emissions of organic carbon (OC), elemental carbon (EC), methane ($CH_4$), nitric oxide ($NO_x$), non-methane volatile organic compounds (NMVOCs), sulfur dioxide ($SO_2$), ammonia ($NH_3$), carbon monoxide (CO), carbon dioxide ($CO_2$) and fine particles ($PM_{2.5}$) were $3.64\times10^3$, $2.87\times10^2$, $3.05\times10^3$, $1.82\times10^3$, $6.4\times10^3$, $2.12\times10^2$, $4.67\times10^2$, $4.59\times10^4$, $9.39\times10^5$ and $4.13\times10^3$ Gg in these years, respectively. Crop straw burning was the largest contributor for all pollutant emissions, by 84%-96%. For the forest, shrubland and grassland fire burning, forest fire burning emissions contributed the most and emissions from grassland fire was negligible due to few grass coverage in this region. High pollutant emissions concentrated in the connection area of Shandong, Henan, Jiangsu and Anhui, with emission intensity higher than 100 ton per square kilometers, which was related to the frequent agricultural activities in these regions. Peak emission of pollutants occurred in summer and autumn harvest periods including May, June, September and October, at which ~50% of the total pollutants emission were emitted in these months . This study highlights the importance in controlling the crop straw burning emissions. From December to March, the crop residue burning emissions decreased, while the emissions from forest, shrubland and grassland exhibited their highest values, leading to another small peak emissions of pollutants. Obvious regional differences in seasonal variations of OBB were observed due to different local biomass types and environmental conditions. Rural population, agricultural output, economic levels, local burning habits, social customs and management policies were all influencing factors for OBB emissions.

## 1. Introduction

Open biomass burning (OBB), which includes forest, shrubland, grassland and crop residue fire burning (van der Werf et al., 2010; Qiu et al., 2016), is one of the most important sources for gaseous and particulate matter (PM) especially for fine particulate particles ($PM_{2.5}$) and associated carbonaceous aerosols (elemental carbon (EC) and organic carbon (OC)) (Zha, 2013; Yan et al., 2014; Zong et al., 2016; Zhou et al., 2017). Previous studies have shown that the OBB contributed to approximately 40% of the annual average submicron EC emission and 65% of primary OC emission globally (Bond et al., 2013), and contributed more than 45% of $PM_{2.5}$ concentration on days of heavy air pollution (Deng, 2011). The pollutants with high emission amounts from OBB posed significant impacts on regional and global climate change, air quality and human health (Seiler and Crutzen, 1980; Crutzen and Andreae, 1990; Andreae and Merlet, 2001; Bond et al., 2004; Akagi et al., 2011; Zhang et al., 2016).

From 1970s (Crutzen et al., 1979), emission estimation of biomass burning has been a research hot topic from global (Seiler and Crutzen et al., 1980; Levine, 1995; Liousse et al., 1995; Bond et al., 2004; Randerson et al., 2012; Kaiser et al., 2012) to regional scale (Yevich and Logan, 2003; Chang et al., 2010; Liousse et al., 2010; Li et al., 2017). China is suffering from severe air pollution with hundred million tons of biomass open burned each year (Zhang et al., 2015). The quantitative estimation of pollutants emission for the whole China (Streets et al., 2003; Tian et al., 2002; Cao et al., 2005; Zhou et al., 2017) or a certain region (Liu et al., 2015; Zhou et al., 2015; Jin et al., 2017) is also a vital practice, which is the base for assessing the impact of OBB on air regional quality deterioration. The Central and Eastern China (CEC), including the Central China (Hunan, Henan and Hubei) and the Eastern China (part of the North Plain of China (Shandong), the Yangtze River delta (YRD, including Zhejiang, Jiangsu, Anhui and Shanghai) and part of the Pan-Pearl River delta (Fujian and Jiangxi)) (Figure 1), is an area with plenty of vegetation coverage (as listed in Figure S1 of Supplementary File). Yin et al (2017) have indicated that the crop residue fire burning in summer harvest time can lead to the increase of $PM_{2.5}$ concentration in China's middle-east region. As one of the most heavily polluted regions in China (Chang et al., 2009; Fu et al., 2013), many large cities are included in this region, such as Nanjing, Wuhan, Shanghai and Hangzhou. Former studies have highlighted the role of OBB on worsening air quality regionally or at megacities, especially for crop residue burning at harvest periods (Yamaji et al., 2010; Zhu et al., 2010; Yin et al., 2011; Huang et al., 2012b; Su et al., 2012; Cheng et al., 2014; Zhou et al., 2016; Zhang et al., 2017).

Previous studies mainly focused on crop residue burning emissions with relatively low spatial and temporal resolution (Yamaji et al., 2010; Huang et al., 2012b), which may limit its adoption in air quality modeling to give an accurate result. An accurate estimation of monthly emissions from OBB with a long timescale and high spatial resolution is still limited. It should be noted that, the OBB activities owned spatial-temporal variation properties and have changed greatly during the last two decades in China, especially for forestland fire burning (Huang et al., 2011) and crop residue burning, considering the implementation of related policies (Table S1 and Table S2). As a big agricultural country, the Chinese government has placed a high priority on environmental pollution prevention caused by OBB. From 1965 to 2015, 51 management documents for crop straw have been formulated and 34 documents were intensively issued after 2008 (Chen et al., 2016). Up to now, few studies have accurately estimated the biomass burning emissions in a long time period (Fu et al., 2013; Cheng et al., 2014). The role of the pollution prevention policies on the spatial-temporal variation of pollutants emitted needs to be better clarified.

In addition, most previous studies have adopted the top-down method (Seiler and Crutzen et al., 1980) to estimate the OBB emission by national or provincial statistical data and then the total emission amounts of pollutants were re-allocated in grids by population, land cover area, or even equal sharing, which is one of the key reasons for the high uncertainties of OBB emission inventories (Streets et al., 2003; Klimont and Streets., 2007; Gadde et al., 2009; He et al., 2013; Zhou et al., 2015; Zhou et al., 2017). Quantitative estimation of biomass burning was highly improved by the satellite observations of fire burned area or active burning fires (Freitas et al., 2005; Wooster et al., 2005; Roy et al., 2008; Giglio et al., 2008; Roy et al., 2008; Reid et al., 2009; Sofiev et al., 2009; Giglio et al., 2010; Liousse et al., 2010; Huang et al., 2012; Li et al., 2016). The improvement of spatial-temporal distribution evolution was achieved by active fire products (e.g., the AVHRR fire count product (Setzer and Pereira, 1991), MODIS active fire satellite products (Cooke et al., 1996) and VIRS fire count product (Ito et al., 2007)). The burned area detection was improved by burned area products (e.g., GBA2000 product (Ito and Penner, 2004; Korontzi, 2005), MODIS burned area dataset (Ito et al., 2007) and Global Fire Emissions Database (GFED) (Randerson et al., 2012)). However, satellite observation also exhibited weakness in estimating fire burning emissions (Duncan et al., 2003; He et al., 2015). One is the burned area product, which provides fire burned areas of the whole month. It is limited by the lower pixel resolutions. The size of many small burn scars is below the detection limit of these products (Eva and Lambin, 1998; Laris, 2005; McCarty et al., 2009; Roy and Boschetti, 2009). Therefore, the contribution of small fires to fire burned area and corresponding fire burning emissions are still poorly understood (Randerson et al., 2012). The other is the active fire product, which can provides information on small fire locations, occurrence time and small fire burned area (Prins and Menzel, 1992; Giglio et al., 2006; Chuvieco et al., 2008; Roberts et al., 2009; Aragao and Shimabukuro, 2010; Bowman et al., 2011; Lin et al., 2012; Arino et al., 2012). The uncertainty of fire detection is mainly due to the limitation of satellite overpass periods. To reduce the uncertainty of emission estimation by satellite products, the combination of two satellite dataset has been proved to be an effective practice recently (Qiu et al., 2016).

The lack of local biomass data (biomass loading data and vegetation speciation data) and local emission factors could introduce uncertainty in emission estimates. Currently, local biomass loading data need to be updated and accurately measured. Local high spatial-resolution vegetation speciation data has been rarely adopted in OBB estimations. Meanwhile, a lot of researches about OBB have used the same emission factors for pollutants emitted from OBB without considering the various biomass species and combustion conditions (Andela et al., 2013; Giglio et al., 2013). All these should be considered and improved in the establishment of OBB emission inventory.

In this study, the multiple satellite data (MCD14 ML and MCD64Al), local high spatial-resolution of vegetation speciation data, updated local biomass loading data, local emission factors and survey results were used to estimate multi-year OBB emissions from 2003 to 2015 in CEC. High spatial-temporal resolution of emission allocation was achieved. The possible driving factors like local habits, social customs, rural population, economic level, agricultural production, energy and pollution control policies which may impact the spatial distribution and temporal variation of OBB emissions were explored. They have been overlooked in previous studies (Song et al., 2009; Chen et al., 2013; Shi et al., 2015). The results here will provide scientific evidence for policy making on controlling OBB emission and modeling its regional impact on air quality, climate and human health. The methods are also helpful for other regions for OBB emission estimation.

## 2. Methods

### 2.1 Estimation of burned areas

OBB emissions in CEC were initially estimated based on the local biomass data (biomass loading data and vegetation speciation data), satellite burned area data (Figure S2) and emission factors. Fire burning emission amount was calculated by the following equation (Wiedinmyer et al., 2011; Shi et al, 2015).

$$E_{i,x,t} = \sum_{j=1}^{n} BA_{x,t} \times CE_{x} \times BL_{x} \times EF_{i,j} \qquad (1)$$

where j stands the different aggregated vegetation types; i stands for different pollutant species; $E_{i,x,t}$ is the emission amount of pollutant i in location x and month t; $BA_{x,t}$ is the total burned area ($km^2$) of aggregated vegetation class in location x and month t; $CE_x$ is defined as the combustion efficiency in location x; $BL_x$ is the biomass fuel loading (kg) in location x; $EF_{i,j}$ is the emission factor of pollutant specie i for vegetation type j.

MODIS burned area product (MCD64AL: http://modis-fire.umd.edu/) and MODIS active fire product (MCD14 ML: https://earthdata.nasa.gov/faq#ed-firms-faq) were combined to obtain accurate open biomass burned area data. MCD64Al had a 500 m spatial resolution and monthly temporal resolution, which could accurately detect the burning area at 500 m pixel. A much lower pixel resolution burning was difficult to detect by this satellite. Therefore, we used MODIS active fire product MCD14 ML as a supplemental tool to obtain the small fire burned area. The active fire detection method based on thermal anomalies could detect fires as low as 1/20 of a pixel. We resampled the two fire products data into 1 km×1 km grid. The total burned area in each grid cell was estimated by the following equation (Randerson et al., 2012).

$$BA_{total(i,t,j)} = BA_{MCD64AL(i,t,j)} + BA_{sf(i,t,j)} \qquad (2)$$

where $BA_{total(i,t,j)}$ is the total fire burned area in grid cell i, month t and aggregated vegetation class j; $BA_{MCD64AL(i,t,j)}$ is the MCD64AL burned area in grid cell i, month t and aggregated vegetation class j; $BA_{sf(i,t,j)}$ is the small fire burned area in grid cell i, month t and aggregated vegetation class j.

$BA_{MCD64AL(i,t,j)}$ was directly detected from MCD64AL product. MCD14ML active fire points in each grid included two parts: active fires points with or near MCD64A1 burned area ($FC_{in}$) and active fires outside the MCD64AL burning area ($FC_{out}$). $BA_{sf(i,t,j)}$ was the burned area of $FC_{out}$. The $BA_{sf(i,t,j)}$ was used as supplement. Due to the active fire product existed as the fire points and could not directly obtain the burned area data, the burned area of small fire was estimated based on the following method (Randerson et al., 2012).

$$BA_{sf\ (i,\ t,\ j)} = FC_{out\ (i,\ t,\ j)} \times \alpha_{(r,\ s,\ j)} \times \gamma_{(r,\ s,\ j)} \quad (3)$$

where $BA_{sf(i,t,j)}$ is the small fire burned area of $F_{out}$ in grid cell i, month t and aggregated vegetation class j; $FC_{out(i,t,j)}$ is the total number of MCD14 ML active fires outside of the burned area in grid cell i, month t and aggregated vegetation class j; α is the ratio of $BA_{MCD64A1}$ to $F_{in}$ and α is equal to the value of surrounding grid cell if $BA_{MCD64A1}$ is equal to 0; γ is an additional unit less scalar which indicates the difference between $F_{in}$ and $F_{out}$ and γ is assumed equal to 1 in this research; r denotes the burning region; s indicates the burning period.

