

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 replies are shown in blue.

1. *Language, the grammar needs to be edited.*

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

2. *The discussion in this study should take into account that all conclusions are drawn using only surface measurements. However, especially when discussing foreign sources the authors should be careful. I understand that vertical EC/OC observations might not be available, but at least this should be mentioned.*

We added explicit statements in the Conclusion to address this:

“Our analyses are based on surface measurements. Aircraft observations of EC and OC concentrations may provide additional constraints on the contribution of foreign sources.”

3. *What are the uncertainties of the measurements? Can these uncertainties be included in the modeling estimates? How much of the top-down estimates could be explained by observational biases?*

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 $\pm 0.78 \text{ TgC yr}^{-1}$, 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.”

- 4. The paper only discusses emission uncertainties, but does not take into account that aerosol processes such as secondary aerosol formation, transport and removal might be responsible for a portion of the disagreement between model and observations.***

Removal and transport are accounted for by our chemical transport model. We tested the uncertainty in removal rates by tripling the timescale of carbonaceous aerosols converting from hydrophobic to hydrophilic but found the effect to be less than 5% (Sect. 2.1). We also found that meteorology factors cannot explain the difference between observation and model results (Sect. 3). We added explicit text in the Conclusion on this point:

“Sensitivity simulations show that meteorological factors and uncertainties in the aerosol removal rates cannot explain the low biases in the simulated concentrations. This suggests an underestimation of carbonaceous aerosol sources on a national scale.”

We discuss the contribution of secondary formation to OC explicitly in Section 5. We also added explicit statements about the secondary formation of OC being severely underestimated in the Abstract and in the Conclusion:

“Secondary production of OC is also underestimated in our model, either due to error in VOC precursor emissions or missing secondary formation pathways.”

- 5. Page 3, L8: VOC is not the only possible precursor of SOA. A small portion can come from condensing primary organics, but this might be just a small fraction.***

We modified and added to the text in Sect. 1 and Sect. 2.1 to reflect this point:

“OC can also be produced in the atmosphere as part of secondary organic aerosol (SOA) from gaseous organic compounds.”

“Recent studies indicate that additional secondary formation pathways, such as the aqueous reactions of dicarbonyls and condensation of semi-volatile and intermediate volatility organic compounds, may be important (Robinson et al., 2007; Carlton et al., 2009; Hallquist et al., 2009; Jimenez et al., 2009).”

“Additional SOA production from semi-volatile and intermediate volatility organic compounds have recently been developed for GEOS-Chem (Pye and Seinfeld, 2010), but that pathway is not included in the present study.”

- 6. Can the authors comment on the currently used historical CMIP5 (Lamarque et al 2010) inventory?***

We added explicit statements about our emission estimates being much higher than those

of Lamarque et al. (2010):

“In particular, our emission estimates for the year 2006 for both EC and OC are more than 120% higher than the emissions estimated by Lamarque et al. (2010) for the year 2000, which are used in the upcoming Fifth Assessment Report (AR5) by the Intergovernmental Panel on Climate Change (IPCC).”

7. *Figure 1 -3 should be enlarged.*

Figures 1, 2, and 3 have been enlarged following the Referee's suggestion.