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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.

yuzhao@nju.edu.cn

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We thank very much for the valuable comments from reviewer 2, which help us improve the quality of our manuscript. Following is our point-by-point responses to the comments and corresponding revisions.

1. This paper attempts to update an emission inventory of anthropogenic organic carbon (OC, including primary OC and secondary OC) and elemental carbon (EC) based on previous reported observational studies. I have serious concerns about the section 3 especially on SOC part because of the limitation of SOC estimation method used in

the study. Without more comprehensive understanding of assumptions and limitations of EC tracer method, which is used to be estimate SOC, the present work towards SOC part is inaccurate or even erroneous. The paper should clear state in the paper what are new findings about carbonaceous aerosols in China. I suggest including more new findings towards carbonaceous aerosols studies (observations and source apportionment studies) in China. Discussion of source and POC/SOC source apportionment based on OC/EC ratios alone is not sufficient. From the method used in the current work, SOC and biomass burning OC could not be separated, therefore, the conclusion of the enhanced SOC across in China can be misleading. Similarly, the trend of OC/EC ratios should be carefully examined by re-considering measurement uncertainty in different studies by different analytic methods. In my opinion, the methodology applied by the authors does not allow distinction of SOC and POC in a national scale.

Response and revisions:

We thank the reviewer for his/her very crucial comment. In this general comment, the reviewer raised three important issues: (1) the limitation of SOC determination with EC-tracer method; (2) limitation in source apportionment from OC/EC or POC/SOC values; and (3) uncertainties in national-scale pattern of carbonaceous aerosols from different sampling and analytic methods. We will respond those issues and present the corresponding revisions in detail in the answers of Questions 10, 9, and 7-8, respectively. We would also like to make a quick response to issue (1) here, that none of the SOC results we apply in the current work were estimated by us but the original studies, although we present the principal method in the revised manuscript (revised equations (3) and (4)). We thus assume that the individual condition for SOC formation during each campaign period has been considered by corresponding study and the most suitable way to determine (OC/EC)pri in eq. (3) and thereby SOC was applied by the original study. The estimated SOC level was then the "best guess" for each campaign. We have clarified this in lines 501-504 in the revised manuscript. In addition, new studies on carbonaceous aerosol observation and source apportionment in China

have been reviewed and added to current work. Table S4 in the supplement has been expanded by including more results from campaign studies. New datapoints have also been included in the revised Figures 4 and 8.

We also follow the reviewer's comment and briefly summarize the new findings of this work in Conclusion section (with detailed information described in Section 2, 3 and 4), mainly in the following aspects.

First, the work presents an updated emission inventory of anthropogenic OC and EC from China, with an improved source category framework that incorporates the most recent emission factor data from domestic measurements. (We also want to clarify here that SOC is NOT included in the emission inventory which focuses only on primary emissions.) From the new information, in particular, OC emissions are estimated to be smaller than previous inventories. Second, by reviewing existing observation studies on OC and EC concentrations over the country, the work provide a comprehensive picture of carbonaceous aerosol pollution in China during a period of rapid economic development and improved pollution controls. Finally, the estimated levels and inter-annual trends of primary carbonaceous aerosol emissions are evaluated using available observed ambient concentrations in the country.

2. Table 1 does not include EF for open biomass burning. Because field burning of agricultural residues is a very common practice during China's harvest season and is thought to be major sources of OC and EC in specific seasons, this should be considered into the inventory. EF in small stoves would not be the same as that in open burning. I suggest also list the other EFs (transportation) in this table.

Response and revisions:

We thank and agree with the reviewer's important comment. Yes, the open biomass burning is an important source of carbonaceous aerosols and it has already been included in the emission inventory. Different EFs for biofuel stoves and open burning are applied in the current work. Table 1, however, focuses on the stoves because con-

siderable field measurements on stoves have been recently conducted but the results were not fully used in previous inventories. We thus incorporate those newly published data and update the EF with probability distribution, and present the results in Table 1. For biomass open burning, very few new field measurements are available, thus we keep the values and probability distributions unchanged from our previous inventories (Zhao et al., 2011). We have clearly stated this in lines 207-210 in the revised manuscript. We have also added a table (Table S1 in the supplement) to present the EFs of transportation, as suggested by the reviewer.

