

Authors' Responses to Reviewer Comments

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

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

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

Response:

We thank you for your comments, which were very helpful for revising and improving our paper. Generally, two approaches are employed for developing a biomass burning emission inventory: a “top-down” approach and a “bottom-up” approach. Regarding a consideration of new methods, we point out the overwhelming importance of employing refined and updated data to develop a more accurate emission inventory with a much higher temporal-spatial resolution. In fact, highly detailed emission information is extremely important for investigating the causes of air pollution (e.g., air quality modeling) and developing a targeted strategy for the control of pollution accompanying biomass burning, particularly for conditions prevalent in recent years owing to the altered pattern of energy consumption in rural areas and the increasing pollution problems associated with dramatic urbanization caused by the economic development in China. Currently, as far as we are aware, few studies have developed a comprehensive biomass burning emission inventory in China, particularly after 2007, because of a lack of detailed statistical data regarding firewood consumption.

Furthermore, the source-specific emission factors (EFs) used in emission estimation need to be updated based on a systematic combination of localized measurements conducted in China. In addition to EFs, the activity data is also a key factor for improving an emission inventory. Moreover, several key sources of information related to biomass emission estimation must be updated, such as the proportion of crop straw domestic combustion and in-field burning, and the uneven temporal distribution coefficient, which reflects recent conditions in different regions of China. In fact, the current biomass burning emission inventory for China is generally at a province-level resolution because detailed activity data is not publicly available. It is obvious that the resolution of activity data determines the preliminary resolution of an emission inventory. An emission inventory with a coarse preliminary resolution could result in greater uncertainty in grid emissions generated according to source-based gridded spatial surrogates (e.g., population) using GIS technology.

The main contributions of our work are summarized as follows.

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

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

Third, the high-resolution temporal (monthly and daily) and spatial (1 km × 1 km) biomass burning emission inventory presented in this study includes major precursors of complex pollution systems, greenhouse gases, and heavy metals released from biomass burning such as SO₂, NO_x, PM₁₀, PM_{2.5}, VOCs, NH₃, CO, EC, OC, CO₂, CH₄, and Hg with further breakdowns of PM_{2.5} particles and VOCs.

In addition, we have carefully taken the reviewer's remaining comments related to

methods and data into consideration during the revision of our paper (e.g., emission estimation of forest and grassland fires). Please see the following point-by-point responses.

"Specific comments"

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

Response:

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

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

Second, with the increasing economic development and income of rural residents, tremendous changes have taken place in the pattern of rural resident consumption. Industrial products are increasingly used in the lives of rural residents. Therefore, the composition of rural human waste tends to be largely a product of urbanization Human waste in rural areas of China is mainly inorganic garbage, such as waste plastics, waste

paper, waste glass, and hazardous waste, and some organic garbage (e.g., crop residue waste and kitchen waste) (Ma et al., 2002; Chai et al., 2012; Yan et al., 2014). More than 80% of the garbage in rural areas is discarded carelessly without any further processing (Guan and Qiu, 2008; Wang et al., 2011; Yao et al., 2009).

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

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

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

2.2.3 Biomass burning of forest/grassland fires. The estimation of burned biomass in this very simple method have lots of problems. AR is the damaged area, in fact, it is the burned area, they are far different. Burned area data were usually derived from satellite data for such a large area of China. It is basically wrong that the authors used the statistics data to allocate them according to the fire counts. Since fire counts does not linearly correspond with the burned area. Please refer to MCD64A1 burned area product with 500 m resolution, which has been validated in many ecosystems. Fire

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

Response:

We thank you for pointing out these problems with the methods and data employed in the emission estimation of forest and grassland fires. As the reviewer mentioned, the damaged area is different from the burned area. Our description in the original manuscript was unfortunately confusing, and we have revised the description to clarify our meaning. Furthermore, as the reviewer pointed out, the burned area should be obtained from satellite data rather than statistical data and allocation based on the fire

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

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

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

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

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

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

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

The corresponding figures and tables have been revised.