## 2.2 Biomass fuel loading

For forestland, most previous studies used the forest biomass loading data from Fang et al (1996). The forest biomass loading data in recent years need to be updated. In this study, the forest loading data between 2003 and 2008 was collected from Fang et al (1996). From 2008-2015, the forest loading data was calculated based on the $8^{th}$ Chinese National Forest Resource Inventory (Xu, 2014). The forest biomass density data (Table 1) was estimated by the following equation:

$$B_{i,\ r} = T_{i,\ r} / A_{i,\ r} \quad (4)$$

where i stands for different forest species (broadleaf forest, coniferous forest and mixed forest); r means each province; $B_{i,r}$ is the biomass density of forest specie i in province r; $T_{i,r}$ means the total biomass of forest specie i in province r; $A_{i,r}$ denotes the total area of forest specie i in province r.

The total biomass of different forest species was calculated based on the forest stock volume method as follows (Fang et al., 1996):

$$T_{i,\ r} = \sum_{j=1}^{n} E_{j,\ r} = \sum_{j=1}^{n} (aV_{j,\ r} + b) \quad (5)$$

where j stands for different tree types of forest specie i; $E_{j,r}$ means the biomass of different tree type j in province r; $V_{j,r}$ indicates the forest stock volume of different tree type j in province r; a and b are set as correlation coefficient.

The correlation coefficient “a” and “b” for different tree types were derived from previous studies (Fang et al., 1996; Tian et al., 2011; Lu et al., 2012; Li et al., 2014; Wang et al., 2014; Wen et al., 2014) (Table 2). $A_{j,r}$ and $V_{j,r}$ was collected from the $8^{th}$ Chinese National Forest Continuous Inventory. As shown in Table 1, the forest biomass density in recent years has changed a lot, which highlighted the importance of the updation for improving the emission estimation.

For grassland and shrubland, local biomass density data were collected (Pu et al, 2004; Hu et al, 2006)in Table 1. To determine the accurate provincial amounts of crop residue burning, we gathered the production of different species of crops from the China Statistical Yearbook (NBSC, 2004-2016). Detailed data of crop residue to production ratio (dry matter) were collected from local statistical data (Table 3) and the updated data for crop straw burned ratio were derived from survey results (Table 4). Using the updated biomass data, the accuracy of the estimation of OBB emission is expected to be improved.

## 2.3 Combustion efficiency

In previous studies (Wang et al., 2008; Tian et al., 2011), the combustion efficiency (CE) of OBB was mainly set as a constant, which may bias the emission estimation. To improve the accuracy, for cropland, the CE was set as 0.68 for soya bean and 0.93 for other types (Koopmans and Koppejan, 1997; Wang and Zhang, 2008; Zhang et al., 2011). For forest, shrubland and grassland, the CE of fires at each grid cell was assumed as a function of forest cover of corresponding grid cell (Ito et al, 2004; Wiedinmyer et al, 2006). If areas with tree coverage exceeding 60%, the CE for woody and herbaceous cover was set as 0.3 and 0.9, respectively; the CE was set as 0 and 0.98 for woody and herbaceous cover with tree coverage less than 40%; for 40-60% tree cover of fires, the CE was defined as 0.3 for woody fuels and the calculation of herbaceous areas was referred to the following equation:

$$CE_s = e^{-0.13\times TB} \quad (6)$$

where TB stands for the percent tree cover for fires in each grid cell.

It should be noted that though we improved the selection of CE values for different biomass burning types by reviewing literatures, the CE value should not be a constant during burning and the pollution emissions were not uniform in different burning phase, such as smoldering (Kondo et al., 2011) and flaming burning (Burling et al., 2010). Emission inventory in this research and currently published papers (Wang and Zhang, 2008; Zhang et al., 2011; Lu et al., 2011) were estimated for a long time period or a whole year with the timescale as month, instead of hour. Therefore, the CE values used here reflected the average biomass burning condition. In the future, for researches on developing emission inventory with hourly or daily resolution, corresponding high time-resolution activity data and emission factors for different burning stages should be considered.

## 2.4 Emission Factors

Emission factors (EFs) of different OBB were summarized in Table 5. EFs for cropland burning were mainly collected from previous researches carried out in CEC (Tang et al., 2014). As the lack of EFs research on some crop species conducted in CEC and forest, grassland and shrubland conducted in China, EFs were collected from previous researches (Cao et al., 2008; Wang et al., 2008; Akagi et al., 2011; He et al., 2015). In addition, some emission factors measured by our research group in CEC were included in this study.

**2.5 Spatial and temporal allocation**

In order to estimate high spatial resolution of OBB emission in CEC, a high resolution vegetation map (1:1 000 000) (Figure S1) together with the burned area of every open biomass species was used. All the data were relocated into a 1 km×1 km grid to identify and estimate spatial variations of OBB emission. The monthly distribution of OBB emissions were estimated based on the monthly burned area of different vegetation cover types.

The emission in t-th grid was calculated by the following equation:

$$E_{t,j} = BA_{t,j} / BA_{i,j} \times E_{i,j} \quad (7)$$

Where j means different biomass species; i denotes different provinces; $E_{t,j}$ is the emission of different biomass specie j in t-th grid; $BA_{t,j}$ is the burned area in t-th grid cell; $BA_{i,j}$ is the total burn area of different vegetation types in province i; $E_{i,j}$ is the total emission amounts from OBB in province i.

**2.6 Other factors influencing OBB emission**

Several detailed statistics data in the NBSC were collected, such as rural population, per capita net income of rural residents, agricultural output and forestry output in each province and each year. They may impact the OBB emission. Correlation analysis between the OBB emissions and these influencing factors were conducted. Rural population data in 2003, 2004 and 2010 were lack as the detailed data was not reported in NBSC.

**2.7 Uncertainty analysis**

The Monte Carlo method together with the crystal software was used to evaluate the estimation uncertainty quantitatively for all the pollutants. Pollutant emissions were estimated from 20,000 Monte Carlo simulations with a 95% coincidence interval.

**3. Results and Discussion**

**3.1 Accumulated pollutants emission from OBB in CEC**

Table 6 presented the cumulative OBB emission amounts during 2003-2015 and multi-year emissions of different provinces were detailedly listed in Table S3. By the end of 2015, the cumulative emissions of OC, EC, $CH_4$, $NO_x$, NMVOCs, $SO_2$, $NH_3$, CO, $CO_2$ and $PM_{2.5}$ were $3.64\times10^3$, $2.87\times10^2$, $3.05\times10^3$, $1.82\times10^3$, $6.4\times10^3$, $2.12\times10^2$, $4.67\times10^2$, $4.59\times10^4$, $9.39\times10^5$ and $4.13\times10^3$ Gg, respectively. For better revealing the spatial-temporal variation of OBB emissions, the $PM_{2.5}$ variation was detailedly discussed as an example. From 2013 to 2015, the highest emission amounts of $PM_{2.5}$ were found in Henan and Shandong, accounting for 27.93% and 24.35% of the total emission amounts, respectively. The lowest emission appeared in Zhejiang and Shanghai, which only contributed for 4.05% and 0.43%. For other provinces, Hunan, Hubei, Fujian, Anhui, Jiangxi and Jiangsu accounted from 5.52% to 10.13% of the whole emission.

The contributions of different biomass burning types for various pollutants were shown in Figure 2a. Cropland burning contributed the most emission for all the pollutants, by84%-96%. The forest fire exhibited higher emission of $NH_3$, $SO_2$, NMVOCs and $PM_{2.5}$, accounting for 12%, 11%, 7% and 5% of corresponding total emission, respectively. As shown in Figure 2b, for cropland, wheat, corn and rice straw burning were the top three emission source types for all the pollutants. Corn straw burning contributed the most to $SO_2$ (48%), $NO_x$ (37%), NMVOCs (33%), CO (32%) and $CO_2$ (28%) emission. Highest contributions of EC (45%), OC (33%) and $CH_4$ (32%) from rice straw burning was found, while wheat straw burning contributed the most (31%) to $PM_{2.5}$ emission.

In Figure 3, except for Fujian, cropland burning emission was the largest contributor to$PM_{2.5}$ emission, with the contributions ranging from 75.25% (Jiangxi) to almost 100% (Shanghai). The higher rural agglomeration, abundant crop production and more crop residue burning activities in these provinces can explain the higher contributions. Shanghai is one of the most developed cities in China. The highest contribution of cropland burning is not related with its high levels of agricultural activities, but is due to the lack of emissions from other open biomass burning sources. Highest contribution from the forest fire burning and shrubland fire burning were found in Fujian as 45.29% and in Jiangxi as 23.95%, respectively. For forest fire burning, the southern provinces (Fujian, Zhejiang, Jiangxi, Hunan, Hubei and Anhui) exhibited higher values, varying from 3.66% (Hubei) to 38.3% (Fujian) and for shrubland fire burning, the contributions varied from 1.5% (Hubei) to 7.23% (Zhejiang). The relative high emission contributions of forest and shrubland fire burning in the southern provinces can be explained by the large forest and shrubland coverage, frequent human forestry activities, low precipitation and dry weather in spring and winter (Cao et al., 2015), which may easily lead to forest and shurbland fires. While for the northern provinces (Shandong, Henan and Jiangsu), the contributions ranged around 0.76%-1.97%, which can be neglected. $PM_{2.5}$ emission from grassland in CEC was negligible with the following provinces holding higher contributions: Jiangxi (0.8%), Hunan (0.25%), Fujian (0.11%) and Anhui (0.1%).

From Figure 4, emissions from wheat and corn straw burning mainly concentrated in Shandong and Henan (totally accounting for 82% and 78% of the whole emissions, respectively) and the rice straw burning exhibited higher concentrations in Hunan, Jiangxi and Hubei provinces, by 25%, 18% and 16%, respectively. The total contributions of rapeseed, cotton, potato and peanut straw burning to the $PM_{2.5}$ emission were relatively small, accounting for 21%-24% of the total emissions. Most emissions from cotton, peanut and potato straw burning located in Shandong (totally accounting for 35%, 35% and 20%) and Henan (totally accounting for 19%, 40% and 15%). Hubei (32%) and Hunan (31%) were the major provinces for rapeseed straw burning emissions. In addition, emissions from soya bean, sugar cane, tobacco, sesame and sugar beet straw burning were negligible, which never exceeded 1% of total crop residue burning emission in this study.

## 3.2 Temporal variation and spatial distribution for OBB emissions in CEC

### 3.2.1 Yearly variation

Multi-year emissions of OBB from 2003 to 2015 in CEC were shown in Figure 5. The multi-year variation of OBB emissions for various pollutants was similar (Figure 6).

The increase of crops residue burning dominated the significant growth of OBB emission. Pollutants emitted from OBB all increased obviously from 2003 to 2008. Then with the adoption of strict control policies (Table S1 in Supplement), the growth of crops residue burning emission gradually slow down. The forest, shrubland and grassland fire burning were related to weather conditions and human activities. Their emissions were difficult to predict and control and existed random yearly variation. Therefore, we discussed the multi-year variation during 2003-2015 instead of the overall trend for the whole period (Figure S3). Take $PM_{2.5}$ as example, emission exhibited clearly increasing trend from 2003 (256 Gg) to 2008 (353 Gg) and then decreased in the following two years to 322 Gg. After 2010, there existed higher (2011, 2013 and 2015) and lower values (2010, 2012 and 2014) alternately. The values in 2011, 2013 and 2015 all did not exceed the peak values in 2008.

Emissions from forest, shrubland and grassland fire burning have an obvious trend of declining from 2003 to 2006 and rising from 2006 to 2008. Peak emissions for $PM_{2.5}$ from forest, shrubland and grassland fire burning were found in 2008, as 49 Gg, 8.9 Gg and 0.7 Gg, respectively. In 2008, intensive policies for utilization of straw energy (Table S1) and strengthening the forestry fires prevention (Table S2) were published, which effectively limited the emissions from forest and shrubland fire burning as Figure 6a shown. Obvious decreasing was found from 2008 to 2010, down to 19 Gg, 4.8 Gg and 0.24 Gg, respectively. Then they exhibited inter-annual oscillation from 2010 to 2015, with higher emission in 2011, 2013 and 2015 and lower emission in 2012 and 2014 (Jin et al., 2017a). The multi-year tendency for forest, shrubland and grassland fire burning were mainly affected by the variations in climate, management measures and other human forcing. It can also conclude that the yearly fluctuation of pollutants from OBB was mainly impacted by the emission of forest, shrubland and grassland fire burning, but not the crop residue burning.

The emission of $PM_{2.5}$ from crop residue burning exhibited quite different yearly variation trend with other three types of biomass burning, which gradually increased from 2003 (228 Gg) to 2015 (323 Gg), by 29%. The increase of crop residue production can primarily explain the increasing of pollutant emission. Meanwhile, as shown in Figure S6 and Table S1, the controlling of pollutants from crop residue burning in China started from 1965s. In 2000, the law for prevention of air pollution was published. Then in 2003, the regulations on straw banning and comprehensive utilization were released. In Figure 6, we found that the emission of $PM_{2.5}$ from crop residue burning significantly increased from 2003 (228 Gg) to 2008 (294 Gg), due to the increase of crops production and deficiency of strict control policies in this period (Table S1). Although emissions from forest, shrubland and grassland fire burning fluctuated markedly during this period, the obvious increase of crops residue burning dominated the total growth of OBB emission from 2003 to 2008 as their higher emission amounts. From 2008 to 2015, strict policies were developed to improve the straw energy utilization and reduce the air pollution raised by its burning. However, it has to say, the policies may not be well implemented, with the annual averaged increasing amounts of 7.3 Gg for $PM_{2.5}$. From Figure 6b, the large contributions to $PM_{2.5}$ (22%-28% and 29%-33%) and increasing trends for corn straw burning and wheat straw burning could be found, which should be further focused. The contribution of rice straw burning has slightly decreased in research period, by about 19% . Other types of biomass totally accounted for averaged 25% of $PM_{2.5}$ emission and all exhibited slightly increasing trend from 2003 to 2015, by about 21%-29%.

