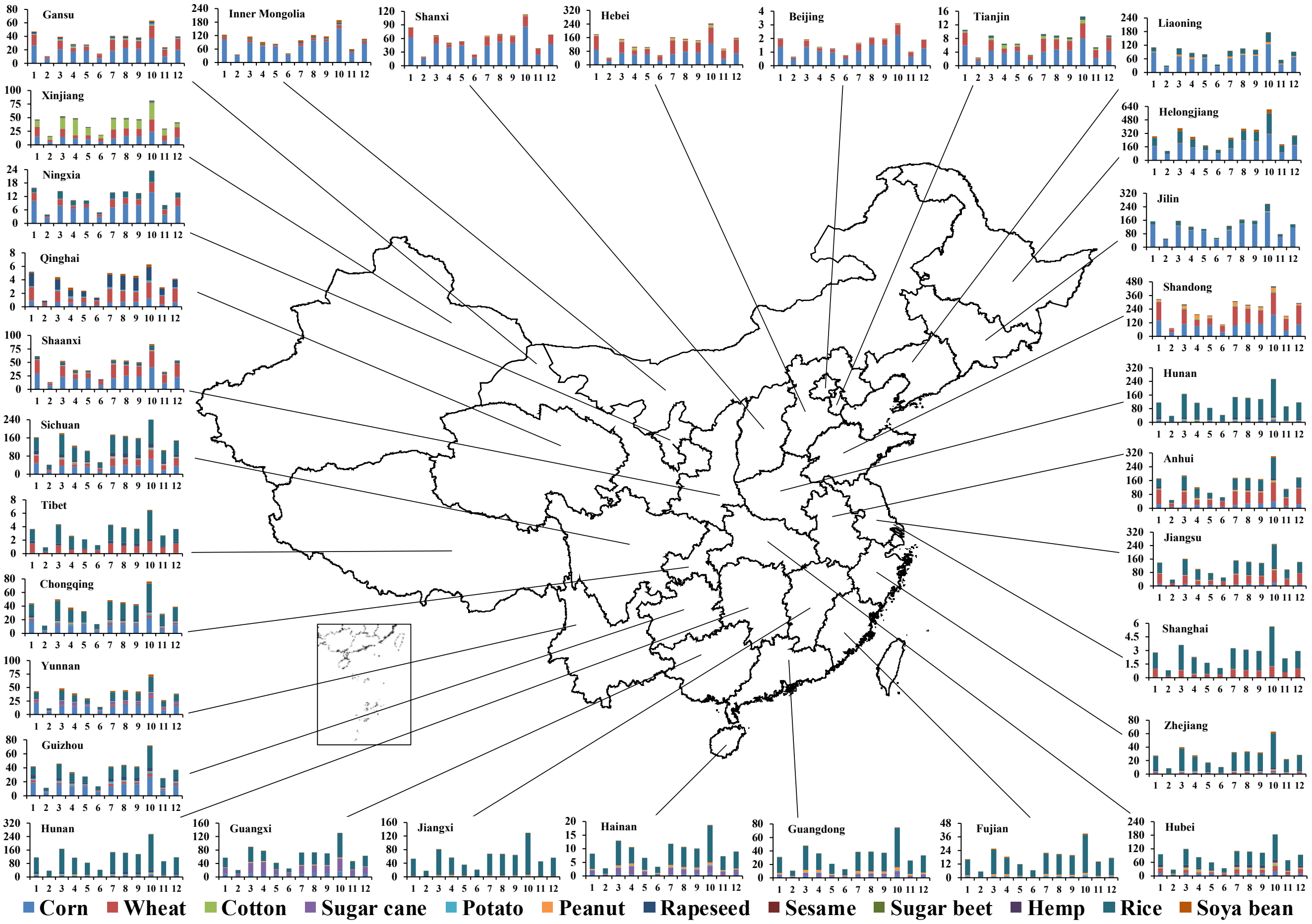

Figure 5. Contributions of different crop straw types to the emission in each province (Gg). Note that numbers 1–12 represent the pollutants of $SO_2 \times 10$, NO_x, NMVOC, $NH_3 \times 10$, EC $\times$ 10, OC, CO/10, PM_{10}, $PM_{2.5}$, $CO_2/100$, CH_4, and Hg $\times$ 1 000 000, respectively.

(e.g. Hebei, Jiangsu, Shandong, Anhui, Jiangxi, and Hunan), where the land is relatively flat and gives priority to agricultural activity, with a substantial amount of crop straw produced from a relatively small area. The greatest numbers of counties with low intensity are concentrated in Tibet, Qinghai, and Xinjiang provinces. In addition, it could be found that some rural counties in Heilongjiang, Jilin, and Liaoning provinces show substantial emissions, but relatively lower intensity (e.g. Nenjiang in Heilongjiang, Dunhua in Jilin, and Chaoyang in Liaoning) due to the large area of these counties.

