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 the careful and insightful review. The comments and suggestions have contributed greatly to improving our paper. Our point-by-point responses to the comments are listed as follows.

Authors Reply to Referee 1 #

Page 8 Line 23-26: 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 the 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 constant CF cannot reflect the spatial variation on the fraction of burned aboveground biomass when a fire happens. CF was controlled by both land cover type and moisture condition. Land cover type decides the maximum and minimum of the combustion efficiency. But the moisture condition determines how much the fuel can be burned during fires. If the fuel is too dry, almost 90% can be combusted, but when it is too wet, only 5% can be burned (van der Werf et al., 2006; 2010). This condition (ranging from 5 to 90%) significantly affects the final emission estimation. Many existing studies dealed with the problem by using vegetation fraction, leaf area index or NDVI to reflect the real condition of CF pixel by pixel since they vary greatly (Zhang et al., 2008, Atmos Environ).

Response:

We thank you very much for your comments. In our paper, a comprehensive biomass burning emission inventory, including domestic straw burning, in-field straw burning, firewood and livestock excrement combustion, and forest and grassland fire for mainland China was developed. As for the part mentioned by the reviewer, the CF was used for the estimation of the forest and grassland fires emission. According to our result, the contribution to total emission for most pollutants of the forest and grassland fire ranged from 0.9% to 3.7%. In other studies, the forest and grassland fire is also not the main biomass burning emission source in China (Lu et al., 2011; Huang et al., 2012). Therefore, considering the information we could obtain currently, 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), Chen et al. (2013), and Zhang et al. (2013) used the similar approach to consider CFs. Moreover, according to the current studies, vegetation type could reflect the moisture condition to some extent (Chang, 1986; Niu, 2000; Ren and Lin, 2013). According to the consideration mentioned above, the CF value selection would not have important impact on the emission inventory result in this study.

Human waste open burning accounted for a large proportion in developing countries due to without efficient burning facility, especially in rural area. China is a good example. The study from Wiedinmyer et al. (2014) estimated the global emissions of trace gases, particulate matter, and hazardous air pollutants from open burning of domestic waste, China is the largest contributor worldwide. Please refer to the large emissions from this part.

Response:

We thank you very much for your suggestion. The human waste burning indeed accounted for a large proportion in developing countries. However, the aim of this study is a comprehensive emission inventory of biomass burning. According to our previous response, the open burning of the biomass waste has been included in our paper, and the human waste irrelevant to the biomass is not the target in this study. Moreover, Few EFs of human waste burning are categorized into specific waste types in current studies (the detailed literature review could be found in the previous response). The specific human 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 the introduced emissions that are irrelevant to biomass burning. With more studies on the specific characteristics and EFs of human waste burning, further study could be conduct about the detailed and reliable emission inventory of human waste burning.

The Monte Carlo model was used to allocate the uncertainty. How did the author run this model? There is a strange phenomenon in this process since the input data for emission estimate are very with Street et al (2003) and van der Werf et al. (2006; 2010), Huang et al. (2011). And the uncertainties you defined in this study is the same with others, therefore, how the uncertainty ranges are so narrow than the others? That means why your estimation has more confidence than others. This is unbelievable.

Response:

Thanks very much for your comments. The details about the Monte Carlo model running has been described in Sect. 3.6 (Lines 11-19 on Page 18) of the revised manuscript:

"Activity data (Zheng et al., 2009) and EFs (Zhao et al., 2011) are assumed to be normal distributions. The coefficients of variation (CV, the standard deviation divided by the mean) of activity data and EFs were obtained from literature review. CV of activity data for firewood and 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 product 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 factors (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. The range of emissions

were calculated by averaging 20000 Monte Carlo simulations with a 95% confidence interval……"

It could be found from the supplement S8 and S9 that CV of EFs in our paper is generally lower than that in Street et al. (2003). This leads to the narrower uncertainty range of emission estimation than others to some extent.

The author stress the Emission Factors (EFs) in their study. The emission factors indeed have spatial variations with geographical characteristics. I welcome the authors to employ the regional EF to reflect the real condition. All EF actually can only be derived from experiment, the authors just used the EF from literature. Your study China covers a large area with strong geographical characteristics, the EF in different ecosystem located in different climatic zones vary greatly. Only one EF value cannot reflect the real situation in China.

The authors contributed their originality to the other studies by highlighting the employed EF in China, but the EF you used actually has minor differences or we can say they are identical to many studies outside China (Andreae and Merlet, 2001; Akagi et al., 2011). therefore, you cannot conclude assertively at Page 20 Line 3-5 below.

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.

Besides, the emission is calculated by using activity data and emission factors. Before excluding the potential different activity data between this study and others, it is irresponsible to judge a conclusion that localized EF is the only reason that contributes your emissions comparable to others.

Response:

Thanks very much for your suggestion.

First, in order to develop the comprehensive and detailed biomass burning emission inventory which could reflect the actual situation in China, we have try to take full account of the domestic and in-field straw burning EFs measured in China for 12 pollutants and the specific biomass burning source (including 12 crop straw) according to the extensive literature review. Based on this premise, it's difficult to further select the region-specific EFs for various areas in China due to the limited measurement. That is why few study on the biomass burning emission inventory is developed based on the EFs with regional difference. With more studies on the EFs measurements in different region in China, the emission inventory could be further improved based on the detailed EFs which could reflect spatial variations with geographical characteristics.

Second, the Table 5 and Table 6 show that most of the literature we mentioned is from the measurement in China and they show obvious difference to the studies outside China. For example, the in-field wheat EF_{SO2} in Li et al. (2007) is 0.85 while is 0.4 in Andreae and Merlet (2001), the domestic rice EF_{nox} in Zhang et al. (2008) is 1.81 while is 1.1 in Andreae and Merlet (2001). We think that the localized EFs selection as much as possible for the various pollutants and the specific biomass burning source is a significant factor for the comprehensive emission inventory improvement.

In addition to the EFs, detailed activity data are also important for a reliable emission inventory, such as detailed activity data with high spatial resolution, the province-specific domestic/in-field straw burning percentage that reflects the status of China in recent years, and so on. The detailed activity data could reduce the uncertainty of emission inventory to some extent because they could reflect the actual situation better. As for the sentence mentioned by the reviewer, we have made the corresponding modification in the revised manuscript in Lines 1-3 on Page 19:

"…As the detailed activity data could also reduce the uncertainty of emission inventory to some extent because they could reflect the actual situation better, in spite of the uncertainty exists in this study, our emission inventory is relatively reliable due to the selection of localized EFs and the detailed activity data."

Figure 7 has a fundamental serious problem. Based on the reply from the authors, we think that. But please refer to the urban center of Beijing, the $PM_{2.5}$ annual emissions is very low, there was scarce open burning, firewood burning. But comparing to other counties in North China Plain, their amounts are similar to Beijing. If you attribute the low emission in Beijing center to its urban area. That means the counties with low emissions in Shandong, Jiangsu and Anhui provinces are all urban areas, which are larger than Beijing. This is a paradox. The strange results appeared many polygons with obvious administrative boundaries, where the emissions were significantly lower than its surrounding areas. The authors failed in improving this figures and understanding the real meaning.

Please refer to Huang et al. (2011) on biomass burning emissions in China.

Response:

Thanks very much for your comment. It should be noted that there are 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. There is little crop yield, cultivated land and rural population. Therefore, in Fig. 7, the biomass burning emission of these areas were significantly lower than its surrounding areas, such as the Dongcheng and Xicheng district in Beijing, the Bincheng and Chengyang district in Shandong, the Qixia and Yuhuatai district in Jiangsu, and the Shushan and Yaohai district in Anhui. The central urban areas in Shandong, Jiangsu, and Anhui provinces are indeed larger than those in Beijing.

As for the literature mentioned by the reviewer (Huang et al, 2011), the EFs used for emission estimation were from one literature (Cao et al., 2005). The emission was allocated by the population density. Because the spatial allocation figures in Huang et al. (2011) represent the distribution for the total emission including not only biomass burning but also other source categories, it could not reflect the spatial distribution characteristic of the biomass burning emission. The biomass burning emission in our study were first estimated based on the straw data at county scale and the EFs summarized from local tests in the latest researches, and then allocated to grid cells according to the rural population density. We think the grid emission allocated from an emission inventory with improved preliminary resolution (i.e., county-level resolution) based on the appropriate surrogate could better reflect the actual situation.

The author stressed the updated biomass burning emissions in China. The study period was selected to be 2012. There is no sign to show the reason for 2012 selection. A comprehensive biomass burning emissions inventory lasting for only one year has little implications. China released strict controls and restrictions on its open biomass burning recently with the decreasing amount of crop residue burning from year to year, therefore, only one-year emission inventory has no meaning.

Response:

Thanks very much for your comment. As for the novelty and contribution of this study, we have given a detailed statement from several perspectives which could be found in the reply to the first comment from Referee #1 in our former response letter. As for the trends analysis, there are several studies that focused on the inter annual variation of open biomass burning emission (Song et a., 2009; van der Werf et al., 2010; Shon, 2015; Sun et al., 2016).

In Figure 9, the largest monthly variations of the $PM_{2.5}$ were found in in-field straw. Actually the in-field straw has strong seasonal variations peaking during field crop rotation twice or three times within a year. The farmer burns the residue for land clearing for next plantation. That means at least two peaks can be found on emissions. And then the emissions will be very low. But we cannot find the monthly variations in the South.

Response:

Thanks very much for your comment. The seasonal variation peaking twice or three times within a year could be found in Figure 9 for the in-field straw burning emission of various regions. As for the south regions, there are three relatively higher in-field straw burning emission occurred in February, April and August than other months. These periods are consistent with local sowing and harvest times in south region. February, April, and August are the sowing season of beans, the harvest season of the first-round and second-round crop (e.g., rice), respectively (CAAS, 1984; MOA, 2000). The corresponding description could be found in Lines 26-27 on Page 16 and Lines 1-4 on Page 17 of the revised manuscript:

"…As indicated by Fig. 9, as for the south regions, there are three relatively higher in-field straw burning emission occurred in February, April and August than other months. These periods are consistent with local sowing and harvest times in south region. The crops in these areas are sown earlier than in northern areas because of the climate differences. February, April, and August are the sowing season of beans, the harvest season of the first-round and second-round crop (e.g., rice), respectively (CAAS, 1984; MOA, 2000) …"

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Authors Reply to Referee 2 #

In many places I found myself a bit confused by the usage of crop straw, unsure if the authors were referring to in-field burning of crop residue or domestic burning. I suggest the authors consistently use “in-field straw” to describe in-field burning of crop straw and consistently use “domestic straw” when referring to domestic burning of crop straw. I imagine the authors use “crop residue” since not all crop residue is straw, e.g. residue from potatoes, beets, peanuts, and cotton. This should be clarified early on for the readers. Where the author are referring to all in-field burning of crop residue types they should consistently use "in-field crop residue" not "in-field straw".

Response:

Thanks very much for your comments, which were very helpful for improving our paper. We are truly sorry about this unclear description. According to the reviewer's advice, we have made the corresponding modification about the "in-field straw" and "domestic straw" through the full text in the revised manuscript. The "straw burning" without specific description means the total straw burning including in-field and domestic burning. In our paper, the straw includes crop stalk and residue. These statements have been clarified in the "General description" in Sect.2.1 (Lines 3-6 on Page 6):

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

In the manuscript text the authors need to define all chemical formulas upon their first use, e.g. at

P2, L13 "Active trace gases (e.g., SO_2, NO_x, NMVOCs, NH_3) released from biomass burning are the major precursors of…" and other places in the Introduction.

Response:

Thanks very much for your suggestion. We have made the corresponding modification in the revised manuscript in Lines 18-20 on Page 2, Lines 24-26 on Page 2 and Line 1 on Page 3, Line 28 on Page 3 and Lines 1-2 on Page 4, and Lines 12-14 on Page 5:

Lines 18-20 on Page 2: "Active trace gases (e.g., sulfur dioxide (SO_2), nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), ammonia (NH_3)) released from biomass burning are the major precursors of secondary *inorganic/organic aerosols and tropospheric ozone (O_3) in the atmosphere."*

Lines 24-26 on Page 2 and Line 1 on Page 3: "Primary particles (e.g., elemental carbon (EC) and organic carbon (OC)) discharged by biomass burning not only impact visibility, but also have an influence on climate due to the positive effects of the absorption of light and cloud condensation (IPCC, 2011). Biomass burning is also a significant source of greenhouse gases such as methane (CH_4) and carbon dioxide (CO_2) (Andreae and Merlet, 2001), which contribute to global warming (Sun et al., 2016)."

Line 28 on Page 3 and Lines 1-2 on Page 4: "Zhang et al. (2008) measured CO_2, carbon monoxide (CO), nitric oxide(NO), nitrogen dioxide (NO_2), NO_x, and PM EFs of rice, wheat, and corn straw. And Wang et al. (2009) *launched a study on characteristics of gaseous pollutants from biofuel stoves in China."*

Lines 12-14 on Page 5: "The gaseous and particulate pollutants examined in this research included SO_2, NOx, particulate matter with a diameter below 10 μm (PM_{10}), particulate matter with a diameter below 2.5 μm ($PM_{2.5}$), NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and mercury (Hg)."

Frequent use of "higher" when authors should use "largest" or "highest" or need an explicit comparison, for example:

P1 L25 "…wheat straw burning has higher contribution to CO and Hg emissions." – "higher" compared to what?

P1 L26: "Heilongjiang, Shandong, and Henan provinces located in northeast and central-south region of China have higher emissions."

And similar misuse in many other places. These instances need to be corrected.

Response:

Thanks very much for your comments. We are truly sorry for the confusing description. We have made the corresponding modification through the full text in the revised manuscript.

Some examples in the revised manuscript are mentioned below:

Lines 25-27 on Page 1 and Line 1 on Page 2: "…As for the straw burning emission of various crops, corn straw burning has the largest contribution to all of the pollutants *considered except for CH_4; rice straw burning has highest contribution to CH_4 and the second largest contribution to other pollutants except for SO_2, OC, and Hg; wheat straw burning is the second largest contributor to SO_2, OC, and Hg and the third largest contributor to other pollutants…"*

Lines 3-4 on Page 2: "Heilongjiang, Shandong, and Henan provinces located in northeast and central-south region of China have higher emissions compared with other provinces in China."

P1 L21-22: Based on Figure 2, perhaps state that: "Domestic straw burning is the largest source of biomass burning emissions for all the pollutants considered except for NH_3, EC (firewood), and NO_x (in-field crop residue)"

Response:

We thank you for your suggestion. We have revised the expression in Lines 22-23 on Page 1:

"Domestic straw burning is the largest source of biomass burning emissions for all the pollutants considered except for NH_3, EC (firewood), and NO_x (in-field straw)."

