Response to comments of Anonymous Referee#2

Dear Referee#2:

We sincerely appreciate your comments which help us to improve the manuscript effectively. The replies to the comments are given below.

(Annotation: The YELLOW HIGHLIGHTs are additions or revise in the manuscript or in the appendix.)

General comments:

This paper measured the emission factors of five miscellaneous sources including the burning of sacrificial incense and joss paper, traditional Chinese barbecue, Chinese style cooking, and fireworks burning. The emission inventories and their spatial and temporal (yearly and monthly) distributions were compiled based on the measurements and surveys. The paper is very informative and provides the results of emissions from some missing sources. However, my major concerns are the reliability of the underlying data in this study. For example, are thirty-eight measurements were convincible enough for these poorly understood miscellaneous sources? Of course, I know that such experiments are rare and labor-intensive. Is the questionnaire sufficiently representative? I believe the questionnaires were done rigorously, but this should be explained. As a kind of unconventional emission sources, more uncertainty analysis and validation might be necessary.

Response:

Thanks for your positive comment and suggestions on this manuscript. We quite agree with you that the representativeness of 38 trials is still inadequate. I think the scientists have no alternative but to do when establishing a new emission inventory with limited data. In fact, former emission inventories have also been established using a single emission factor (Li et al., 2007; Kang et al., 2016). What's more, there are precedents for emission inventory establishment with a relatively small amount of actually measured emission factors (Chen et al., 2009; Shen et al., 2012). We believe it can be accepted at current situation. As the first emission inventory of these types of sources, we try our best to obtain effective datasets with limited funding. The questionnaire was designed carefully and the data was checked. The validation was done. More descriptions of the questionnaire were added in the text and the validation of emission inventory was added in the appendix. Detailed responses for the comments are listed below.

Chen, Y., Zhi, G., Feng, Y., Liu, D., Zhang, G., Li, J., Sheng, G., and Fu, J.: Measurements of black and organic carbon emission factors for household coal combustion in China: Implication for emission reduction, Environ. Sci. Technol., 43, 9495–9500, https://doi.org/10.1021/es9021766, 2009.

Li, X., Wang, S., Duan, L., Hao, J., Li, C., Chen, Y., and Yang, L.: Particulate and Trace Gas Emissions from Open Burning of Wheat Straw and Corn Stover in China, Environ. Sci. Technol., 41, 6052–6058, https://doi.org/10.1021/es0705137, 2007.

Shen, G., Wei, S., Wei, W., Zhang, Y., Min, Y., Wang, B., Wang, R., Li, W., Shen, H., Huang, Y., Yang, Y., Wang, W., Wang, X., Wag, X., and Tao, S.: Emission Factors, Size Distributions, and Emission Inventories of Carbonaceous Particulate Matter from Residential Wood Combustion in Rural China, Environ. Sci. Technol., 46, 4207–4214, https://doi.org/10.1021/es203957u, 2012.

Kang, Y., Liu, M., Song, Y., Huang, X., Yao, H., Cai, X., Zhang, H., Kang, L., Liu, X., Yan, X., He, H., Zhang, Q., Shao, M., and Zhu, T.: High-resolution ammonia emissions inventories in China from 1980 to 2012, Atmos. Chem. Phys., 16, 2043–2058, https://doi.org/10.5194/acp-16-2043-2016, 2016.

Specific comments:

Line 119: “This analyzer was developed by the Key Laboratory of Environmental Optics& Technology (Anhui Institute of Optics and Fine Mechanics, CAS) based on the thermaloptical method (Ding et al., 2014). The analyzer showed reliable stability and repeatability.” How the analyzer stable and repeatable should be described here. What about the response, accuracy, time resolution of this device? How to calibrate?

Response:

Thanks for the comments. The instrument stability, repeatability, accuracy, time resolution, calibration, etc., are added in detail in **Text S1** as following.

Text S1: The online carbonaceous aerosol analyzer (OCAA) was developed by the Key Laboratory of Environmental Optics & Technology, Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences (Ding et al., 2014). Repeated testing of standard samples showed a relative standard deviation of 1.5% for the analysis of OCAA. When the sampling flow is 8 L min^{-1} and the sampling time is 30 min, the OCAA can detect the lowest concentration of particulate matter containing carbon as 0.23 μgC m^{-3}. The time resolution of the OCAA is 1 or 2 h. In addition, the OCAA can be set to a single sampling time as needed. The pure substance sucrose of OC was used to configure a series of sucrose solutions with different concentrations. The sucrose solutions with the same volume and different concentrations were used for OCAA analysis. The ratio of peak area between sucrose solution and internal standard was taken as the ordinate, and the carbon content of sucrose solution was taken as the abscissa to obtain the standard curve for instrument calibration.