$PM_{2.5}$ emission intensities per capita are illustrated in Fig. 6c. Because of the diversity of population density and biomass energy utilization, the emissions intensities per capita among various counties present obvious differences. The counties with emission intensities of more than 10 kg cap^{-1} year^{-1} are mainly distributed in Heilongjiang, Jilin, Tibet, and Sichuan provinces. The high emission intensities in north-eastern China are mainly attributed to the large amount of biomass burning emissions from straw and firewood burning. The high emission intensities in south-western China occur mainly because these regions are less economically developed (depending on non-commercial energy sources such as straw and firewood) and are prone to forest and grassland fires. Moreover, the populations of these regions are relatively small. The counties with lower emission intensities per capita compared to other provinces, concentrated in Henan, Guangdong, and Shanxi provinces, are attributed to the high numbers of people living there.

3.3 Spatial distribution of biomass burning emissions

As the pollutants all showed a similar emission distribution, $PM_{2.5}$ was taken as an example to discuss the grid emission distribution. Figure 7 shows the $1 \times 1\,km^2$ grid distribution, and illustrates that high biomass emissions are distributed in Henan, Heilongjiang, Shandong, Anhui, Hebei, and Sichuan provinces; these high-emission areas are mainly scattered in the major agricultural regions of north-eastern to central-southern China, showing a zonal distribution. The biomass burning emissions are concentrated in the regions with great agricultural and rural activity and low economic

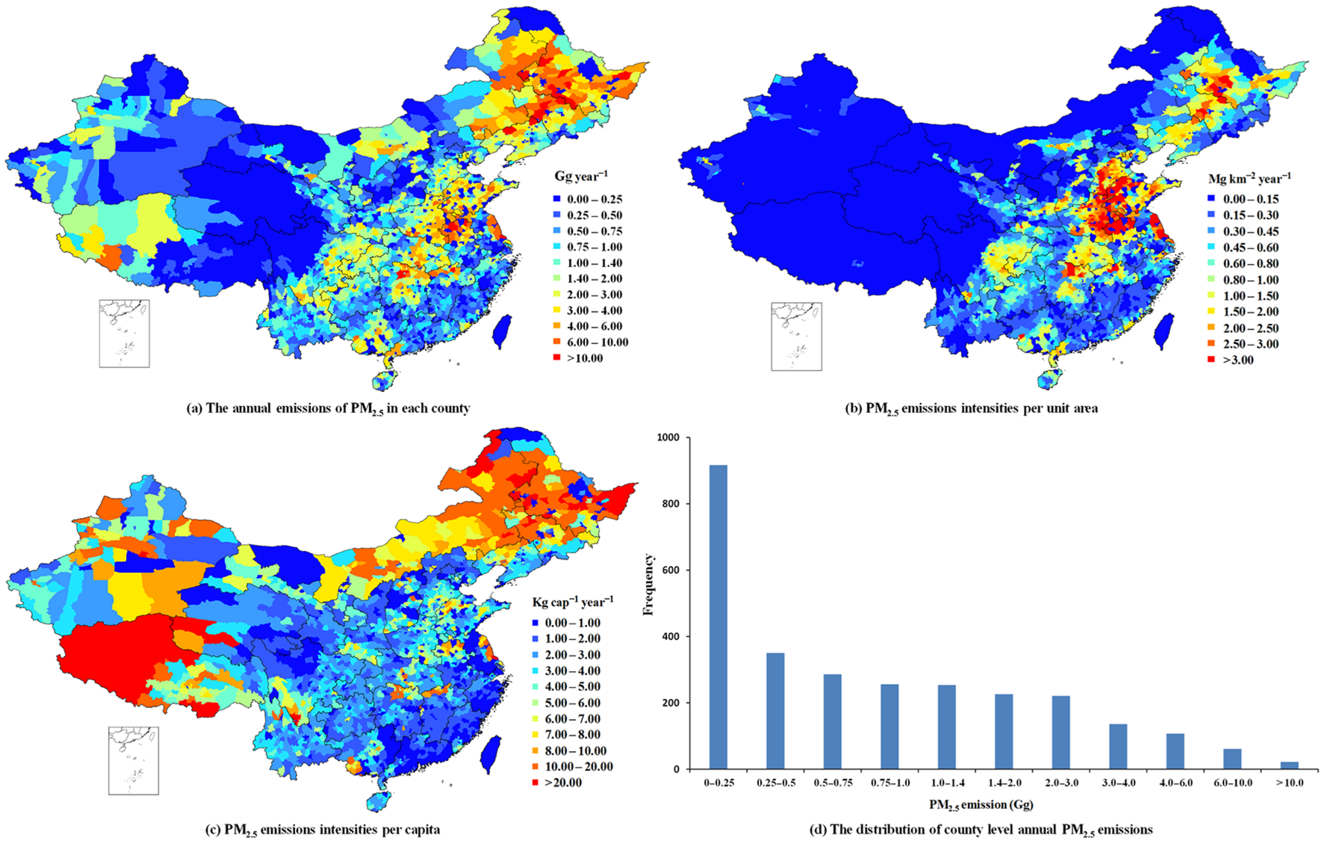

Figure 6. Biomass emission inventory at county resolution and intensity (PM$_{2.5}$).