3. Sec 2.3: "...residential sector is estimated to have accounted for 52±3 % of total EC emissions..". The uncertainty of 3% seems very small. Did the authors use error prolongation from all possible uncertainties from all factors in the equation? Or 3% only represent standard derivation of results from different years in 2000-2012. Please clarify it and show the detailed approach to estimate uncertainty in the SI.

Response and revisions:

We thank the reviewer's comment and admit that we did not clearly indicate the meaning in the original manuscript. The number with a range does not indicate the uncertainty but the variation of sector shares for different years. We have modified the sentence in lines 238-239 in the revised manuscript (and similar sentences in the second and third paragraphs of Section 2.3 have also been modified):

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

Regarding uncertainty analysis, we have briefly described the approach in lines 160-166 in the revised manuscript. There are too many data and assumptions involved in the analysis, and most of them were substantially described in our previous work (Zhao et al., 2011). In current work, therefore, we address the main improvements (particularly on emission factors with probability distributions for small stoves) in details in Section 2.2 in the revised manuscript.

4. "During the period, emissions from the residential sector increased by 34 %, principally due to the growth of coal consumption". Uncertainty should be given after 34%. How about the variation of EF from coal combustion during 2000-2012? If EF from coal is decreased due to high combustion efficiency in the modern stove, the total emissions will not simply dependent on coal consumption.

Response and revisions:

We thank the reviewer's important comment. The uncertainties of the growth rates have been calculated and provided in the revised manuscript in lines 241-242 in the revised manuscript: "During the period, emissions from the residential sector increased by 34% (95% CI: 23%-61%), principally due to the growth of coal consumption". Correspondingly, we have moved the description of uncertainty analysis approach to Section 2.1 (lines 160-166 in the revised manuscript), earlier than this part that includes the results of emission uncertainty.

We agree with the reviewer that the average EF from coal combustion would probably decrease due to improved combustion efficiency. However, current available field measurements are still insufficient to provide a clear time-series trends of EC emission factors. We acknowledged this limitation in lines 647-651 in Section 4.1. In this case, we follow the reviewer's suggestion and explore the effects of changed coal type on emission factors from residential coal combustion. As can be seen in Table 1, the EC EFs from briquette combustion are smaller than those from chuck combustion. Thus we evaluate the trends in average EC EF from residential coal combustion and the national fraction of briquette use for 2000-2012, and find they are in consistent with each other, as shown in the added Figure S1 in the revised supplement. At the national scale, the fraction of briquette use is still small, and it started to decrease from 2006 following the growth in earlier years, leading to similar values and thereby the national averages of EC EF for 2000 and 2012. Therefore the difference in annual EC emissions for the two years comes mainly from the different coal combustion levels. Although there is no significant change in EF at national scale, the fractions of briquette

use varied considerably by region, and diverged levels and trends in provincial-level emission factors are found and used in this work. We have added these discussions in lines 203-207 in the revised manuscript.

5. Line 28 in 8991 line 1 in Page 8992: the trend of OC/EC ratios in different years is very likely smaller than the uncertainty. For example, OC/EC uncertainty alone in any thermal-optical methods will not be better than 10% (Schmid et al., 2001; Schauer et al., 2003; Hitzenberger et al. , 2006). The EF in the current study includes literature values from previous studies in which different OC/EC method were used. As a result, the uncertainty in OC/EC ratio alone will be easily higher than 20%. One should carefully evaluate such a trend when OC/EC is decreased from 1.58 in 2000 to 1.52 in 2012 by <4%.