P1 L 24: "As for the straw burning emission of various crops, corn straw burning has the largest contribution to EC, NO_x and SO_2 emissions"

Here any everywhere else specify "in-field" or "domestic" straw burning."

Response:

Thanks very much for your comment. According to our reply to the first comment of Referee 2 #, we have made the corresponding modification about the "in-field straw" and "domestic straw" through the full text in the revised manuscript. In addition, the "straw burning" without specific description refers to the total straw burning including in-field and domestic burning. Here is an example of it.

P2, L2: "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."

This statement is a bit confusing as written and not too useful once deciphered. Change to something like: “The months of April, May, June and October account for X% of emissions from in-field crop residue burning”.

Response:

Thanks very much for your comment. We have made the modification in the revised manuscript in Lines 6-7 on Page 2:

“The months of April, May, June and October account for 65% of emissions from in-field crop residue burning.”

P2, L3: “While as for EC, the emission in February, January, October and December are relatively higher due to the biomass domestic burning in heating season.”

See previous comment.

Response:

Thanks very much for your comment. We have made the corresponding modification in the revised manuscript in Lines 8-9 on Page 2:

“While as for EC, the emissions in February, January, October, November and December are relatively higher than other months due to the domestic biomass burning in heating season.”

P2, L3: P2, L4: “There’s regional difference in monthly variation due to the diversity of main planted crop and the climate conditions.”

“Variation” of what? Please clarify.

Response:

Thanks very much for your comment. We have made the corresponding modification in the revised manuscript in Lines 9-10 on Page 2:

“There’s regional difference in monthly variation of emission due to the diversity of main planted crop and the climate conditions.”

P2, 13: Biomass burning emission have an important role in climate system, independent of anthropogenic forcing / change. Suggest delete “change”

Response:

Thanks very much for your suggestion. We have made the corresponding modification through the full text in the revised manuscript.

P5, L25: Since E and A are in units of Mg/yr and EF is in units of g/kg, the equation should multiplied by 0.001 (.001 kg per g). This must be a typo as the emission estimates in this study are similar to other inventories (Fig 10) and not high by a factor 100,000.

Response:

Thanks very much for your comment. We are truly sorry about this mistake. We have revised the equation in the revised manuscript in Line 12 on Page 6:

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

P10, L2-4: This first two sentences are awkward and bit confusing. I believe the authors intend something like: "Detailed speciation of NMVOC and $PM_{2.5}$ emissions in necessary to model gas and aerosol chemistry and simulate the impact of biomass burning on atmospheric composition and it has received extensive attention by domestic scholars in recent years (refs…)"

Response:

Thanks very much for your suggestion. We have made the corresponding revision in revised manuscript in Lines 14-15 on Page 10:

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

P10 L18-19: Based on Figure 2, perhaps rephrase to stress that domestic straw burning is the largest source of biomass burning emissions for all the pollutants considered except for NH_3, EC (firewood), and NO_x (in-field crop residue).

Response:

Thanks very much for your comment. We have made the corresponding revision in revised manuscript in Lines 9-10 on Page 11:

"*…Domestic straw burning is the largest source of biomass burning emissions for SO_2 (57.8%), PM_{10} (42.8%), $PM_{2.5}$ (42.0%), NMVOC (49.2%), CO (58.1%), OC (41.9%), CO_2 (38.8%), CH_4 (53.2%), and Hg (37.4%)…*"

P11, L15-17: "Among the various crops, corn straw burning has large contribution to all of the chemical species except for CH_4. Rice straw has the largest contribution to CO_2, NMVOC, CH_4 and NH_3 emissions, accounting for 32.90%, 32.43%, 31.61% and 30.12%, respectively;"

These statements are contradictory. Which has the larger contribution corn or rice? Should this read "corn straw burning has the largest contribution to all of the chemical species…" and "Rice straw has the second largest contribution to…"?

Response:

Thanks very much for your comment. We are truly sorry for the confusing description. We have made the corresponding modification in the revised manuscript in Lines 8-11 on Page 12:

"Among the various crops, corn straw burning has the largest contribution to all of the pollutants except for CH_4. Rice straw burning is the largest contributor to CH_4 and the second largest contributor to other pollutants except for SO_2, OC, and Hg. Wheat straw burning is the second largest contributor to SO_2, OC, and Hg and the third largest contributor to other pollutants."

P11, L21-22: "In addition, Fig. 3a and Fig. 3b indicate that for most of the chemical species, the contribution of in-field corn residue burning is larger than that of domestic burning, except for SO_2, EC and CO_2."

This statement is incorrect. Figure 3a and Figure 3b show percentages within groups (in-field and domestic). The magnitudes of emissions from in-field and domestic cannot be inferred from Fig 3a and Fig 3b. This statement must be supported by a different figure or table.

Response:

Thanks very much for your suggestion. We are truly sorry for the unclear description. Through the Fig. 3a and Fig. 3b, we want to show the contribution of each straw burning emission to the in-field and domestic straw burning emission, respectively. We have revised the expression in revised manuscript in Line 16-26 on Page 12:

"...In addition, Fig. 3a and Fig. 3b show the contribution of each straw burning emission to the in-field and domestic straw burning emission, respectively. Similar to Fig. 3c, corn, rice, and wheat straw are the main contributors whether for in-field or domestic burning emission. However, the dominant contributor of certain pollutants are different in in-field and domestic straw burning: for SO_2 and CO_2, rice straw is the largest contributor to in-field straw burning emission while corn straw is the largest contributor to domestic straw burning emission; for NO_x and VOC, corn straw contributes most to in-field straw burning emission while rice straw contributes most to domestic straw burning emission; for CO and CH_4, corn straw has the largest contribution to in-field straw burning emission while wheat straw has the largest contribution to domestic straw burning emission."

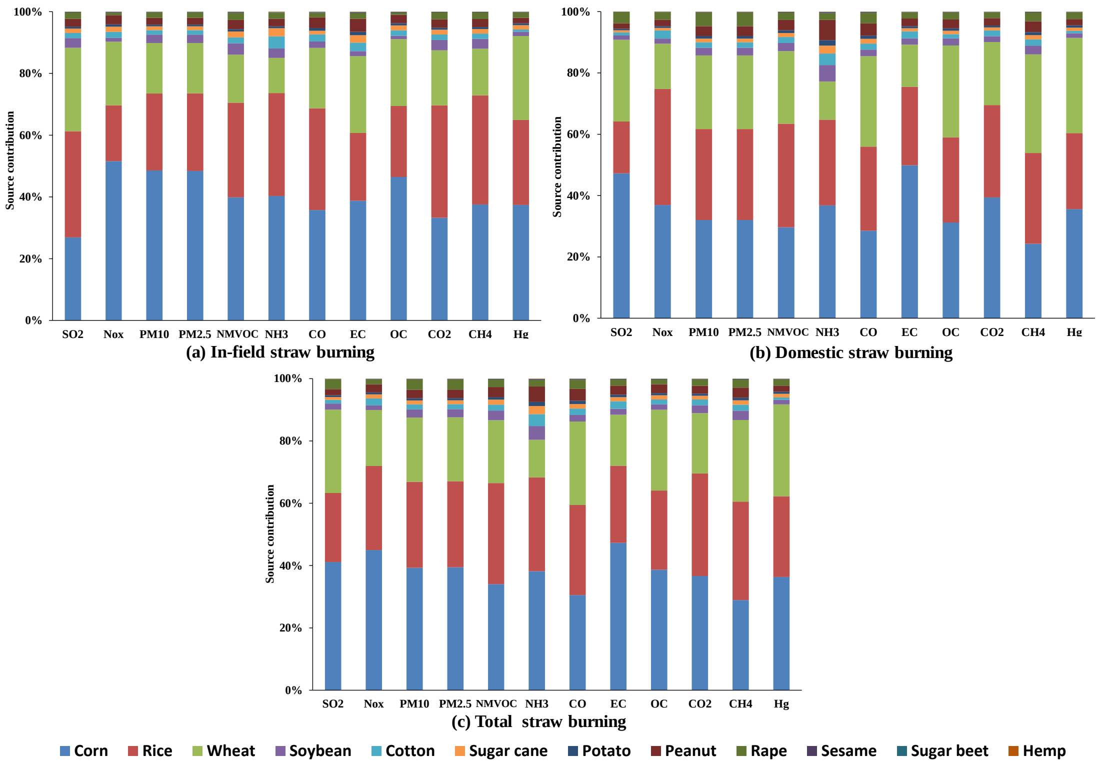

Figure 3. Contributions of 12 crop straw types to total straws burning emissions for various species.

P11, L22-24: "Contrary to that for corn straw, emissions of all chemical species (except for SO_2, NO_x and EC) from wheat straw domestic burning is greater than those from in-field crop residue burning. For rice straw, the contribution of in-field crop residue burning to NO_x, PM_{10}, $PM_{2.5}$, NMVOC, EC and OC emissions is larger than domestic burning."

These statements are cannot be supported by Figure 3. Please refer the reader to figures or tables that support these statements.

Response:

Thanks very much for your suggestion. We are truly sorry for the unclear description. Similar to the reply mentioned above, the unclear statements have been revised. The modified content could be found in revised manuscript in Line 16-26 on Page 12:

"…In addition, Fig. 3a and Fig. 3b show the contribution of each straw burning emission to the in-field and domestic straw burning emission, respectively. Similar to Fig. 3c, corn, rice, and wheat straw are the main contributors whether for in-field or domestic burning emission. However, the dominant contributor of certain pollutants are different in in-field and domestic straw burning: for SO_2 and CO_2, rice straw is the largest contributor to in-field straw burning emission while corn straw is the largest contributor to domestic straw burning emission; for NO_x and VOC, corn straw contributes most to in-field straw burning emission while rice straw contributes most to domestic straw burning emission; for CO and CH_4, corn straw has the largest contribution to in-field straw burning emission while wheat straw has the largest contribution to domestic straw burning emission."

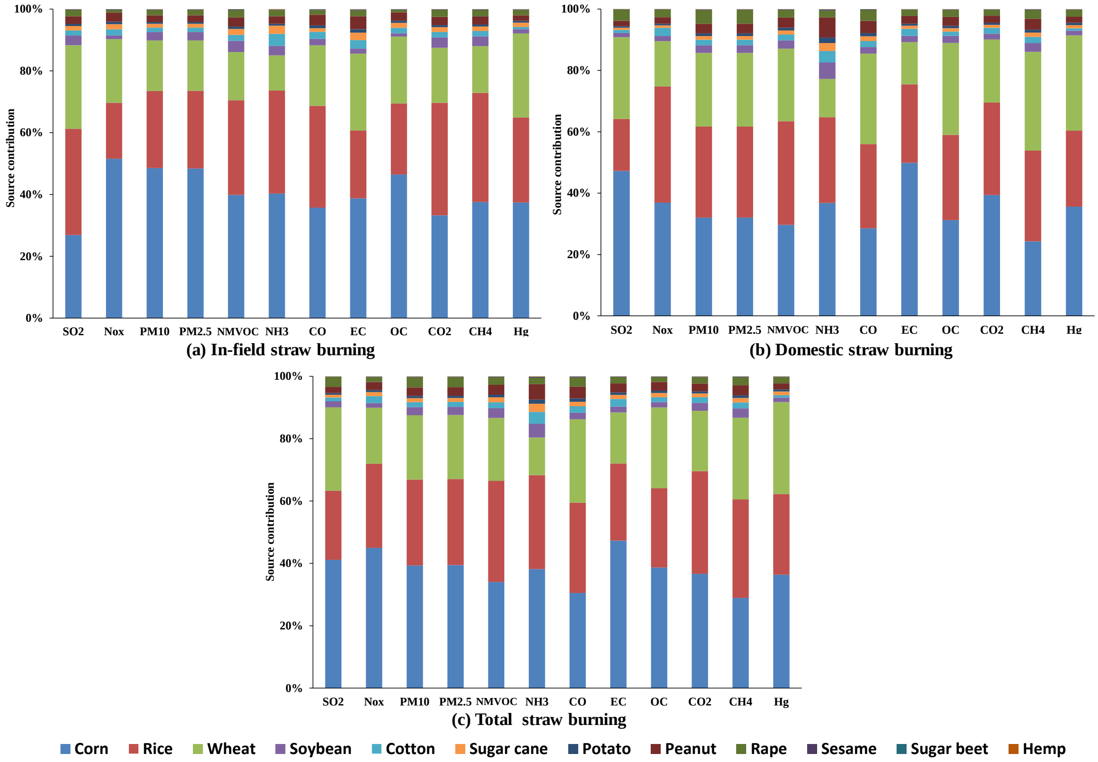

Figure 3. Contributions of 12 crop straw types to total straws burning emissions for various species.

Technical

The manuscript has many instances where English usage needs to be improved. Here is a list of some, but not all instances.

P3, L1: insert "frequently" between "more" and "burned"

P3, L10: insert "associated" between "the" and "environmental"

P3, L12: change "inventory" to "inventories"

P3, L24: spelling, change "in-filed' to "in-field"

P6, L22: Maybe change "investigation" to "review"

P6, L22: change "collect" to "estimate" or "derive"

P9, L3: change "research" to "review"

P9, L6: insert "burning" between "biomass" and "emission" and change "emission" to "emissions"

P10, L5: new paragraph not needed.

P10, L16: delete "the" before "domestic straw…"

P11, L10: delete "change" it is not needed, see earlier comment for P2, L13.

P12, L8: change "shown" to "provided"

P14, L 7: Change "The most of high values" to "Most of the high values"

P15, L6: Change "Figure 8 shows the 12 species emissions in each month…" to "Figure 8 shows the monthly emissions of all 12 species considered"

P15, L7-8: "Besides, the in-field burning of crop residue mainly in the harvest season and thus shows the obvious monthly variation features."

This statement is unclear and needs to be rewritten. Do the authors mean to say that "the monthly variability in in-field emissions reflects the timing of the harvest seasons"?

P16, L16: change "The total PM2.5 emission of biomass burning emission …" to "Total PM2.5 emissions from biomass burning…"

P18, L11: change "The emission involves…" to "The emission inventory includes…"

Figure 2 caption: add "s" "source" and delete "the"

Figure 6d caption is awkward change to something like: "The distribution of county level annual $PM_{2.5}$ emissions"

Response:

Thanks very much for your careful review and suggestion. We have made the corresponding modification through the full text in the revised manuscript. The details could be found in the revised manuscript.