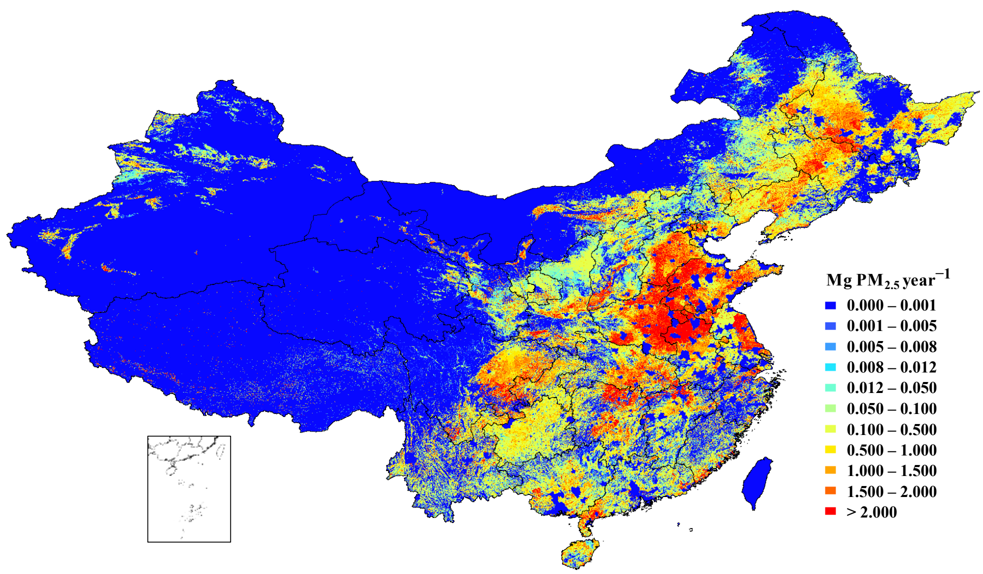

Figure 7. Gridded distribution of PM$_{2.5}$ annual emissions.

