Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018 © Author(s) 2018. CC BY 4.0 License.



5



# Estimating the open biomass burning emissions in Central and

- 2 Eastern China from 2003 to 2015 based on satellite observation
- 3 Jian Wu <sup>1</sup>, Shaofei Kong <sup>2</sup>, Fangqi Wu <sup>2</sup>, Yi Cheng <sup>2</sup>, Shurui Zheng <sup>2</sup>, Qin Yan <sup>1</sup>, Huang Zheng <sup>2</sup>, Guowei Yang <sup>2</sup>,
- 4 Mingming Zheng <sup>1</sup>, Dantong Liu <sup>3</sup>, Delong Zhao <sup>4</sup> and Shihua Qi <sup>1,5</sup>
- 6 Department of Environmental Science and Technology, School of Environmental Studies, China University of
- 7 Geosciences, Wuhan, 430074, China
- 8 Department of Atmospheric Sciences, School of Environmental Studies, China University of Geosciences, Wuhan,
- 9 430074, China
- 3 Centre for Atmospheric Sciences, School of Earth and Environmental Sciences, University of Manchester,
- 11 Manchester M13 9PL, UK
- <sup>4</sup> Beijing Weather Modification Office, Beijing 100089, China
- 13 State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan,
- 14 430074, China

15

17

- 16 Correspondence to: Shaofei Kong (kongshaofei@cug.edu.cn); Shihua Qi (shihuaqi@cug.edu.cn)
- 18 Abstract. Open biomass burning (OBB) has significant impacts on air pollution, climate change and potential
- 19 human health. OBB has raised wide attention but with few focus on the annual variation of pollutant emission.
- 20 Central and Eastern China (CEC) is one of the most polluted regions in China. This study aims to provide a state-of
- 21 the-art estimation of the pollutant emissions from OBB in CEC from 2003 to 2015, by adopting the satellite
- 22 observation dataset (the burned area product (MCD64Al) and the active fire product (MCD14 ML)), local biomass
- data (updated biomass loading data and high-resolution vegetation data) and local emission factors. Monthly
- emissions of pollutants were estimated and allocated into a  $1 \times 1$  km spatial grid for four types of OBB including
- 25 grassland, shrubland, forest and cropland. From 2003 to 2015, the emissions from forest, shrubland and grassland
- 26 fire burning had a minor annual variation whereas the emissions from crop straw burning steadily increased. The
- 27 cumulative emissions of OC, EC, CH<sub>4</sub>, NO<sub>3</sub>, NMVOC, SO<sub>2</sub>, NH<sub>3</sub>, CO, CO<sub>2</sub> and PM<sub>2.5</sub> were 3.64×10<sup>3</sup>, 2.87×10<sup>2</sup>,
- 28  $3.05 \times 10^3$ ,  $1.82 \times 10^3$ ,  $6.4 \times 10^3$ ,  $2.12 \times 10^2$ ,  $4.67 \times 10^3$ ,  $4.59 \times 10^4$ ,  $9.39 \times 10^5$  and  $4.13 \times 10^2$  Gg in these years,
- 29 respectively. For cropland, corn straw burning was the largest contributor for all pollutant emissions, by 84%-96%.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018







Among the forest, shrubland, 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 were populated in the connection area of Shandong, Henan, Jiangsu and Anhui, with emission intensity higher than 100 ton per pixel, which was related to the frequent agricultural activities in these regions. The monthly emission peak of pollutants occurred in summer and autumn harvest periods including May, June, September and October, at which period ~50% of pollutants were emitted for OBB. This study highlights the importance in controlling the crops straw burning emission. From December to March of the next year, 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, local burning habits, anthropological activities and management policies are all influence factors for OBB emissions. The successful adoption of double satellite dataset for long term estimation of pollutants from OBB with a high spatial resolution can support the assessing of OBB on regional air-quality, especially for harvest periods or dry seasons. It is also useful to evaluate the effects of annual OBB management policies in different regions.

Keywords: emission estimation; open biomass burning; Central and Eastern China; high spatial-temporal resolution;

46 satellite dataset

# 1. Introduction

Open biomass burning (OBB), which includes forest, shrubland, grassland and crop residues fire burning (van der Werf et al., 2010; Qiu et al., 2016), is one of the most important sources of gaseous and particulate matter (PM) especially for fine particulate matter (PM<sub>2.5</sub>) and associated carbonaceous aerosols (elemental carbon (EC) and organic carbon (OC)) (Zha et al., 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 average annual submicron EC emission and 65% of primary OC emission globally (Bond et al., 2013), and impacted more than 45% of PM<sub>2.5</sub> concentration on days of heavy air pollution (Deng, 2011). The pollutants of high emission amounts from OBB have 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 the research in 1970s (Crutzen et al., 1979), multi-scale estimation of biomass burning emissions has been a research hot topic from global (Seiler and Crutzen et al., 1980; Levine, 1995; Liousse et al., 1995; Bond et

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





60 al., 2004; Randerson et al., 2012; Kaiser et al., 2012) to regional scale (Yevich and Logan, 2003; Chang et al., 2010; 61 Liousse et al., 2010; Li et al., 2017). China is suffering from severe air pollution with hundred millions of open 62 biomass burned each year (Zhang et al., 2015). The quantitative estimation of pollutants emission for the whole 63 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 64 65 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 (Anhui and Shandong), the Yangtze River delta 66 (YRD, including Zhejiang, Jiangsu and Shanghai) and part of the Pan-Pearl River delta (Fujian and Jiangxi)) 67 68 (Figure 1), is an area with plenty of vegetation coverage (Figure S1). Yin et al (2017) indicated that the crop residue fire burning in summer harvest time can lead to the increase of PM<sub>2.5</sub> concentration in China's middle-east region. 69 70 As one of the most heavily polluted regions in China (Chang et al., 2009; Fu et al., 2013), many large cities are 71 included in this region, such as Nanjing, Wuhan, Shanghai and Hangzhou. Former studies have highlighted the role 72 of OBB on worsening air quality regionally or at megacities, especially for crop residue burning at harvest periods 73 (Yamaji et al., 2010; Zhu et al., 2010; Yin et al., 2011; Huang et al., 2012b; Su et al., 2012; Cheng et al., 2014; 74 Zhou et al., 2016; Zhang et al., 2017). 75 Previous studies mainly focused on crop residue burning emissions with relatively low spatial and temporal 76 resolution (Yamaji et al., 2010; Huang et al., 2012b), which may limit its adoption in air quality modeling to give 77 an accurate result. An accurate estimation of monthly emissions from OBB with a long timescale and high spatial 78 resolution is still limited. It should be noted that, the OBB activities owned spatial-temporal variation properties 79 and have changed greatly during the last two decades in China, especially for forestland fire burning (Huang et al., 80 2011) and crop residue burning, in view of related policies implementation (as listed in Table S1 and Table S2 of 81 Supplementary File). As a big agricultural country, the Chinese government has placed a high priority on 82 environmental pollution prevention caused by OBB. From 1965 to 2015, 51 policy documents for crop straw 83 management has been formulated and 34 policy documents were introduced after 2008 (Chen et al., 2016). Up to 84 now, few studies have accurate estimated the biomass burning emissions in a long time period (Fu et al., 2013; 85 Cheng et al., 2014). The role of the pollution prevention policy on the spatial-temporal variation of pollutants 86 emitted needs to be better clarified. 87 In addition, most previous studies used the top-down method (Seiler and Crutzen et al., 1980) to estimate emission amounts from OBB by national or provincial statistical data and then the total emission amounts 88 pollutants were re-allocated in grids by population, land cover area, or even equal sharing, which is one of the key 89

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





90 reasons for the high uncertainties of OBB emission inventories (Streets et al., 2003; Klimont and Streets., 2007; 91 Gadde et al., 2009; He et al., 2013; Zhou et al., 2015; Zhou et al., 2017). Quantitative estimates of biomass burning 92 were highly improved by the satellite observations of fire burned area or active burning fires (Freitas et al., 2005; 93 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 spatio-temporal 94 95 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 96 97 burned area detection was improved by burned area products (e.g., GBA2000 product (Ito and Penner, 2004; 98 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 99 100 emissions (Duncan et al., 2003; He et al., 2015). One is the burned area product, which provides fire burned areas 101 of the whole month, is limited by the lower pixel resolutions. The size of many small burn scars was below the 102 detection limit of these products (Eva and Lambin, 1998; Laris, 2005; McCarty et al., 2009; Roy and Boschetti, 103 2009). Therefore, the contribution of small fires to fire burned areas and corresponding fire burning emissions are 104 still poorly understood (Randerson et al., 2012). The other is the active fire product, which can provide information on small fire locations, occurrence time and small fire burned area (Prins and Menzel, 1992; Giglio et al., 2006; 105 106 Chuvieco et al., 2008; Roberts et al., 2009; Aragao and Shimabukuro, 2010; Bowman et al., 2011; Lin et al., 2012; 107 Arino et al., 2012). The uncertainty of fire detection was mainly due to the limitation of satellite overpass periods. 108 To reduce the uncertainty of emission estimation by satellite products mainly raised by the missing of small burning 109 areas, the combination of two satellite dataset has recently been proved to be an effective practice (Qiu et al., 2016). 110 The lack of local biomass data (biomass loading data and vegetation speciation data) and local emission 111 factors could also introduce uncertainty in emission estimates. At present, the local biomass loading data has not 112 been updated and still needs to be accurately measured. In addition, local high spatial-resolution vegetation 113 speciation data was also rarely adopted in OBB estimations. Meanwhile, a lot of researches about OBB have used 114 the same emission factors without considering the various biomass species and combustion conditions (Andela et 115 al., 2013; Giglio et al., 2013). All these should be considered and improved in the establishment of OBB emission 116 inventory. 117 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 118 to estimate historical OBB emissions from 2003 to 2015 in CEC. High spatial-temporal resolution of emission 119

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





allocation was achieved. The possible driving factors like rural population, economic level, agricultural production, energy and pollution control policies and anthropogenic activities which may impact the spatial distribution and temporal variation of OBB emissions were explored. They had also 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 Figure 2), and emission factors. Fire burning emission amounts are calculated by the following equation (Wiedinmyer et al., 2011; Shi et al, 2015).

$$E_{i} = \sum_{i=1}^{n} BA_{x,\,t} \times CE \times BL_{x} \times EF_{i,\,j} \tag{1} \label{eq:energy}$$

where j stands the different aggregated vegetation types; i stands for different pollutant species;  $E_i$  is the emission amounts of different pollutants;  $BA_{x,t}$  is the total burned area (km<sup>2</sup>) of aggregated vegetation class in location x and time t;  $CE_x$  is defined as the fraction of OBB; BL is the biomass fuel loading (kg) in different location x;  $EF_{i,j}$  is the emission factor of species i for the j vegetation types.

MODIS burned area product (MCD64AL: http://modis-fire.umd.edu/) and the 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 has a 500 m spatial resolution and monthly temporal resolution, which can accurately detect the burning area at 500 m pixel. Nevertheless, a much lower pixel resolution burning is difficult to detect by this satellite. The detection of burned area was often affected by weather conditions or cloud cover. Therefore, we use MODIS active fire product MCD14 ML as a supplemental tool to obtain the contribution of small fire burned area. The active fire detection method based on thermal anomalies could detect fires as low as 1/20 of a pixel and could identify much smaller burned areas. However, the active fire product existed as the fire points and could not directly obtain the burned area data. In order to obtain smaller burned areas less than 500 m × 500 m of a land grid cell, the burned areas of small fire were estimated based on the following method (Randerson et al., 2017).

