Authors' Responses to Reviewer Comments

Manuscript: A comprehensive biomass burning emission inventory with high spatial and temporal resolution in China (Ref. No.: acp-2016-560)

We are very grateful to the respectful referee for your careful and insightful review. Your comments and suggestions have contributed greatly to improving our paper. Our point-by-point responses to your comments are listed as follows.

Anonymous Referee #1

General comments: In general, this study quantified a comprehensive biomass burning emissions including indoor and outdoor biomass burning emissions and fits the requirement of East Asia emissions assessment. However, it is difficult to find anything new to the scientific world. Since there is nothing new on used methods or data. And some methods and data usually reduce some errors and uncertainties.

Response:

We thank you for your comments, which were very helpful for revising and improving our paper. Generally, two approaches are employed for developing a biomass burning emission inventory: a "top-down" approach and a "bottom-up" approach. Regarding a consideration of new methods, we point out the overwhelming importance of employing refined and updated data to develop a more accurate emission inventory with a much higher temporal-spatial resolution. In fact, highly detailed emission information is extremely important for investigating the causes of air pollution (e.g., air quality modeling) and developing a targeted strategy for the control of pollution accompanying biomass burning, particularly for conditions prevalent in recent years owing to the altered pattern of energy consumption in rural areas and the increasing pollution problems associated with dramatic urbanization caused by the economic development in China. Currently, as far as we are aware, few studies have developed a comprehensive biomass burning emission inventory in China, particularly after 2007, because of a lack of detailed statistical data regarding firewood consumption. Furthermore, the source-specific emission factors (EFs) used in emission estimation need to be updated based on a systematic combination of localized measurements conducted in China. In addition to EFs, the activity data is also a key factor for improving an emission inventory. Moreover, several key sources of information related to biomass emission estimation must be updated, such as the proportion of crop straw domestic combustion and in-field burning, and the uneven temporal distribution coefficient, which reflects recent conditions in different regions of China. In fact, the current biomass burning emission inventory for China is generally at a province-level resolution because detailed activity data is not publicly available. It is obvious that the resolution of activity data determines the preliminary resolution of an emission inventory. An emission inventory with a coarse preliminary resolution could result in greater uncertainty in grid emissions generated according to source-based gridded spatial surrogates (e.g., population) using GIS technology.

The main contributions of our work are summarized as follows.

First, a comprehensive biomass burning emission inventory, including crop straw domestic combustion and in-field burning, firewood and livestock excrement combustion, and forest and grassland fires for mainland China was developed based on detailed data (county-level data and satellite data) and updated source-specific EFs for the first time.

Second, a range of important information representing the recent status for emissions estimation in China were obtained from field investigation, a systematic combination of the latest research, and regression analysis (e.g., province-specific straw domestic combustion/in-field burning ratio, detailed firewood combustion quantities, and non-uniform temporal distribution coefficient).

Third, the high-resolution temporal (monthly and daily) and spatial (1 km × 1 km) biomass burning emission inventory presented in this study includes major precursors of complex pollution systems, greenhouse gases, and heavy metals released from biomass burning such as SO_2, NO_x, PM_{10}, $PM_{2.5}$, VOCs, NH_3, CO, EC, OC, CO_2, CH_4, and Hg with further breakdowns of $PM_{2.5}$ particles and VOCs.

In addition, we have carefully taken the reviewer's remaining comments related to methods and data into consideration during the revision of our paper (e.g., emission estimation of forest and grassland fires). Please see the following point-by-point responses.

"Specific comments"

The biomass burning includes firewood burning and in-field burning. There is another large contributor of human waste burning that should not be overlooked, especially in rural area of the developing countries. Since this is a comprehensive inventory, I suggested the authors can add this part. Wiedinmyer, C.; Yokelson, R. J.; Gullett, B. K.Global emissions of trace gases, particulate matter, and hazardous air pollutants from open burning of domestic waste Environ. Sci. Technol. 2014, 48(16) 9523–9530, DOI: 10.1021/es502250z Shi, Y., Matsunaga, T., and Yamaguchi, Y., High-resolution mapping of biomass burning emissions in three tropical regions, Environmental Science and Technology, Environ. Sci. Technol., 2015, 49 (18), pp10806–10814. DOI: 10.1021/acs.est.5b01598

Response:

We thank you very much for your suggestion. In view of your proposal, we have conducted an extensive literature review.

First, in this study, we developed a detailed emission inventory of biomass burning, including crop straw domestic combustion and in-field burning, firewood and livestock excrement combustion, and forest and grassland fires. These are important sources of biomass burning that are considered in the literature, including studies focused on domestic combustion and open burning in China (He et al., 2011; Chen et al., 2013; Zhang et al., 2013) and other regions (Shon et al., 2015; van der Werf et al., 2010; Bhardwaj et al., 2016) in recent years.

Second, with the increasing economic development and income of rural residents, tremendous changes have taken place in the pattern of rural resident consumption. Industrial products are increasingly used in the lives of rural residents. Therefore, the composition of rural human waste tends to be largely a product of urbanization Human waste in rural areas of China is mainly inorganic garbage, such as waste plastics, waste paper, waste glass, and hazardous waste, and some organic garbage (e.g., crop residue waste and kitchen waste) (Ma et al., 2002; Chai et al., 2012; Yan et al., 2014). More than 80% of the garbage in rural areas is discarded carelessly without any further processing (Guan and Qiu, 2008; Wang et al., 2011; Yao et al., 2009).

Third, among the sources of rural human waste, the primary biomass wastes that may be burnt in the rural areas of China are crop residue waste and livestock excrement (Tian et al., 2011; Zhou et al., 2013; Zhang et al., 2013). These wastes have been considered in our study.

Fourth, we have added a review of studies focused on the emissions of human waste burning, including the two articles recommend by the reviewer, such as Park et al. (2013), Wiedinmyer et al. (2014), Shi et al. (2015), and Maasikmets et al. (2016). Few EFs of human waste burning are categorized into specific waste types (particularly with regard to biomass waste). In addition, the specific waste production in rural areas is difficult to obtain currently in China. If we attempt to estimate the emissions of human waste burning based on non-specific EFs and waste production in our biomass burning study, the results of the biomass burning emission inventory will be overestimated due to introduced emissions that are independent of biomass burning.

Owing to the reasons discussed above, we did not estimate the emissions of human waste burning in the current study. Further studies on the specific characteristics of human waste burning emission must be conducted, which would then allow the development of an elaborate emission inventory of waste burning related to biomass based on detailed EFs and relevant activity data investigations.

2.2.3 Biomass burning of forest/grassland fires. The estimation of burned biomass in this very simple method have lots of problems. AR is the damaged area, in fact, it is the burned area, they are far different. Burned area data were usually derived from satellite data for such a large area of China. It is basically wrong that the authors used the statistics data to allocate them according to the fire counts. Since fire counts does not linearly correspond with the burned area. Please refer to MCD64A1 burned area product with 500 m resolution, which has been validated in many ecosystems. Fire consumes great amount of biomass when burning happens. And this biomass usually cost several years to recover to its previous condition. The authors failed in considering the reduction of biomass of this month due to fire as the beginning of the next month. Therefore, I suggested the authors should consider the reduction of biomass when it is used as the base for the next month. Besides, the biomass used in this study within each province are even. The biomass density was constrained by precipitation, air temperature and vapor pressure controlled gross primary production, respiration, etc. The used constant data cannot reflect the heterogeneity of the biomass. Combustion factor is strongly controlled by fuel types and moisture conditions and vary widely from pixel to pixel. The authors set the combustion factors for each fuel type as constant, which cannot depict the differences between moisture and dry fuels types. Since dry fuels can burn mostly while wet fuels burn less completely. I suggested the authors should consider the moisture condition of the fuel types and revised them into spatial and temporal variable parameters, which can really reflect the condition of each pixel. Since there are many available satellite products on burned area, ecosystem productivity model estimated biomass density and moisture condition, we really do not suggest the authors used the county-level data and allocate them into each pixel. The estimation of biomass burning emissions by using the bottom-up method should use the pixel-based high-resolution datasets to describe its process. van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos.Chem. Phys., 10, 11707-11735, doi:10.5194/acp-10-11707-2010, 2010.

Response:

We thank you for pointing out these problems with the methods and data employed in the emission estimation of forest and grassland fires. As the reviewer mentioned, the damaged area is different from the burned area. Our description in the original manuscript was unfortunately confusing, and we have revised the description to clarify our meaning. Furthermore, as the reviewer pointed out, the burned area should be obtained from satellite data rather than statistical data and allocation based on the fire counts. According to the reviewer's suggestion, we have updated our methods and data employed in this study, and have re-calculated the pixel-based emission of forest and grassland fires using the bottom-up method. The burned area data is derived from the MCD64A1 burned area satellite product with a 500 m resolution. As for the biomass density, we agree with the reviewer that the value within each province should not be constant because this cannot reflect the heterogeneity of the biomass. The vegetation type of each pixel where forest and grass fires have occurred can be determined according to the land cover data. Therefore, the biomass density was determined according to the vegetation type of different provinces based on localized studies in China. As for the biomass density reduction over continuous months due to fires, we compared the distribution of burned areas due to forest and grassland fires in different months for 2012 in China, and found that little overlap between burning areas is observed among various months. This is mainly due to the fact that forest and grassland fires are accidental events, which can occur only with the confluence of three elements, i.e., forest and grassland fuel, fire, and meteorological conditions (Wei et al., 2014). Therefore, we did not consider the reduction of biomass density in different months. A similar consideration of biomass density can be found in recent studies (Song et al., 2009; Zhang et al., 2013; Qiu et al., 2016). As for the combustion factor (CF), we also noticed that CFs for each fuel type should not be constant. Considering the information we could obtain, we used specific CFs according to the vegetation type in each pixel based on literature review (Michel et al., 2005; Kasischke et al., 2000; Hurst et al., 1994). He et al. (2011), Zhang et al. (2013), and Chen et al. (2013) used a similar approach to consider CFs. The methodology employed for the estimation of the biomass burning emission owing to forest and grassland fires in Sect. 2.2.3 of the manuscript was modified accordingly, as follows.

"2.2.3 Estimation of biomass burning emission of forest/grassland fires

The burning mass of forest/grassland can be calculated from the annual mass of forest/grassland burned (kg/yr) as follows:

$$A = \sum_{j=1}^{10} BAx,j \times FLx,j \times CFj, \quad (3)$$

where subscripts *j*, and *x* represent the land cover type, and location, respectively, $BA_{x,j}$ is the burned area (m^2) at *x* where belongs to *j*, $FL_{x,j}$ is the biomass fuel loading (the aboveground biomass density in this study; kg/m^2) at *x* where belongs to *j*, and CF_j is the combustion factor (the fraction of burned aboveground biomass) at *j*.

Burned area data for 2012 were derived from the moderate-resolution imaging spectroradiometer (MODIS) direct broadcast burned area product (MCD64A1; http://modis-fire.umd.edu). This product employs an automated algorithm for mapping MODIS post-fire burned areas, and deriving the approximate burn date within each burn cell combined with surface reflectance, land cover products, and daily active fires. The MCD64A1 product has a primary spatial resolution of 500 m and a temporal resolution of 1 month. The extent of burning over a Julian day and its temporal uncertainty are specified for each burn cell. The burned areas within an approximate Julian day can be extracted from the original 500 m resolution map.

Earlier research on the estimation of *FL* values for forest and grassland typically employed an averaged value of aboveground biomass density. However, these values do not well reflect the spatial variations of *FL* for each vegetation type. In this study, numerous local *FL* were collected for each province and vegetation type. The type of vegetation burned in each pixel was determined by the 1 km resolution MODIS Land Cover product produced by Ran et al. (2010). We considered 10 vegetation types as forest and grassland (i.e., evergreen needleleaf forest, evergreen broadleaf forest, deciduous needleleaf forest, deciduous broadleaf forest, mixed forest, closed shrublands, open shrublands, woody savannas, savannas, and grassland). The values of *FL* employed in this study are listed in Table 4. As for *CF*, it has usually been set as a constant in previous literature. In our paper, *CF* values were collected for each vegetation type, and the *CF* in each pixel was determined by the MODIS Land Cover product and the *CF* of typical vegetation. The *CF* of forest, closed shrublands, open shrublands, woody savannas, and grassland were set as 0.25, 0.5, 0.85, 0.4, and 0.95, respectively (Michel et al., 2005; Kasischke et al., 2000; Hurst et al., 1994)."

The corresponding figures and tables have been revised.

As for 2012, this study estimated 665.989Tg CO2 list in Table 7 and there is almost no forest and grassland fires based on Figure 2 CO2 chart and Figure 8 CO2 chart. But actually, by using the ecosystem production model integrated with fire emission process, Global Fire Emissions Database v4 (GFED4) estimated outdoor biomass burning emissions (forest, savanna and agriculture) with 54 Tg CO2 in 2012. Authors should explain this large differences due to their used methods and datasets.

Response:

We thank you very much for your comment. According to the reviewer's suggestion, we have re-estimated forest and grassland fire emission based on the bottom-up approach and high-resolution satellite data. The total annual forest and grassland fire emission of CO_2 was determined in this way to be 10.9 Tg, where it was 1.68 Tg in the original manuscript. The total biomass burning emission of CO_2 is therefore 675 Tg. Figures 2, 8, and other relevant figures have been revised accordingly. As for the large differences between the CO_2 emissions reported in the Global Fire Emissions Database v4 (GFED4) and those reported in the present study, these could be attributed to the following reasons.

First, the biomass burning emission inventory in this study included the crop straw domestic combustion, an important biomass burning source in China, with a contribution accounting for 68% of the total CO_2 emission. This source was not included in the GFED4.

Second, the emissions of crop straw open burning were estimated in the present study based on the specific EFs and the amount burned for each type of crop straw. The emission in GFED4 was estimated according to the burned area based on a constant EF. The agricultural burned area was derived from the MODIS MCD64A1 product (~500 m resolution). Despite the efforts made to improve the direct broadcast mapping algorithm employed in the MCD64A1 product, the product has a minimum detectable burn area size, which is greater than the size of many agriculture waste burn sites. Therefore, numerous small and scattered agricultural fires would not be detected

(McCarty et al., 2007; Giglio et al., 2013; Shi et al., 2015). This is particularly the case in China, where the open burning of crop residue tends to be conducted by individual families (Liu et al., 2015), resulting in agricultural burning that often occurs over small areas, which is then undetected. The results in the present study were compared with the results of other research. The CO_2 emission of agricultural crop residue open burning in China in 2012 was estimated at 184 Tg by Sun et al. (2016), which is similar to the 207 Tg value obtained in our study. In addition, the total CO_2 emission by biomass burning published in most literature (Cao et al., 2005; Yan et al., 2006; Lu et al., 2011; Tian et al., 2011) is similar to the value obtained in our work, with differences ranging from 0.7% to 40.0%. This comparison indicates that the CO_2 emission estimated in our paper is relatively credible.

Figure 7:

In North China Plain, there are many polygons in blue with small amounts of $PM_{2.5}$, which were far lower than their surrounding areas, the sudden reduction of these polygon values may be attributable to the used county-level data, we suggested the authors changed this dataset since it is unreasonable of these polygons with small amount.

Response:

We thank you very much for your comment. Figure 7 presents a 1 km × 1 km grid reflecting biomass burning emission, including crop straw domestic combustion and in-field burning, firewood and livestock excrement combustion, and forest and grassland fires. Actually, the North China Plain includes several urban areas surrounded by suburban and rural areas. The main fuel used in these urban areas is commodity energy (e.g., coal, natural gas, and electricity) rather than biomass fuel. Therefore, these urban areas produce little biomass burning emission. However, previous studies could not account for these actual conditions because the gridded emission was allocated from an emission inventory with coarse preliminary resolution (e.g., a provincial or prefectural level resolution prior to spatial allocation) based on gridded surrogates (e.g., rural population). In the present study, the grid emission was allocated from an emission inventory with improved preliminary resolution (i.e., county-level resolution), which could reflect the low use of biomass fuel (e.g., crop straw, firewood, and livestock excrement) within several urban regions of the North China Plain. Consequently, the use of the relatively high-resolution emission inventory allocation could better represent the actual conditions.

We have emphasized the description in Sect. 3.3 (Lines 1-15 on Page 16) of the manuscript:

"The scarce population and crop yield in part of southwest and northwest areas, and lower agricultural activity in downtown areas result in lower emissions. Specially, some urban areas in the north China Plain are surrounded by suburban and rural areas, the main fuel used in these urban areas is commodity energy. Besides, there is no agricultural activity in the field. Therefore, these areas produce little biomass burning emission. However, error will be brought in grid emissions if they are allocated from the emission inventory at coarse preliminary resolution (e.g., provincial or prefectural resolution before spatial allocation) based on the gridded surrogates (e.g., rural population). Consequently, gridded emissions, which were obtained through spatial allocation from emission inventory at county resolution, could better represent the actual situation."

Figure 10: This study estimates SO2, NOx, which are comparable to Lu et al., (2011). What is the reason for the underestimation of PM10, VOC, NH3, CH4 and overestimation of EC and OC relative to Lu et al.,(2011). Why these emissions agreed well in NOx, but large differences on other gases?

Response:

We thank you very much for your question. The differences in the cited pollutant emissions between this study and that of Lu et al. (2011) are most likely attributable to the selection of EFs. The higher estimations of EC and OC relative to those obtained by Lu et al. (2011) are mainly due to our use of higher EC and OC EFs for crop straw and firewood domestic combustion. The EC and OC EFs employed in the present study were selected from the work of Li et al. (2007), which were measured in representative rural areas across China to determine the characteristics of household biofuel combustion emission. The EFs employed in Lu et al. (2011) were constant values for different crop straw types, and were derived from measurements conducted outside of China (Reddy and Chandra, 2002). The lower values of CH_4 and NH_3 emissions relative to those obtained by Lu et al. (2011) is mainly due to the lower EFs employed in the present study for crop domestic burning. The crop domestic EFs for CH_4 and NH_3 employed in Lu et al. (2011) were constant values for different crop straw types. The EFs used in the present study were specific for each type of crop straw, which were updated according to published reports of localized measurements conducted in China. The lower estimations of VOC and PM_{10} relative to those obtained by Lu et al. (2011) are the result of the employment of lower EFs for in-field crop residue burning and firewood combustion. The VOC and PM_{10} EFs employed in Lu et al. (2011) did not distinguish between different crop straw types, and were derived from measurements conducted outside of China (Street et al., 2003; Reddy and Chandra, 2002). The specific EFs for various crop straws employed in our study were derived from the Chinese guide for compiling atmospheric pollutant emission inventories for biomass burning published in 2014.

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Anonymous Referee #2

General comments:

Review of "A comprehensive biomass burning emission inventory with high spatial and temporal resolution in China" This study addresses what the authors believe are key weaknesses of current biomass burning emission inventory for China: 1. Missing sources (in particular firewood), 2. Incomplete or source specific EF, 3. Estimates of crop straw utilization and it's variability across regions/provinces, 4. Province level resolution of available inventories is not appropriate for modeling / evaluation emission impacts of atmospheric chemistry, climate or health.

Response:

We thank you very much for your careful and insightful review. We have taken the following comments into consideration in revision. Please see the following point-by-point responses.

Specific Comments:

P1, Ln 25-26 "Corn, rice and wheat represent the major crop straws, with their total emission contribution exceeding 80% for each pollutant." Please clarify for which pollutants ("each pollutant") crop straw combustion accounts for 80% of total emissions. Do they refer to SO_2, CO, CH_4, and Hg? Or all pollutants listed in lines 20-21? Do the authors mean that the combined emissions of corn, rice, and wheat account for 80% of the inventory total emissions of specific pollutants?

Response:

We thank you very much for your comment. We are truly sorry for the confusing sentence. In this sentence, each pollutant refers to all pollutants listed in lines 18-19.

We have revised this sentence in the revised version in Lines 26-27 on Page 1:

"Corn, rice and wheat represent the major crop straws. The combined emissions of these three straw types account for 80% of the total straw burned emissions for each specific pollutants mentioned in this study. "

Statements regarding emissions of EC and NH_3 are contradictory, please clarify / correct: P1, Ln 24-25 "...firewood contributes most to EC and NH3 emission." P1, Ln 26-27: "Corn straw burning has the greatest contribution to EC, NOx and SO_2 emissions; rice straw burning is dominant contributor to CO_2, VOC, CH_4 and NH_3 emissions.

Response:

We thank you very much for your comment. We are truly sorry about this unclear description. This sentence means that firewood contributes most to EC and NH_3 emission compared with other sources (indoor straw, In-field crop residue, livestock excrement, forest and grassland fire). As for the various crop straw types, corn straw burning has more contribution to EC, NOx and SO_2 emissions, and rice straw burning has more contribution to CO_2, NMVOC, CH_4 and NH_3 emissions compared with other straw types. We have revised this sentence in the revised version in Line 27 on Page 1 and Lines 1-2 on Page 2:

" As for the straw burning emission of various crops , corn straw burning has the largest contribution to EC, NOx and SO_2 emissions; rice straw burning has higher contribution to CO_2, NMVOC, CH_4 and NH_3 emissions; wheat straw burning has higher contribution to CO and Hg emissions."

P1, Ln 31: "The temporal distribution shows that higher emissions occurred in April, September, and October during the whole year." This statement is unclear. Do the authors mean that the combined emissions from April, September, and October exceeded emissions for the remainder of the year? Please clarify

Response:

We thank you very much for your comment. We are truly sorry for the confusing sentence. This sentence means that as for the emission from each month, these months have higher emission. According to our calculation, April, May, June and October are the top four months with higher emissions, due to the in-field crop residue burning. While as for EC, the emission in February, January, October and December are relatively higher due to the biomass domestic burning in heating season. We have made the corresponding revision in the revised manuscript in the P2L6-7:

"…April, May, June and October are the top four months with higher emissions, due to the in-field crop residue burning. While as for EC, the emission in February, January, October and December are relatively higher due to the biomass domestic burning in heating season."

P2, Ln3: "haolocarbon" important to secondary chemistry?

Response:

We thank you very much for your comment. We are truly sorry for the confusing description. Haolocarbon is unimportant to secondary chemistry. Ethylene, propylene, toluene, mp-xylene and ethyl benzene are major species of NMVOC, which are important to secondary chemistry. We have made the corresponding modification in the revised manuscript in Lines 10-12 on Page 2, Lines 18-25 on Page 18 and Lines 18-19 on Page 21:

Lines 10-12 on Page 2*: "The species with relatively higher contribution to NMVOC emission include ethylene, propylene, toluene, mp-xylene and ethyl benzene, which are key species for the formation of secondary air pollution."*

Lines 18-25 on Page 18: "The total NMVOC emission is 3474 Gg in this study. The alkenes are the major contributor of biomass burning NMVOC emissions. The contribution of alkenes to the total NMVOC emission is approximately 34%, more than that of alkane (28%), aromatics (24%), alkynes (13%) and others (1%). Among these species, ethylene, acetylene, propylene and 1-butylene are the major species of alkenes and alkynes, with the total contribution accounting for 40.1%. Ethane, n-propane, n-butane, and n-dodecane are the main species of alkanes, with the total contribution accounting for 14.0%. Benzene, toluene, styrene, mp-xylene and ethyl benzene are the major species of aromatics, with the total contribution of 16.6%. Several species *mentioned above are key for the formation of secondary air pollution, such as ethylene, propylene, toluene, mp-xylene and ethyl benzene (Huang et al., 2011). It illustrates that the biomass burning emission control is urgently needed for the air quality improvement. Detailed NMVOC species emission is shown in the Supplement (Fig. S5)."*

Lines 18-19 on Page 21: "Several species with higher contribution to NMVOC (e.g., ethylene, propylene, toluene, mp-xylene and ethyl benzene) are key species for the formation of secondary air pollution."