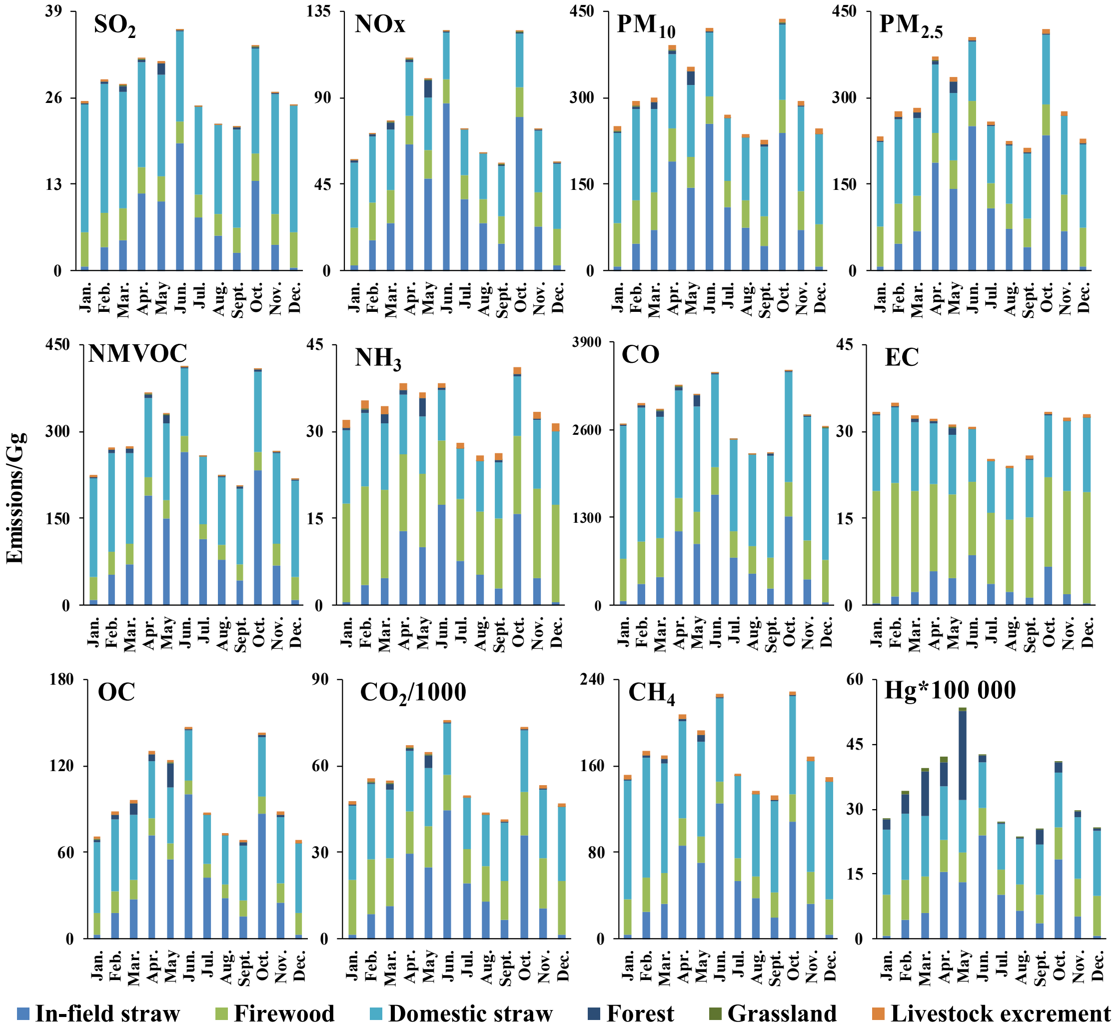

Figure 8. Monthly variation of different biomass sources emission for each pollutant.

income. These regions are characterized by dense population, abundant cultivated areas, and tree resources. Low emissions are mainly distributed in parts of south-western China, in the north-western regions, and in downtown areas of the majority of urban areas. The scarce population and crop yields in parts of south-western China and of the north-west areas, and low agricultural activity in downtown areas, result in low emissions. In particular, some urban areas in the North China Plain are surrounded by suburban and rural areas, and the main fuel used in these urban areas is commodity energy. Besides, there is no agricultural activity in the fields. Therefore, a small amount of biomass burning emissions are produced by these areas. However, error will be introduced in grid emissions if they are allocated from the emission inventory at coarse preliminary resolution (e.g. provincial or prefectural resolution before spatial allocation) based on the gridded surrogates (e.g. rural population). Consequently, gridded emissions, which were obtained through spatial allocation from the emission inventory at county resolution, could better represent the actual situation.

3.4 Temporal variation in biomass burning emission

Figure 8 shows the monthly emission of all 12 pollutants considered, indicating that there are different monthly emission variations for each pollutant. The pollutants showing large monthly variations were SO_2, NO_x, PM_{10}, OC, NMVOC, and $PM_{2.5}$. The in-field burning of crop residue mainly occurred in the harvest season, thus showing the obvious monthly variation features. The sources of NH_3, CO, and EC emissions are dominated by straw and firewood domestic burning, and the contributions of these two kinds of sources to the total emissions of these pollutants are 73.1, 75.9, and 86.9 %, respectively. The temporal distribution of these two sources was more uniform compared to in-field straw burning at the monthly scale, and thus monthly emissions of these three pollutants showed less temporal distinction. In addi-