146 
$$BA_{sf(i,t,v)} = FC_{out(i,t,v)} \times \alpha_{(r,s,v)} \times \gamma_{(r,s,v)}$$
 (2)

where  $BA_{sf}$  is the small fire burned area in grid cell i, month t, and aggregated vegetation class v;  $FC_{out}$  is the total number of MCD14 ML active fires outside of the burned area in each 1 km×1 km grid cell;  $\alpha$  is the ratio of

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018







BA<sub>MCD64A1</sub> to the number of active fires with or near the MCD64A1 burned area (km<sup>2</sup>) and  $\alpha$  is equal to the value of surrounding grid cell if BA<sub>MCD64A1</sub> is equal to 0;  $\gamma$  is an additional unit less scalar which indicates the difference between the active fires in MCD64A1 burning area and active fires outside the burning area and  $\gamma$  is assumed

# 2.2 Biomass fuel loadings

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, for forest land, the biomass loading data between 2003 and 2008 was collected from Fang et al (1996). From 2008-2015, the update forest loading data was calculated based on the 8<sup>th</sup> Chinese National Forest Resource Inventory. The forest biomass density data were estimated (Table 1) by the following equation:

$$B_i = \frac{T_i}{A_i} \tag{3}$$

equal to 1 in this research; r denotes the burning region. s indicates the burning period.

where i stands for different forest species (broadleaf forest, needleleaf forest and mixed forest); B is the biomass density; T means the total biomass; A denotes the total area of forest.

The total biomass of different forest species were calculated based on the forest stock volume derived biomass method. The specific calculated method of different forest biomass 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). Meanwhile, the forest stock volume data and the total area of forest were collected from the 8<sup>th</sup> Chinese National Forest Continuous Inventory. As shown in Table 1, the forest biomass density in recent years has changed a lot in recent years, which highlighted the updates for improving the emission inventories of OBB.

For grassland and shrubland, local biomass density data were also collected (Pu et al, 2004; Hu et al, 2006) as listed 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 2003-2015). The detailed data of crop-specific 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 is mainly set as a constant, which may bias the emission estimates. To improve the accuracy, for cropland, the CE was set as 0.68 for legumes and 0.93 for other types (Koopmans and Koppejan, 1997; Wang and Zhang, 2008; Zhang et al., 2011).

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018 © Author(s) 2018. CC BY 4.0 License.



185

186

187

188

189190

191

192193

194 195

196

197

199 200

201

202

203

204

205



For grassland and shrubland, 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:

183 
$$CE_s = e^{-0.13 \times TB}$$
 (4)

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

## 2.4 Emission Factors

Emission factors (EFs) of different OBB were summarized in Table 5. EFs for cropland burning were mainly collected from previous research 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 similar researches (Cao et al, 2008; Tian et al, 2011; GFED4, Version 4; Akagi et al, 2011; He et al, 2015). In addition, some emission factors measured by our previous research 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) as Figure S1 shown together with the burned area of every opening biomass species were 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 emissions in t-th grid were calculated using the following equation:

198 
$$E_{t, j} = \frac{BA_{t, j}}{BA_{i, j}} \times E_{i, j}$$
 (5)

Where  $E_{t,j}$  is the emissions of different biomass species 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 The influence factors for the OBB emission

Several detailed statistics data in the NBSC were collected, such as the rural population, the 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.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018 © Author(s) 2018. CC BY 4.0 License.



206

207

211

212

235



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 of all the pollutant emissions. 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 shows the cumulative OBB emission amounts during 2003-2015 and historical emissions from 213 different provinces were detailedly listed in Table S3. By the end of 2015, the cumulative emissions of OC, EC, 214 CH<sub>4</sub>, NO<sub>x</sub>, non-methane volatile organic compounds (NMVOCs), SO<sub>2</sub>, NH<sub>3</sub>, CO, CO<sub>2</sub> and PM<sub>2.5</sub> were 3.64×10<sup>3</sup>, 215 216  $2.87 \times 10^2$ ,  $3.05 \times 10^3$ ,  $1.82 \times 10^3$ ,  $6.4 \times 10^3$ ,  $2.12 \times 10^2$ ,  $4.67 \times 10^3$ ,  $4.59 \times 10^4$ ,  $9.39 \times 10^5$  and  $4.13 \times 10^2$  Gg, 217 respectively. In the following section, for better revealing the spatial-temporal variation of OBB emissions, the 218 PM<sub>2.5</sub> variation was detailedly discussed as an example. At the province level from 2013 to 2015, the highest 219 emission amounts of PM<sub>2.5</sub> were found in Henan and Shandong, accounting for 28% and 24% of the total emission 220 amounts, respectively. The lowest emission appeared in Zhejiang and Shanghai, which only contributed for 4% and 221 0.4%. For other provinces, Hunan, Hubei, Fujian, Anhui, Jiangxi and Jiangsu accounted from 5.5% to 10.1% of the 222 whole emission. 223 The contributions of different types of biomass sources for various pollutants were shown in Figure 3a. 224 Cropland burning contributed the most emission for all the pollutants, from 84%-96%. The forest fire also 225 exhibited higher emission of NH3, SO2, NMVOC and PM2.5, accounting for 12%, 11%,7% and 5% of 226 corresponding total emission, respectively. As shown in Figure 3b, for the croplands, wheat, corn and rice straw 227 burning were the top three emission source types for all the pollutants. Corn straw burning contributed the most to 228 SO<sub>2</sub> (48%), NO<sub>x</sub> (37%), NMVOCs (33%), CO (32%) and CO<sub>2</sub> (28%) emission. Highest contributions of EC (45%), 229 OC (33%) and CH<sub>4</sub> (32%) from rice straw burning was found, while wheat straw burning contributed the most 230 (31%) to PM<sub>2.5</sub> emission. 231 In Figure 4, except for Fujian, cropland burning emission was the largest contributor to the PM2.5 emission, 232 with the contributions ranging from 78% (Hunan) to almost 100% (Shanghai). The higher rural agglomeration, 233 abundant crops production and more cropland residue burning activities in these provinces can explain the higher contributions. In Shanghai, one of the most developed cities in China, the highest contribution of cropland burning 234

is not related with the high levels of agricultural activities, but is only due to the lack of emissions from other open

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018







biomass burning sources. Highest contribution from the forest fire burning and shrubland fire burning were found in Fujian as 46% and in Hunan as 21%, respectively. For forest fire burning, the provinces located in South China exhibited higher values, varying from 6% (Hubei) to 44% (Fujian) and for shrubland fire burning, the contributions varied in 1.5% (Hubei)-7.5% (Zhejiang). While for the Northern provinces (Shandong, Henan, Jiangsu and Hubei), the contributions ranged around 0.03% and 1%, respectively, which can be neglected. This is mostly due to the suitable weather conditions, a relative large forest and shrubland coverages and frequent human forestry activities in those provinces as Figure 2 shown. PM<sub>2.5</sub> emissions from grassland were negligible with the following provinces holding the higher contributions: Jiangxi (0.8%), Hunan (0.25%), Anhui (0.1%) and Fujian (0.1%).

From Figure 5, emissions from wheat and corn straw burning mainly concentrated in Shangdong 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<sub>2.5</sub> emission were relatively small, occupied by 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 emission in this study.

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

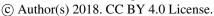
# 3.2.1 Yearly variation

Historical emissions of OBB from 2003 to 2015 in CEC were shown in Figure 6. The multi-year variation tendency of OBB emissions for various pollutants was similar. Take PM<sub>2.5</sub> as example, emissions exhibited clearly increasing trends 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.

In 2008, intensive policies for utilization of straw energy (Table S1) and strengthening the forest fire and grassland fire prevention (Table S2) were published, which effectively limited the emissions from forest and shrubland burning as Figure 7a shown. Peak emissions for PM<sub>2.5</sub> from forest, shrubland and grassland burning were found in 2008, as 49 Gg, 8.9 Gg and 0.7 Gg, respectively. Obvious decreasing was found from 2008 to 2010, to 19 Gg, 4.8 Gg and 0.24 Gg, respectively. Then they exhibited inter-annual oscillation from 2010 to 2015, with higher emission amounts in odd years and lower emission amounts in even years (Jin et al., 2017a). The multi-year

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018







tendency for forest, shrubland and grassland were mainly affected by the variations in climate, management measures and other human forcing. It can also conclude that the yearly variation trends of pollutants from OBB were mainly impacted by the emission from forest, shrubland and grassland burning, but not the crop burning.

The emissions of PM<sub>2.5</sub> from crop 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 explain the increasing of pollutant emission. Meanwhile, from Table S1, the controlling of pollutants from crop residue burning in China started from 1970s, and in 2000, the law for prevention of air pollution was published. Then in 2003, the regulations on straw banning and comprehensive utilization were released. From 2005 to 2015, all the policies were to improve the straw energy utilization to 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<sub>2.5</sub>. From Figure 7b, the large contributions to PM<sub>2.5</sub> (22%-28% and 29%-33%) and increasing trends for corn burning and wheat burning could be found, which should be further focused. The contributions from rice burning slightly decreased in the past decade, by about 19% from 2003 to 2015. Other types of biomass totally accounted for averaged 25% of PM<sub>2.5</sub> emission and all exhibited slightly decreasing trend from 2003 to 2015, increased by about 21%-29%.

Figure 8 showed that the crop burning emission in Henan, Shandong, Anhui, Jiangsu, Hubei, Hunan and Jiangxi exhibited obvious increasing trends, which suggested the importance of crop burning control in these provinces. For Fujian and Zhejiang, no obvious increase for crop burning emissions was found, implying that the emissions have 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 residue 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 scattering 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 10, the PM<sub>2.5</sub> 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

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.



296



297 PM<sub>2.5</sub> 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 (14.3 298 299 Gg-65.9 Gg). As the emission amounts of crop fire burning were one or two magnitude higher than other three 300 types of biomass burning, the month variation of total PM<sub>2.5</sub> variation was mainly controlled by the crop biomass 301 burning (Zhang et al., 2016). The periods with highest PM<sub>2.5</sub> emissions were just the summer and autumn harvest 302 times, when the burning activities are more frequent. The peak of open biomass fire burning occurs in May and 303 June totally accounted for 42% of the whole PM<sub>2.5</sub> emission in 2003-2015, which is caused by the harvest of winter 304 wheat, especially in Henan, Shandong, Jiangsu and Anhui (Figure 9b). 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). 305 306 Though the open biomass burning was strictly forbidden in recent years, scattered burning activities still existed in 307 China. The small peak of open biomass burning emission in September to October (totally accounted for 13.8% of 308 the whole PM<sub>2.5</sub> emission in 2003-2015) can be attributed to the burning of corn straw after corn harvest. As shown 309 in Figure S4, in recent years, the emissions in CEC and major agricultural province during harvest time have shown 310 a rapid decline, in accordance with the change tendency of burned area due to increased government management. 311 Considering of the increase tendency of crops straw burning from year to year, it is worth noting that fire burning 312 out of harvest season as a way of circumventing governmental polices needs to continue to be well regulated. From 313 December to February of the next year, the crop residue burning emissions decreased to the lowest level in the 314 whole year (18.9% of the whole PM2.5 emission in 2003-2015). However, during December to March, the 315 emissions of PM<sub>2.5</sub> from forest, shrubland and grassland exhibited their peak values, totally occupied by 67% of the whole PM<sub>2.5</sub> emission of forest, shrubland and grassland fire burning in 2003-2015. 316 317 Figure 10 clearly listed the monthly average emissions of PM2.5 from OBB in different provinces. These 318 provinces were classified based on the correlation of emissions in each month of 2003-2015. Henan, Shandong, 319 Anhui and Jiangsu provinces (R<sup>2</sup> higher than 0.92, P<0.01), as one of the largest and contiguous wheat planting 320 areas in China (Fang et al., 2014), have two crop rotations. The highest monthly emissions were observed for 321 winter wheat harvesting (sown in October and harvested from May to June) and corn harvesting (sown in middle 322 June and harvested from September to October). A large proportion of crop straw is always burnt directly after the 323 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 324 325 (R<sup>2</sup> higher than 0.9, P<0.01), the largest monthly emissions were observed with forest and shrubland fire burning

The monthly PM<sub>2.5</sub> emission variation of different OBB in CEC was shown in Figure 9a. The total monthly

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.



