Response to Editor

We thank the Editor and all the reviewers for insightful comments on improving the paper. We have endeavored to address all questions raised by the reviewers. Please find below our point-by-point responses with all relevant changes, and a marked-up manuscript version is also attached after the point-by-point responses.

Reply to anonymous referee #4

In this paper "Constraining Black Carbon Aerosol over Southeast Asia using OMI Aerosol Absorption Optical Depth (AAOD) and the Adjoint of GEOS-Chem" the authors make use of the OMI absorbing AOD dataset to produce posterior estimates of BC emissions in Asia with the GEOS-Chem adjoint. They find extremely large increases in BC emissions from multiple datasets are required for the model to account for the AAOD from OMI, and the posterior emissions still underestimate the surface concentrations in some regions.

It appears that the considerable concerns of the previous reviewer have been addressed to some extent, although the results raise new questions - partly because of presentation (e.g. for which period are the WRF-Chem simulations displayed in Fig S3?). It is frustrating that these are not presented in a way that is directly comparable with the results of the GEOS-Chem simulation.

Reply: We have repeated the test using WRF-Chem for the same time period (April 2006) and clarified the time period used in the text, see p35 and Figure S3.

Aside from this, I have some concerns over the apparent lack of consideration of the uncertainty in the assumed optical properties of BC (and the unrepresented brown carbon), it is unclear how much this may contribute to the large changes in prior and posterior BC emissions and agreement with observations.

Reply: The absorbing aerosols in GEOS-Chem only include BC, OC and dust, while the brown carbon has not yet been taken into account. In this study, the simulated BC is a proxy of all absorbing carbonaceous aerosols, and the constraints on emissions by AAOD are recognized to be on absorbing carbonaceous emissions. While the attribution of ambient aerosol absorption to BC may be a reasonable approximation in areas dominated by fresh soot emissions, it may lead to misleading estimates of the AAOD when other light absorbing particles were present since brown carbon contributes 28% on average of the total absorption at 440 nm [Bahadur et al., 2012]. This undoubtedly resulted in overestimation of BC emissions after optimization in the areas where brown carbon and other absorbing aerosols were part of the observed AAOD. We performed a sensitivity experiments (SenExpt.) by removing the brown carbon contribution (~30%, see above reference) to the total OMI AAOD observation, since GEOS-Chem does not include brown carbon. The differences of the optimized anthropogenic emissions are lower by up to 30% over the major source regions compare to the standard optimized results [see Fig. A below]. The positive difference over southeastern China is related to the penalty term,

as we used the same regularization parameter value as in the standard optimization. The large low biases of simulated surface OC concentrations over eastern China [Fu et al., 2012], even up to a factor of 7 at the sites of XIA in April (see Fig. B below), likely resulted in overestimation of BC emissions after optimization in the areas where brown carbon and other absorbing aerosols were considered in the observed AAOD. Therefore, the overestimates of surface BC concentrations after optimization at XIA, GUC and TYS (Fig. 18a) are possibly attributed to the underestimate of absorbing OC (brown carbon).

We have updated the text on page 35 as:

"The overestimates of optimized surface BC concentrations at XIA, GUC and TYS (Fig. 18a) are possibly attributed to the underestimate of absorbing OC and associated brown carbon, the latter which is not included in the model."

and on page 45 as:

"Therefore, in this study, the simulated BC is effectively a proxy of all absorbing carbonaceous aerosols, and the resulting constraints on emissions are thus best interpreted as constraints on absorbing carbonaceous emissions."

"We performed a sensitivity experiment by removing 30% of the total absorption from the OMI AAOD observation, since GEOS-Chem does not include brown carbon. The optimized anthropogenic emissions are lower by up to 30% over the major source regions compared to the standard results. Given that the model has large low biases of surface OC concentrations over eastern China [Fu et al., 2012], the overestimated BC concentrations after optimization at XIA, GUC and TYS (Fig. 18a) may possibly be attributed to the underestimation of absorbing OC (brown carbon)."