Line 129: "*M* was the fuel consumption quality (kg)" What is fuel consumption quality?

Response:

Thank you for this comment. There is an ambiguity here. We wanted to describe the mass of the materials used in each tail of experiments. We have revised it in the manuscript (Line 130–131: *M* was the mass of the material used in each tail of experiments (kg).)

Line 170: "The original consumptions of sacrificial incenses, joss paper, and fireworks, were from a household investigation. We got the per capita consumption of sacrificial incenses, joss paper, and fireworks in each province. The data were adjusted to overcome the problem of insufficient sample size. In China, sacrificial activities mean honoring ancestors, and they mainly take place in temples or graveyards. Most traditional graveyards would be placed in hills that might be covered with vegetation. The data on the consumption of sacrificial incenses and joss paper will be revised based on the number of temples (data from POI) and frequency of forest fires caused by sacrifices (data from China Forestry Statistical Yearbook)" This paragraph mainly introduces the quantification method of FMS activity, which I think is very important. However, the description is somewhat simple and obscure. For example, how the household investigation was conducted? The authors only provided some brief introductions in Text S2. But how the questionnaires distributed in different regions, different cities, different ages, and even different nationalities? How to prove that there is an inevitable connection between burning incense and forest fires? What's more, for some plain areas, the incense burning activities are not carried out on the hills.

Response:

Thanks for this query and comment. We quite agree with you that this section is important. According to the suggestion, more details on the questionnaires has been added in **Text S3**. Unfortunately, when we designed the questionnaire, we didn't take into account the gender and nationalities of the respondents. It can be improved in future studies.

In this paper, there are actually two data items of the forest fire. One is the total forest fires. Another dataset, which is stated in the statistical yearbook, is the forest fires caused by sacrificial activities (the index of it in the statistical yearbook is "Forest fires, Number of confirmed combustion sources, Fire used in sacrifice"). The data used in this work is the proportion of forest fire numbers caused by sacrifice to the total forest fire numbers in each province. We believe that this proportion can represent the active level of sacrificial activities in a province. Possible errors in the description of hills and graveyards have also been noted and corrected (Line 170–172: China is a mountainous country with rolling terrain. Most of the inhabitants of non-plain areas chose hills that might cover vegetation as the site of graveyards.).

Text S3: To understand the consumption of sacrificial incenses, joss paper, and fireworks, we have organized household investigations in China. We have investigated the population during the Chinese New Year, address (in the urban or rural region), the time when local fireworks were prohibited, the date or festivals of setting off fireworks, the date or festivals of burning joss paper and sacrificial incense, the quantities of fireworks, joss paper, and sacrificial incense that per capita consumed each year. We did not design the questionnaire to ask about the gender, or nationality of the respondents, but in the process of the questionnaire, we tried to ask the older person of each family. Our questionnaire was based on provinces (27 provinces were covered), and the distribution of the questionnaire is shown in **Figure S15**. Since the burning of fireworks is concentrated during the Chinese New Year, and the population migration during the Chinese New Year is huge in China. The registration or permanent population commonly mentioned in the questionnaire was not applicable in our work. Some families did not give accurate data on the consumption of fireworks, but the approximate volume of fireworks or the number of Xiang of firecrackers (firecrackers are made of thousands of small units connected in series, each unit can be called "Xiang" in Chinese; we thought the firecrackers were also a kind of fireworks). The consumption of this family was estimated according to the local quality of unit volume or Xiang. Similar situations can be observed for the sacrificial incense and joss paper. In addition to the festivals like Chinese New Year or Lantern festivals, some respondents also gave the dates of marriage, funeral, childbirth, and housewarming, that would burn fireworks. Given that these were only relatively occasions, thus these dates were considered as other days than the festivals mentioned in **Text S5** and **Figure 7**. In addition, in the survey, we found that some residents were not clear about the specific quality of fireworks set off at each festival, but would flexibly be changed according to the quantity of fireworks or firecrackers purchased in the year (such as the number of fireworks boxes and the number of whole rolls of firecrackers). Therefore, we considered the proportion of the occurrence number for each festival to the number for all dates in the questionnaire as the proportion of fireworks set off during the festival in the whole year. For example, if the word "Chinese Spring Festival" appeared 100 times in the questionnaire of a province, and the word "Chinese Spring Festival", "Chinese New Year's eve", "Lantern Festival", and other possible words, have appeared 250 times, then we consider that the fireworks set off during the Spring Festival in this province account for 100/250=40% of the whole year. Finally, 2461 valid questionnaires were collected.