Figure 7 showed that the crop residue burning emission in Henan, Shandong, Anhui, Jiangsu, Hubei, Hunan and Jiangxi exhibited obvious increasing trends, which suggested the importance of crop residue burning control in these provinces. For Fujian and Zhejiang, no obvious increase for cropland burning emission was found, implying that the emission has been well controlled in these years. It should be noted that in Fujian and Zhejiang, the main crop is rice. While in other provinces, the main crops are corn and wheat especially for northern provinces. To conclude, pollutants emitted from crop straw burning (wheat, corn and rice) are still now the key sources for air pollution, in view of its increasing emission trend. The randomness of burning activities and corresponding widespread and scattered distribution make it difficult to control them. The wheat and corn emissions at northern provinces and rice burning emissions at southern provinces should be controlled specially in the future.

In Figure 8, the $PM_{2.5}$ emission from crop residue burning exhibited higher amounts for Henan and Shandong province in 2015, as 100 Gg and 82 Gg, respectively, which are 200%-1200% times of those for other provinces. As the main source regions for air pollution of Yangtze River Delta (YRD) and Beijing-Tijin-Hebei (BTH) region (Fu et al., 2013; Zhou et al., 2015), the enforced and effective control of crop residue burning in the two provinces at summer and autumn harvest periods are important for improving the air quality of these regions.

### 3.2.2 Monthly distribution

The monthly $PM_{2.5}$ emission variation of different OBB in CEC was shown in Figure 8a. $PM_{2.5}$ emission held higher amounts in May and June (90.4 Gg-179.3 Gg), followed by December to March of next year (32.2 Gg-127.3 Gg) and September-October (8.2 Gg-89.2 Gg), and was lowest during July-August (14.3 Gg-65.9 Gg). As the emission amounts of cropland fire burning was one or two magnitude higher than other three types of biomass burning, the monthly variation of total $PM_{2.5}$ emission was dominantly controlled by the crop residue fire burning (Zhang et al., 2016). The periods with highest $PM_{2.5}$ emissions were the summer and autumn harvest times, when the burning activities are more frequent. The peak of open biomass fire burning occurred in May and June, totally accounted for 42% of the whole $PM_{2.5}$ emission in 2003-2015, which is caused by the harvest and open residue burning of winter wheat, especially in Henan, Shandong, Jiangsu and Anhui (Figure 8b). Large amounts of wheat straw were burned after the harvest to increase the soil fertility and prepare for following corn cultivation (Levine et al., 1995). The small peak of open biomass burning emission in September to October (totally accounted for 13.82% of the whole $PM_{2.5}$ emission in 2003-2015) can be attributed to the burning of corn straw after corn harvest. Though the open biomass burning was strictly forbidden in recent years, scattered burning activities still existed in these regions. As shown in Figure S4, the $PM_{2.5}$ emissions in CEC and major agricultural provinces during harvest time have shown a rapid decline in recent years, in accordance with the change tendency of burned area due to increased government management. Considering of the yearly increasing fact of crops straw burning, it is worth noting that fire burning out of harvest season as a way of circumventing governmental polices needs to be well regulated. From December to February, the crop residue burning emission decreased to the lowest level in the whole year (18.9% of the whole $PM_{2.5}$ emission in 2003-2015). However, the emissions of $PM_{2.5}$ from forest, shrubland and grassland burning achieved peak values from December to March, being 67% of that in 2003-2015.

Figure 9 clearly listed the monthly average emissions of $PM_{2.5}$ from OBB in different provinces. These provinces were classified based on the correlation between their monthly emissions of 2003-2015. Henan, Shandong, Anhui and Jiangsu provinces ($R^2$ higher than 0.92, P<0.01), as one of the largest and contiguous wheat planting areas in China (Fang et al., 2014), have two crop rotations. The highest monthly emissions were observed for winter wheat harvesting (sown in October and harvested from May to June) and corn harvesting (sown in middle June and harvested from September to October). A large proportion of crop straw were always burnt directly after the crop harvest (MEPC, 2015). For Hubei province, agricultural emissions fluctuated over the period from February to October with several peaks due to that different crop species matured in succession. In Jiangxi, Fujian and Hunan ($R^2$ higher than 0.9, P<0.01), the largest monthly emissions were observed with forest and shrubland fire burning during the time between December and March, which is the dry season in these provinces (Li et al., 2014; Li et al., 2015). While in other months, the emissions were limited. For Shanghai and Zhejiang ($R^2$ = 0.7, P<0.01), lowest levels of $PM_{2.5}$ emission were found, with peak values occurring in summer and autumn harvest periods. Obvious two peaks were found for April-May and July-August periods, which may reflect the rice harvesting at these times. To sum up, the regional differences of monthly $PM_{2.5}$ emissions from OBB were mainly caused by the different biomass burning types and times as well as corresponding environmental conditions.

### 3.2.3 Spatial distribution within 1 km×1 km of $PM_{2.5}$ emitted from OBB in CEC

The spatial distribution of $PM_{2.5}$ emitted from OBB within 1 km×1 km resolution was mapped based on the burned area and a high-resolution vegetation map (1:1 000000) in CEC. The multi-year averaged spatial distributions of $PM_{2.5}$ emission were shown in Figure 10. It can be found that the OBB was widespread and scattered. The average emissions intensity of $PM_{2.5}$ ranged from 0 to 15 tons per pixel in most provinces. The variation range is mainly caused by the social-economic development level, rural population and agricultural activities. The highest value in different provinces was all mainly raised by the cropland fire burning due to the centralized burning in a relatively small area. Some pixels with high emissions exceeding more than 100 tons each year were found in Henan, Shandong and Hunan. It can be attributed to the large amounts of crop straws in these provinces. The pixels of high emission intensity more than 70 tons from crop straw burning were also found in Hubei, Jiangsu and Anhui. For forest and shrubland fire burning, the high emission points (more than 30 tons per pixel) were found in Fujian and Jiangxi. Lower emission intensities in Zhejiang (lower than 10 tons per pixel on average) and Shanghai (lower than 7 tons on average) were mainly due to the highly developed economy and limited agricultural activities (Su et al., 2012). In addition, northern Anhui and eastern Jiangsu featured high emissions of OBB with a relatively lower intensity (lower than 15 tons per pixel on average), which may be due to that the crop straw were burned in a large area in these regions.

Though the emission intensities varied in the past ten years, the areas with high emission amounts remain similar. They were mainly located in the main agricultural areas in eastern Henan, southern Shandong, northern Anhui, northern Jiangsu, eastern Hubei and northern Hunan. This result is in accordance with formers (Huang, et al., 2012b). The junction regions of the four provinces (Henan, Shandong, Anhui and Jiangsu) should be paid more attention, where the pollutants emission from OBB jointed together. This was similar to a recent research (Jin et al., 2017b). This region belongs to HuangHuai Plain, with large area of cropland and low economic development levels. The open burning activities and corresponding banning policies are both abundant in village scale. The game of "cat and mouse" is frequently acted. More effective policies for guiding or helping farmers to utilize straw energy rather than banning crop residue burning arbitrarily should be considered sincerely. In Zhejiang and Shanghai, OBB emissions are sparsely scattered, due to the relatively developed economic level, scarce biomass sources and limited agricultural activities. The recycling of crop straw faces many difficulties due in part to its high cost and the relative low price of crop straw. Improving policies for effectively utilizing crop residue straw is also an important challenge for the government.

Figure 11 highlights the spatial distribution of $PM_{2.5}$ emitted from OBB in different seasons of 2015. Emissions were more concentrated in summer, followed by winter. In summer, the emission was concentrated in the connection regions of Henan, Shandong, Anhui and Jiangsu, which is mainly raised by the crop residue burning as discussed before. In winter, Jiangxi, Hunan and Fujian showed the higher emission intensities from forest and shrubland burning.

### 3.3 The impact of social-economic factors on OBB emission

Emissions from OBB were found to be in line with the local burning habit, social customs, rural population, local economic level, agricultural level and pollution controlling policies. Local burning habits have a great influence on different types of OBB emissions. According to our survey, in agricultural provinces, such as Henan, Shandong, Jiangsu and Anhui, people always burn crop straws in sowing and harvest seasons. Despite the strict implementation of crop residue burning management policies, the burning habit is difficult to change in a short time. Less crop residue production and crop burning activities are found in Jiangxi and Fujian, where people are accustomed to use crop straw to feed draught animals and produce biogas instead of open burning directly. Emission from crop residue burning is low. However, due to the rich forest and shrubland resources, wood is served as the staple household fuel, which mainly comes from felling trees or collecting branches. These human activities can lead to an increase of forest and shrubland fire burning, resulting in the elevated levels of corresponding emission in these provinces.

Social customs also pose impact on OBB emissions. Biomass burning emissions in April can be enhanced by human burning activities in the tomb-sweeping day. The tomb-sweeping day (often in April 4 or April 5) is a time to memorize the death. People sweep graves and burn sacrifices by ignited straw, which can easily cause grass, shrub and forest fires (Qiu et al., 2016). The fire points at the tomb-sweeping day in CEC occupied by 22%-38% of the whole fire points in April in some years (Figure S5). The Chinese government has also introduced policies to prevent forest, shrubland and grassland fires on tomb-sweeping day (Table S2). The wildfires caused by biomass burning from late January to early February are partially related to the firework burning in the Spring Festival (Zuo, 2004). The firework burning activities for celebration and official sacrifices to ancestors in the Spring Festival easily lead to grass, shrub and forest fires. All these activities can affect the emission levels and air quality in a short time scale.

In order to understand the impact of the rural population, local economic level and agricultural level, correlation analysis between $PM_{2.5}$ emissions from OBB and statistics data (rural population, per capita net income of rural residents, agricultural output (crop straw burning) and forestry output (forest, shrubland and grassland burning) in different provinces were conducted. Significant positive correlations were found between the rural population, agricultural output and the $PM_{2.5}$ emissions from crops straw burning ($R^2$ higher than 0.58, $P<0.01$) for the whole CEC (Figure 12a). According to our survey, the high rural population and agricultural output indicate that agricultural activities are quite important in a certain region. With more crops residue produced, it can easily cause high emissions from cropland fire burning. No significant correlations were found for $PM_{2.5}$ emission from crop straw burning with the income of rural residents (Figure 15), which indicates that the rural economic level in different regions in CEC have no relationship with the $PM_{2.5}$ emission. Then we calculated the correlations between the change tendency of $PM_{2.5}$ emission from crops fire burning and the multi-year variation of other three social-economic factors as Table 7 shown for different provinces. Significant positive correlations were found for $PM_{2.5}$ emission with per capita income of rural residents and agricultural output (most $R^2$ higher than 0.59, $P<0.01$) and negative correlation were found for $PM_{2.5}$ emission with rural population (most $R^2$ higher than 0.73, $P<0.01$) except for the provinces of Shanghai, Zhejiang and Fujian, which are underdeveloped agricultural provinces. From 2003 to 2015, with the increase of agricultural outputs, more crop residue was produced. However, rapid economic development and less rural population in each province lead to the popular of commercial energy and clean energy in rural area. It decreased the demands in using crop residue as fuel. As a consequence, more crop residues were directly burned in the agricultural field. But it was not suitable for Shanghai, Zhejiang and Fujian (most $R^2$ lower than 0.19, $P>0.05$), which holds less crop residue production and high utilization efficiency of crop straws.

Positive correlations were also found between forestry output and $PM_{2.5}$ emission from forestland, shrubland and grassland fire burning ($R^2$ =0.14, $P<0.01$) in the whole CEC (Figure 12b), which indicated that human forestry activities played positive role on open fire burning (Yan et al., 2006). According to our survey, human forest activities such as felling trees or picking up branches from trees can easily cause more forest and shrubland burning. However, compared with the crops straw burning, no correlation was found between $PM_{2.5}$ emission and other statistics data (the rural population and the per capita net income of rural residents) (Figure 13b and Table S4). It may indicate that the forestry fire burning activities were not predominantly associated with the rural human living activity. According to previous studies, forestry fire burning was affected by environmental conditions and human activities with environmental factors having a larger impact (Chen et al., 2013).

