We thank the Referee for the constructive feedback. We respond to each specific comment below. The original comments by the Referee are shown in bold italics. Our reply is shown in blue.

1. ***One key conclusion of this manuscript is that EC and OC emissions of the bottom-up emission inventory (INTEX-B) in China are severely underestimated at the national level. After reviewing the methodology carefully, the reviewer thinks the results shown in this study are based on two strong assumptions. First, the EC and OC observations of 10 rural and background sites were reliable, and second, the distribution of emissions in the INTEX-B inventory (i.e., relative values in different cells of gridded emissions) is correct. However, both of them could be problematic. For EC and OC measurements, although the authors used some criteria to filter available datasets reported in the literature, they could still contain large uncertainties, and the "top-down" estimates would be very sensitive to these data. For INTEX-B gridded emissions and seasonality, the uncertainties could be even larger since gridding was based on surrogates like population, land cover, road network, etc., and residential seasonality was estimated based on the dependence of stove operation on mean temperature. Therefore, it is not appropriate to apply "domain-wide scalar scale factors" to INTEX-B gridded inventory directly (Eq. 1). As shown in Fig. 6 and some discussion in Sections 3 and 4 of the manuscript, the simulation results based on the original INTEX-B inventory reproduced the EC measurements in some sites, but underestimated that of several other sites. However, using the "top-down" inventory, simulations were improved for some sites, but overestimated others. This result clearly implies that the distribution of current INTEX-B inventory contains errors. Some places are underestimated, and some places are overestimated. Hence, it is arbitrary to directly conclude that China's EC emissions are severely underestimated at country-wide level, because the distribution errors of emissions alone could also explains the authors' results to some extent. This is also consistent with Hakami et al. (2005) and Kondo et al. (2011)'s results.***
   a. We agree that there is considerable uncertainty in the measured monthly mean surface concentrations used in this study. We added explicit statements in Section 2.3 about the uncertainties of the measurements:

      "Both EC and OC concentrations have considerable day-to-day variability at all sites. The normalized standard deviations of the monthly mean concentrations at background and rural sites are 46% for EC and 33% for OC."

      We also added standard deviations of the observed EC and OC monthly mean concentrations in Fig. 6 and Fig. 7.

      We evaluated the uncertainty of the top-down emission estimates by calculating the standard deviation of the scaling factors using a bootstrapping technique (Sect. 4.1):

      "The standard deviation of the top-down EC emission estimate is ±0.78 TgC yr^{-1}, calculated by combining the top-down uncertainties (standard deviation of the scaling factors from the multiple regression calculated by bootstrapping) in quadrature and assuming the bottom-up uncertainty for biomass burning emissions."

In addition, we added two sensitivity tests on the multiple regression (Sect. 4.1):

"We conduct two sensitivity tests to test the robustness of our multiple regression. First, we add up $c_{\text{residential}}$ and $c_{\text{non-residential}}$ in Eq. (1) and fit the observations against the combined anthropogenic contribution. The resulting estimate for the total anthropogenic EC emissions is 2.91 TgC yr^{-1}. In a second test, we remove the observations at Dunhuang and Gaolanshan from the multiple regression. The resulting estimate for total anthropogenic EC emissions is 2.58 TgC yr^{-1}. Both estimates are within 12% of our original top-down estimate shown in Table 1."

b. We agree that biases between the carbonaceous aerosol measurements and model results suggest not only an underestimation of emissions on a national scale, but also errors in the spatiotemporal distribution of emissions in the bottom-up inventories. We modified the text to point out that the underestimation of the anthropogenic sources is on a national scale, and that the spatiotemporal distributions of emissions are misrepresented.

(Abstract) "Our analysis points to four shortcomings in the current bottom-up inventories of Chinese carbonaceous aerosols: (1) the anthropogenic source is underestimated on a national scale, particularly for OC; (2) the spatiotemporal distributions of emissions are misrepresented; (3) there is a missing source in western China, likely associated with the use of biofuels or other low-quality fuels for heating; and (4) sources in fall are not well represented, either because the seasonal shifting of emissions and/or secondary formation are poorly captured or because specific fall emission events are missing."

(Sect. 3) "This suggests that the EC emissions in the model are too low and misrepresented in space and time."

(Sect. 3) "This suggests that primary emissions and secondary formation of OC are both poorly represented in the model."

(Sect. 3) "This shows that (1) the bottom-up OC emissions are too low and misrepresented spatiotemporally, and (2) the representation of secondary formation in the model is poor."

(Sect. 4.3) "As shown in Fig. 9, the regression slopes for simulated versus observed OC are improved for all seasons, but the model remains unable to capture the variability in the observations. Again, this shows that both the primary emissions and the secondary formations of OC are poorly simulated in the model."