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

Ying Zhou[1,2], Xiaofan Xing[1,2], Jianlei Lang[1,2], Dongsheng Chen[1,2], Shuiyuan Cheng[1,2,3], Lin Wei[1,2], Xiao Wei[4], Chao Liu[5]

[1]Key Laboratory of Beijing on Regional Air Pollution Control, Beijing University of Technology, Beijing 100124, China
[2]College of Environmental & Energy Engineering, Beijing University of Technology, Beijing 100124, China
[3]Collaborative Innovation Center of Electric Vehicles, Beijing 100081, China
[4]Beijing Municipal Research Institute of Environmental Protection, Beijing 100037, China
[5]Environmental Meteorological Center of China Meteorological Administration, Beijing 100081, China

Correspondence to: Ying Zhou (y.zhou@bjut.edu.cn) and Shuiyuan Cheng (bjutpaper@gmail.com)

Abstract. Biomass burning injects many different gases and aerosols into the atmosphere, which could have a harmful effect on air quality, climate, and human health. In this study, a comprehensive biomass burning emission inventory including ~~crop straw domestic combustion and in-field crop residue~~domestic and in-field straw burning, firewood, and livestock excrement burning, forest, and grassland fire was developed for mainland China in 2012 based on county-level activity data, satellite data, and updated source-specific emission factors (EFs). The emission inventory within 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 combustion and in-field crop residue burning~~domestic and in-field straw burning, detailed firewood burning quantities, uneven temporal distribution coefficient) was obtained from field investigation, systematic combing of the latest research and regression analysis of statistical data. The established emission inventory includes the major precursors of complex pollution, greenhouse gases, and heavy metal released from biomass burning. The results show that the emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and Hg in 2012 are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34380.4 Gg, 369.7 Gg, 1189.5 Gg, 675299.0 Gg, 2092.4 Gg, and 4.12 Mg, respectively. ~~Straw domestic~~Domestic straw burning, in-field ~~crop residue~~straw burning, and firewood burning are identified as the dominant biomass burning sources. The largest contributing source is different for various pollutants. Domestic straw burning is the largest source of biomass burning emissions for all the pollutants considered except for NH_3, EC (firewood), and NO_x (in-field straw).~~Straw domestic burning is the major source of SO_2, CO, CH_4 and NMVOC emission; firewood contributes most to EC and NH_3 emission.~~ Corn, rice, and wheat represent the major crop straws. The combined emission~~s~~ 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 all of the pollutants considered except for CH_4; rice straw burning has highest contribution to CH_4 and the second largest contribution to other pollutants except for SO_2, OC, and Hg; wheat straw burning is the second largest contributor to SO_2, OC, and Hg and the third largest contributor to other pollutantsAs for the straw burning emission of various crops, corn straw burning has the largest 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 higher contribution to CO and Hg emissions. Heilongjiang, Shandong, and Henan provinces located in northeast and central-south region of China have higher emissions compared with other provinces in China. 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 more agricultural and rural activity. The months of April, May, June and October account for 65% of emissions from in-field crop residue burning.,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 January, February, October, November and December are relatively higher than other months due to the biomass domestic burning in heating season. There's regional difference in monthly variation of emission 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 NMVOC emission include ethylene, propylene, toluene, mp-xylene, and ethyl benzene, which are key species for the formation of secondary air pollution. The detailed biomass burning emission inventory developed by this study could provide useful information for air quality modelling and 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., sulfur dioxide（SO_2), nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), ammonia (NH_3))(e.g., SO_2, NO_x, 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 pollutants 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., elemental carbon (EC) and organic carbon (OC))(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 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 large agricultural country and thus once consumed a large amount of biofuels (e.g., straw 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, straws were more frequently 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 in-field burning of straw 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 labour costs but also delayed sowing of the next crop. Thus, the phenomenon of straw in-field burning continued to occur. The amount of in-field ~~crop residue~~straw 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 associated 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 inventories~~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 straw burning in field and Cao et al. (2008) measured EFs for the domestic burning of rice straw, wheat straw, corn straw, and cotton straw. Zhang et al. (2008) measured

CO_2, carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO_2), NO_x, and PM EFs of rice, wheat, and corn straw. 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 in-filedin-field burning of sugar cane leaves and rice straw in southeast China. Ni et al. (2015) conducted laboratory burning tests to determine the EFs of wheat straw, rice straw, and corn straw, considering the impacts of the fuel moisture content.

Based on the local EFs, emission inventories that focused on certain provinces (Li et al., 2015; He 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; 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 domestic and in-field straw burning percentage, 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 residuestraw 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 residuedomestic and in-field straw burning in 2007 for seven and three regions of China, respectively. Thus, there is limited information about the percentage of straw used as fuel or waste in the field that reflects the status of China in recent years for different provinces. Moreover, because of the lack of firewood consumption report in the China Energy Statistical Yearbook (NBSC, 2009-2015), few studies have developed a comprehensive biomass burning emission inventory in China in recent years. 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 domesticdomestic and in-field straw 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. Activity data at coarse resolution are likely to be associated with great~~er~~ 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~~domestic and in-field straw burning, firewood, and livestock excrement burning, 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~~domestic/in-field straw burning percentage, detailed firewood burning quantities and uneven temporal distribution coefficient) were obtained from a field investigation, systematic combing of latest research and regression analysis of statistical data. A 1-km resolution emission inventory was generated using GIS software. The gaseous and particulate pollutants examined in this research included SO_2, NO_x, particulate matter with a diameter below 10 µm (PM_{10}), particulate matter with a diameter below 2.5 µm ($PM_{2.5}$), NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and mercury (Hg), covering the major precursors of complex pollution, greenhouse gases, and heavy metals released from biomass burning. The detailed emission inventory given by this paper could provide valuable information to support 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, speciation of $PM_{2.5}$ and NMVOC~~s~~. 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, 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 NMVOC species. Uncertainty in biomass burning emission estimates is described in Sect. 3.6. The comparison between this study and other studies appears in Sect. 3.7. 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 burning and open burning. Domestic burning mainly involves domestic straw (straw burned as fuel indoors), firewood, and livestock excrement (mainly used in pastoral and semi-pastoral areas) burning. Open burning includes in-field straw burning (straw burned as waste outdoors, including crop stalk and residue), forest and grassland fire. Straw burning without specific description in this paper refers to the total straw burning including in-field and domestic straw burning.~~The biomass burning considered in this study is mainly divided into two categories, domestic burning and open burning. Domestic burning 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 sources classification are shown in Table 1.

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

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

where subscripts i and j represent the type of pollutant and biomass burning source, respectively; E is the annual typical pollutant emission (Mg/yr); A is annual amount of dry biomass burned (Mg/yr), 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 domestic burning and in-field crop residue~~domestic and in-field straw burning can be calculated using Eq. (2) as follows:

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

where subscripts i and k represent region (district or county) and crop type, respectively; $A_{i,k}$ is the annual burning mass of each crop straw in each region (Mg/yr); $P_{i,k}$ is the amount of crop-specific yields per year in each region (Mg/yr); N_k is the straw-to-product ratio of each straw type~~-~~; $R_{i,k}$ is the domestic or in-field straw burning percentage~~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 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. 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 maps at prefecture and county resolution are ~~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 represent the recent status in China needs to be updated because of the continued economic development and the gradual implementation of national control policies for in-field ~~crop residue~~straw burning. In this study, we conducted a detailed ~~investigation~~ review of recent literature to derive~~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). $R_{i,k}$ is summarized in Table 2. According to our estimation, the amount of ~~straw domestic and in-field burning~~domestic and in-field straw burning for China in 2012 was 0.26 billion Mg and 0.19 billion Mg, respectively, which is similar to other recently published results for 2012 (0.26 billion Mg domestic straw burning, Tian et al., 2014) and 2009 (0.215 billion Mg in-field ~~crop residue~~straw 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 2008. For more recent years, we obtained the total firewood consumption for China in 2012 and for each province in 2010 (Tian et al., 2014; 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 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):

$$A = (\sum_{j=1}^{10} BA_{x,j} \times FL_{x,j} \times CF_{j}) \times 10^{-6}, \quad (3)$$

where subscripts j, and x represent the land cover type, and location, respectively; A is the annual burning mass of forest and grassland fire (Mg); $BA_{x,j}$ 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 burned) of land cover type j.

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

2.2.4 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~~ burning mass of livestock manure ~~burned~~ (Mg/yr); S represents the amount of each livestock type in pastoral and semi-pastoral areas at the end of the year (head/yr); Y is a single livestock annual fecal output per year (Mg/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 large animals, single cattle annual manure output was 10 Mg and single horse annual manure output was 7.3 Mg (Li and Zhao, 2008). The livestock annual manure output of other animals was set at 8 Mg, according to Tian et al. (2011). The C value was set as 18% (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 EF from localized measurement in the literature.~~we prioritized the use of localized measured EFs from the literature.~~ Second, for the biomass sources or crop types which lacked of localized measurements, we prioritized results from developing foreign countries similar to our country above those of developed countries. Third, when localized measurement~~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~~ review~~on EFs~~, the resultant EFs ~~for~~ of domestic and open burning for each ~~chemical species~~pollutant 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 burning emissions, and to provide grid based data for the air quality model simulation, the biomass burning inventory in this study 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 in-field ~~crop residue~~straw 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 surrogate~~s~~ of in-field ~~crop residue~~straw 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 emissions of forest and grassland fire are were estimated in 500m resolution, it can be reshaped into 1km grid using GIS software. The emissions of straw, firewood, and livestock excrement burning were treated as area sources and the spatial surrogates used to distribute these biomass sources were 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 in-field crop residuestraw burning can be estimated based on the number of typical specific fire points, and the monthly/daily emission of forest and grassland fire emission can be calculated by the Julian day emission of forest and grassland fire. For domestic biomass source, the monthly emission of each source can be estimated based on the monthly uneven coefficient which was derived from our survey questionnaire. Details of the questionnaire survey are presented in the Supplement (S3). The daily domestic emission is equally allocated from the monthly emission.

2.6 Speciation of NMVOCs and $PM_{2.5}$

Detailed speciation of NMVOC and $PM_{2.5}$ emissions is necessary to model gas and aerosol chemistry and simulate the impact of biomass burning on atmospheric composition and it has received extensive attention by domestic scholars in recent years The detailed species emission of NMVOCs and $PM_{2.5}$ is necessary information of model simulation for different chemical mechanism selection (e.g., CB05). The speciation of NMVOCs 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 NMVOC, $PM_{2.5}$ 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 NMVOC source profile mainly refers to data from Liu et al. (2008) and Akagi et al. (2011), including species covering alkane, alkene, alkyne, aromatic, 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}, $PM_{2.5}$, NMVOC, 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 Mg, respectively. The contribution of different sources to the total emissions of various pollutants is shown in Fig. 2. It shows that ~~the straw domestic~~domestic straw burning, in-field ~~crop residue~~straw burning, and firewood burning are the dominant biomass burning sources with the total contribution ranging from 86.02% to 97.58% for various pollutants. However, the largest contributing source to different pollutants are not similar. Domestic straw burning is the largest source of biomass burning emissions for SO_2 (57.8%), PM_{10} (42.8%), $PM_{2.5}$ (42.0%), NMVOC (49.2%), CO (58.1%), OC (41.9%), CO_2 (38.8%), CH_4 (53.2%), and Hg (37.4%).~~Compared with other sources, straw domestic burning contributed most to SO_2, CO, CH_4 and NMVOC, accounting for 57.8%, 58.1%, 53.2% and 49.2% of total emissions, respectively.~~ It has a direct impact on residents. Moreover, the prolonged exposure under high domestic straw burning emission (e.g., SO_2, CO, CH_4, and Hg) can cause many adverse health effects (e.g. acute respiratory infections and chronic bronchitis) (Emily and Martin, 2008). The contribution of firewood to each pollutant~~s~~ cannot be neglected, especially for EC (51.3%) and NH_3 (41.2%). According to the localized measurement of EF by Li et al. (2009), the EF_{EC} for firewood (1.49 g/kg) is 3.5 times of the average of in-field straw (0.43 g/kg). EF_{NH3} of firewood is larger than the average of various straws. This results in a large contribution by firewood for these two pollutants. The contribution of ~~straw domestic burning and in-field crop residue burning~~domestic and in-field straw burning to NO_x, PM_{10}, $PM_{2.5}$, NMVOC, Hg, OC, and CO_2 is nearly equal. Straw burning has an important influence on indoor air quality and outdoor atmospheric environment.

In addition to the sources mentioned above, the contribution of livestock excrement burning, forest and grassland fire is relatively small. It is mainly due to the small amount of biomass consumption. The biomass fuel consumptions of these three biomass sources are 10614Gg, 6647Gg, and 505 Gg, respectively, which ~~is~~ are significantly lower than that of ~~straw domestic combustion~~domestic straw burning (201582 Gg), in-field ~~crop residue~~straw burning (147178 Gg), and firewood burning (127250 Gg). The contributions of livestock excrement burning to PM_{10}, $PM_{2.5}$, NH_3, EC, OC, CO_2, and CH_4 are 2.52%, 2.47%, 3.44%, 1.52%, 1.96%, 1.67%, and 2.10%, respectively. The contribution of forest and grassland fire to biomass burning emissions for most pollutants 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 Sect. 3.1.1, straw burning is the important biomass burning source with considerable influence on the pollutants that most strongly impact the air quality, climate,~~ change~~ and human health. Furthermore, the major crop straw types contribution was analysed. Figure 3 shows the contributions of 12 different crop straw types ~~of crop straw domestic burning and in-field crop residue burning to total straws burning emissions~~ for various ~~species~~ pollutants in 2012 from the perspective of the mainland China. Figure 3c indicates that corn, rice, and wheat straw are the major crop~~s~~ straws burned as fuel and~~or~~ as waste in China. The contribution is more than 80% to the total straw burning emissions of all pollutants studied in this paper. Corn, rice, and wheat are the major three food crops in China with large planting area (the output of these three kinds of grain accounts for 70% of the total grain output in China, NBSC, 2013c), resulting in a large amount of straw production. Among the various crops, corn straw burning has the largest contribution to all of the pollutants except for CH_4. Rice straw burning is the largest contributor to CH_4 and the second largest contributor to other pollutants except for SO_2, OC, and Hg. Wheat straw burning is the second largest contributor to SO_2, OC, and Hg and the third largest contributor to other pollutants.~~corn straw burning has large contribution to all of the chemical species except for CH_4. H_4 an~~Rice ~~straw has the largest contribution to CO_2, NMVOC, CH_4 and NH_3 emissions, accounting for 32.90%, 32.43%, 31.61% and 30.12%, respectively; wheat straw has 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 straw burning to the various pollutants is relatively small, accounting for 8.1–19.2% of the total emissions for all pollutants considered; the contribution of sesame, sugar beet, and hemp straw burning to various pollutants is negligible, never exceeding 0.5%. In addition, ~~Fig. 3a and Fig. 3b indicate that for most of the chemical species, the contribution of in-field corn residue burning is larger than that of 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 domestic burning is greater than those from in-field crop residue burning. For rice straw, the contribution of in-field crop residue burning to NO_x, PM_{10}, $PM_{2.5}$, NMVOC, EC and OC emissions is larger than domestic burning.~~ Fig. 3a and Fig. 3b show the contribution of each straw burning emission to the in-field and domestic straw burning emission, respectively. Similar to Fig. 3c, corn, rice, and wheat straw are the main contributors whether for in-field or domestic burning emission. However, the dominant contributor of certain pollutants are different in in-field and domestic straw burning: for SO_2 and CO_2, rice straw is the largest contributor to in-field straw burning emission while corn straw is the largest contributor to domestic straw burning emission; for NO_x and VOC, corn straw contributes most to in-field straw burning emission while rice straw contributes most to domestic straw burning emission; for CO and CH_4, corn straw has the largest contribution to in-field straw burning emission while wheat straw has the largest contribution to domestic straw burning emission.