355



326 during the time between December and March in the next year, which is the dry seasons in these provinces (Li et al., 327 2014; Li et al., 2015). And in other months, the emissions were limited. For Shanghai and Zhejiang ( $R^2 = 0.7$ , 328 P<0.01), lowest levels of PM<sub>2.5</sub> emissions were found, with peak values also occurred in summer and autumn 329 harvest periods. Obvious two peaks were found for April-May and July-August periods, which may reflect the rice 330 harvesting at these times. To sum up, these regional differences of monthly PM<sub>2.5</sub> emissions from OBB were 331 mainly caused by the different biomass burning types and times as well as corresponding environmental conditions. 3.2.3 Spatial distribution within 1 km  $\times$  1 km of PM<sub>2.5</sub> emitted from OBB in CEC 332 333 The spatial distribution of PM<sub>2.5</sub> 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 334 distributions of PM<sub>2.5</sub> emission are shown in Figure 11. It can be found that the OBB was widespread and scattered. 335 336 The average emissions intensity of PM<sub>2.5</sub> ranged from 0 to 15 tons per pixel in most provinces. The variation range 337 is mainly caused by the social-economic development level, rural population and agricultural activities. The highest 338 value in different provinces was all caused by the crops fire burning due to the centralized burning of them in a 339 relatively small area. Some pixels with high emissions exceeding more than 100 tons each year were found in 340 Henan, Shandong and Hunan. It can be attributed to the large amounts of crop straws in these provinces. The pixels 341 of high emission intensity more than 70 tons from crop straw burning were also found in Hubei, Jiangsu and Anhui. 342 For forest and shrubland fire burning, the high emission points from (more than 30 tons per pixel) were found in 343 Fujian and Jiangxi. Lower emission intensities in Zhejiang (lower than 10 tons per pixel on average) and Shanghai 344 (lower than 7 tons on average) were mainly due to the highly developed economy and limited agricultural activities 345 (Su et al., 2012). In addition, northern Anhui and eastern Jiangsu were found high emissions of OBB with a 346 relatively lower intensity (lower than 15 tons per pixel on average), which may be due to that the crop straw was 347 burned in a large area in these regions. 348 Though the emission intensities varied in the past ten years, the areas with high emission amounts are 349 uniformed. They were mainly located in the main agricultural areas in eastern Henan, southern Shandong, northern 350 Anhui, northern Jiangsu, eastern Hubei and northern Hunan. This result is in accordance with formers (Huang, et 351 al., 2012b). The junction regions of the four provinces-Henan, Shandong, Anhui and Jiangsu should be paid more 352 attention, where the pollutants emission from OBB jointed together. This was similar to a recent research (Jin et al., 353 2017b). This region belongs to HuangHuai Plain, with large area of croplands and low economic development levels. The opening burning activities and corresponding banning policies are both abundant in village scale. The 354

game of "cat and mouse" is frequently acted. More effective policies for guiding or helping farmers to utilize straw

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





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 low 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 12 highlights the spatial distribution of PM<sub>2.5</sub> emitted from OBB in different seasons of 2015. Emissions were more concentrated in summer, followed by winter. In summer, the emission was mainly concentrated in the connection regions of Henan, Shandong, Anhui and Jiangsu, mainly raised by crop straw 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, anthropogenic activities, rural population, local economic level, agricultural level and pollution controlling policies. Local burning habits have a great influence on different types of OBB emissions. In 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. The 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 in forest and shrubland burning, resulting in the elevated levels of corresponding emission in these provinces.

Some anthropogenic activities 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 their 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 can occupied by 22%-38% of the whole file points in April in CEC in some years (Figure S3). 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

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.



386

387

388

389

390

391

392

393

394

395 396

397

398

399

400

401 402

403

404

405

406

407

408

409

410

411

412

413

414

415



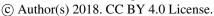
the Spring Festival can easily lead to grass, shrub and forest fires. All these activities can affect the emission levels in a short time scale.

In order to understand the impact of the rural population, local economic level and agricultural level, correlation analysis between PM<sub>2.5</sub> emissions from OBB and statistics data (the rural population, the per capita net income of rural residents and agricultural output (crop straw burning) and forestry output (forest, shrubland and grassland burning)) in different provinces were conducted. For crop residue burning, significant positive correlations were found between the rural population, agricultural output and the PM2.5 emissions from crops straw burning for the whole CEC (Figure 13a). The high rural population and agricultural output indicates 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<sub>2.5</sub> 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<sub>2.5</sub> emission. Then we calculated the correlations between the change tendency of PM2.5 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<sub>2.5</sub> emission and per capita income of rural residents and agricultural output (most R<sup>2</sup> higher than 0.58, P<0.01) and negative correlation were found for PM<sub>2.5</sub> emission with rural population (most R<sup>2</sup> higher than 0.7, 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 of this, more crop residues were directly burned in the agricultural field. But it was not suitable to Shanghai, Zhejiang and Fujian, where holds less crop residue production and high utilization efficiency of crop straws.

Positive correlations were also found between forestry output and PM<sub>2.5</sub> emission from forestland, shrubland and grassland burning (most R<sup>2</sup> higher than 0.5, P<0.05) in the whole CEC (Figure 13b), and it 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<sub>2.5</sub> emission and other statistics data (the rural population and the per capita net income of rural residents) (Figure 13b and Table S5). It may indicate that the forestry fire burning activities were not predominantly affected by the rural human living activity. According to previous studies, forestry fire burning was affected by environmental

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018







conditions and human activities with environmental factors have 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 OC, EC, CH<sub>4</sub>, NO<sub>x</sub>, NMVOC, NH<sub>3</sub>, CO<sub>2</sub> and CO emissions are close, with the differences ranging from -41% to 12 %. The difference in SO<sub>2</sub> and PM<sub>2.5</sub> emission is relative high. The differences were mainly caused by the accuracy of biomass data, the burned ratio for various crop types and the selection of EFs. The results in this study can decrease the uncertainty from statistical data for forest, shrubland and grassland fire burning, as there are limited forestry statistical data. Compared with Huang et al. (2012), who use the same emission factors for different crops straw, the estimate in our study is more accuracy. An obvious underestimation of PM<sub>2.5</sub> emission from crop straw burning were found in Jin et al. (2017), in which all the crop species in the study were not 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 products, 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 some actual agricultural fire activities could not be detected (van der Werf et al., 2010). The lower emission of CO<sub>2</sub>, NMVOC, SO<sub>2</sub> and NOx in our study is due to the smaller EFs values used. Our emission estimation of the pollutants is more similar to the results in 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<sub>4</sub>, NO<sub>x</sub>, NMVOC, NH<sub>3</sub> and CO<sub>2</sub>, 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. Meanwhile, the updated forest loading data also reduced the uncertainty of pollutant emissions from forest fire burning. At the same time, the EFs used for various biomass burning types, the crop specific residue to production ratio data and the burned ratio for various crop types were all localized in CEC in this study. The combination of multiple satellite products with local EFs data and updated local biomass data can improve the estimation of pollutant emission from OBB effectively.

# 3.5 Uncertainty analysis

Emission uncertainties in our study were associated with the fire satellite products, biomass fuel loading data, combustion efficiency and emission factors. The estimation for large fires was proved to be reliable for burned area product MCD64AL (Giglio et al., 2013). For the active fire product MCD14ML, the uncertainty was mainly caused by the satellite passing time. The small fires which burned in 10:30 am-1:30 pm could not be captured by MCD14ML. The uncertainty of biomass fuel loading data was estimated to be approximately 50% (Shi et al., 2015)

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





446 and the uncertainty of EFs of each pollutant ranging from 0.03 to 0.85. At last, in order to evaluate the estimation 447 uncertainty quantitatively, the Monte Carlo method was used. Pollutant emissions were estimated from 20, 000 448 Monte Carlo simulations with a 95% coincidence interval. Table 9 shows the emission uncertainty for different 449 pollutants for each year of 2003-2015. On average, the uncertainty of the estimated OC, EC, CH<sub>4</sub>, NO<sub>x</sub>, NMVOCs, CO, SO<sub>2</sub>, NH<sub>3</sub>, CO<sub>2</sub> and PM<sub>2.5</sub> were (-30%, 30%), (-48%, 48%), (-20%, 20%), (-20%, 20%), (-45%, 45%), (-18%, 450 451 18%), (-45%, 45%), (-35%, 35%), (-3%, 3%) and (-36%, 36%), respectively. 452 Compared with previous studies, for the emission estimation of forest burning, the uncertainty was improved by the updated forest fuel loading data. For cropland, the uncertainty was improved by the adoption of local 453 454 grain-straw ratio data and the crop residue burned ratio data based on survey results. Compared with the constant combustion efficiency in previous researches, the activity combustion efficiency data could also reduce the 455 456 uncertainty as they could more accurately reflect the actual combustion conditions (Chen et al., 2013). Meanwhile, 457 the local measured EFs data for different biomass burning species from previous researches also improved the 458 accuracy of the estimation. Therefore, due to the adoption of multiple satellite products, local high resolution 459 vegetation data, updated local biomass distribution data and local emission factors, the estimation of emissions in

# 4 Conclusions

our study is relatively more reliable.

460

461

462

463

464

465

466 467

468

469

470

471

472

473

474 475 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 burning were considered.

Crop residue burning was the major source type for pollutant emissions, followed by forest fire and shrubland 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. 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<sub>2</sub>, NO<sub>X</sub>, CO, NMVOC, CO<sub>2</sub>, NH<sub>3</sub>; OC, EC and CH<sub>4</sub> emissions were mainly produced by rice straw burning; wheat straw burning was the largest contributor to PM<sub>2.5</sub>. The spatial distribution of opening biomass residue burning in different years is similar. The high emissions are mainly found in the major agricultural areas in eastern Henan, southern Shandong, northern Anhui, northern Jiangsu, eastern

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





Hubei and northern Hunan, due to their abundant agricultural activities cultivated areas and low straw utilization efficiency.

From 2003 to 2015, the multi-year tendency of opening biomass residue burning emission for various pollutants is 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 minor fluctuation from year to year, 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 from May to 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 the fire burning out of harvest season should also be paid more attention in recent years. Meanwhile, emissions from forest and shrubland accounted for the vast majority of total emissions in December to March of the next year. The rural population, agricultural output and economic levels impacted on the emission of crop residue burning while the emissions from forestland, shrubland and grassland burning were more affected by environmental conditions.

The estimation of historical emissions by satellite data in this study will provide a fundamental role in assessing the role of pollution prevention policies on open burning activities published in the last decade. The high-spatial  $(1 \times 1 \text{ km})$  resolution monthly emission inventory is also useful in modeling regional air quality and human health risks in the future.

# Acknowledgements

This study was financially supported by the Key Program of Ministry of Science and Technology of the
People's Republic of China (2016YFA0602002; 2017YFC0212602), the Key Program for Technical Innovation of
Hubei Province (2017ACA089). The research was also supported by the Fundamental Research Funds for the
Central Universities, China University of Geosciences, Wuhan.

