Authors' Responses to Reviewer Comments

Manuscript: Estimating the open biomass burning emissions in Central and Eastern China from 2003 to 2015 based on satellite observation (Ref. No.: acp-2018-282)

We are very grateful for your careful and insightful comments, which contributed greatly to improve this manuscript. We carefully answered them point-by-point as below and corrected the corresponding parts in the manuscript (in red color).

This manuscript discussed the open biomass burning emissions in Central and Eastern China including several provinces with different vegetation and also the emissions in different years and seasons. The authors also estimated the pollutions from open biomass burning in this area from 2003 to 2015. The spatial and temporal distribution of open biomass burning provides a high resolution result to relevant researchers and could be meaningful in policy making. And there are some technical questions that need the authors to clarify (see additional comments).

Response:

Thanks a lot for your positive comments on this manuscript. We have improved this manuscript by answering the reviewers' comments and advice.

Additional comments:

1. Line 27: The initialism needs to be explained for the first use, i.e. OC, EC and NMVOC.

Response:

Thanks for this suggestion. We have corrected it in the manuscript (in Line 30-32): "...organic carbon (OC), elemental carbon (EC), methane (CH_4), nitric oxide (NO_X), non-methane volatile organic compounds (NMVOCs), sulfur dioxide (SO_2), ammonia (NH_3), carbon monoxide (CO), carbon dioxide (CO_2) and fine particles ($PM_{2.5}$)..."

2. Line 100-101: "". Please review the structure of this sentence.

Response:

We have polished it as shown in Line 106-107.

"One is the burned area product, which provides fire burned areas of the whole month. It is limited by the lower pixel resolution."

3. Section 2.3: Although the author set different CE values for different vegetation, the CE was set as a constant during each open burning process. Please discuss about it and provide a reasonable explanation, because the CE should not be a constant during burning and the pollution emissions were not uniform in different phase.

Response:

We really appreciate for this comment. We highly agree with you that the CE should not be a constant during burning and the pollution emissions were not uniform in different burning phase. However, the emission inventories in this research and currently published papers were estimated for a long time period or a whole year with the time scale as month, instead of hour. Therefore, the "CE" values used here reflected the average biomass burning condition for the whole open burning processes, which can not reflect the different combustion phase (like smoldering and flaming, etc.).

It is the research hotspot in developing emission inventory with high time resolution, which needs both the high time-resolution activity data and emission factors for different burning stages. It is also now being considered in our research group.

We accepted this suggestion and added corresponding discussion in the manuscript (Line 212-219).

4. Line 219-221: The contributions presented in this part were with different significant figures or decimal digits. Please explain why you use different significant figures or decimal digits for different contributors?

Response:

Thank you very much for this comment. We are truly sorry for the confusion in the use of different significant figures or decimal digits. We have made the revision in the manuscript, by using the same decimal digits for different contributors (Line 254-256).

5. Line 257: Please explain the increasing trend of OBB emission from 2003 to 2008. It seems that the explanation in Line 269-280 was not convincing enough.

Response:

Thanks for this suggestion. In Figure 6, we found that the emissions of $PM_{2.5}$ from crop burning significantly increased from 2003 (228 Gg) to 2008 (294 Gg), due to the increase of crops production and deficiency of strict control policies in this period (Table S1). Emissions from forest, shrubland and grassland burning exhibited an obvious declining trend from 2003 to 2006 and then increased from 2006 to 2008, which maybe related with the different weather conditions and human forestry activities. Although emissions from forest, shrubland and grassland burning fluctuated markedly during this period, the obvious increase of crops residue burning dominated the total growth of OBB emission from 2003 to 2008.

Corresponding revision and discussion has been added in the manuscript (Line 323-327).

6. Line 452-460: Please specify how much do your research improve the uncertainties in OBB emission and contribution estimation.

Response:

Thanks for this query. In this study, we improve the uncertainty estimation method based on reliable multiple satellites data. In addition, the uncertainties of active data sets (biomass loading data and local EFs) were improved. Multiple satellites can better obtain burned area data; different active data were more suitable because they could better reflect the actual situation in Central and Eastern China. Meanwhile, the field survey also helped to improve the uncertainty and reliability of our emission results. Through these improvements, the uncertainty of pollutant emission was improved.

As shown in Table 1, the uncertainty ranges of different pollutant emissions were narrowed.

Table 1. Uncertainty ranges of different pollutants in emission estimation.

Pollutant	Uncertainty ranges		
	Our study	Wang et al., 2008	Qiu et al., 2016
OC	±30%	±148%	-72% - 213%
EC	±48%	±132%	-67% - 204%
CH_4	±20%	±77%	-32% - 81%
NO_X	±20%	±80%	-36% - 92%
NMVOCs	±45%	±71%	-24% - 78%
SO_2	±45%	±108%	-47% - 121%
NH_3	±35%	±137%	-61% - 152%
CO	±18%	±86%	-52% - 105%
CO_2	±3%	±60%	
$PM_{2.5}$	±36%	±142%	-77% - 213%

Response

Wang, S. X. and Zhang, C. Y.: Spatial and Temporal Distribution of Air Pollutant Emissions from Open

Burning of Crop Residues in China, Science paper Online, 3, 329–333, 2008 (in Chinese).

Qiu, X., Duan, L., Chai, F., Wang, S., Yu, Q. and Wang, S.: Deriving High-Resolution Emission Inventory of OBB in China based on Satellite Observations, Environ. Sci. Technol., 50(21), 11779-11786, doi:10.1021/acs.est.6b02705, 2016.

Authors' Responses to Reviewer Comments

Manuscript: Estimating the open biomass burning emissions in Central and Eastern China from 2003 to 2015 based on satellite observation (Ref. No.: acp-2018-282).

Thanks a lot for your positive and constructive comments and suggestions. We carefully checked the issues and answered them point-by-point as below. Corresponding corrections were done in the manuscript (in red color).

General comment:

The uncertainty discussion in Sect. 3.5 can be improved. Presumably for the Monte Carlo simulations, some uncertainty range had to be selected for each individual variable involved in the emissions calculation, but it is not clear what this range is for some variables, such as for example burned area. Also, the authors do not explain what is it that makes specific pollutants (e.g. EC) have larger uncertainty and others smaller? Which of the variables needed for calculating emissions have the biggest role in driving uncertainty? This will help focus future efforts in order to reduce uncertainty in emissions in this area. Finally, when the authors say that "Compared with previous studies, for the emission estimation of forest burning, the uncertainty was improved" or "For cropland, the uncertainty was improved. . .", do they mean that the other studies estimated uncertainty ranges which were wider, or that the datasets used here are better/more suitable?

The use of English can be polished throughout the manuscript. Not that the language has major problems - more or less it is fine. But almost in every sentence (or every other sentence) I found myself thinking that the choice of words, grammar or syntax could be improved. I have made some suggestions, but there are more places where things can be improved.

General response:

We really appreciate for your constructive suggestions and comments, which will be helpful for improving this manuscript. We are regretful for the unclear description. The following is an explanation for the parameters used in Monte Carlo simulation.

We assess the emission uncertainty of specific pollutants by the Monte Carlo simulation, which was widely used in uncertainty estimation in previous studies (e.g., Streets et al., 2003; Li et al., 2016). The variables involved in emission calculation included burned area, biomass loading data, combustion efficiency and emission factors. According to previous researches, the estimation of fires burned area was proved to be reliable by the adoption of burned area product MCD64AL (Giglio et al., 2013) and active fire product MCD14ML (Randerson et al., 2012). It is difficult to assess the uncertainty of the satellite-derived data for the burned land area (Hoelzemann et al., 2004, Chang et al., 2010). Although some active fires which burned out at 10:30 am-1:30 pm each day could not be captured by MCD14ML, the burned area used in this study were more reliable due to the combination of multiple satellite datasets (MCD64AL and MCD14ML). The uncertainties in this study were mainly caused by biomass loading data, combustion efficiency and emission factors. These data were assumed to be normal distribution (Zhao et al., 2011). The uncertainty of biomass loading data and combustion efficiency was estimated with a standard deviation of approximately 50% of the mean value (Shi et al., 2015). The uncertainty for emission factors was cited from references as listed in Table S5, with the uncertainty for each pollutant mainly ranged from 0.03 to 0.85. The reliable of emission factors play the most important role in driving uncertainty. Considering all these parameters, 20,000 Monte Carlo simulations were performed to evaluate the estimation uncertainty quantitatively for pollutant emissions with 95% coincidence level. We have made corresponding revision in the manuscript (Line 512-526). In addition, Table S5 has also been added in the supplementary file.

Compared with previous studies, the uncertainties were improved in our study due to the datasets used here were better and more suitable: multiple satellites can obtain more reliable and detailed burned area data. Different active data (biomass loading data and local EFs) involved in emission estimation were more suitable because they could better reflect the actual situation in the research region. Overall, the uncertainty ranges of different pollutant emissions were narrowed and more reliable, which could better reflect the real emission.

In addition, we have improved the choice of words, grammar or syntax in the revised manuscript. All the authors have revised the manuscript for another time independently.

We have carefully taken the reviewer's suggestion into consideration during the revision of our paper. Please see the following point-by-point responses.

Specific Comments:

Line 19: Please change “few focus” to “little focus”.

Response:

It has been corrected (in Line 19).

Line 33: Is "per pixel" informative for the average reader?

Response:

Thanks for this query. In this study, all the data were relocated into a 1 km×1 km grid to identify and estimate spatial variation of open biomass burning emission. “per pixel” in line 33 was the same as 1 km×1 km. It has been changed into “per square kilometers” (Line 37-38).

Line 35: Please change “for” to “from”.

Response:

It has been deleted (Line 40).

Lines 41-43: I think that these sentences would be better suited for the beginning of the abstract, rather than the end.

Response:

We accepted this suggestion. The sentences have been moved in Line 23-26.

Line 84:“accurate”-“accurately”

Response:

It has been corrected (Line 90).

Line 114: “the same emissions factors”: The same to what?

Response:

Thank you very much for this comment. We are truly sorry for the unclear description. It means the same emission factors for different pollutants emitted from open biomass burning. We have made the corresponding revision in the manuscript (Line 121).

“... have used the same emission factors for pollutants emitted from OBB without considering ...”

Line 119: "historical" might be too strong a word here. Suggest using "of recent years" or something similar.

Response:

Thanks for your suggestion. We have corrected the sentence as *"…estimate multi-year OBB emissions from 2003 to 2015…"* (Line 126)

Line 131: Shouldn't "E" also be indexed with x and t?

Response:

Sorry for this error. We have changed the "E_i" to "$E_{i,x,t}$" in the manuscript. In addition, the description of "$E_{i,x,t}$" have been changed to "emission amount of different pollutants in location x and time t" in the revised manuscript (Line 139-140).

Line 134: Mention explicitly that "CE" stands for combustion efficiency. Also, CE does not have a subscript x in the equation, whereas it does in this line.

Response:

We have changed the "CE" into "CE_x". The description of "CE_x" have been changed to "the combustion efficiency of open biomass burning in location x" in the manuscript (Line 141).

Line 147: Is v the same as j in the earlier equation? Then why change the symbol? Same lower down for r and s.

Response:

Sorry for the confusing description. In fact, "v" in the equation is the same as "j" in the earlier equation. We have made the corresponding revision in the manuscript (Line 162).

Considering the difference of definitions, "r" and "s" are not replaced by "i" and "t" in equation 2 in the manuscript.

Line 148: Not clear (to me at least) what "outside of the burned area" means here. I think generally Equation 2 would benefit from a somewhat clearer explanation for the readers not familiar with the technical details of the products.

Response:

Thanks for this suggestion.

In this study, we re-sampled the two fire products data into a 1 km × 1 km grid. The total burned area in each grid cell was estimated by the following equation (Randerson et al., 2012).

$$BA_{total(i,t,j)} = BA_{MCD64AL(i,t,j)} + BA_{sf(i,t,j)} \quad (1)$$

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

$BA_{MCD64AL(i,t,v)}$ was directly detected from MCD64AL product. MCD14ML active fire points in each grid included two parts: active fires points with or near the MCD64A1 burned area (FC_{in}) and active fires outside the MCD64AL burning area (FC_{out}). $BA_{sf(i,t,j)}$ was the burned area of FC_{out}. The $BA_{sf(i,t,j)}$ was used as supplement.