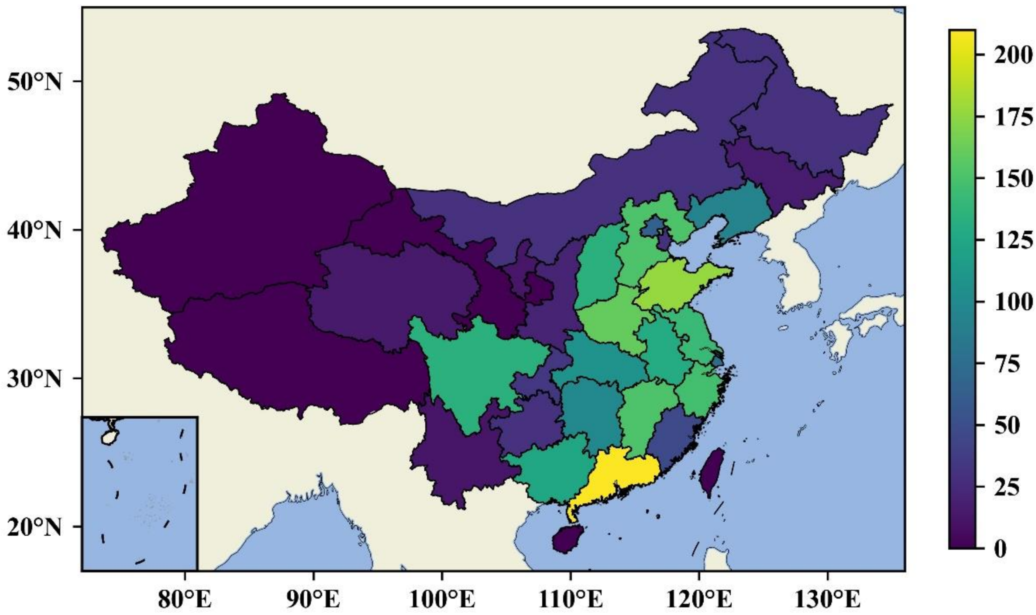

Figure S15 The distribution of questionnaire numbers obtained in each province.

Table 1: Before these emission factors can be applied to the estimation of emission inventories, some remarks about the reliability of these emission factors are required. Also, why are the BC and EC emission factors so different for some sources?

Response:

Thanks for the suggestion and query. As the lack of reports on relevant emission factors, we cannot make a detailed comparison with formers. We have added possible comparison of emission factors in the manuscript (Line 263–271: Multiple factors, such as fuel properties (Chen et al., 2009; Shen et al., 2014; Cheng et al., 2019), combustion condition (Cheng et al., 2019), and stove properties (Shen et al., 2014; Chen et al., 2015), affected the emission of CA from combustion sources. Similarly, CA emissions from FMS were dominated by diverse factors. Results in previous studies were also applicable in this study. For example, the emissions from environmental or aromatic incense were lower (Lee and Wang, 2004; Lui et al., 2016), and cooking fatty pork generated higher emissions (Saito et al., 2014). In addition, the previous study showed higher EF_{OC} (0.779 g kg^{-1}) and EF_{EC} (0.339 g kg^{-1}) for sacrificial offerings (Zhang et al., 2019b). The huge differences in EFs were highly possible (Liu et al., 2015), and more detailed research is needed to expand the datasets of EFs for FMS in the future.)

We think that the large difference between BC and EC emission factors is caused by the characteristics of the source itself and the different analytical methods used by the instruments. According to different measurement techniques, the analytical result of the thermal method was called elemental carbon (EC), and the result of the optical method was named as black carbon (BC). Since BC (or EC) was the mixture of multiple substances, different measurement methods might emphasize different components, leading to the bias in the final concentrations and poor comparability of the measurement results. In addition, FMS is a kind of less studied source, and the regulars obtained from other sources might not be applicable to FMS.

Figure 4: Is there any evidence to prove the surge in emissions of firework in 2014 was correct, rather than a statistical error?

Response:

Thank you for this query. This comment is consistent with that raised by another reviewer. We are very pleased to have such a pertinent comment. We also realize that answering this problem is quite necessary. The surge in consumption was due to the huge sale volumes in statistic data, which might caused by the destocking after that the air pollution control plan was implemented. We attempted to collect the direct observation data. The Air Pollution Prevention and Control Action Plan was published on September 10, 2013, and the Chinese New Year's eve (CNE) and Chinese Spring Festival (CSF) were on February 9, and February 10. The CNE and CSF in 2014 were on January 30 and January 31. Therefore we did not collect the observation data of air quality for the CNE and CSF of 2013. Corresponding data for the year of 2014 were only available for about half of the cities. Thus, we adopted other datasets which have been cited for many times (DOI: 10.5281/zenodo.6398971; Wei et al., 2021; Wei et al., 2020) to verify our results. Considering that the firework burning was mainly concentrated in rural regions after 2013, we conducted a correlation analysis between the FMS **ACS_{BrC} emissions on New Year's Eve** and the **$PM_{2.5}$ concentration in non-urban regions on New Year's Eve** after 2013, and the result can be accepted (r=0.85, p<0.05). The relationship was also significant between 2000 and 2018 (r=0.59, p<0.01). The variation trends of the emission and $PM_{2.5}$ concentration were similar. Therefore, we believe that the emission peak in 2014 is possible. The relative discussion was added in **Text S6** and Line 354–355 (The surge in sales might have been caused by destocking after that the Air Pollution Prevention and Control Action Plan (APPCP) was implemented.)