### 3.4 Comparison with others

Emission data from OBB in CEC during the past several years have been compared with other studies for the similar year (Table 8). Compared with the emissions derived from Wang et al. (2008) based on statistical data, the differences of OC, EC, $CH_4$, $NO_x$, NMVOCs, $NH_3$, $CO_2$ and CO emissions ranged from -41% to 12%. For $SO_2$ (121%) and $PM_{2.5}$ (288%) emission, the differences were relative high. All these differences were mainly caused by the selection of EFs. The EFs employed in Wang et al.(2008) were constant values for different biomass species. In addition, the crop residue to production ratio data and the burned ratio for various crop types were all specific to CEC in this study based on literatures and survey results, which increased the reliable of these data. Similarly, Huang et al. (2012) used the same $EF_S$ of different crops straw burning for emission calculation. Compared with Wang et al. (2008) and Huang et al. (2012),,the estimate in our study is believed to be more accuracy. An obvious underestimation of $PM_{2.5}$ emission from crop straw burning were found in Jin et al. (2017), in which not all the crop species were considered.

The estimation based on satellite observation was prevalent recently. Compared to Zhou et al. (2017) who estimated the pollutant emission amounts from MODIS burned area product, the results in this study were much higher. The reason may be that when using a single satellite data set, pollutant emission can be underestimated due to that some actual fire activities could not be detected (van der Werf et al., 2010). The lower emission of $CO_2$, NMVOCs, $SO_2$ and $NO_x$ in our study is due to the adoption of more accurate and suitable EFs values as those in a previous study (Tang et al, 2014). Our emission estimation of the pollutants is more similar to the results of Qiu et al. (2016), who also used multiple satellite products (MCD14 ML and MCD64Al) to estimate the OBB emissions of China in 2013, with the differences of the two studies ranging from -42% to 22%. For $CH_4$, $NO_x$, NMVOCs, $NH_3$ and $CO_2$, the differences were less than 10%. The reason for the differences is due to the use of updated local biomass data and EFs in this study. Therefore, the combination of multiple satellite products with local EFs data and updated local biomass data (updated forest loading data, the crop residue to production ratio data and the burned ratio for various crop types) are likely to have improved the estimation of pollutant emission from OBB effectively.

### 3.5 Uncertainty analysis

Emission uncertainties in this study were associated with the satellite fire products, biomass fuel loading data, combustion efficiency and emission factors. It is difficult to assess the uncertainty of the satellite-derived data for burned land area (Hoelzemann et al., 2004, Chang et al., 2010). The estimation of fire burned area were proved to be reliable by using the burned area product MCD64AL (Giglio et al., 2013) and active fire product MCD14ML (Randerson et al., 2012). Although some active fires which burned out at 10:30 am-1:30 pm each day could not be captured by MCD14ML, the burned area used in this study were more reliable due to the combination of multiple satellite dataset (MCD64AL and MCD14ML). The uncertainties in this study were mainly caused by biomass loading data, combustion efficiency and emission factors. These data were assumed to be normal distributions (Zhao et al., 2011). The uncertainty of biomass loading data and combustion efficiency was estimated to be approximately 50% (Shi et al., 2015) and the uncertainty of EFs of each pollutant mainly ranged from 0.03 to 0.85 (Table S5). The reliable of emission factors played the most important role in driving uncertainty. Considering all these parameters, 20,000 Monte Carlo simulations were performed to evaluate the estimation uncertainty quantitatively for pollutant emissions with 95% coincidence level. Table 9 showed the emission uncertainty for different pollutants from 2003-2015. On average, the uncertainty of the estimated OC, EC, $CH_4$, $NO_x$, NMVOCs, CO, $SO_2$, $NH_3$, $CO_2$ and $PM_{2.5}$ were (-30%, 30%), (-48%, 48%), (-20%, 20%), (-20%, 20%), (-45%, 45%), (-18%, 18%), (-45%, 45%), (-35%, 35%), (-3%, 3%) and (-36%, 36%), respectively.

Compared with previous studies, the uncertainty was improved in our study due to the datasets used here were better and more suitable. The reliable multiple satellites could better obtain burned area data. The local EFs data, updated forest loading data, the adoption of local crop residue to production ratio data and the crop residue burned ratio data based on survey results improved the emission estimation of forestry and cropland burning as they could better reflect the actual situation in this region. Compared with the constant combustion efficiency in previous researches, the activity combustion efficiency data could also reduce the uncertainty as they could more accurately reflect the actual combustion conditions (Chen et al., 2013) Therefore, due to the adoption of multiple satellite products, updated local biomass data and local emission factors, the uncertainty ranges of different pollutant emissions were narrowed and reliable in this study, which could better reflect the real emission.

## 4 Conclusions

In this study, a combination of the burned area product (MCD64Al) with the active fire product (MCD14 ML), as well as local high resolution vegetation speciation data, updated local biomass data, local emission factors and survey results were used to estimate the pollutant emissions from open burning in Central and Eastern China (CEC) from 2003 to 2015. The emission from crop residue, forest, shrubland and grassland fire burning were considered.

Crop residue burning was the major source type for pollutant emissions, followed by forest and shrubland fire burning. The grassland fire burning emissions were negligible in CEC. For cropland, the fire burning was mainly concentrated in agricultural provinces, such as Henan and Shandong. For forest and shrubland, the fire burning was mainly concentrated in Fujian, Jiangxi and Hunan provinces, with abundant forest resources. Wheat, corn and rice straw were the major three types of crop straws for pollutant emission. Wheat and corn straw burning dominated in Shangdong and Henan and the rice straw burning dominated in Hunan, Jiangxi and Hubei provinces. For various pollutant emissions, corn straw burning was the largest contributor to $SO_2$, $NO_x$, CO, NMVOCs, $CO_2$, $NH_3$. OC, EC and $CH_4$ emission was mainly produced by rice straw burning. Wheat straw burning was the largest contributor to $PM_{2.5}$. The spatial distribution of open biomass residue burning in different years was similar. The high emissions were mainly found in the major agricultural areas in eastern Henan, southern Shandong, northern Anhui, northern Jiangsu, eastern Hubei and northern Hunan, due to their abundant agricultural cultivated areas and low straw utilization efficiency.

From 2003 to 2015, the multi-year tendency of opening biomass residue burning emission for various pollutants was similar. Emissions from crop straw burning continued to increase, due to the gradual increase of crop residue production. While emissions from forest, shrubland and grassland fire burning exhibited yearly fluctuations, which was mainly influenced by the environmental conditions, management measures and other human driving factors. Monthly distributions revealed that the pollutant emissions were at the highest levels in May and June, with the lowest emissions in July and August. The high emissions in May, June and October were mainly caused by crop straw burning in sowing and harvest times. It is worth noting that the fire burning activities at harvest season need to be regulated continuously by local governments and emissions from forest and shrubland burning accounted for the vast majority of total emissions in December to March should also be paid attention. The emission of crop residue burning was associated with the rural population, agricultural output and economic levels while the environmental conditions play an important role in the emissions from forestland, shrubland and grassland fire burning.

The estimation of mulit-year open biomass burning emissions by satellite data in this study will provide an objective and creditable evidences for assessing the role of pollution prevention policies on open burning activities issued in the last decade. The high-spatial (1×1 km) resolution emission inventory in month scale is also useful in modeling regional air quality and human health risks in the future.

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**Table 1.** Forest, shrubland and grassland biomass fuel loading (kt $km^{-2}$) in each province.

| Province | Forest (2003-2008)[a] | Forest (2009-2015)[*] | Shrubland[b] | Grassland[c] |
|---|---|---|---|---|
| Shandong | 4.26 | 2.95 | 6.94 | 0.78 |
| Henan | 5.66 | 4.16 | 6.94 | 0.77 |
| Anhui | 6.32 | 3.61 | 12.2 | 0.77 |
| Jiangsu | 4.7 | 2.64 | 6.86 | 0.72 |
| Hubei | 5.34 | 3.28 | 7.87 | 0.88 |
| Hunan | 4.79 | 2.52 | 17.4 | 0.8 |
| Jiangxi | 4.75 | 3.08 | 18.5 | 0.76 |
| Fujian | 6.29 | 5.91 | 18.9 | 0.85 |
| Zhejiang | 3.51 | 3.11 | 18.4 | 0.86 |
| Shanghai | 6.09 | 2.99 | 6.86 | 0.93 |

References: [a] Fang et al. (1996); [b] Pu et al. (2004); [c] Hu et al. (2006); [*] This study.

**Table 2.** Parameters of linear regression model for biomass and stock volume of dominant tree species.

| Tree species | a | b | Tree species | a | b |
|---|---|---|---|---|---|
| Larix | 0.967[a] | 5.7598[a] | Cinnamomum camphora | 1.0357[a] | 8.0591[a] |
| Pinus koraiensis | 0.5185[a] | 18.22[a] | Phoebe | 1.0357[a] | 8.0591[a] |
| Pinus sylvestris var. mongolica | 1.11[a] | | Elm | 0.7564[f] | 8.3013[f] |
| Pinus densiflora | 1.0945[b] | 2.004[b] | Robinia | 0.7564[a] | 8.3103[a] |
| Pinus thunbergii parl | 0.5168[b] | 33.237[b] | Schima superba | 0.76[e] | 8.31[e] |
| Chinese pine | 0.7554[a] | 5.0928[a] | Sweetgum | 0.76[e] | 8.31[e] |
| Pinus armandi | 0.5856[a] | 18.7435[a] | Other hard broad leaf | 0.7564[b] | 8.3103[b] |
| Pinus massoniana | 0.52[a] | | Tilia | 0.7975[b] | 0.4204[b] |
| Pinus yunnanensis | 0.52[a] | | Sassafras | 1.0357[a] | 8.0591[a] |
| Pinus kesiya var. langbiamensis | 0.510[b] | 1.045[b] | Populus | 0.4754[a] | 30.603[a] |
| Pinus densata | 0.5168[b] | 33.237[b] | Salix | 0.4754[c] | 30.6034[c] |
| Foreign pine | 0.5168 | 33.2378 | Paulownia | 0.8956[d] | 0.0048[d] |
| Pinus elliottii | 0.51[e] | 1.05[e] | Eucalyptus | 0.7893[a] | 6.9306[a] |
| Pinus taeda | 0.5168[f] | 33.2378[f] | Rich acacia | 0.4754[a] | 30.60[a] |
| Mount huangshan pine | 0.5168[f] | 33.2378[f] | Casuarina equisetifolia | 0.7441[b] | 3.2377[b] |
| Joe pine | 0.5168[f] | 33.237[f] | Melia azedarach | 0.4754[b] | 30.603[b] |
| Other pine | 0.5168[a] | 33.2378[a] | Other soft broad leaf | 0.4754[b] | 30.603[b] |
| Cunninghamia lanceolata | 0.399[a] | 22.54[a] | Coniferous mixed | 0.5168[f] | 33.2378[f] |
| Cryptomeria fortunei | 0.4158[a] | 41.3318[a] | Broad-leaved mixed | 0.8392[b] | 9.4157[b] |
| Metasequoia | 0.4158[a] | 41.3318[a] | Coniferous and broad-leaved mixed | 0.7143[b] | 16.9154[b] |
| Taxodium ascendens | 0.399[a] | 22.541[a] | Betula | 0.9644[a] | 0.8485[a] |
| Abies | 0.4642[a] | 47.499 | White birch | 0.9644[a] | 0.8485[a] |
| Picea | 0.4642[a] | 47.499[a] | Betula costata | 0.9644[a] | 0.8485[a] |
| Tsuga | 0.4158[a] | 41.3318[a] | Water, beard and yellow | 0.7975[b] | 0.4202[b] |
| Keteleeria | 0.4158 | 41.3318 | Manchurian Ash | 0.798[c] | 0.42[c] |
| Cupressus | 0.6129[a] | 26.1451[a] | Juglans mandshurica | 0.798[c] | 0.42[c] |
| Yew | 0.4642[b] | 47.499[b] | Amur corktree | 0.798[c] | 0.42[c] |
| Other fir | 0.399[a] | 22.541[a] | Quercus | 1.3288[a] | -3.8999[a] |

References: [a] Fang et al. (1996); [b] Wen et al. (2014); [c] Lu et al. (2012); [d] Tian et al (2011); [e] Wang et al (2014); [f] Li et al. (2014).