MCD14ML data could only give the fire points data, but not for the burned area data. Therefore, we used equation 2 to calculate $BA_{sf(i,t,j)}$:

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

Where $BA_{sf(i,t,j)}$ is the small fire burned area of FC_{out} in grid cell i, month t, and aggregated vegetation class j; $FC_{out(i,t,j)}$ is the MCD14 ML active fires outside of the burned area in grid cell i, month t and aggregated vegetation class j; $\alpha_{(r,s,j)}$ and $\gamma_{(r,s,j)}$ are set as coefficients of burned area calculation in burning region r and burning period s, for biomass species j. α is allocated as the ratio of $BA_{MCD64A1}$ to FC_{in} in each grid. It had units of km^2 per active fire and was used to estimate the burned states of FC_{in} in each grid cell. γ is an additional unit less scalar which is used to estimate the difference between F_{in} and F_{out}. According to Randerson et al. (2012), γ is assumed equal to 1 in China.

We have made the revision correspondingly in the manuscript (Line 150-158).

Lines 154-158: So, is the biomass basically a step function? Is this justifiable? Is this expected to generate any artifacts in the results?

Response:

Thanks for this query. It is not just a step function.

The forest biomass density data was defined as the ratio of the total biomass to the total area of different forest species (broadleaf forest, coniferous forest and mixed forest):

$$B_{i,r}=T_{i,r}/A_{i,r} \quad (3)$$

where i stands for different forest species; r means different provinces; $B_{i,r}$ is the biomass density of forest species i in province r; $T_{i,r}$ means the total biomass of forest specie i in province r; $A_{i,r}$ denotes the total area of forest species i in province r.

To obtain $B_{i,r}$, we need to calculated $T_{i,r}$ and $A_{i,r}$, respectively.

$A_{i,r}$ was collected from the 8th Chinese National Forest Resource Inventory (Xu, 2014).

Since different forest species are consist of various tree types, we obtain $T_{i,r}$ by calculating the biomass of each tree types (Fang et al., 1996):

$$T_{i,r} = \sum_{j=1}^{n} E_{j,r} \quad (4)$$

where j stands for different tree types of forest specie i; $E_{j,r}$ is the biomass of tree type j in province r.

$E_{j,r}$ was calculated based on the forest stock volume as following (Fang et al., 1996):

$$E_{j,r} = aV_{j,r} + b \quad (5)$$

Where $E_{j,r}$ is the biomass of tree type j in province r; $V_{j,r}$ indicates the forest stock volume (m^{-3}) of tree type j in province r; a and b were set as correlation coefficient.

“a” and “b” for different tree types were derived from previous studies (Fang et al., 1996; Tian et al., 2011; Lu et al., 2012; Li et al., 2014; Wang et al., 2014; Wen et al., 2014) (Table 2 in manuscript), $V_{j,r}$ was collected from the 8th Chinese National Forest Resource Inventory.

In our study, the correlation coefficient “a” and “b”, the calculated method and the 8th Chinese National Forest Resource Inventory were all proved to be reliable (Fang et al., 1996; Lu et al., 2012). Therefore, the result of the biomass data in our study is justifiable. However, the correlation coefficient “a” and “b” in current studies have not shown the difference in the same tree species of each region, which may cause uncertainty in results. We will improve this in our future research.

Sorry for the unclear description in the original manuscript. We have made the revision correspondingly in the manuscript (Line 182-187).

Lines 188-189: What does “similar” mean here?

Response:

Sorry for the confusing sentence. "similar" in line 188-189 means researches which used emission factors (EFs) of cropland, forest, shrubland and grassland in China or foreign regions. We added these EFs in our calculation due to the shortage of EFs for some crop species burning in CEC and the shortage of EFs for pollutants emitted from forest, grassland and shrubland burning in China. To avoid misleading information, we have changed "similar research" to "previous researches" in the manuscript (Line 224).

Line 190: Is there a reference for this previous research?

Response:

Thanks for this comment. Emission factors of OC and EC for corn straw burning were derived from our experimental data which have not been published now (as shown in following Table 1). The corn straw is from Nanjing, which is one of the most important cities in CEC.

Meanwhile, we are so sorry for a mistake in Table 5. Table 5 in the original manuscript was a temporary table due to we tried to use our own EFs of OC and EC for forest fire burning in CEC. The OC (2.6 g/kg) and EC (0.11 g/kg) of coniferous forest were measured for pine wood and the OC (1.1 g/kg) and EC (0.31 g/kg) of broadleaf forest were measured for cotton wood (as shown in following Table 1). However, we found that the open wood burning in our experimental were not truly equal to forest fire. Therefore, we choose the emission factors of forest fire from Akagi et al. (2011) in our emission estimation. Emission factors and emission results for forest fire in our study were corresponding to Akagi et al. (2011), instead of our own data. We forget to update Table 5 in the original manuscript until we found this problem recently. We are so sorry for this mistake. We have made the revision in the manuscript (Table 5).

Table 1. The original experimental data for EFs calculating

Types	fuel consumption/kg	dilution ratio	Particle size/μm	Sampling flow	The number of sampling film	OC (ug)	EC (ug)
Pine wood	0.325	60	9.0~10.0	28.3	XQ116	52.2496	1.5072
		60	5.8~9.0		XQ117	31.6512	1.5072
		60	4.7~5.8		XQ118	45.7184	1.5072
		60	3.3~4.7		XQ119	51.2448	1.5072
		60	2.1~3.3		XQ120	53.7568	1.5072
		60	1.1~2.1		XQ121	87.92	1.5072
		60	0.65~1.1		XQ122	396.896	2.0096
		60	0.43~0.65		XQ123	632.0192	1.5072
		60	≤0.43		XQ124	693.312	1.5072
Cotton wood	0.31	60	9.0~10.0	28.3	XQ125	40.6944	1.5072
		60	5.8~9.0		XQ126	19.0912	1.5072
		60	4.7~5.8		XQ127	21.1008	1.5072
		60	3.3~4.7		XQ128	33.6608	1.5072
		60	2.1~3.3		XQ129	21.1008	1.5072

		60	1.1~2.1		XQ130	46.2208	7.536
		60	0.65~1.1		XQ131	37.1776	19.0912
		60	0.43~0.65		XQ132	80.384	16.0768
		60	≤0.43		XQ133	485.3184	118.064
Corn straw	0.31	60	9.0~10.0	28.3	XQ143	67.824	1.5072
		60	5.8~9.0		XQ144	40.192	1.5072
		60	4.7~5.8		XQ145	36.1728	1.5072
		60	3.3~4.7		XQ146	47.2256	1.5072
		60	2.1~3.3		XQ147	36.1728	1.5072
		60	1.1~2.1		XQ148	39.1872	1.5072
		60	0.65~1.1		XQ149	67.3216	4.5216
		60	0.43~0.65		XQ150	99.4752	1.5072
		60	≤0.43		XQ151	601.8752	59.7856

Line 193:“opening”-“open”

Response:

It has been corrected (Line 228).

Line 202: Maybe the title should be “Other factors influencing OBB emission”?

Response:

Thanks for your suggestion. It has been corrected (Line 237).

Line 209:“20, 000”-“20,000”

Response:

It has been corrected (Line 244).

Lines 214-215: It should be made clear somewhere that $PM_{2.5}$ and OC/EC are not totally different things, i.e. PM will include some OC and EC.

Response:

Thank you very much for this query. We are really sorry for the mistake. It is really a clerical error. Emission of $PM_{2.5}$ was 4.13×10^{3} Gg instead of 4.13×10^{2} Gg. It has been corrected in the text (Line 251).

Line 217: Suggest removing “section”.

Response:

It has been deleted (Line 251).

Line 240:“This is mostly”: Which? For example, what are the “suitable weather conditions” in this case?

Response:

Thank you very much for this query. “This” means the relative high contributions of forest and shrubland fire burning emissions in southern provinces. Corresponding correction has been done in the text as following (Line 275-277).

"The relative high emission contributions of forest and shrubland fire burning in the southern provinces can be explained by the large forest and shrubland coverage, frequent human forestry activities, low precipitation and dry weather in spring and winter (Cao et al., 2015), which may easily lead to forest and shurbland fires."

Figure 6:"opening"-"open"

Response:

It has been corrected.

Lines 255-259: There is confusion between trends and short-term (interannual) variations here. Can the authors perhaps provide the overall trend for each species for the whole period, and whether it is statistically significant?

Response:

Thanks for this query. The variation tendency of different pollutants and different biomass types were shown in following Figure 1. For different pollutant species, we found that the emissions have similar increasing tendency (R^2 higher than 0.32, $P<0.05$). For different biomass types, cropland fire burning emission have a significant increasing tendency ($R^2=0.87$, $P<0.01$), while no significantly variation trend was observed for forest, shrubland and grassland fire burning emissions in research period ($P>0.05$).

On the process of data analysis, we think discussing the interannual variation is more meaningful than listing the changing tendency during the whole research period. It could be found that the increase of crop residue burning dominated the significant growth of OBB emission from 2003 to 2008. Then with the adoption of strict control policies (Table S1 in Supplementary file), the growth of crops residue burning emission gradually slow down. Meanwhile, the forest, shrubland and grassland fire burning were mainly affected by weather conditions and human activities as discussed above. Their emissions were difficult to predict and control, and exhibited random yearly variation of pollutant emission. Therefore, we initially discussed the mulit-year variation during 2003-2015 instead of the overall trend for the whole period in this study.

We adopted the suggestion and added these discussions in the text (Line 296-301). And the following Figure 1 was also added in the supplementary file (Figure S3).

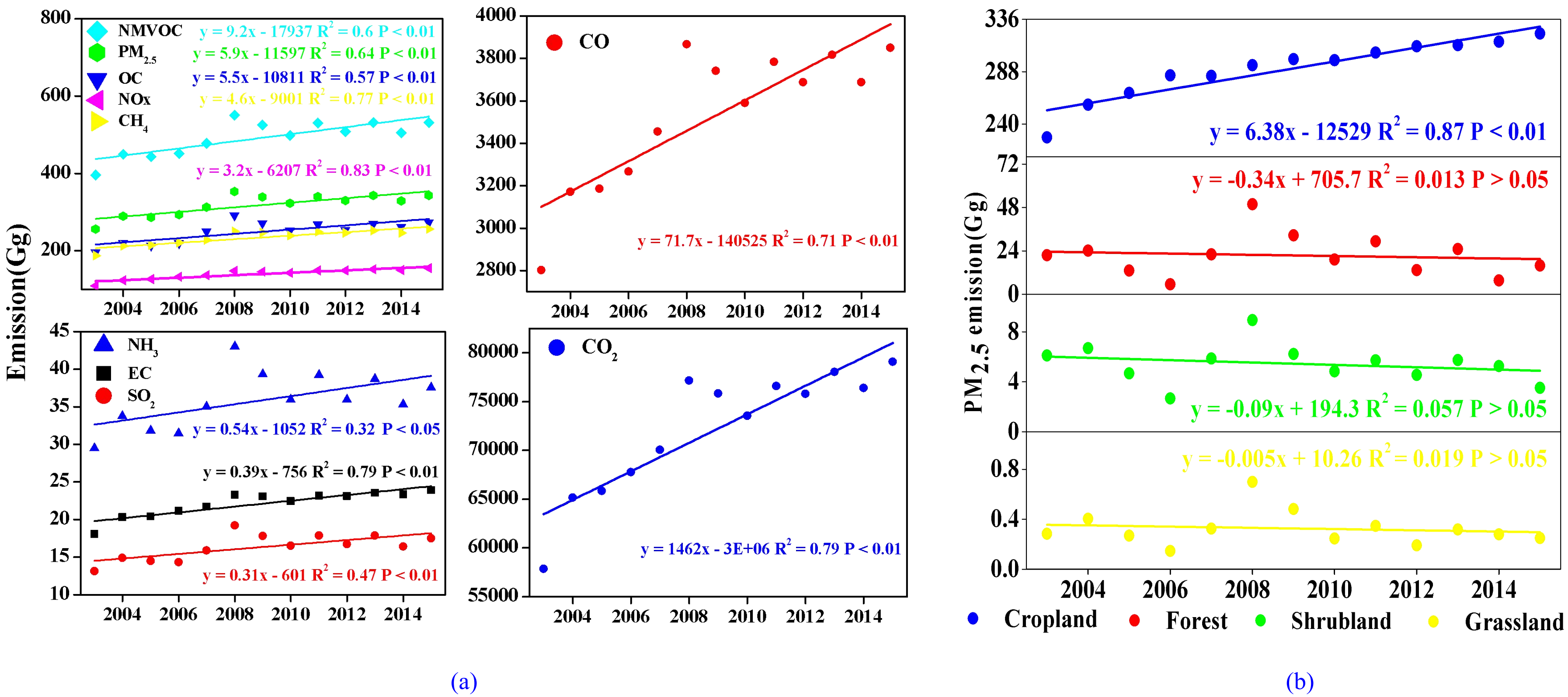

Figure 1. The changing tendency of different pollutant emissions (a) and $PM_{2.5}$ emission from different biomass types (b) during 2003 to 2015.