Text S6: The surge in ACS_{BrC} emission in 2014 might be out of the ordinary. We attempted to use the $PM_{2.5}$ concentration dataset (Wei et al., 2020, 2021) to verify the accuracy of the inventory. The ACS_{BrC} emissions mainly came from fireworks burning (**Figure S8**), and most of the fireworks were burnt in rural regions during the Chinese New Year (**Section 3.3.3**). We conducted a correlation analysis between the FMS ACS_{BrC} emissions and $PM_{2.5}$ concentration in non-urban regions for the New Year's Eve. The results were shown in **Figure S16**. There was a positive correlation (r=0.59, p<0.01) between the ACS_{BrC} emissions and $PM_{2.5}$ concentration. The correlation (r=0.85, p<0.05) was even higher if we focus only on the period after 2013. Thus, the emission surge in 2014 was possible. However, more accurate verification is still needed to be carried out by chemical transport models in the future.

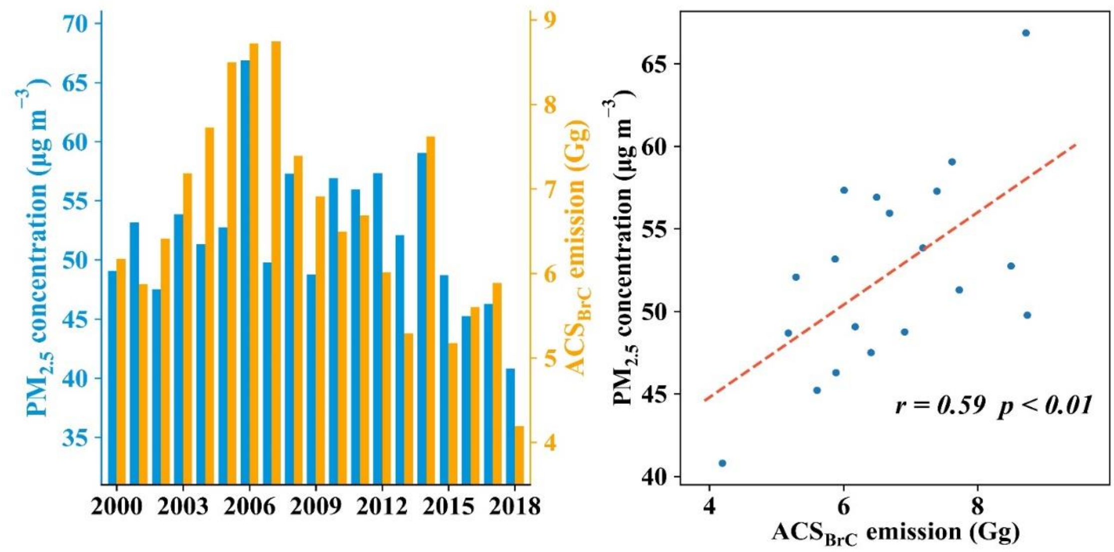

Figure S16 $PM_{2.5}$ concentration and ACS_{BrC} emission from FMS in rural China in 2000–2018 and the correlation between them.

Wei, J., Li, Z., Lyapustin, A., Sun, L., Peng, Y., Xue, W., Su, T., and Cribb, M. Reconstructing 1-km-resolution high-quality $PM_{2.5}$ data records from 2000 to 2018 in China: spatiotemporal variations and policy implications. *Remote Sensing of Environment*, 2021, 252, 112136. https://doi.org/10.1016/j.rse.2020.112136

Wei, J., Li, Z., Cribb, M., Huang, W., Xue, W., Sun, L., Guo, J., Peng, Y., Li, J., Lyapustin, A., Liu, L., Wu, H., and Song, Y. Improved 1 km resolution $PM_{2.5}$ estimates across China using enhanced space-time extremely randomized trees, *Atmospheric Chemistry and Physics*, 2020, 20(6), 3273-3289. https://doi.org/10.5194/acp-20-3273-2020