3.2 Emissions from different regions

3.2.1 Total emissions for different provinces

The total biomass burning emissions in 31 provinces in 2012 are presented in Table 7. These results indicate that Heilongjiang, Shandong, Henan, Hubei, Anhui, Sichuan, Jilin, Inner Mongolia, Hunan, and Jiangsu province are the major contributors, with the total emission contributions ranging from 53% to 65% for various pollutants. The province with most contribution to total emission of NO_x, PM_{10}, $PM_{2.5}$, NMVOC, NH_3, OC, CH_4, Hg, and CO_2 is Heilongjiang; while Shandong province has the highest emission of SO_2, CO, and EC. 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~~ provided below.

3.2.2 Contributions by biomass sources of each province

The emission of detailed biomass sources of each province is presented in Fig. 4. The province with major contribution to total pollutant emissions for each biomass source are various. Straw burning emissions are mainly distributed in Shandong, Henan, Heilongjiang, Hebei, Anhui, Sichuan, Jilin, and Hunan 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 produces~~ a~~ large emissions are Hunan, Yunnan, Hubei, Hebei, Sichuan, Guangdong, Shaanxi, Liaoning, and Jiangxi province. More than 54% firewood burning 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 burning emissions are 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 are mainly distributed in Tibet, Yunnan, 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 pollutant emissions in Heilongjiang (79−97%), Ningxia (87−98%), Shandong (74−95%), Jilin (74−95%), Henan (61−93%), Anhui (51−91%), and Shanxi (61−90%) province. The economic income of the rural areas in these provinces is relatively low. A large number of straws are consumed as main non−commodity energy. In addition, firewood resources are scarce in these areas and as a result, the usage of straw is very high. Figure 4 also indicates that, for most provinces (e.g. Beijing, Tianjin, and Hebei), the contribution of the domestic straw burning is greater than in-field ~~crop residue~~straw 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 in-field ~~crop residue~~straw burning is higher than that of ~~straw domestic burning~~domestic straw 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 (47−90%), Guangdong (31−83%), Yunan (31−79%), Fujian (30−81%), Hainan (26−77%), and Guizhou (27−74%) province. The straw amounts in the rural areas of these provinces are relative low. Firewood is the main~~ly~~ non−commodity energy used by rural people. It is worth noting that though the biomass fuel consumption in Beijing is small, compared with~~to~~ straw burning emission contribution (9%−41%), firewood emission (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~~straw 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 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 province is considerable (exceeding 10%), which is 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 are mainly distributed in Henan, Shandong, Anhui, Hebei, Jiangsu, Sichuan, Shaanxi, Hubei, and Shanxi province. More than 89% wheat crop straw burning emission is contributed by these provinces. Rice crop straw burning emissions are mainly distributed in Heilongjiang, Hunan, Jiangsu, Sichuan, Anhui, Hubei, Guangxi, Guangdong, and Zhejiang province, with the total contribution more than 71%. The water condition, light, and heat are better for the cultivation of rice in the South. Low temperature, long sunshine duration, and the large temperature difference between day and night are suitable for wheat growing in the North. In addition, soybean, cotton, sugar cane, potato, peanut, and rape straw have a small contribution to the various pollutants, and these straws are mainly distributed in Heilongjiang, Xinjiang, Guangxi, Sichuan, Henan, and Sichuan province, respectively.

3.2.4 Emissions intensity at county resolution

At county resolution, we found that the spatial distributions of emissions for various pollutants are similar, 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 or districts. The distribution of county level annual $PM_{2.5}$ emissions was~~The number of counties within different emission ranges were~~ shown in Fig. 6d. The spatial diversity of various counties emission is obvious. There are 406 ~~counties~~ districts without biomass burning, because they are mainly distributed in the urban areas of developed cities, such as the Dongcheng and Xicheng districts in Beijing, the Jing'an district in Shanghai. The total emission of 32.3% of districts and counties (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 4.0 Gg, including the regions with low crop yield or scarce population. The emission of 30.9% of the total districts and counties (875) were more than the average emission across all counties (1.245 Gg). The two largest emission (approximately 16 Gg) appeared in Longjiang and Wuchang ~~which~~ where are major grain-producing counties in Heilongjiang province.

Figure 6b shows the $PM_{2.5}$ emissions intensities per unit area. Most of the high values~~The most of high values~~ (more than 3 Mg km^{-2} yr^{-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 giving~~e~~ priority to agricultural activity, with a substantial amount of crop straw from a relatively small area. The most counties with low~~er~~ 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 relatively 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 obvious difference. The counties with emission intensity more than 10 kg per^{-1} yr^{-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, population in there are relatively small. The counties with lower emissions intensities per capita compared with other provinces concentrated in Henan, Guangdong, and Shanxi provinces, attributed to the large amount of people there.

3.3 Spatial distribution of biomass burning emissions

As pollutants showed a similar emission distribution, $PM_{2.5}$ was taken as an example to discuss 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; these areas with high emission are mainly scattered in major agricultural region of China’s northeast to central–south, showing a zonal distribution. The biomass burning emissions are concentrated in the regions with great~~er~~ agricultural and rural activity, and low~~er~~ economic income. These regions are 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 population and crop yield in part of southwest, northwest areas, and low~~er~~ agricultural activity in downtown areas result in low~~er~~ 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. 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.

3.4 Temporal variation in biomass burning emission

Figure 8 shows the monthly emission of all 12 pollutants considered~~12 species emissions in each month~~, indicating that there are different monthly emission variations ~~in~~ for ~~the~~ each pollutant~~ emissions~~. The pollutants showing large monthly variation were SO_2, NO_x, PM_{10}, OC, NMVOC, and $PM_{2.5}$. ~~Besides, the~~The in-field burning of crop residue mainly occurred 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 domestic burning and the contributions of these two kinds of source to the total emissions of these pollutants are 73.1%, 75.9%, and 86.9%, respectively. The temporal distribution of these two sources was more uniform compared with~~to~~ in-field ~~crop residue~~ straw burning at the monthly scale, and thus monthly emissions of these three pollutants showed less temporal distinction. ~~Despite the temporal variations of some pollutants at the monthly scale~~In addition, the overall trends of emissions ~~to~~ for ~~most~~ other pollutants show a certain similarity: April, May, June, and October are the top four months with high~~er~~ emissions, mainly due to the in-field ~~crop residue~~straw burning. The total emission of these months account for 65% of emissions from in-field straw burning. While as for EC, the emissions in January, February, October, November and December are relatively higher than other months due to the biomass domestic burning in heating season.

Burning activity mainly occurs in the harvest season (~~crop residue~~in-field straw burning) or crop sowing season (clearing the cultivated land and increasing the soil fertility for the next sowing) and it varies by burning habit in different regions. In addition, the sowing and harvest seasons vary in different regions because of 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, 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, as for the south regions, there are three relatively higher in-field straw burning emission occurred in February, April and August than other ~~the south regions have relatively small peaks of $PM_{2.5}$ emissions in February, April,~~

and August, thesemonths. These periods are consistent with local sowing and harvest times in south region. The crops in these areas are sown earlier than in northern areas because of the climate differences.As a result of the climate differences, crops in these areas are sown earlier than in northern areas. February, April, and August 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; MOA, 2000). 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, owing to the burning 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, 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 regions 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 crop residuestraw burning, northern winter wheat and corn are mainly harvested in June and October, respectively. April and May are the sowing seasons of spring rice and 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 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 which are shown in the Supplement (Fig. S3). It could be found that the main emission peaks appeared in early April, early June, and the whole month of October. 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 NMVOC species

The totalTotal $PM_{2.5}$ emission of from biomass burning emission in this study is 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 33.7% 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 46.63%. Additionally, 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 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 factor~~EFs 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 factors (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. The range of emissions were calculated by averaging 20000 Monte Carlo simulations with a 95% confidence interval~~And then ran 20000 Monte Carlo simulations to estimate the range of emissions with a 95% confidence interval~~. 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 burning (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 with~~to~~ other pollutants. The total uncertainty for emissions of these pollutants are (−54%, 54%), (−49%, 48%), and (−61%, 61%), respectively. NH_3, EC, and SO_2 exist the highest uncertainties in livestock excrement burning, forest, and grassland fire. The ~~emission factor~~EFs 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.

As the detailed activity data could also reduce the uncertainty of emission inventory to some extent because they could reflect the actual situation better, in spite of the uncertainty exists in this study, our emission inventory is relatively reliable due to the selection of localized EFs and the detailed activity data.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 inventories published after 2000 have been compared with this study (Fig. 10). It could be found that the relatively high difference (range from −80% to 366% for various pollutants) 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 −34% to 15%. While the PM_{10}, NMVOC, CH_4, and NH_3 emission in this study is lower than Lu et al. (2011). The EFs of PM_{10}, NMVOC, CH_4, and NH_3 for various crop types used in this study is generally lower than the EF without specific crop types in Lu et al. (2011). The SO_2, NO_x, CH_4, and CO_2 emissions in this study are close to those in Tian et al. (2011), with the difference ranging from −49% to 40%. The difference of CO emission is relatively high. The major emission difference of the straw domesticdomestic straw burning, in-field crop residue straw burning, and firewood burning 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 residuein-field straws) 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 inventory includesThe emission involves domestic and in field straw burning, firewood, and livestock excrement burning, forest, and grassland fire. The total annual emissions of SO_2, NO_x, PM_{10}, $PM_{2.5}$, NMVOC, NH_3, CO, EC, OC, CO_2, CH_4, and Hg are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34380.4 Gg, 369.7 Gg, 1189.5 Gg, 675299.0 Gg, 2092.4 Gg, and 4.12 Mg, respectively.

The ~~straw domestic~~domestic straw burning, in-field ~~crop residue~~straw burning, and firewood burning are the major biomass burning sources, while the largest contributing source to various pollutants is different. ~~Straw domestic burning is the major source of SO_2, CO, CH_4 and NMVOC emission; while firewood contributes most to EC and NH_3 emission~~ Domestic straw burning contributes most to all of the pollutants considered except for NO_x, NH_3, and EC emission; firewood contributes most to EC and NH_3 emission; and in-field straw burning is the largest contributor of NO_x. In terms of crop straw burning, corn, rice, and wheat straw are the major crop types, with the total contribution exceeding 80% for each pollutant of straw burning~~burned~~ emissions. Corn straw burning has the greatest contribution to EC, NO_x, and SO_2 emissions; rice and wheat straw burning has the second and the third greatest contribution to most of the pollutants considered, respectively~~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 is mainly distributed in the northern China, while the rice straw burning emission is concentrated in the southern China. Gridded emission~~s~~ result ~~show~~ indicates 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 high~~er~~ emissions in April, May, June, and October were mainly due to the burning activity before sowing and harvesting of main crops. Regional differences of temporal distribution are attributed ~~in~~ to 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 high~~er~~ contribution to NMVOCs (e.g., ethylene, propylene, toluene, mp-xylene, and ethyl benzene) 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 parameters in the emission estimation. More localized EF of different biomass fuel types within diverse burning conditions, more detained $PM_{2.5}$ and NMVOC source profiles that contain as much components as possible still needs to expand in the future. In addition, the high~~er~~ 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 sources.

Table 2. Straw domestic and in-field crop residue burning proportionsDomestic and in-field straw burning percentage of each province.

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

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

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

Table 6. Emission factors used in the estimation of 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. Contributions of different sources to the total biomass burning emissions in China, 2012.

Figure 3. Contributions of 12 crop straw types for various pollutants in China, 2012.Contributions of 12 crop straw types to total straws burning 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 pollutant.

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 researches.

Table 1. The classification of biomass burning emission sources.

I	II	III
Domestic burning	Firewood	Firewood
	Livestock excrement	Cattles
		Horses
		Donkeys
		Mules
		Camels
	Straw	Corn, wheat, cotton, sugar cane, potato, peanut, rapeseed, sesame, sugar beet, hemp, rice, soybean
Open burning	In-field straw	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

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

Province	Domestic straw burning percentage	In-field straw burning percentage	Province	Domestic straw burning percentage	In-field straw burning percentage
Beijing	0.0923[a]	0.096[b]	Hubei	0.283[j]	0.197[o]
Tianjin	0.42[a]	0.165*	Hunan	0.4[c]	0.2[c]
Hebei	0.35*	0.165*	Guangdong	0.17*	0.197*
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. Straw-to-product ratio (N_k), dry matter fraction (D_k), and combustion efficiency (CE_k) of crop straw used in this study.

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

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

Province	Biomass fuel loadings (g/m^2)				
	Needleleaf Forest[a,d]	Broadleaf Forest[a,e]	Mixed Forest[a,f]	Shrublands[b,g]	Grassland[c,h]
Heilongjiang	8140	7610	7875	1387	180
Jilin	9340	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

[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 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 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 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)

Pollutant	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.