Line 265: Presumably it is just coincidental that they are odd and even years. Therefore maybe not worth the emphasis?

Response:

Thanks for this query. Sorry for this unclear description. As discussed above, the forestry fire burning was affected by environmental conditions and human activities with environmental factors having a larger impact (Chen et al., 2013), so their emissions are random. Initially, it is just a general description (Line 312-313). We have corrected the sentences as *"with higher emission in 2011, 2013 and 2015 and lower emission in 2012 and 2014."*

Line 280: Increased or decreased?

Response:

Sorry for this mistake, it has been corrected as *"...increasing trend from 2003 to 2015, by about 21%-29%."* (Line 334).

Line 288:"scattering"-"scattered"

Response:

It has been corrected (Line 343).

Line 315:"totally occupied. . .": Please improve phrasing.

Response:

It has been corrected as following:

"However, the emissions of $PM_{2.5}$ from forest, shrubland and grassland burning achieved peak values from December to March, being 67% of that in 2003-2015." (Line 372-373)

Line 318:"based on the correlation of emissions in each month"-"based on the correlation between their monthly emissions"

Response:

It has been corrected (Line 378).

Line 328:“occurred”-“occurring”

Response:

It has been corrected (Line 389).

Line 345:“were found”-“featured”

Response:

It has been corrected (Line 407).

Lines 348-349:“are uniformed”-“remain similar”

Response:

It has been corrected (Line 409-410).

Line 378: May need to improve terminology a bit. For example, why is “local burning habits” not part of “anthropogenic activities”? What is the distinction?

Response:

Thanks for this query. To our knowledge, “burning habit” lead to anthropogenic activity, which is not anthropogenic activity itself. Sweep graves in tomb-sweeping day and celebration in Spring Festival are anthropogenic activities, which caused by social customs. In order to avoid unclear clarification, we change “some anthropogenic activities” to “social customs” through the full text in the revised manuscript.

Line 380: “People sweep their graves”: The authors may want to change the wording as “their” might be a bit misleading here...

Response:

Thank you very much for your comment. We are truly sorry for the misunderstanding. We have made the revision in revised manuscript (Line 441).

“People sweep graves and burn sacrifices by ignited straw...”

Line 382:“file”-“fire”

Response:

It has been corrected (Line 443).

Line 406:“where”-“which”

Response:

It has been corrected (Line 468).

Line 416:“have”-“having”

Response:

It has been corrected (Line 478).

Line 420: Maybe “close” is a bit of an overstatement? They should not be expected to be very close anyway as there seem to have been several improvements in the current study.

Response:

Thank you very much for this comment. We agree with you that they should not be expected to be very close anyway as there are several improvements in this study. Actually, we use “close” to describe the objective results instead of emphasizing the similarity of the results. The EFs employed in Wang et al. (2008) were constant values for different biomass species burning. The interaction between some undervalued EFs variables and overvalued EFs variables in Wang et al. (2008) may lead to this “close” result. As shown in following Table 2, the uncertainty range of different pollutant species were much smaller than those in Wang et al. (2008), which verified that our results were improved. We are so sorry for the confusing description. We have made the revision in the manuscript (Line 482-483).

Table 2. Uncertainty ranges of different pollutants in emission estimates

Pollutant	Uncertainty ranges	
	Our study	Wang et al., 2008
OC	±30%	±148%
EC	±48%	±132%
CH_4	±20%	±77%
NO_X	±20%	±80%
NMVOC	±45%	±71%
SO_2	±45%	±108%
NH_3	±35%	±137%
CO	±18%	±86%
CO_2	±3%	±60%
$PM_{2.5}$	±36%	±142%

Line 421-422:"The differences were mainly caused. . .": How do the authors know that these are the causes of differences? And which of the factors may be more responsible?

Response:

Thank you very much for your comment. By comparing with different methods and parameters (e.g. biomass loading data, EFs) involved in the processing of emission estimation in previous studies, we find the reasons for the differences in our research. In previous studies, the differences were resulted from different methods and parameters adopted. Compared with previous studies, the combination of multiple satellite products with local EFs data and updated local biomass loading data are likely to improve the estimation of pollutant emission from OBB effectively in this study. Please see the detailed response to the "General Response" above. In order to give a clearer description, we have made revision in the manuscript (Line 485-489).

Line 431: Are lower EFs used here more justifiable?

Response:

Thanks for this query. We are sorry for the unclear description. The justifiable of EFs used in our study is not because of the lower values, but some of EFs were collected from previous research carried out in CEC (Tang et al, 2014), which were more accurate and suitable in CEC. It has been corrected in the text (Line 498-499).

Lines 436-439: This can be combined into one sentence to avoid repetition.

Response:

We have made corresponding revision in the manuscript (Line 506-509).

Line 438:"localized in CEC"-"specific to CEC"

Response:

The sentence has been deleted and combined according to the above suggestion.

Line 439:"can improve"-"are likely to have improved"

Response:

It has been corrected (Line 508).

Line 446: Do 0.03 and 0.85 refer to relative uncertainties?

Response:

Sorry for the unclear description. The uncertainty for the emission factors was cited from references in the supplementary Table S5, mainly ranged from 0.03-0.85 for each pollutant.

Line 446:"At last"-"Finally"

Response:

It has been deleted.

Line 474:"opening"-"open"

Response:

It has been corrected (Line 558).

Line 487:“of the next year” is not needed.

Response:

It has been deleted (Line 572).

Line 488:“impacted on the emission”: This implies causality but the analysis has been based on correlations, which can only be suggestive of possible associations but not conclusive for cause-effect relationships.

Response:

Thank you very much for this suggestion. We quite agree with your opinion that possible associations does not means conclusive for cause-effect relationships. The sentences have been corrected as following (Line 572-574).

“The emission of crop residue burning was associated with the rural population, agricultural output and economic levels while the environmental conditions play an important role in the emissions from forestland, shrubland and grassland burning.”

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Estimating the open biomass burning emissions in Central and Eastern China from 2003 to 2015 based on satellite observation

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Abstract. Open biomass burning (OBB) has significant impacts on air pollution, climate change and potential human health. OBB has raised wide attention but with little~~few~~ focus on the annual variation of pollutant emission. Central and Eastern China (CEC) is one of the most polluted regions in China. This study aims to provide a state-of the-art estimation of the pollutant emissions from OBB in CEC from 2003 to 2015, by adopting the satellite observation dataset (the burned area product (MCD64Al) and the active fire product (MCD14 ML)), local biomass data (updated biomass loading data and high-resolution vegetation data) and local emission factors. The successful adoption of double satellite dataset for long term estimation of pollutants from OBB with a high spatial resolution can support the assessing of OBB on regional air-quality, especially for harvest periods or dry seasons. It is also useful to evaluate the effects of annual OBB management policies in different regions. Here, m~~M~~onthly emissions of pollutants were estimated and allocated into a 1×1 km spatial grid for four types of OBB including grassland, shrubland, forest and cropland. From 2003 to 2015, the emissions from forest, shrubland and grassland fire burning ~~had~~ held ~~a minor~~ annual fluctuation~~variation~~ whereas the emissions from crop straw burning steadily increased.

The cumulative emissions of organic carbon (OC), elemental carbon (EC), methane (CH_4), nitric oxide (NO_x), non-methane volatile organic compounds (NMVOCs), sulfur dioxide (SO_2), ammonia (NH_3), carbon monoxide (CO), carbon dioxide (CO_2) and fine particles ($PM_{2.5}$) were 3.64×10^3, 2.87×10^2, 3.05×10^3, 1.82×10^3, 6.4×10^3, 2.12×10^2, 4.67×10^2, 4.59×10^4, 9.39×10^5 and 4.13×10^3 Gg in these years, respectively. ~~For cropland, corn~~Crop straw burning was the largest contributor for all pollutant emissions, by 84%-96%. ~~Among~~ For the forest, shrubland~~,~~ and grassland fire burning, forest fire burning emissions contributed the most and emissions from grassland fire was negligible due to few grass coverage in this region. High pollutant emissions concentrated~~were populated~~ in the connection area of Shandong, Henan, Jiangsu and Anhui, with emission intensity higher than 100 ton per square kilometers~~ per pixel~~, which was related to the frequent agricultural activities in these regions. Peak~~The monthly~~ emission~~ peak~~ of pollutants occurred in summer and autumn harvest periods including May, June, September and October, at which~~ period~~ ~50% of the whole pollutants emission were emitted in these months ~~for OBB~~. This study highlights the importance in controlling the crop~~s~~ straw burning emissions. From December to March~~ of the next year~~, the crop residue burning emissions decreased, while the emissions from forest, shrubland and grassland exhibited their highest values, leading to another small peak emissions of pollutants. Obvious regional differences in seasonal variations of OBB were observed due to different local biomass types and environmental conditions. Rural population, agricultural output, economic levels, local burning habits, social customs~~anthropological activities~~ and management policies were~~are~~ all influence factors for OBB emissions. ~~The successful adoption of double satellite dataset for long term estimation of pollutants from OBB with a high spatial resolution can support the assessing of OBB on regional air-quality, especially for harvest periods or dry seasons. It is also useful to evaluate the effects of annual OBB management policies in different regions.~~

1. Introduction

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

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

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

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

The lack of local biomass data (biomass loading data and vegetation speciation data) and local emission factors could also introduce uncertainty in emission estimates. CurrentlyAt present, the local biomass loading data has not been updated and still needs to be updated and accurately measured. In addition, Llocal high spatial-resolution vegetation speciation data has beenwas also rarely adopted in OBB estimations. Meanwhile, a lot of researches about OBB have used the same emission factors for pollutants emitted from OBB without considering the various biomass species and combustion conditions (Andela et al., 2013; Giglio et al., 2013). All these should be considered and improved in the establishment of OBB emission inventory.

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

2. Methods

2.1 Estimation of burned areas

OBB emissions in CEC were initially estimated based on the local biomass data (biomass loading data and vegetation speciation data), satellite burned area data (Figure S2 and Figure 2) and emission factors. Fire burning emission amounts are was calculated by the following equation (Wiedinmyer et al., 2011; Shi et al, 2015).

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

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

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

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

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

$BA_{MCD64AL(i,t,j)}$ was directly detected from MCD64AL product. MCD14ML active fire points in each grid included two parts: active fires points with or near MCD64A1 burned area (FC_{in}) and active fires outside the MCD64AL burning area (FC_{out}). $BA_{sf(i,t,j)}$ was the burned area of FC_{out}. The $BA_{sf(i,t,j)}$ was used as supplement. Due to the active fire product existed as the fire points and could not directly obtain the burned area data, ~~. In order to obtain smaller burned areas less than 500 m ×500 m of a land grid cell,~~ the burned area of small fire was estimated based on the following method (Randerson et al., 2012).

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

(~~2~~3)

where $BA_{sf(i,t,j)}$ is the small fire burned area of F_{out} in grid cell i, month t~~,~~ and aggregated vegetation class ~~v~~j; $FC_{out(i,t,j)}$ is the total number of MCD14 ML active fires outside of the burned area in grid cell i, month t and aggregated vegetation class j~~each 1 km×1 km grid cell~~; α is the ratio of $BA_{MCD64A1}$ to ~~the~~ F_{in}~~number of active fires with or near the MCD64A1 burned area (km²)~~ and α is equal to the value of surrounding grid cell if $BA_{MCD64A1}$ is equal to 0; γ is an additional unit less scalar which indicates the difference between ~~the active fires in MCD64Al burning are~~F_{in}~~a~~ and ~~active fires outside the burning area~~F_{out} and γ is assumed equal to 1 in this research; r denotes the burning region; s indicates the burning period.