# References

- 502 Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D. and Wennberg, P.
- 503 O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmospheric
- 504 Chemistry and Physics, 11(9), 4039-4072, doi:10.5194/acp-11-4039-2011, 2011.
- 505 Andreae, M. O. and Rosenfeld, D.: Aerosol-cloud precipitation interactions. Part 1, The nature and sources of

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 506 cloud-active aerosols, Earth Sci. Rev., 89, 13–41, doi:10.1016/j.earscirev.2008.03.001, 2008.
- 507 Andela, N., Schultz, M., Van, der W., Van Leeuwen, T. T., Kaiser, J. W., Wooster, M. J., Heil, A. and Remy, S.:
- Assessment of the Global Fire Assimilation System (GFASv1), MACC-II Deliverable D\_31.2, 2013.
- 509 Andreae, M. O and Merlet, P: Emission of trace gases and aerosols from biomass burning, Global Biogeochemical
- 510 Cycles, 15(4), 955-966, 2001.
- 511 Ao, H. J., Zou, Y. B., Shen, J. B., Peng, S. B., Tang, Q. Y. and Feng, Y. h.: Effects of fertilizer-N application for
- double early rice on the yield, nitrogen use efficiency and soil nitrogen content of double rice, 13(5), 772-780,
- 513 2007 (in Chinese).
- 514 Aragao, L. E. O. C. and Shimabukuro, Y. E.: The Incidence of Fire in Amazonian Forests with Implications for
- 515 REDD, Science, 328(5983), 1275-1278, doi:10.1126/science.1186925, 2010.
- 516 Arino, O., Casadio, S. and Serpe, D.: Global night-time fire season timing and fire count trends using the ATSR
- instrument series, Remote Sensing of Environment, 116, 226-238, doi:10.1016/j.rse.2011.05.025, 2012.
- Zhu, B., Su, J. F., Han, Z. W., Y, C and Wang, T. J.: Analysis of a serious air pollution event resulting from crop
- 519 residue burning over Nanjing and surrounding regions., China Environmental Science, 30 (5), 585-592, 2010.
- 520 Bond, T. C.: A technology-based global inventory of black and organic carbon emissions from combustion, Journal
- of Geophysical Research, 109(D14), doi:10.1029/2003JD003697, 2004.
- 522 Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S.,
- 523 Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schultz, M.,
- Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z.,
- 525 Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender,
- 526 C. S.: Bounding the role of black carbon in the climate system: A scientific assessment: BLACK CARBON IN
- 527 THE CLIMATE SYSTEM, Journal of Geophysical Research: Atmospheres, 118 (11), 5380-5552,
- 528 doi:10.1002/jgrd.50171, 2013.
- 529 Bowman, D. M. J. S., Balch, J., Artaxo, P., Bond, W. J., Cochrane, M. A., D'Antonio, C. M., DeFries, R., Johnston,
- 530 F. H., Keeley, J. E., Krawchuk, M. A., Kull, C. A., Mack, M., Moritz, M. A., Pyne, S., Roos, C. I., Scott, A. C.,
- Sodhi, N. S. and Swetnam, T. W.: The human dimension of fire regimes on Earth: The human dimension of
- 532 fire regimes on Earth, Journal of Biogeography, 38 (12), 2223-2236, doi:10.1111/j.1365-2699.2011.02595.x,
- 533 2011
- 534 Cao, G. L., Zhang, X. Y., Wang, D., and Deng, F. C.: Inventory of Atmospheric Pollutants Discharged from
- Biomass Burning in China Continent, China Environment Science, 25, 389-393, 2005 (in Chinese).

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 536 Cao, G. L., Zhang, X. Y., Gong, S. L., and Zheng, F. C.: Investigation on emission factors of particulate matter and
- 537 gaseous pollutants from crop residue burning, Journal of Environmental Sciences, 20 (1), 50-55, 2008.
- 538 Chang, D. and Song, Y.: Estimates of biomass burning emissions in tropical Asia based on satellite-derived data,
- 539 Atmospheric Chemistry and Physics, 10(5), 2335-2351, doi:10.5194/acp-10-2335-2010, 2010.
- 540 Chang, D., Song, Y. and Liu, B.: Visibility trends in six megacities in China 1973-2007, Atmospheric Research,
- 541 94(2), 161-167, doi:10.1016/j.atmosres.2009.05.006, 2009.
- 542 Chen, C., Wang, H., Zhang, W., Hu, D., Chen, L. and Wang, X.: High-resolution inventory of mercury emissions
- from biomass burning in China for 2000-2010 and a projection for 2020: MERCURY EMISSION FROM
- BIOMASS BURNING, Journal of Geophysical Research: Atmospheres, 118(21), 12,248-12,256, doi:
- 545 10.1002/2013JD019734, 2013.
- 546 Chen, C. L., Yang, Y. and Xie, G. H.: Study of the development of crop straw management policy in China, Journal
- of China Agricultural University, 21, 1-11, 2016 (in Chinese).
- 548 Chen, H. F., Lin, R. Y., Liang, Y. Y., Zheng, L. D., Liang, K. J., and Lin, W. X.: Dry-matter accumulation and
- transportation in first-rice crop of early rice-ratoon rice under different cultivation patterns, 16(1), 129-133,
- 550 2008 (in Chinese).
- 551 Cheng, Z., Wang, S., Fu, X., Watson, J. G., Jiang, J., Fu, Q., Chen, C., Xu, B., Yu, J., Chow, J. C. and Hao, J.:
- Impact of biomass burning on haze pollution in the Yangtze River delta, China: a case study in summer 2011,
- 553 Atmospheric Chemistry and Physics, 14(9), 4573-4585, doi:10.5194/acp-14-4573-2014, 2014.
- 554 Chuvieco, E., Giglio, L. and Justice, C.: Global characterization of fire activity: toward defining fire regimes from
- 555 Earth observation data, Global Change Biology, 14(7), 1488-1502, doi:10.1111/j.1365-2486.2008.01585.x,
- 556 2008.
- 557 Cooke, W. F., Koffi, B. and Grégoire, J.-M.: Seasonality of vegetation fires in Africa from remote sensing data and
- application to a global chemistry model, Journal of Geophysical Research Atmospheres, 101(D15),
- 559 21051-21066, 1996.
- 560 Crutzen, P. J., Heidt, L. E., Krasnec, J. P., Pollock, W. H., and Seiler, W.: Biomass Burning as a source of
- 561 Atmospheric Gases CO, H<sub>2</sub>, N<sub>2</sub>O, NO, CH<sub>3</sub>CL and COS, Nature, 282, 253–256, doi:10.1038/282253a0, 1979.
- 562 Crutzen, P. J. and Andreae, M. O.: Biomass burning in the tropics: impact on atmospheric chemistry and
- 563 biogeochemical cycles, Science, 250, 1669-1678, 1990.
- 564 Deng. C. R.: Identification of biomass burning source in aerosols and the formation mechanism of haze, PhD
- dissertation, Fudan University, Shanghai, 2011 (in Chinese).

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 566 Duncan, B. N.: Interannual and seasonal variability of biomass burning emissions constrained by satellite
- 567 observations, Journal of Geophysical Research, 108(D2), doi:10.1029/2002JD002378, 2003.
- 568 EPD: Guide for compiling atmospheric pollutant emission inventory for biomass burning, Environmental
- 569 Protection Department, available at:
- 570 http://www.zhb.gov.cn/gkml/hbb/bgg/201501/t20150107\_293955.htm, 2014 (in Chinese).
- 571 Fang, J. Y., Liu, G. H. and Xu, S. L.: Biomass and net production of forest vegetation in China, Acta Ecologica
- 572 Sinica, 16(5), 497-508, 1996.
- 573 Fang, S., Qi, Y., Han, G., Zhou, G. and Cammarano, D.: Meteorological drought trend in winter and spring from
- 574 1961 to 2010 and its possible impacts on wheat in wheat planting area of China, Scientia Agricultura Sinica,
- 575 47(9), 1754-1763, 2014.
- 576 Freitas, S. R., Longo, K. M., Dias, M. A. F. S., Dias, P. L. S., Chatfield, R., Prins, E., Artaxo, P., Grell, G. A. and
- 577 Recuero, F. S.: Monitoring the transport of biomass burning emissions in South America, Environmental Fluid
- 578 Mechanics, 5(1-2), 135-167, 2005.
- 579 Fu, X., Wang, S., Zhao, B., Xing, J., Cheng, Z., Liu, H. and Hao, J.: Emission inventory of primary pollutants and
- 580 chemical speciation in 2010 for the Yangtze River Delta region, China, Atmospheric Environment, 70, 39-50,
- 581 doi:10.1016/j.atmosenv.2012.12.034, 2013.
- 582 Gadde, B., Bonnet, S., Menke, C. and Garivait, S.: Air pollutant emissions from rice straw open field burning in
- 583 India, Thailand and the Philippines, Environmental Pollution, 157(5), 1554-1558,
- 584 doi:10.1016/j.envpol.2009.01.004, 2009.
- 585 Giglio, L., Csiszar, I. and Justice, C. O.: Global distribution and seasonality of active fires as observed with the
- Terra and Aqua Moderate Resolution Imaging Spectroradiometer (MODIS) sensors: Global fire distribution
- 587 and seasonality, Journal of Geophysical Research: Biogeosciences, 111(G2), n/a-n/a,
- 588 doi:10.1029/2005JG000142, 2006.
- 589 Giglio, L., Randerson, J. T. and Werf, G. R. V. D.: Analysis of daily, monthly, and annual burned area using the
- 590 fourth generation global fire emissions database (GFED4), Journal of Geophysical Research: Biogeosciences,
- 591 118(1), 317-328, 2013a.
- 592 Giglio, L., Randerson, J. T. and van der Werf, G. R.: Analysis of daily, monthly, and annual burned area using the
- 593 fourth-generation global fire emissions database (GFED4): ANALYSIS OF BURNED AREA, Journal of
- 594 Geophysical Research: Biogeosciences, 118(1), 317-328, doi:10.1002/jgrg.20042, 2013b.
- 595 He, M., Zheng, J., Yin, Shasha and Zhang, Y.: Trends, temporal and spatial characteristics, and uncertainties in