**Table 3.** Detailed crop residue to production ratio data for each province

| Province | Rice | Corn | Wheat | Cotton | Rapeseed | Soy bean | Sugar cane | Peanut | Potato | Sesame | Sugar beet | Tobacco |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Anhui | 1.09[a] | 1[a] | 1.12[a] | 3.35[a] | 2.98[a] | 1.52[a] | 0.34[a] | 1.26[a] | 0.53[a] | 2.01[a] | 0.37[a] | 0.71[a] |
| Fujian | 0.85[b] | 1.04[c] | 1.17[c] | 2.91[d] | 2.87[d] | 1.5[d] | 0.43[d] | 1.08[m] | 0.57[d] | 2.01[d] | 0.43[d] | 0.56[d] |
| Henan | 1[c] | 0.96[c] | 1.08[h] | 2.41[i] | 2.87[d] | 1.5[d] | 0.34[d] | 0.89[d] | 0.57[d] | 1.78[d] | 0.43[d] | 0.49[d] |
| Hubei | 1.17[e] | 1.04[c] | 1.17[c] | 4.09[j] | 3.17[k] | 1.5[d] | 0.43[d] | 1.14[d] | 0.57[d] | 2.01[d] | 0.43[d] | 0.71[d] |
| Hunan | 0.94[f] | 1.11[g] | 1.17[c] | 2.91[d] | 3[l] | 1.5[d] | 0.43[d] | 1.38[n] | 0.57[d] | 2.23[d] | 0.43[d] | 0.85[d] |
| Jiangsu | 1.04[a] | 1[a] | 1.41[c] | 2.61[i] | 2.98[a] | 1.52[a] | 0.34[a] | 1.26[a] | 0.53[a] | 2.01[a] | 0.37[a] | 0.71[a] |
| Jiangxi | 1[c] | 1.04[c] | 1.17[c] | 2.91[d] | 2.87[d] | 1.5[d] | 0.43[d] | 1.14[d] | 0.57[d] | 2.01[d] | 0.43[d] | 0.71[d] |
| Shandong | 1[c] | 0.96[c] | 1.33[c] | 2.91[d] | 2.87[d] | 1.5[d] | 0.43[d] | 0.85[d] | 0.57[d] | 2.01[d] | 0.43[d] | 0.71[d] |
| Shanghai | 1.28[a] | 0.93[a] | 1.09[a] | 3.35[a] | 2.98[a] | 1.52[a] | 0.34[a] | 1.26[a] | 0.53[a] | 2.01[a] | 0.37[a] | 0.71[a] |
| Zhejiang | 1.07[a] | 0.96[a] | 1.2[a] | 3.35[a] | 2.98[a] | 1.52[a] | 0.34[a] | 1.26[a] | 0.53[a] | 2.01[a] | 0.37[a] | 0.71[a] |

References: [a] Zhu et al. (2017); [b] Chen et al. (2008); [c] Xie et al. (2011a); [d] Xie et al (2011b); [e] Zeng et al (2007); [f] Ao et al. (2007); [g] Lei et al. (2009); [h] Zhao et al. (2008); [i] Xue et al. (2006); [j] Yu et al (2009); [k] Zou et al (2008); [l] Liu et al. (2010); [m] Tang et al. (2009); [n] Li et al. (2008).

**Table 4.** Detailed crops straw burned ratio data for each province.

| Region | Crops straw burning percentage |
|---|---|
| Anhui | 0.10[a] |
| Fujian | 0.188[b] |
| Henan | 0.208[c] |
| Hubei | 0.207[c] |
| Hunan | 0.278[c] |
| Jiangsu | 0.10[a] |
| Jiangxi | 0.18[c] |
| Shandong | 0.178[c] |
| Shanghai | 0.148[d] |
| Zhejiang | 0.319[c] |

References: [a] Tian (2011); [b] Huang (2014); [c] Peng et al; (2016).[d] Zhou et al (2017).

**Table 5.** The emission factors of open biomass burning emissions for various pollutants (g $kg^{-1}$ dry matter)

| Vegetation | OC | EC | CO | $CH_4$ | $NO_x$ | NMVOCs | $SO_2$ | $NH_3$ | $CO_2$ | $PM_{2.5}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| Corn | 1.457[*] | 0.14* | 70.2[a] | 4.4[b] | 3.36[a] | 10[c] | 0.45[c] | 0.68[g] | 1261[f] | 5[c] |
| Rice | 1.96[a] | 0.52[c] | 52.32[c] | 3.9[b] | 1.42[d] | 6.05[f] | 0.147[a] | 0.53[g] | 791[f] | 3.03[d] |
| Wheat | 2.7[b] | 0.49[a] | 61.90[c] | 3.4[b] | 1.19[d] | 7.5[c] | 0.147[c] | 0.37[b] | 1557[f] | 7.6[a] |
| Cotton | 3.06[c] | 0.57[f] | 70.29[c] | 4.4[b] | 2.98[c] | 10[c] | 0.23[c] | 0.68[b] | 1445[h] | 11.7[c] |
| Rapeseed | 1.08[d] | 0.23[d] | 34.3[d] | 3.9[b] | 1.12[d] | 8.64[c] | 0.25[c] | 0.53[g] | 1445[h] | 5.76[c] |
| Soya bean | 1.05[d] | 0.13[d] | 32.3[d] | 3.9[b] | 1.08[d] | 8.64[c] | 0.25[c] | 0.53[g] | 1445[h] | 3.32[d] |
| Sugar cane | 2.03[c] | 0.41[c] | 40.08[f] | 3.9[b] | 2.03[c] | 11.02[f] | 0.25[c] | 0.53[g] | 1445[h] | 4.12[f] |
| Peanut | 2.03[c] | 0.41[c] | 55.13[c] | 3.9[b] | 2.11[c] | 8.64[c] | 0.25[c] | 0.53[g] | 1445[h] | 5.76[c] |
| Potato | 2.03[c] | 0.41[c] | 55.13[c] | 3.9[b] | 2.11[c] | 8.64[c] | 0.25[c] | 0.53[g] | 1445[h] | 5.76[c] |
| Tobacco | 2.03[c] | 0.41[c] | 55.13[c] | 3.9[b] | 2.11[c] | 8.64[c] | 0.25[c] | 0.53[g] | 1445[h] | 5.76[c] |
| Sesame | 2.03[c] | 0.41[c] | 55.13[c] | 3.9 [b] | 2.11[c] | 8.64[c] | 0.25[c] | 0.53[g] | 1445[h] | 5.76[c] |
| Sugar beet | 2.03[c] | 0.41[c] | 55.13[c] | 3.9[b] | 2.11[c] | 8.64[c] | 0.25[c] | 0.53[g] | 1445[h] | 5.76[c] |
| Coniferous forest | 7.8[e] | 0.2[e] | 118[e] | 6[e] | 2.4[e] | 28[e] | 1[i] | 3.5[e] | 1514[e] | 9.7[e] |
| Broadleaf forest | 9.2[e] | 0.6[e] | 102[e] | 5[e] | 1.3[e] | 11[e] | 1[e] | 1.5[e] | 1630[e] | 13[e] |
| Mixed forest | 9.2[e] | 0.6[e] | 102[e] | 5[e] | 1.3[e] | 14[e] | 1[i] | 1.5[e] | 1630[e] | 9.7[e] |
| Grassland | 2.6[e] | 0.4[e] | 59[e] | 1.5[e] | 2.8[e] | 9.3[e] | 0.5[e] | 0.5[e] | 1692[e] | 5.4[e] |
| Shrubland | 6.6[e] | 0.5[e] | 68[e] | 2.6[e] | 3.9[e] | 4.8[e] | 0.7[e] | 1.2[e] | 1716[e] | 9.3[e] |

References: [a] Cao et al. (2008); [b] Li et al. (2007); [c] He et al. (2015); [d] Tang et al. (2014); [e] Akagi et al. (2011); [f] Zhang et al. (2008); [g] EPD (2014); [h] Wang et al. (2008); [i] Andreae and Rosenfeld (2008); [*] This study.

**Table 6.** Cumulative emissions of major pollutants from open biomass burning in Central and Eastern China during 2003-2015 (Gg)

| Province | OC | EC | $CH_4$ | $NO_x$ | NMVOCs | $SO_2$ | $NH_3$ | CO | $CO_2$ | $PM_{2.5}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| Shandong | 783.9 | 48.56 | 669.4 | 479.3 | 1505 | 54.55 | 95.56 | 10880 | 226705 | 1007 |
| Henan | 1068 | 63.19 | 738.3 | 512.1 | 1629 | 54.23 | 101.3 | 11869 | 260239 | 1155 |
| Anhui | 238.2 | 20.24 | 197.7 | 115 | 410 | 12.94 | 29.75 | 2939 | 63623 | 283.1 |
| Jiangsu | 201.6 | 19.88 | 178 | 98.48 | 341 | 9.29 | 23.89 | 2543 | 53106 | 228.5 |
| Hubei | 234.2 | 33.92 | 337.7 | 173.1 | 660.7 | 19.86 | 48.5 | 4555 | 97788 | 415.8 |
| Hunan | 202 | 40.34 | 376.8 | 179.1 | 738.4 | 24.33 | 64.3 | 5239 | 96338 | 418.8 |
| Jiangxi | 132.8 | 27.88 | 236.1 | 109 | 447.6 | 14.2 | 40.55 | 3305 | 57692 | 252.3 |
| Fujian | 97.15 | 15.15 | 148.1 | 71.14 | 347.4 | 12.81 | 34.45 | 2285 | 40095 | 190.2 |
| Zhejiang | 91.41 | 16.22 | 147.9 | 70.53 | 290.9 | 9.62 | 25.83 | 2055 | 39142 | 167.8 |
| Shanghai | 14.34 | 2.09 | 17.14 | 8.56 | 29.89 | 0.76 | 2.29 | 233.8 | 4392 | 17.88 |
| Total | 3064 | 287.5 | 3047 | 1816 | 6399 | 212.6 | 466.5 | 45904 | 939120 | 4136 |

**Table 7.** Correlation of the variation tendency between $PM_{2.5}$ emission from crops straw burning and rural population, agricultural output, per capita incomes of rural residents in each province from 2003 to 2015.

| $PM_{2.5}$ emission (Gg) | Rural population (10 thousand) | Per capita income of rural residents (RMB) | Agricultural output (0.1 billion RMB) |
|---|---|---|---|
| Shanghai | y = -0.001x + 1.64<br>$R^2$ = 0.17 P > 0.05 | y = -5E-06x + 1.4<br>$R^2$ = 0.09 P > 0.05 | y = 7E-05x + 1.36<br>$R^2$ = 0.0005 P > 0.05 |
| Zhejiang | y = 0.002x + 6.19<br>$R^2$ = 0.06 P > 0.05 | y = -6E-05x + 10.47<br>$R^2$ = 0.19 P > 0.05 | y = -0.001x + 10.72<br>$R^2$ = 0.19 P > 0.05 |
| Fujian | y = -0.0002x + 8.219<br>$R^2$ = 0.01 P > 0.05 | y = -3E-05x + 8.1884<br>$R^2$ = 0.06 P > 0.05 | y = -0.0002x + 8.2144<br>$R^2$ = 0.06 P > 0.05 |
| **Jiangsu** | **y = -0.002x + 23.41**<br>**$R^2$ = 0.8 P < 0.01** | **y = 0.0002x + 15.33**<br>**$R^2$ = 0.66 P < 0.01** | **y = 0.001x + 15.18**<br>**$R^2$ = 0.69 P < 0.01** |
| **Hubei** | **y = -0.008x + 56.19**<br>**$R^2$ = 0.94 P < 0.01** | **y = 0.0009x + 25.39**<br>**$R^2$ = 0.86 P < 0.01** | **y = 0.004x + 24.31**<br>**$R^2$ = 0.92 P < 0.01** |
| **Anhui** | **y = -0.005x + 37.11**<br>**$R^2$ = 0.91 P < 0.01** | **y = 0.0007x + 16.12**<br>**$R^2$ = 0.79 P < 0.01** | **y = 0.004x + 14.5**<br>**$R^2$ = 0.85 P < 0.01** |
| **Hunan** | **y = -0.01x + 62.66**<br>**$R^2$ = 0.78 P < 0.01** | **y = 0.0008x + 20.66**<br>**$R^2$ = 0.8 P < 0.01** | **y = 0.003x + 20.1**<br>**$R^2$ = 0.91 P < 0.01** |
| **Jiangxi** | **y = -0.008x + 33.73**<br>**$R^2$ = 0.92 P < 0.01** | **y = 0.0006x + 11.19**<br>**$R^2$ = 0.82 P < 0.01** | **y = 0.006x + 9.84**<br>**$R^2$ = 0.87 P < 0.01** |
| **Henan** | **y = -0.01x + 150.14**<br>**$R^2$ = 0.8 P < 0.01** | **y = 0.003x + 70.41**<br>**$R^2$ = 0.59 P < 0.01** | **y = 0.008x + 62.79**<br>**$R^2$ = 0.72 P < 0.01** |
| **Shandong** | **y = -0.009x + 122.46**<br>**$R^2$ = 0.73 P < 0.01** | **y = 0.0014x + 66.48**<br>**$R^2$ = 0.66 P < 0.01** | **y = 0.004x + 62.11**<br>**$R^2$ = 0.77 P < 0.01** |

