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Interactive comment on “Patterns in atmospheric carbonaceous aerosols in China: emission estimates and observed concentrations” by H. Cui et al.

H. Cui et al.

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We thank very much for the valuable comments from reviewer 1, which help us improve the quality of our manuscript. Following is our point-by-point responses to the comments and corresponding revisions.

1. The manuscript presented an updated emission inventory of anthropogenic organic carbon (OC) and elemental carbon (EC) from China, and a thorough analysis of the characteristics of carbonaceous aerosol including spatial, temporal distributions, size distribution, and share of secondary organic compound (SOC) by reviewing existing

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observation studies. The manuscript also used observations to test the levels and inter-annual trends of the calculated emission inventory and proposed possible improvements for future emission estimation. Overall, the manuscript is well organized, professionally written with adequate data, tables and figures, and falls in the scope of Atmospheric Chemistry and Physics. The manuscript has provided a more detailed and broader view on the current situation of carbonaceous aerosols in China. I would recommend the publication of this manuscript in Atmospheric Chemistry and Physics after the following comments have been addressed.

Response and revisions:

We thank the reviewer's positive comment on the paper.

2. The authors should clearly highlight the improvement of emission estimation in this study compared with their previous work, or even with other studies, at the beginning of the methodology part. It has been mentioned sparsely in the manuscript on the difference from previous work done by the same group. However, I think this point merits a more systematic and detailed discussion.

Response and revisions:

We thank the reviewer's important comment. We have added a paragraph in lines 151-159 in the revised manuscript to briefly summarize the main improvement in the method used in this work compared to previous inventories:

Compared to previous inventories, improvements are made in the method of current work. First, activity data of certain categories (e.g., biofuel use) are updated with the latest available information, as described later in this section. Second, more detailed classification is applied for residential combustion to better differentiate the emission characteristics of various subcategories. The third, the emission factor database is modified compared to previous work (Zhao et al., 2011; Lu et al., 2011), with the most recent results from local field measurements incorporated. Clear difference in emis-

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sion factors for given sources is found from previous inventory studies. Finally, the temporal and spatial variability in emission factors is better understood with more local information combined. The details for the latter three will be provided in Section 2.3.

3. The authors should provide more explanation on the sector categories. Kerosene and brick kiln have been recently identified as two important sources of black carbon in South Asia. Have the authors conducted any work on estimate BC and OC emissions from these two sources in China? How are these two sources incorporated in the emission inventory presented in this study?

Response and revisions:

We thank the reviewer's comment. For brick production, we include the emissions in the other industrial process (PRO) category. For kerosene use, some studies indicate that the BC emissions from household lighting were probably underestimated in previous inventory in developing countries such as India (Lam et al., 2012). In China, however, the fraction of kerosene in household fossil use is very small (<1%) according to national energy statistics, and the kerosene lighting is rarely seen even in rural regions, due to increased use of electricity. In this paper, therefore, we do not specify the kerosene use but include its emissions in the industrial and residential oil combustion categories. To better indicate the source category, we have added Table S2 in the revised supplement and have provided the detailed emission values by category.

4. Please check the format of the references in the main text, as most of them have first name initial for the in-text citation, which need to be removed.

Response and revisions:

We thank the reviewer's reminder. Yes, some references in the main text have first name initial, in order to differentiate papers that were published at the same year, by different first authors with the same family name.

For example, Q. Zhang et al. (2012) and X. Zhang et al. (2012) indicate the following

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two papers, respectively:

Zhang, Q., He, K. B., and Huo, H.: Cleaning China's air, *Nature*, 484, 161-162, 2012.

Zhang, X. Y., Wang, Y. Q., Niu, T., Zhang, X. C., Gong, S. L., Zhang, Y. M., and Sun, J. Y.: Atmospheric aerosol compositions in China: spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols, *Atmos. Chem. Phys.*, 12, 779-799, 2012.

5. First and second paragraph in Section 2.3, how did the authors calculate the uncertainty associated with the share of different sectors?

Response and revisions:

We thank the reviewer's comment and admit that we did not clearly indicate the meaning. The number with a range does not indicate the uncertainty but the variation of sector shares for different years. We have modified the sentences in the related paragraphs of Section 2.3. Following is an example in lines 238-239 in the revised manuscript:

During the research period, the share of residential sector to total EC emissions is estimated to range 49-55% for different years.

6. Page 8994, Line 14 – 15, I would suggest the authors presented the percentage difference between current and previous studies, instead of absolute difference of emission values. Response and revisions:

We thank the reviewer's suggestion and the percentage differences are presented instead of absolute values in lines 322-326 in the revised manuscript:

Our estimates of OC emissions are roughly 33-47% lower than those of Lu et al. (2011) for different years, and 23-27% lower than those of Zhang et al. (2009), Lei et al. (2011) and REAS 2, even though they did not include the emissions from biomass open burning, an important OC source that is estimated to contribute 400-600 Gg OC

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emissions per year according to Lu et al. (2011) and this work.

7. Page 8994, Line 16, the authors should be clear here about biomass open burning. Does it include forest fire? I would assume not because that is not anthropogenic, but the authors should clarify this point.

Response and revisions:

We thank the reviewer's reminder. Biomass open burning in this paper does not include forest fire. We have clarified this in lines 136-137 in the revised manuscript:

Residential biomass combustion contains household biofuel use and open biomass burning (forest fire not included).

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Lei, Y., Zhang, Q., He, K. B., and Streets, D. G.: Primary anthropogenic aerosol emission trends for China, 1990–2005, *Atmos. Chem. Phys.*, 11, 931-954, 2011.

Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010, *Atmos. Chem. Phys.*, 11, 9839-9864, 2011.

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