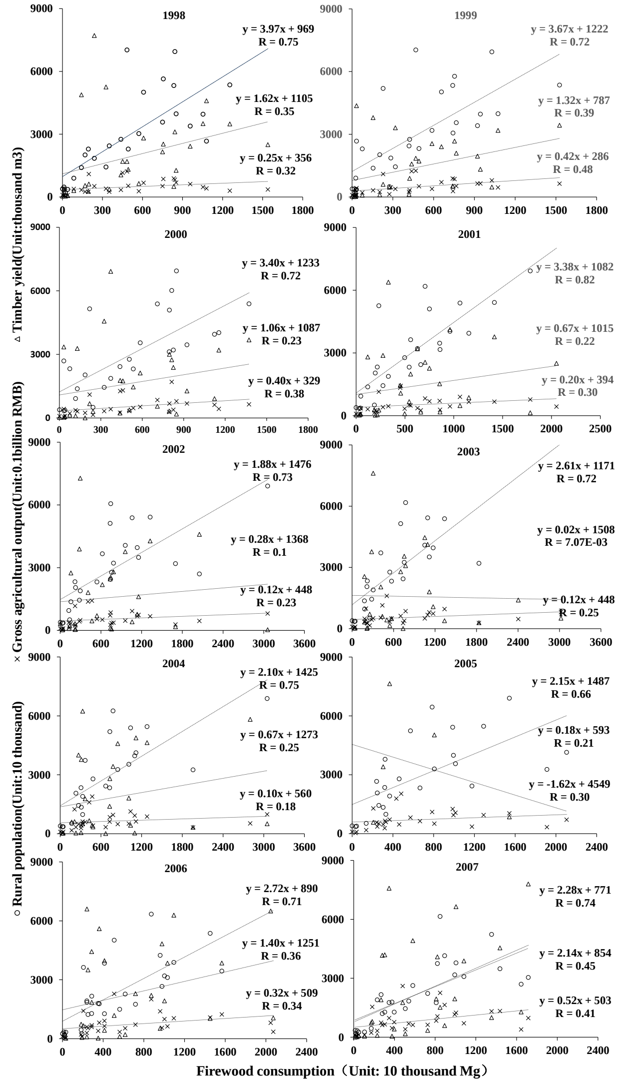


Figure 1. Regression analysis between firewood consumption at province resolution and (1) rural population, (2) gross agricultural output, and (3) timber yield, respectively.

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.

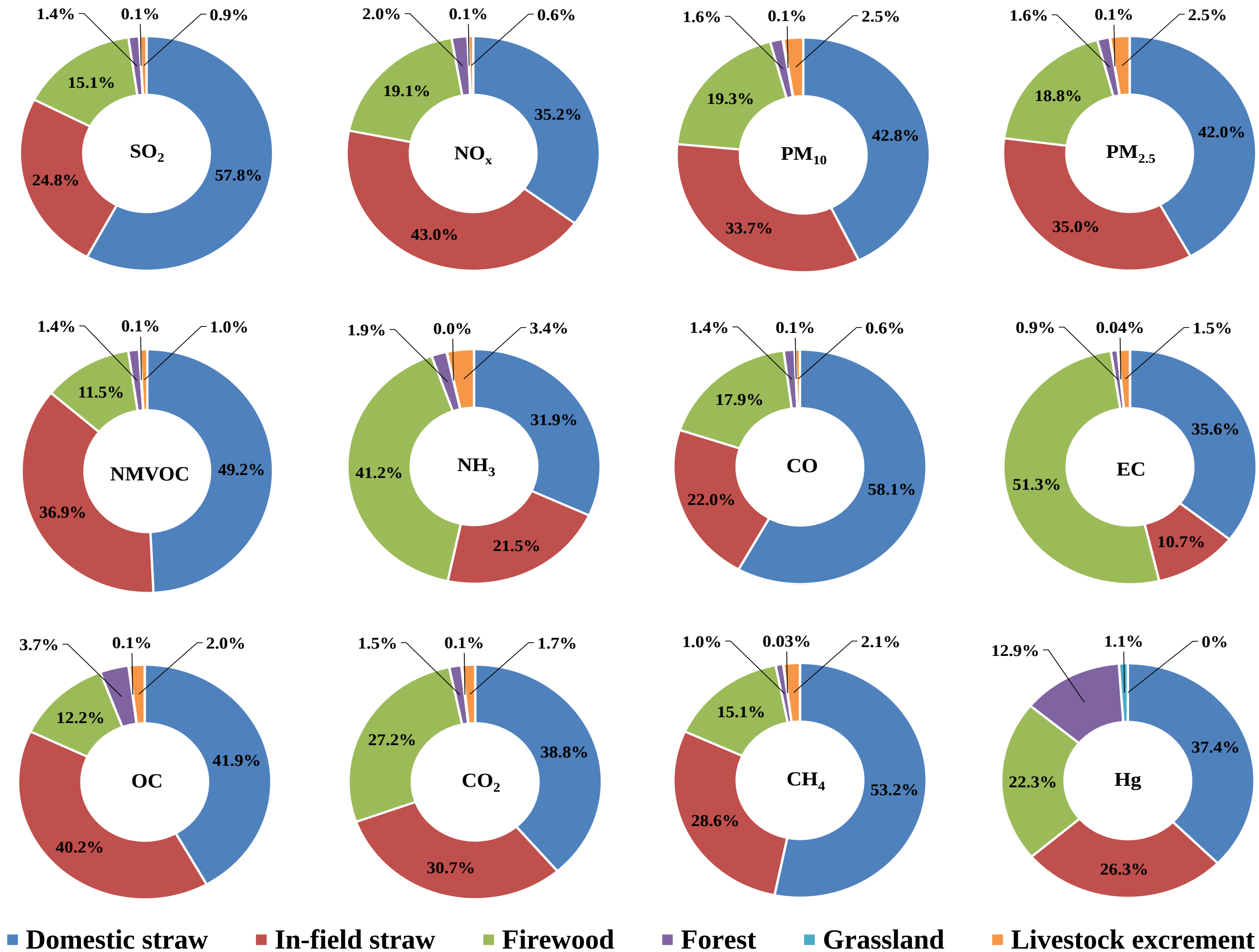


Figure 2. Contributions of different sources to ~~the~~ total biomass burning emissions in China, 2012.

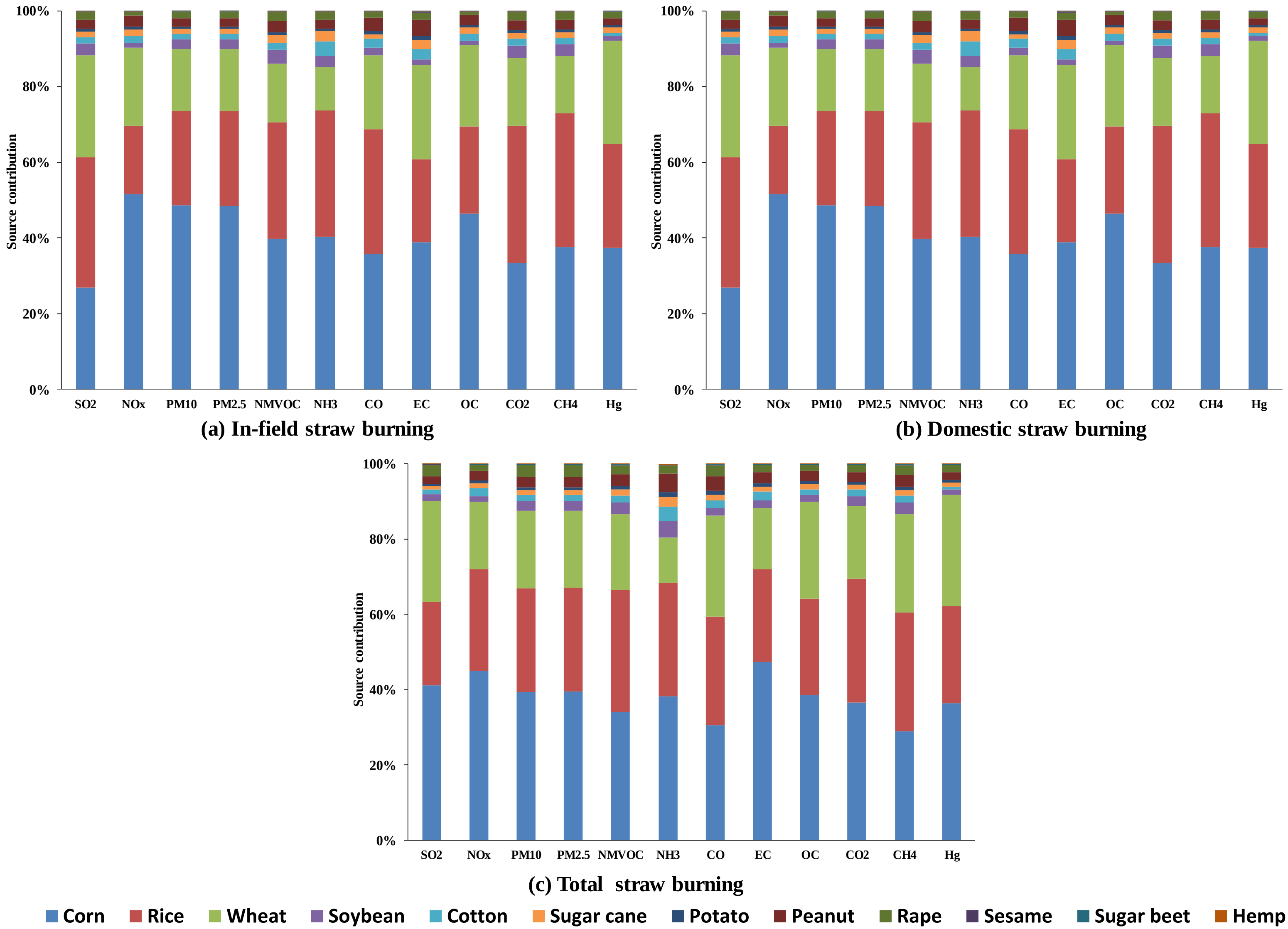


Figure 3. Contributions of 12 crop straw types ~~to total straws burning emissions~~ for various ~~species~~pollutants in China, 2012.

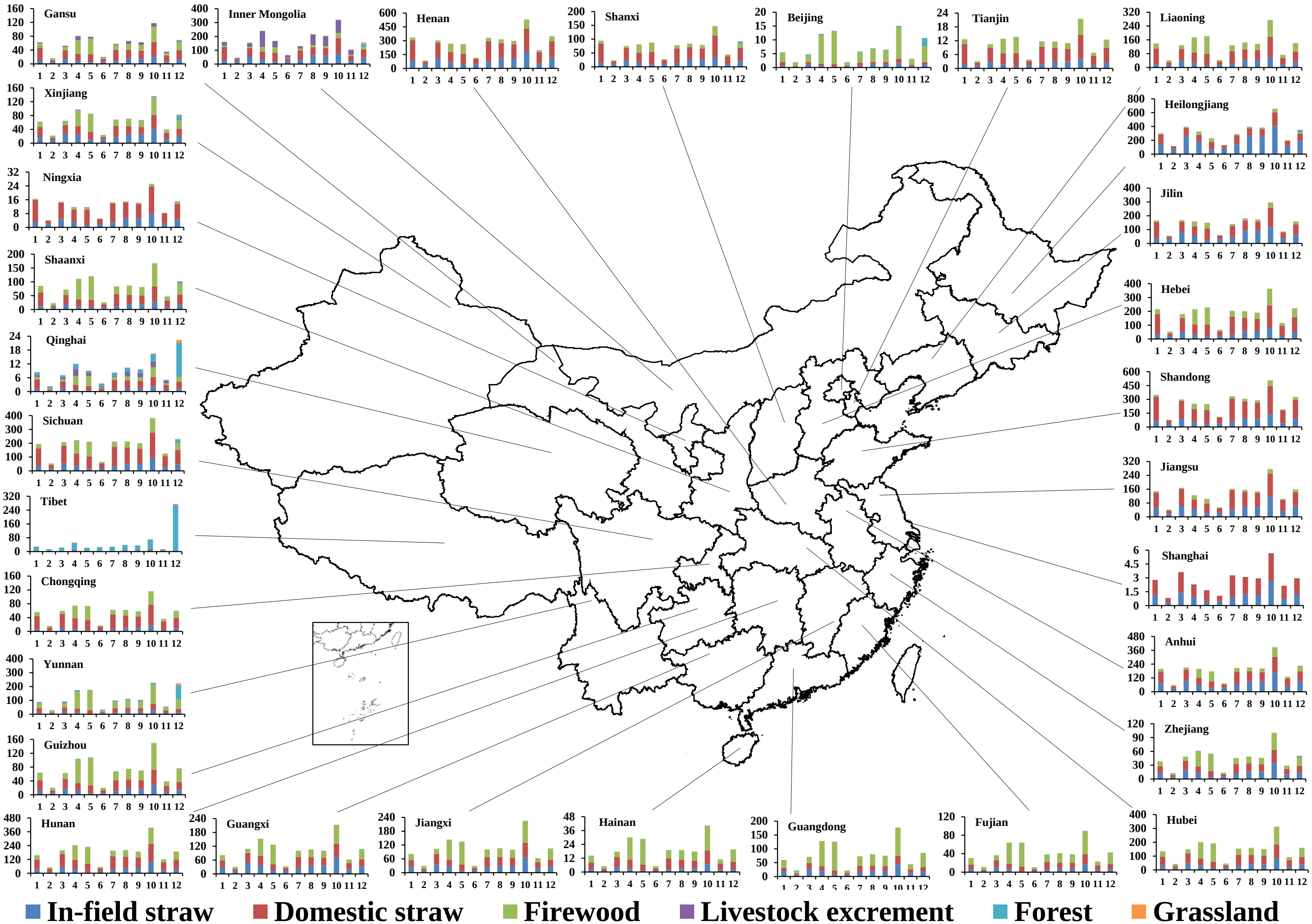


Figure 4. Contributions of different biomass sources to the emission in each province (Gg).

Note: The numbers 1–12 represent the pollutant of $SO_2\times10$, NO_x, NMVOC, $NH_3\times10$, EC×10, OC, CO/10, PM_{10}, $PM_{2.5}$, $CO_2/100$, CH_4, and Hg×1000000, respectively.