2.2 Biomass fuel loading

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

$$B_{i,r} = T_{i,r} / A_{i,r} \quad B_i = \frac{T_i}{A_i} \qquad (34)$$

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

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

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

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

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

For grassland and shrubland, local biomass density data were also collected (Pu et al, 2004; Hu et al, 2006) as listed in Table 1. To determine the accurate provincial amounts of crop residue burning, we gathered the production of different species of crops from the China Statistical Yearbook (NBSC, 2003-2015). The dDetailed data of crop-specific residue to production ratio (dry matter) were collected from local statistical data (Table 3) and the updated data for crop straw burned ratio were derived from survey results (Table 4). Using the updated biomass data, the accuracy of the estimation of OBB emission is expected to be improved.

2.3 Combustion efficiency

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

$$CE_s = e^{-0.13 \times TB} \qquad (~~4~~6)$$

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

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

2.4 Emission Factors

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

2.5 Spatial and temporal allocation

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

The emission~~s~~ in t-th grid was~~were~~ calculated ~~using~~ by the following equation:

$$E_{t,j} = BA_{t,j} / BA_{i,j} \times E_{i,j} \qquad (~~5~~7)$$

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

2.6 Other factors influencing OBB emission~~The influence factors for the OBB emission~~

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

2.7 Uncertainty analysis

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

3. Results and Discussion

3.1 Accumulated pollutants emission from OBB in CEC

Table 6 presented~~shows~~ the cumulative OBB emission amounts during 2003-2015 and ~~historical~~multi-year emissions ~~from~~of different provinces were detailedly listed in Table S3. By the end of 2015, the cumulative emissions of OC, EC, CH_4, NO_x, NMVOCs, SO_2, NH_3, CO, CO_2 and $PM_{2.5}$ were 3.64×10^3, 2.87×10^2, 3.05×10^3, 1.82×10^3, 6.4×10^3, 2.12×10^2, 4.67×10^{32}, 4.59×10^4, 9.39×10^5 and 4.13×10^{23} Gg, respectively. ~~In the following section, f~~For better revealing the spatial-temporal variation of OBB emissions, the $PM_{2.5}$ variation was detailedly discussed as an example. ~~At the province level~~ F~~f~~rom 2013 to 2015, the highest emission amounts of $PM_{2.5}$ were found in Henan and Shandong, accounting for 27.93~~28~~% and ~~24~~24.35% of the total emission amounts, respectively. The lowest emission appeared in Zhejiang and Shanghai, which only contributed for 4.05% and 0.43%. For other provinces, Hunan, Hubei, Fujian, Anhui, Jiangxi and Jiangsu accounted from 5.52% to 10.13% of the whole emission.

The contributions of different biomass burning types~~of biomass sources~~ for various pollutants were shown in Figure 3a. Cropland burning contributed the most emission for all the pollutants, by~~from~~ 84%-96%. The forest fire ~~also~~ exhibited higher emission of NH_3, SO_2, NMVOCs and $PM_{2.5}$, accounting for 12%, 11%, 7% and 5% of corresponding total emission, respectively. As shown in Figure 3b, for ~~the~~ cropland~~s~~, wheat, corn and rice straw burning were the top three emission source types for all the pollutants. Corn straw burning contributed the most to SO_2 (48%), NO_x (37%), NMVOCs (33%), CO (32%) and CO_2 (28%) emission. Highest contributions of EC (45%), OC (33%) and CH_4 (32%) from rice straw burning was found, while wheat straw burning contributed the most

(31%) to $PM_{2.5}$ emission.

In Figure 4, except for Fujian, cropland burning emission was the largest contributor to ~~the~~ $PM_{2.5}$ emission, with the contributions ranging from 75.25% (Jiangxi) to almost 100% (Shanghai). The higher rural agglomeration, abundant crop~~s~~ production and more crop residue burning activities in these provinces can explain the higher contributions. ~~In~~ Shanghai~~,~~ is one of the most developed cities in China~~,~~. ~~the~~ The highest contribution of cropland burning is not related with its~~the~~ high levels of agricultural activities, but is ~~only~~ due to the lack of emissions from other open biomass burning sources. Highest contribution from the forest fire burning and shrubland fire burning were found in Fujian as 45.29% and in Jiangxi as 23.95%, respectively. For forest fire burning, the ~~Southern~~ southern provinces (Fujian, Zhejiang, Jiangxi, Hunan, Hubei and~~,~~ Anhui) exhibited higher values, varying from 3.66% (Hubei) to 38.3% (Fujian) and for shrubland fire burning, the contributions varied ~~in~~ from 1.5% (Hubei) to 7.23% (Zhejiang). The relative high emission contributions of forest and shrubland fire burning in the southern provinces can be explained by the large forest and shrubland coverage, frequent human forestry activities, low precipitation and dry weather in spring and winter (Cao et al., 2015), which may easily lead to forest and shurbland fires. While for the ~~Northern~~ northern provinces (Shandong, Henan~~,~~ and Jiangsu), the contributions ranged around 0.76%-1.97%, ~~respectively,~~ which can be neglected. ~~This is mostly due to the suitable weather conditions, a relative large forest and shrubland coverages and frequent human forestry activities in those provinces as Figure 2 shown.~~ $PM_{2.5}$ emission from grassland in CEC was negligible with the following provinces holding ~~the~~ higher contributions: Jiangxi (0.8%), Hunan (0.25%), Fujian (0.11%) and Anhui (0.1%).

From Figure 5, emissions from wheat and corn straw burning mainly concentrated in Shandong and Henan (totally accounting for 82% and 78% of the whole emissions, respectively) and the rice straw burning exhibited higher concentrations in Hunan, Jiangxi and Hubei provinces, by 25%, 18% and 16%, respectively. The total contributions of rapeseed, cotton, potato and peanut straw burning to the $PM_{2.5}$ emission were relatively small, accounting for~~occupied by~~ 21%-24% of the total emissions. Most emissions from cotton, peanut and potato straw burning located in Shandong (totally accounting for 35%, 35% and 20%) and Henan (totally accounting for 19%, 40% and 15%). Hubei (32%) and Hunan (31%) were the major provinces for rapeseed straw burning emissions. In addition, emissions from soya bean, sugar cane, tobacco, sesame and sugar beet straw burning were negligible, which never exceeded 1% of total crop residue burning emission in this study.

3.2 Temporal variation and spatial distribution for OBB emissions in CEC

3.2.1 Yearly variation

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

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

Emissions from forest, shrubland and grassland fire burning have an obvious trend of declining from 2003 to 2006 and rising from 2006 to 2008. Peak emissions for $PM_{2.5}$ from forest, shrubland and grassland fire burning were found in 2008, as 49 Gg, 8.9 Gg and 0.7 Gg, respectively. In 2008, intensive policies for utilization of straw energy (Table S1) and strengthening the forestry fires prevention (Table S2) were published, which effectively limited the emissions from forest and shrubland fire burning as Figure 7a shown. ~~Peak emissions for $PM_{2.5}$ from forest, shrubland and grassland burning were found in 2008, as 49 Gg, 8.9 Gg and 0.7 Gg, respectively.~~ Obvious decreasing was found from 2008 to 2010, down to 19 Gg, 4.8 Gg and 0.24 Gg, respectively. Then they exhibited inter-annual oscillation from 2010 to 2015, with higher emission ~~amounts~~ in ~~odd~~2011, 2013 and 2015~~ years~~ and lower emission ~~amounts~~ in 2012 and 2014~~ even years~~ (Jin et al., 2017a). The multi-year tendency for forest, shrubland and grassland fire burning were mainly affected by the variations in climate, management measures and other human forcing. It can also conclude that the yearly fluctuation~~variation trends~~ of pollutants from OBB ~~were~~ was mainly impacted by the emission ~~from~~ of forest, shrubland and grassland fire burning, but not the crop residue burning.

The emission of $PM_{2.5}$ from crop residue burning exhibited quite different yearly variation trend with other three types of biomass burning, which gradually increased from 2003 (228 Gg) to 2015 (323 Gg), by 29%. The increase of crop residue production can primarily explain the increasing of pollutant emission. Meanwhile, ~~from~~ as shown in Figure S6 and Table S1, the controlling of pollutants from crop residue burning in China started from 1965s. ~~and i~~In 2000, the law for prevention of air pollution was published. Then in 2003, the regulations on straw banning and comprehensive utilization were released. In Figure 6, we found that the emission of $PM_{2.5}$ from crop residue burning significantly increased from 2003 (228 Gg) to 2008 (294 Gg), due to the increase of crops production and deficiency of strict control policies in this period (Table S1). Although emissions from forest, shrubland and grassland fire burning fluctuated markedly during this period, the obvious increase of crops residue burning dominated the total growth of OBB emission from 2003 to 2008 as their higher emission amounts. From 2008 to 2015, ~~Strict~~strict policies were developed to improve the straw energy utilization ~~to~~and reduce the air pollution raised by its burning. However, it has to say, the policies may not be well implemented, with the annual averaged increasing amounts of 7.3 Gg for $PM_{2.5}$. From Figure 7b, the large contributions to $PM_{2.5}$ (22%-28% and 29%-33%) and increasing trends for corn straw burning and wheat straw burning could be found, which should be further focused. The contribution~~s~~ ~~from~~ of rice straw burning has slightly ~~decreased~~ decreased~~in the past decade~~ in research period, by about 19% ~~from 2003 to 2015~~. Other types of biomass totally accounted for averaged 25% of $PM_{2.5}$ emission and all exhibited slightly ~~decreasing~~ increasing trend from 2003 to 2015, ~~increased~~ by about 21%-29%.

Figure 8 showed that the crop residue burning emission in Henan, Shandong, Anhui, Jiangsu, Hubei, Hunan and Jiangxi exhibited obvious increasing trends, which suggested the importance of crop residue burning control in these provinces. For Fujian and Zhejiang, no obvious increase for cropland burning emission was found, implying that the emission~~s~~ ~~have~~ has been well controlled in these years. It should be noted that in Fujian and Zhejiang, the main crop is rice. W~~w~~hile in other provinces, the main crops are corn and wheat especially for ~~Northern~~ northern provinces. To conclude, pollutants emitted from crop straw burning (wheat, corn and rice) are still now the key sources for air pollution, in view of its increasing emission trend. The randomness of burning activities and corresponding widespread and ~~scattering~~ scattered distribution make it difficult to control them. The wheat and corn emissions at ~~Northern~~ northern provinces and rice burning emissions at ~~Southern~~ southern provinces should be controlled specially in the future.

In Figure ~~10~~8, the $PM_{2.5}$ emission from crop residue burning exhibited higher amounts for Henan and Shandong province in 2015, as 100 Gg and 82 Gg, respectively, which are 200%-1200% times of those for other provinces. As the main source regions for air pollution of Yangtze River Delta (YRD) and Beijing-Tijin-Hebei (BTH) region (Fu et al., 2013; Zhou et al., 2015), the enforced and effective control of crop residue burning in the two provinces at summer and autumn harvest periods are important for improving the air quality of these regions.

3.2.2 Monthly distribution

The monthly $PM_{2.5}$ emission variation of different OBB in CEC was shown in Figure 9a. ~~The total monthly~~ $PM_{2.5}$ emission held higher amounts in May and June (90.4 Gg-179.3 Gg), followed by December to March of next year (32.2 Gg-127.3 Gg) and September-October (8.2 Gg-89.2 Gg), and was lowest during July-August (14.3

Gg-65.9 Gg). As the emission amounts of cropland fire burning was one or two magnitude higher than other three types of biomass burning, the monthly variation of total $PM_{2.5}$ emission~~variation~~ was dominantly~~mainly~~ controlled by the crop residue fire burning (Zhang et al., 2016). The periods with highest $PM_{2.5}$ emissions were ~~just~~ the summer and autumn harvest times, when the burning activities are more frequent. The peak of open biomass fire burning occurred~~s~~ in May and June, ~~–~~totally accounted for 42% of the whole $PM_{2.5}$ emission in 2003-2015, which is caused by the harvest and open residue burning of winter wheat, especially in Henan, Shandong, Jiangsu and Anhui (Figure 9b). Large amounts of wheat straw were burned after the harvest to increase the soil fertility and prepare for following corn cultivation (Levine et al., 1995). ~~Though the open biomass burning was strictly forbidden in recent years, scattered burning activities still existed in China.~~ The small peak of open biomass burning emission in September to October (totally accounted for 13.82% of the whole $PM_{2.5}$ emission in 2003-2015) can be attributed to the burning of corn straw after corn harvest. Though the open biomass burning was strictly forbidden in recent years, scattered burning activities still existed in these regions. As shown in Figure S4, ~~in recent years,~~ the $PM_{2.5}$ emissions in CEC and major agricultural provinces during harvest time have shown a rapid decline in recent years, in accordance with the change tendency of burned area due to increased government management. Considering of the yearly increasing fact ~~increase tendency~~of crops straw burning~~ from year to year~~, it is worth noting that fire burning out of harvest season as a way of circumventing governmental polices needs to be well regulated. From December to February~~ of the next year~~, the crop residue burning emission decreased to the lowest level in the whole year (18.9% of the whole $PM_{2.5}$ emission in 2003-2015). However, the emissions of $PM_{2.5}$ from forest, shrubland and grassland burning achieved peak values from December to March, being 67% of that in 2003-2015.~~However, during December to March, the emissions of $PM_{2.5}$ from forest, shrubland and grassland exhibited their peak values, totally occupied by 67% of the whole $PM_{2.5}$ emission of forest, shrubland and grassland fire burning in 2003-2015~~.