Response and revisions:

We thank the reviewer and agree with his/her important comment. As indicated by the papers suggested by the reviewer, uncertainties definitely exist in OC/EC measurements for both ambient concentrations and emissions, with different, or even the same methods/instruments/protocols (Schmid et al., 2001; Schauer et al., 2003). Therefore, we first checked the analytical methods of emission factors that are included in our database for emission inventory development. For residential combustion, as summarized in Table S3 in the revised manuscript, around 77% of the samples in selected field measurements were analyzed with the TOT method, while the left with TOR. For industrial and transportation sector, most studies apply TOR (Wang et al., 2009; Zhang et al., 2009a, b). We thus acknowledge that the different analytical methods potentially generate big uncertainty for OC and EC EF measurements. Second, in the revised manuscript, we have estimated the uncertainty of annual OC to EC emission ratio, (OC/EC)emi, from the bottom-up emission inventory, using the Monte-Carlo simulation. For 2000 and 2012, the 95% CIs of (OC/EC)emi are calculated at (0.81-2.31) and (0.76-2.21), i.e., -48%~+47% and -48~+49% around the central estimates (lines 257-260 in the revised manuscript). Those values are much bigger than 4%, the differ-

ence between "best guess" of (OC/EC)emi for the two years, as judged by the reviewer. Therefore, we agree with the reviewer that the relatively small inter-annual variability of (OC/EC)emi needs to be carefully and cautiously evaluated. We want to note that the uncertainties of primary emissions and thereby (OC/EC)emi are dominated by parameters related with emission factors by source, as shown in Table 2. We assume that there is no inter-annual variation in those parameters during the research period, even though high uncertainties exist for them in any given year. The changes in emissions and (OC/EC)emi over time at the sector level are thus driven mainly by the varied activity levels and fractions of different emission sources, and they are less associated with the uncertainty for individual year, whether the uncertainty is big or small. Even small decrease in (OC/EC)emi with large uncertainties, given that they are generated using the same methods, is worth noting. We have added those discussions in lines 267-280 in Section 2.3 of the revised manuscript.

6. Sec 2.3. In this section, I think only primary OC is estimated, so it is better to use POC instead of OC when only primary OC is considered.

Response and revisions:

We thank the reviewer's reminder. Yes, in the emission inventory, only primary OC is estimated. The word "emissions" itself indicates the fact. We also state "primary carbonaceous aerosols" in the title of Section 2.

7. Page 8995 lines 19-22: Please clearly OC and EC uncertainty from different analytical methods. If uncertainty of OC and EC is larger than 25%, it indeed would sometimes lead some statistical errors for comparison in this work. For examples, without evaluation of uncertainty from OC and EC measurements in different studies, it is not scientific sound to conclude that "ambient EC concentrations in north are higher than those in south" (page 8998 lines 13-15) because the difference between north and south in urban and remote regions may be not statistical significant if the uncertainty of EC measurement is included in the comparison. The difference of OC/EC ratios be-

tween the north and south may be smaller than the uncertainty from different analytic methods conducted in different years.

Response and revisions:

We thank the reviewer's important comment. We agree with the reviewer that different sampling/analytical methods can lead to considerable uncertainties in observation of ambient carbonaceous aerosol concentrations, similar to emission factor measurement as we response to Q5. Most of the selected observation studies that were at least seasonally representative, however, just applied one method, and very few of them compared the results from different measurement methods. Therefore, it is difficult to directly provide the uncertainty of every individual measurement that is included in this work, and to clearly estimate the uncertainty at the national scale. In limited cities, e.g., Beijing, a few studies have been conducted to evaluate the influence of measurement methods and thermal-optical temperature protocols on OC to EC ratio (Cheng et al., 2011; 2014), and the uncertainty from different methods/protocols reaches 10-40%, which cannot be imply ignored.