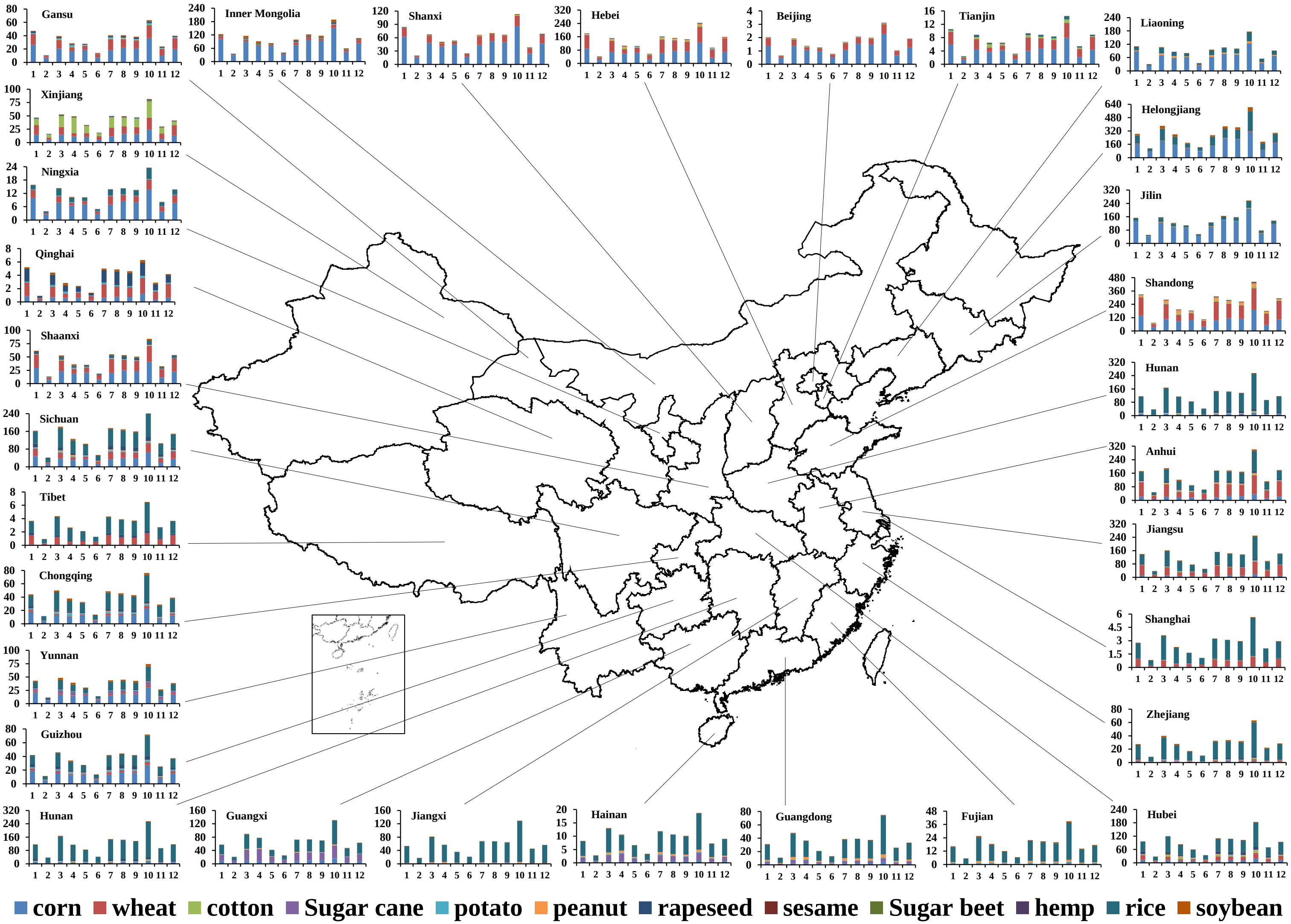
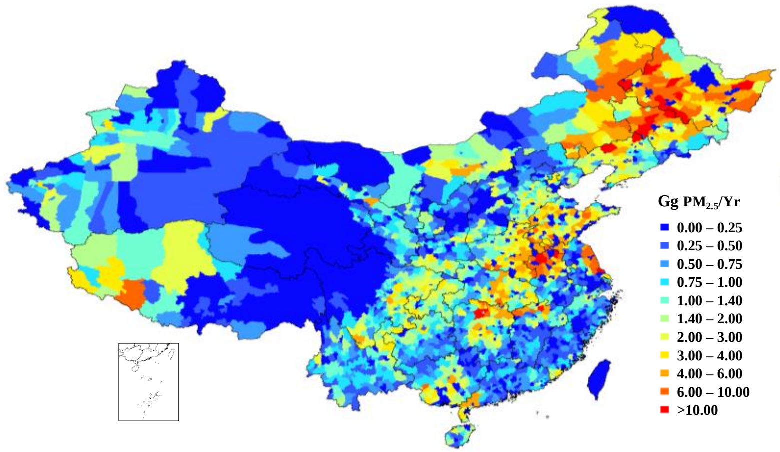
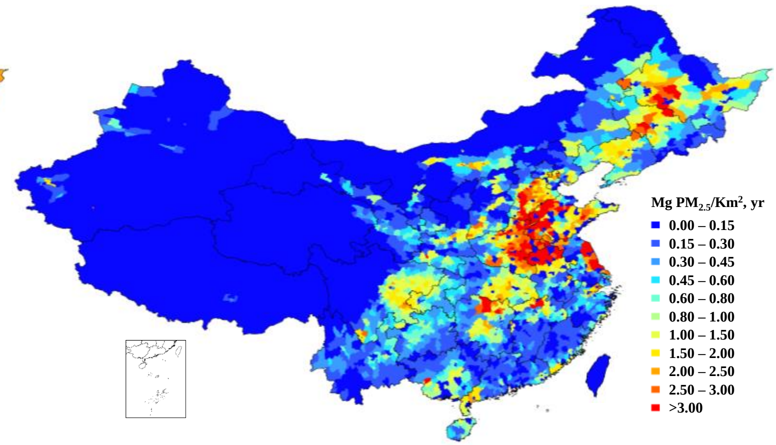


Figure 5. Contributions of different crop straw types to the emission in each province (Gg).

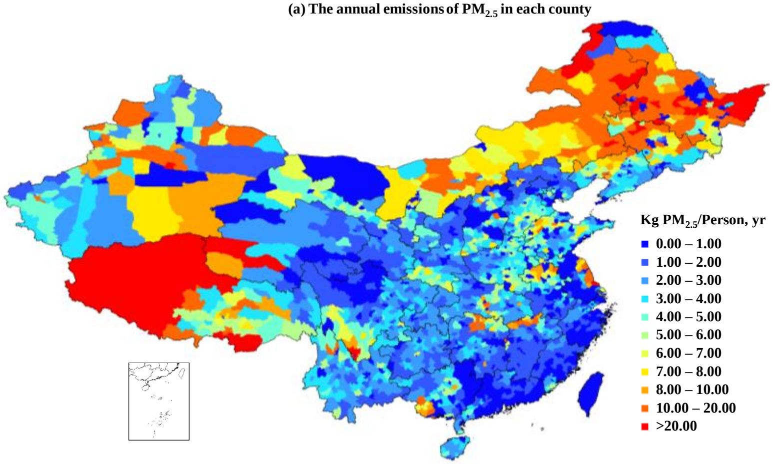
Note: The numbers 1–12 represent the pollutant of SO_2×10, NO_x, NMVOC, NH_3×10, EC×10, OC, CO/10, PM_{10}, $PM_{2.5}$, CO_2/100, CH_4, and Hg×1000000, respectively.



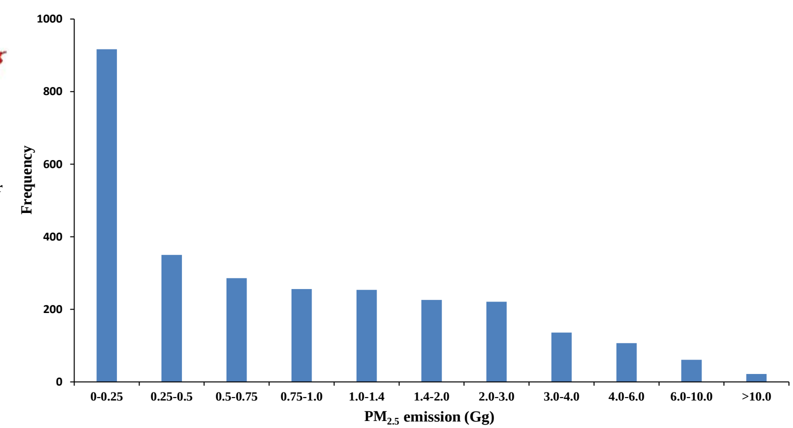
(a) The annual emissions of $PM_{2.5}$ in each county



(b) $PM_{2.5}$ emissions intensities per unit area



(c) $PM_{2.5}$ emissions intensities per capita



(d) The distribution of county level annual $PM_{2.5}$ emissions

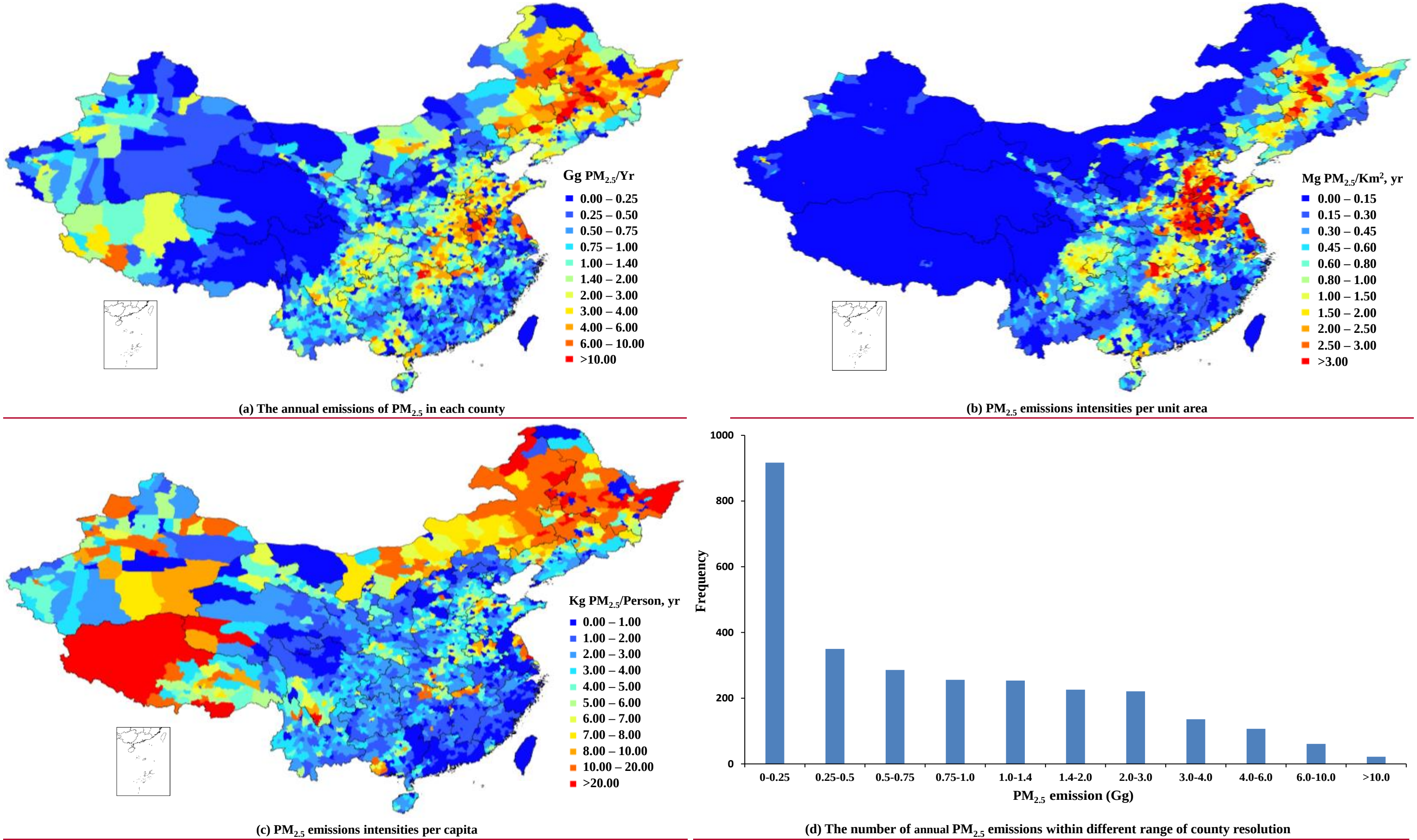


Figure 6. Biomass emission inventory at county resolution and intensity ($PM_{2.5}$).