(Sect. 6) "Our top-down emission estimates do lead to high biases at Taiyangshan in central China. This indicates that the emissions in southern China may be biased high in our top-down estimate, potentially consistent with the findings of Hakami et al. (2005)."

(Conclusion) "In summary, our analysis points to four shortcomings in the current bottom-up inventories of Chinese carbonaceous aerosols. Firstly, the anthropogenic source is underestimated on a national scale, particularly for OC, likely due to uncertainties in emissions from small industries, residential combustion, and transportation (Zhang et al., 2009; Lu et al., 2011; Qin and Xie, 2011). Secondly, the spatial and temporal distributions of emissions are incorrect in the current bottom-up inventories. Thirdly, there is a missing source in western China, characterized by a high OC/EC emission ratio and a strong enhancement during the colder months, likely associated with the use of biofuels or other low-quality fuels for heating. The fourth issue is that sources in fall are not well represented, either because the seasonal shifting of emissions and/or secondary formation is poorly captured or because specific fall emission events are missing."

2. ***For "top-down" OC estimates, except for the factors mentioned above, uncertainties of VOC inventories and inadequate secondary aerosol formation mechanism in current models also have large effect on final results.***

   We agree that the simulation of secondary OC in the model is wrong, likely due to error in both the VOC precursor emission inventor and the SOC formation mechanism. We modified and modified the texts throughout the paper to emphasis this point:

(Abstract) "In addition, secondary production of OC in China is severely underestimated."

(Section 3) "Moreover, the model cannot capture the observed OC variability; the correlation coefficients *r* range from 0.08 to 0.26. This shows that (1) the bottom-up OC emissions are too low and spatially and/or temporally misrepresented, and (2) the representation of secondary formation in the model is poor."

(Section 3) "The variability of rural OC away from the urban OC versus EC regression line reflects additional secondary production not represented in the model or primary OC sources not represented in the bottom-up inventories (e.g., at Dunhuang and Gaolanshan)."

(Section 4.3) "Again, this shows that both the primary emissions and the secondary formations of OC are poorly simulated in the model."

3. ***The authors compared their "top-down" estimates with previous "bottom-up" inventories in Section 6, and commented that their estimates are still within the uncertainties of previous studies. As mentioned in the manuscript, their emission estimates "are near the upper limits of" "95% confidence intervals". However, from statistical point of view, such high boundary values are statistically not likely to reach.***
Upon closer inspection, we find that our top-down estimates for the year 2006 are well within the range of uncertainties reported by bottom-up inventories for the years 2005 to 2010. We modified the text:

    "Our top-down emission estimates for both EC and OC for the year 2006 are within the range of uncertainties reported by bottom-up inventories for the years 2005 to 2010 (Zhang et al., 2009; Lei et al., 2011; Lu et al., 2011; Qin and Xie 2011)."

4. ***On the other hand, the authors attributed one reason of low estimates in "bottom-up" inventories to the unrealistic emission factors used by emission inventory developer. However, based on some recent field experiments in India and China (e.g., Venkataraman et al., 2005; Chen et al., 2009), the EC and OC emission factors of biofuels and residential coals in developing countries are actually lower than those used in Streets et al. (2003a), Bond et al. (2004), and Zhang et al. (2009). It means the "bottom-up" estimates could be even smaller when applied with emission factor measurements not from "western societies".***
We thank the reviewer for pointing out the Chen et al. (2009) reference. Chen et al. (2009) calculated very low BC emissions from residential coal burning. This is contradictory to the observed BC concentration enhancements in winter. Moreover, a very recent paper by Qin and Xie (2011) used Chen et al. (2009) but still calculated anthropogenic BC emissions (1.14 Tg yr^{-1} for 2000) similar to that of Ohara et al. (2007) (1.09 Tg yr^{-1}) and Zhang et al. (1.60 Tg yr^{-1} for 2001). We read Chen et al. (2009) carefully and found that reason for their low emission estimates is due to a large assumed share of briquettes. There is currently no good information on the share of briquettes versus raw coal in the residential sector in China. This is another reason why the emission factors are highly uncertain. We added text in Section 6 to explain this point:

    "In particular, the emission factors from residential combustion can vary by an order of magnitude depending on the share of low- versus high-quality fuel (e.g., briquettes versus raw coal) (Zhang et al., 2009; Lu et al., 2011), but there is currently no good statistics for that information."

5. ***In sum, it is very risky and not convincing to conclude that "bottom-up" EC and OC emissions in China in year 2006 are severely underestimated based on the results presented because of the shortcomings of the methodology used in this study. The most possible situation for the current China's EC and OC emission inventory may be that the national total is OK (or at least not severely underestimated), but the spatial and***

temporal variations of emissions are problematic. The reviewer would recommend the authors to add relevant discussion in Section 4, and revise some conclusions correspondingly (e.g., abstract, section 4, 6, and 7).

