

## ***Interactive comment on “Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution” by T.-M. Fu et al.***

### **Anonymous Referee #2**

Received and published: 14 December 2011

#### General comments:

In this work, Fu et al. used a well constructed GEOS-Chem model to simulate the EC and OC concentrations at 10 rural and background sites in China. Using a “multiple regression” technique, the authors provided a “top-down” emission estimates of EC and OC, and the secondary OC formation in China was quantified. The manuscript is well organized and the methodology is feasible, and it may be of great interest to the China’s EC and OC modelers and emission inventory developers. However, one of the key conclusions seems to be too arbitrary and not convincing to me that the “bottom-up” EC and OC emissions in China in year 2006 are severely underestimated (>60%) at

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the country level. In addition, there are some grammatical errors. I would recommend the manuscript for publication after the comments listed below are fully addressed. Grammar-checking by native English speaker is also highly recommended.

Specific comments:

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 explain the authors’ results to some extent. This is also

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consistent with Hakami et al. (2005) and Kondo et al. (2011)'s results. 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.

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. 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”.

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 author s to add relevant discussion in Section 4, and revise some conclusions correspondingly (e.g., abstract, section 4, 6, and 7).

Page 28228, line 4-6. Page 28229, line 1-13. 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.

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(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. Additionally, “Duncan et al. (2003)” is not included in the reference list.

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 Gaolan-shan 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.

Page 28237-28238. 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.

Page 28239, line 25. 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.

Technical corrections:

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

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

Page 28222, line 5. “are emitted” should be “is emitted”.

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

Page 28222, line 27. Add “developed” between “inventories” and “by”.

Page 28223, line 3. Add “developed” between “inventories” and “by”.

Page 28223, line 18. Add “out” between “16” and “of”.

Page 28224, line 23. Add “have been” between “studies” and “measured”.

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

Page 28227, line 28. “tests to by” should be “tests by”. And please rewrite this long sentence.

Page 28228, line 3. Add comma after “OC”.

Page 28230, line 2. “whose emissions” should be “emissions of which”.

Page 28235, line 19-21. Please rewrite this sentence.

Page 28241, line 1. “concentration improve” should be “concentrations are improved”.

Page 28241, line 6. “OC improve” should be “OC are improved”.

Page 28241, line 8. Put “either” after “the model”.

Page 28241, line 25. “emission estimates do a better job than...”. Please change to other words.

Page 28242, line 2. “suggest” should be “suggests”. “is capturing” should be “captures”.

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Discussion Paper



Page 28242, line 4-6. Please rewrite this sentence.

Page 28242, line 12. Add “high” between “of” and “precursor”.

Page 28243, line 3. Change “the findings” to “those”.

Page 28243, line 4. “a larger isoprene emission” should be “larger isoprene emissions”.

Page 28243, line 8. “double” should be “twice higher”.

Page 28244, line 4. “Ohara et al., 2007” is not included in the reference list.

Page 28248, line 1. “statics” should be “statistics”.

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