Reply to anonymous referee #2

This paper is outlining a method and procedure by which emissions of BC are estimated from South Asia, East Asia, and Southeast Asia, using various combinations of modeled fields and some information of UV absorption from OMI. The idea behind the paper is interesting. If it is executed correctly, it will provide a worthwhile and significant step forward. However, at the present time, the paper is far too undeveloped, it lacks clarity and reproducibility, it mis-uses measurements, it shows a lack of understanding of absorption in the UV and how that is different from absorption in the visible, it confuses model values in the vertical as being equivalent with measurements in the vertical, and makes gross assumptions in the model space. In addition, although ex tremely long, there still is a lack of clarity and precision, specifically with regards to the aerosol chemical and physical change assumptions, and on the 4d-var components. Hence, it is actually impossible to know what has been done, or to reproduce what has been done. Furthermore, the conclusions are not supported based on the evidence as provided in the figures, as outlined in detail below.

For these reasons, I suggest that the paper be rejected. I would urge re-submission of a completely revised scientific effort at an appropriate stage of development.
Reply: We have emphasized the sections of the manuscript that address the reviewers most fundamental science concerns (namely, the use of AAOD as an indicator of BC, and the uncertainties in our inversion owing to biomass burning). We have clarified our discussion of uncertainties owing to aerosol vertical distribution and provided more details of the aerosol mechanisms employed in this model. While there were no specific comments below related to 4D-Var components, we do include a succinct overview of the approach with ample references to previous studies that cover the method in more detail. Lastly, the results here use publically available OMI data, and the code used for the inversion will become part of the publically available GEOS-Chem adjoint model, meaning that all results are readily reproducible. We thus encourage the reviewer and editor to reconsider this manuscript.

1. Title, Abstract, and throughout the paper: Your definition of Southeast Asia is not standard, and must be changed throughout the paper, including in the title itself. Scientifically, this is justifiable as well, since the climatology of most of Greater China, Koreas, and Japan is far different from that of very Southern Greater China and ASEAN. Furthermore, the Indian sub-continent is also significantly different.
Reply: The name "Southeast Asia" was adopted in accordance with the GEOS-Chem model's definition of this domain. Admittedly, this domain is a mix of the traditional SE Asia and Eastern Asian regions (http://en.wikipedia.org/wiki/Southeast_Asia, http://en.wikipedia.org/wiki/East_Asia). However, as the domain was defined in the first line of the abstract and visually portrayed in numerous figures, it seems there is little ambiguity concerning the region of study. Nevertheless, we have removed "Southeastern" from the title, and changed the first line of the abstract to read "in the region referred to here as Southeastern Asia (70°E–150°E, 11°S–55°N)" and the introduction to read "The Asian region referred to here as Southeast Asia (70°E–150°E,

11°S–55°N).”

2. At 388nm there is still a significant absorbing fraction from dust, OC, and Sulfate. Hence, this is not a good proxy for BC, at least as compared to AERONET and other sources that use visible and near IR. The paper does not seem to take this into account very well. The authors even acknowledge this when they point out that the worst fitting AERONET SSA is at 440nm (blue) over dusty regions. Naturally the OMI results are far more error prone. This needs to be re-thought out before it can proceed.
Reply: We do in fact explicitly consider the contribution of dust and OC to total AAOD. The topic of sections 2.5 and 4.1 is the parsing of BC-related information from the AAOD measurement. We consider both constraints from our own modeling work as well as the use of “flags” in the retrievals themselves for carbonaceous aerosol. We also included extensive discussion of this issue in the second to last paragraph of the conclusions.

While section 2.5 has been significantly re-organized and re-written for clarity following comments from reviewer 1, the calculations presented here are unchanged. The originally submitted manuscript thus did take into account most factors raised here by reviewer 2.

Further, the reviewer’s concerns are likely overstated, as BC is found to contribute more than 90% to AAOD over urban regions.

Lastly, while pure sulfate aerosol is not a significant absorber, a sulfate coating can enhance absorption by other species. While this mechanism is not treated in our externally mixed aerosol model, this point is raised as a source of uncertainty in Section 6.

3. Geos Chem, like most global-scale models significantly underestimate the vertical heights of aerosols in this part of the world. This is due to significant impacts of convection, urban heat co-released with the aerosols, fire, and other dynamical and chemical properties not captured by these models. The fact is that the GEOS-CHEM heights were used instead of measurements from CALIPSO, and that they were found to be so different. Since CALIPSO is measurement based, these heights are the ones that should be used. This shows that GEOS-CHEM’s ability to model the distribution is in error, and hence that the results are untrustworthy.
Reply: We do agree that the GEOS-Chem model still has bias in simulating the aerosol layer height. This has been examined in detail via comparison to CALIOP in van Donkelaar et al. [2013].