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 596 biomass burning emissions in the Pearl River Delta, China, Atmospheric Environment, 45(24), 4051-4059,
- 597 2011.
- 598 He, M., Wang, X. R., Han, L., Feng, X. Q., and Mao, X.: Emission Inventory of Crop Residues Field Burning and
- 599 Its Temporal and Spatial Distribution in Sichuan Province, Environment Science, 36, 1208-1216, 2015 (in
- 600 Chinese).
- Huang, X., Li, M., Friedli, H. R., Song, Y., Chang, D. and Zhu, L.: Mercury Emissions from Biomass Burning in
- 602 China, Environmental Science & Technology, 45(21), 9442-9448, doi: 10.1021/es202224e, 2011.
- 603 Huang, X., Li, M., Li, J. and Song, Y.: A high-resolution emission inventory of crop burning in fields in China
- based on MODIS Thermal Anomalies/Fire products, Atmospheric Environment, 50, 9-15,
- doi:10.1016/j.atmosenv.2012.01.017, 2012a.
- 606 Huang, X., Song, Y., Li, M., Li, J. and Zhu, T.: Harvest season, high polluted season in East China, Environmental
- Research Letters, 7(4), 044033, doi:10.1088/1748-9326/7/4/044033, 2012b.
- 608 Hugh, E. and Eric F, L.: Remote Sensing of Biomass Burning in Tropical Regions: Sampling Issues and
- Multisensor Approach, 1998.
- 610 Hui-Feng, H. U. and Bo-Jie, F. U.: Vegetation carbon storage of major shrublands in China, Journal of Plant
- 611 Ecology, 30(4), 539-544, 2006.
- 612 Ito, A.: Global estimates of biomass burning emissions based on satellite imagery for the year 2000, Journal of
- Geophysical Research, 109(D14), doi: 10.1029/2003JD004423, 2004.
- 614 Ito, A., Ito, A. and Akimoto, H.: Seasonal and interannual variations in CO and BC emissions from OBB in
- 615 Southern Africa during 1998-2005: SEASONAL CO/BC EMISSIONS, Global Biogeochemical Cycles, 21(2),
- 616 n/a-n/a, doi: 10.1029/2006GB002848, 2007.
- 517 Jin, Q. F., Ma, X. Q., A., Wang, W. H., Yang, S. Y. and Guo, F. T.: Temporal and spatial dynamics of pollutants
- emission from forest fires in Fujian during 2000-2010, China environment science, 37, 476-485, 2017a (in
- Chinese).
- 620 Jin, Q. F., Ma, X. Q., A., Wang, W. H., Yang, S. Y. and Guo, F. T.: Temporal and spatial variations of PM\_(2.5)
- emissions from crop straw burning in eastern China during 2000—2014, Acta Scientiae Circumstantiae, 37,
- 622 460-468, 2017b (in Chinese).
- 623 Kaiser, J. W., Benedetti, A., Detmers, R., Heil, A., Morcrette, J. J., Schultz, M. G., Van, der W., Wooster, M. J.
- 624 and Xu, W.: Assimilation of FRP Observations for Global Fire Emission Estimation in MACC-II, in EGU
- General Assembly Conference, p. 10521., 2012a.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 626 Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J.-J., Razinger, M.,
- 627 Schultz, M. G., Suttie, M. and van der Werf, G. R.: Biomass burning emissions estimated with a global fire
- 628 assimilation system based on observed fire radiative power, Biogeosciences, 9(1), 527-554,
- 629 doi:10.5194/bg-9-527-2012, 2012b.
- 630 Klimont, Z. and Streets, D.: Emission inventories and projections for assessing hemispheric or intercontinental
- transport, Hemispheric Transport of Air Pollution, 2007.
- 632 Koopmans, A. and Koppejan, J.: Agricultural and Forest Residues-Generation, Utilization and Availability, 6, 1997.
- 633 Laris, P. S.: Spatiotemporal problems with detecting and mapping mosaic fire regimes with coarse-resolution
- satellite data in savanna environments, Remote Sensing of Environment, 99(4), 412-424,
- 635 doi:10.1016/j.rse.2005.09.012, 2005.
- 636 Lei, E., Tang, Q. Y., Luo, H. B., Chen, L. J.: Comparison of late maturing spring maize varieties in paddy field and
- its correlation analysis, Crop research, 23(1), 24-29, 2009 (in Chinese).
- 638 Levine, J. S., Iii, W. R. C., Jr, D. R. C. and Winstead, E. L.: A DRIVER FOR GLOBAL CHANGE, Environ. Sci.
- 639 Technol., 1995.
- 640 Li, C., Hu, Y., Zhang, F., Chen, J., Ma, Z., Ye, X., Yang, X., Wang, L., Tang, X., Zhang, R., Mu, M., Wang, G., Kan,
- 641 H., Wang, X. and Mellouki, A.: Multi-pollutant emissions from the burning of major agricultural residues in
- China and the related health-economic effects, Atmospheric Chemistry and Physics, 17(8), 4957-4988,
- doi:10.5194/acp-17-4957-2017, 2017.
- Li, H. K., Lei, Y. C. and Zeng, W. S.: Forest Carbon Storage in China Estimated Using Forestry Inventory Data,
- Scientla silvae sinicae, 47, 7-12, 2011 (in Chinese).
- 646 Li, J., Li, Y., Bo, Y. and Xie, S.: High-resolution historical emission inventories of crop residue burning in fields in
- 647 China for the period 1990-2013, Atmospheric Environment, 138, 152-161,
- 648 doi:10.1016/j.atmosenv.2016.05.002, 2016.
- 649 Li, L., Liu, W. D., Zou, D. S. and Liu, F.: The correlation between main characteristics and pod yield in peanut
- genotypes under natural waterlogging stress, 30(1), 62-70, 2008 (in Chinese).
- 651 Li, S. M., Yang C. Q., Wang, H. N. and Ge, L. Q.: Carbon storage of forest stands in Shandong Province estimated
- by forestry inventory data, Chinese Journal of Applied Ecology, 25(8), 2215–2220, 2014 (in Chinese).
- 653 Li, X. H., Wang, S. X., Duan, L., Hao, J., Li, C., Chen, Y. S., and Yang, L.: Particulate and trace gas emissions from
- open burning of wheat straw and corn stover in China, Environ. Sci. Technol., 41, 6052-6058, doi:
- 655 10.1021/es0705137, 2007.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 656 Li, Y. P., Wang, J. S., Li Y. H., Wang, S. P. and Sha, S.: Study of the sustainability of droughts in China, Journal of
- Glaciology and Geocryology, 36, 1131-1142, 2014 (in Chinese).
- 658 Li, W. J., ZUO J. Q., Song, Y. L., Liu, J. P., LI, Y., Shen, Y. Y., and Li, J. X.: Changes in Spatio-Temporal
- 659 Distribution of Drought/Flood Disaster in Southern China Under Global Climate Warming, Meteorological
- Monthly, 3, 261-271, 2015 (in Chinese).
- 661 Lin, H. W., Jin, Yufang, GIGLIO, L., Foley, J. A. and RANDERSON, J. T.: Evaluating greenhouse gas emissions
- 662 inventories for agricultural burning using satellite observations of active fires, Ecological Applications, 22(4),
- 663 1345-1364, 2012.
- 664 Liousse, C., Devaux, C., Dulac, F. and Cachier, H.: Aging of savanna biomass burning aerosols: Consequences on
- their optical properties, Journal of Atmospheric Chemistry, 22(1-2), 1-17, 1995.
- 666 Liousse, C., Guillaume, B., Grégoire, J. M., Mallet, M., Galy, C., Pont, V., Akpo, A., Bedou, M., Cast éra, P.,
- Dungall, L., Gardrat, E., Granier, C., Konar é, A., Malavelle, F., Mariscal, A., Mieville, A., Rosset, R., Ser ça,
- D., Solmon, F., Tummon, F., Assamoi, E., Yobou é, V. and Van Velthoven, P.: Updated African biomass burning
- emission inventories in the framework of the AMMA-IDAF program, with an evaluation of combustion
- aerosols, Atmospheric Chemistry and Physics, 10(19), 9631-9646, doi:10.5194/acp-10-9631-2010, 2010.
- 671 Liu, D. M., Liu, Q., Rong, X. M., Peng, J. W., Xie, G. X., Zhang, Y. P. and Song, H. X.: Influences of
- 672 photosynthesis and dry matter accumulation of different oilseed rape cultivars on nitrogen use efficiency,
- Hunan agricultural sciences, 34(9), 29-31, 2010 (in Chinese).
- 674 Liu, M., Song, Y., Yao, H., Kang, Y., Li, M., Huang, X. and Hu, M.: Estimating emissions from agricultural fires in
- the North China Plain based on MODIS fire radiative power, Atmospheric Environment, 112, 326-334,
- doi:10.1016/j.atmosenv.2015.04.058, 2015.
- 677 Lu, J. L., Liang, S. L. and Liu, J.: Study on Estimation of Forest Biomass and Carbon Storage of Shanxi Province,
- 678 Chinese Agricultural Science Bulletin, (31), 51–56, 2012.
- 679 McCarty, J. L., Korontzi, S., Justice, C. O. and Loboda, T.: The spatial and temporal distribution of crop residue
- burning in the contiguous United States, Science of The Total Environment, 407(21), 5701-5712,
- doi:10.1016/j.scitotenv.2009.07.009, 2009.
- 682 MEPC (Ministry of Environmental Protection of China), 2015. Crop residue burning report.
- 683 http://www.zhb.gov.cn/.
- 684 Piao, S. L., Fang, J. Y., He, J. S. and Xiao, Y.: Spatial distribution of grassland biomass in China, Acta
- 685 Phytoecologica Sinica, 28(4), 491-498, 2004.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 686 PJ, C. and MO, A.: Biomass Burning in the Tropics: Impact on Atmospheric Chemistry and Biogeochemical Cycles,
- Springer International Publishing, 2016.
- 688 Prins, E. M. and Menzel, W. P.: Geostationary satellite detection of bio mass burning in South America,
- 689 International Journal of Remote Sensing, 13(15), 2783-2799, 1992.
- 690 Qiu, X., Duan, L., Chai, F., Wang, S., Yu, Q. and Wang, S.: Deriving High-Resolution Emission Inventory of OBB
- 691 in China based on Satellite Observations, Environmental Science & Technology, 50(21), 11779-11786,
- 692 doi:10.1021/acs.est.6b02705, 2016.
- 693 Randerson, J. T., Chen, Y., van der Werf, G. R., Rogers, B. M. and Morton, D. C.: Global burned area and biomass
- burning emissions from small fires: BURNED AREA FROM SMALL FIRES, Journal of Geophysical
- Research: Biogeosciences, 117(G4), n/a-n/a, doi:10.1029/2012JG002128, 2012.
- 696 Reid, J. S., Hyer, E. J., Prins, E. M., Westphal, D. L., Zhang, J., Wang, J., Christopher, S. A., Curtis, C. A., Schmidt,
- 697 C. C., Eleuterio, D. P., Richardson, K. A. and Hoffman, J. P.: Global Monitoring and Forecasting of
- 698 Biomass-Burning Smoke: Description of and Lessons From the Fire Locating and Modeling of Burning
- 699 Emissions (FLAMBE) Program, IEEE Journal of Selected Topics in Applied Earth Observations and Remote
- 700 Sensing, 2(3), 144-162, doi:10.1109/JSTARS.2009.2027443, 2009.
- 701 Roberts, G., Wooster, M. J. and Lagoudakis, E.: Annual and diurnal African biomass burning temporal dynamics,
- 702 Biogeosciences, 6(5), 849-866, 2009.
- 703 Roy, D. P. and Boschetti, L.: Southern Africa Validation of the MODIS, L3JRC, and GlobCarbon Burned-Area
- 704 Products, IEEE Transactions on Geoscience and Remote Sensing, 47(4), 1032-1044,
- 705 doi:10.1109/TGRS.2008.2009000, 2009.
- 706 Roy, D. P., Ju, J., Lewis, P., Schaaf, C., Gao, F., Hansen, M. and Lindquist, E.: Multi-temporal MODIS-Landsat
- 707 data fusion for relative radiometric normalization, gap filling, and prediction of Landsat data, Remote Sensing
- 708 of Environment, 112(6), 3112-3130, doi:10.1016/j.rse.2008.03.009, 2008.
- 709 Seiler, W. and Crutzen, P. J.: Estimates of gross and net fluxes of carbon between the biosphere and the atmosphere
- 710 from biomass burning, Climatic Change, 2(3), 207-247, 1980.
- 711 Setzer, A. W. and Pereira, M. C.: Amazonia Biomass Burnings in 1987 and an Estimate of Their Tropospheric
- 712 Emissions, Ambio, 20(1), 19-22, 1991.
- 713 Shi, Y., Matsunaga, T., Saito, M., Yamaguchi, Y. and Chen, X.: Comparison of global inventories of CO<sub>2</sub> emissions
- 714 from biomass burning during 2002-2011 derived from multiple satellite products, Environmental Pollution,
- 715 206, 479-487, doi:10.1016/j.envpol.2015.08.009, 2015a.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 716 Shi, Y., Matsunaga, T. and Yamaguchi, Y.: High-Resolution Mapping of Biomass Burning Emissions in Three
- 717 Tropical Regions, Environmental Science & Technology, 49(18), 10806-10814, doi:10.1021/acs.est.5b01598,
- 718 2015b.
- 719 Sofiev, M., Vankevich, R., Lotjonen, M., Prank, M., Petukhov, V., Ermakova, T., Koskinen, J. and Kukkonen, J.: An
- 720 operational system for the assimilation of the satellite information on wild-land fires for the needs of air
- 721 quality modelling and forecasting, Atmospheric Chemistry and Physics, 9(18), 6833-6847, doi:
- 722 10.5194/acp-9-6833-2009, 2009.
- 723 Song, Y., Liu, B., Miao, W., Chang, D. and Zhang, Y.: Spatiotemporal variation in nonagricultural open fire
- 724 emissions in China from 2000 to 2007: OPEN FIRE EMISSIONS IN CHINA, Global Biogeochemical Cycles,
- 725 23(2), n/a-n/a, doi: 10.1029/2008GB003344, 2009.
- 726 Streets, D. G., Yarber, K. F., Woo, J.-H. and Carmichael, G. R.: Biomass burning in Asia: Annual and seasonal
- 727 estimates and atmospheric emissions, Global Biogeochemical Cycles, 17(4), n/a-n/a,
- 728 doi:10.1029/2003GB002040, 2003.
- 729 Su, J. Feng., Zhu, B., Kang, H. Q., Wang, H. L. and Wang, T. J.: Applications of Pollutants Released form Crop
- 730 Residues at Open Burning in Yangtze River Delta Region in Air Quality Model, Environment Science, 33,
- 731 1418-1424, 2012 (in Chinese).
- 732 Sun, J., Peng, H., Chen, J., Wang, X., Wei, M., Li, W., Yang, L., Zhang, Q., Wang, W. and Mellouki, A.: An
- 733 estimation of CO<sub>2</sub> emission via agricultural crop residue open field burning in China from 1996 to 2013,
- 734 Journal of Cleaner Production, 112, 2625-2631, doi:10.1016/j.jclepro.2015.09.112, 2016.
- 735 Tao, S.: Study on the Effect of Crop Production on the Air Quality in Wuhan, Journal of Residuals Science and
- 736 Technology, 14(S1), S41-S45, doi:10.12783/issn.1544-8053/14/S1/5, 2017.
- 737 Tang, X. B., Huang, C., Lou, S. R., Qiao, L. P., Wang, H. L., Zhou, M., Chen, M. H., Chen, C. H., Wang, Q., Li, G.
- 738 L., Li, L., Huang, H. Y., and Zhang, G. F.: Emission Factors and PM Chemical Composition Study of Biomass
- 739 Burning in the Yangtze River Delta Region, Environment Science, 35, 1623–1632, 2014 (in Chinese).
- 740 Tang, Z. X., Xu, R, R. and Lan, X. L.: Breeding of a new peanut variety fuhua 3 and the physiological foundation
- of high yield, Chinese agricultural science bulletin, 25(23), 232-237, 2009 (in Chinese).
- 742 Tian, H., Hao, J., Yongqi, L. U. and Zhou, Z.: Evaluation of SO\_2 and NO\_x emissions resulted from biomass fuels
- utilization in China, Acta Scientiae Circumstantiae, 22(2), 204-208, 2002.
- 744 Tian, H., Zhao, D. and Wang, Y.: Emission inventories of atmospheric pollutants discharged from biomass burning
- in China, Acta Scientiae Circumstantiae, 31(2), 349-357, 2011 (in Chinese).