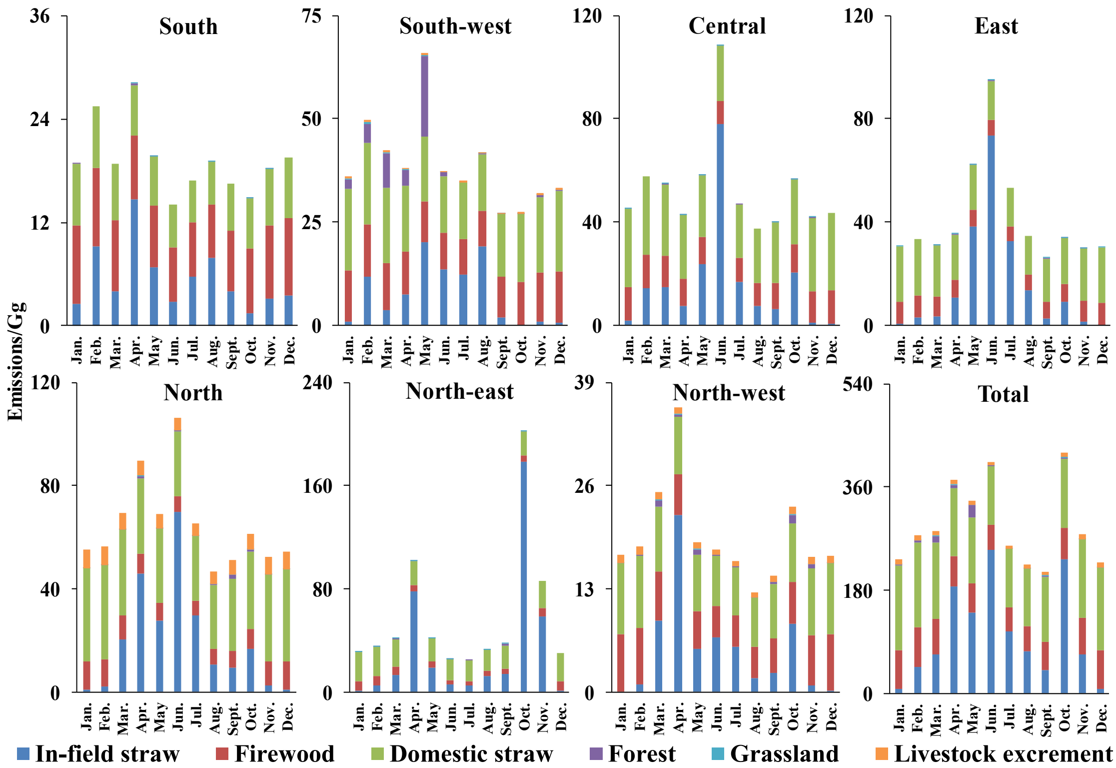

Figure 9. Monthly variation of different biomass sources emission for $PM_{2.5}$ emissions in different regions.

tion, the overall trends of emissions for other pollutants show a certain similarity: April, May, June, and October are the top four months with high emissions, mainly due to in-field straw burning. The total emission of these months account for 65 % of the emissions from in-field straw burning, while, as for EC, the emissions in January, February, October, November, and December are relatively higher than in other months due to domestic biomass burning in the cold season.

Burning activity mainly occurs in the harvest season (in-field straw burning) or crop-sowing season (clearing the cultivated land and increasing the soil fertility for the next sowing), and it varies by burning habit in different regions. In addition, the sowing and harvest seasons vary in different regions because of climatic conditions. Because of the differences in burning activity and climatic conditions in various regions, monthly emission features vary regionally, so to consider this, we divided China into seven areas, again taking $PM_{2.5}$ as an example to analyse the pollutant emission characteristics (Fig. 9). Regions located in southern China (including Fujian, Guangdong, Hainan, and Guangxi provinces) and south-western China (including Chongqing, Sichuan, Guizhou, Yunnan, and Tibet provinces) have climates that are highly suited to arable agriculture because of sufficient temperatures and abundant rainfall. As indicated by Fig. 9, for the southern regions, there are three relatively higher in-field straw burning emissions occurring in February, April, and August rather than in other months. These periods are consistent with local sowing and harvest times in the southern regions. The crops in these areas are sown earlier than in northern areas because of the climatic differences. February, April, and August are the sowing season for beans and the harvest seasons of first- and second-round crops (e.g. rice), respectively (CAAS, 1984; MOA, 2000). For the south-western region, the emission peaks are mainly distributed in February, May, and August, which differs from the southern regions due to the inclusion of May, owing to the burning of rapeseed straw and large emissions from forest fires.

For the central region (including Henan, Hubei, and Hunan provinces), the main crops are winter wheat and summer corn, and the harvest seasons of these two crops are the end of May and the end of September (MOA, 2000), respectively. The peak emissions in the eastern region (including Shanghai, Jiangsu, Zhejiang, Anhui, and Jiangxi provinces) are mainly distributed from May to July, and May, June, and July are the harvest seasons of rapeseed, wheat, and rice in the eastern regions, respectively. The northern plains of China (including Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia, and Shandong provinces) include the largest agricultural area in the country, accounting for 34 % of the rural population, 27 % of the farmland, and 35 % of the harvest crops (NBSC, 2013c). These regions differ from the eastern and central parts firstly in the usage of firewood, since there firewood

Table 8. Uncertainty ranges of different pollutants in emission estimates (min, max). Unit for emission estimate is Gg.

Pollutant	Emission estimate	Uncertainty ranges*	Previous study (Streets et al., 2003)
SO_2	337	(−54, 54 %)	(−245, 245 %)
NO_x	991	(−37, 37 %)	(−220, 220 %)
PM_{10}	3728	(−7, 6 %)	
$PM_{2.5}$	3527	(−13, 1 %)	
NMVOC	3474	(−9, 9 %)	(−210, 210 %)
NH_3	401	(−49, 48 %)	(−240, 240 %)
CO	34 380	(−4 %, 4 %)	(−250 %, 250 %)
EC	370	(−61, 61 %)	(−430, 430 %)
OC	1190	(−20, 19 %)	(−420, 420 %)
CO_2	675 299	(−3, 3 %)	
CH_4	2092	(−9, 9 %)	(−195, 195 %)
Hg	0.00412	(−31, 32 %)	

* 95 % confidence interval.

is also used as heating energy and therefore the consumption of firewood in winter is greater than in summer. In addition, for in-field straw burning, northern winter wheat and corn are mainly harvested in June and October, respectively. April and May are the sowing seasons of spring rice and soya beans. The north-eastern region (including Liaoning, Jilin, and Heilongjiang provinces) shows high values in October, April, and November. The high value in April was a result of burning activity. The peak in October was mainly due to the harvesting of corn, and November is the harvest season for rice. In the north-western region (including Shaanxi, Gansu, Qinghai, Ningxia, and Xinjiang provinces), the peaks in March, April and October are mainly due to burning activities for the next corn sowing, wheat sowing, and corn harvesting seasons, respectively.