Please see response 1(b).

6. ***The authors claimed that they use "state-of-the-science bottom-up emission inventories for EC, OC, and VOC". However, the reviewer does not think biomass burning emissions developed by Streets et al. (2003b) are "state-of-the-science" for Asia in 2006. As the authors mentioned in the manuscript, Streets et al. (2003b)'s results represented average burning activities for the mid-1990s. And the following sentence that "van der Werf et al. (2010) showed that Chinese biomass burning emission total for the year 2006 is similar to the average annual biomass burning emission total between the years 1997 and 2009 base on satellite observations" does not prove that Streets et al. (2003b)'s mid-1990s estimates are appropriate for year 2006. The reviewer is wondering why the authors did not use van der Werf et al. (2010)'s monthly gridded datasets for 2006 directly.***

   We actually started out using the GFED3 inventory (van der Werf et al., 2010) but found the biomass burning emissions to be too low (EC: 0.01 TgC yr^{-1}; OC: 0.011 TgC yr^{-1}) due to underestimation of the in-field crop burning. We added text in Section 2.2 to explain why we choose to use the inventory developed by Streets et al. (2003b):

   > "Biomass burning emissions of EC and OC for China and rest of East and South Asia are taken from Streets et al. (2003b), which represents average burning activities for the mid-1990s. There are several more recent biomass burning emission inventories based on satellite burnt area observations (van der Werf et al., 2006, 2010; Song et al., 2010), including the widely-used Global Fire Emissions Database (GFED3) developed by van der Werf et al. (2010). However, these inventories either underestimate or entirely exclude the contribution from in-field crop residue burning, which has been shown to be an important seasonal biomass burning source of carbonaceous aerosols in China (Yang et al., 2008; T. Zhang et al., 2008; Huang et al., 2012). We use the inventory developed by Streets et al. (2003b) for the sake of completeness in the present study. A very recent study by Huang et al. (2012) provided new emission estimates from the in-field crop residue burning in China, and we will explore the use of that inventory in a future study. However, as will be shown in Sect. 4.1, the available measurements used for constraining EC emissions are not sensitive to biomass burning emissions. Thus the choice of biomass burning emission inventories will not have a large impact on the top-down estimates of anthropogenic EC emissions presented in Sect. 4.1."

7. ***"Duncan et al. (2003)" is not included in the reference list.***

   Added. Thank you.

8. ***The authors points out that "there is a missing source in western China, likely***

associated with the use of biofuels or other low quality fuels for heating". Based on the results presented in the manuscript, the reviewer totally agrees with the authors findings. The question is why the authors did not omit the observation data of Dunhuang and Gaolanshan in their multiple regression process, as they had known the emissions in these two sites were wrong. Including observations of these two sites probably makes the "top-down" estimates of EC and OC much higher.

We found that excluding data from Dunhuang and Gaolanshan does not alter the regression result much. This is because there is very little emission in the bottom-up inventory at Dunhuang and Gaolanshan. Thus, the observations at these two sites have very little leverage against the regression. We added text in Sect. 4.1 to describe the sensitivity tests:

> "We conduct two sensitivity tests to test the robustness of our multiple regression. First, we add up $\boldsymbol{c}_{\text{residential}}$ and $\boldsymbol{c}_{\text{non-residential}}$ in Eq. (1) and fit the observation against the combined anthropogenic contribution. The resulting estimate for the total anthropogenic emissions is 2.91 TgC yr^{-1}. In a second test, we remove the observations at Dunhuang and Gaolanshan from the multiple regression. The resulting estimate for total anthropogenic emissions is 2.58 TgC yr^{-1}. Both estimates are similar to our original top-down estimate shown in Table 1."

9. ***The authors used multiple regression method to obtain "domain-wide scalar scale factors" for Chinese anthropogenic residential and anthropogenic non-residential sources. However, they also commented that "the distinction between residential and non-residential sources in the multiple regression is somewhat arbitrary", "and not distinguishable with the limited observation data." In this case, the reviewer is wondering why the authors did not treat the anthropogenic emissions as a whole sector, and instead used a single "domain-wide scalar scale factor" directly to it.***

   We conducted a sensitivity test to do this, but found the resulting emission estimate to be similar to that of our original top-down estimate. We added text in Sect. 4.1 to describe the sensitivity test:

   > "We conduct two sensitivity tests to test the robustness of our multiple regression. First, we add up $\boldsymbol{c}_{\text{residential}}$ and $\boldsymbol{c}_{\text{non-residential}}$ in Eq. (1) and fit the observation against the combined anthropogenic contribution. The resulting estimate for the total anthropogenic EC emissions is 2.91 TgC yr^{-1}. In a second test, we remove the observations at Dunhuang and Gaolanshan from the multiple regression. The resulting estimate for total anthropogenic EC emissions is 2.58 TgC yr^{-1}. Both estimates are similar to our original top-down estimate shown in Table 1."