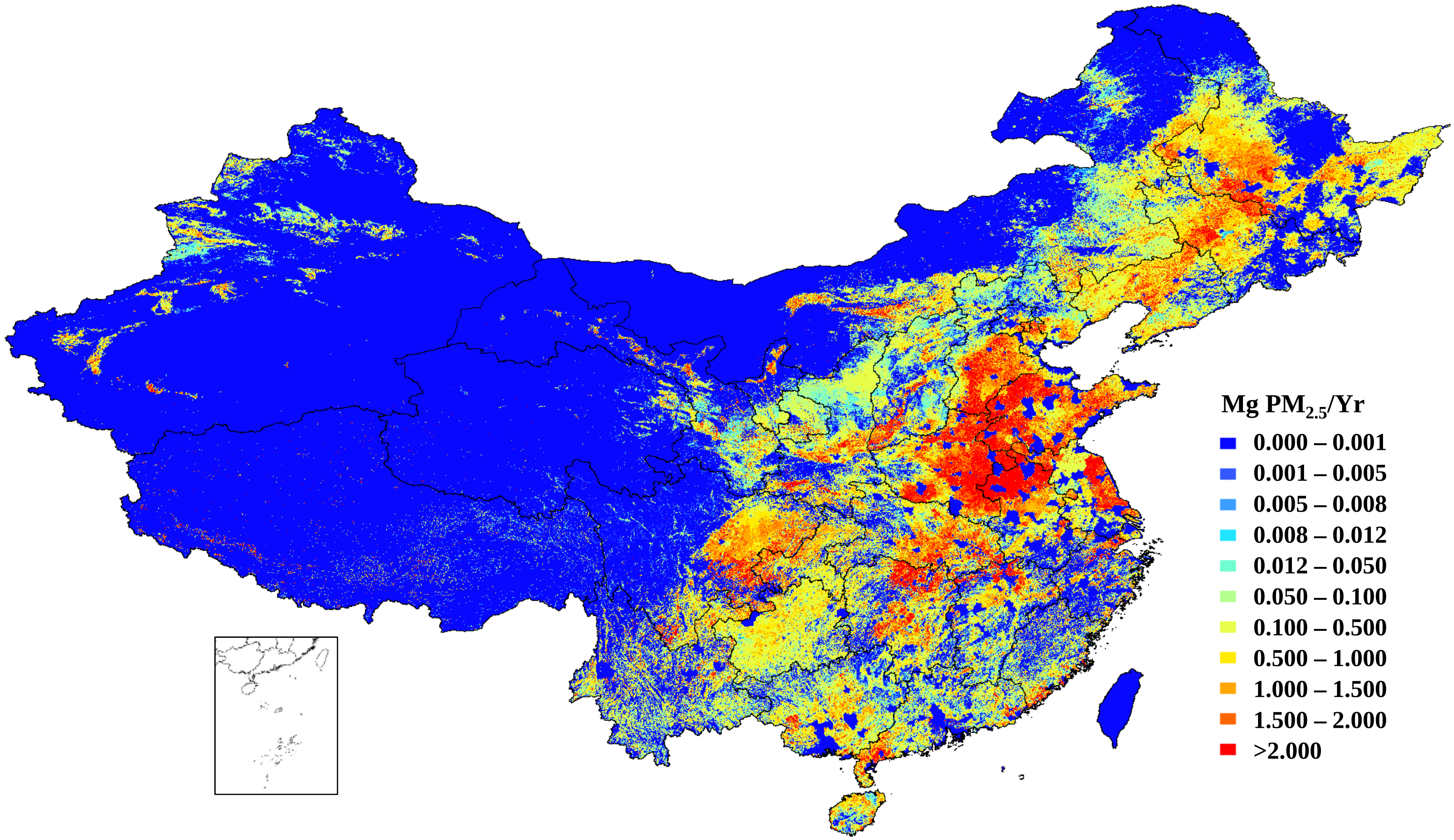
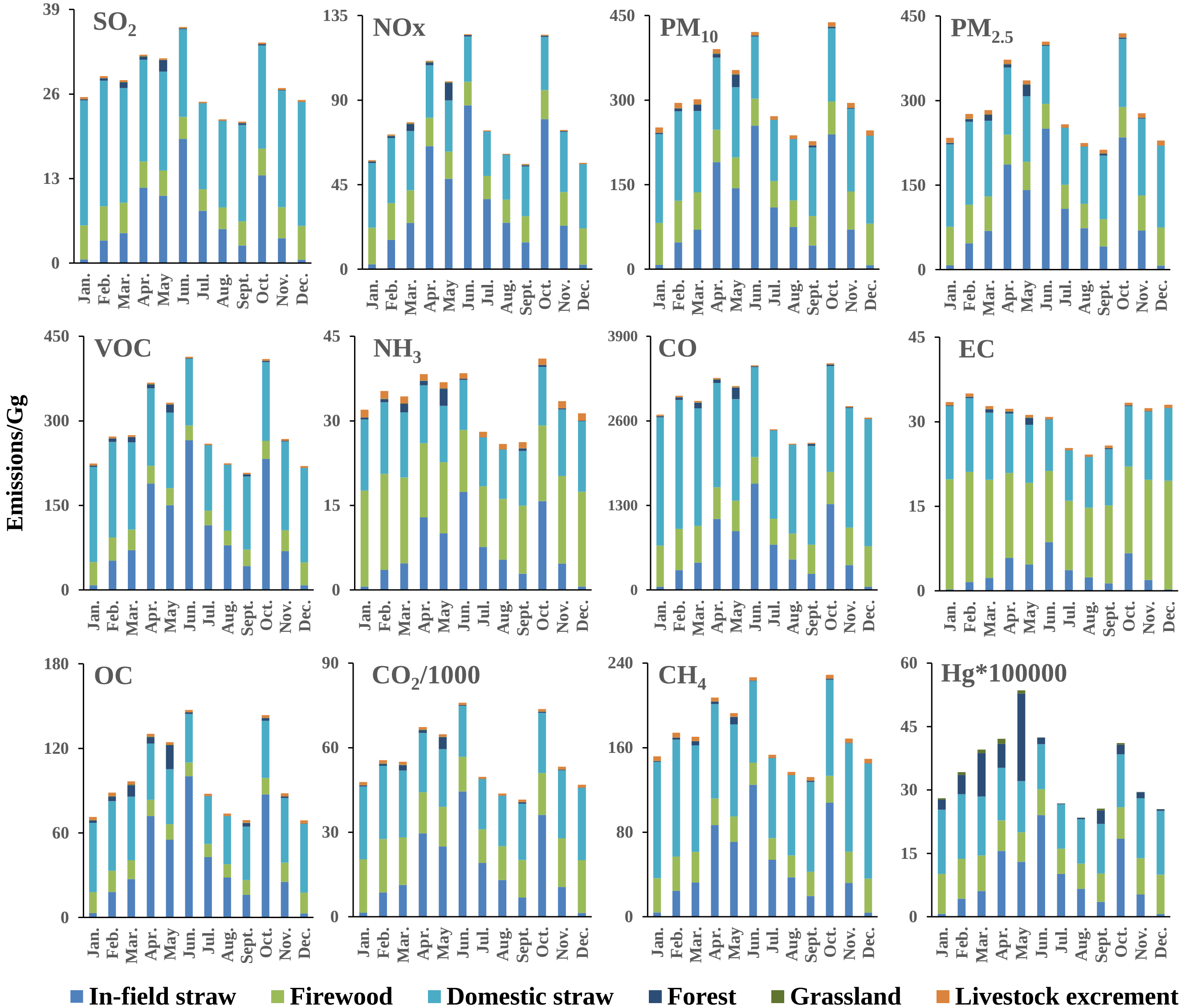


Figure 7. Gridded distribution of $PM_{2.5}$ annual emissions.



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

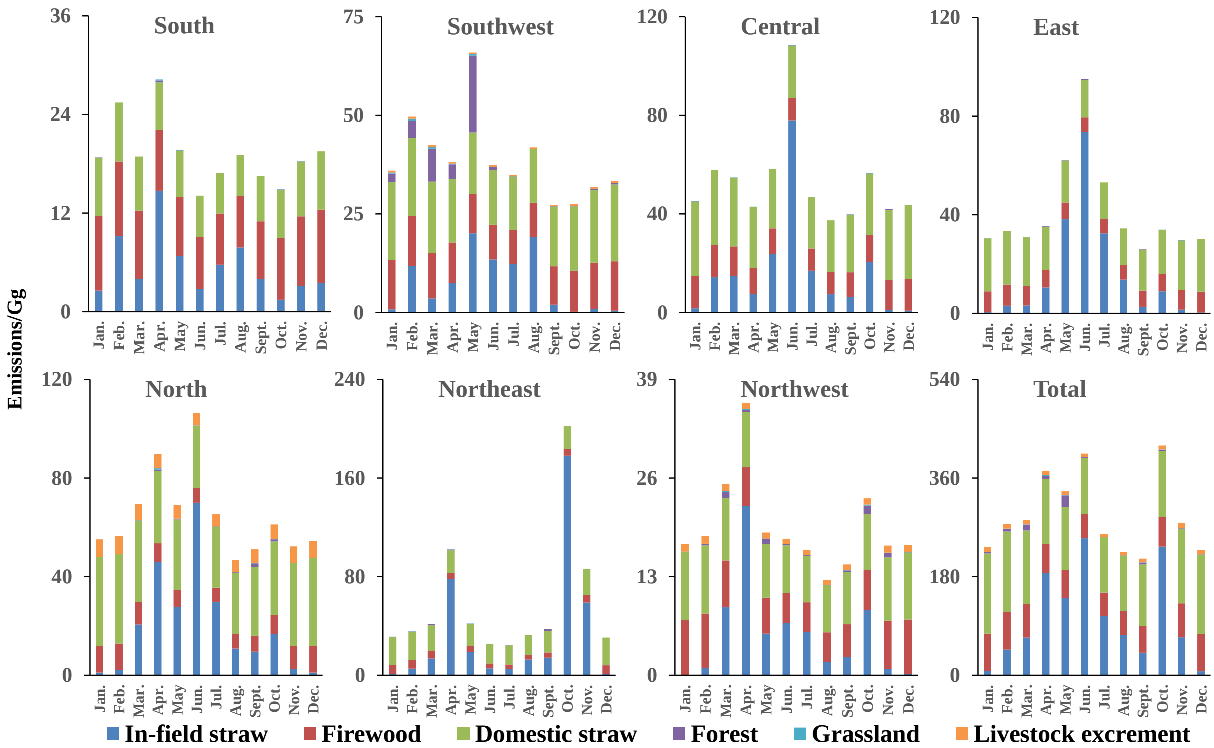


Figure 9. Monthly variation of different biomass sources emission for $PM_{2.5}$ emissions in different regions.

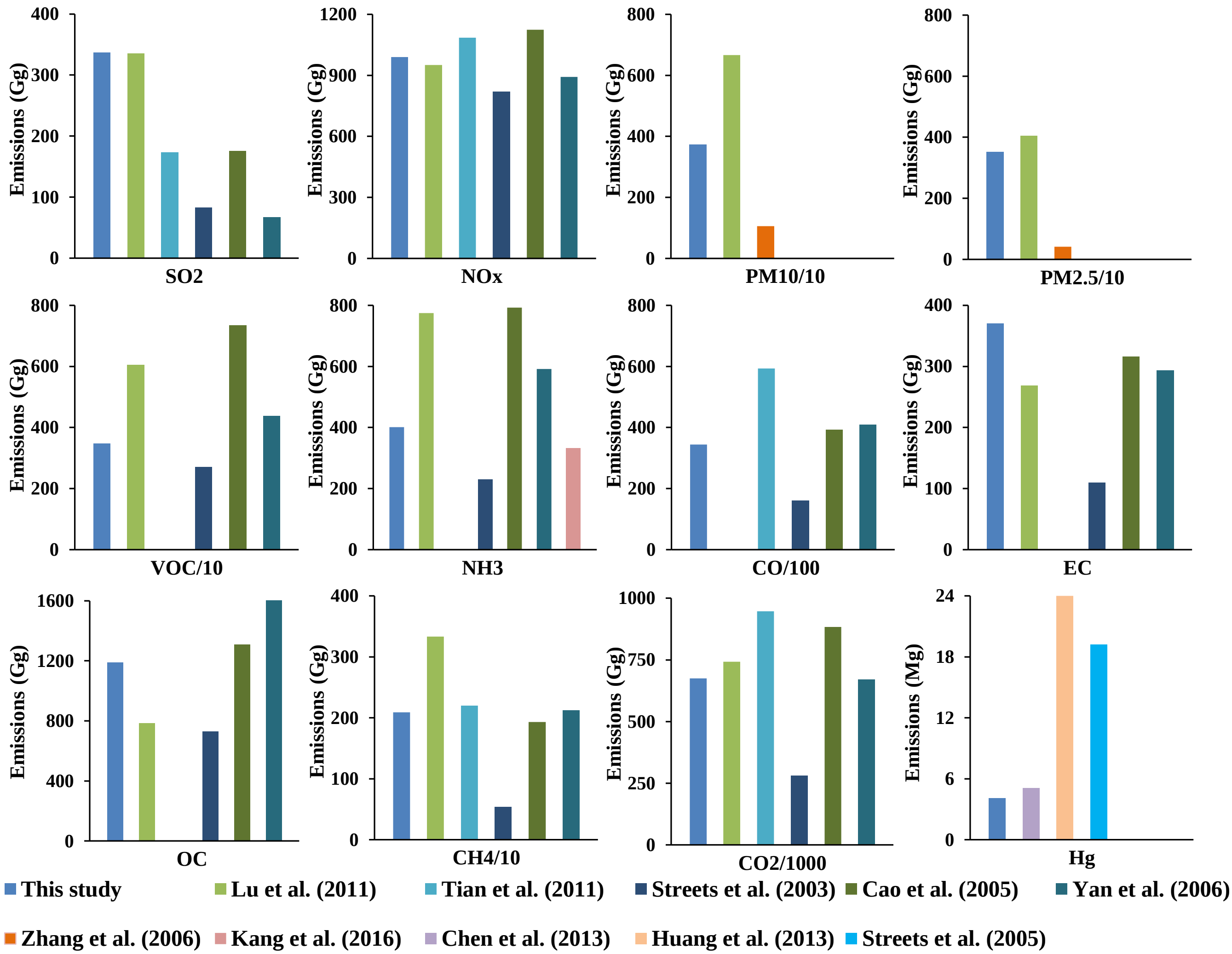


Figure 10. Comparison of the emissions inventory derived by this study with the emissions estimated by previous researches.

Supplement of

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

Ying Zhou [1,2], Xiaofan Xing [1,2], Jianlei Lang [1,2], Dongsheng Chen [1,2], Shuiyuan Cheng [1,2,3], Lin Wei[1,2], Xiao Wei [4], Chao Liu [5]

[1] Key Laboratory of Beijing on Regional Air Pollution Control, Beijing University of Technology, Beijing 100124, China

[2] College of Environmental & Energy Engineering, Beijing University of Technology, Beijing 100124, China

[3] Collaborative Innovation Center of Electric Vehicles, Beijing 100081, China

[4] Beijing Municipal Research Institute of Environmental Protection, Beijing 100037, China

[5] Environmental Meteorological Center of China Meteorological Administration, Beijing 100081, China

Correspondence to: Ying Zhou (y.zhou@bjut.edu.cn) and Shuiyuan Cheng (bjutpaper@gmail.com)

Supporting Information

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

Section S2: Figure S2 Maps 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 NMVOC 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.