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

Response:

We thank you very much for your comment. According to the reviewer's suggestion, we have re-estimated forest and grassland fire emission based on the bottom-up approach and high-resolution satellite data. The total annual forest and grassland fire emission of CO₂ was determined in this way to be 10.9 Tg, where it was 1.68 Tg in the original manuscript. The total biomass burning emission of CO₂ is therefore 675 Tg. Figures 2, 8, and other relevant figures have been revised accordingly. As for the large differences between the CO₂ emissions reported in the Global Fire Emissions Database v4 (GFED4) and those reported in the present study, these could be attributed to the following reasons.

First, the biomass burning emission inventory in this study included the crop straw domestic combustion, an important biomass burning source in China, with a contribution accounting for 68% of the total CO₂ emission. This source was not included in the GFED4.

Second, the emissions of crop straw open burning were estimated in the present study based on the specific EFs and the amount burned for each type of crop straw. The emission in GFED4 was estimated according to the burned area based on a constant EF. The agricultural burned area was derived from the MODIS MCD64A1 product (~500 m resolution). Despite the efforts made to improve the direct broadcast mapping algorithm employed in the MCD64A1 product, the product has a minimum detectable burn area size, which is greater than the size of many agriculture waste burn sites. Therefore, numerous small and scattered agricultural fires would not be detected (McCarty et al., 2007; Giglio et al., 2013; Shi et al., 2015). This is particularly the case in China, where the open burning of crop residue tends to be conducted by individual

families (Liu et al., 2015), resulting in agricultural burning that often occurs over small areas, which is then undetected. The results in the present study were compared with the results of other research. The CO₂ emission of agricultural crop residue open burning in China in 2012 was estimated at 184 Tg by Sun et al. (2016), which is similar to the 207 Tg value obtained in our study. In addition, the total CO₂ emission by biomass burning published in most literature (Cao et al., 2005; Yan et al., 2006; Lu et al., 2011; Tian et al., 2011) is similar to the value obtained in our work, with differences ranging from 0.7% to 40.0%. This comparison indicates that the CO₂ emission estimated in our paper is relatively credible.

Figure 7:

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

Response:

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

excrement) within several urban regions of the North China Plain. Consequently, the use of the relatively high-resolution emission inventory allocation could better represent the actual conditions.

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

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

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

Response:

We thank you very much for your question. The differences in the cited pollutant emissions between this study and that of Lu et al. (2011) are most likely attributable to the selection of EFs. The higher estimations of EC and OC relative to those obtained by Lu et al. (2011) are mainly due to our use of higher EC and OC EFs for crop straw and firewood domestic combustion. The EC and OC EFs employed in the present study were selected from the work of Li et al. (2007), which were measured in representative rural areas across China to determine the characteristics of household biofuel combustion emission. The EFs employed in Lu et al. (2011) were constant values for

different crop straw types, and were derived from measurements conducted outside of China (Reddy and Chandra, 2002). The lower values of CH₄ and NH₃ emissions relative to those obtained by Lu et al. (2011) is mainly due to the lower EFs employed in the present study for crop domestic burning. The crop domestic EFs for CH₄ and NH₃ employed in Lu et al. (2011) were constant values for different crop straw types. The EFs used in the present study were specific for each type of crop straw, which were updated according to published reports of localized measurements conducted in China. The lower estimations of VOC and PM₁₀ relative to those obtained by Lu et al. (2011) are the result of the employment of lower EFs for in-field crop residue burning and firewood combustion. The VOC and PM₁₀ EFs employed in Lu et al. (2011) did not distinguish between different crop straw types, and were derived from measurements conducted outside of China (Street et al., 2003; Reddy and Chandra, 2002). The specific EFs for various crop straws employed in our study were derived from the Chinese guide for compiling atmospheric pollutant emission inventories for biomass burning published in 2014.