10. ***The reviewer does not understand why the authors use the winter ratios for April to September and the summer ratios for the rest of the year.***

    This was a typo and has been corrected. Thank you.

11. ***There are some grammatical errors in the manuscript. Some sentences are too long to***

follow. The reviewer strongly suggests the manuscript to be checked by native speakers after revision.

The manuscript has been edited by a native English speaker following the Referee's suggestion.

12. ***Page 28222, line5. "are emitted" should be "is emitted".***

    Fixed as suggested. Thank you.

13. ***Page 28222, line 13. "constructed "from the bottom up" based on" change to "constructed from the "bottom up" approach based on ".***

    Fixed as suggested. Thank you.

14. ***Page 28222, line 27. Add "developed" between "inventories" and "by".***

    Fixed as suggested. Thank you.

15. ***Page 28223, line 3. Add "developed" between "inventories" and "by".***

    Fixed as suggested. Thank you.

16. ***Page 28223, line 18. Add "out" between "16" and "of".***

    Fixed as suggested. Thank you.

17. ***Page 28224, line 23. Add "have been" between "studies" and "measured".***

    Changed to:

    > "In the past decade, many studies have measured EC and OC …"

18. ***Page 28227, line 4-7. Please rewrite this sentence.***

    Changed to:

    > "The equilibrium partitioning is as described by Chung and Seinfeld (2002), except that in this study the semi-volatile SOA is allowed to partition onto all preexisting OC and inorganic aqueous aerosols. This provides an upper estimate of semi-volatile SOA production."

19. ***Page 28227, line 28. "tests to by" should be "tests by". And please rewrite this long sentence.***

    Changed to:

    > "We also conduct sensitivity tests by turning off Chinese EC and OC emissions from each of the three source sectors (anthropogenic non-residential, anthropogenic residential, and biomass burning), in turn and all at once. This is done to evaluate the contributions to surface concentrations from each source sector and from non-Chinese sources."

20. ***Page 28228, line 3. Add comma after "OC".***

    Fixed as suggested. Thank you.

21. ***Page 28230, line 2. "whose emissions" should be "emissions of which".***
    Fixed as suggested. Thank you.

22. ***Page 28235, line 19-21. Please rewrite this sentence.***
    Changed to:
    > "This shows that (1) the bottom-up OC emissions too low and spatially and/or temporally misrepresented, and (2) the representation of secondary formation in the model is poor."

23. ***Page 28241, line 1. "concentration improve" should be "concentrations are improved".***
    Fixed as suggested. Thank you.

24. ***Page 28241, line 6. "OC improve" should be "OC are improved".***
    Fixed as suggested. Thank you.

25. ***Page 28241, line 8. Put "either" after "the model".***
    Fixed as suggested. Thank you.

26. ***Page 28241, line 25. "emission estimates do a better job than: : :". Please change to other words.***
    Changed to:
    > "This indicates that the top-down estimates better represent the EC and OC emissions affecting Chinese urban and non-urban air, compared to the bottom-up inventories."

27. ***Page 28242, line 2. "suggest" should be "suggests". "is capturing" should be "captures".***
    Fixed as suggested. Thank you.

28. ***Page 28242, line 4-6. Please rewrite this sentence.***
    Changed to:
    > "However, the model OC low-bias is significantly correlated with observed EC in eastern China for spring, fall, and winter, as well as in western China year-round (not shown). This suggests that the primary OC sources are still too low in our top-down estimate."

29. ***Page 28242, line 12. Add "high" between "of" and "precursor".***
    Fixed as suggested. Thank you.

30. ***Page 28243, line 3. Change "the findings" to "those".***
    Changed "the findings" to "the results"

31. ***Page 28243, line 4. "a larger isoprene emission" should be "larger isoprene emissions".***

Fixed as suggested. Thank you.

32. Page 28243, line 8. "double" should be "twice higher".
Changed "double" to "approximately twice". Thank you.

33. Page 28244, line 4. "Ohara et al., 2007" is not included in the reference list.
Added. Thank you.

34. Page 28248, line 1. "statics" should be "statistics".
Fixed as suggested. Thank you.

References

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Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., Hayasaka, T., An Asian emission inventory of anthropogenic emission sources for the period 1980-2020, Atmos. Chem. Phys., 7, 4419-4444, doi:10.5194/acp-7-4419-2007, 2007.

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