Therefore, we expanded Table S4 in the revised supplement by including new results from available campaigns and examined the details of OC and EC measurement methods of every selected studies, including the instrument (online or offline), sampling (denuder included or not), temperature protocol (IMPROVE or NIOSH), and measurement method (TOT or TOR). As summarized in Table S4, IMPROVE_TOR was applied by 65 out of 97 campaign studies (43 out of 59 for urban studies) and was thus the most frequently applied method across the country. In the revised manuscript, therefore, we recalculated the average OC, EC and OC/EC for studies applying the IMPROVE_TOR method for northern urban, southern urban, suburban, rural and remote sites. The averaged OC and EC concentrations in northern urban sites remain higher than those in southern ones, and the gradient of carbonaceous aerosol concentrations and OC/EC from remote to urban sites keeps. Thus we believe the pattern of OC and EC concentrations across the country is reasonable although the result is undoubtedly influenced

by the different measurement methods. We have added the discussions in lines 388-401 in the revised manuscript. We also agree with the reviewer that the difference in OC/EC between north and south urban sites is less conclusive, as urban sites are more influenced by local primary emissions than remote/rural sites. We respond and make corresponding revision on this issue in the answer of Q8 as below.

8. Page 8998 lines 14-16: it is not meaningful to compare OC/EC in north and in south because these results have been obtained in different years (even sometimes in different seasons) and by different methods. There could be many other factors (year/season to year/season variability and/or measurement uncertainty) controlling OC/EC ratios. Response and revisions:

We thank the reviewer's comment and agree that the OC/EC can be affected by the uncertainty from different measurement periods and methods, besides emissions and SOC formation condition. To improve the comparison, therefore, we have made two revisions in the manuscript. First, we expanded the database by including new results from available seasonally representative campaigns, and examined the year and season when the campaign was conducted for every individual study. (Since there are not enough studies for every year between 2000 and 2010, we split the whole dataset into 2000-2005 and 2006-2010.) As shown in the added Figure S3 in the revised supplement, the distribution in north and south does not differ much, implying limited bias from sampling time on the spatial pattern of OC/EC (lines 430-432 in the revised manuscript). Second, as shown in the revised Figure 4, totally 513 data points are included, and 292 and 160 out of them are measured with IMPROVE_TOR and NIOSH_TOT method, respectively. Similar as Figure 4, we have added Figure S4 and S5 in the revised supplement, including data obtained with IMPROVE_TOR and NIOSH_TOT method, respectively. Similar pattern of OC/EC in north and south and gradient of OC/EC can be found as that from Figure 4 (note the rural and remote sites are merged, so are the suburban and urban sites in Figures S4 and S5, attributed mainly to less data points available from a given analytical method). Thus we believe

the uncertainty from measurement period or method would not lead to big bias on the results in OC/EC pattern across the country. We should admit, however, that uncertainty exists in the comparison, as suggested by the reviewer. Since current available campaigns using a certain analytical method for a given type of site (particularly for remote site) are still insufficient, the results can be easily influenced by limited studies. To better understand the OC/EC pattern across the country, more campaigns on varied locations, particularly at remote sites, are recommended. We have added the discussion in lines 446-456 in the revised manuscript.

9. Page 8999 line 1-5: the statement needs references. Biomass burning and coal combustion emissions also could contribute substantially (sometimes may be dominant over vehicle emissions) in both the JJJ and YRD regions. The source apportionment in urban regions of China is still not clear; such a statement should be avoided. More discussions and new literatures should be included in this section.

Response and revisions:

We thank the reviewer's important comment and suggestion. First we need to admit that we did not describe carefully in the original manuscript and the text was ambiguous. We do not aim mainly at source apportionment for a given region (i.e., analyzing the contribution of different emission sources to pollution), since it is relatively complicated and cannot be well conducted depending only on OC/EC, as pointed by the reviewer. Instead, we want to infer and compare the pollution sources for different regions (urban vs. rural, and developed vs. developing), based on the emission inventory and observation of carbonaceous aerosols. In this case, we agree with the reviewer that biomass burning and coal combustion emissions could also contribute substantially (sometimes even dominate) compared with transportation, in JJJ and YRD regions. Transportation in those developed areas, however, is believed to contribute more to air pollution than it does in developing and rural regions. This can be partly supported by the lower OC/EC ratio observed in developed urban areas than rural or developing areas, as the emission ratio of OC to EC for transportation is low among various source

categories. Moreover, the current emission inventory also indicates the larger fraction of vehicles in EC emissions. For example, transportation is estimated to account for 37% of total EC emissions in the YRD provinces (Shanghai, Jiangsu and Zhejiang) for 2012, clearly larger than the national average level at 17%. We have revised the text to avoid confusion and added the discussions in lines 460-465 in the revised manuscript.