Figure 10 clearly listed the monthly average emissions of $PM_{2.5}$ from OBB in different provinces. These provinces were classified based on the correlation between their monthly emissions~~based on the correlation of emissions in each month~~ of 2003-2015. Henan, Shandong, Anhui and Jiangsu provinces (R^2 higher than 0.92, $P<0.01$), as one of the largest and contiguous wheat planting areas in China (Fang et al., 2014), have two crop rotations. The highest monthly emissions were observed for winter wheat harvesting (sown in October and harvested from May to June) and corn harvesting (sown in middle June and harvested from September to October). A large proportion of crop straw were always burnt directly after the crop harvest (MEPC, 2015). For Hubei province, agricultural emissions fluctuated over the period from February to October with several peaks due to that different crop species matured in succession. In Jiangxi, Fujian and Hunan (R^2 higher than 0.9, P<0.01), the largest monthly emissions were observed with forest and shrubland fire burning during the time between December and March~~ in the next year~~, which is the dry season in these provinces (Li et al., 2014; Li et al., 2015). ~~And~~ While in other months, the emissions were limited. For Shanghai and Zhejiang ($R^2 = 0.7$, P<0.01), lowest levels of $PM_{2.5}$ emission were found, with peak values ~~also occurred~~ occurring in summer and autumn harvest periods. Obvious two peaks were found for April-May and July-August periods, which may reflect the rice harvesting at these times. To sum up, these regional differences of monthly $PM_{2.5}$ emissions from OBB were mainly caused by the different biomass burning types and times as well as corresponding environmental conditions.

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

The spatial distribution of $PM_{2.5}$ emitted from OBB within 1 km×1 km resolution was mapped based on the burned area and a high-resolution vegetation map (1:1 000000) in CEC. The multi-year averaged spatial distributions of $PM_{2.5}$ emission ~~are~~ were shown in Figure 11. It can be found that the OBB was widespread and scattered. The average emissions intensity of $PM_{2.5}$ ranged from 0 to 15 tons per pixel in most provinces. The variation range is mainly caused by the social-economic development level, rural population and agricultural activities. The highest value in different provinces was all ~~caused~~ mainly raised by the ~~crops~~ cropland fire burning due to the centralized burning ~~of them~~ in a relatively small area. Some pixels with high emissions exceeding more than 100 tons each year were found in Henan, Shandong and Hunan. It can be attributed to the large amounts of crop straws in these provinces. The pixels of high emission intensity more than 70 tons from crop straw burning were also found in Hubei, Jiangsu and Anhui. For forest and shrubland fire burning, the high emission points ~~from~~ (more than 30 tons per pixel) were found in Fujian and Jiangxi. Lower emission intensities in Zhejiang (lower than 10 tons per pixel on average) and Shanghai (lower than 7 tons on average) were mainly due to the highly developed economy and limited agricultural activities (Su et al., 2012). In addition, northern Anhui and eastern Jiangsu ~~were found~~featured high emissions of OBB with a relatively lower intensity (lower than 15 tons per pixel on average), which may be due to that the crop straw ~~was~~ were burned in a large area in these regions.

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

Figure 12 highlights the spatial distribution of $PM_{2.5}$ emitted from OBB in different seasons of 2015. Emissions were more concentrated in summer, followed by winter. In summer, the emission was ~~mainly~~ concentrated in the connection regions of Henan, Shandong, Anhui and Jiangsu, which is mainly raised by the crop ~~straw~~ residue burning as discussed before. In winter, Jiangxi, Hunan and Fujian showed the higher emission intensities from forest and shrubland burning.

3.3 The impact of social-economic factors on OBB emission

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

Social customs~~Some anthropogenic activities~~ also pose impact on OBB emissions. Biomass burning emissions in April can be enhanced by human burning activities in the tomb-sweeping day. The tomb-sweeping day (often in April 4 or April 5) is a time to memorize the death. People sweep ~~their~~ graves and burn sacrifices by ignited straw, which can easily cause grass, shrub and forest fires (Qiu et al., 2016). The fire points at the tomb-sweeping day in CEC ~~can~~ occupied by 22%-38% of the whole ~~file~~ fire points in April in some years (Figure ~~S3~~S5). The Chinese government has also introduced policies to prevent forest, shrubland and grassland fires on tomb-sweeping day

(Table S2). The wildfires caused by biomass burning from late January to early February are partially related to the firework burning in the Spring Festival (Zuo, 2004). The firework burning activities for celebration and official sacrifices to ancestors in the Spring Festival can easily lead to grass, shrub and forest fires. All these activities can affect the emission levels and air quality in a short time scale.

In order to understand the impact of the rural population, local economic level and agricultural level, correlation analysis between $PM_{2.5}$ emissions from OBB and statistics data (the rural population, the per capita net income of rural residents and , agricultural output (crop straw burning) and forestry output (forest, shrubland and grassland burning) in different provinces were conducted. For crop residue burning, sSignificant positive correlations were found between the rural population, agricultural output and the $PM_{2.5}$ emissions from crops straw burning (R^2 higher than 0.58, P<0.01) for the whole CEC (Figure 13a). According to our survey, Tthe high rural population and agricultural output indicates that agricultural activities are quite important in a certain region. With more crops residue produced, it can easily cause high emissions from cropland fire burning. No significant correlations were found for $PM_{2.5}$ emission from crop straw burning with the income of rural residents (Figure 15), which indicates that the rural economic level in different regions in CEC have no relationship with the $PM_{2.5}$ emission. Then we calculated the correlations between the change tendency of $PM_{2.5}$ emission from crops fire burning and the multi-year variation of other three social-economic factors as Table 7 shown for different provinces. Significant positive correlations were found for $PM_{2.5}$ emission and with per capita income of rural residents and agricultural output (most R^2 higher than 0.59, P<0.01) and negative correlation were found for $PM_{2.5}$ emission with rural population (most R^2 higher than 0.73, P<0.01) except for the provinces of Shanghai, Zhejiang and Fujian, which are underdeveloped agricultural provinces. From 2003 to 2015, with the increase of agricultural outputs, more crop residue was produced. However, rapid economic development and less rural population in each province lead to the popular of commercial energy and clean energy in rural area. It decreased the demands in using crop residue as fuel. As a consequence of this, more crop residues were directly burned in the agricultural field. But it was not suitable to for Shanghai, Zhejiang and Fujian (most R^2 lower than 0.19, P>0.05), where which holds less crop residue production and high utilization efficiency of crop straws.

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

3.4 Comparison with others

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

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

3.5 Uncertainty analysis

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

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

4 Conclusions

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

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

From 2003 to 2015, the multi-year tendency of opening biomass residue burning emission for various pollutants ~~is~~ was similar. Emissions from crop straw burning continued to increase, due to the gradual increase of crop residue production. While emissions from forest, shrubland and grassland fire burning exhibited yearly ~~minor~~

fluctuations~~ from year to year~~, which was mainly influenced by the environmental conditions, management measures and other human driving factors. Monthly distributions revealed that the pollutant emissions were at the highest levels in May and June, with the lowest emissions in July and August. The high emissions ~~from~~ in May~~ to~~ , June and October were mainly caused by crop straw burning in sowing and harvest times. It is worth noting that the fire burning activities at harvest season need to be regulated continuously by local governments and ~~the fire burning out of harvest season should also be paid more attention in recent years. Meanwhile,~~ emissions from forest and shrubland burning accounted for the vast majority of total emissions in December to March should also be paid attention~~ of the next year~~. The emission of crop residue burning was associated with the rural population, agricultural output and economic levels while the environmental conditions play an important role in the emissions from forestland, shrubland and grassland fire burning.~~The rural population, agricultural output and economic levels impacted on the emission of crop residue burning while the emissions from forestland, shrubland and grassland burning were more affected by environmental conditions.~~

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

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

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

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

Table 2. Parameters of ~~biomass-stem volume regression functions~~linear regression model for biomass and stock volume of dominant tree species~~ in forests (B=aV+b)~~.

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

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

~~B = aV + b: B indicates the total biomass of different tree species (t); V indicates the forest stock volume (m^{-3}).~~

Table 3. ~~The d~~Detailed crop ~~specific~~ residue to production ratio data for each province

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

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

Table 4. ~~The d~~Detailed crops straw burned ratio data for each province~~from survey results~~.

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

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

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

Vegetation	OC	EC	CO	CH_4	NO_x	NMVOCs	SO_2	NH_3	CO_2	$PM_{2.5}$
Corn	1.457[*]	0.14*	70.2[a]	4.4[b]	3.36[a]	10[c]	0.45[c]	0.68[g]	1261[f]	5[c]
Rice	1.96[a]	0.52[c]	52.32[c]	3.9[b]	1.42[d]	6.05[f]	0.147[a]	0.53[g]	791[f]	3.03[d]
Wheat	2.7[b]	0.49[a]	61.90[c]	3.4[b]	1.19[d]	7.5[c]	0.147[c]	0.37[b]	1557[f]	7.6[a]
Cotton	3.06[c]	0.57[f]	70.29[c]	4.4[b]	2.98[c]	10[c]	0.23[c]	0.68[b]	1445[h]	11.7[c]
Rapeseed	1.08[d]	0.23[d]	34.3[d]	3.9[b]	1.12[d]	8.64[c]	0.25[c]	0.53[g]	1445[h]	5.76[c]
Soya bean	1.05[d]	0.13[d]	32.3[d]	3.9[b]	1.08[d]	8.64[c]	0.25[c]	0.53[g]	1445[h]	3.32[d]
Sugar cane	2.03[c]	0.41[c]	40.08[f]	3.9[b]	2.03[c]	11.02[f]	0.25[c]	0.53[g]	1445[h]	4.12[f]
Peanut	2.03[c]	0.41[c]	55.13[c]	3.9[b]	2.11[c]	8.64[c]	0.25[c]	0.53[g]	1445[h]	5.76[c]
Potato	2.03[c]	0.41[c]	55.13[c]	3.9[b]	2.11[c]	8.64[c]	0.25[c]	0.53[g]	1445[h]	5.76[c]
Tobacco	2.03[c]	0.41[c]	55.13[c]	3.9[b]	2.11[c]	8.64[c]	0.25[c]	0.53[g]	1445[h]	5.76[c]
Sesame	2.03[c]	0.41[c]	55.13[c]	3.9 [b]	2.11[c]	8.64[c]	0.25[c]	0.53[g]	1445[h]	5.76[c]
Sugar beet	2.03[c]	0.41[c]	55.13[c]	3.9[b]	2.11[c]	8.64[c]	0.25[c]	0.53[g]	1445[h]	5.76[c]
Coniferous forest	7.8[e] ~~2.65[j]~~	0.2[e] ~~0.11[j]~~	118[e]	6[e]	2.4[e]	28[e]	1[i]	3.5[e]	1514[e]	9.7[e]
Broadleaf forest	9.2[e] ~~1.181*~~	0.6[e] ~~0.31[e]~~	102[e]	5[e]	1.3[e]	11[e]	1[e]	1.5[e]	1630[e]	13[e]
Mixed forest	9.2[e]	0.6[e]	102[e]	5[e]	1.3[e]	14[e]	1[i]	1.5[e]	1630[e]	9.7[e]
Grassland	2.6[e]	0.4[e]	59[e]	1.5[e]	2.8[e]	9.3[e]	0.5[e]	0.5[e]	1692[e]	5.4[e]
Shrubland	6.6[e]	0.5[e]	68[e]	2.6[e]	3.9[e]	4.8[e]	0.7[e]	1.2[e]	1716[e]	9.3[e]

References: [a] Cao et al. (2008); [b] Li et al. (2007); [c] He et al. (2015); [d] Tang et al. (2014); [e] Akagi et al. (2011); [f] Zhang et al. (~~2009~~2008); [g] EPD (2014); [h] Wang et al. (2008); [i] Andreae and Rosenfeld (2008); [*] This study.