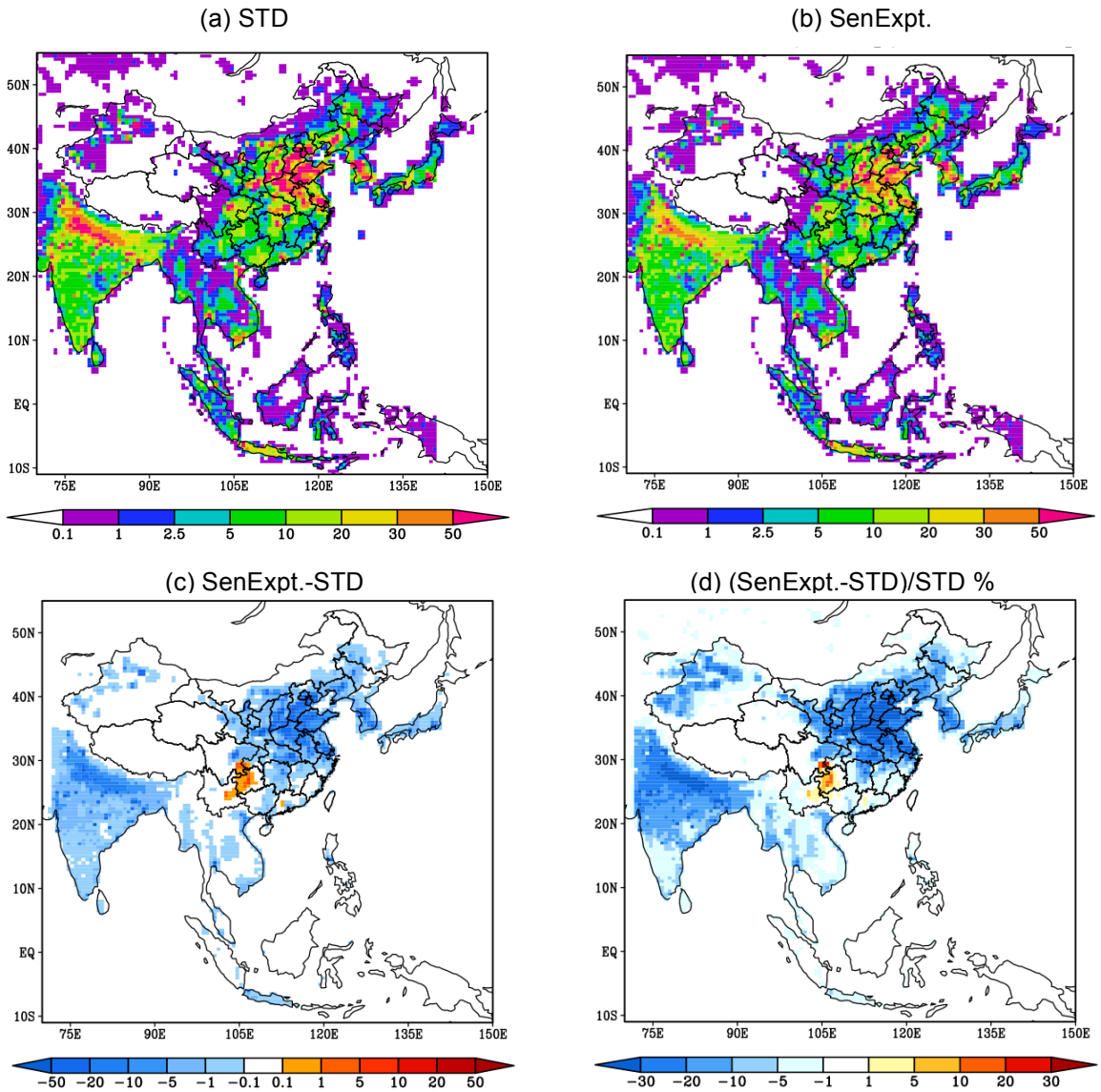

Fig. A Optimized anthropogenic BC emissions of (a) standard optimization; (b) sensitivity optimization by taking out the 30% brown carbon from observation; (c) absolute difference between (a) and (b); (d) percentage differences between (a) and (b).