**Table 8.** Comparison of the emissions with previous studies in different years (Gg)

| Reference | Year | OC | EC | $CH_4$ | $NO_x$ | NMVOCs | $SO_2$ | $NH_3$ | CO | $CO_2$ | $PM_{2.5}$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Wang et al., 2008 | 2006 | 252 | 25.8 | 197 | 189 | 459 | 31.8 | 44.1 | 3841 | 81225 | 1138 |
| This study | | 215.3 | 21.13 | 220.7 | 131.9 | 451.1 | 14.33 | 31.46 | 3267 | 67753 | 293.09 |
| Huang et al., 2012 | 2006 | 54 | 17.4 | 136 | 123 | 1196 | 8.1 | 50.6 | 2379 | 36886 | 146 |
| This study | | 209.8 | 20.67 | 215.8 | 129.1 | 436.4 | 13.56 | 29.64 | 3172 | 66088 | 283.3 |
| Qiu et al., 2016 | 2013 | 222 | 41.5 | 243 | 168 | 591 | 30.2 | 46.9 | 3273 | 78633 | 475 |
| This study | | 258.2 | 23.53 | 252.1 | 151.2 | 531.5 | 17.86 | 38.67 | 3817 | 78050 | 343.44 |
| Zhou et al., 2017 | 2012 | 185 | 16.9 | 254 | 160 | 543 | 40.4 | 34.5 | 3330 | 92797 | 484 |
| This study | | 248.6 | 23.11 | 245.7 | 148.5 | 507.8 | 16.71 | 35.92 | 3688 | 75785 | 329.46 |

**Table 9.** The uncertainty estimation of open biomass burning emissions for various pollutants from 2003 to 2015.

| Year | OC | EC | CO | $CH_4$ | $NO_x$ | NMVOC | $SO_2$ | $NH_3$ | $CO_2$ | $PM_{2.5}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| 2003 | (-31%, 31%) | (-46%, 46%) | (-20%, 20%) | (-20%, 20%) | (-23%, 23%) | (-52%, 53% | (-52%, 51%) | (-33%, 33%) | (-3%, 3%) | (-44%, 44%) |
| 2004 | (-29%, 29%) | (-47%, 48%) | (-21%, 21%) | (-22%, 22%) | (-24%, 24%) | (-45%, 45%) | (-56%, 58%) | (-34%, 34%) | (-3%, 3%) | (-47%, 47%) |
| 2005 | (-31%, 31%) | (-42%, 44%) | (-16%, 16%) | (-16%, 17%) | (-19%, 19%) | (-41%, 40%) | (-44%, 44%) | (-32%, 33%) | (2%, 3%) | (-35%, 34%) |
| 2006 | (-32%, 33%) | (-44%, 44%) | (-13%, 13%) | (-14%, 14%) | (-16%, 17%) | (-43%, 43%) | (-34%, 35%) | (-34%, 34%) | (-3%, 3%) | (-25%, 25%) |
| 2007 | (-30%, 30%) | (-46%, 46%) | (-18%, 19%) | (-19%, 19%) | (-22%, 22%) | (-50%, 51%) | (-50%, 50%) | (-33%, 34%) | (-3%, 3%) | (-42%, 42%) |
| 2008 | (-26%, 26%) | (-52%, 53%) | (-25%, 25%) | (-28%, 28%) | (-29%, 29%) | (-69%, 69%) | (-62%, 61%) | (-38%, 39%) | (-3%, 3%) | (-55 %, 56%) |
| 2009 | (-28%, 28%) | (-48%, 48%) | (-21%, 21%) | (-21%, 22%) | (-24%, 24%) | (-59%, 59%) | (-54%, 54%) | (-34%, 35%) | (-3%, 3%) | (-47%, 47%) |
| 2010 | (-31%, 31%) | (-44%, 44%) | (-16%, 17%) | (-17%, 17%) | (-19%, 19%) | (-45%, 46%) | (-42%, 42%) | (-33%, 34%) | (-3%, 3%) | (-34%, 34%) |
| 2011 | (-29%, 29%) | (-46%, 46%) | (-18%, 18%) | (-19%, 19%) | (-21%, 21%) | (-52%, 53%) | (-47%, 47%) | (-34%, 35%) | (-3%, 3%) | (-40%, 40%) |
| 2012 | (-32%, 33%) | (-44%, 44%) | (-14%, 14%) | (-14%, 14%) | (-17%, 17%) | (-35%, 35%) | (-35%, 35%) | (-34%, 35%) | (-3%, 3%) | (-27%, 26%) |
| 2013 | (-30%, 30%) | (-44%, 44%) | (-16%, 16%) | (-17%, 17%) | (-20%, 20%) | (-51%, 51%) | (-42%, 43%) | (-33%, 34%) | (-3%, 3%) | (-36%, 36%) |
| 2014 | (-32%, 32%) | (-45%, 46%) | (-15%, 15%) | (-16%, 16%) | (-19%, 18%) | (-43%, 43%) | (-42%, 42%) | (-35%, 35%) | (-3%, 3%) | (-33%, 33%) |
| 2015 | (-31%, 31%) | (-44%, 44%) | (-14%, 146%) | (-14%, 13%) | (-17%, 17%) | (-41%, 41%) | (-34%, 34%) | (-34%, 35%) | (-3%, 3%) | (-26%, 26%) |

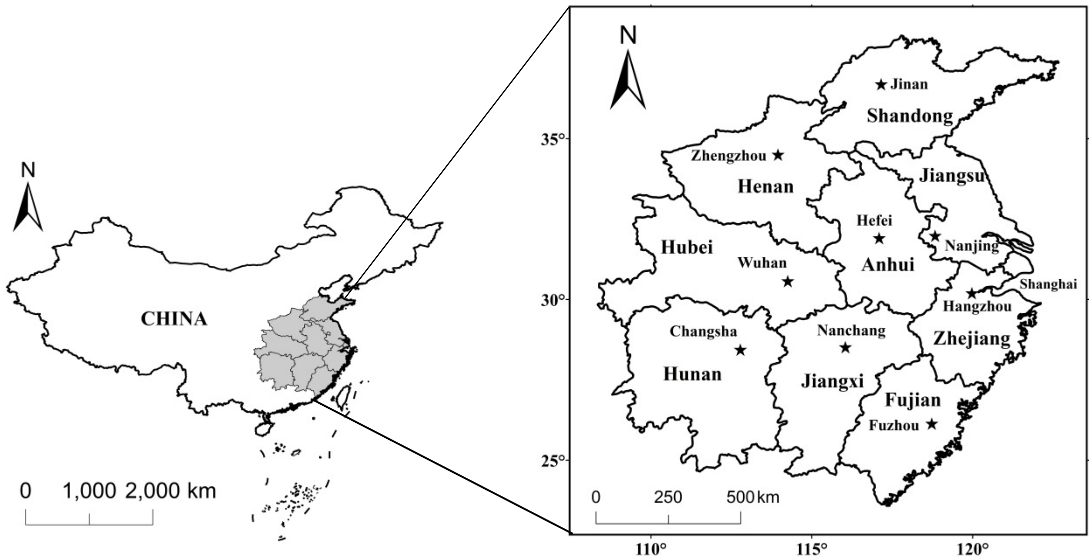



**Figure 1.** Location of Central and Eastern China and the key megacitie

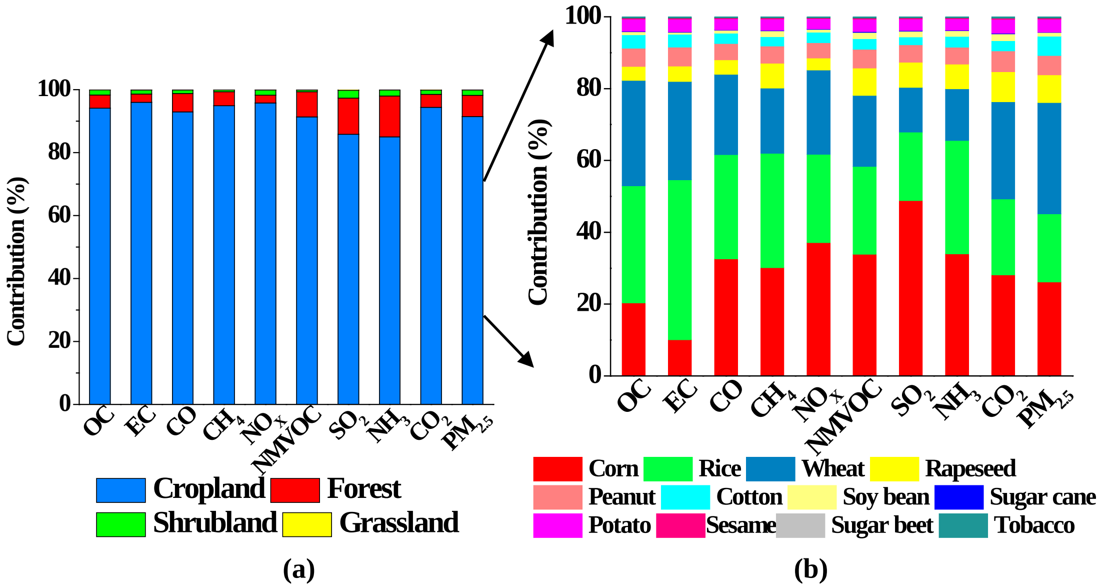
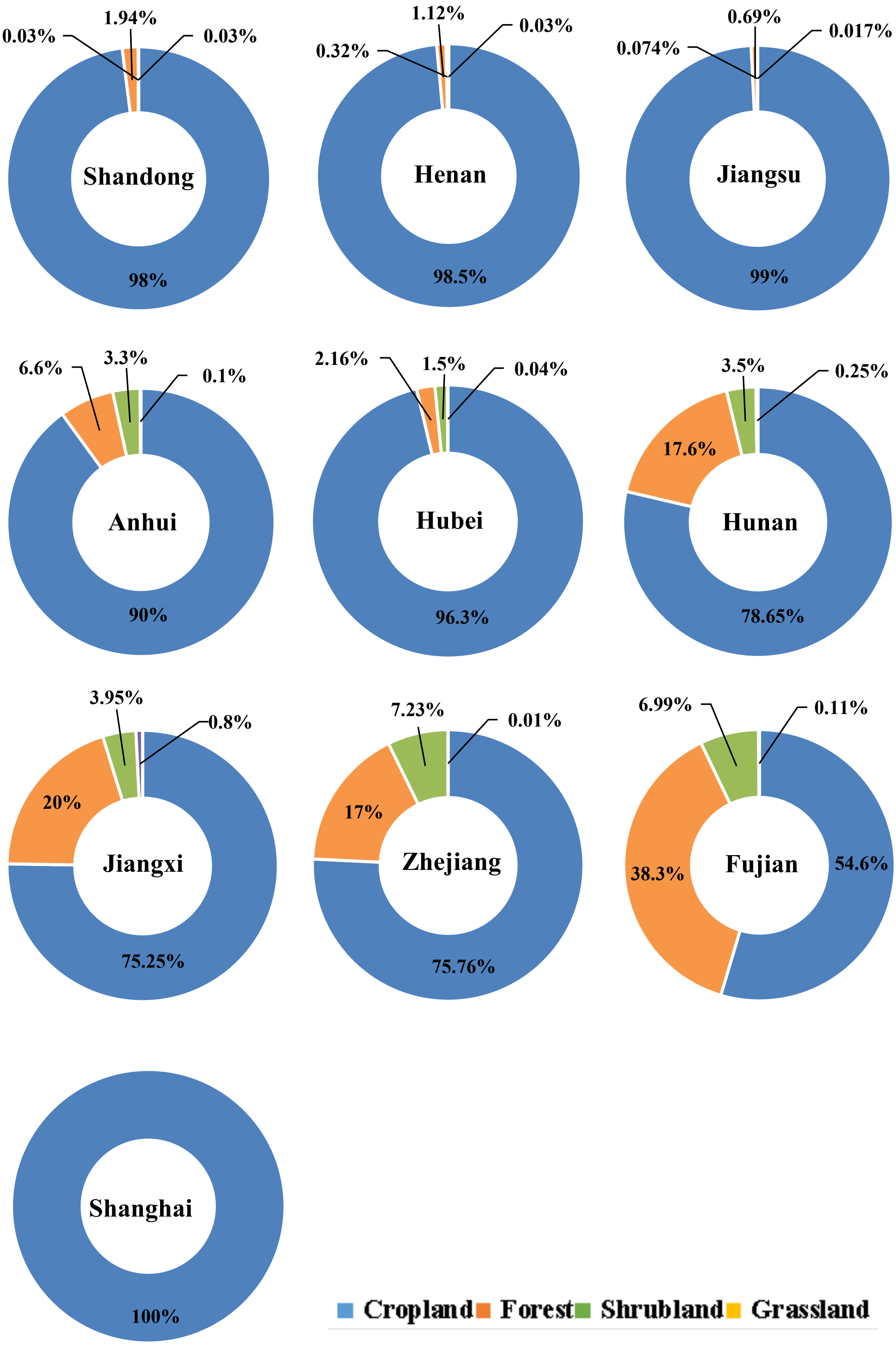



**Figure 2.** The mean contributions of different types of biomass to biomass burning pollutant emission (a) and the mean contributions of different types of crops to cropland accumulative pollutant emission (b) from 2003 to 2015.