Second, we agree with the reviewer that source apportionment is still unclear in urban China, and OC/EC observation could not provide comprehensive information for it. Since the analysis here is related with (but does not focus mainly on) source apportionment, we follow the reviewer's suggestion and have added literatures on source apportionment with other methods. Although the results differ and depend largely on region, period and the method used, transportation is identified as an important source of carbonaceous aerosols, particularly in developed cities. For example, the contribution of transportation to OC in winter Beijing could exceed 70% with a simple ratio method (Zhang et al., 2007). The contribution of transportation to total carbonaceous aerosols were estimated to range 47-96% in autumn and winter in urban Xian, with principle component analysis (PCA) method (Cao et al., 2005). Using chemical mass balance (CMB) method, biomass burning and transportation are estimated to be the most important source of OC in Hong Kong (Li et al., 2012; Hu et al., 2010), while transportation could contribute 30% of excess OC for the Pearl River Delta (PRD) region compared to Hong Kong (Zheng et al., 2011). Studies using isotopic tracer method indicated less contribution of biomass burning but more from fossil fuel in developed urban regions compared to rural and developing ones (Huang et al., 2010; Niu et al., 2013). We have added the discussions in lines 475-487 in the revised manuscript.

10. Page 8999 Lines 17-28: EC-tracer method is a traditional (and very old) method to estimate SOC which is often used in the same campaign by the same OC/EC analytic method. There are several disadvantages in the SOC estimation as already discussed in the current paper. However, such an approach could not be applied in different regions of China when OC and EC results have been derived from different campaigns

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with different methods.

Response and revisions:

We thank the reviewer's crucial comment. First we admit that we did not fully describe EC-tracer method with eq. (3) in the original manuscript. In the revised manuscript, we have revised the equations as follows:

$POC = (OC/EC)pri \times EC + OCnc$

$SOC = OCtot - POC$

where POC and SOC indicate the primary and secondary organic carbon, respectively; OCtot and EC are the observed total OC and EC concentrations, respectively; (OC/EC)pri is the ratio of primary OC and EC emissions with the contribution of SOC excluded (Castro et al., 1999); and OCnc is the OC emissions from non-combustion sources and it is usually small and sometimes overlooked in the calculation.

In the method, determination of (OC/EC)pri is crucial to SOC estimate. (OC/EC)pri can be determined by various ways, including the OC to EC ratio from emission inventory (i.e., (OC/EC)emi), OC to EC concentration ratio from observation when SOC formation is weak and thus the concentrations are dominated by emissions, or the lowest OC to EC concentration ratio (i.e., (OC/EC)min) during the observation. It depends largely on primary emissions and atmospheric chemistry condition during individual campaigns, as pointed by the reviewer. We have added the discussions in lines 492-501 in the revised manuscript.

Second, we need to clarify that none of the SOC results included in the manuscript were calculated with a uniform method of determining the (OC/EC)pri (e.g., as the (OC/EC)min). Instead, they were estimated with different methods by original studies, with the SOC formation condition considered during corresponding campaign periods. Thus we assume that the most suitable way to determine (OC/EC)pri and thereby SOC was applied by the original study. The estimated SOC level was then the "best guess"

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for each campaign. Therefore, we mean reviewing and summarizing those studies across the country could provide general information of SOC formation at the national scale. We have stated this in lines 501-504 in the revised manuscript.