Furthermore, the daily $PM_{2.5}$ emissions were estimated according to the monthly emissions and the biomass sources' daily non-uniformity coefficient, both of which are shown in the Supplement (Fig. S3). It could be found that the main emission peaks appeared in early April, early June, and over the entire month of October. This is due to (1) burning activities for the next sowing in the southern, south-western, and north-eastern regions; (2) the harvest season of winter wheat in the central, eastern, and northern regions; and (3) the harvest season of corn in the central, north-eastern, and north-western regions.

3.5 Emissions of $PM_{2.5}$ and NMVOC species

Total $PM_{2.5}$ emission from biomass burning in this study was 3527 Gg. According to our calculation based on the method described in Sect. 2.6, OC was the largest contributor of $PM_{2.5}$, accounting for 33.7 % of total emissions. Cl^-, EC, K^+, NH_4^+, K, and SO_4^{2-} are also the major species of $PM_{2.5}$, and the contribution of these species was 46.63 %. Additionally, there are several species that have less emission (e.g. Al, Si, and Mg). Detailed $PM_{2.5}$ component emissions are presented in the Supplement (Fig. S4).

The total NMVOC emission in this study was 3474 Gg. The alkenes are the major contributor of biomass burning NMVOC emissions. The contribution of alkenes to the total NMVOC emission was approximately 34 %, more than that of alkane (28 %), aromatics (24 %), alkynes (13 %), and others (1%). Among these species, ethylene, acetylene, propylene, and 1-butylene are the major species of alkenes and alkynes, with the total contribution accounting for 40.1 %. Ethane, *n*-propane, *n*-butane, and *n*-dodecane are the main species of alkanes, with the total contribution accounting for 14.0 %. Benzene, toluene, styrene, mp-xylene, and ethyl benzene are the major species of aromatics, with a total contribution of 16.6 %. Several species mentioned above are key for the formation of secondary air pollution, such as ethylene, propylene, toluene, mp-xylene, and ethyl benzene (Huang et al., 2011). This illustrates that biomass burning emission control is urgently needed for air-quality improvement. Detailed NMVOC species emissions are shown in the Supplement (Fig. S5).

3.6 Uncertainties in biomass burning emission estimates

The Monte Carlo method is used to analyse the uncertainty of this emission inventory, as it has been used in uncertainty estimation for many inventory studies (e.g. Streets et al., 2003; Zhao et al., 2011, 2012). Activity data (Zheng et al., 2009) and EFs (Zhao et al., 2011) are assumed to be normal distributions. The coefficients of variation (CV, the standard deviation divided by the mean) of activity data and EFs were obtained from the literature. The CVs of activity data for firewood and straw burning were set as 20 % (Zhao et al., 2011; Ni et al., 2015). As the source of activity data for livestock excrement is the same as that for crop straw burning (i.e.