0.03%
1.94%
0.03%
Shandong
98%
1.12%
0.32%
0.03%
Henan
98.5%
0.69%
0.074%
0.017%
Jiangsu
99%
6.6%
3.3%
0.1%
Anhui
90%
2.16%
1.5%
0.04%
Hubei
96.3%
3.5%
0.25%
17.6%
Hunan
78.65%
3.95%
0.8%
20%
Jiangxi
75.25%
7.23%
0.01%
17%
Zhejiang
75.76%
6.99%
0.11%
38.3%
Fujian
54.6%
Shanghai
100%
Cropland
Forest
Shrubland
Grassland

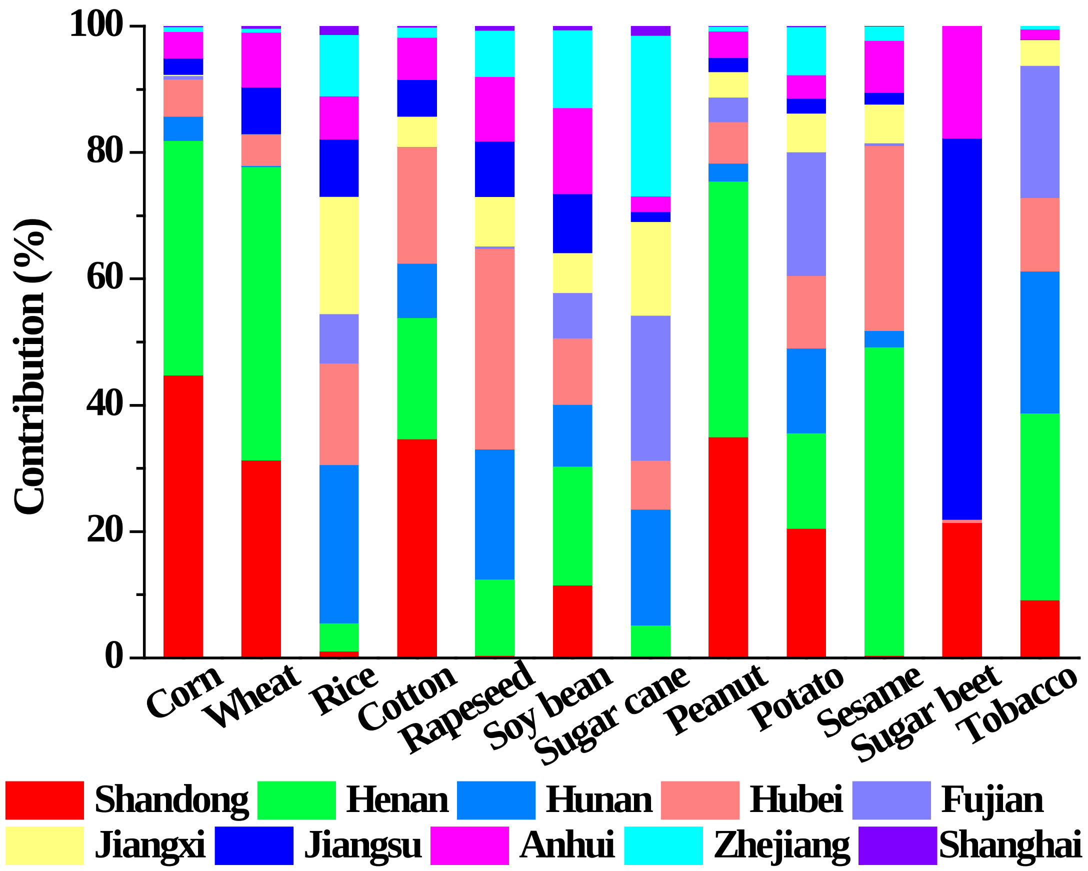



**Figure 4.** The averaged contributions of various crops straw burning to cropland $PM_{2.5}$ emission in different provinces.

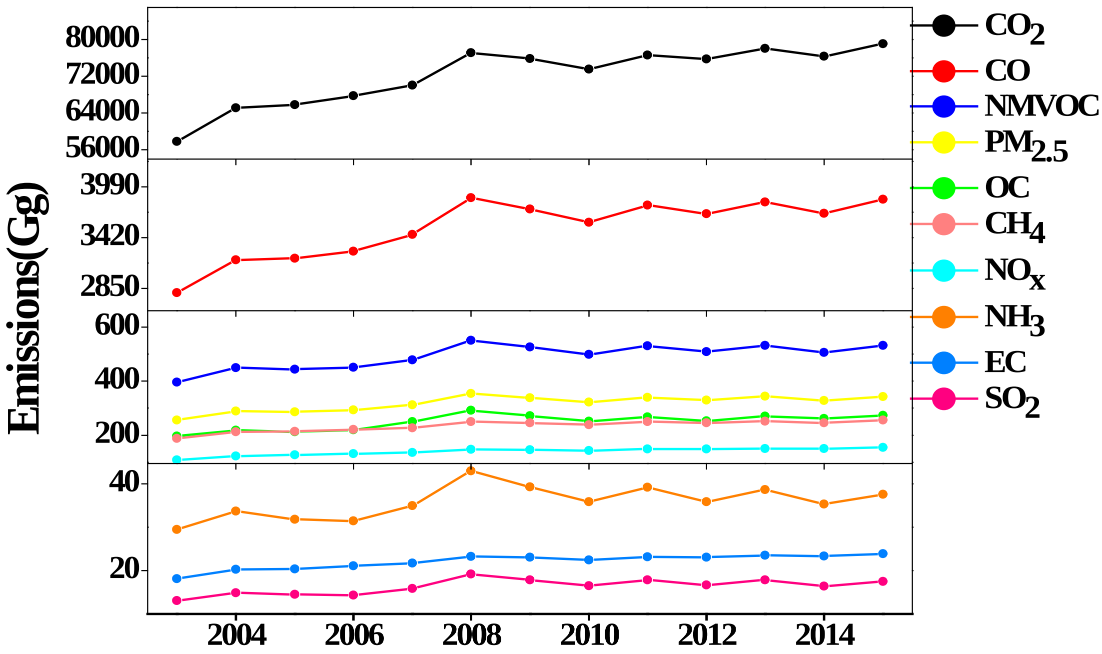
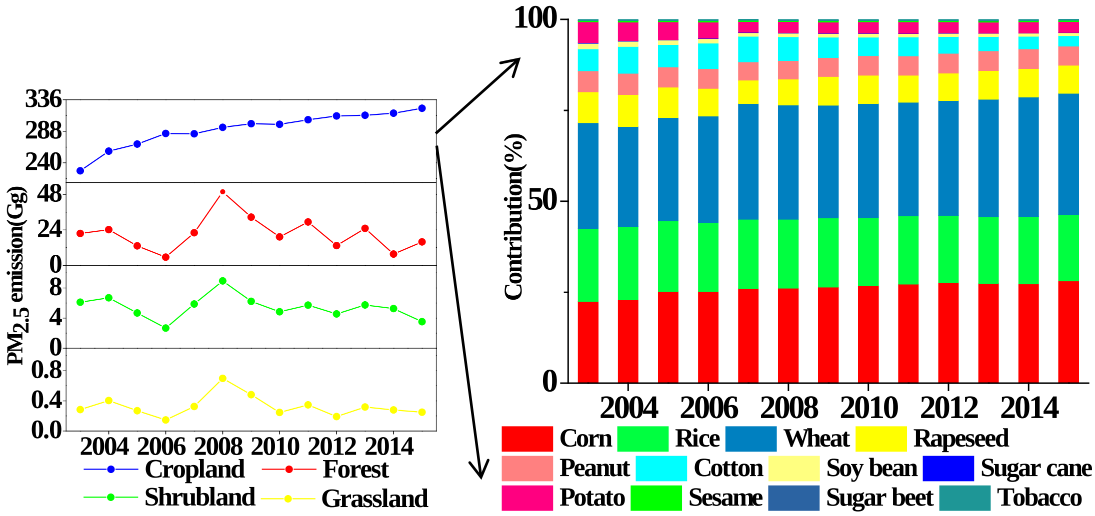



**Figure 5.** Yearly emissions of open biomass burning from 2003 to 2015.




(a) (b)

**Figure 6.** The multi-year $PM_{2.5}$ emission of different opening biomass burning sources (a) and various crop types (b) from 2003 to 2015.

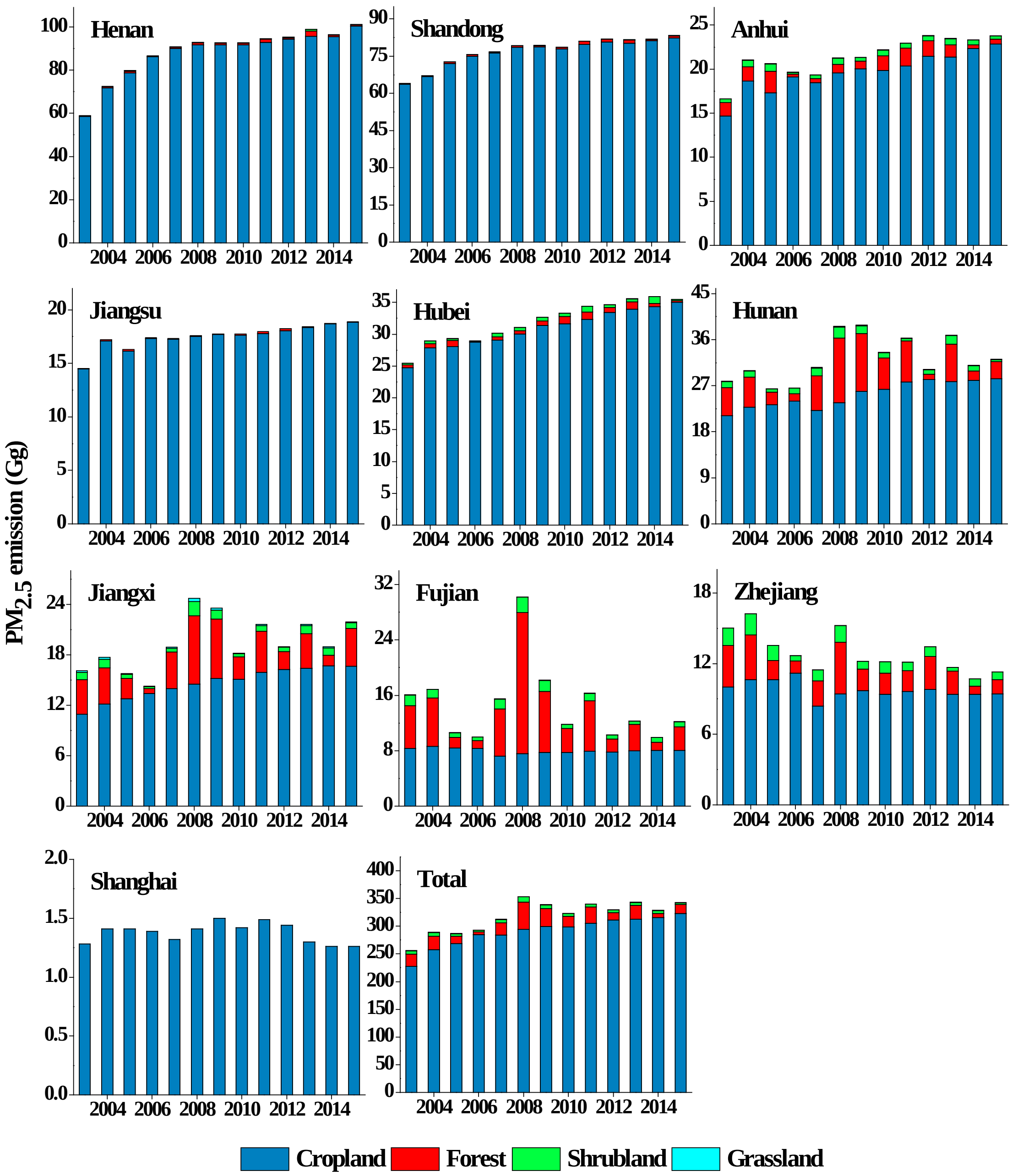



**Figure 7.** The multi-year $PM_{2.5}$ emission of the four types of biomass burning in different provinces from 2003 to 2015.