P2, Ln17 change "critical" to "significant"

Response:

We thank you very much for your suggestion. We have made the revision in revised manuscript in Line 24 on Page 2:

"...Biomass burning is also a significant source of greenhouse gases such as methane (CH_4) and carbon dioxide (CO_2) ..."

P2, L2: "The amount of straw outdoor burning in China in 2009 is 0.215 billion tons (MA, 2011)." This should statement should be qualified e.g. change: "is" to "was estimated as". Also, please provide a couple lines describing the data and the source of data, since the citation is not readily accessible.

Response:

Thanks very much for your comment. We have made the corresponding modification in the revised manuscript in Lines 11-12 on Page 3:

"...The amount of in-field crop residue burning in China in 2009 was estimated as 0.215 billion Mg. The data is obtained from the government report on the investigation and evaluation of crop straw resources in various provinces in China (MA, 2011)."

In addition, we have added the sources of the report in the reference list in the revised manuscript:

"MA: Investigation and Evaluation Report on Crop Straw Resources in China, Ministry of Agriculture, 2011, available at http://www.kjs.moa.gov.cn/, (in Chinese)."

P4 L3: Please provide a citation for the "energy statistical yearbook" and provide a brief (1 sentence) description of the yearbook. P4, L7 "statistical yearbook" is this the "energy statistical yearbook"? Please clarify. P4, L11 "yearbook" is this the "energy statistical yearbook"? Please clarify.

Response:

We thank you very much for your comment. We have added the corresponding citation, description and clarification. Detailed content in the revised manuscript was listed below.

Lines 20-23 on Page 4: "Moreover, because of the lack of firewood consumption record in the China Energy Statistical Yearbook (NBSC, 2009-2015), few studies have developed a comprehensive biomass burning emission inventory in China in recent years. China Energy Statistical Yearbook provides official information on the energy construction, production and consumption, including the detailed firewood consumption in various regions. However, the firewood consumption data is no longer contained in the NBSC (2009-2015) since 2008."

We have added the corresponding citation in the reference list in the revised manuscript:

"National Bureau of Statistics of China (NBSC): China Energy Statistical Yearbook 2009−2015, China Statistics Press, Beijing, 2009−2015, available at http://www.stats.gov.cn/tjsj/tjcbw/, (in Chinese)."

P4, L7 Here "statistical yearbook" refer generically to all statistical yearbook. Actually, the detailed firewood consumption could be not obtained from any other yearbook in addition to energy statistical yearbook.

P4, L11 Here "yearbook" refer generically to all statistical yearbook (e.g. China Statistical Yearbook, China Rural Statistical Yearbook, etc.) in China.

We have made the corresponding revision in Lines 25-28 on Page 4 and Lines 1-3 on Page 5:

"…First, not all biomass burning sources have been included in recent years, especially since 2008, because of the lack of firewood consumption data in the various statistical yearbooks (e.g. China Energy Statistical Yearbook, China statistical

yearbook, China rural statistical yearbook). Second, the source-specific EFs used in emission estimation need to be updated based on the systematic combing of local tests in the latest research. Third, the proportion of crop straw domestic burning and in-field crop residue burning, which could reflect the recent conditions of different provinces in China needs to be investigated. Fourth, the current biomass burning emission inventory for China is generally at province resolution because detailed activity data cannot be directly obtained from the various statistical yearbooks in China…"

P5, L1 delete "including" before "domestic combustion" The authors use the term "field burning" to refer to burning of crop residue in the field, and grassland and forest fires. The widely used terminology in biomass burning research refers to in-field crop residue burning, grassland, and forest fires as "open burning". The author should use this terminology not "field burning" when referring to the combined crop residue, grassland, and forest burning. The use of "field burning" by the authors is inconsistent with accepted terminology and is confusing, a forest is not a "field".

Response:

We thank you very much for your comment. We have made the corresponding modification:

"The biomass burning considered in this study is mainly divided into two categories, domestic combustion and open burning. Domestic combustion mainly involves crop straw, firewood and livestock excrement (mainly used in pastoral and semi-pastoral areas) burning. Open burning includes in-field crop residue burning, forest and grassland fire."

Similar description has been revised through the full text in the revised manuscript.

P5, L11 What is the unit "a" in Mg/a?

Response:

We thank you very much for your comment. We have changed the unit "Mg/a" to "Mg/yr" in the manuscript. In addition, all the "a" in the unit have been changed to "yr" through the full text in the revised manuscript.

P5, L23: Please clarify the source of data in Figure S1 (statistical yearbook?) and note that it is prefecture level.

Response:

Thanks very much for your suggestion. The source of data in Figure S1 is from China Statistical Yearbook (NBSC, 2013b). We have added the corresponding content in the revised manuscript in Lines 24-26 on Page 6 and Lines 1-2 on Page 7, and in the supplement in Figure S1:

Lines 24-26 on Page 6 and Lines 1-2 on Page 7: *"There are currently no statistics on the amount of each crop yield at the county resolution ($P_{i,k}$) in various yearbooks in China. Therefore, in this study, we conducted a correlation analysis between grain yield and crop yield at prefecture resolution, and found a good correlation (R = 0.747, detailed analysis is provided in the Supplement, Fig. S1). The grain yield at prefecture resolution was summarized from China Statistical Yearbook in 2012 (NBSC, 2013b). The crop yield at prefecture resolution was summarized from statistical yearbooks edited by National Bureau of Statistics in 2012 for each province.…"*

Figure S1 in the revised supplement:

"S1 The correlation between crop yield and grain yield at prefecture resolution.

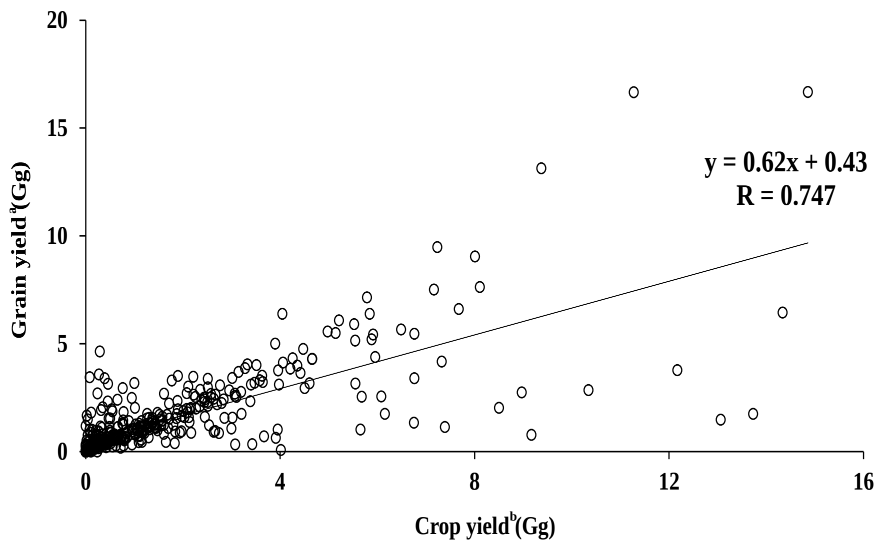

Figure S1 The correlation between crop yield and grain yield at prefecture resolution.
Note: [a] NBSC (2013); [b] a range of statistical yearbooks edited by National Bureau of Statistics in 2012 for each province.

Table 3. Are the superscripts denoting reference for N_k also the references for D_k and

CE_k? Please clarify.

Response:

Thank you very much for the comment. We are truly sorry for the missing reference of D_k and CE_k. The reference for D_k and CE_k is He et al. (2015). We have add the corresponding reference in Table 3 on Page 38:

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

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

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

Section 2.2.2 Firewood. Please clarify exactly which regression equation(s) were used to predict firewood consumption.

Response:

Thanks very much for your comment. According our correlation analysis between firewood consumption and other factors that may have a relationship with the firewood (rural population, gross agricultural and timber yield), we found that the rural population and firewood consumption have the best correlation relationship. Because the regression equation is various for the different historical years (Figure 1, 1998-2007), the regression analysis results was used to find the main factor which could be applied to calculate the detailed firewood consumption. Then the detailed firewood consumption was estimated based on the rural population.

P6, L26 What is the unit "a" in ton/a? Is a = "annum"? If so I recommend using year ("yr") instead. Also, is this metric ton?

Response:

We thank you very much for your suggestion. We have changed the "a" to "yr" in the units through the full text in the revised manuscript. In addition, we have changed the "ton" to "Mg" in the units through the full text.

P6, L26: "damaged area" = "burned area"? I assume by "damage area" the authors mean burned area. I recommend the authors use "burned area" instead of "damaged area" for consistency with biomass burning literature and accepted terminology. From an ecological standpoint a burned forest or grassland is not generally "damaged" since fire is a natural part of many if not most ecosystems.

Response:

We thank you very much for your suggestion. We are truly sorry for the confusing description. In fact, the "damaged area" in the original manuscript means "burned area". We have made the corresponding modification through the full text in the revised manuscript.

P6, L29: More details are needed on the data used and the method used to determine the spatial and temporal distribution of burned area. 1. Describe the "damaged area" data from NBSC (2013c) and NBSC (2013d). a. Is the data county level, prefecture level, province level? b. What is the time resolution of the data (annual, monthly, daily)? c. How was the data collected, e.g. is it based on administrative reports from local land management agencies? d. Does the dataset include both wildfires and fires used for ecosystem management, e.g. clearing logging debris or rangeland burning for grazing? 2. Provide a web link to where the references NBSC (2013c) and NBSC (2013d) can be accessed.

Response:

We thank you very much for your comment.

1. The response about the "damaged area" data from NBSC (2013c) and NBSC (2013d):

1.a. The data of damaged area from NBSC (2013c) and NBSC (2013d) is at provincial-level;

1.b. The time resolution of the data is annual resolution;

1.c. The data was collected from the China Statistical Yearbook published by National Bureau of Statistics of China;

1.d. This dataset did not include the fires used for ecosystem management.

Considering the low temporal and spatial resolution of statistics data for burned area, we have updated our methods and data employed in this study, and have re-calculated the pixel-based emission of forest and grassland fire using the bottom-up method. The daily burned area data is derived from the moderate-resolution imaging spectroradiometer (MODIS) direct broadcast burned area satellite product (MCD64A1; http://modis-fire.umd.edu) with a primary spatial resolution of 500 m. Detailed description about the methodology employed for the estimation of the biomass burning emission owing to forest and grassland fire was listed in Sect. 2.2.3 of the revised manuscript**:**

"2.2.3 Forest and grassland burning

The burning mass of forest/grassland can be calculated from the annual mass of forest/grassland burned (Mg/yr) as Eq. (3):

$$A = \left(\sum_{j=1}^{10} BAx,j \times FLx,j \times CFj\right) \times 10^{-6}, \quad (3)$$

where subscripts j, and x represent the land cover type, and location, respectively, $BA_{x,j}$ is the burned area (m^2) of land cover type j at x, $FL_{x,j}$ is the biomass fuel loading (the aboveground biomass density in this study; g/m^2) of land cover type j at x, and CF_j is the combustion factor (the fraction of burned aboveground biomass) of land cover type j.

Burned area data for 2012 were derived from the moderate-resolution imaging spectroradiometer (MODIS) direct broadcast burned area product (MCD64A1; http://modis-fire.umd.edu). This product employs an automated algorithm for mapping

MODIS post-fire burned areas, and deriving the approximate burn date within each burn cell combined with surface reflectance, land cover products, and daily active fires. The MCD64A1 product has a primary spatial resolution of 500 m and a temporal resolution of 1 month. The extent of burning over a Julian day and its temporal uncertainty are specified for each burn cell. The burned areas within an approximate Julian day can be extracted from the original 500 m resolution map.

Earlier research on the estimation of FL values for forest and grassland typically employed an averaged value of aboveground biomass density. However, these values do not well reflect the spatial variations of FL for each vegetation type. In this study, numerous local FL were collected for each province and vegetation type. The type of vegetation burned in each pixel was determined by the 1 km resolution MODIS Land Cover product produced by Ran et al. (2010). We considered 10 vegetation types as forest and grassland (i.e., evergreen needleleaf forest, evergreen broadleaf forest, deciduous needleleaf forest, deciduous broadleaf forest, mixed forest, closed shrublands, open shrublands, woody savannas, savannas, and grassland). The values of FL employed in this study are listed in Table 4. As for CF, it has usually been set as a constant in previous literature. In our paper, CF values were collected for each vegetation type, and the CF in each pixel was determined by the MODIS Land Cover product and the CF of typical vegetation. The CF of forest, closed shrublands, open shrublands, woody savannas, and grassland were set as 0.25, 0.5, 0.85, 0.4, and 0.95, respectively (Michel et al., 2005; Kasischke et al., 2000; Hurst et al., 1994).”

2. The web link of the references NBSC (2013c) has been added in the reference list in the revised manuscript. The references NBSC (2013d) have been deleted in the revised manuscript:

“*National Bureau of Statistics of China (NBSC): China Statistical Yearbook 2013, China Statistics Press, Beijing, 2013c,* ***available at http://www.stats.gov.cn/tjsj/ndsj/****, (in Chinese)*.”

P6, L31: A more detailed description of the forest and grassland biomass and combustion efficiency data is needed: 1. Provide a web link to where the references Tian et al. (2003), Lu et al. (2011), and EPD (2013) can be accessed. If they are not accessible, the work is not reproducible. 2. What forest components does the biomass number listed in Table 4 include? Organic soil/duff, litter, down dead wood, understory herbs and shrubs? 3. Is the forest biomass derived from forest inventory data? 4. The combustion efficiency of forests is 0.1 – 0.2. Is this because much of the forest biomass numbers include boles and branches of live trees which do not burn? 5. Does grassland category include shrub lands? 6. Please comment on how the biomass loadings and consumption estimates used in this study compare with those used in previous global emission inventories (e.g. GFED, van der Werf et al., 2010; FiNN, Wiedinmyer et al., 2011) and surveys of fuel consumption (e.g. van Leeuwen et al., 2014) and studies of grassland biomass in China (e.g. Ni, 2004; Ma et al. 2016; Zhao et al., 2014) 7. The value of 1800 kg/ha (180 g/m^2) used in this study compares reasonably well with Ni (2004) study of northern China Northern (325.5 g/m^2).

Response:

Thanks very much for your comment.

Considering the coarse resolution of statistical data and the lack of fires used for ecosystem management, we have changed the dataset and method employed in this study, and we re-calculated the pixel-based emission of forest and grassland fire. Detailed description could be found in the response to the comment mentioned above..

Besides, we further elaborate on the biomass fuel loadings and combustion factor data. As the values of biomass fuel loadings are various from vegetation types and provinces, in this study, numerous local biomass fuel loadings were collected for various vegetation types and provinces (Fang et al., 1996; Fang et al., 1998; Pu et al., 2004; Hu et al., 2006). Combustion factor of various vegetation types were derived from Michel et al. (2005), Levine et al. (2000), Kasischke et al. (2000), and Hurst et al. (1994).

Detailed description about the methodology employed for the estimation of the biomass burning emission owing to forest and grassland fire was listed in Sect. 2.2.3 of the revised manuscript. Please see the response to the comment mentioned above.

In addition, the specific responses to the comments are listed below:

1. The web link of the references Tian et al. (2003), Lu et al. (2011), and EPD (2014)

has been added in the reference list in the revised manuscript

"Tian, X. R., Shu, L. F. and Wang, M. Y.: Direct Carbon Emissions from Chinese Forest Fires, 1991−2000, Fire Safety Science, 12, 6−10, 2003, http://hzkx.ustc.edu.cn/ch/reader/view_abstract.aspx?flag=1&file_no=200312002&journal_id=hzkx, (in Chinese).

Lu, B., Kong, S. F., Han, B., Wang, X. Y. and Bai, Z. P.: Inventory of Atmospheric Pollutants Discharged from Biomass Burning in China Continent in 2007, China Environmental Science, 31, 186−194, 2011, http://manu36.magtech.com.cn/Jweb_zghjkx/CN/Y2011/V31/I2/186, (in Chinese).

EPD: Guide for compiling atmospheric pollutant emission inventory for biomass burning, Environmental Protection Department, 2014, http://www.zhb.gov.cn/gkml/hbb/bgg/201501/t20150107_293955.htm, (in Chinese)."

2. The forest components in the revised manuscript include trunk, branch and leaves of trees.
3. The forest biomass in the revised manuscript is not derived from forest inventory data. The forest inventory data only provides the trunks biomass of the forest. In addition, the data used in our paper also involves branches and leaves biomass of trees.
4. The combustion efficiency of forests in the revised manuscript was set as 0.25 according to Michel et al. (2005).
5. Grassland category in the revised manuscript including woody savannas, savannas and grasslands. shrub lands are included in the forest category.
6. The detailed description on the estimation of biomass loadings and consumption in this study could be found in the response to the comment mentioned above (Sect. 2.2.3). The biomass fuel loadings data are collected from the research made in China (Fang et al., 1996; Fang et al., 1998; Pu et al., 2004; Hu et al., 2006), and the data is often used in recent studies on the estimation of biomass burning emission and proved to be credible. (Song et al., 2008; Qiu et al., 2016).

7. In revised manuscript, the biomass fuel loadings of grassland are different from province and vegetation type.

P7, L1 Units for grassland biomass are kg/ha while other quantities are listed as kg/hm^2. While these units are equivalent, please be consistent by using either ha or hm^2 throughout the manuscript.

Response:

Accepted. We have made the corresponding modification through the full text in the revised manuscript.

Table 4. Note the units for forest biomass.

Response:

Accepted. The unit of forest biomass is g/m^2, which has been added in Table 4 in the revised manuscript.

P7, L10: Provide a web link to where the references EOCAIY (2013) and NBSC (2013b) can be accessed

Response:

Thanks very much for your comment.

The web link of NBSC (2013b) has been added in the reference list in the revised manuscript:

"National Bureau of Statistics of China (NBSC): China Statistical Yearbook for Regional Economy 2013, China Statistics Press, Beijing, 2013b, available at http://www.stats.gov.cn/tjsj/tjcbw/, (in Chinese)."

China Animal Industry Yearbook (EOCAIY 2013) is edited by the editorial committee of Chinese animal husbandry, which is a reference about the information of animal husbandry and veterinary medicine, feed and forage industry. It is widely used in recent research, such as Kang et al. (2016) and Huang et al. (2012). However, it is not publicly available online.

Section 2.3 EFs The VOC emissions factors for forest, grasslands, open residue burning, and feces burning seem quite low compared to those reported in extensive reviews such as Akagi et al. (2011). I imagine the difference is that the VOC category in Table 5 & 6 include only a subset of VOC present in biomass smoke and measured in other studies. Please comment in the differences.

Response:

Thanks very much for your comment. The NMVOC emission factors were updated based on a systematic combination of localized measurements conducted in China. According to our examination about the references selected for In-field crop residue burning and feces burning in this study, NMVOC emission factor include alkane, alkene, alkyne and aromatics with C2-C12. In the revised manuscript, the EF for forest and grassland fire was selected from Akagi et al. (2011) due to the lack of localized measurement. The emission of NMVOCs species has been revised according to the species corresponding to emission factor.

Section 2.4 Spatial Distribution Is the land use data of Ran et al. (2010) publicly available? If so, please provide a web link where it may be accessed.

Response:

Thanks very much for your comment. The spatial distribution of the land use data is MODIS land cover data which is processed by Ran et al. (2010). The web link of the reference has been added in the reference list in the revised manuscript:

"Ran, Y. H., Li, X. and Lu, L.: Evaluation of four remote sensing based land cover products over China, Int. J. Remote Sens., 31, 391−401, available at http://www.tandfonline.com/doi/abs/10.1080/01431160902893451, 2010."

Figure 2 is not very useful. It should be replaced with or augmented with table that provides the total emissions and percent of each species by source.

Response:

We thank you very much for your suggestion. As the total emissions of each species have already been mentioned in the manuscript and in the Table 7, we marked the percent of each species by source in the Fig. 2.

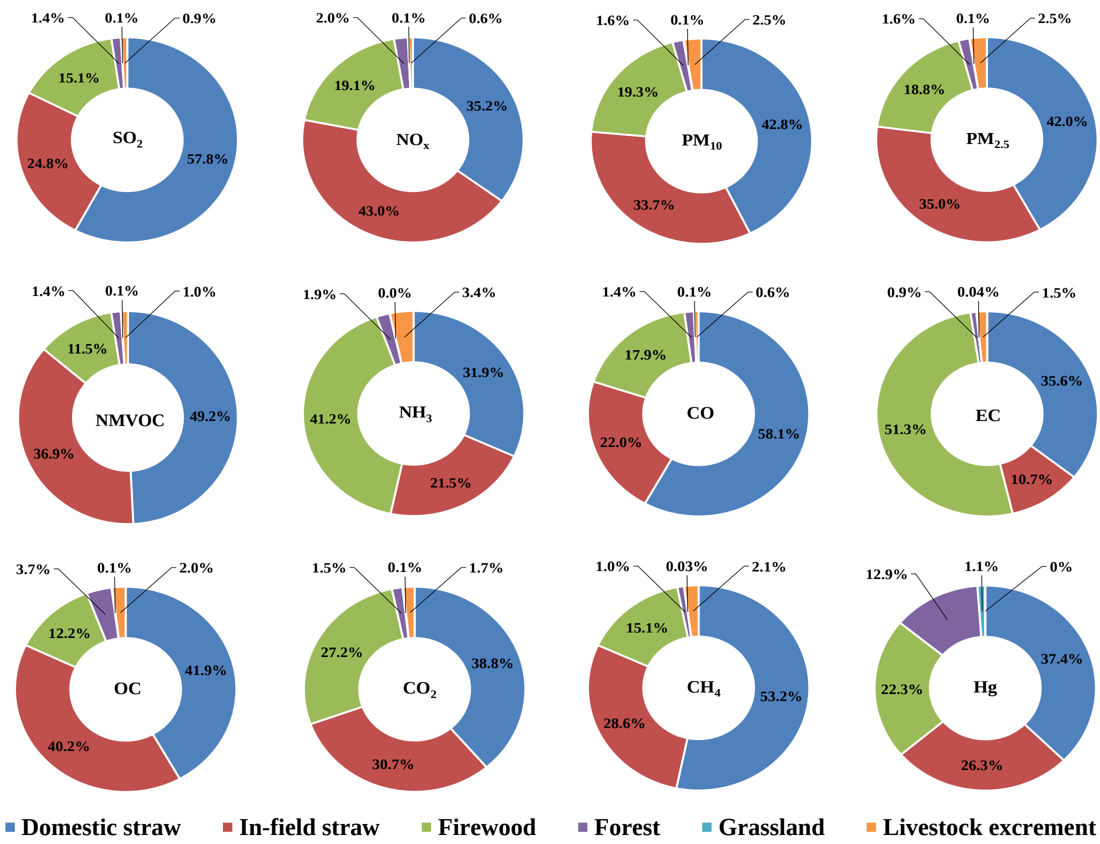

Figure 2. Contribution of different source to the total biomass burning emissions in China, 2012.

Results and discussion. Please include a table providing total annual fuel consumption and emissions for the 12 species in Table 7 by source. This will is needed to compare current paper to previous studies that may have focused on only a subset of sources.