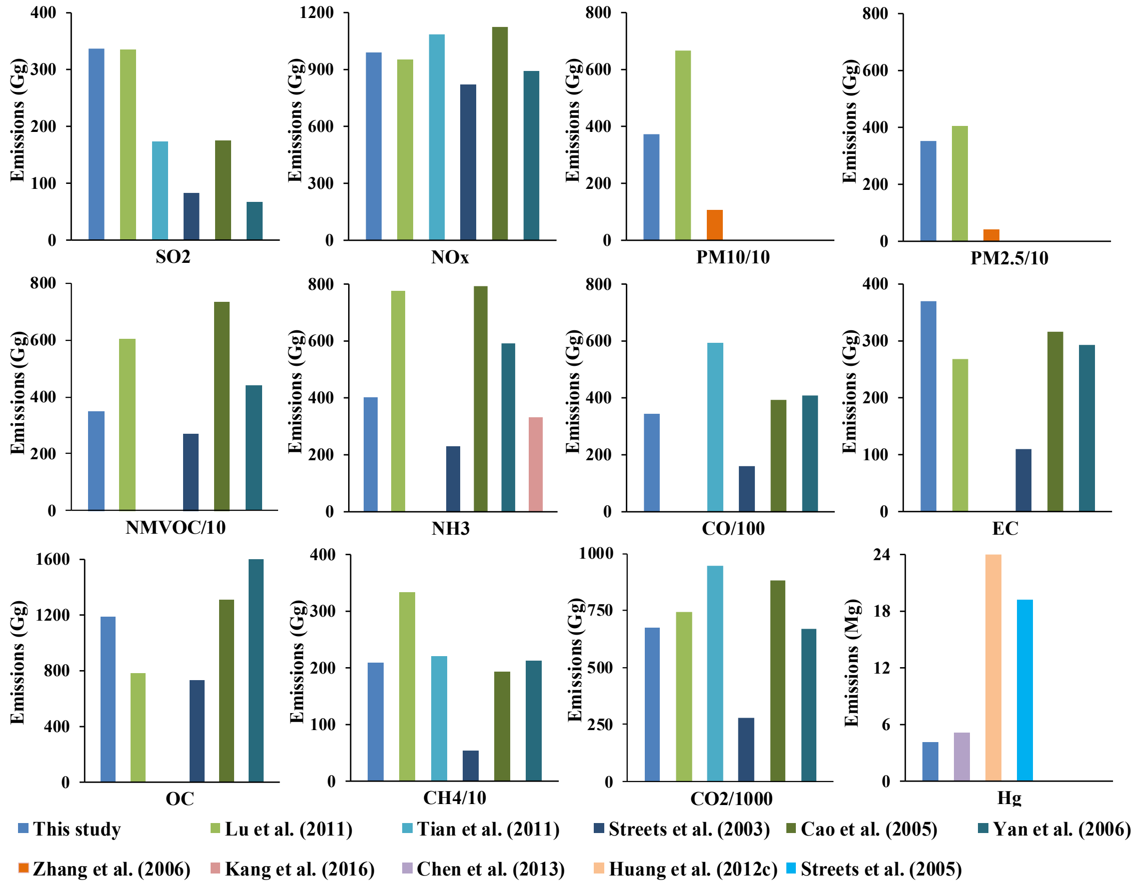

Figure 10. Comparison of the emissions inventory derived by this study with the emissions estimated by previous studies.

government statistical data), the CV was also set as 20 %. MCD64A1-burned data products have been shown to be reliable in large fires (Giglio et al., 2013), and the CV of burned area of forest and grassland fires is from the reported standard deviation (Giglio et al., 2010). The biomass fuel loadings (Saatchi et al., 2011; Shi et al., 2015) and combustion factors (van der Werf et al., 2010) of forest and grassland fires were within a CV of approximately 50 %. The CVs of the EFs for each pollutant for each biomass burning type are shown in the Supplement (Sects. S8 and S9). The range of emissions was calculated by averaging 20 000 Monte Carlo simulations with a 95 % confidence interval. From the perspective of source, the uncertainty of forest fires (ranging from −624 to 631 % for all pollutants) is the highest, following by grassland burning (ranging from −378 to 290 % for all pollutants), livestock excrement burning (ranging from −300 to 295 % for all pollutants), and firewood burning (ranging from −189 to 188 % for all pollutants). The uncertainty of crop straw burning (ranging from −114 to 114 % for all pollutants) is the smallest. Uncertainty ranges of different pollutants in emission estimation are presented in Table 8. The total uncertainties of SO_2, NH_3, and EC are large compared to other pollutants. The total uncertainty for emissions of these pollutants are (−54, 54 %), (−49, 48 %), and (−61, 61 %), respectively. For NH_3, EC, and SO_2, the highest uncertainties exist in livestock excrement burning and in forest and grassland fires. Large uncertainties exist in the EFs used in emission estimation of livestock excrement burning, which is mainly due to the lack of localized measurements of EFs. The large uncertainties in forest and grassland fire emission are due to the uncertainty of biomass fuel loadings and combustion factors used in the estimation. The detailed activity data could also reduce the uncertainty in the emission inventory to some extent because they could better reflect the actual situation. In spite of the uncertainty existing in this study our emission inventory is relatively reliable due to the selection of localized EFs and detailed activity data.