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 746 Tian, X. L., Xia, J., Xia, H. B. and Ni, J.: Forest biomass and its spatial pattern in Guizhou Province, Chinese
- 747 Journal of Applied Ecology, 22(2), 287–297, 2014 (in Chinese).
- van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries,
- 749 R. S., Jin, Y. and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna,
- 750 forest, agricultural, and peat fires (1997-2009), Atmospheric Chemistry and Physics, 10(23), 11707-11735,
- 751 doi:10.5194/acp-10-11707-2010, 2010.
- 752 Wang, K. D. and Deng, L. Y.: Dynamics of forest vegetation carbon stock in Fujian Province based on national
- 753 forest inventories, Journal of Fujian College of Forestry, 34(2), 145–151, 2014 (in Chinese).
- 754 Wang, S. X. and Zhang, C. Y.: Spatial and Temporal Distribution of Air Pollutant Emissions from Open Burning of
- 755 Crop Residues in China, Science paper Online, 3, 329–333, 2008 (in Chinese).
- 756 Wen, X. R., Jiang, L. X., Liu, L., Lin, G. Z., Zheng, Y., Xie, X. J. and She, G. H.: Forest biomass, spatial
- 757 distribution analysis and productivity estimation in Jiangsu Province, 29(1), 36–40, 2014 (in Chinese).
- 758 Wiedinmyer, C., Quayle, B., Geron, C., Belote, A., McKenzie, D., Zhang, X., O'Neill, S. and Wynne, K. K.:
- 759 Estimating emissions from fires in North America for air quality modeling, Atmospheric Environment,
- 760 40(19), 3419-3432, doi:10.1016/j.atmosenv.2006.02.010, 2006.
- 761 Wiedinmyer, C., Akagi, S. K., Yokelson, R. J. and Emmons, L. K.: The Fire INventory from NCAR (FINN) a high
- 762 resolution global model to estimate the emissions from open burning, Geoscientific Model Development
- 763 Discussions, 3(4), 625-641, 2011.
- 764 Wooster, M. J., Roberts, G., Perry, G. L. W. and Kaufman, Y. J.: Retrieval of biomass combustion rates and totals
- 765 from fire radiative power observations: FRP derivation and calibration relationships between biomass
- 766 consumption and fire radiative energy release, Journal of Geophysical Research, 110 (D24), doi:
- 767 10.1029/2005JD006318, 2005.
- 768 Xie, G. H., Han, D. Q., Wang, X. Y. and Lv, R. H.: Harvest index and residue factor of cereal crops in China,
- Journal of China agricultural university, 16(1), 1–8, 2011a (in Chinese).
- 770 Xie, G. H., Han, D. Q., Wang, X. Y. and Lv, R. H.: Harvest index and residue factor of non-cereal crops in China,
- Journal of China agricultural university, 16(1), 1–8, 2011b (in Chinese).
- 772 Yamaji, K., Li, J., Uno, I., Kanaya, Y., Irie, H., Takigawa, M., Komazaki, Y., Pochanart, P., Liu, Y., Tanimoto, H.,
- 773 Ohara, T., Yan, X., Wang, Z. and Akimoto, H.: Impact of open crop residual burning on air quality over
- 774 Central Eastern China during the Mount Tai Experiment 2006 (MTX2006), Atmospheric Chemistry and
- 775 Physics, 10(15), 7353-7368, doi:10.5194/acp-10-7353-2010, 2010.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 776 Xue, X. P., Wang, J. G., Guo, W. Q., Chen, B. L., You, J. and Zhou, Z. G.: Effect of nitrogen applied levels on the
- dynamics of biomass, nitrogen accumulation and nitrogen fertilization recovery rate of cotton after initial
- 778 flowering, Acta Ecologica Sinica, 26(11), 3632-3640, 2006 (in Chinese).
- 779 Yan, X., Ohara, T. and Akimoto, H.: Bottom-up estimate of biomass burning in mainland China, Atmospheric
- 780 Environment, 40(27), 5262-5273, doi:10.1016/j.atmosenv.2006.04.040, 2006.
- 781 Yan, W. L., Liu, D. Y., Sun Y., Wei, J. S., and Pu, M. J.: Analysis of the Sustained Fog and Haze Event Resulting
- 782 from Crop Burning Residue in Jiangsu Province, Climatic and Environmental Research, 19, 237-247, 2014 (in
- 783 Chinese).
- 784 Yevich, R. and Logan, J. A.: An assessment of biofuel use and burning of agricultural waste in the developing
- 785 world, Global Biogeochemical Cycles, 17(4), n/a-n/a, doi:10.1029/2002GB001952, 2003.
- 786 Yin, S., Wang, X., Xiao, Y., Tani, H., Zhong, G. and Sun, Z.: Study on spatial distribution of crop residue burning
- 787 and PM<sub>2.5</sub> change in China, Environmental Pollution, 220, 204–221, doi:10.1016/j.envpol.2016.09.040, 2017.
- 788 Yin, S., Wang, X., Xiao, Y.PM<sub>2.5</sub>, Tani, H., Zhong, G. and Sun, Z.: Study on spatial distribution of crop residue
- purning and change in China, Environmental Pollution, 220, 204-221, doi:10.1016/j.envpol.2016.09.040,
- 790 2017.
- 791 Yu, L. X., Zhang, J. H., Liu, L. Q., Chen, Q. Q., Zhou, Y., Wang, X. G., Xia, S. B. and Bie, S.: Study on high yield
- 792 mechanism of hybrid cotton, 48(9), 2084-2086, 2009 (in Chinese).
- 793 Zeng, J. M., Cui, K. H., Huang, J. L., He, F., Peng, S. B.: Responses of physio-biochemical properties to
- 794 N-fertilizer application and its relationship with nitrogen use efficiency in rice, Acta agronomica sinica, 33(7),
- 795 1168-1176, 2007 (in Chinese).
- 796 Zha, S.: Agricultural Fires and Their Potential Impacts on Regional Air Quality over China, Aerosol & Air Quality
- 797 Research, 13(3), 992-1001, 2013.
- 798 Zhang, H., Hu, D., Chen, J., Ye, X., Wang, S. X., Hao, J. M., Wang, L., Zhang, R. and An, Z.: Particle Size
- 799 Distribution and Polycyclic Aromatic Hydrocarbons Emissions from Agricultural Crop Residue Burning,
- 800 Environmental Science & Technology, 45(13), 5477-5482, doi:10.1021/es1037904, 2011.
- 801 Zhang, H., Wang, S., Hao, J., Wang, X., Wang, S., Chai, F. and Li, M.: Air pollution and control action in Beijing,
- Journal of Cleaner Production, 112, 1519-1527, doi:10.1016/j.jclepro.2015.04.092, 2016.
- 803 Zhang, H.F.: A laboratory study on emission characteristics of gaseous and particulate pollutants emitted from
- agricultural crop residue burning in China. PHD thesis, Fudan University, 2009 (in Chinese).
- 805 Zhang, L., Liu, Y. and Hao, L.: Contributions of open crop straw burning emissions to PM<sub>2.5</sub> concentrations in