Response:

We thank you very much for your suggestion. We have added the total annual fuel consumption by source in order to discuss the result (Sect. 3.1.1 in lines 15-25 on Page 11 and lines 1-13 on Page 12). The emissions for the 12 species by source can be calculated through the total emission in Table 7 and the percent of each species showed in Fig. 2.

lines 5-8 on Page 12: *"...In addition to the sources mentioned above, the contribution of livestock excrement burning, forest and grassland fire is relatively small. It is mainly due to the small amount of biomass fuel consumption. The biomass fuel consumption of these three biomass sources are 10614 Gg, 6647 Gg and 505 Gg, respectively, which is* *significantly lower than that of straw domestic combustion (201582 Gg), in-field crop residue burning (147178 Gg) and firewood combustion (127250 Gg)…"*

P9 L15-16 Please note the total annual burned area of forest and grasslands.

Response:

We thank you very much for your comment. The total burned area of forest and grasslands are 3587 and 4241 km^2, respectively. As the discussion content in P9 L15-16 in the original manuscript is about the contributions by each biomass burning sources. The fuel consumption has more direct influence on emission estimation compared with burned area of forest and grasslands. Therefore, we gave the fuel consumption here:

"…In addition to the sources mentioned above, the contribution of livestock excrement burning, forest and grassland fire is relatively small. It is mainly due to the small amount of biomass fuel consumption. The biomass fuel consumption of these three biomass sources are 10614 Gg, 6647 Gg and 505 Gg, respectively, which is significantly lower than that of straw domestic combustion (201582 Gg), in-field crop residue burning (147178 Gg) and firewood combustion (127250 Gg). The contribution of livestock excrement burning to PM10, PM2.5, NH3, EC, OC, CO2 and CH4 is 2.52%, 2.47%, 3.44%, 1.52%, 1.96%, 1.67% and 2.10%, respectively. The contribution of forest and grassland fire to biomass burning emissions to most chemical species in China is small (0.9–3.7%), except for the contribution of forest fire to Hg emissions (14.0%)."

Figures 4 & 5 are difficult to read and the data would be better presented as tables, perhaps in the supplement.

Response:

Thanks very much for your comment. Considering the occupied space of many information in Figures 4 & 5, the result is more suitable to present through figures. A furthermore quality improvement of the Figure 4 and Figure 5 has been made. The data in the figures could be read. In addition, the reader can get the detailed data freely through contacting us after the paper acceptation. Considering the importance of the result, it is better to present it in the main body of the manuscript.

P13, L13-14: Are specific crops typically burned harvest season, sowing season, or both? Or does it vary by region and practice?

Response:

Thanks very much for your comment. The specific crops typically burned in harvest season or sowing season, and it varies according to the burning habit in different regions. For example, wheat crop straw in the north often burned in its harvest season while rice crop straw in the south often burned in its sowing season to clear the cultivated land and increasing the soil fertility for the next sowing. In addition, due to the difference of climate conditions, the harvest and sowing season vary in various regions. Therefore, we discussed the temporal variation in biomass burning emission in different regions. We have revised the explanation in Lines 5-8 on Page 17:

"Burning activity mainly occurs in the harvest season (crop residue burning) or crop sowing season (clearing the cultivated land and increasing the soil fertility for the next sowing), and it varies according to the burning habit in different regions. In addition, the sowing and harvest seasons vary in different regions because of the climate conditions. Because of the differences in burning activity and climate conditions in various regions, monthly emission features vary regionally and to consider this, we divided China into seven areas…"

Section 3.6 Please describe how parameters were estimated for the PDFs used in the Monte Carlo simulation.

Response:

We thank you very much for your comment. We have made the corresponding modification in Lines 2-10 on Page 19:

"The Monte Carlo method is used to analyse the uncertainty of this emission inventory, which was used in uncertainties estimation for many inventories studies (e.g., Streets et al., 2003; Zhao et al., 2011; Zhao et al., 2012). Activity data (Zheng et al., 2009) and EFs (Zhao et al., 2011) are assumed to be normal distributions. The coefficients of variation (CV, the standard deviation divided by the mean) of activity data and emission factors were obtained from literature review. CV of activity data for

firewood and crop straw burning were set as 20% (Zhao et al., 2011; Ni et al., 2015). As the data source of activity data for livestock excrement is same as the crop straw burning (i.e., government statistic data), CV is also set as 20%. MCD64A1 burned data products has been shown to be reliable in big fires (Giglio et al., 2013), and the CV of burned area of forest and grassland fire is from the reported standard deviation (Giglio et al., 2010). The biomass fuel loadings (Saatchi et al., 2011; Shi et al., 2015) and combustion factor (van der Werf et al., 2010) of forest and grassland fire were within a CV of approximately 50%. The CV of EF for each pollutant for each biomass burning type is shown in the supplement S8 and S9."

Supplement S8 and S9 have been added in the revised supplement:

S8 CV (coefficients of variation) of biomass domestic burning emission factors.

	Material	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
Domestic burning	Corn	0.5[*]	0.02[a]	0.5[*]	0.27[b]	0.5[*]	0.5[*]	0.85[a]	0.34[b]	0.44[b]	0.04[a]	0.5[*]	0.05[c]
	Wheat	0.5[*]	0.16[a]	0.5[*]	0.23[b]	0.5[*]	0.5[*]	0.89[a]	0.76[b]	0.29[b]	0.07[a]	0.5[*]	0.12[c]
	Cotton	0.5[*]	0.5[*]	0.5[*]	0.26[b]	0.5[*]	0.5[*]	0.5[*]	0.39[b]	0.55[b]	0.5[*]	0.5[*]	0.33[c]
	Cane	0.5[*]	0.5[*]	0.5[*]	0.26[b]	0.5[*]	0.5[*]	0.5[*]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.32[c]
	Potato	0.5[*]	0.5[*]	0.5[*]	0.26[b]	0.5[*]	0.5[*]	0.5[*]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.53[c]
	Peanut	0.5[*]	0.5[*]	0.5[*]	0.26[b]	0.5[*]	0.5[*]	0.5[*]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.03[c]
	Rape	0.5[*]	1.21[d]	0.5[*]	0.15[b]	0.26[d]	0.5[*]	0.26[d]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.3[c]
	Sesame	0.5[*]	1.78[d]	0.5[*]	0.26[b]	0.24[d]	0.5[*]	0.29[d]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.3[c]
	Beet	0.5[*]	0.5[*]	0.5[*]	0.26[b]	0.5[*]	0.5[*]	0.5[*]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.3[c]
	Hemp	0.5[*]	0.5[*]	0.5[*]	0.26[b]	0.5[*]	0.5[*]	0.5[*]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.3[c]
	Rice	0.5[*]	0.05[a]	0.5[*]	0.29[b]	0.5[*]	0.5[*]	0.06[a]	0.65[b]	0.5[b]	0.01[a]	0.5[*]	0.46[c]
	Soybean	0.5[*]	1.78[d]	0.5[*]	0.26[b]	0.76[d]	0.5[*]	0.44[d]	0.63[b]	0.45[b]	0.5[*]	0.5[*]	0.74[c]
	Firewood	0.5[*]	1.42[d]	0.5[*]	0.16[b]	0.15[d]	0.5[*]	0.39[d]	0.46[b]	0.35[b]	0.5[*]	0.5[*]	1.17[c]
	Feces	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]	0.8[*]

Table S1 CV (coefficients of variation) of biomass domestic burning emission factors.

Note: Lowercase letters indicate the data source.

*Sources are from the following: [a] Zhang et al. (2008). [b] Li et al. (2009). [c] Chen et al. (2013). [d] Zhang et al. (2013). * Expert judgment data from Wei et al. (2011).*

S9 CV (coefficients of variation) of biomass open burning emission factors.

	Material	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
Open burning	Corn	0.45[b]	0.42[b]	0.5[*]	0.09[b]	0.53[b]	0.76[b]	0.08[b]	0.33[b]	0.39[b]	0.01[b]	0.22[b]	0.05[a]
	Wheat	0.67[b]	0.52[b]	0.5[*]	0.54[b]	0.25[b]	0.38[b]	0.41[b]	0.32[b]	0.26[b]	0.03[b]	0.25[b]	0.12[a]
	Cotton	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.33[a]
	Cane	0.5[*]	0.32[d]	0.19[d]	0.16[d]	0.71[d]	0.5[*]	0.61[d]	1.57[d]	0.2[d]	0.18[d]	0.5[*]	0.32[a]
	Potato	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.53[a]
	Peanut	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.03[a]
	Rape	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Sesame	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Beet	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Hemp	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Rice	0.5[*]	0.8[d]	0.88[d]	0.17[d]	0.75[d]	0.5[*]	1.19[d]	1.38[d]	1.53[d]	0.14[d]	0.5[*]	0.46[a]
	Soybean	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.74[a]
	Evergreen Needleleaf Forest	0.3[c]	0.39[c]	0.25[d]	0.25[d]	0.31[e]	0.66[e]	0.38[e]	1[f]	0.62[f]	0.08[e]	0.52[e]	0.52[g]
	Evergreen Broadleaf Forest	0.4[e]	0.54[e]	0.25[d]	0.25[d]	-	1.58[h]	0.29[e]	0.6[e]	0.57[e]	0.04[e]	0.39[e]	0.52[g]
	Deciduous Needleleaf Forest	0.3[c]	0.23[c]	0.25[d]	0.25[d]	0.31[e]	0.66[e]	0.38[r]	1[f]	0.62[f]	0.08[e]	0.52[r]	0.52[g]
	Deciduous Broadleaf Forest	0.3[c]	0.46[e]	0.25[d]	0.25[d]	0.79[e]	0.27[e]	0.19[e]	0.33[e]	0.52[e]	0.02[e]	0.18[e]	0.52[g]
	Mixed Forest	0.3[c]	0.46[e]	0.25[d]	0.25[d]	0.62[e]	0.27[e]	0.19[e]	0.33[e]	0.52[e]	0.02[e]	0.18[e]	0.52[g]
	Closed Shrublands	0.44[e]	0.21[e]	0.25[d]	0.25[d]	0.48[e]	0.33[e]	0.25[e]	0.4[f]	0.18[f]	0.02[e]	0.35[e]	0.74[h,g]
	Open Shrublands	0.44[e]	0.21[e]	0.25[d]	0.25[d]	0.48[e]	0.33[e]	0.25[e]	0.4[f]	0.18[f]	0.02[e]	0.35[e]	0.74[h,g]
	Woody Savannas	0.44[e]	0.21[e]	0.25[d]	0.25[d]	0.48[e]	0.33[e]	0.25[e]	0.4[f]	0.18[f]	0.02[e]	0.35[e]	0.52[h]
	Savannas	0.63[e]	0.29[e]	0.25[d]	0.25[d]	0.25[e]	0.8[e]	0.29[e]	0.5[e]	0.46[e]	0.02[e]	0.6[e]	0.52[h]
	Grasslands	0.63[e]	0.29[e]	0.25[d]	0.25[d]	0.25[e]	0.8[e]	0.29[e]	0.5[e]	0.46[e]	0.02[e]	0.6[e]	0.52[h]

Table S2 CV (coefficients of variation) of biomass open burning emission factors.

Note: Lowercase letters indicate the data source.

Sources are from the following: [a] *Chen et al. (2013).* [b] *Li et al. (2007).* [c] *Andreae and Rosenfeld (2008).* [d] *Song et al. (2009).* [e] *Akagi et al. (2011).*[f] *McMeekin et al. (2008).* [g] *Friedli et al. (2003).* [h] *Streets et al. (2005). * Expert judgment data from Wei et al. (2011).*

Figure S4. Please note the data sources used to derive the non-carbon PM components.

Response:

We thank you very much for your suggestion. The $PM_{2.5}$ speciation is obtained from Li et al., (2007) and Waston et al., (2001), which have been described in Sect. 2.6 in the original manuscript. In addition, we have added the data sources of the $PM_{2.5}$ speciation in Figure S4.

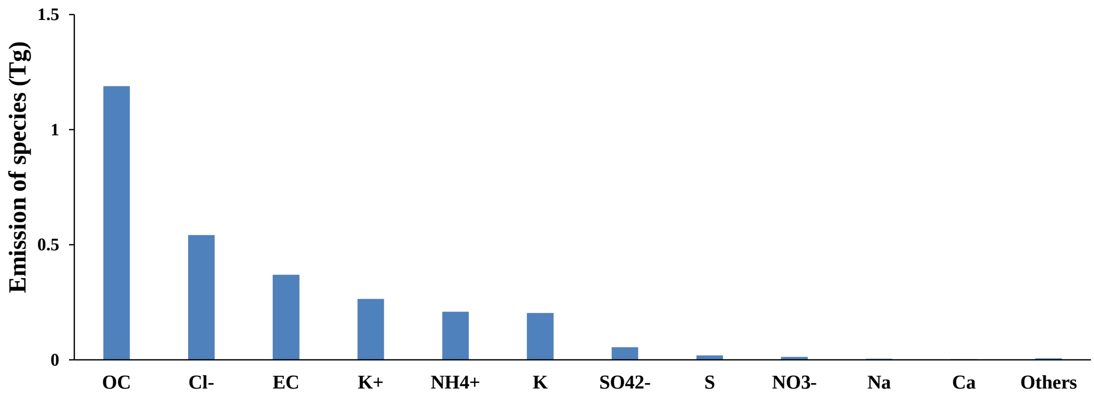

Figure S4 Emission of $PM_{2.5}$ species from biomass burning.

Note: Species in others include Al，Si，Mg，Fe，Pb，Zn，Ba，Ti，Ni，Cr，Mn，Sr，V，Cd，As，Zr，Se，Ag，Sb，Sc，Mo，Ga，Tl，Co and Hg. $PM_{2.5}$ speciation profile is obtained from Li et al., (2007) and Waston et al., (2001).

TECHINCAL The manuscript contains many minor grammatical errors, here are a few: P13, Ln3-4: Change “This is because the main contribution of these species emission sources is from straw outdoor burning” to “This is because straw outdoor burning is the main source for these species” P13, Ln 4: change “The outdoor burning straw mainly occurs in...” to “The outdoor burning of straw occurs mainly in...” P13, L19 insert “a” between “have” and “relatively” and change “peak” to “peaks” P13, L20: change “discrepancies” to “differences” P13, L28: change “while” to “where” P14, L7: change ‘peak” to “peaks” P14, L14: Change “Besides” to “Additionally” P16, L23-25 Sentence beginning “More localized EF of...” is jumbled and must be rewritten.

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Response:

We thank you very much for your helpful suggestion. We have made the corresponding modification in the revised manuscript.

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

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Abstract. Biomass burning injects many different gases and aerosols into the atmosphere, which could have a harmful effect on air quality, climate change and human health. In this study, a comprehensive biomass burning emission inventory including crop straw domestic combustion and in-field crop residue burning, firewood and livestock excrement combustion, forest and grassland fire was developed for mainland China in 2012 based on county-level activity data, satellite date, and updated source-specific emission factors (EFs). The emission inventory within 1 × 1 km grid was generated using geographical information system (GIS) technology according to source-based spatial surrogates. A range of key information related to emission estimation (e.g., province-specific proportion of crop straw domestic ~~burning~~ combustion and in-field crop residue burning~~open burning~~, detailed firewood combustion quantities, uneven temporal distribution coefficient) was obtained from field investigation, systematic combing of the latest research and regression analysis of statistical data. The established emission inventory includes the major precursors of complex pollution, greenhouse gases and heavy metal released from biomass burning. The results show that the emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, ~~VOC~~NMVOC, NH_3, CO, EC, OC, CO_2, CH_4 and Hg in 2012 ~~were 332.8 Gg, 972.5 Gg, 3676.0 Gg, 3479.4 Gg, 3429.6 Gg, 395.8 Gg, 33987.9 Gg, 367.1 Gg, 1151.7 Gg, 665989.0 Gg, 2076.5 Gg and 3.65 Mg, respectively. Indoor and outdoor burning of straw~~ are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34380.4 Gg, 369.7 Gg, 1189.5 Gg, 675299.0 Gg, 2092.4 Gg and 4.12 Mg, respectively. Straw domestic burning, in-field crop residue burning, and firewood combustion are identified as the dominant biomass burning sources. The largest contributing source is different for various pollutants. Straw ~~indoor~~ domestic burning is the major source of SO_2, CO, CH_4 and ~~Hg~~ NMVOC emission; firewood contributes most to EC and NH_3 emission. ~~Corn, rice and wheat represent the major crop straws, with their total emission contribution exceeding 80% for each pollutant. Corn straw burning has the greatest contribution to EC, NO_x and SO_2 emissions; rice straw burning is dominant contributor to CO_2, VOC, CH_4 and NH_3 emissions~~Corn, rice and wheat represent the major crop straws. The combined emissions of these three straw types account for 80% of the total straw burned emissions for each specific pollutant mentioned in this study. As for the straw burning emission of various crops, corn straw burning has the largest contribution to EC, NOx and SO_2 emissions; rice straw burning has higher contribution to CO_2, NMVOC, CH_4 and NH_3 emissions; wheat straw burning has higher contribution to CO and Hg emissions. Heilongjiang, Shandong, and Henan provinces located in northeast and central-south region of China have higher emissions. Gridded emissions, which were obtained through spatial allocation based on the gridded rural population and fire point data from emission inventory at county resolution, could better represent the actual situation. Higher biomass burning emissions are concentrated in the areas with greater agricultural and rural activity. The temporal distribution shows that April, May, June and October are the top four months with higher emissions, due to the in-field crop residue burning. While as for EC, the emission in February, January, October and December are relatively higher due to the biomass domestic burning in heating season.~~The temporal distribution shows that higher emissions occurred in April, September, and October during the whole year.~~ There's regional difference in monthly variation due to the diversity of main planted crop and the climate conditions. Furthermore, $PM_{2.5}$ component results showed that OC, Cl^-, EC, K^+, NH_4^+, K element and SO_4^{2-} ~~-~~are the main $PM_{2.5}$ species accounting for 80% of the total emissions. The species with relatively higher contribution to ~~VOC~~NMVOCs emission includ e~~ing~~ ethylene, propylene, toluene~~,~~, mp-xylene ~~and~~ and ethyl benzene, ~~halocarbons~~ which are key species for the formation of secondary air pollution. The detailed biomass burning emission inventory generated by this study could provide useful information for air quality modelling and support the development of appropriate pollution control strategies.

Keywords: Biomass burning; Emission inventory; High resolution; Species

1 Introduction

Biomass burning is considered a significant source of gas and particulate matter (PM), resulting in a major impact on atmospheric chemistry, climate change and human health. Active trace gases (e.g., SO_2, NO_x, ~~VOC~~NMVOCs, NH_3) released from biomass burning are the major precursors of secondary inorganic/organic aerosols and tropospheric ozone (O_3) in the atmosphere (Penner et al., 1992; Kaufman and Fraser, 1997; Koppmann et al., 2005; Langmann et al., 2009). Several studies have indicated that observed local and regional air pollution could be attributed to the chemical species emitted from biomass burning (Huang et al., 2012b; Zha et al., 2013; Cheng et al., 2014; Yan et al., 2014; Zong et al., 2016). The emission factor (EF) of some biomass burning pollutants is even greater than coal burning, which is widely recognized as a major pollution source (Zheng et al., 2009； Fu et al., 2013). Primary particles (e.g., BC and OC) discharged by biomass burning not only impact visibility, but also have an influence on climate change due to the positive effects of the absorption of light and cloud condensation (IPCC, 2011). Biomass burning is also a ~~critical~~ significant source of greenhouse gases such as methane (CH_4) and carbon dioxide (CO_2) (Andreae and Merlet, 2001), which contribute to global warming (Sun et al., 2016). Moreover, several reports (Fernandez et al., 2001; Huang et al., 2012b; Shi and Yamaguchi, 2014) reveal that the long-term or short-term exposure to PM (e.g., BC emitted from indoor biomass burning) can cause adverse effects to human health, such as decreased lung function, increased respiratory diseases and lung cancer mortality. Furthermore, studies have identified that indoor biomass burning could bring adverse health effects on residents (Jiang and Bell, 2008; Fullerton et al., 2008).

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

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

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

Based on the local EFs, emission inventories that focused on certain provinces (Li et al., 2015; He at al., 2015) or city group regions (He at al., 2011; Fu et al., 2013) were developed. In our previous study, we reported an emission inventory with high resolution in the Beijing–Tianjin–Hebei region of China (Zhou et al., 2015). To produce a national emission inventory, several studies of biomass burning have been carried out without distinguishing the detailed crop straws (Lu et al., 2011; Yan et al., 2006; Tian et al., 2011). Moreover, there are several studies that have focused on certain pollutants (Huang et al., 2012d; Chen et al., 2013; Zhang et al., 2013a; Kang et al., 2016; Li et al., 2016), and certain crop straws (Zhang et al., 2008; Hong, et al., 2016; Sun, et al., 2016). In recent years, the comprehensive biomass emission inventory is limited. Most of recent studies are concentrated upon biomass open burning, including the multi-year trend analysis on certain or multiple pollutants (Wang and Zhang, 2008; Song et al., 2009; ~~Huang et al., 2012c;~~ Shi et al., 2014; Shon, 2015; Xu et al., 2016; Zhang et al., 2016). Few studies have covered recent firewood burning (see next paragraph for details regarding the reason for this). In addition to the EF, detailed activity data are also important for a reliable emission inventory, such as straw domestic or in-~~–~~field crop residue burning ratios, which are not currently publicly available. Gao et al. (2002) produced a study on the percentage of straw used as fuel and for direct incineration in 2000. Wang et al. (2008) investigated the percentage of in-field crop residue~~crop open~~ burning in 2006 of six regions in China, which were divided according to the similarities of agriculture, climate, economy and region. Tian et al. (2011) estimated the proportion of crop straw domestic burning and in-field crop residue~~open~~ burning in 2007 for seven and three regions of China, respectively. Thus, there is limited information about the ratio of straw used as fuel and~~to that~~ crop residue burning~~t~~ in the field that reflects the status of China in recent years for different provinces. ~~Moreover, because of the lack of firewood consumption in the energy statistical yearbook after 2007, there are few reports containing a comprehensive biomass burning emission inventory for China.~~ Moreover, because of the lack of firewood consumption record in the China Energy Statistical Yearbook (NBSC, 2009-2015), few studies have developed a comprehensive biomass burning emission inventory in China in recent years. China Energy Statistical Yearbook provides official information on the energy construction, production and consumption, including the detailed firewood consumption in various regions. However, the firewood consumption data is no longer contained in the NBSC (2009-2015) since 2008, as a result, there are few literature containing a comprehensive biomass burning emission inventory for China.

Consequently, we have identified several weaknesses in the current biomass burning emission inventories. First, not all biomass burning sources have been included in recent years, especially since 2008, because of the lack of firewood consumption data in the various statistical yearbooks (e.g. China Energy Statistical Yearbook, China statistical yearbook, China rural statistical yearbook). Second, the source-specific EFs used in emission estimation need to be updated based on the systematic combing of local tests in the latest research. Third, the proportion of crop straw domestic burning and in-field crop residue burning, which could reflect the recent conditions of different provinces in China needs to be investigated. Fourth, the current biomass burning emission inventory for China is generally at province resolution because detailed activity data cannot be directly obtained from the various statistical yearbooks in ChinaFirst, not all biomass burning sources have been included in recent years, especially after 2007, because of the lack of firewood consumption data in the statistical yearbook. Second, the source-specific EFs used in emission estimation need to be updated based on the systematic combing of local tests in the latest research. Third, the proportion of crop straw domestic burning and open burning, which could reflect the recent situation of different provinces in China needs to be investigated. Fourth, the current biomass burning emission inventory for China is generally at province resolution because detailed activity data cannot be directly obtained from the yearbook. Activity data at coarse resolution are likely to be associated with greater uncertainty in grid emissions generated according to source-based gridded spatial surrogates (e.g., population) using GIS technology (Zheng et al., 2014). As a result, it is of great importance to develop an integrated and model-ready biomass burning emission inventory with high spatial and temporal resolution.