3.7 Comparison to other studies

In this paper, the national biomass burning emission inventories published after 2000 have been compared to those produced by our study (see Fig. 10). It could be found that the relatively high difference (range from −80 to 366 % for various pollutants) occurs between our estimation and the results of earlier studies (e.g. papers published before 2006) due to economic development and EF localization. Compared to recent studies, the SO_2, NO_x, $PM_{2.5}$, EC, and OC emissions of our estimation are close to those derived from Lu et al. (2011), with the difference ranging from −34 to 15 %, while the PM_{10}, NMVOC, CH_4, and NH_3 emissions in this study are lower than those of Lu et al. (2011). The EFs of PM_{10}, NMVOC, CH_4, and NH_3 for various crop types used in this study are generally lower than the EFs without specific crop types in Lu et al. (2011). The SO_2, NO_x, CH_4, and CO_2 emissions in this study are close to those in Tian et al. (2011), with the difference ranging from −49 to 40 %. The difference in CO emission is relatively high. The major emission differences of domestic straw burning, in-field straw burning, and firewood burning between our paper and that of Tian et al. (2011) are −78, −17, and −122 %, respectively. The reason for this is also the selection of EFs. Our localized EFs for crop and firewood burning are lower than the EFs reported in Tian et al. (2011). In addition, for NH_3 emission, compared to the earlier studies, our estimation is close to that derived from recent research (Kang et al., 2016). The difference is less than 17 %. For Hg emissions, our estimation is lower than that of Huang et al. (2012c), but is close to that of Chen et al. (2013). The EF of Hg is classified by stems and leaves (40 and 100 $ng\,g^{-1}$ for firewood; 35 and 319 $ng\,g^{-1}$ for in-field straws) in Huang et al. (2012c), which is higher than the localized EF classified by specific crop (the mean EF is 6.08 $ng\,g^{-1}$) and firewood (7.2 $ng\,g^{-1}$).

4 Conclusions

In this study, a comprehensive biomass burning emission inventory with high spatial and temporal resolution was developed for mainland China for 2012, based on county-level activity data, satellite data, and updated source-specific EFs. The emission inventory includes domestic and in-field straw burning, firewood and livestock excrement burning, and burning by forest and grassland fires. The total annual emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and Hg are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34 380.4 Gg, 369.7 Gg, 1189.5 Gg, 675 299.0 Gg, 2092.4 Gg, and 4.12 Mg, respectively.

Domestic straw burning, in-field straw burning, and firewood burning are the major biomass burning sources, while the largest contributing source to various pollutants is different. Domestic straw burning contributes most to all of the pollutants considered, except for NO_x, NH_3, and EC emission; firewood contributes most to EC and NH_3 emission; and in-field straw burning is the largest contributor of NO_x. In terms of crop straw burning, corn, rice, and wheat straw are the major crop types, with the total contribution exceeding 80 % for each pollutant of straw burning emissions. Corn straw burning has the greatest contribution to EC, NO_x, and SO_2 emissions; rice and wheat straw burning have the second and the third greatest contributions to most of the pollutants considered, respectively. Straw burning emissions are concentrated in agricultural provinces. Firewood burning emissions are mainly distributed in the southern regions of China, where tree resources are abundant. Corn and wheat straw burning emission is mainly distributed in northern China, while rice straw burning emission is concentrated in southern China. Gridded emission results indicate that high emissions are concentrated in the north-eastern and central-southern regions of China, which have more agricultural and rural activity. It is also illustrated that gridded emissions, which were obtained through spatial allocation from the emission inventory at county resolution instead of province or prefecture resolution, could better reflect the actual situation. Monthly distributions reveal that the high emissions in April, May, June, and October were mainly due to the burning activity before sowing and harvesting of main crops. Regional differences in temporal distribution are attributed to the diversity of main planted crops and the climatic conditions in each region. OC, Cl^-, EC, K^+, NH_4^+, K, and SO_4^{2-} are the major $PM_{2.5}$ species, with a total contribution of 80 %. Several species with high contribution to NMVOCs (e.g. ethylene, propylene, toluene, mp-xylene, and ethyl benzene) are key species for the formation of secondary air pollution. Comparison to other studies shows that the emission inventory in this study is relatively reliable. The detailed emission inventory given by this paper could provide detailed information to support further biomass burning pollution research and the development of a targeted control strategy for all regions of the Chinese mainland.

EF and speciation of chemical species are the key parameters in emission estimation. More localized EFs of different biomass fuel types within diverse burning conditions and more detailed $PM_{2.5}$ and NMVOC source profiles that contain as many components as possible are still needed for future studies. In addition, high-temporal-resolution (e.g. hourly resolution) satellite data are necessary to provide hourly emission information for the numerical simulation of biomass burning pollution research and effective control.

5 Data availability

The emission data are archived at Key Laboratory of Beijing on Regional Air Pollution Control and are available on request (y.zhou@bjut.edu.cn).

The Supplement related to this article is available online at doi:10.5194/acp-17-2839-2017-supplement.

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

Acknowledgements. The MODIS Thermal Anomalies/Fire products were provided by the Land Process Distributed Active Archive Center (LPDAAC). The China Land Cover product and the 1 km population distribution data set was provided by the National Science & Technology Infrastructure of China, National Earth System Science Data Sharing Infrastructure (http://www.geodata.cn). This study was supported by the Natural Sciences Foundation of China (Grant No. 91644110 and 51408014), the National Science and Technology Support Project of China (No. 2014BAC23B02).

Edited by: G. Frost
Reviewed by: two anonymous referees

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