Finally, we agree with the reviewer that EC-tracer method has limitation. Determination of (OC/EC)pri is arbitrary and unable to obtain single OC/EC ratio that represented a mixture of primary sources varying in time and space (Yuan et al., 2006). For example, overestimate of SOC can be made during the period of biomass open burning with very high emission ratio of OC to EC (Ding et al., 2012). Although occasional irregular contributions from sources with a primary OC to EC ratio vastly different from the usual mix of sources could cause errors in estimated SOC, EC-tracer method, as the most widely applied approach across the country, is believed to provide reasonable SOC level at monthly or seasonal average when high frequency measurements are used (Folidori et al., 2006). Some recent studies made improvement on the EC-tracer method. Chen et al. (2014) combined the EC-tracer method and potassium mass balance to reduce the impacts of biomass burning on SOC calculation. Day et al. (2015) modified the criterion of (OC/EC)pri by choosing EC/OC points that are two standard deviations above the mean value, and demonstrated better performance of SOC estimation by comparing the results with those from chemical transport model. We have added those discussions in lines 511-524 in the revised manuscript.

11. Page 9001 Lines 17-25: A high SOC fraction in OC in winter could be also due to high OC/EC ratios in aerosols from biomass burning and/or coal combustion. This part should be POC, which is mistaken as SOC by the EC-tracer method.

Response and revisions:

We thank and agree with the reviewer's comment. EC-tracer method tends to overestimate SOC during the biomass open burning period, as suggested by campaign studies in other cities in the country (Ding et al., 2012; Feng et al., 2013), and we've added this point in lines 556-558 in the revised manuscript.

12. Page 9001 lines 26 to Page 9002 lines 2: Huang et al., (2014) revealed that a possible mechanism of high SOC formation in winter, but this does not mean that SOC formation in summer (under condition of high temperature and more sunlight) is smaller than SOC in winter. Can we consider enhanced condensation of semi-volatile organic compounds at colder temperatures simply as an increase in SOC? SOC generally should imply some photochemical transformations prior to aerosol partitioning, which may not be the case here. Please clear give the definition of SOC.

Response and revisions:

We thank the important comment and agree with the reviewer, as we have stated in lines 572-575 in the revised manuscript: Although the absolute SOC levels are higher in winter, the oxidation reactions from VOC to OC are implied to be faster in summer because of higher temperature and more abundant VOC precursors, accelerating SOC formation and thus elevating SOC/OC.

We also agree that SOC indicates the aerosols from photochemical reactions, as we defined in the Introduction section (lines 63-65 in the revised manuscript). We thus remove the explanation of condensation of semi-volatile organic compounds at colder temperatures. According to original campaign study (P. Zhao et al., 2013), high SOC levels (in terms of absolute concentrations) in winter resulted largely from the stable atmosphere and low temperatures, which can facilitate the accumulation of air pollutants. Lower temperature may also contribute to increase the SOA concentration through the favorable partitioning of oxidation products into the particle phase (Folidori et al., 2006). We have modified the text in lines 558-561 in the revised manuscript. In addition, as we respond to Q11, SOC might also be overestimated by the original study using the EC-tracer method for winter when biomass open burning occurs.

13. Page 8986, Lines 4 Q. Zhang et al., 2012 should be changed to Zhang et al., 2012. The similar mistakes should be corrected in the MS.

Response and revisions:

We thank the reviewer's reminder. Some references in the main text have first name initial, in order to differentiate papers that were published in 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 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.

14. Page 9006 Lines 1-3: This contrast could be also due to the uncertainty from OC/EC measurement and inventory estimation. And the inter-annual trend can be easily biased from the selection of literature values used in the paper.

Response and revisions:

We thank the reviewer's comment and have stressed the point in line 666 in the revised manuscript. We also admit that selection of literatures values affects the inter-annual trend. In the revised manuscript, we have tried our best to collect the results from existing campaign studies, and to select and include the data points in the analysis under the same rule. New observation data have also been added to the analysis. The conclusion still holds that EC concentrations have slightly decreased but OC/EC has slightly increased. Thus the influence of data selection on the inter-annual trend is believed to be limited.

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