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

The remainder of this paper is structured as follows. Section 2 describes the methodology including the emission estimation method, the selection and handling of activity data and corresponding parameters, determination of EFs, spatial and temporal allocation, and speciation of $PM_{2.5}$ and VOCNMVOCs. Section 3.1 describes the total emission in China, and the contribution of various biomass burning sources and crop straws. Section 3.2 describes the emission from different regions, and contributions of different biomass sources and crop straws of each province. Spatial and temporal distribution of biomass burning emissions is discussed in Secs. 3.3 and 3.4, respectively. Section 3.5 presents the emissions of $PM_{2.5}$ and VOCNMVOC species. Uncertainty in biomass burning emission estimates is described in Section 3.6. The comparison between this study and other studies appears in Section 3.7. Section 4 summarizes the conclusions.

2 Methodology

2.1 General description

The biomass burning considered in this study is mainly divided into two categories, domestic combustion and open burning. Domestic combustion mainly involves crop straw, firewood and livestock excrement (mainly used in pastoral and semi-pastoral areas) burning. Open burning includes in-field crop residue burning, forest and grassland fire. Details of the source classifications are shown in Table 1.The biomass burning considered in this study is mainly divided into two categories including domestic combustion and in field burning. Domestic combustion mainly involves crop straw, firewood and livestock excrement (mainly used in pastoral and semi-pastoral areas). In field burning includes seasonal crop residue waste burning, grassland and forest fires. Details of the source classifications are shown in Table 1.

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

$$E_i = \sum(A_i \times EF_{i,j}) \times / 1000, \quad (1)$$

where subscripts *i* and *j* represent the type of pollutant and biomass burning source; E is the annual typical pollutant emission (Mg/yra); A is annual amount of dry biomass burned (Mg/yra), for which the detailed calculation method is shown in Sec. 2.2; and EF is the emission factor (g/kg), for which a detailed description is presented in Sec. 2.3.

2.2 Activity data

2.2.1 Straw burning

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

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

where subscripts *i* and *k* represent region (district or county) and crop type, respectively; $A_{i,k}$ is the annual burning mass of crop straw (Mg/yrton/year); $P_{i,k}$ is the amount of crop-specific yields per year (Mg/yrton/year); N_k is the residue-to-production ratio of each straw type (ton/tonMg/Mg); $R_{i,k}$ is percentage of crop straw burned as fuel or in field burning; D_k is dry matter fraction of each straw type; and CE_k is the combustion efficiency of each straw type.

There are currently no statistics on the amount of each crop yield at the county resolution ($P_{i,k}$) in the statisticalvarious yearbooks in China. Therefore, in this study, we conducted a correlation analysis between grain yield and crop yield at prefecture resolution, and found a good correlation (R = 0.747, detailed analysis is provided in the Supplement, Fig. S1). The grain yield at prefecture resolution was summarized from China Statistical

Yearbook in 2012 (NBSC, 2013b). The crop yield at prefecture resolution was summarized from statistical yearbooks edited by National Bureau of Statistics in 2012 for each province.Next, the $P_{i,k}$ was calculated based on the various types of crop yield at prefecture resolution and grain yield at county resolution, which was summarized from a range of statistical yearbooks in 2012 for each province (including the city yearbooks which are publically available for some cities), NBSC (2013a) and NBSC (2013b). The total straw amount of China in 2012 calculated in this study is 832.5 Tg, which is similar to NDRC (2014) (817.4 Tg) Next, the $P_{i,k}$ at county level was calculated based on the various types of crop yield at prefecture resolution and grain yield at county resolution. Grain yield at county resolution was summarized from a range of statistical yearbooks edited by National Bureau of Statistics in 2012 for each province and city, NBSC (2013a) and NBSC (2013b). The total straw amount of China in 2012 calculated in this study is 832.5 Tg, which is similar to the data of Chinese governmental annual statistical reports about the straw utilization and burning (NDRC, 2014; the amount of straw can be collected is 817.4 Tg). The map at prefecture and county resolution is shown in Fig. S2 in the Supplement.

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

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

2.2.2 Firewood

Firewood consumption is recorded as non-commodity energy in the China energy statistical yearbook. However, detailed firewood consumption has not been publicly available since 20087. For more recent years, we obtained the total firewood consumption for China in 2012 and for each province in 2010 (Tian et al., 2014; IEA, 2012). However, these data could not support the development of an emission inventory at high resolution. There are several detailed statistics available in the yearbook, such as the rural population, gross agricultural output and timber yield, which are likely to have a relationship with the firewood consumption. Therefore, we produced a correlation analysis between the three statistics and the firewood consumption of each province for different years in which the firewood consumption data were available at province resolution, as shown in Fig. 1. The best correlation relationship was found between rural population and firewood consumption. The correlation coefficient for the different years ranged from 0.66 to 0.82, therefore, we choose rural population as the surrogate to calculate the detailed firewood consumption. . The firewood consumption at county resolution was obtained based on the rural population at county resolution and the total firewood consumption reported by Tian et al. (2014) and IEA (2012). China's rural population, gross agriculture output and timber yield of each province come from NBSC (1999-2008a). Firewood consumption comes from NBSC (1999-2008b).

2.2.3 ~~Biomass burning of forest/grassland fires~~Forest and grassland burning

The burning mass of forest/grassland can be calculated from the annual mass of forest/grassland burned (Mg/yr) as Eq. (3):~~The burning mass of forest/grassland can be calculated using Eq. (3) as follows:~~

$$A = \left(\sum_{j=1}^{10} BAx,j \times FLx,j \times CFj\right) \times 10^{-6}, \quad (3)$$

where subscripts j, and x represent the land cover type, and location, respectively, $BA_{x,j}$ is the burned area (m^2) of land cover type j at x, $FL_{x,j}$ is the biomass fuel loading (the aboveground biomass density in this study; g/m^2) of land cover type j at x, and CF_j is the combustion factor (the fraction of burned aboveground biomass) of land cover type j.

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

Earlier research on the estimation of FL values for forest and grassland typically employed an averaged value of aboveground biomass density. However, these values do not well reflect the spatial variations of FL for each vegetation type. In this study, numerous local FL were collected for each province and vegetation type. The type of vegetation burned in each pixel was determined by the 1 km resolution MODIS Land Cover product produced by Ran et al. (2010). We considered 10 vegetation types as forest and grassland (i.e., evergreen needleleaf forest, evergreen broadleaf forest, deciduous needleleaf forest, deciduous broadleaf forest, mixed forest, closed shrublands, open shrublands, woody savannas, savannas, and grassland). The values of FL employed in this study are listed in Table 4. As for CF, it has usually been set as a constant in previous literature. In our paper, CF values were collected for each vegetation type, and the CF in each pixel was determined by the MODIS Land Cover product and the CF of typical vegetation. The CF of forest, closed shrublands, open shrublands, woody savannas, and grassland were set as 0.25, 0.5, 0.85, 0.4, and 0.95, respectively (Michel et al., 2005; Kasischke et al., 2000; Hurst et al., 1994).

$A = AR \times B \times \eta,$ (3)

where A is the annual burning amount of forest/grassland (ton/a); AR is the damaged area of grassland or forest fire per year (hm^2/a); B represents the dry biomass density of grassland or forest (ton/hm^2); and η is the combustion efficiency of grassland or forest.

The damaged area of forest fire and grassland fire could be obtained from NBSC (2013c) and NBSC (2013d), respectively. The specific locations at which fire occurred can be determined according to Moderate Resolution Imaging Spectroradiometer (MODIS) fire point data; the B and η of forest is different from the climatic zones, which is shown in Table 4; B for grassland is 1800 kg/ha (Lu et al., 2011), and η for grassland is 80.0% according to the 'Guide for the Preparation of Atmospheric Pollutant Emission Inventories for Biomass Burning' (EPD, 2014).

2.2.4 Livestock manure

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

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

where, A is the annual discharge of livestock manure burned (tonMg/yra); S represents the amount of each livestock type in pastoral and semi-pastoral land at the end of the year (head/yra); Y is a single livestock annual fecal output per year (tonMg/head); C represents livestock manure dry matter fraction; and R is the proportion of total livestock manure directly combusted.

The S values were taken from the China governmental annual statistical reports, including EOCAIY (2013) and NBSC (2013c). The Y values were related to the large animals only. Among these, single cattle annual manure output was 10 tons Mg and single horse annual manure output was 7.3 tons Mg (Li and Zhao, 2008). The livestock annual manure output of other animals was set at 8 tonsMg, according to Tian et al. (2011). The C value was set as 18% (Tian et al., 2011) and R was 20% (Li, 2007a; Liu and Shen, 2007). Since not all regions use livestock manure in biomass burning, we consider only the pastoral and semi-pastoral areas including Tibet, Inner Mongolia, Gansu, Xinjiang, Qinghai province in this study (Tian et al., 2011).

2.3 Determination of EFs

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

2.4 Spatial distribution

In order to obtain the detailed spatial distribution characteristics of biomass emission, and to provide grid based data for the air quality model simulation, the biomass burning inventory in this study ~~is~~ assigned into 1 × 1 km grid cells based on the source-specific surrogate. We applied GIS software as the main tool to produce the spatial distribution. In this paper, the approaches used to determine spatial distribution varied between biomass sources; thus, we selected different methods of spatial allocation according to the homologous source characteristics. ~~The regions in which open biomass burning occurred can be located according to the MODIS fire counts data (van der Werf et al., 2006; Liu et al., 2015). Detailed description about the MODIS fire data are shown in Supplement (S4). The outdoor biomass sources (e.g. outdoor straw burning, forest fire, and grassland fire) were treated as point sources that were located based on MODIS fire data, and land use data provided by Ran et al. (2010). Farmland fire point, forest fire point and grassland fire point are the spatial surrogates of outdoor straw burning, forest fire and grassland fire, respectively.~~ The regions in which in-field crop residue burning occurred can be located according to the MODIS fire counts data (MOD14/MYD14) (van der Werf et al., 2006; Huang et al., 2012e). Farmland fire point is the spatial surrogates of in-field crop residue burning, land use data (MODIS Land cover) is provided by Ran et al. (2010). Detailed description about the MODIS fire counts data (MOD14/MYD14) are shown in Supplement (S4). As for forest and grassland fire, the emission of forest and grassland fire are estimated in 500m resolution, it can be resampled into 1km grid using GIS software. The emissions of straw, firewood, and livestock excrement combustion were treated as area sources and the spatial surrogates used to distribute these biomass sources were population density of different land use types (e.g. rural population density, grassland population density) (Zheng et al., 2009; Huang et al., 2012c). The population density of different land use types is according to the land use data provided by Ran et al. (2010) and 1 km grid population distribution data provided by Fu et al. (2014). Detailed calculation method and equation of gridded emission are presented in Supplement (S4).

2.5 Temporal distribution

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

2.6 Speciation of VOCNMVOCs and $PM_{2.5}$

The detailed species emission of VOCNMVOCs and $PM_{2.5}$ is necessary information of model simulation for different chemical mechanism selection (e.g., CB05). The speciation of VOCNMVOCs and $PM_{2.5}$ is the main research object of the chemical composition of the atmospheric emission source, which has received extensive attention by domestic scholars in recent years (Song et al., 2007; Li et al., 2007c; Liu et al., 2008).

In this study, the species emission was mainly estimated based on the total emission, and VOCNMVOC and $PM_{2.5}$ source profiles (mass fraction) of biomass sources collected from literature review. In terms of the data selection, we prioritized domestic measurement with the species as much as possible. Therefore, the VOCNMVOCs source profile mainly refers to data from Liu et al. (2008) and Akagi et al. (2011), including 91 species covering alkane, alkene, alkyne, benzene series compoundsaromatic and so on; the $PM_{2.5}$ source profile data is cited from the work of Li et al. (2007c) and Watson et al. (2001), including 36 species, such as element, ion and so on.

3 Results and discussion

3.1 Total emissions in China

3.1.1 Contributions by biomass burning sources

The annual emissions of biomass burning in mainland China are presented in Table 7; The total annual emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, VOCNMVOC, NH_3, CO, EC, OC, CO_2, CH_4 and Hg for Chinese mainland in 2012 are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34380.4 Gg, 369.7 Gg, 1189.5 Gg, 675299.0 Gg, 2092.4 Gg and 4.12 Mg332.8 Gg, 972.5 Gg, 3676.0 Gg, 3479.4 Gg, 3429.6 Gg, 395.8 Gg, 33987.9 Gg, 367.1 Gg, 1151.7 Gg, 665989.0 Gg, 2076.5 Gg and 3.65 Mg, respectively. The contribution of different sources to the total emissions of various pollutantsto various chemical species is shown in Fig. 2. It shows that the straw indoordomestic burning, outdoor in-field crop residue burning and firewood combustion are the dominant biomass burning sources with the total contribution ranging from 95.986.02% to 99.197.58% for various species. However, the largest contributing sources to different species are not similar. Compared with other sources, straw indoor domestic burning contributed most to SO_2, CO, CH_4 and HgNMVOC, accounting for 58.557.8%, 58.758.1%, 53.653.2% and 42.249.2% of total emissions, respectively. Straw indoor domestic burning has a direct impact on residents and the prolonged exposure under high indoor domestic biomass burning emission (e.g., SO_2, CO, CH_4 and Hg) can cause many adverse health effects (e.g. acute respiratory infections and chronic bronchitis) (Emily and Martin, 2008). The contribution of firewood to each species cannot be neglected, especially for EC (51.651.3%) and NH_3

(~~41.8~~41.2%). According to the localized measurement of EF by Li et al. (2009), the average EC EF for firewood (1.49 g/kg) is 3.5 times of crop residue (0.43 g/kg). EF of firewood NH_3 is larger than the average of various straws. This results in a large contribution by firewood for these two species. The contribution of straw ~~indoor~~ domestic burning and ~~outdoor~~ in-field crop residue burning to NO_x, PM_{10}, $PM_{2.5}$, ~~VOC~~NMVOC, Hg, OC and CO_2 is nearly equal. Straw burning has an important influence on indoor air quality and ~~the~~ outdoor atmospheric environment.

In addition to the sources mentioned above, the contribution of livestock excrement burning, forest and grassland fire~~s~~ is relatively small. It is mainly due to the small amount of biomass fuel consumption. The biomass fuel consumption of these three biomass sources are 10614Gg, 6647Gg and 505 Gg, respectively, which is significantly lower than that of straw domestic combustion (201582 Gg), in-field crop residue burning (147178 Gg) and firewood combustion (127250 Gg). ~~The contribution of livestock excrement burning to PM_{10}, $PM_{2.5}$, NH_3, EC, OC, CO_2 and CH_4 is 2.55%, 2.51%, 3.49%, 1.53%, 2.03%, 1.69% and 2.12%, respectively. The contribution of forest and grass fires to biomass burning emissions to most chemical species in China is negligible (0.19–0.66%), except for the contribution of forest fire to Hg emissions (2.65%).~~ The contribution of livestock excrement burning to PM_{10}, $PM_{2.5}$, NH_3, EC, OC, CO_2 and CH_4 is 2.52%, 2.47%, 3.44%, 1.52%, 1.96%, 1.67% and 2.10%, respectively. The contribution of forest and grassland fires to biomass burning emissions to most chemical species in China is small (0.9–3.7%), except for the contribution of forest fire to Hg emissions (14.0%).

3.1.2 Contributions by various crop straw

As mentioned in Section 3.1.1, straw burning is the important biomass burning source with considerable influence on the chemical species that most strongly impact the air quality, climate change and human health. Furthermore, the major crop straw type contribution was analysed. Figure 3 shows the contributions of 12 different types of crop straw ~~indoor~~ domestic burning and ~~outdoor~~ in-field crop residue burning to total straws burning emissions for various species ~~to various chemical species~~ in 2012 from the perspective of the ~~whole country~~ mainland China. Figure 3c indicates that corn, rice and wheat straw are the major crops straw burned as fuel or as waste in China. The contribution is more than 80% to the total straw burned emissions of all pollutants studied in this paper. Corn, rice and wheat are the three major food crops in China with large planting area (the output of these three kinds of grain accounts for 70% of the total grain output in China, NBSC, 2013c), resulting in a large amount of straw production. Among the various crops, corn straw burning has ~~the greatest~~large contribution to all of the chemical species except for CH_4. Rice straw ~~is major contributor~~ has the largest contribution to CO_2, ~~VOC~~NMVOC, CH_4 and NH_3 emissions, accounting for 32.90%, 32.43%, 31.61% and 30.12%, respectively; wheat straw has a ~~a~~ considerable contribution to Hg, SO_2 and OC emissions, accounting for 29.46%, 26.47% and 25.91%, respectively. Compared with the three kinds of crop mentioned above, the total contribution of soybean, cotton, sugar cane, potato, peanut and rape to the various chemical species is relatively small, accounting for 8.1–19.2% of the total emissions for all pollutants; the contribution of sesame, sugar beet and hemp burning to various chemical species emission is negligible, never exceeding 0.5%. In addition, Fig. 3a and Fig. 3b indicate that for most of the chemical species, the contribution of ~~corn straw outdoor~~ in-field corn residue burning is larger than that of ~~indoor~~ domestic burning, except for SO_2, EC and CO_2. Contrary to that for corn straw, emissions of all chemical species (except for SO_2, NO_x and EC) from wheat straw ~~indoor~~ domestic burning is greater than those from ~~outdoor~~ in-field crop residue burning. For rice straw, the contribution of ~~outdoor~~ in-field crop residue burning to NO_x, PM_{10}, $PM_{2.5}$, ~~VOC~~NMVOC, EC and OC emissions is larger than ~~indoor~~ domestic burning.

3.2 Emissions from different regions

3.2.1 Total emissions for different provinces

The total biomass burning emissions in ~~the~~ 31 provinces in 2012 are presented in Table 7. These results indicate that Heilongjiang, Shandong, Henan, Hubei, Anhui, Sichuan, Jilin, Inner Mongolia, Hunan and Jiangsu province are the major contributors, with the total emission contributions ~~of various pollutant emissions between~~ ranging from ~~54~~53% to ~~and 66~~65% for various ~~species~~pollutants. The province with most contribution to total emission of NO_x, PM_{10}, $PM_{2.5}$, ~~VOC~~NMVOC, NH_3, OC, CH_4, Hg and CO_2 is Heilongjiang; while Shandong province has the highest emission of SO_2, CO and ~~,~~ EC ~~and Hg~~. It could be attributed to different types of biomass consumption in each province due to geographical location, climate conditions and population density. Detailed discussion about the contribution by biomass source and crop straw type of different regions is shown below.

3.2.2 Contributions by biomass sources of each province

The emission of detailed biomass sources of each province is presented in Fig. 4. The province~~s~~ with major contribution to total pollutants emissions for each biomass source are various. Straw burning emissions mainly distributed in Shandong, Henan, Heilongjiang, Hebei, Anhui, Sichuan, Jilin and Hunan province. The total contribution of these provinces to various pollutants is more than 58%. It is due to the large amount of cultivated land in the north plain region as cultivated land in this region prioritizes economic crops that produce rich straw resources. Several regions in which firewood produce a large emission are Hunan, Yunnan, Hubei, Hebei, Sichuan, Guangdong, Shaanxi, Liaoning and Jiangxi province. More than 54% firewood combustion emission is contributed by these provinces. These areas are mainly distributed in the south of China, a mountainous region in which the forest cover is higher than 30% (NBSC, 2013c). Livestock excrement combustion emissions mainly distributed in Tibet, Inner Mongolia, Gansu, Xinjiang, and Qinghai province, since only pastoral and semi−pastoral areas burn livestock manure as fuel in China. Emissions from forest and grassland fire~~s~~ are mainly distributed in Tibet, Yunnan, ~~Zhejiang~~Heilongjiang, Xinjiang, Inner Mongolia and Sichuan province. This is owing to the high vegetation cover and climatic conditions in these areas.

The contribution of biomass sources to total emissions in each province is also distinct. Straw burning has a large contribution to various pollutants emissions in Heilongjiang (79−~~95~~97%), Ningxia (~~84~~87−98%), Shandong (~~73~~74−95%), Jilin (~~73~~74−95%), Henan (~~60~~61−~~91~~93%), Anhui (~~50~~51−91%) and ~~Sichuan~~ Shanxi (~~57~~61−~~89~~90%) province. The economic income of the rural areas in these provinces is relatively low. A large number of crop residues ~~is~~ are consumed as main non−commodity energy. In addition, firewood resources ~~is~~ are scarce in these areas and as a result, the usage of straw is very high. Figure 4 also indicates that, for most provinces (e.g. Beijing, Tianjin, and Hebei), the contribution of the ~~indoor~~ domestic straw burning is greater than ~~outdoor straw~~ in-field crop residue burning. This is mainly attributable to the gradual response to the prohibition of burning straw and the introduction of straw resource utilization measures. The emission contribution of ~~straw burned~~ in~~-~~field crop residue burning~~s~~ is higher than that of straw domestic burning in Hebei, Heilongjiang and Anhui province. It suggests that the prohibition of burning straw measures in these provinces still needs to be strengthened. Several regions in which firewood produce a large component of total emissions of various pollutants are Beijing (~~56~~47−~~76~~90%), Guangdong (31−83%), Yunan (~~37~~31−~~81~~79%), Fujian (~~29~~30−~~80~~81%), Hainan (26−~~64~~77%) and Guizhou (27−74%) province. The straw amounts in the rural areas of these provinces are relative low. Firewood is the mainly non−commodity energy used by rural people. It is worth noting that though the biomass fuel consumption in Beijing is small, ~~the firewood is main biofuel due to the server restriction of straw open burning. C~~compared to straw burning emission contribution (9%−~~45~~41%), firewood emission (~~55~~47~~%~~−90%) represents a large proportion of the total biomass burning in Beijing. It is mainly due to the server restriction of in-field crop residue burning. Firewood gradually replaces straw as the main non−commodity biomass energy source in suburban Beijing in recent years (Wang, 2010; Liu, 2012). In addition, Tibet and Inner Mongolia are the major provinces where livestock excrement produces a large component of total pollutant emissions. Less crop straw and little firewood is used as a fuel source and thus fierce has a large contribution to total biomass emissions in these provinces. Forest and grassland fire~~s~~ have a small contribution to pollutant emissions in each province. The contribution of Hg emission by forest fire in Inner-Mongolia, Sichuan, Yunnan, Qinghai, Tibet and Xinjiang~~Jiangsu, Zhejiang, Fujian, Hunan, Yunan, Tibet and Qinghai~~ province is considerable (exceeding 10%), which mainly due to the high EF of Hg for forest fire.