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

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

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

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

Table 8. Comparison of the emissions with previous studies in different years (Gg ~~yr^{-1}~~)

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

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

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

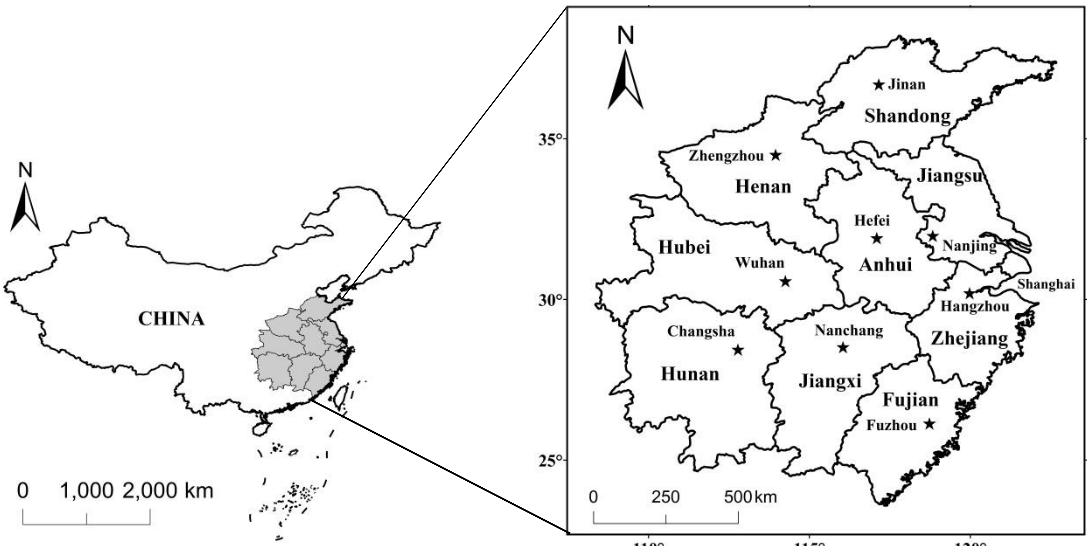


Figure 1. Location of Central and Eastern China and the key megacitie

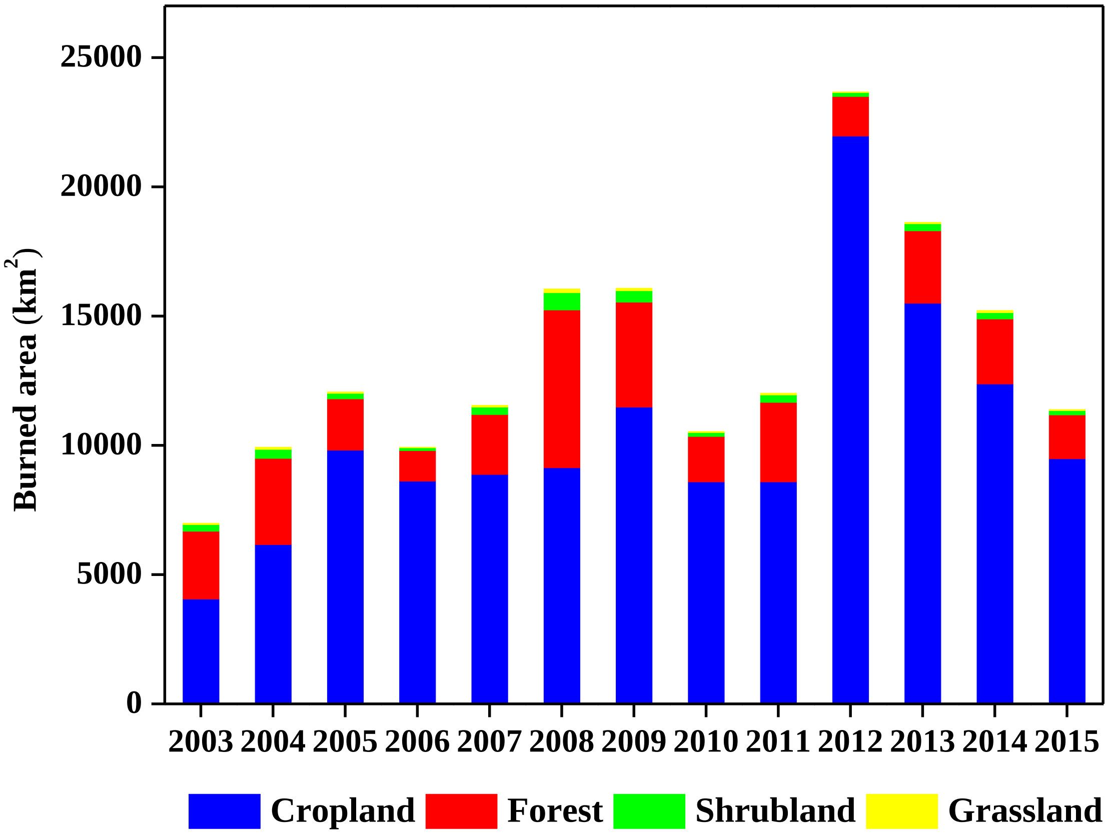
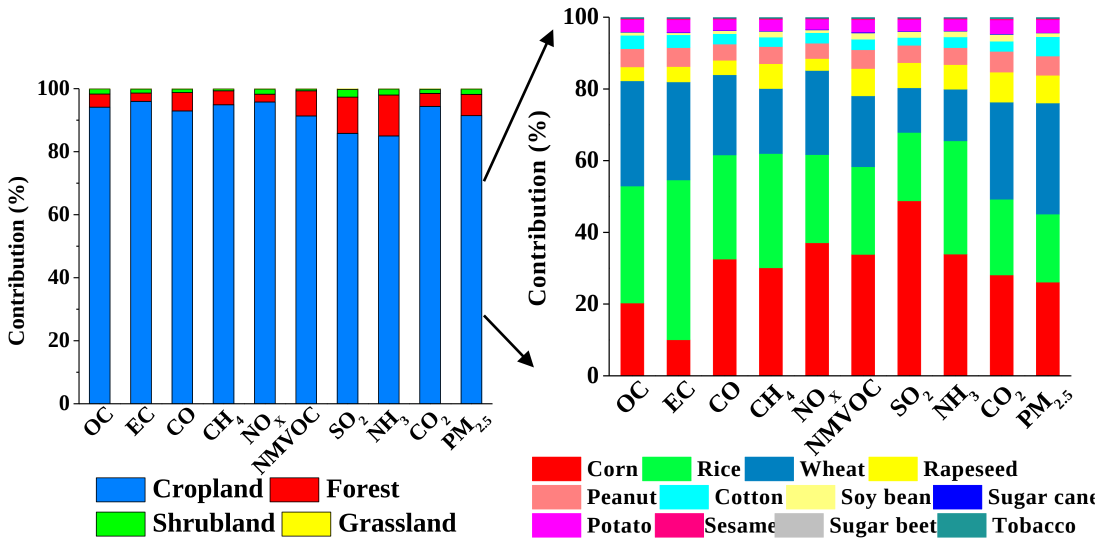


Figure 2. The integrated open biomass burned area in Central and Eastern China from 2003 to 2015.



(a) (b)

Figure 3. The mean contributions of different types of biomass to biomass burning pollutant emission (a) and ~~and~~ the mean contributions of different types of crops to ~~the whole~~ cropland accumulative pollutant emission (b)~~s of pollutants in Central and Eastern China~~ from 2003 ~~-~~to 2015.

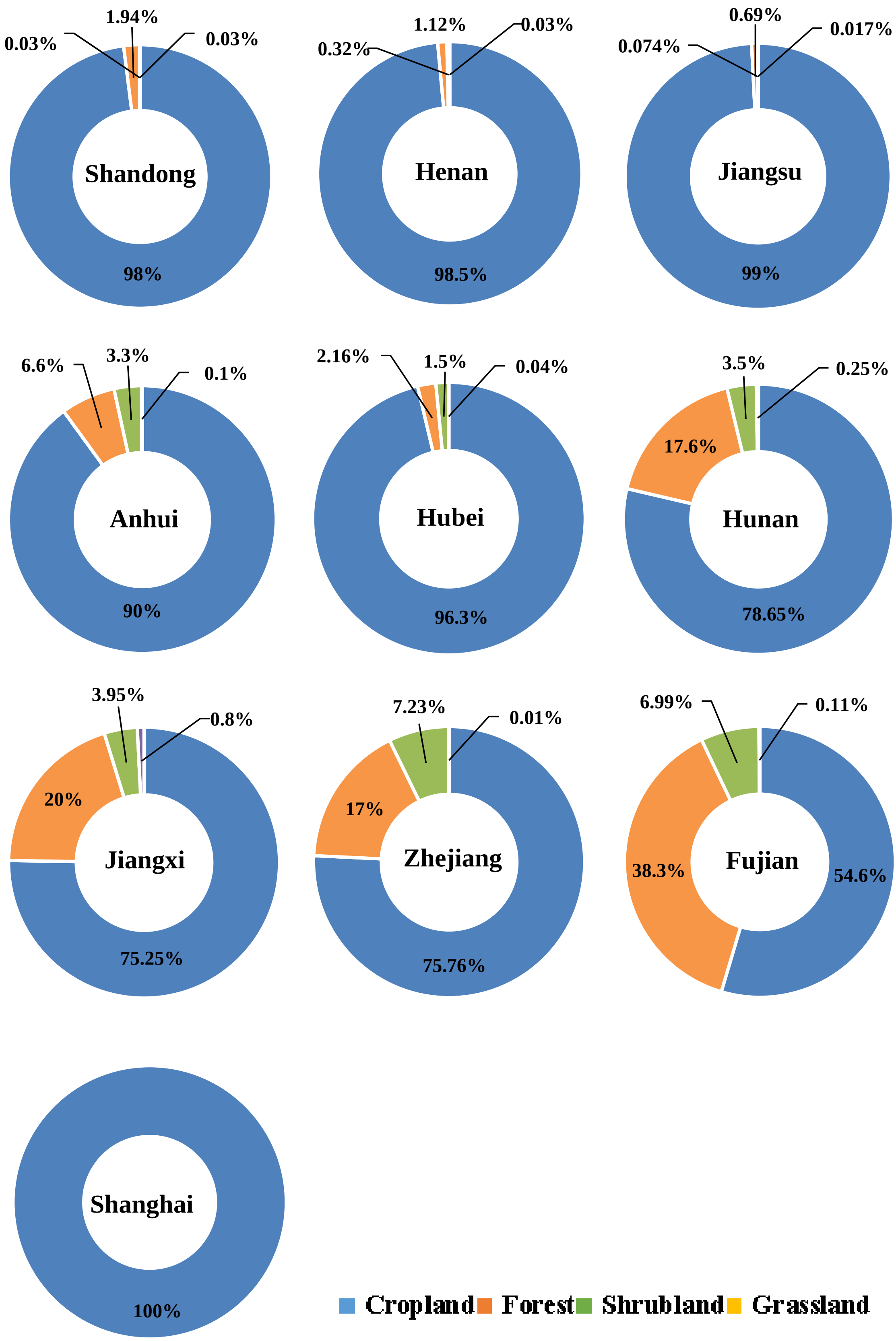


Figure 4. The averaged contributions of different biomass burning types to $PM_{2.5}$ emission in each province.