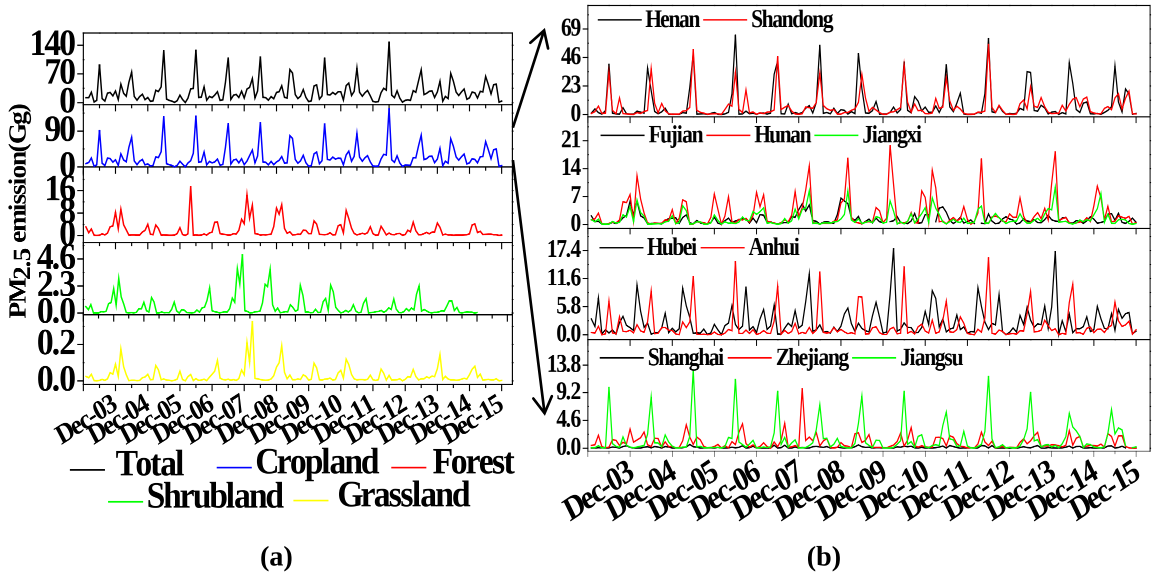



**Figure 8.** The monthly $PM_{2.5}$ emission of different open biomass burning from 2003 to 2015 for the whole Central and Eastern China (a) and each province (b).

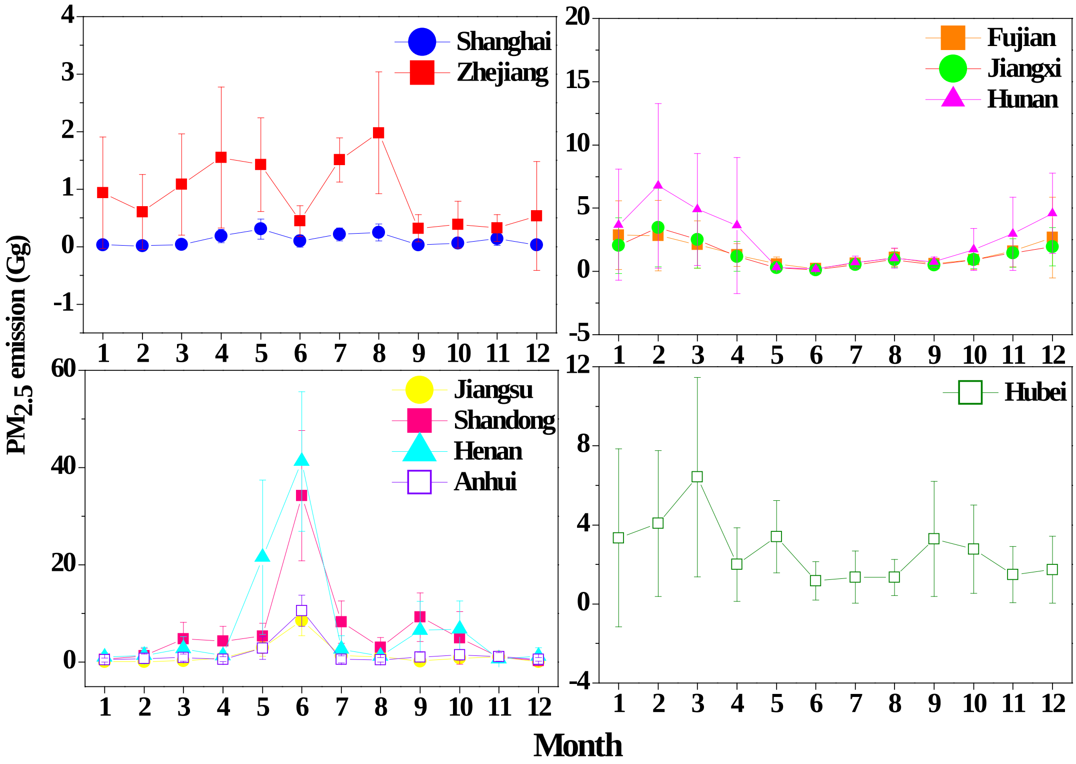


**Figure 9.** The monthly $PM_{2.5}$ emission from open biomass burning in each province.

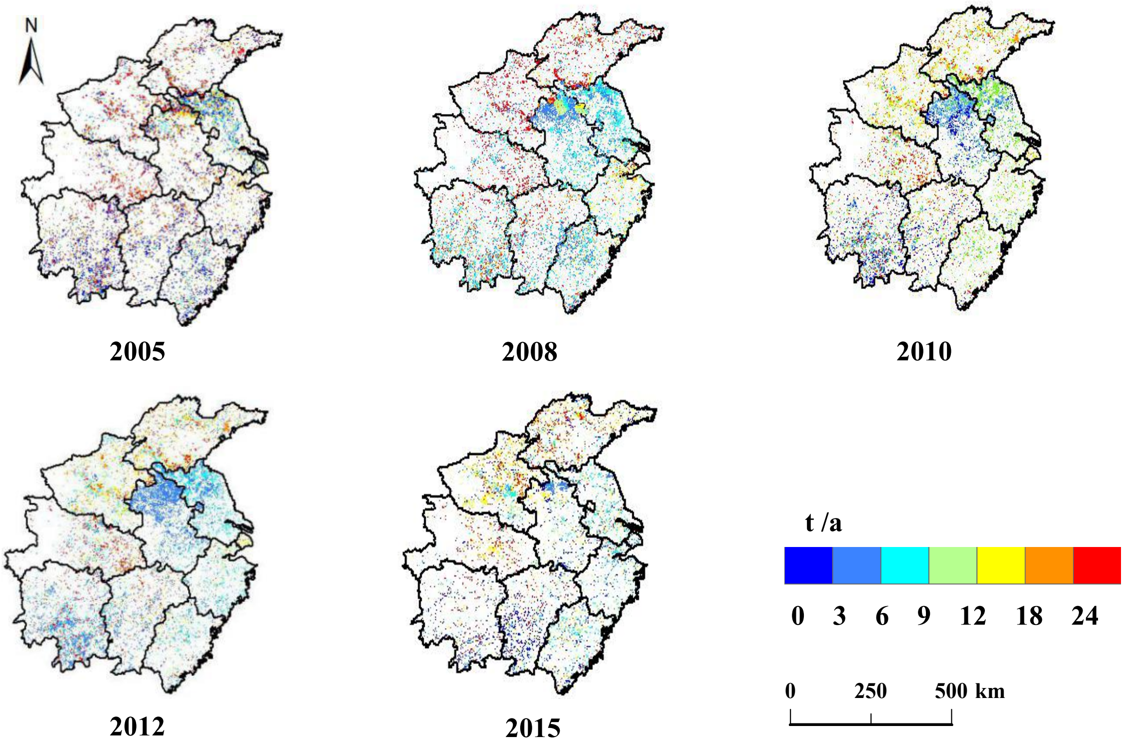



**Figure 10.** Annual spatial distribution (1 km × 1 km) of $PM_{2.5}$ emission from opening biomass burning in Central and Eastern China.

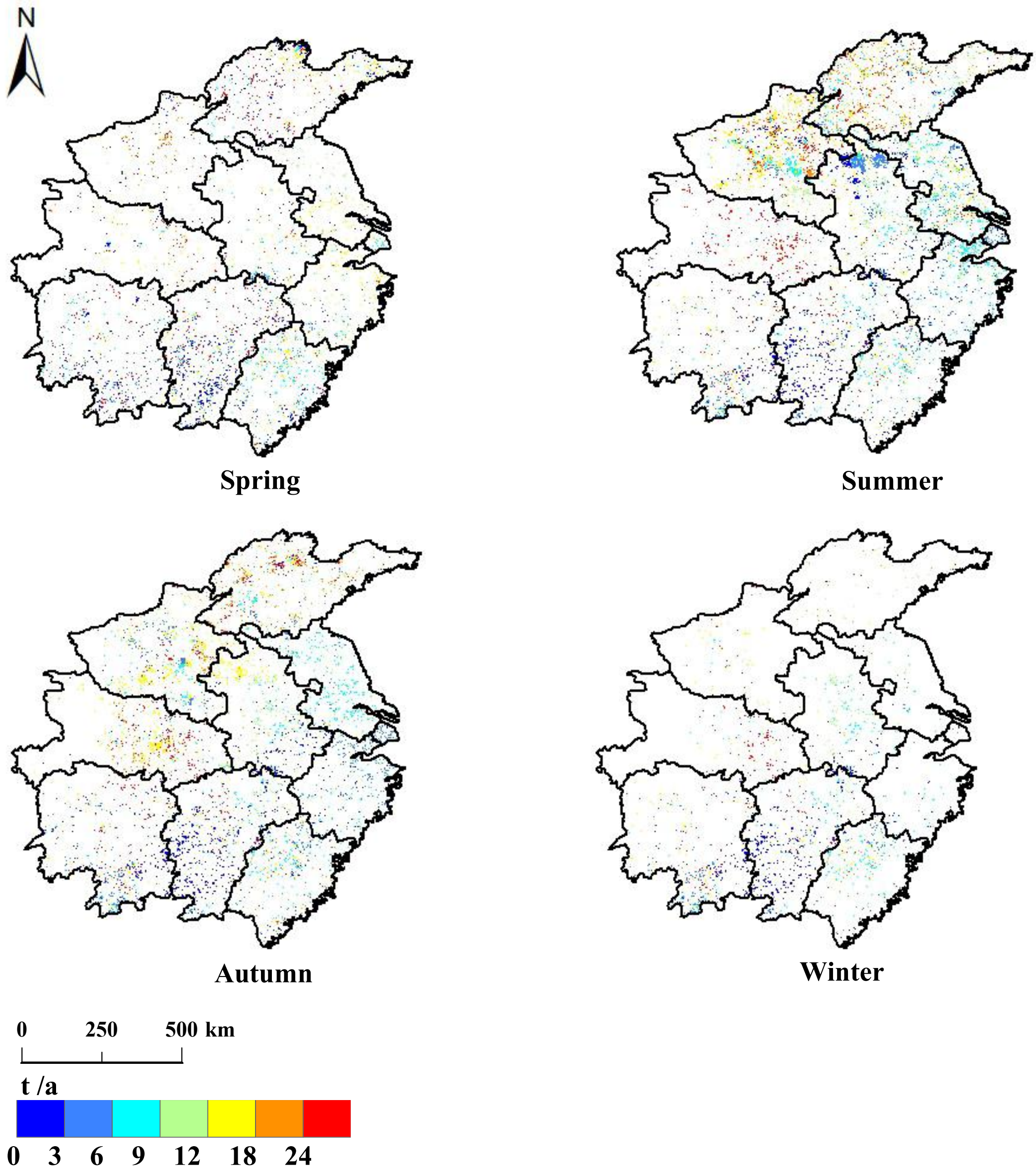
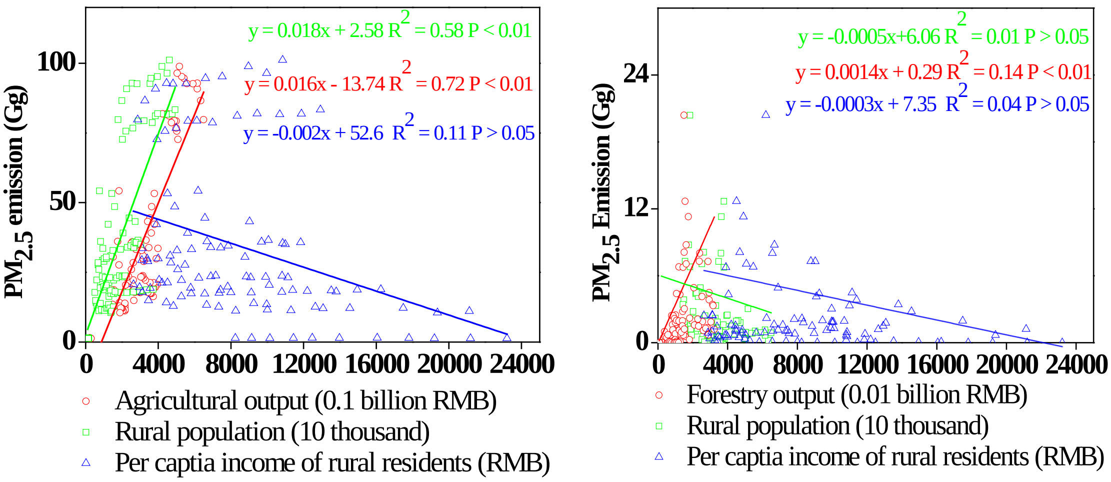



**Figure 11.** Seasonal emission distribution (1 km × 1 km) of $PM_{2.5}$ in 2015 from opening biomass burning in Central and Eastern China.




(a) (b)

**Figure 12.** Correlation between $PM_{2.5}$ emission from crop residue burning and agricultural output , rural population, per captia incomes of rural residents (a) and correlation between $PM_{2.5}$ emission from forestry fire burning and forestry output, rural population, per captia incomes of rural residents (b) in different provinces from 2003 to 2015.