3.2.3 Contributions from different crop straws of each province

As the largest biomass source, crop straw burning represents a major contribution to the total emissions from biomass burning. The 12 different types of straw burning emission of each province are further analysed in Fig. 5. The corn straw burning emission is concentrated in Heilongjiang, Shandong, Inner Mongolia, Hebei, Henan, Shanxi and Sichuan province, with the total contribution more than 72%. Wheat crop straw emissions mainly distributed in Henan, Shandong, Anhui, Hebei, Jiangsu, Sichuan, Shaanxi, Hubei and Shanxi province. More than 89% wheat~~-~~ crop straw combustion emission is contributed by these provinces. Rice crop straw combustion emissions mainly distributed in Heilongjiang, Hunan, Jiangsu, Sichuan, Anhui, Hubei, Guangxi, Guangdong and Zhejiang province, with the total contribution more than 71%. The water condition, light and heat are better for the cultivation of rice in the S~~s~~outh. Low temperature, long sunshine duration, and the large temperature difference between day and night are suitable for wheat growing in the N~~n~~orth. In addition, soybean, cotton, sugar cane, potato, peanut and rape straw have a small contribution to the various chemical species, and these straw are mainly distributed in Heilongjiang, Xinjiang, Guangxi, Sichuan, Henan and Sichuan province, respectively.

3.2.4 Emissions intensity at county resolution

At county resolution, we found that the spatial distributions of emissions for various chemical species are similar, taking $PM_{2.5}$ as an example to analyse the emission intensity (e.g., per unit area, per capita) at county resolution. Figure 6a shows the county-level geographic distribution of $PM_{2.5}$ emissions in 2836 counties. The number~~s~~ of counties within different emission ranges were shown in Fig. 6d. The spatial diversity of various counties emission is obvious. There are ~~403~~ 406 counties without biomass burning, because they are mainly distributed in the urban areas of developed cities, such as the Dongcheng and Xicheng districts in Beijing, the Jing'an district in Shanghai. The total emission of ~~the 33.4~~32.3% of ~~the total~~ districts and counties (~~948~~917) in China were less than 0.25 Gg. The cumulative frequency analysis result indicated that the emission in most of the counties (i.e., more than 90%) were less than ~~3.2~~4.0 Gg, including the regions with low crop yield or scarce population. The emission of ~~the 31.1~~30.9% of the total districts and counties (~~883~~875) were more than the average emission across all counties (~~1.227~~1.245 Gg). The two largest emission (approximately 16 Gg) appeared in Longjiang and Wuchang which are major grain-producing counties in Heilongjiang province.

Figure 6b shows the $PM_{2.5}$ emissions intensities per unit area. The most of high values (more than 3 Mg km^{-2} yr~~a~~$^{-1}$) mainly appeared in the north and central region of China (e.g., Hebei, Jiangsu, Shandong, Anhui, Jiangxi, Hunan), where the land is relatively flat and give priority to agricultural activity, with a substantial amount of crop straw from a relatively small area. The most counties with lower intensity concentrated in Tibet, Qinghai and Xinjiang province. In addition, it could be found that some rural counties in Heilongjiang, Jilin, and Liaoning provinces show substantial emissions, but relative lower intensity (e.g., Nenjiang in Heilongjiang, ~~,~~ Dunhua in Jilin, Chaoyang in Liaoning) due to the large area of these counties.

$PM_{2.5}$ emissions intensities per capita is illustrated in Fig. 6c. Because of the diversity of population density and biomass energy utilization, the emissions intensities per capita among various counties present~~s~~ obvious difference. The counties with emission intensity more than ~~20~~ 10 kg per^{-1} yr~~a~~$^{-1}$ are mainly distributed in Heilongjiang, Jilin, Tibet and Sichuan province. The high emission intensity in northeast China are mainly attributed to the large amount of biomass burning emissions from straw and firewood burning. The high emission intensity in southwest China mainly because these regions are less economically developed (depending on non-commercial energy as straw, firewood) and prone to forest and grassland fire burning. Besides, ~~and~~ population in there are relatively ~~low~~small. The ~~most~~ counties with lower emissions intensities per capita concentrated in Henan, Guangdong, and Shanxi provinces, attributed to the large amount of people there.~~.~~

3.3 Spatial distribution of biomass burning emissions

As chemical species showed a similar distribution, $PM_{2.5}$ wasis taken as an example to discussion the grid emission distribution. Figure 7 shows the 1 ×1 km grid distribution. It illustrates that high biomass emissions are distributed in Henan, Heilongjiang, Shandong, Anhui, Hebei and Sichuan provinces; the these areas with higher high emission are mainly scattered in major agricultural region of China's northeast to central–south China's major agricultural region, showing a zonal distribution. The biomass burning emissions are concentrated in the regions with greater agricultural and rural activity, and lower economic income. These regions characterized by dense population, abundant cultivated areas and tree resources. Low emissions are mainly distributed in the part of southwest, northwest regions and downtown areas of the majority of urban areas. The scarce populations and crop yields in part of southwest and northwest areas, and lower agricultural activity in downtown areas result in lower emissions. Specially, some urban areas in the north China Plain are surrounded by suburban and rural areas, the main fuel used in these urban areas is commodity energy. Besides, there is no agricultural activity in the field. Therefore, little biomass burning emission produced by these areas. In addition, it should be noted that grid distribution result allocated from emission at county resolution could reflect the fact that there is no grid emission within several urban regions in eastern China. However, The error will be brought in grid emissions if they are allocated from the emission inventory at coarse preliminary resolution (e.g., provincial or prefectural resolution before spatial allocation) based on the gridded surrogates (e.g., rural population). Consequently, gridded emissions, which were obtained through spatial allocation from emission inventory at county resolution, could better represent the actual situation.

3.4 Temporal variation in biomass burning emission

Figure 8 shows the 12 species emissions in each month, indicating that there are different monthly variationsnces in the chemical species emissions in different months. The chemical species showing large monthly variations were SO_2, NO_x, PM_{10}, OC, VOC and $PM_{2.5}$. This is because the main contribution of these species emission sources is from straw outdoor burning. The outdoor burning straw mainly occurs in the harvest season and thus shows the obvious monthly variation features. The chemical species showing large monthly variation were SO_2, NO_x, PM_{10}, OC, NMVOC and $PM_{2.5}$. This is because in-field crop residue burning is the main source for these species. Besides, the in-field burning of crop residue mainly in the harvest season and thus shows the obvious monthly variation features. The sources of NH_3, CO and EC emissions are dominated by straw and firewood indoor domestic burning and the contributions of these two kinds of source to the total emissions of these species are 74.0873.1%, 76.875.9%, and 87.5186.9%, respectively. The temporal distribution of these two sources was more uniform compared to in-field crop residue burning at the monthly scale, and thus monthly emissions of these three chemical species showed less temporal distinction. Despite the temporal variations of some pollutants at the monthly scale, the overall trends of emissions to mosteach species show a certain similarity: April, June and October are the top four months with peak higher emissions and the contribution of these three months to the totals for various pollutant are 8.7

~~11.2%, 8.4–12.7% and 9.0–12.8%, respectively, due to the crop straw outdoor burning of the main crop in the harvest season. While as for EC, the emission in February, January, October and December are relatively higher due to the biomass domestic burning in heating season.~~ April, May, June and October are the top four months with higher emissions, due to the in-field crop residue burning. While as for EC, the emission in February, January, October and December are relatively higher due to the biomass domestic burning in heating season.

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

For the central region (including Henan, Hubei and Hunan provinces), the main crops are winter wheat and summer corn, and the harvest season of these two crops are the end of May and the end of September (MOA, 2000), respectively.~~.~~ The peak emissions in the east region (including Shanghai, Jiangsu, Zhejiang, Anhui and Jiangxi provinces) are mainly distributed from May to July, ~~while~~ where May, June and July are the harvest seasons of rapeseed, wheat and rice in east region, respectively. The northern plains of China (including Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia and Shandong provinces), include the largest agricultural area in the country, accounting for 34% of the rural population, 27% of the farmland and 35% of the harvest crops (NBSC, 2013c). These~~is~~ region differs from the eastern and central parts firstly in the usage of firewood, since here firewood is also used as heating energy and therefore the consumption of firewood in winter is greater than in summer. In addition, for the in-field ~~burning of outdoor straw~~crop residue burning, northern winter wheat and corn are mainly harvested in June and October, respectively, and April and May are the sowing seasons of spring rice and soybeans. Northeast region (including Liaoning, Jilin and Heilongjiang provinces) shows high value in October, April and November. The high value in April was a result of burning activity. The peak in October was mainly due to the harvesting of corn and November is the harvest season for rice. In the northwest region (including Shaanxi, Gansu, Qinghai, Ningxia and Xinjiang provinces), the peaks in March-April and October are~~is~~ due to burning activities for next sowing and corn harvesting, respectively.

Furthermore, the daily $PM_{2.5}$ emissions are estimated according to the monthly emissions and the biomass sources daily non-uniformity coefficient, and are shown in the Supplement (Fig. S3). It could be found that the main emission peaks appeared~~is~~ in early April, early June and the whole month of December. This is due to (1) burning activities for the next sowing in the south, southwest, and northeast regions; (2) the harvest season of winter wheat in the central, east, and north regions; and (3) the harvest season of corn in the central, northeast, northwest regions.

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

The total $PM_{2.5}$ emission of biomass burning emission in this study is ~~3479~~3527 Gg. According to our calculation based on the method described in Sec. 2.6., OC is the largest contributor of $PM_{2.5}$ accounting for ~~33.1~~33.7% of total emission. Cl^-, EC, K^+, NH_4^+, K, and SO_4^{2-} are also the major species of $PM_{2.5}$, and the contribution of these species is ~~48.29~~46.63%. Additionally~~Besides~~, there are several species have less emission (e.g. Al, Si, Mg). Detailed $PM_{2.5}$ components emissions are presented in Supplement (Fig. S4).

~~The total VOC emission is 3429.6 Gg in this study. The alkenes are the major contributor of biomass burning VOC emissions. The contribution of alkenes to the total VOC emission is approximately 39%, more than it of alkane (25%), benzene series compounds (18.4%), alkynes (11.1%) and halocarbons (6.5%). Among the 91 species, ethylene, acetylene, propylene and 1-butylene are the major species of alkenes and alkynes, with the total contribution accounting for 29.7%. Methyl chloride, ethane, n-propane, isobutene, and n-butane are the main species of alkanes and halocarbons, with the total contribution accounting for 17.5%. Benzene, toluene, mp-xylene and styrene are the major species of benzene series compounds, with the total contribution of 12.1%. Several species mentioned above are key for the formation of secondary air pollution, such as ethylene, propylene, toluene, mp-xylene and halocarbons (Huang et al., 2011). It illustrates that the biomass burning emission control is very necessary for the air quality improvement. Detailed VOC species emission is shown in the Supplement (Fig. S5).~~

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

3.6 Uncertainties in biomass burning emission estimates

The Monte Carlo method is used to analyse the uncertainty of this emission inventory, which was used in uncertainties estimation for many inventories studies (e.g., Streets et al., 2003; Zhao et al., 2011; Zhao et al., 2012). Activity data (Zheng et al., 2009) and EFs (Zhao et al., 2011) are assumed to be normal distributions. The coefficients of variation (CV, the standard deviation divided by the mean) of activity data and emission factors were obtained from literature review. CV of activity data for firewood and crop straw burning were set as 20% (Zhao et al., 2011; Ni et al., 2015). As the data source of activity data for livestock excrement is same as the crop straw burning (i.e., government statistic data), CV is also set as 20%. MCD64A1 burned data products has been shown to be reliable in big fires (Giglio et al., 2013), and the CV of burned area of forest and grassland fire is from the reported standard deviation (Giglio et al., 2010). The biomass fuel loadings (Saatchi et al., 2011; Shi et al., 2015) and combustion factor (van der Werf et al., 2010) of forest and grassland fire were within a CV of approximately 50%. The CV of EF for each pollutant for each biomass burning type is shown in the supplement S8 and S9The Monte Carlo method is used to analyse the uncertainty of this emission inventory, which was used in uncertainties estimation for many inventories studies (e.g., Streets et al., 2003; Zhao et al., 2011; Zhao et al., 2012). Activity data are assumed to be uniform distributions (Zheng et al., 2009) and EFs are assumed to be lognormal distributions (Zhao et al., 2011). . And thenWe ran 10000 20000 Monte Carlo simulations to estimate the range of emissions with a 95% confidence interval. Uncertainty ranges of different pollutants in emission estimation are in Table 8. The uncertainty of NH_3, SO_2, EC, PM_{10}, and OC are large compared to other chemical species. The total uncertainty for emissions of these species are (−78%, 76%), (−76%, 75%), (−103%, 37%), (−37%, 80%), and (−78%, 24%), respectively. NH_3, EC, and SO_2 exist the highest uncertainties in livestock excrement combustion. The parameters used in activity data estimation of livestock excrement exist large uncertainties, such as the proportion of total livestock manure directly combusted. Besides, the localized measurements of EF for livestock manure are limited, resulting in the large uncertainty of emission. In addition to livestock excrement source, straw burning is another large uncertainty contributor to NH_3, EC, and SO_2 due to the parameter used in the estimation of straw burning amount (e.g., percentage of crop straw burned as fuel or in−field burning). The PM_{10} uncertainty is attributed in straw burning with the uncertainty ranging from −46.65% to 101.94%. It is mainly due to the limited localized measurements of EF. The uncertainty of OC is mainly attributed to firewood burning (−179.82% to 60.75%). Because the firewood consumption could not be directly obtained from yearbook after 2007, the data used in this study is estimated by statistical regression. This process may bring uncertainty to some extent. From the perspective of source, the uncertainty of forest fire (ranging from −624% to 631% for all pollutants) is the highest, following by grassland burning (ranging from−378% to 290% for all pollutants), livestock excrement (ranging from −300% to 295% for all pollutants), and firewood combustion (ranging from −189% to 188% for all pollutants). The uncertainty of crop straw (ranging from −114% to 114% for all pollutants) is the smallest. Uncertainty ranges of different pollutants in emission estimation are in Table 8. The total uncertainty of SO_2, NH_3 and EC are large compared to other chemical species. The total uncertainty for emissions of these species are (−54%, 54%), (−49%, 48%) and (−61%, 61%), respectively. NH_3, EC and SO_2 exist the highest uncertainties in livestock excrement combustion, forest and grassland fire. The emission factors used in emission estimation of livestock excrement exist large uncertainties, which is mainly due to lack of localized measurements of EF. The large uncertainty of forest and grassland fire emission due to the uncertainty of biomass fuel loadings and combustion factor used in the estimation. Though the uncertainty exists in this study, compared with the limited research of national and comprehensive emission with uncertainty analysis (Table 8), our emission inventory is relatively reliable due to the selection of localized and specific crop EFs.

3.7 Comparison with other studies

In this paper, the national biomass burning emission inventory published after 2000 has been compared with this study (Fig. 10). It could be found that the relatively high difference (range from −80% to ~~426~~366% for various species) occur between our estimation and earlier studies (e.g., published paper before 2006) due to the economic development and EF localization. Compared with recent studies, the SO_2, NO_x, $PM_{2.5}$, EC, and OC emissions of our estimation are close to those derived from Lu et al. (2011), with the difference ranging from −~~32~~34% to ~~16~~15%. While the PM_{10}, ~~VOC~~NMVOC, CH_4 and NH_3 emission in this study is lower than Lu et al. (2011). The EFs of PM_{10}, ~~VOC~~NMVOC, CH_4 and NH_3 for various crop types used in this study is generally lower than the EF without ~~disgusting~~ specific crop types in Lu et al. (2011). The SO_2, NO_x, CH_4 and CO_2 emissions in this study are close to those in Tian et al. (2011), with the difference ranging from −~~48~~49% to ~~42~~40%. The difference of CO emission is relatively high. The major emission difference of the straw ~~indoor~~ domestic burning, ~~straw outdoor~~ in-field crop residue burning, and firewood combustion between our paper and Tian's et al. (2011) research are −78%, −17%, and −122%. The reason is also the selection of EF. Our localized EF for crop and firewood is lower than EFs in Tian et al. (2011). In addition, for NH_3 emission, compared with the earlier studies, our estimation is close to that derived from recent research (Kang et al., 2016). The difference is less than 17%. For Hg emission, our estimation is lower than Huang et al. (2012d), but is close to Chen et al. (2013). The EF of Hg is classified by stems and leaves (40 ng/g and 100 ng/g for firewood; 35 ng/g and 319 ng/g for crop residues) in Huang et al. (2012d), which is higher than the localized EF classified by specific crop (mean EF is 6.08 ng/g) and firewood (7.2 ng/g).

4 Conclusions

In this study, a comprehensive biomass burning emission inventory with high spatial and temporal resolution was developed for mainland China in 2012, based on the county-level activity data, satellite data and updated source-specific EFs. The emission involves crop straw domestic combustion and in field burning, firewood and livestock excrement combustion, forest and grassland fire~~s~~. The total annual emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, ~~VOC~~NMVOC, NH_3, CO, EC, OC, CO_2, CH_4 and Hg are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34380.4 Gg, 369.7 Gg, 1189.5 Gg, 675299.0 Gg, 2092.4 Gg and 4.12 Mg~~332.8 Gg, 972.5 Gg, 3676.0 Gg, 3479.4 Gg, 3429.6 Gg, 395.8 Gg, 33987.9 Gg, 367.1 Gg, 1151.7 Gg, 665989.0 Gg, 2076.5 Gg, and 3.65 Mg~~, respectively.

The straw ~~indoor~~ domestic burning, ~~and outdoor~~ in-field crop residue burning and firewood combustion are the major biomass burning sources, while the largest contributing source to various pollutants is different. Straw ~~indoor~~ domestic burning is the major source of SO_2, CO, CH_4 and ~~Hg~~ NMVOC emission, while firewood contributes most to EC and NH_3 emission. ~~Corn, rice and wheat straw are the major crop types, with the total contribution exceeding 80% for each pollutant. Corn straw burning has the greatest contribution to EC, NO_x and SO_2 emissions; rice straw burning is dominant contributor to CO_2, VOC, CH_4 and NH_3 emissions.~~ In terms of crop straw burning, corn, rice and wheat straw are the major crop types, with the total contribution exceeding 80% for each pollutant of straw burned emissions. Corn straw burning has the greatest contribution to EC, NO_x and SO_2 emissions; rice straw burning has higher contribution to CO_2, NMVOC, CH_4 and NH_3 emissions; wheat straw burning has a considerable contribution to Hg and OC. Straw burning emissions are concentrated in agricultural provinces. Firewood burning emissions are mainly distributed in southern regions of China, where the tree resource is abundant. The corn and wheat straw burning emission are mainly distributed in the northern China, while the rice straw burning emission is concentrated in the southern China. Gridded emissions result show that high emission is concentrated in northeast and central–south region of China with more agricultural and rural activity. It also illustrates that gridded emissions, which were obtained through spatial allocation from emission inventory at county resolution instead of province or prefecture resolution, could better reflect the actual situation. Monthly distributions reveal the higher emissions in April, ~~September~~ May, June and October due to the burning activity before sowing and harvesting of main crops. Regional differences of temporal distribution are attributed in the diversity of main planted crop and the climate conditions in each region. OC, Cl^-, EC, K^+, NH_4^+, K, and SO_4^{2-} are the major $PM_{2.5}$ species, with the total contribution of 80%. Several species with higher contribution to ~~VOC~~NMVOCs (e.g., ethylene, propylene, toluene, mp-xylene and ethyl benzene~~halocarbons~~) are key species for the formation of secondary air pollution. The comparison with other studies presents that the emission inventory in this study is relatively reliable. The detailed emission inventory given by this paper could provide detailed information to support the further biomass burning pollution research and the development of a targeted control strategy of all regions across the Chinese mainland.

EF and speciation of chemical species are the key parameter in the emission estimation. More localized EF of different biomass fuel types ~~indoor and outdoor burning~~ within diverse burning conditions, more detained $PM_{2.5}$ and ~~VOC~~NMVOC source profiles that contain as much components as possible still needs to expand in the future. In addition, the higher temporal resolution (e.g. hourly resolution) satellite data are necessary to provide hourly emission information for the numerical simulation of biomass burning pollution research and effective control.

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Table Captions List:

Table 1. The classification of biomass burning emission source.

Table 2. ~~Indoor~~ Straw domestic and ~~outdoor~~ in-field ~~straw~~ crop residue burning proportions of each province.

Table 3. Residue-to-production ratio (N_k), dry matter fraction (D_k) and combustion efficiency (CE_k) used in this study.

Table 4. Forest and grassland biomass fuel loadings in each province.~~Forest biomass and combustion efficiency of each province.~~

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

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

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

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

Figure Captions List:

Figure1. Regression analysis between firewood consumption at province resolution and (1) rural population, (2) gross agricultural output, and (3) timber yield, respectively.

Figure 2. Contribution of different source to the total biomass burning emissions in China, 2012.

Figure 3. Contributions of 12 crop straw types to total straws burning emissions for various species.

~~Contributions of 12 crop straws burning to total pollutant emissions for various species.~~

Figure 4. Contributions of different biomass sources to the emission in each province (Gg).

Figure 5. Contributions of different crop straw types to the emission in each province (Gg).

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

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

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

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

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

Table 1. The classification of biomass burning emission source.

I	II	III
Domestic combustion	Firewood	Firewood
	Livestock excrement	Cattles
		Horses
		Donkeys
		Mules
		Camels
	Crop straw	Corn, wheat, cotton, sugar cane, potato, peanut, rapeseed, sesame, sugar beet, hemp, rice, soybean
Open burning	In-field crop residue	Corn, wheat, cotton, sugar cane, potato, peanut, rapeseed, sesame, sugar beet, hemp, rice, soybean
	Forest	Evergreen Needleleaf Forest
		Evergreen Broadleaf Forest
		Deciduous Needleleaf Forest
		Deciduous Broadleaf Forest
		Mixed Forest
		Closed Shrublands
		Open Shrublands
		Evergreen Needleleaf Forest
	Grassland	Woody Savannas
		Savannas
		Grasslands

~~I~~	~~II~~	~~III~~
~~Domestic combustion~~	~~Firewood~~	~~Firewood~~
	~~Livestock excrement~~	~~Cattles~~
		~~Horses~~

		Donkeys Mules Camels
	Crop straw	Corn, wheat, cotton, sugar cane, potato, peanut, rapeseed, sesame, sugar beet, hemp, rice, soybean
In-field burning	Crop straw	Corn, wheat, cotton, sugar cane, potato, peanut, rapeseed, sesame, sugar beet, hemp, rice, soybean
	Grassland fire	Grassland fire
	Forest fire	Tropics South subtropical zone Central subtropical zone North subtropical zone Warm temperate zone Temperate zone Cold temperate zone Tibetan region

Table 2. Indoor Straw domestic and outdoor in-field straw crop residue burning proportions of each province.

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

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

* The result from our questionnaire.