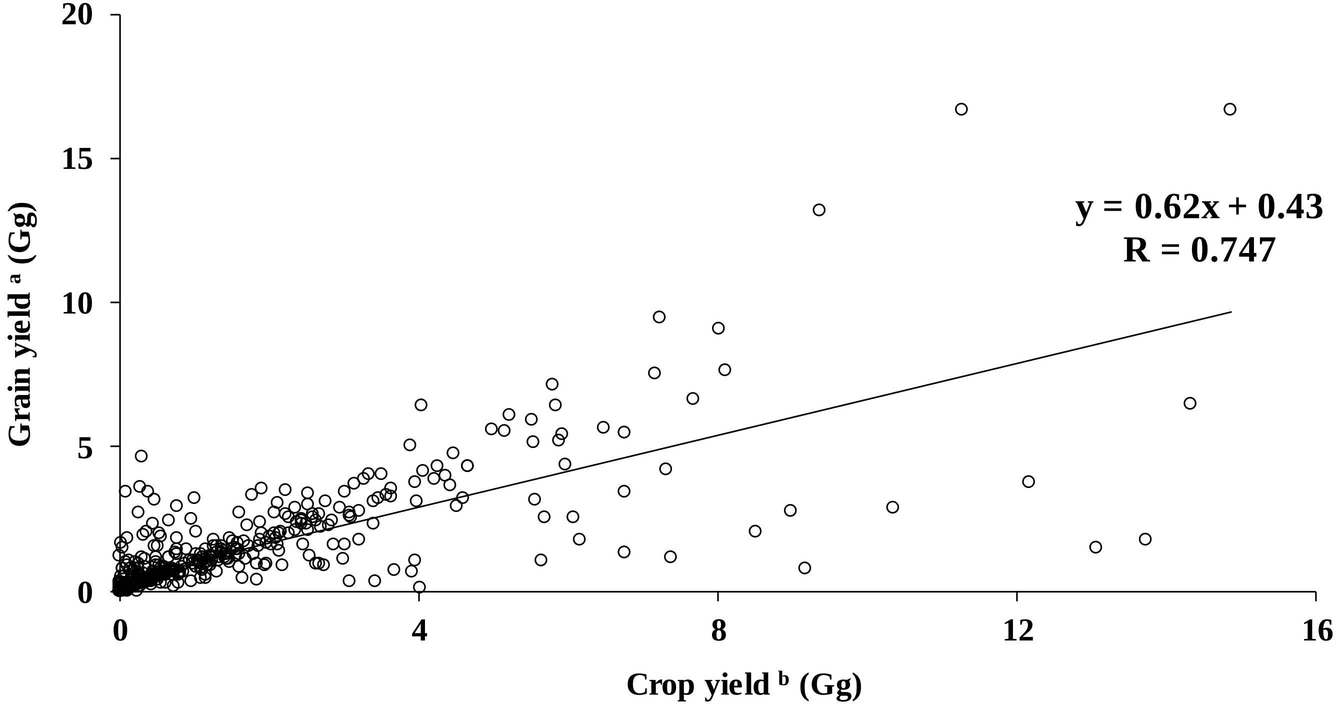
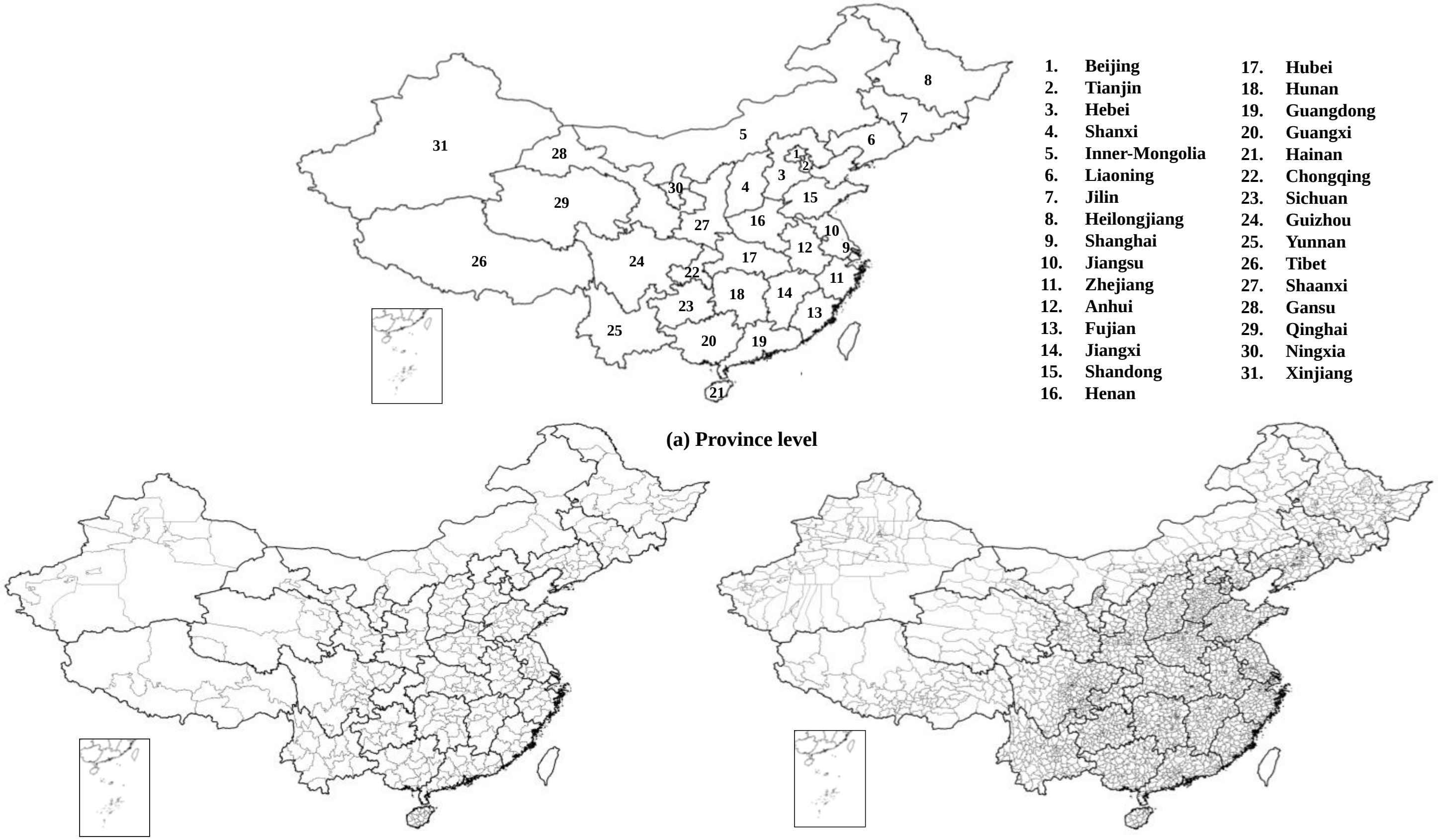


Figure S1 The correlation between crop yield and grain yield at prefecture resolution.

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

S2 Maps showing the prefecture and county resolution.



(a) Province level

(b) Prefecture level

(c) County level

Figure S2 Maps 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 domestic and in-field straw 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 launched 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 domestic burning was calculated using Eqs. (1) and (2), respectively, as follows:

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

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

where *m* is the *m*-th grid and *n* represents the *n*-th county; $E_{m\text{-}open}$ and $E_{n\text{-}open}$ represent the emissions of the *m*-th grid and *n*-th county for biomass open burning (in-field straw burning), respectively; $E_{m\text{-}domestic}$ and $E_{n\text{-}domestic}$ represent the emissions of the *m*-th grid and *n*-th county for biomass domestic 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.

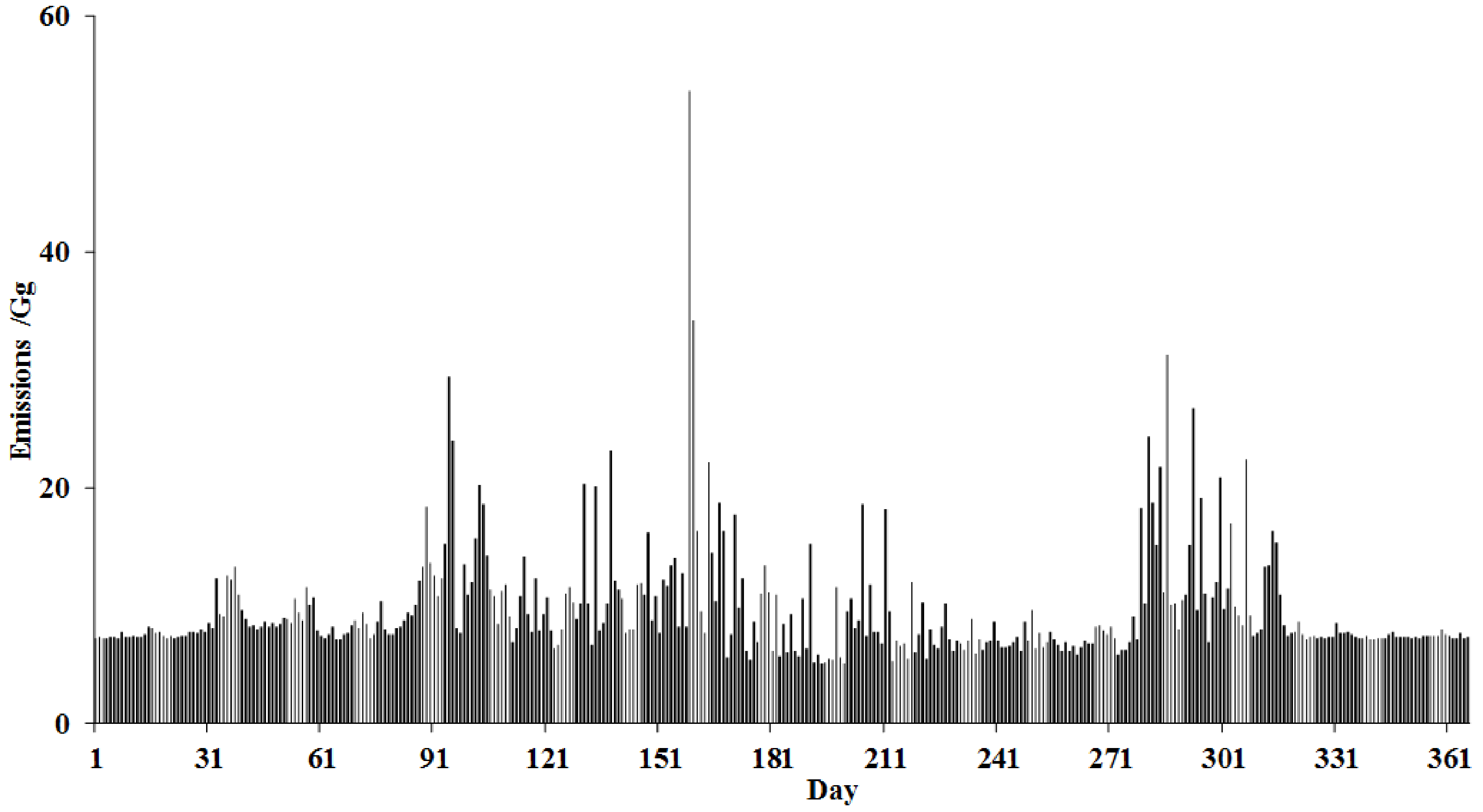


Figure S3 Daily $PM_{2.5}$ biomass burning emissions variation in 2012.

S6 Emission of $PM_{2.5}$ species from biomass burning

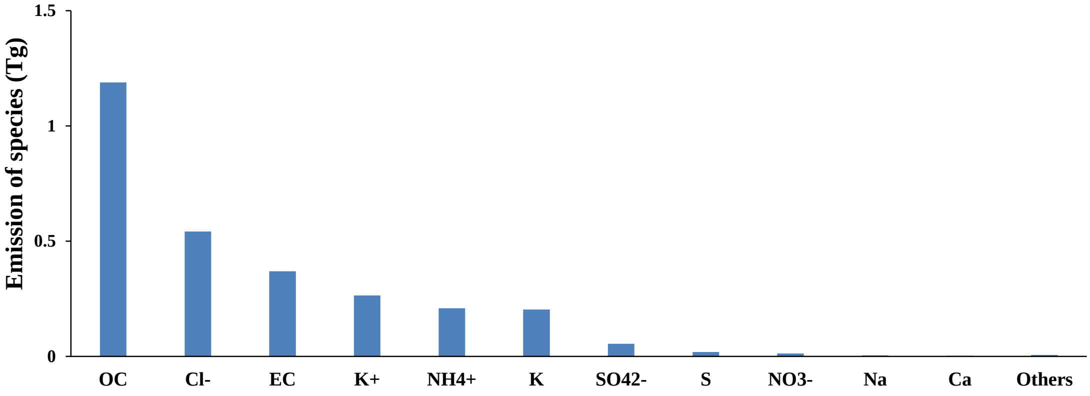


Figure S4 Emission of $PM_{2.5}$ species from biomass burning.

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 NMVOC species from biomass burning.

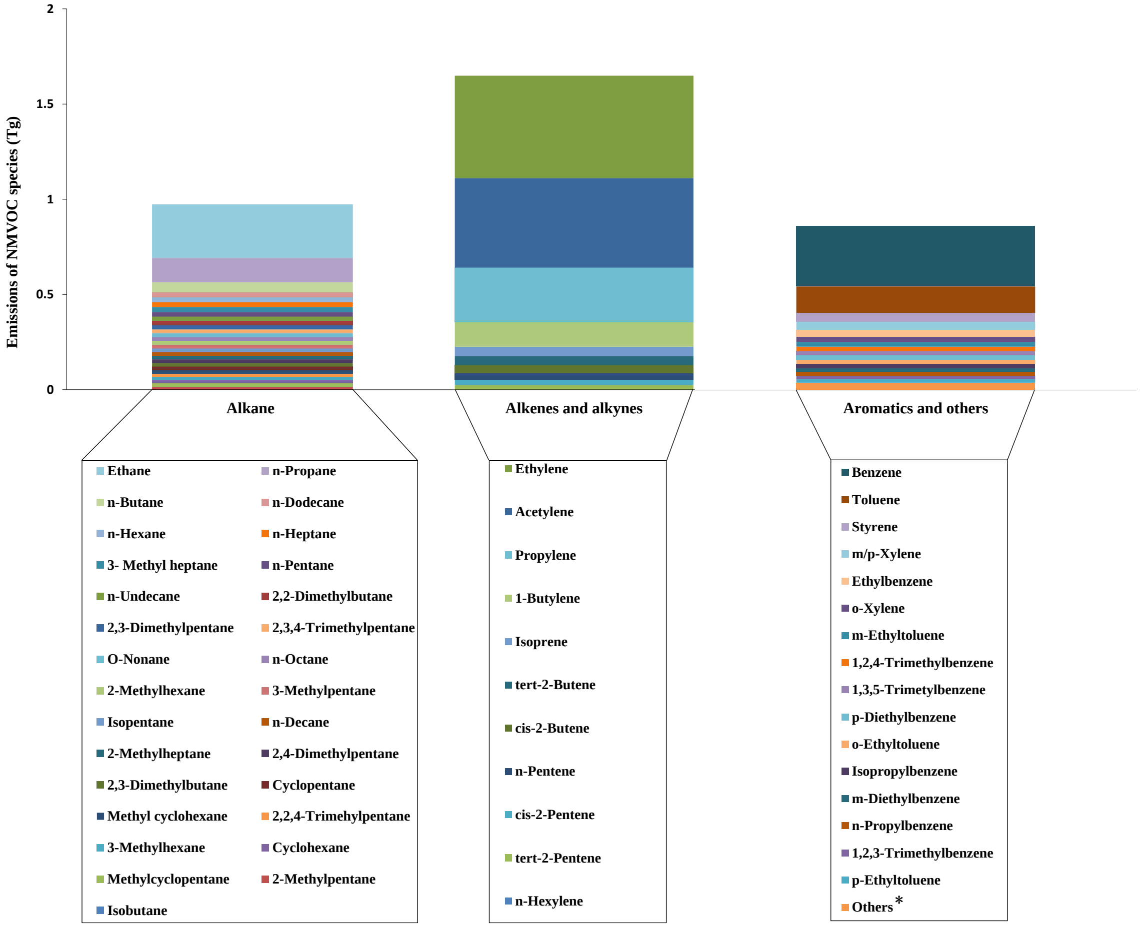


Figure S5 Emission of NMVOC species from biomass burning.

Note: *Species in others include aldehyde, ethers, alcohols, esters, ketone and acids.

S8 Table S1 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[*]

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 Table S2 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]

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]

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