Reference

- Chai, C. Y.: Study on applicable technology for rural solid waste classification and recycling, Master Thesis, Zhejiang University, China, Shanghai, 2012 (in Chinese).
- Chen, C., Wang, H. H., Zhang, W., Hu, D., Chen, L. and Wang, X. J.: High-resolution inventory of mercury emissions from biomass burning in China for 2000–2010 and a projection for 2020, *Journal of Geophysical Research–Atmospheres*, 118, 12248–12256, doi: 10.1002/2013jd019734, 2013.
- Guang, D. X. and Qiu, C.: Preliminary Research of the Situation and Countermeasures of Pollution of Rural Domestic Garbage, *China Resources Comprehensive Utilization*, 26, 29-31, 2008 (in Chinese).
- Li, X. H., Wang, S. X., Duan, L., Hao, J. M. and Nie, Y. F.: Carbonaceous Aerosol Emissions from Household Biofuel Combustion in China, *Environ. Sci. Technol.*, 43, 6076–6081, doi: 10.1021/es803330j; 2009.
- Ma, X. J., Chen, Y.: Problem of rural living rubbish and its solutions, *Energy and*

- Engineering, 25-27, 2002 (in Chinese).
- Shon, Z. H.: Long-term variations in PM_{2.5} emission from open biomass burning in Northeast Asia derived from satellite-derived data for 2000–2013, *Atmos. Environ.*, 107, 342–350, doi: 10.1016/j.atmosenv.2015.02.038, 2015.
- Sun, J. F., Peng, H. Y., Chen, J. M., Wang, X. M., Wei, M., Li, W. J., Yang, L. X., Zhang, Q. Z., Wang, W. X. and Mellouki, A.: An estimation of CO₂ emission via agricultural crop residue open field burning in China from 1996 to 2013, *Journal of Cleaner Production*, 112, 2625–2631, doi: 10.1016/j.jclepro.2015.09.112, 2016.
- Sun, J. S., Peng, H. Y., Chen, J. M., Wang, X. M., Wei, M., Li, W. J., Yang, L. X., Zhang, Q. Z., Wang, W. X. and Mellouki A.: An estimation of CO₂ emission via agricultural crop residue open field burning in China from 1996 to 2013, *Journal of Cleaner Production*, 112, 2625–2631, 2016.
- Tansey, K., J. M. Gregoire, P. Defourny, R. Leigh, J. F. Pekel, E. van Bogaert, and E. Bartholome (2008), A new, global, multi-annual (2000 – 2007) burnt area product at 1 km resolution, *Geophys. Res. Lett.*, 35, L01401, doi:10.1029/2007GL031567.
- van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S. and Arellano, A. F.: Interannual variability in global biomass burning emissions from 1997 to 2004, *Atmos. Chem. Phys.*, 6, 3423–3441, 2006.
- Yan J., Wang Z. W. Zhou Y. J. and Zhang C.: Generation Status and Treatment Mode of Rural Domestic Refuse in China, *China Environmental Protection Industry*, 49-53, 2014 (in Chinese).
- Zhang, Y. S., Shao, M., Lin, Y., Luan, S. J., Mao, N., Chen, W. T. and Wang, M.: Emission inventory of carbonaceous pollutants from biomass burning in the Pearl River Delta Region, China, *Atmos. Environ.*, 76, 189–199, doi: 10.1016/j.atmosenv.2012.05.055, 2013b.
- Zhou, Y. L. and Chen, S.: The type, causes and treatment of rural domestic waste, *China Population, Resources and Environment*, 23, 178-182, 2013 (in Chinese).
- Giglio, L.; Randerson, J. T.; van der Werf, G. R. Analysis of daily, monthly, and annual burned area using the fourth-generation global fire emissions database (GFED4), *J. Geophys. Res.: Biogeosci.*, 118, 317–328, 2013.