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





- 806 China, Environmental Research Letters, 11(1), 014014, doi:10.1088/1748-9326/11/1/014014, 2016.
- 807 Zhang, J., Cui, M., Fan, D., Zhang, D., Lian, H., Yin, Z. and Li, J.: Relationship between haze and acute
- 808 cardiovascular, cerebrovascular, and respiratory diseases in Beijing, Environmental Science and Pollution
- 809 Research, 22(5), 3920-3925, doi:10.1007/s11356-014-3644-7, 2015.
- 810 Zhang, Y., Tang, L., Croteau, P. L., Favez, O., Sun, Y., Canagaratna, M. R., Wang, Z., Couvidat, F., Albinet, A.,
- Zhang, H., Sciare, J., Pr éν α̂, A. S. H., Jayne, J. T. and Worsnop, D. R.: Field characterization of the PM<sub>2.5</sub>
- Aerosol Chemical Speciation Monitor: insights into the composition, sources, and processes of fine particles
- in eastern China, Atmospheric Chemistry and Physics, 17(23), 14501-14517, doi:10.5194/acp-17-14501-2017,
- 814 2017.
- 815 Zhao, P., Chen, F.: Short-term influences of straw and nitrogen cooperation on nitrogen use and soil nitrate content
- 816 in North Henan, 13(4), 19-23, 2008 (in Chinese).
- 817 Zhou, Y., Cheng, S. Y, Lang, J., Chen, D. S., Zhao, B. B., Liu, C., Xu, R. and Li, T.: A comprehensive ammonia
- emission inventory with high-resolution and its evaluation in the Beijing-Tianjin-Hebei (BTH) region, China,
- 819 Atmospheric Environment, 106, 305-317, doi:10.1016/j.atmosenv.2015.01.069, 2015.
- 820 Zhou, Y., Yue, Y., Lan, L. I., Liu, M. and Zhou, T.: Analysis of a Serious Haze Event Resulting from Crop Residue
- Burning in Central Eastern Hubei, Climatic & Environmental Research, 2016.
- 822 Zhou, Y., Xing, X., Lang, J., Chen, D., Cheng, S., Wei, L., Wei, X. and Liu, C.: A comprehensive biomass burning
- 823 emission inventory with high spatial and temporal resolution in China, Atmospheric Chemistry and Physics,
- 824 17(4), 2839-2864, doi:10.5194/acp-17-2839-2017, 2017.
- 825 Zong, Z., Wang, X., Tian, C., Chen, Y., Qu, L., Ji, L., Zhi, G., Li, J. and Zhang, G.: Source apportionment of PM<sub>2.5</sub>
- at a regional background site in North China using PMF linked with radiocarbon analysis: insight into the
- 827 contribution of biomass burning, Atmospheric Chemistry and Physics, 16(17), 11249-11265,
- 828 doi:10.5194/acp-16-11249-2016, 2016.
- 829 Zou, J., Lu, J. W., Liao, Z. W., Gong, X. M., Wang, H., Zhou, Y. G. and Zhou, H.: Study on response of rapeseed to
- 830 boron application and critical level of soil available B in Hubei province, Scientia Agricultura Sinica, 41(3),
- 831 752-759, 2008 (in Chinese).
- 832 Zuo, Z. G.: The cause and prevention of forest fire in forest area of Southern China, Land Greening, 2004 (in
- 833 Chinese).
- 834 Zhu, L. J., Wang, G. Y., Zhang, Y. L.: Spatial and temporal distribution of crop straw resources in Yangtze River
- Delta area, Guizhou Agricultural Sciences, 45(4), 138-142, 2017 (in Chinese).

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





Table 1. Forest, shrubland and grassland biomass fuel loading (kt km<sup>-2</sup>) in each province.

Province	Forest (2003-2008) <sup>a</sup>	Forest (2009-2015)*	Shrubland <sup>b</sup>	Grassland <sup>c</sup>
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: <sup>a</sup> Fang et al. (1996); <sup>b</sup> Pu et al. (2004); <sup>c</sup> Hu et al. (2006); <sup>\*</sup> This study.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





**Table 2.** Parameters of biomass-stem volume regression functions of dominant tree species in forests (B=aV+b)

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

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

B = aV + b: B indicates the total biomass of different tree species (t); V indicates the forest stock volume (m<sup>-3</sup>).

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-282 Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018 © Author(s) 2018. CC BY 4.0 License.





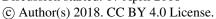
Table 3. The detailed crop specific residue to production ratio data for each province

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

References: <sup>a</sup> Zhu et al. (2017); <sup>b</sup> Chen et al. (2008); <sup>c</sup> Xie et al. (2011a); <sup>d</sup> Xie et al (2011b); <sup>e</sup> Zeng et al (2007); <sup>f</sup> Ao et al. (2007); <sup>g</sup> Lei et al. (2009); <sup>h</sup> Zhao et al. (2008); <sup>i</sup> Xue et al. (2006); <sup>j</sup> Yu et al (2009); <sup>k</sup> Zou et al (2008); <sup>1</sup> Liu et al. (2010); <sup>m</sup> Tang et al. (2009); <sup>n</sup> Li et al. (2008).

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018







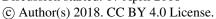
**Table 4.** The detailed crops straw burned ratio data from survey results.

Region	Crops straw burning percentage
Anhui	$0.10^{a}$
Fujian	0.188 <sup>b</sup>
Henan	$0.208^{\circ}$
Hubei	0.207°
Hunan	$0.278^{\circ}$
Jiangsu	$0.10^{a}$
Jiangxi	$0.18^{c}$
Shandong	$0.178^{\circ}$
Shanghai	$0.148^{d}$
Zhejiang	0.319°

References: <sup>a</sup> Tian (2011); <sup>b</sup> Huang (2014); <sup>c</sup> Peng et al; (2016). <sup>d</sup> Zhou et al (2017).

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018







**Table 5.** The emission factors of open biomass burning emissions for various pollutants (g kg<sup>-1</sup> dry matter)

Vegetation	OC	EC	CO	CH <sub>4</sub>	NOx	NMVOCs	SO <sub>2</sub>	NH <sub>3</sub>	$CO_2$	PM <sub>2.5</sub>
Corn	1.457*	0.14*	70. 2 <sup>a</sup>	4. 4 <sup>b</sup>	3.36 <sup>a</sup>	10 <sup>c</sup>	0.45 <sup>h</sup>	0. 68 <sup>g</sup>	1261 <sup>f</sup>	5 <sup>c</sup>
Rice	1. 96 <sup>a</sup>	0. 52 <sup>c</sup>	52. 32 <sup>c</sup>	3. 9 <sup>b</sup>	1.42 <sup>d</sup>	6. 05 <sup>f</sup>	0. 147 <sup>a</sup>	0. 53 <sup>g</sup>	791 <sup>f</sup>	3.03 <sup>d</sup>
Wheat	2.7 <sup>b</sup>	0. 49a	61. 90 <sup>c</sup>	3. 4 <sup>b</sup>	1.19 <sup>d</sup>	7. 5 <sup>c</sup>	0. 147 <sup>c</sup>	0. 37 <sup>b</sup>	1557 <sup>f</sup>	7.6a
Cotton	3.06 <sup>c</sup>	0. 57 <sup>f</sup>	70.29 <sup>c</sup>	4.4 <sup>b</sup>	2. 98 <sup>c</sup>	10 <sup>c</sup>	0.23 <sup>c</sup>	0. 68 <sup>b</sup>	1445 <sup>h</sup>	11.7 <sup>c</sup>
Rapeseed	1.08 <sup>d</sup>	0. 23 <sup>d</sup>	34.3 <sup>d</sup>	3. 9 <sup>b</sup>	1. 12 <sup>d</sup>	8. 64 <sup>c</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	5.76 <sup>c</sup>
Soya bean	1.05 <sup>d</sup>	0. 13 <sup>d</sup>	32.3 <sup>d</sup>	3. 9 <sup>b</sup>	1. 08 <sup>d</sup>	8. 64 <sup>c</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	$3.32^{d}$
Sugar cane	2. 03 <sup>c</sup>	0. 41 <sup>c</sup>	$40.08^{f}$	3. 9 <sup>b</sup>	2. 03 <sup>c</sup>	11. 02 <sup>f</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	$4.12^{f}$
Peanut	2. 03 <sup>c</sup>	0. 41 <sup>c</sup>	55. 13 <sup>c</sup>	3. 9 <sup>b</sup>	2. 11 <sup>c</sup>	8. 64 <sup>c</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	5.76 <sup>c</sup>
Potato	2. 03 <sup>c</sup>	0. 41 <sup>c</sup>	55. 13 <sup>c</sup>	3. 9 <sup>b</sup>	2. 11 <sup>c</sup>	8. 64 <sup>c</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	5.76 <sup>c</sup>
Tobacco	2. 03 <sup>c</sup>	0. 41 <sup>c</sup>	55. 13 <sup>c</sup>	3. 9 <sup>b</sup>	2. 11 <sup>c</sup>	8. 64 <sup>c</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	5.76 <sup>c</sup>
Sesame	2. 03 <sup>c</sup>	0. 41 <sup>c</sup>	55. 13 <sup>c</sup>	3.9 b	2. 11 <sup>c</sup>	8. 64 <sup>c</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	5.76 <sup>c</sup>
Sugar beet	2. 03 <sup>c</sup>	0. 41 <sup>c</sup>	55. 13 <sup>c</sup>	3. 9 <sup>b</sup>	2. 11 <sup>c</sup>	8. 64 <sup>c</sup>	0. 25 <sup>c</sup>	0. 53 <sup>g</sup>	1445 <sup>h</sup>	5.76 <sup>c</sup>
Coniferous forest	2.65 <sup>j</sup>	0.11 <sup>j</sup>	118 <sup>e</sup>	6 <sup>e</sup>	2.4 <sup>e</sup>	28 <sup>e</sup>	$1^{i}$	3.5 <sup>e</sup>	1514 <sup>e</sup>	9.7 <sup>e</sup>
Broadleaf forest	1.181*	0.31 <sup>e</sup>	102e	5 <sup>e</sup>	1.3e	14 <sup>e</sup>	$1^{i}$	1.5 <sup>e</sup>	1630e	13 <sup>e</sup>
Mixed forest	9.2e	0.6e	102e	5 <sup>e</sup>	1.3e	14 <sup>e</sup>	$1^{i}$	1.5 <sup>e</sup>	1630e	9.7 <sup>e</sup>
Grassland	2.6e	0.4 <sup>e</sup>	59e	1.5e	2.8e	9.3 <sup>e</sup>	0.5e	0.5 <sup>e</sup>	1692 <sup>e</sup>	5.4e
Shrubland	6.6 <sup>e</sup>	0.5 <sup>e</sup>	68 <sup>e</sup>	2.6 <sup>e</sup>	3.9e	4.8 <sup>e</sup>	0.7 <sup>e</sup>	1.2 <sup>e</sup>	1716 <sup>e</sup>	9.3 <sup>e</sup>

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. (2009);  $^g$ EPD (2014);  $^h$ Wang et al. (2008);  $^i$ Andreae and Rosenfeld (2008);  $^*$ This study.

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018 © Author(s) 2018. CC BY 4.0 License.





**Table 6.** Cumulative emissions of major pollutants from open biomass burning in Central and Eastern China during 2003-2015 (Gg yr $^{-1}$ )

Province	OC	EC	CH <sub>4</sub>	NOx	NMVOCs	$SO_2$	NH <sub>3</sub>	СО	CO <sub>2</sub>	PM <sub>2.5</sub>
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

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-282 Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018

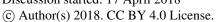






Table 7. Correction analysis of the variation tendency from 2003 to 2015 between PM<sub>2.5</sub> emission from crops straw burning in each province and the rural population, agricultural output and per capita incomes of rural residents.