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

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

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

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

5

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

Province	Climatic zone	Forest biomass*	Combustion efficiency*
Hainan	Tropics	348	0.2
Guangdong	South subtropical zone	178	0.2
Guangxi		178	0.2
Yunnan	South subtropical zone, Central subtropical zone	138	0.2
Fujian	Central subtropical zone	143	0.2
Jiangxi		143	0.2
Chongqing		143	0.2
Zhejiang	Central subtropical zone, North subtropical zone	121	0.2
Guizhou		121	0.2
Sichuan	Central subtropical zone, Tibetan region	118	0.2
Shanghai	North subtropical zone	98	0.2
Anhui		98	0.2
Hubei		98	0.2
Hunan		98	0.2
Jiangsu	North subtropical zone, Warm temperate zone	77	0.2
Beijing	Warm temperate zone	55	0.1
Tianjin		55	0.1
Hebei		55	0.1
Shanxi		55	0.1
Shandong		55	0.1
Henan		55	0.1
Shaanxi		55	0.1
Xinjiang	Warm temperate zone, Temperate zone	106	0.1
Inner Mongolia	Temperate zone	157	0.1
Liaoning		157	0.1
Jilin		157	0.1
Heilongjiang		157	0.1
Gansu		157	0.1
Ningxia		157	0.1
Tibet	Tibetan region	121	0.2

Province	Biomass fuel loadings (g/m^2)				
	Needleleaf Forest[a,d]	Broadleaf Forest[a,e]	Mixed Forest[a,f]	Shrublands[b,g]	Grassland[c,h]
Heilongjiang	8140	7610	7875	1387	180
Jilin	9340	10710	10025	1387	140
Liaoning	2620	8250	5435	1387	160
Inner Mongolia	8140	4470	6305	1387	90
Gansu	8900	6630	7765	1500	90
Ningxia	6910	6280	6595	1386	50
Qinghai	8800	5430	7115	1545	110

Shaanxi	3730	7550	5640	1442	100
Xinjiang	14410	3060	8735	1387	70
Tibet	13990	6490	10240	2007	60
Beijing	1560	6750	4155	1387	170
Hebei	2480	5150	3815	1388	150
Henan	1550	5560	3555	1388	140
Shandong	1280	5660	3470	1387	130
Shanxi	3640	4790	4215	1387	130
Tianjin	0	6760	3380	1378	160
Anhui	1690	10360	6025	2447	140
Hubei	1680	8060	4870	1573	160
Hunan	2080	10650	6365	3471	150
Jiangxi	1820	9370	5595	3699	140
Fujian	2910	9700	6305	3773	160
Guangdong	2060	8970	5515	3702	140
Hainan	4810	9220	7015	3739	150
Jiangsu	2630	5530	4080	1371	120
Shanghai	3060	9250	6155	1371	110
Zhejiang	1710	10500	6105	3682	160
Chongqing	8090	9900	8995	3010	170
Guangxi	2110	9280	5695	3142	150
Guizhou	2210	11410	6810	3431	150
Sichuan	8090	9900	8995	3006	170
Yunnan	5760	14510	10135	3534	150

~~* Tian et al. (2003).~~ [a] Fang et al.(1996,1998). [b] Hu et al. (2006). [c] Pu et al. (2004). And all the biomass here calculated using the aboveground biomass density.

[d] Needleleaf forest including needleleaf deciduous forest and needleleaf evergreen forest.[e] Broadleaved forest including broadleaved deciduous forest and broadleaved evergreen forest. [f] The biomass of mixed forest is the mean of needleleaf forest and broadleaved forest. [g] Shrublands including Closed Shrublands and Open Shrublands. [h] Grassland including Woody Savannas, Savannas and Grasslands.

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

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

Note: Lowercase letters indicate the data source.

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

* The unit of emission factor.

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

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

Note: Lowercase letters indicate the data source.

Sources are from the following: [a] Li et al. (2015). [b] Li et al. (2007c). [c] EPD (2014). [d] Zhang et al. (2013b). [e] Tian et al. (2011). [f] Wang and Zhang (2008). [g] Tang et al. (2014). [h] Ni et al. (2015). [i] Streets et al. (2003). [j] Andreae and Merlet (2001). [k] Chang ~~et al.~~ and Song (2010). [l] Christian et al. (2003). [m] Kanabkaew and Nguyen (2011). [n] Chen et al. (2013). [o] Zhang et al. (2013a). [p] Andreae and Rosenfeld (2008). [r] Akagi et al. (2011). [s] Song et al. (2009). [t] McMeekin et al. (2008). [u] Friedli et al. (2003). [v] Streets et al. (2005).

*The unit of emission factor.

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

Province	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
	unit:Gg											unti:Mg
Beijing	0.5	1.9	6.9	6.5	4.8	1.2	58	1.3	1.9	1507	3.1	0.01
Tianjin	1.2	3.1	11.6	10.9	10.4	1.3	116	1.4	3.7	2136	6.6	0.01
Hebei	21.4	52.8	200.2	188.6	178.7	21.4	2023	22.7	65.9	36308	115.4	0.22
Shanxi	9.4	22.9	83.5	78.8	74.9	8.1	777	8.7	27.5	14668	43.9	0.09
Inner-Mongolia	16.1	45.2	217.5	204.9	154.5	24.1	1309	16.8	65.3	32278	103.6	0.16
Liaoning	13.8	40.7	144.3	136.1	128.2	17.4	1277	18.0	42.5	27369	72.4	0.14
Jilin	16.5	54.3	179.6	171.5	165.6	15.7	1395	14.7	58.3	29529	84.7	0.16
Heilongjiang	30.0	117.5	397.4	383.3	395.4	32.5	2878	22.8	132.1	65619	200.7	0.36
Shanghai	0.3	0.8	3.1	3.0	3.6	0.2	33	0.2	1.1	566	2.1	0.00
Jiangsu	14.7	39.7	154.5	146.8	167.0	12.4	1614	10.2	52.6	27527	102.2	0.16
Zhejiang	3.8	12.4	48.2	45.7	48.2	6.1	451	5.5	13.6	9986	28.6	0.05
Anhui	19.7	56.5	210.1	199.9	209.9	19.7	2046	17.6	71.4	38539	127.7	0.23
Fujian	3.0	10.7	40.6	38.1	36.4	6.3	387	6.4	10.5	8905	22.9	0.04
Jiangxi	8.0	27.4	105.0	99.0	102.7	14.3	998	13.4	28.6	22445	62.0	0.10
Shandong	34.7	77.9	304.3	287.4	296.6	25.2	3318	24.9	108.9	50493	192.2	0.33
Henan	33.1	82.1	313.0	296.5	301.3	26.6	3294	26.0	112.7	52896	194.5	0.35
Hubei	13.2	40.7	158.2	149.0	147.6	19.9	1530	19.1	44.5	31167	91.5	0.16
Hunan	15.6	51.6	199.2	187.4	198.0	24.1	1949	22.8	54.0	39478	118.5	0.19
Guangdong	5.9	21.1	78.9	74.2	70.0	12.8	726	12.5	21.0	17619	43.4	0.08
Guangxi	8.1	29.1	105.6	100.0	108.0	15.2	1001	12.6	31.8	21300	61.5	0.11
Hainan	1.4	5.0	19.1	17.9	17.7	3.0	191	2.9	5.1	4032	11.0	0.02
Chongqing	5.5	15.6	61.3	57.4	58.7	7.4	619	7.3	17.0	11564	35.6	0.06
Sichuan	19.3	53.0	212.1	199.6	206.4	22.1	2115	21.0	63.2	38192	125.1	0.23
Guizhou	6.4	19.5	74.7	70.1	62.9	10.4	679	10.8	19.8	14944	38.9	0.08
Yunnan	8.8	27.6	108.7	101.4	90.7	17.2	972	17.4	31.8	22370	54.0	0.20
Tibet	3.0	15.4	40.6	37.6	24.8	5.4	305	2.4	26.5	7554	14.8	0.30
Shaanxi	8.5	22.2	86.5	81.1	71.6	11.1	832	12.0	25.9	16701	47.0	0.10
Gansu	6.2	15.9	66.6	62.5	52.6	8.1	579	7.8	20.1	11814	35.2	0.07
Qinghai	0.9	2.3	10.5	9.8	7.2	1.2	84	0.9	3.6	1683	5.2	0.02
Ningxia	1.6	4.1	14.9	14.1	14.8	1.2	144	1.2	5.1	2515	8.5	0.02
Xinjiang	6.2	21.8	71.5	67.5	64.9	9.8	682	8.5	23.7	13596	39.5	0.08

Total	336.8	990.7	3728.3	3526.7	3474.2	401.2	34380	369.7	1189.5	675299	2092.4	4.12

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

Species	Emission estimate	Uncertainty ranges *	Previous study
			Street et al., 2003
SO_2	337	(−54%, 54%)	(−245%, 245%)
NO_x	991	(−37%, 37%)	(−220%, 220%)
PM_{10}	3728	(−7%, 6%)	
$PM_{2.5}$	3527	(−13%, 1%)	
NMVOC	3474	(−9%, 9%)	(−210%, 210%)
NH_3	401	(−49%, 48%)	(−240%, 240%)
CO	34380	(−4%, 4%)	(−250%, 250%)
EC	370	(−61%, 61%)	(−430%, 430%)
OC	1190	(−20%, 19%)	(−420%, 420%)
CO_2	675299	(−3%, 3%)	
CH_4	2092	(−9%, 9%)	(−195%, 195%)
Hg	0.00412	(−31%, 32%)	

* 95% confidence interval.

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

Species	Emission estimate	Uncertainty ranges *	Previous study
			Streets et al. （2003）
SO_2	333	(−76%, 75%)	(−245%, 245%)
Nox	972	(−62%, 24%)	(−220%, 220%)
PM_{10}	3676	(−37%, 80%)	
$PM_{2.5}$	3479	(−54%, 34%)	
VOC	3430	(−55%, 21%)	(−210%, 210%)
NH_3	396	(−78%, 76%)	(−240%, 240%)
CO	33988	(−41%, 47%)	(−250%, 250%)
EC	367	(−103%, 37%)	(−430%, 430%)
OC	1152	(−78%, 24%)	(−420%, 420%)
CO_2	665989	(−38%, 43%)	
CH_4	2076	(−55%, 12%)	(−195%, 195%)
Hg	0.00365	(−26%, 51%)	

* 95% cConfidence iInterval.

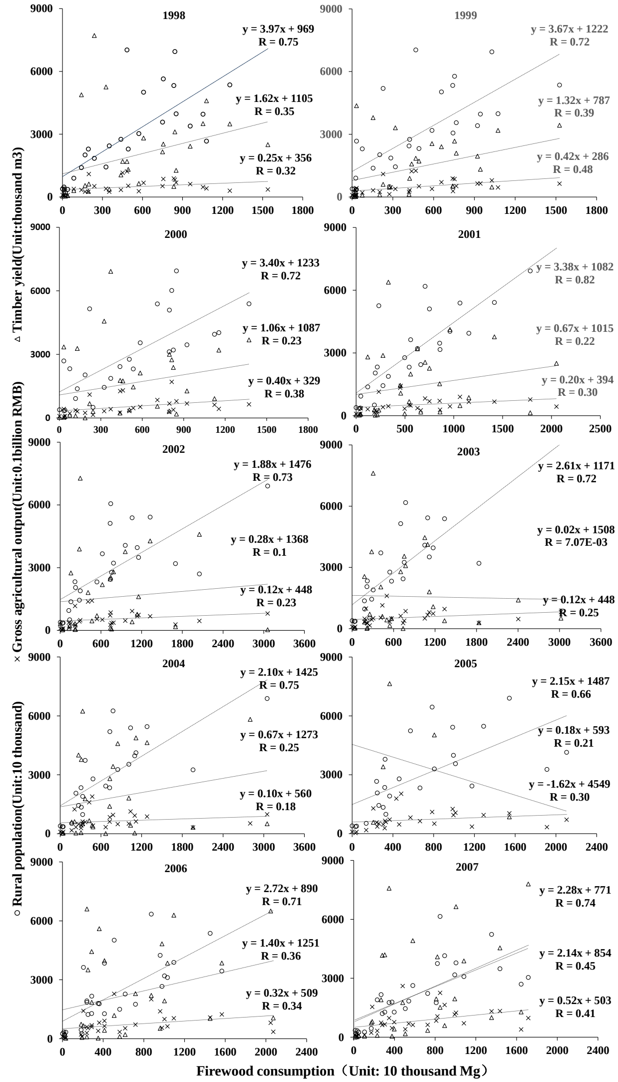
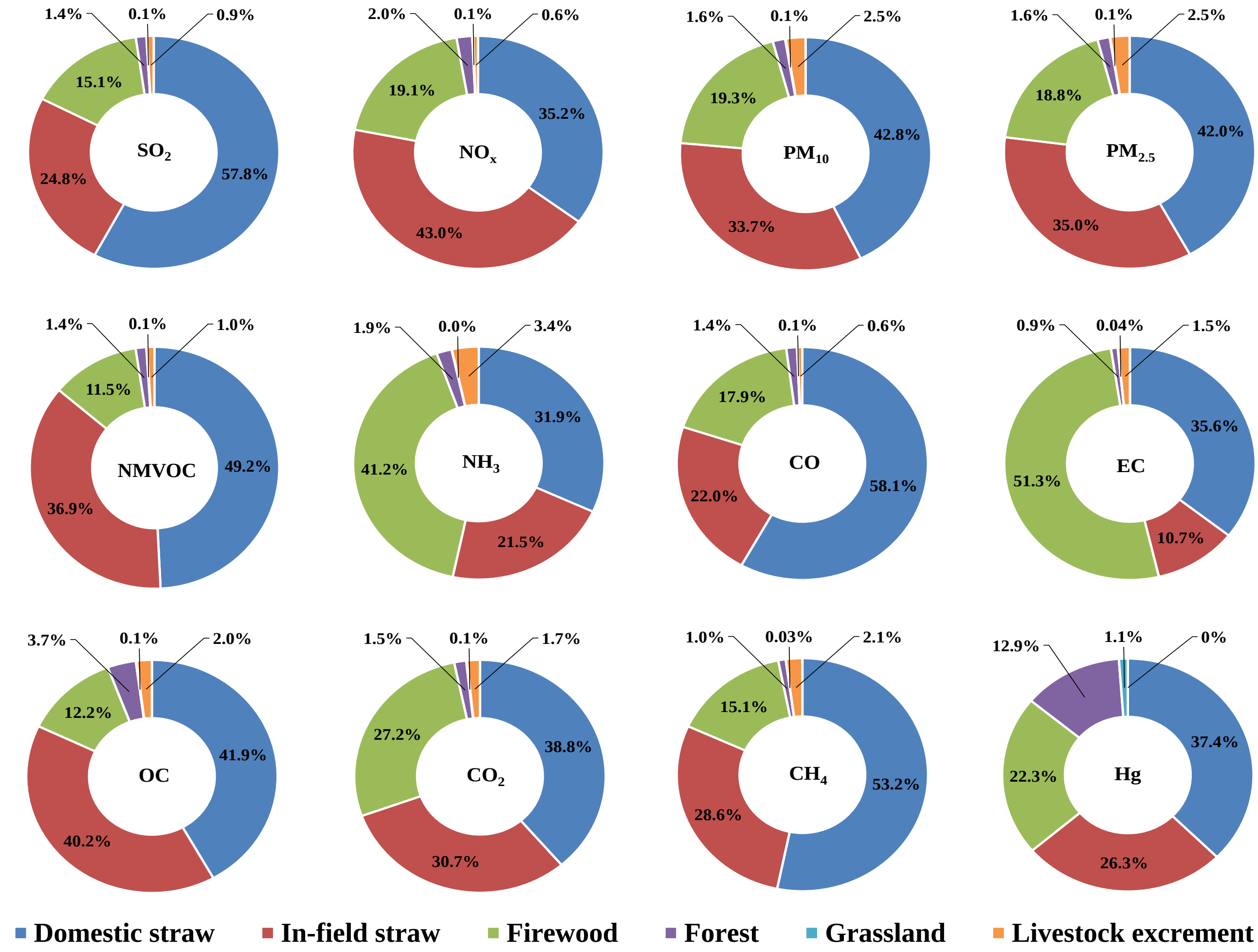


Figure 1. Regression analysis between firewood consumption at province resolution and (1) rural population, (2) gross agricultural output, and (3) timber yield, respectively.

Note: It is referred by circles, crosses and triangles, respectively. The regression equation of each figure is provided in the top, middle and bottom, respectively.



5 **Figure 2. Contribution of different source to the total biomass burning emissions in China, 2012.**

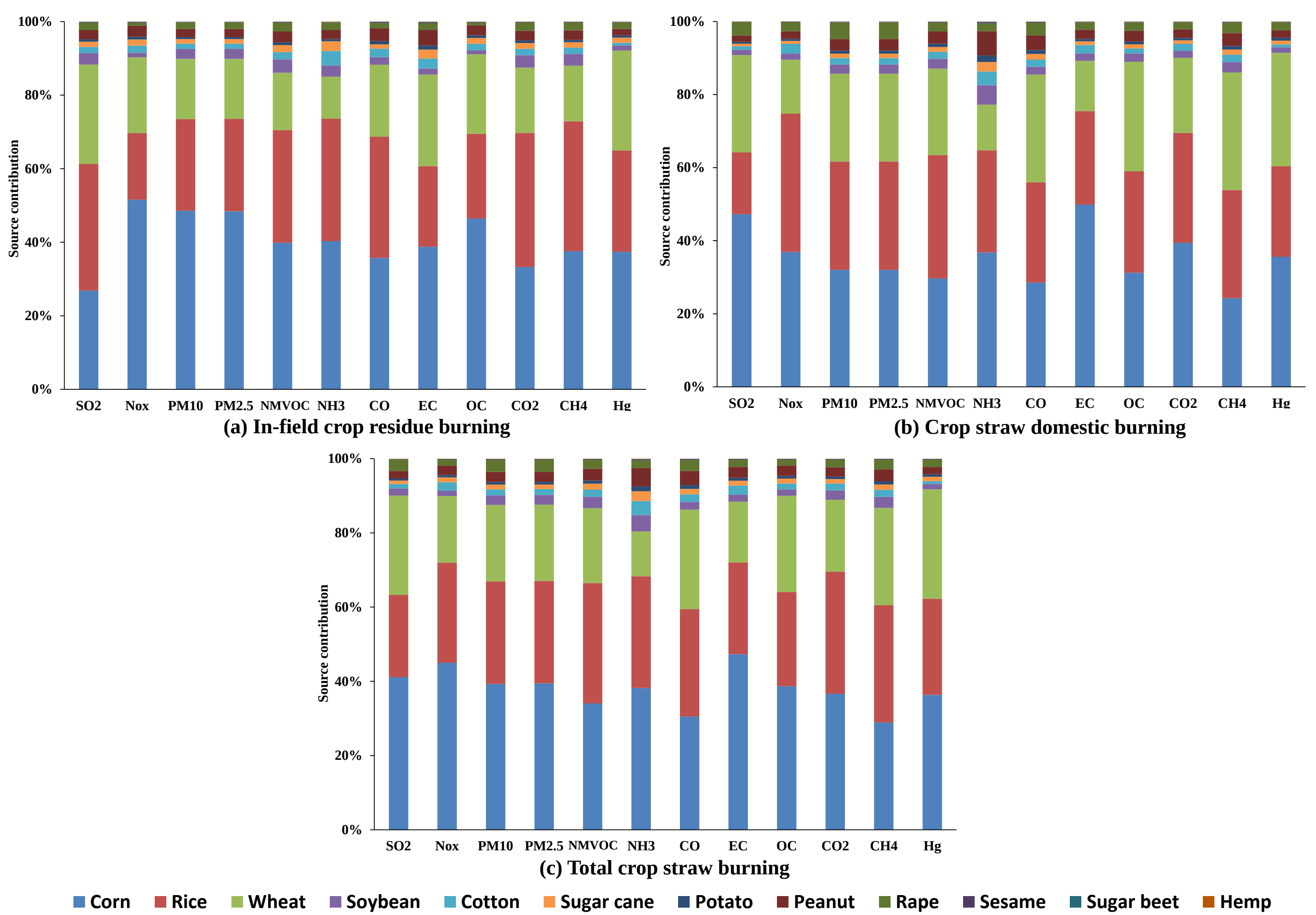


Figure 3. Contributions of 12 crop straw types~~s burning~~ to total ~~pollutant~~ straws burning emissions for various species.

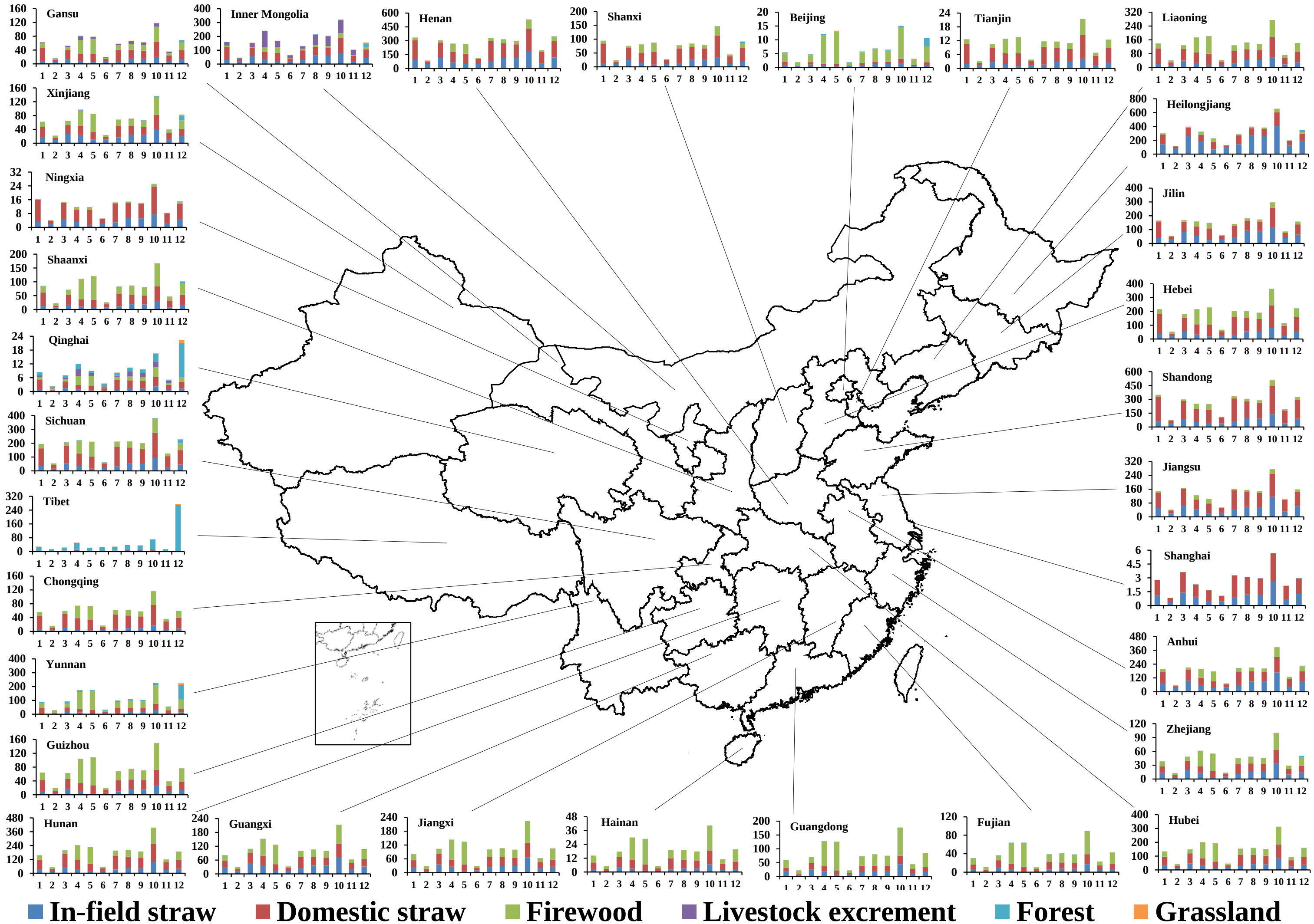


Figure 4. Contributions of different biomass sources to the emission in each province (Gg).

Note: The numbers 1–12 represent the species SO_2×10, NO_x, ~~VOC~~NMVOC, NH_3×10, EC×10, OC, CO/10, PM_{10}, $PM_{2.5}$, CO_2/100, CH_4 and Hg×1000000, respectively.