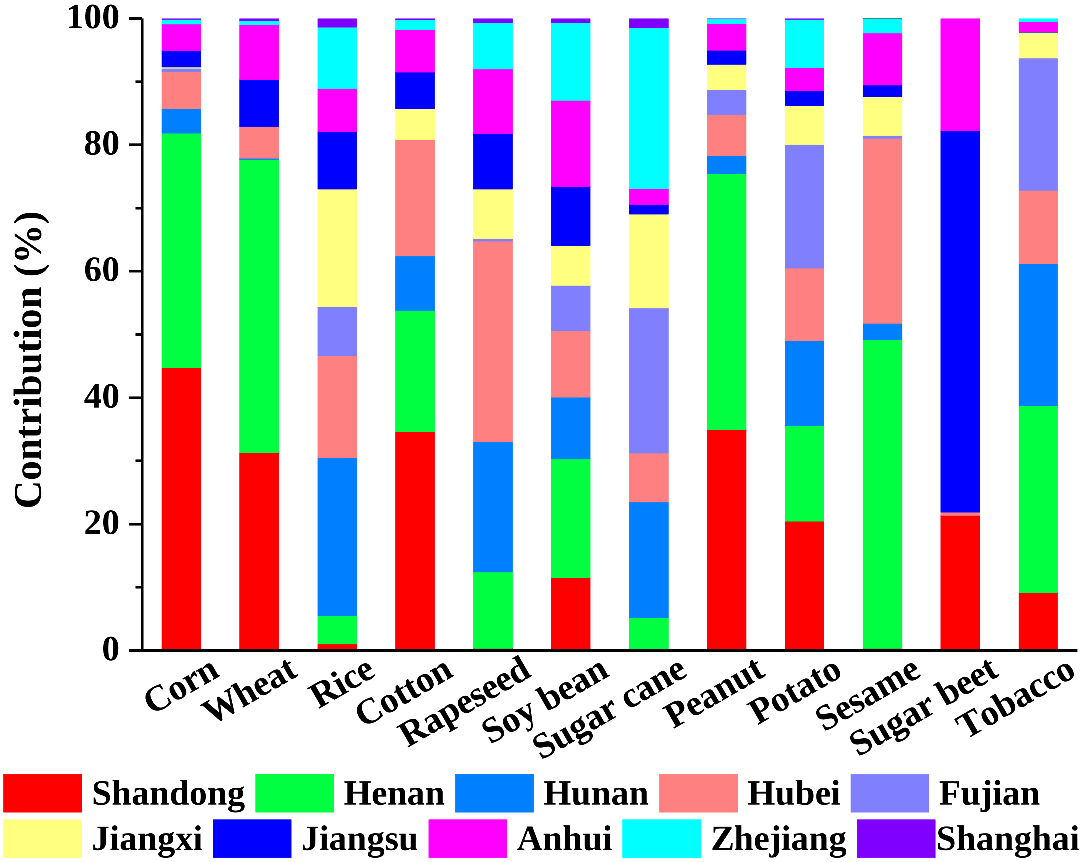


Figure 5. The averaged contributions of various crops straw burning to cropland $PM_{2.5}$ emission in different provinces.

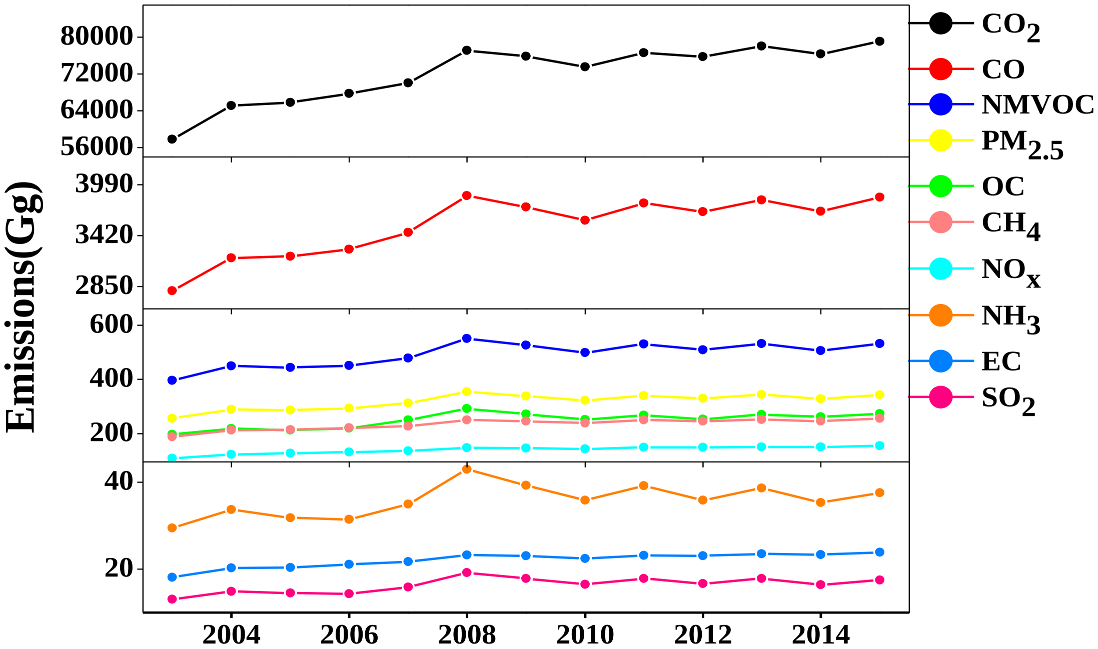


Figure 6. ~~Historical~~ Yearly emissions of open~~ing~~ biomass burning from 2003 to 2015.

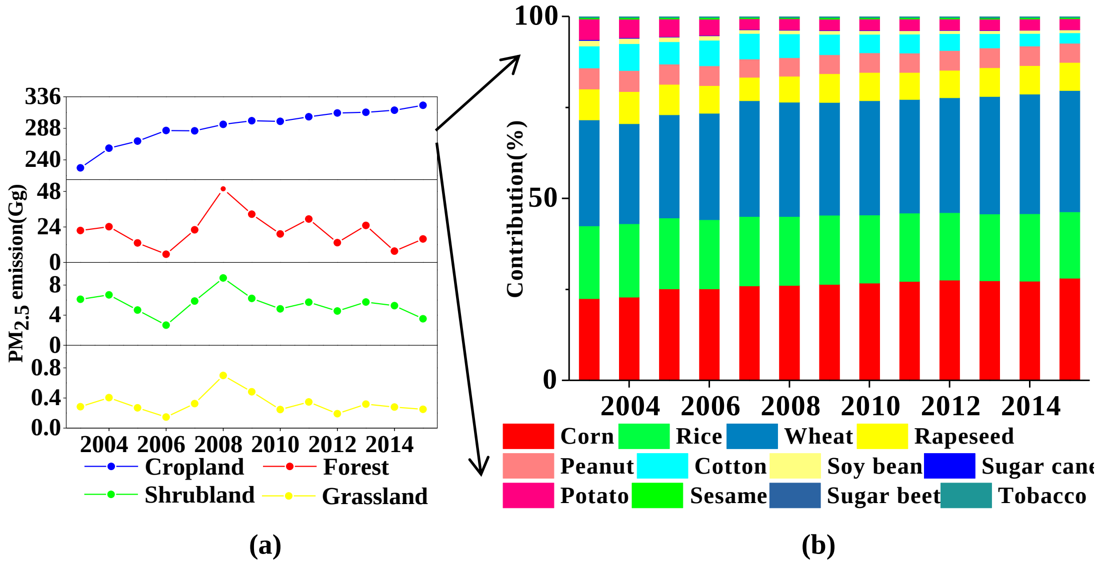


Figure 7. The multi-year $PM_{2.5}$ emissions ~~from~~ of ~~(a):~~ different opening biomass burning sources (a)~~;~~ and ~~(b):~~ various crop types (b) from 2003 to 2015.

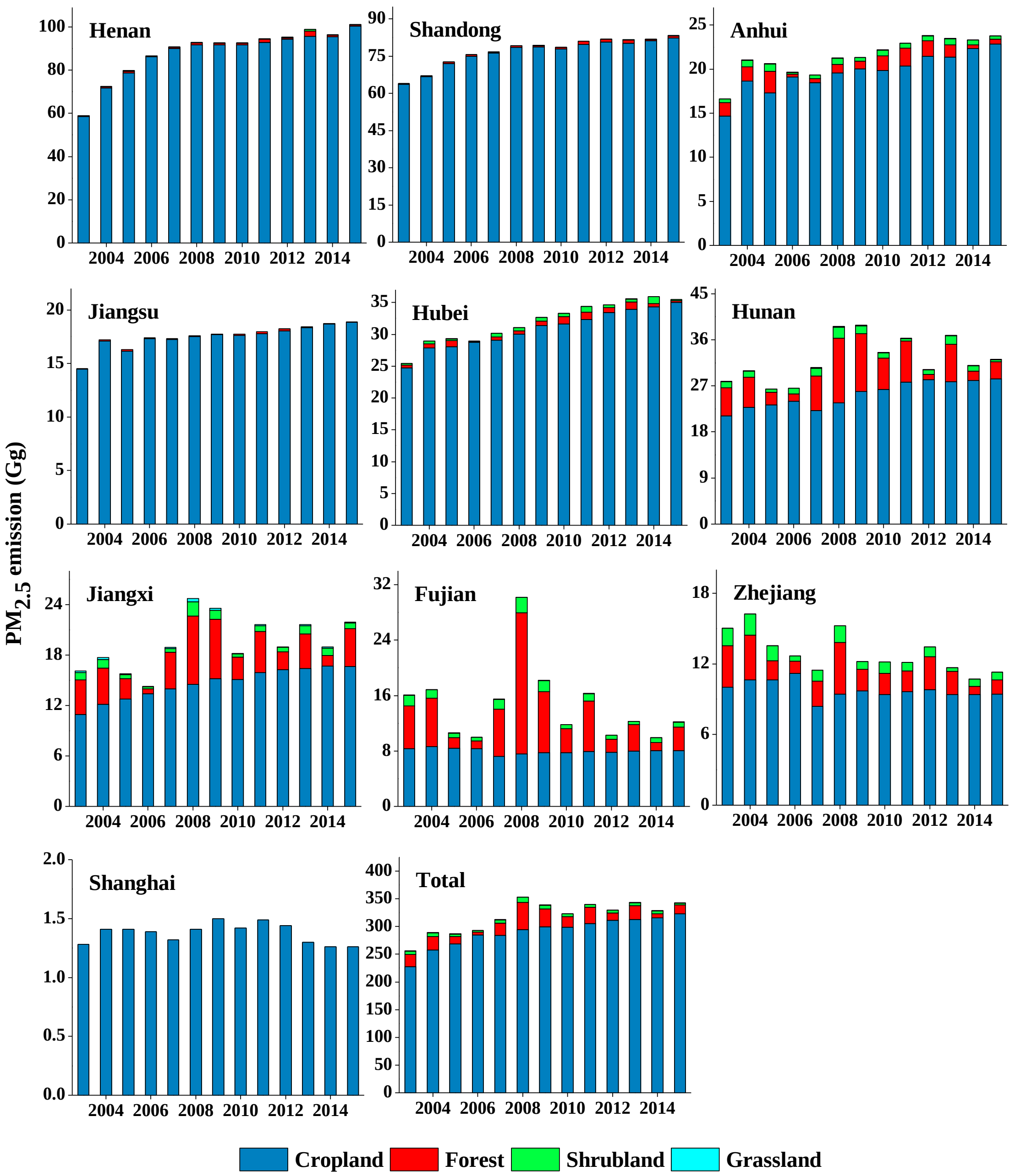


Figure 8. The multi-year $PM_{2.5}$ emission ~~for~~ of the four types of biomass burning in different provinces from 2003 to 2015.

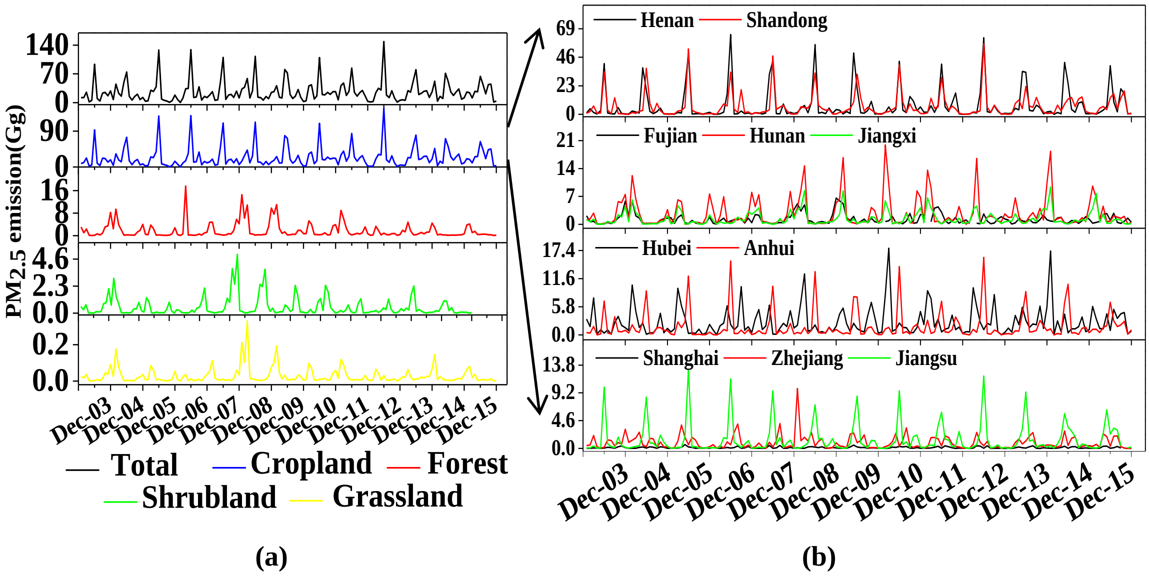


Figure 9. The monthly $PM_{2.5}$ emission of different open biomass burning from 2003 to 2015 for the whole Central and Eastern China (a) and ~~for~~ each province (b).