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

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-282 Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 17 April 2018





**Table 8.** Comparison of the emissions with previous studies in different years (Gg yr<sup>-1</sup>)

Reference	Year	OC	EC	CH <sub>4</sub>	NOx	NMVOCs	$SO_2$	NH <sub>3</sub>	СО	$CO_2$	PM <sub>2.5</sub>
Wang et al., 2008	2006	252	25.8	197	189	459	31.8	44.1	3841	81225	1138
This study		215.3	21.13	3267	220.7	131.9	451.1	14.33	31.46	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	3817	252.1	151.2	531.5	17.86	38.67	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	3688	245.7	148.5	507.8	16.71	35.92	75785	329.46

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





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

Year	OC	EC	CO	CH <sub>4</sub>	NOx	NMVOC	SO <sub>2</sub>	NH <sub>3</sub>	CO <sub>2</sub>	PM <sub>2.5</sub>
2003	(-31%,	(-46%,	(-20%,	(-20%,	(-23%,	(-52%,	(-52%,	(-33%,	(-3%,	(-44%,
	31%)	46%)	20%)	20%)	23%)	53%	51%)	33%)	3%)	44%)
2004	(-29%,	(-47%,	(-21%,	(-22%,	(-24%,	(-45%,	(-56%,	(-34%,	(-3%,	(-47%,
2004	29%)	48%)	21%)	22%)	24%)	45%)	58%)	34%)	3%)	47%)
2005	(-31%,	(-42%,	(-16%,	(-16%,	(-19%,	(-41%,	(-44%,	(-32%,	(2%,	(-35%,
2005	31%)	44%)	16%)	17%)	19%)	40%)	44%)	33%)	3%)	34%)
2006	(-32%,	(-44%,	(-13%,	(-14%,	(-16%,	(-43%,	(-34%,	(-34%,	(-3%,	(-25%,
2000	33%)	44%)	13%)	14%)	17%)	43%)	35%)	34%)	3%)	25%)
2007	(-30%,	(-46%,	(-18%,	(-19%,	(-22%,	(-50%,	(-50%,	(-33%,	(-3%,	(-42%,
2007	30%)	46%)	19%)	19%)	22%)	51%)	50%)	34%)	3%)	42%)
2008	(-26%,	(-52%,	(-25%,	(-28%,	(-29%,	(-69%,	(-62%,	(-38%,	(-3%,	(-55 %,
2008	26%)	53%)	25%)	28%)	29%)	69%)	61%)	39%)	3%)	56%)
2009	(-28%,	(-48%,	(-21%,	(-21%,	(-24%,	(-59%,	(-54%,	(-34%,	(-3%,	(-47%,
2009	28%)	48%)	21%)	22%)	24%)	59%)	54%)	35%)	3%)	47%)
2010	(-31%,	(-44%,	(-16%,	(-17%,	(-19%,	(-45%,	(-42%,	(-33%,	(-3%,	(-34%,
2010	31%)	44%)	17%)	17%)	19%)	46%)	42%)	34%)	3%)	34%)
2011	(-29%,	(-46%,	(-18%,	(-19%,	(-21%,	(-52%,	(-47%,	(-34%,	(-3%,	(-40%,
2011	29%)	46%)	18%)	19%)	21%)	53%)	47%)	35%)	3%)	40%)
2012	(-32%,	(-44%,	(-14%,	(-14%,	(-17%,	(-35%,	(-35%,	(-34%,	(-3%,	(-27%,
2012	33%)	44%)	14%)	14%)	17%)	35%)	35%)	35%)	3%)	26%)
2013	(-30%,	(-44%,	(-16%,	(-17%,	(-20%,	(-51%,	(-42%,	(-33%,	(-3%,	(-36%,
2013	30%)	44%)	16%)	17%)	20%)	51%)	43%)	34%)	3%)	36%)
2014	(-32%,	(-45%,	(-15%,	(-16%,	(-19%,	(-43%,	(-42%,	(-35%,	(-3%,	(-33%,
2014	32%)	46%)	15%)	16%)	18%)	43%)	42%)	35%)	3%)	33%)
2015	(-31%,	(-44%,	(-14%,	(-14%,	(-17%,	(-41%,	(-34%,	(-34%,	(-3%,	(-26%,
2015	31%)	44%)	146%)	13%)	17%)	41%)	34%)	35%)	3%)	26%)





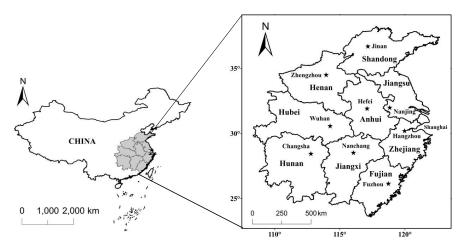
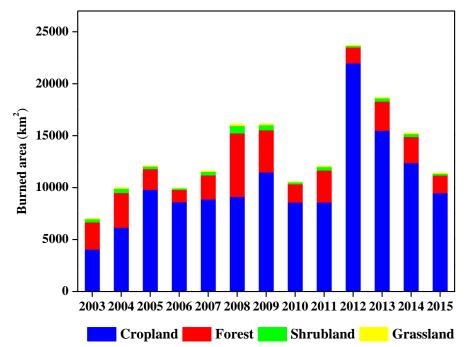


Figure 1. Location of Central and Eastern China and the key megacities.



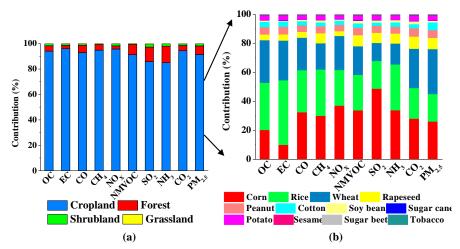




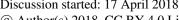
**Figure 2.** The integrated open biomass burned area in Central and Eastern China from 2003 to 2015.







**Figure 3.** The mean contributions of different types of biomass (a) and the contribution of different types of crops to the whole cropland accumulative emissions of pollutants in Central and Eastern China from 2003-2015.







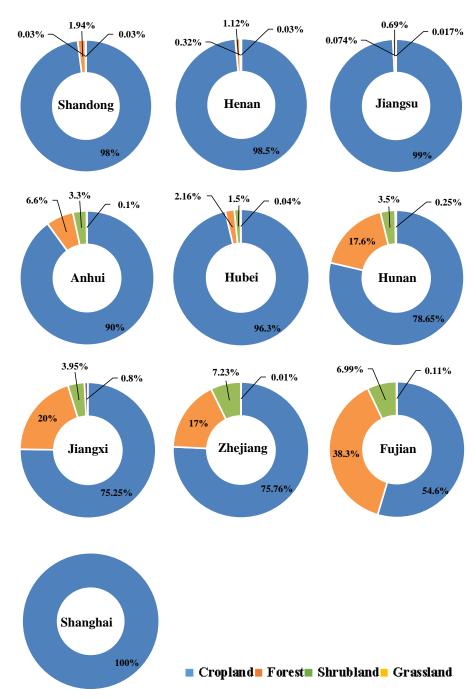


Figure 4. The averaged contributions of different biomass burning types to PM<sub>2.5</sub> emission in each province.





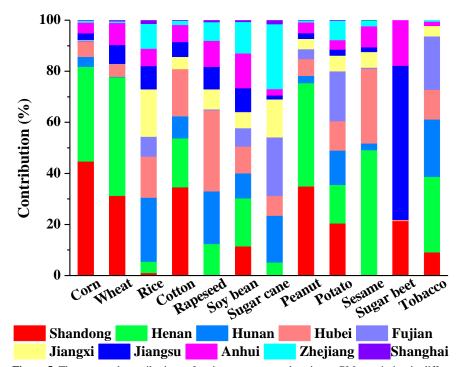


Figure 5. The averaged contributions of various crops straw burning to  $PM_{2.5}$  emission in different provinces.

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





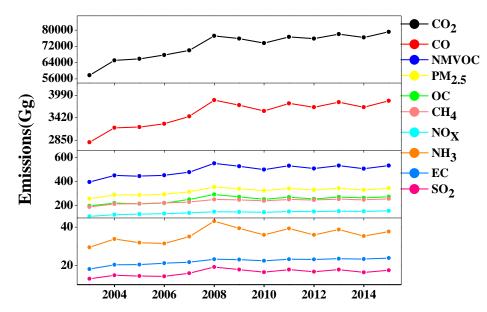
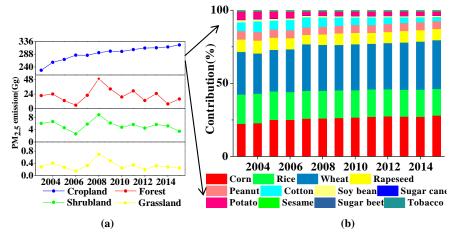


Figure 6. Historical emissions of opening biomass burning from 2003 to 2015.



**Figure 7.** The multi-year  $PM_{2.5}$  emissions from (a): different opening biomass burning sources; (b): various crop types from 2003 to 2015.

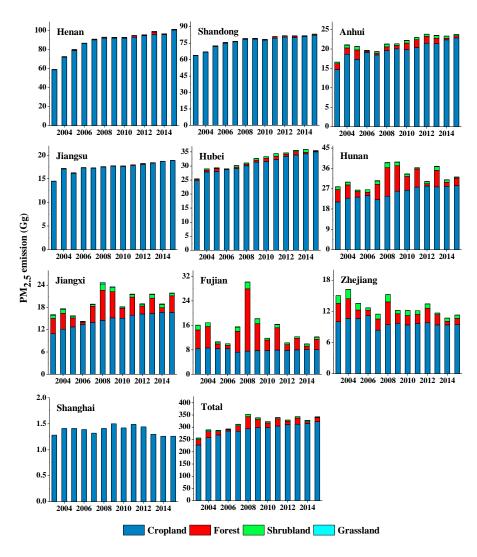
43

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.



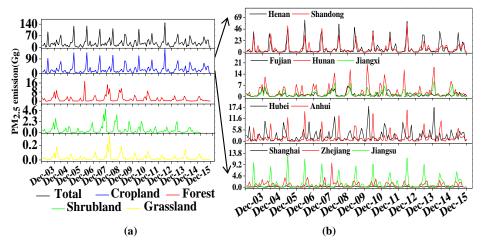




**Figure 8.** The multi-year  $PM_{2.5}$  emission for the four types of biomass burning in different provinces from 2003 to 2015.







**Figure 9.** The monthly  $PM_{2.5}$  emission of different open biomass burning from 2003 to 2015 for the whole Central and Eastern China (a) and for each province (b).

Discussion started: 17 April 2018

© Author(s) 2018. CC BY 4.0 License.





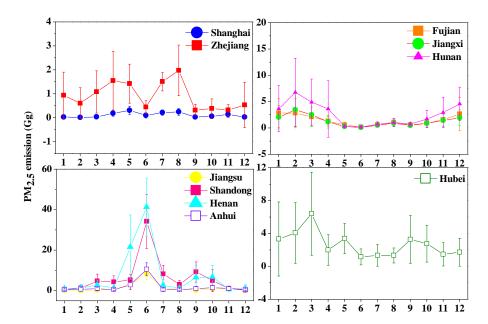
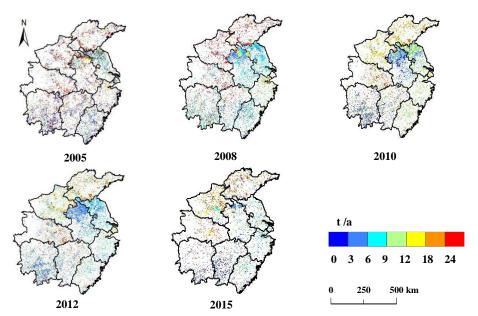


Figure 10. The Monthly  $PM_{2.5}$  emission from open biomass burning in each province.



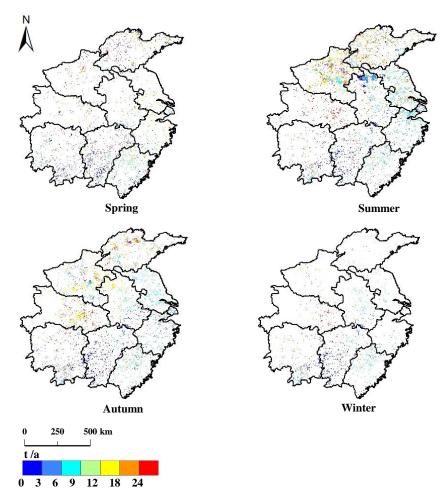




**Figure 11.** Annual spatial distribution (1 km $\times$ 1 km) of PM<sub>2.5</sub> emissions from opening biomass burning in Central and Eastern China.

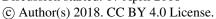






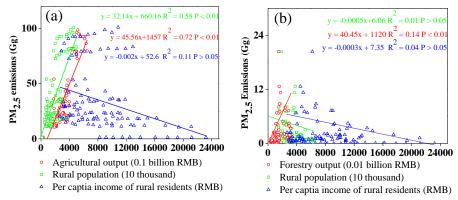
**Figure 12.** Seasonal emission distribution (1 km $\times$ 1 km) of PM<sub>2.5</sub> in 2015 from opening biomass burning in Central and Eastern China.

Discussion started: 17 April 2018









**Figure 13.** Correction analysis between  $PM_{2.5}$  emission from (a) crop residue burning and (b) forestry fire burning in different provinces and agricultural output and forestry output, rural population and per captia incomes of rural residents from 2003-2015.