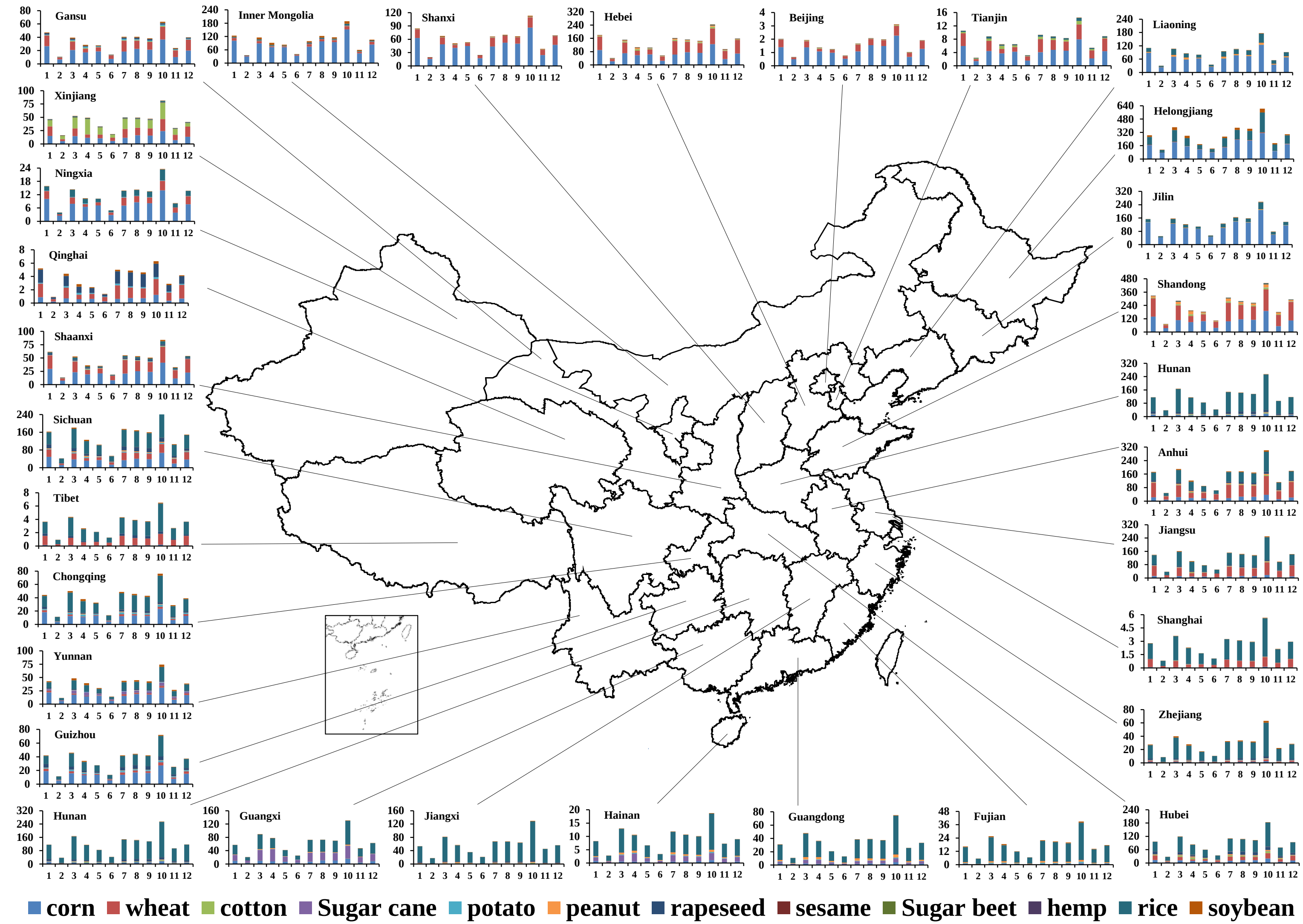


Figure 5. Contributions of different crop straw types to the emission in each province (Gg).

Note: The numbers 1–12 represent the species SO_2×10, NO_x, ~~VOC~~NMVOC, NH_3×10, EC×10, OC, CO/10, PM_{10}, $PM_{2.5}$, CO_2/100, CH_4 and Hg×1000000, respectively.

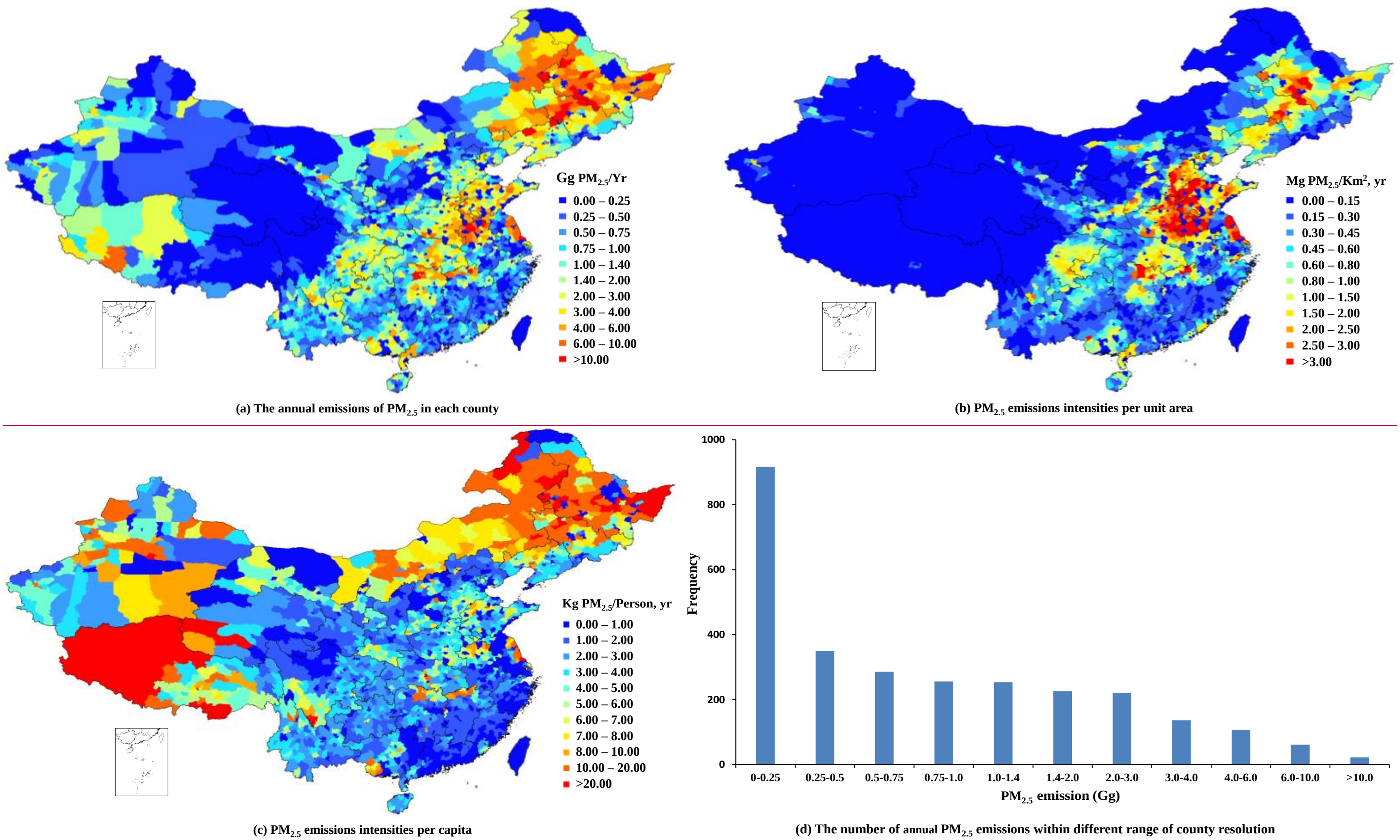


Figure 6. Biomass emission inventory at county resolution and intensity ($PM_{2.5}$).

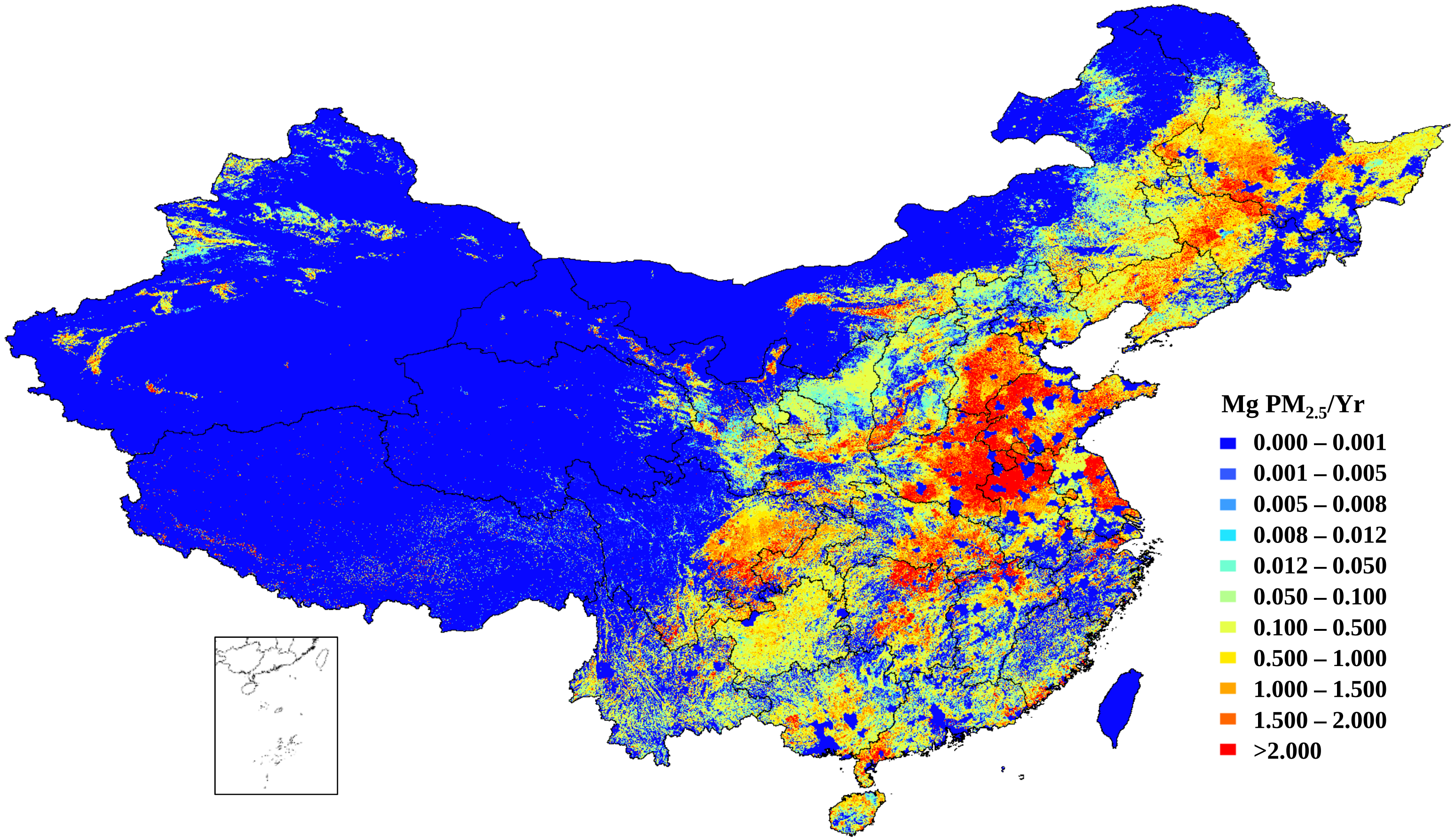


Figure 7. Gridded distribution of $PM_{2.5}$ annual emissions.

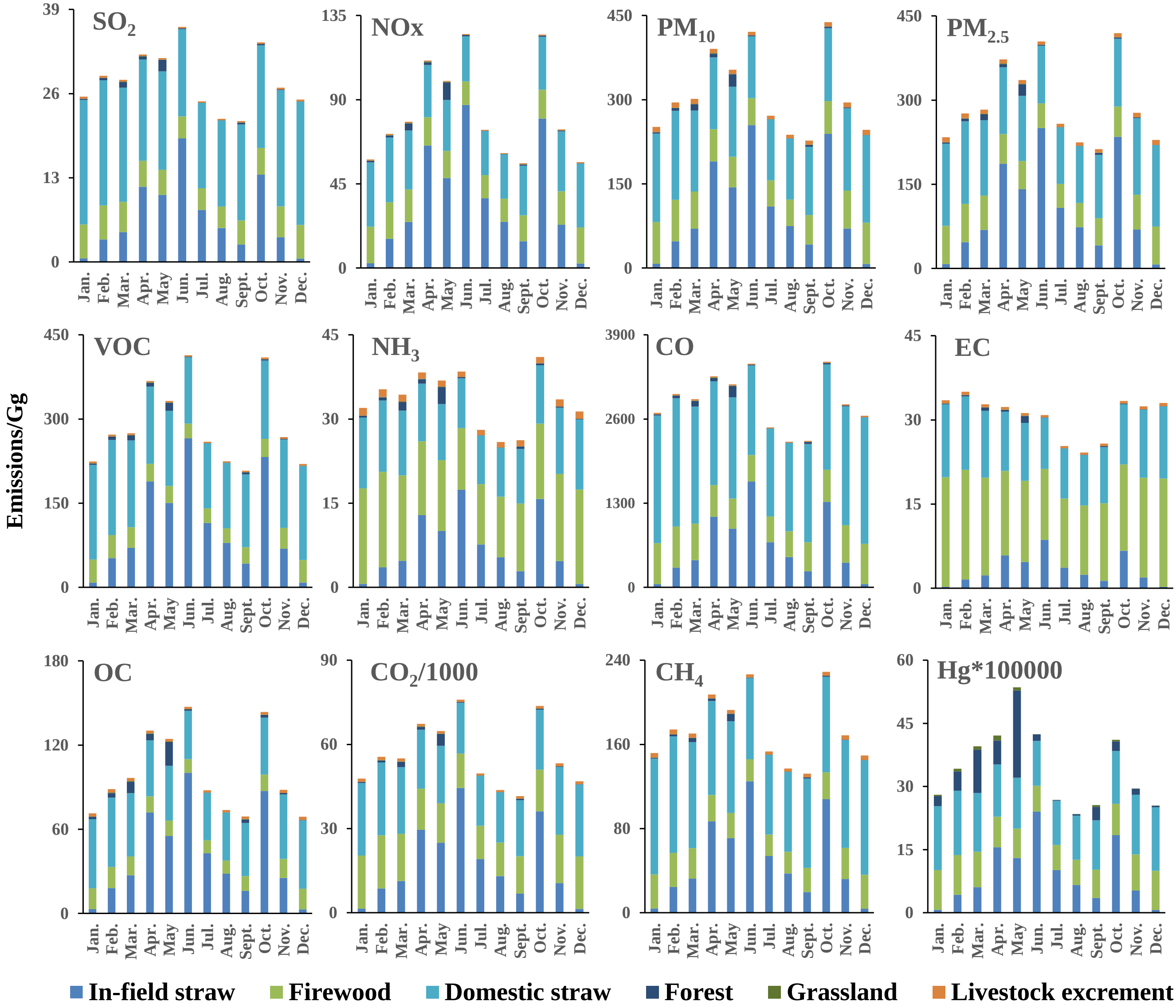


Figure 8. Monthly variation of different biomass sources emission for each chemical species.

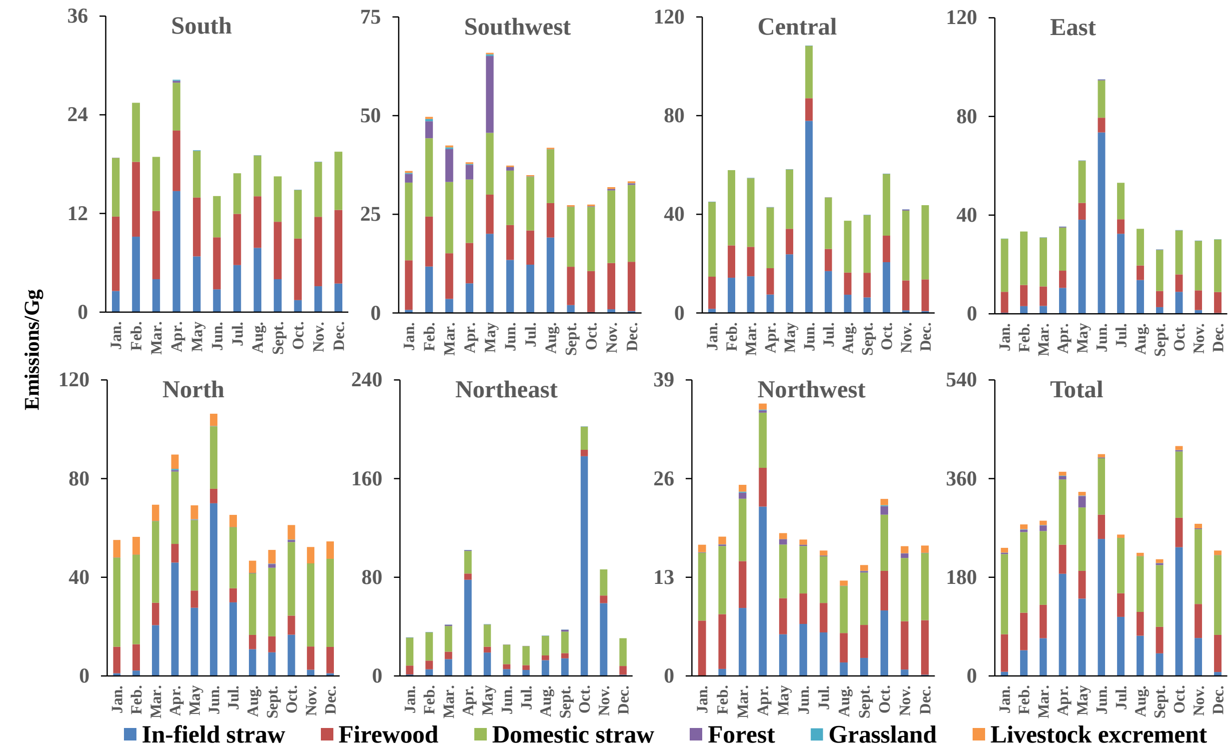


Figure 9. Monthly variation of different biomass sources emission for $PM_{2.5}$ emissions in different regions.

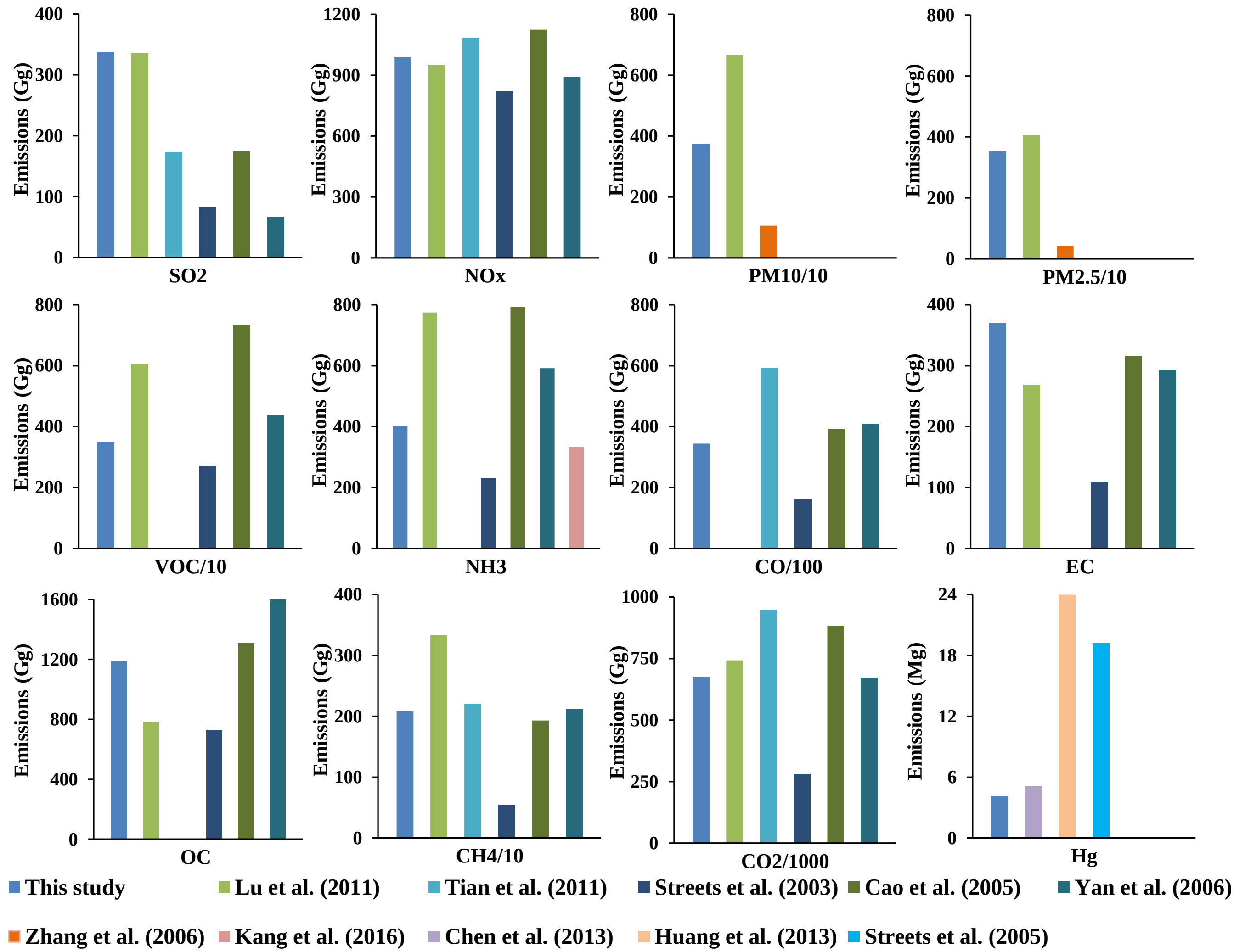


Figure 10. Comparison of the emissions inventory derived by this study with the emissions estimated by previous research.

Supplement of

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

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Supporting Information

Section S1: Figure S1 The correlation between crop yield and grain yield at prefecture resolution.

Section S2: Figure S2 Map showing the prefecture and county resolution.

Section S3: The details about questionnaire field survey.

Section S4: The detailed description about the MODIS fire data and calculation method and equation of gridded emission

Section S5: Figure S3 Daily $PM_{2.5}$ biomass burning emissions variation in 2012.

Section S6: Figure S4 Emission of $PM_{2.5}$ species from biomass burning.

Section S7: Figure S5 Emission of NMVOCs species from biomass burning.

Section S8: Table S1 CV (coefficients of variation) of biomass domestic burning emission factors.

Section S9: Table S2 CV (coefficients of variation) of biomass open burning emission factors

S1 The correlation between crop yield and grain yield at prefecture resolution.