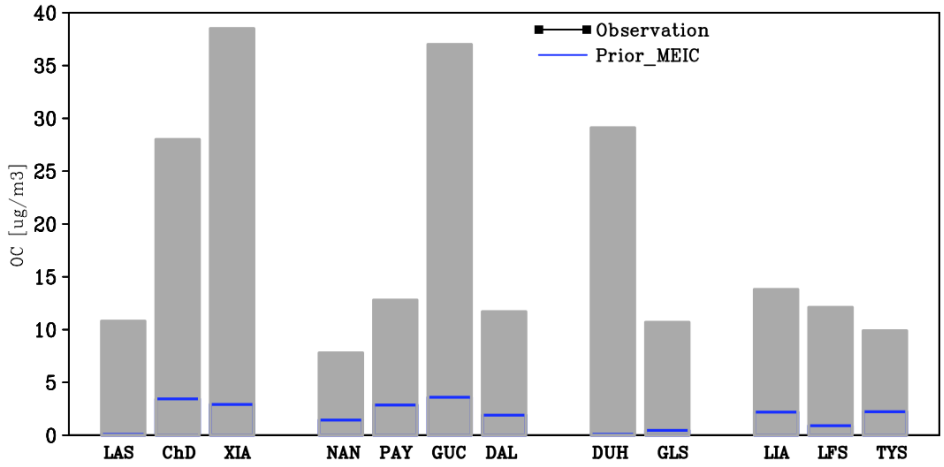

Fig. B Comparison of monthly surface OC concentration from in situ measurements and GEOS-Chem.

Finally, there are minor corrections throughout the text and figures that need addressing before considering for publication.
Reply: We have performed additional proofreading throughout the revised manuscript.

Reply to anonymous referee #5

Review on "Constraining Black Carbon Aerosol over Asia using OMI Aerosol Absorption Optical Depth and the Adjoint of GEOS-Chem" by L. Zhang et al.

I have read previous reviewers' comments and authors' replies. The authors put a considerable effort to accurate black carbon emission over East Asia, Southeast Asia and South Asia by using data assimilation and an adjoint model. Although the method remains some questions, the results still have substantial value for future studies. Therefore, I recommend to publish in ACP.

Based on previous reviewers' comments, using the OMAERUV AAOD is not suitable for this type of inversion problem, I kind agree with that. However, I believe that someone will do a similar work as the authors did in the future. The work itself is not about the scientific contribution only. It opens a discussion of the OMAERUV AAOD usage in data assimilation to accurate black carbon emission. Another value of this work is the conversation between the authors and reviewers. For sure, many people have the same questions as us. I will suggest authors summarize the reviews' comments and authors' replies in the supplementary to extend the value of this paper.

Reply: We thank the reviewer for their comments. The reviews' comments and authors' replies have been summarized in the supplementary.

There is one minor comment. The unit of Table 1 should not be Tg. Is it Tg/year or others if you integrated the domain surface area.
Reply: This table has been removed.

Reference

Bahadur, R., Praveen, P. S., Xu, Y., and Ramanathan, V.: Solar absorption by elemental and brown carbon determined from spectral observations, P. Natl. Acad. Sci. USA, 109, 17366–17371, doi:10.1073/pnas.1205910109, 2012.

Fu, T.-M., Cao, J. J., Zhang, X. Y., Lee, S. C., Zhang, Q., Han, Y. M., Qu, W. J., Han, Z., Zhang, R., Wang, Y. X., Chen, D., and Henze, D. K.: Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution, Atmos. Chem. Phys., 12, 2725-2746, doi:10.5194/acp-12-2725-2012, 2012.