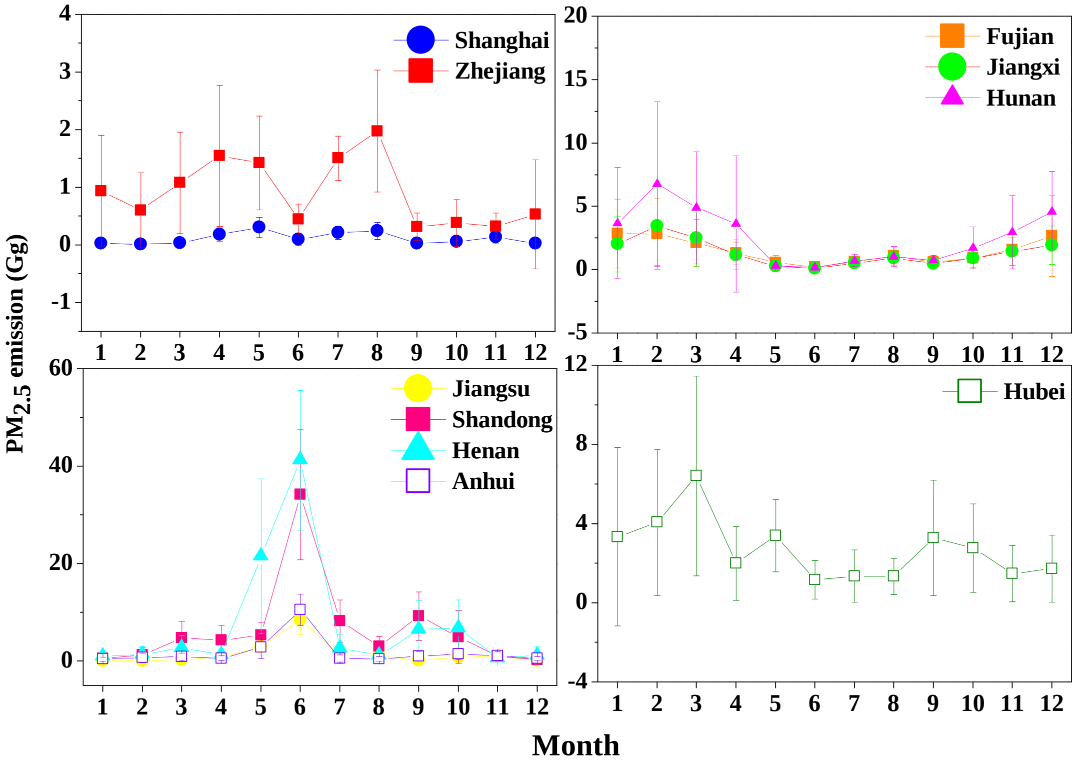


Figure 10. The ~~M~~monthly $PM_{2.5}$ emission from open biomass burning in each province.

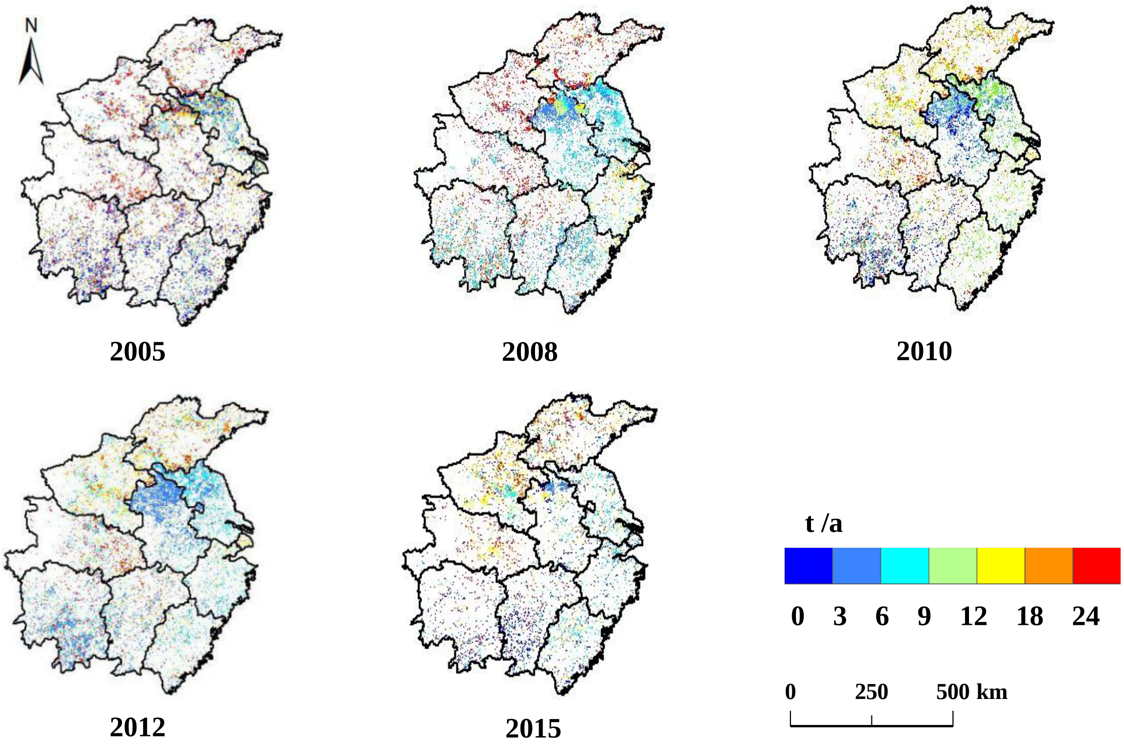


Figure 11. Annual spatial distribution (1 km × 1 km) of $PM_{2.5}$ emissions from opening biomass burning in Central and Eastern China.

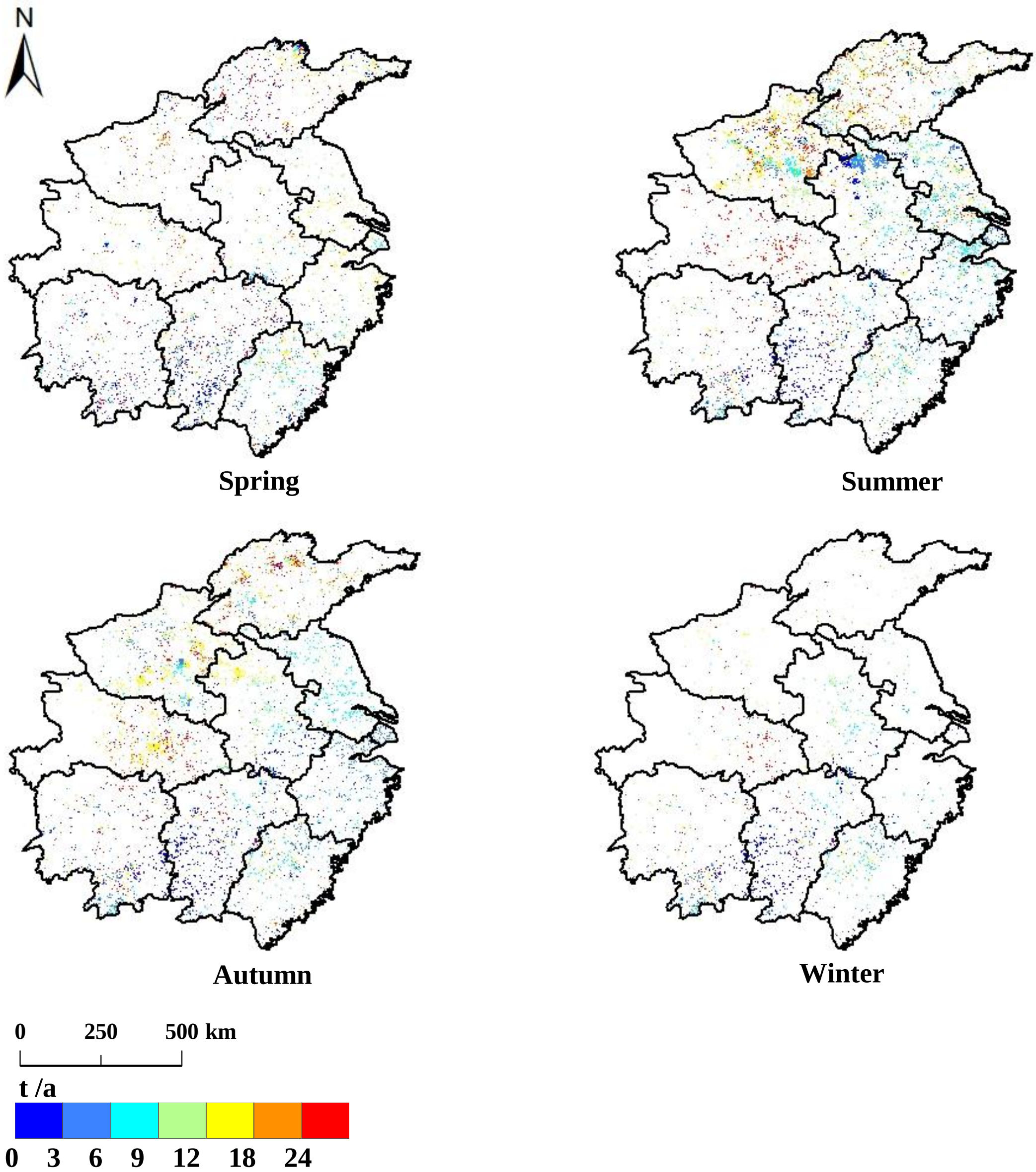
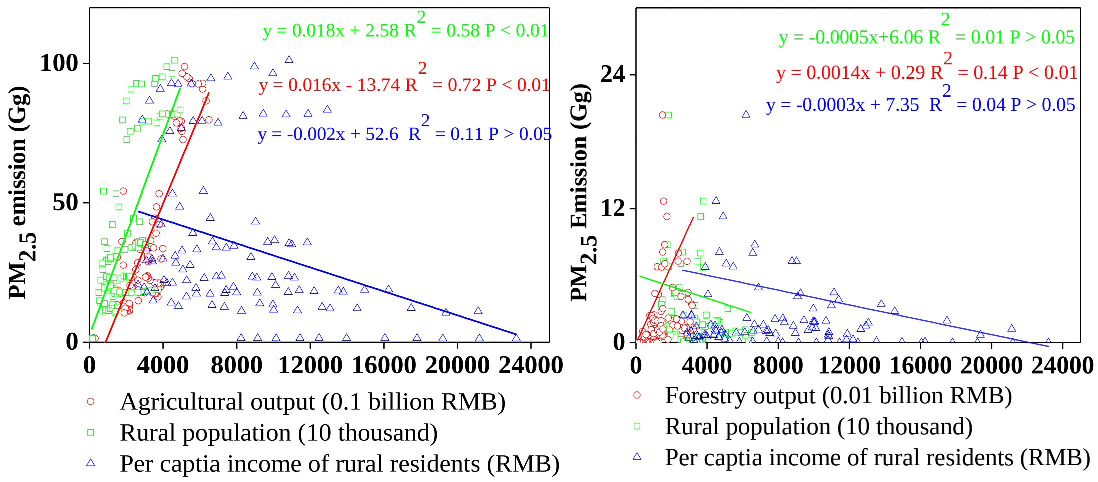


Figure 12. Seasonal emission distribution (1 km×1 km) of $PM_{2.5}$ in 2015 from opening biomass burning in Central and Eastern China.



(a) (b)

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