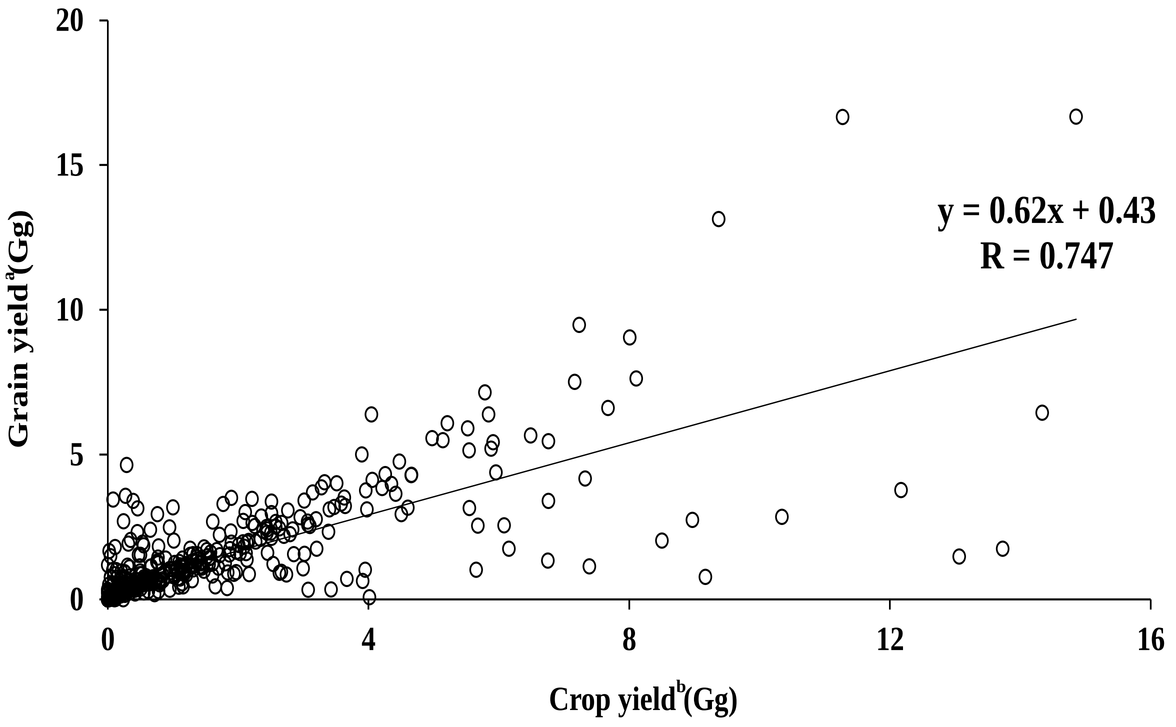
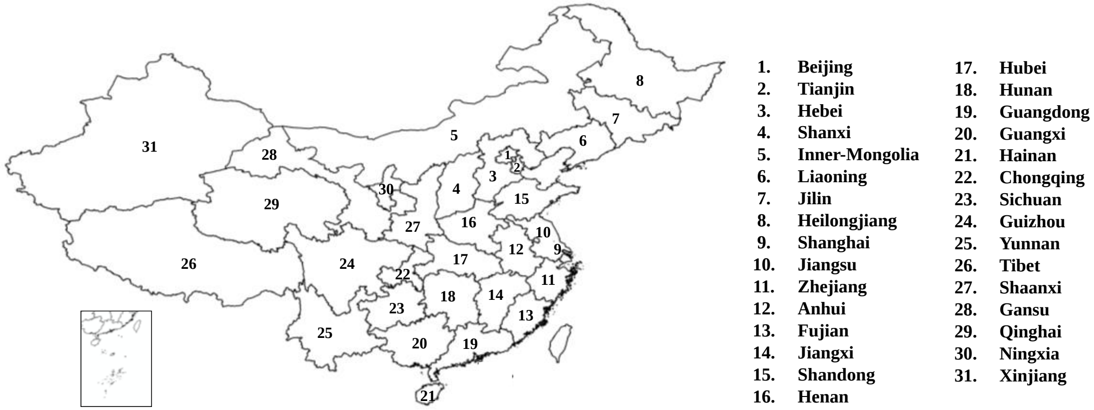


Figure S1 The correlation between crop yield and grain yield at prefecture resolution.

Note: [a] NBSC (2013b); [b] a range of statistical yearbooks edited by National Bureau of Statistics in 2012 for each province.

S2 Map showing the prefecture and county resolution.



(a) Province level

(b) Prefecture level

(c) County level

Figure S2 Map showing the prefecture and county resolution.

S3 The details about questionnaire field survey.

A questionnaire was designed to conduct field investigation during face-to-face interviews with rural resident, in order to obtain the percentage of crop straw indoor burning and outdoor burning and uneven temporal distribution coefficient in several provinces with limited literature reports, including Tianjin, Hebei, Inner Mongolia, Heilongjiang, Shanghai, Zhejiang, Anhui, Jiangxi and Guangdong provinces. Respondents need to provide the detailed address, main cultivated crop type. They selected from a list of cooking and heating fuels, including specific crop straw, firewood, coal, gas, electricity or solar, livestock excrement and other detailed fuels not existing in the list. They also need to provide approximate proportion of crop straw domestic combustion and in field burning, and selected the month of burning the straw as waste, and heating period. The investigation was launch in the representative regions in each province mentioned above, with the integrative consideration about the geographical location, economic development level and population intensity. All the surveyors were trained and tested in their understanding of the questionnaire content. Ultimately, we received 2478 valid questionnaire responses, and at least 200 valid questionnaires in each province.

S4 The detailed description about the MODIS fire data and calculation method and equation of gridded emission

4.1 Detailed description about the MODIS fire data

For the spatiotemporal distributions of biomass open burning, satellite remote sensing has excellent characteristics of wide coverage, high resolution and strong temporal reliability. As a result, satellite remote sensing has been increasingly applied to solving temporal and spatial emission distributions in recent years. The MODIS satellite fire data were taken from FIRM (Fire Information for Resource Management System). The MODIS Thermal Anomalies/Fire 5-Min L2 Swath Product (MOD14/MYD14) within 1km resolution was used in this study. The MOD14 were provided by the Terra satellite with overpass times at 10:30 AM and 10:30 PM local time, while MYD14 were provided by Aqua at 1:30 AM and 1:30 PM local time.

4.2 Detailed calculation method and equation of gridded emission

The mass of biomass emission in each grid of biomass open burning and indoor burning was calculated using Eqs. (1) and (2), respectively, as follows:

$$E_{m-outdoor} = \frac{FC_m}{FC_n} \times E_{n-outdoor} \quad (1)$$

$$E_{m-indoor} = \frac{PO_m}{PO_n} \times E_{n-indoor} \quad (2)$$

where *m* is the *m*-th grid and *n* represents the *n*-th county; $E_{m\text{-}outdoor}$ and $E_{n\text{-}outdoor}$ represent the emissions of the *m*-th grid and *n*-th county for biomass outdoor burning (in-field crop residue burning), respectively; $E_{m\text{-}indoor}$ and $E_{n\text{-}indoor}$ represent the emissions of the *m*-th grid and *n*-th county for biomass indoor burning, respectively; FC_m represents the number of typical fire points of the *m*-th grid; FC_n is the number of total typical fire points of the *n*-th county; PO_m is the number of typical population of the *m*-th grid; finally, PO_n is the number of typical population of the *n*-th county.

S5 Daily $PM_{2.5}$ biomass burning emissions variation in 2012.

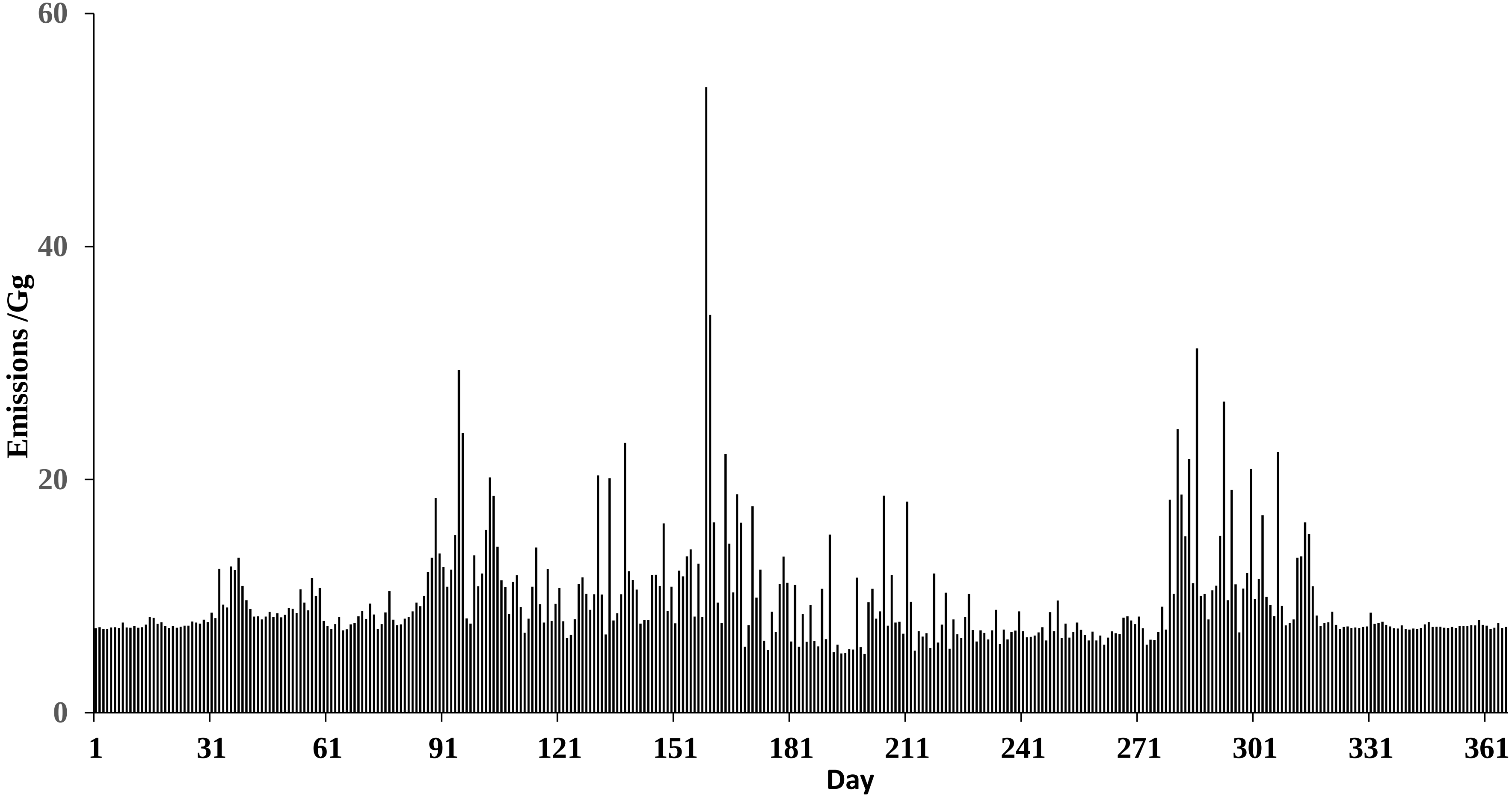


Figure S3 Daily $PM_{2.5}$ biomass burning emissions variation in 2012.

S6 Emission of PM2.5 species from biomass burning

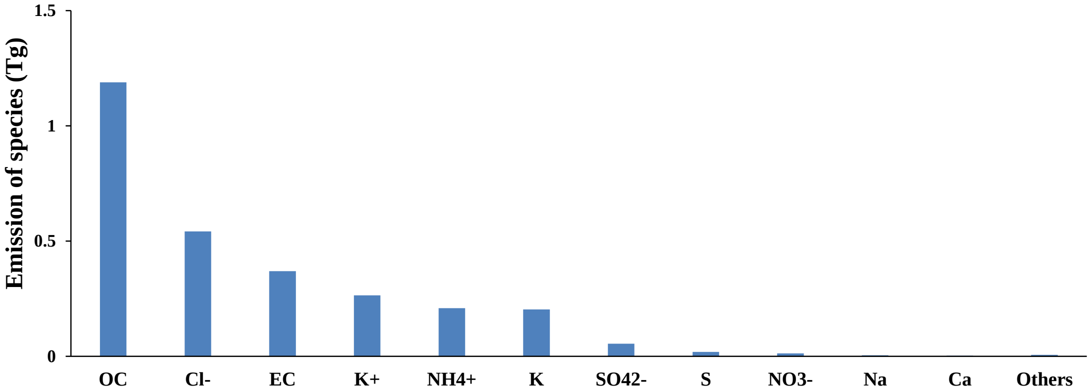


Figure S4 Emission of $PM_{2.5}$ species from biomass burning.

Note: Species in others include Al，Si，Mg，Fe，Pb，Zn，Ba，Ti，Ni，Cr，Mn，Sr，V，Cd，As，Zr，Se，Ag，Sb，Sc，Mo，Ga，Tl，Co and Hg. $PM_{2.5}$ speciation profile is obtained from Li et al., (2007) and Waston et al., (2001).

S7 Emission of NMVOCs species from biomass burning.

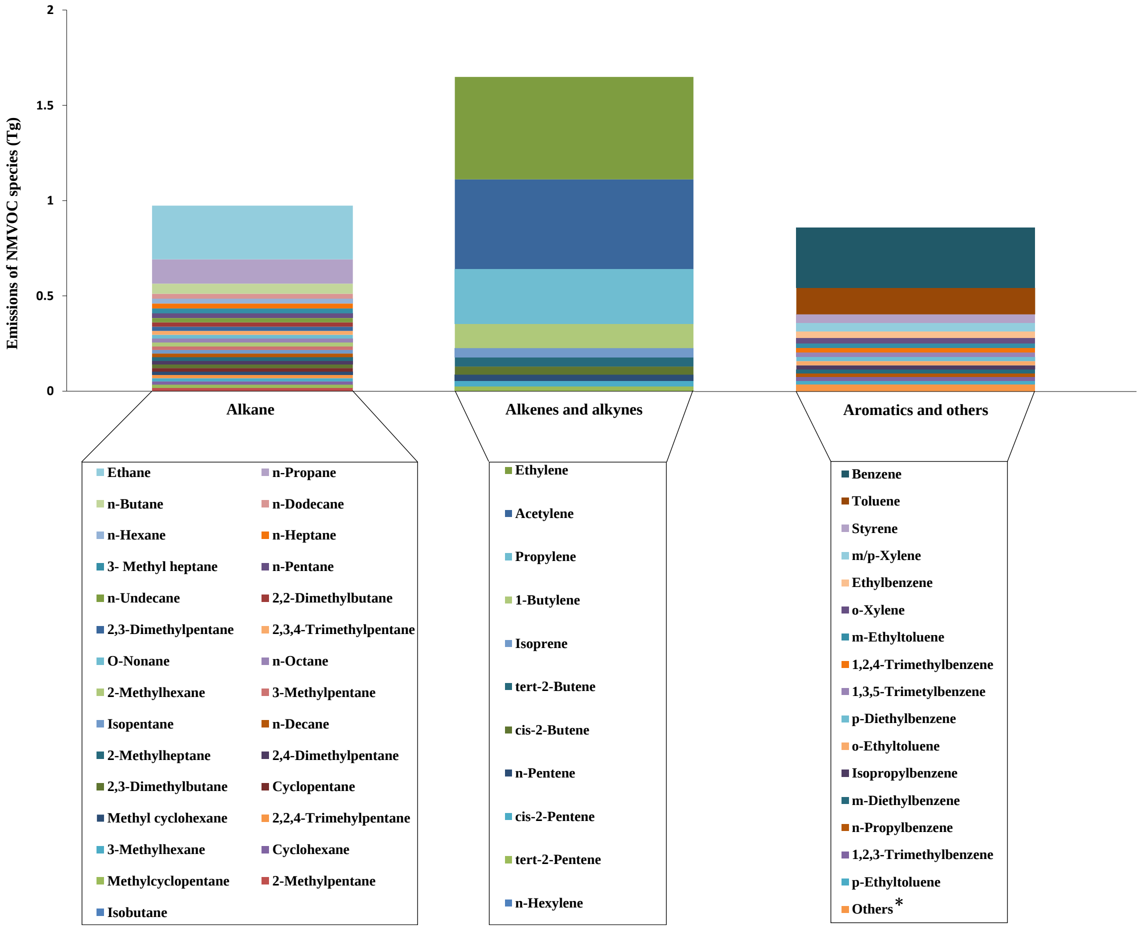


Figure S5 Emission of NMVOC species from biomass burning.

Note: *Species in others include aldehyde, ethers, alcohols, esters, ketone and acids.~~**Figure S5 Emission of VOCs species from biomass burning.**~~

S8 CV (coefficients of variation) of biomass domestic burning emission factors.

	Material	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
Domestic burning	Corn	0.5*	0.02[a]	0.5*	0.27[b]	0.5*	0.5*	0.85[a]	0.34[b]	0.44[b]	0.04[a]	0.5*	0.05[c]
	Wheat	0.5*	0.16[a]	0.5*	0.23[b]	0.5*	0.5*	0.89[a]	0.76[b]	0.29[b]	0.07[a]	0.5*	0.12[c]
	Cotton	0.5*	0.5*	0.5*	0.26[b]	0.5*	0.5*	0.5*	0.39[b]	0.55[b]	0.5*	0.5*	0.33[c]
	Cane	0.5*	0.5*	0.5*	0.26[b]	0.5*	0.5*	0.5*	0.63[b]	0.45[b]	0.5*	0.5*	0.32[c]
	Potato	0.5*	0.5*	0.5*	0.26[b]	0.5*	0.5*	0.5*	0.63[b]	0.45[b]	0.5*	0.5*	0.53[c]
	Peanut	0.5*	0.5*	0.5*	0.26[b]	0.5*	0.5*	0.5*	0.63[b]	0.45[b]	0.5*	0.5*	0.03[c]
	Rape	0.5*	1.21[d]	0.5*	0.15[b]	0.26[d]	0.5*	0.26[d]	0.63[b]	0.45[b]	0.5*	0.5*	0.3[c]
	Sesame	0.5*	1.78[d]	0.5*	0.26[b]	0.24[d]	0.5*	0.29[d]	0.63[b]	0.45[b]	0.5*	0.5*	0.3[c]
	Beet	0.5*	0.5*	0.5*	0.26[b]	0.5*	0.5*	0.5*	0.63[b]	0.45[b]	0.5*	0.5*	0.3[c]
	Hemp	0.5*	0.5*	0.5*	0.26[b]	0.5*	0.5*	0.5*	0.63[b]	0.45[b]	0.5*	0.5*	0.3[c]
	Rice	0.5*	0.05[a]	0.5*	0.29[b]	0.5*	0.5*	0.06[a]	0.65[b]	0.5[b]	0.01[a]	0.5*	0.46[c]
	Soybean	0.5*	1.78[d]	0.5*	0.26[b]	0.76[d]	0.5*	0.44[d]	0.63[b]	0.45[b]	0.5*	0.5*	0.74[c]
	Firewood	0.5*	1.42[d]	0.5*	0.16[b]	0.15[d]	0.5*	0.39[d]	0.46[b]	0.35[b]	0.5*	0.5*	1.17[c]
	Feces	0.8*	0.8*	0.8*	0.8*	0.8*	0.8*	0.8*	0.8*	0.8*	0.8*	0.8*	0.8*

Table S1 CV (coefficients of variation) of biomass domestic burning emission factors.

Note: Lowercase letters indicate the data source.

Sources are from the following: [a] Zhang et al. (2008). [b] Li et al. (2009). [c] Chen et al. (2013). [d] Zhang et al. (2013). * Expert judgment data from Wei et al. (2011).

S9 CV (coefficients of variation) of biomass open burning emission factors

	Material	SO_2	NO_x	PM_{10}	$PM_{2.5}$	NMVOC	NH_3	CO	EC	OC	CO_2	CH_4	Hg
Open burning	Corn	0.45[b]	0.42[b]	0.5[*]	0.09[b]	0.53[b]	0.76[b]	0.08[b]	0.33[b]	0.39[b]	0.01[b]	0.22[b]	0.05[a]
	Wheat	0.67[b]	0.52[b]	0.5[*]	0.54[b]	0.25[b]	0.38[b]	0.41[b]	0.32[b]	0.26[b]	0.03[b]	0.25[b]	0.12[a]
	Cotton	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.33[a]
	Cane	0.5[*]	0.32[d]	0.19[d]	0.16[d]	0.71[d]	0.5[*]	0.61[d]	1.57[d]	0.2[d]	0.18[d]	0.5[*]	0.32[a]
	Potato	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.53[a]
	Peanut	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.03[a]
	Rape	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Sesame	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Beet	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Hemp	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.3[a]
	Rice	0.5[*]	0.8[d]	0.88[d]	0.17[d]	0.75[d]	0.5[*]	1.19[d]	1.38[d]	1.53[d]	0.14[d]	0.5[*]	0.46[a]
	Soybean	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.5[*]	0.74[a]
	Evergreen Needleleaf Forest	0.3[c]	0.39[c]	0.25[d]	0.25[d]	0.31[e]	0.66[e]	0.38[e]	1[f]	0.62[f]	0.08[e]	0.52[e]	0.52[g]
	Evergreen Broadleaf Forest	0.4[e]	0.54[e]	0.25[d]	0.25[d]	-	1.58[h]	0.29[e]	0.6[e]	0.57[e]	0.04[e]	0.39[e]	0.52[g]
	Deciduous Needleleaf Forest	0.3[c]	0.23[c]	0.25[d]	0.25[d]	0.31[e]	0.66[e]	0.38[r]	1[f]	0.62[f]	0.08[e]	0.52[r]	0.52[g]
	Deciduous Broadleaf Forest	0.3[c]	0.46[e]	0.25[d]	0.25[d]	0.79[e]	0.27[e]	0.19[e]	0.33[e]	0.52[e]	0.02[e]	0.18[e]	0.52[g]
	Mixed Forest	0.3[c]	0.46[e]	0.25[d]	0.25[d]	0.62[e]	0.27[e]	0.19[e]	0.33[e]	0.52[e]	0.02[e]	0.18[e]	0.52[g]
	Closed Shrublands	0.44[e]	0.21[e]	0.25[d]	0.25[d]	0.48[e]	0.33[e]	0.25[e]	0.4[f]	0.18[f]	0.02[e]	0.35[e]	0.74[h,g]
	Open Shrublands	0.44[e]	0.21[e]	0.25[d]	0.25[d]	0.48[e]	0.33[e]	0.25[e]	0.4[f]	0.18[f]	0.02[e]	0.35[e]	0.74[h,g]
	Woody Savannas	0.44[e]	0.21[e]	0.25[d]	0.25[d]	0.48[e]	0.33[e]	0.25[e]	0.4[f]	0.18[f]	0.02[e]	0.35[e]	0.52[h]
	Savannas	0.63[e]	0.29[e]	0.25[d]	0.25[d]	0.25[e]	0.8[e]	0.29[e]	0.5[e]	0.46[e]	0.02[e]	0.6[e]	0.52[h]
	Grasslands	0.63[e]	0.29[e]	0.25[d]	0.25[d]	0.25[e]	0.8[e]	0.29[e]	0.5[e]	0.46[e]	0.02[e]	0.6[e]	0.52[h]

Table S2 CV (coefficients of variation) of biomass open burning emission factors

Note: Lowercase letters indicate the data source.

Sources are from the following: [a] Chen et al. (2013). [b] Li et al. (2007). [c] Andreae and Rosenfeld (2008). [d] Song et al. (2009). [e] Akagi et al. (2011). [f] McMeekin et al. (2008). [g] Friedli et al. (2003). [h] Streets et al. (2005). * Expert judgment data from Wei et al. (2011).

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