However, we perhaps did not articulate clearly the point of using GEOS-Chem aerosol layer heights. Our goal is to have a consistent vertical treatment for both the retrieved and the modeled AAOD. The replacement of information used in the retrieval with information from the assimilation model for the sake of consistency is used in other studies as well [Choi et al., 2008; McLinden et al., 2014; Lamsal et al., 2014]. The reason to enforce such consistency is to cleanly evaluate the impact of the observations separate from other issues. Otherwise, an undetermined component of the result would be owing to differences between GEOS-Chem and CALIOP vertical profiles. So using the same vertical treatment (both based on GEOS-Chem aerosol layer height) is to make the comparison between "apple" and "apple" even though the "apple" is not a perfect "apple".

Continuing with this analogy, it would have been even better to have made an "oranges" to "oranges" comparison by instead using CALIOP data to correct all of the GEOS-Chem profiles to have aerosol heights matching those of the retrieval. However, the latter are drawn from a climatology, not from scene-specific knowledge of the vertical profile, and this approach is thus more suitable for considering longer-term averages (e.g., estimating annual average surface $PM_{2.5}$ as in van Donkelaar et al. [2013]).

That being said, we do recognize that improving the vertical distribution of aerosols in GEOS-Chem warrants further attention, and we hope this work helps underpin the importance of addressing this issue further in future studies. We additionally state now in the conclusion: "The results of the optimization may be biased by error in the model's vertical distribution of BC, which has been adjusted in other studies [van Donkelaar et al., 2013]."

4. The carbonaceous aerosol scheme used in GEOS-CHEM, which UNDERPINS this entire paper, has been found to be not reliable in this part of the world. One good example comes from a pair of papers embedded in one of the other papers cited in the text: Cohen and Prinn 2011 and Cohen et al., 2011.
Reply: While we appreciate the value of Cohen et al. [2011] and Cohen and Prinn [2011], we do not find results or references therein pertaining to the specific carbonaceous aerosol scheme used in GEOS-Chem. However, they do make the general point that urban-scale processing can lead to a +8% bias in AAOD [Cohen et al., 2011]. While important, we would hardly conclude the neglecting an 8% bias leads to an "unreliable" model. We will however include reference to this work in our discussion of the impact of model resolution in Section 5.3. We also stated in the abstract of the original manuscript that model resolution error may lead to underestimates in surface concentrations of up to x2.5.

These show that the lifetime of BC and OC are significantly different in these regions of the world due to the strong nonlinear chemistry and physics. Additionally, multiple measurement studies have done by the Koreans and Japanese that underlay this conclusion. Additionally, strong removal differences between the hydrophobic, partially converted, and hydrophilic forms interact non-linearly with convection. And given the large amount of convection present, this will introduce another large error term.
Reply: Indeed, GEOS-Chem is not a perfect model but it is nevertheless valuable for the simulation of atmospheric chemistry and air pollution transport, and has been used for numerous studies (see the GEOS-Chem website: http://geos-chem.org/). The original carbonaceous aerosol simulation in GEOS–Chem was developed by Park et al. [2003] based on the Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) aerosol module. Heald et al. [2011] and Wang et al. [2011] give overviews of the current organic aerosol and BC simulations in GEOS-Chem, respectively. Changes in particle lifetime

owing to combination with other aerosols is not considered in our results since the standard GEOS-Chem version the aerosols treated as external mixtures that do not interact with each other. But the aging of BC in GEOS-Chem for converting hydrophobic BC to hydrophilic BC is similar to most of other models, typically about 1 day [Koch et al., 2009]. The scheme for aerosol scavenging was based on Liu et al., [2001], which did not distinguish between rain and snow. The recent updates by Wang et al. [2011] included corrections to below-cloud and in-cloud scavenging that improved the overestimation of integrated scavenging [Dana and Hales, 1976]. Corresponding updates to the wet scavenging in the GEOS-Chem adjoint might also be helpful for improving the optimized results, which will be our next step. The aerosol internal mixing that includes effects of various physical, chemical, and meteorological processing also play important role in simulating BC concentration and aerosol absorptions. We have included these in the discussion section, see Section 6, the 6th paragraph.

5. GFED has been demonstrated to not be a good product for matching actual observations of aerosols over Southeast Asia, as given by Cohen 2014[1]. It is both low in terms of absolute amount, as well as having timing which is not fully representative, both inter-annually as well as intra-annually. This is especially true for 2006, the year you have chosen, since it was a very strong El-Nino year, and hence the emissions in that year from fires in Southeast Asia were much stronger than a normal year. This is a major problem in terms of the a-priori and needs to be addressed. Furthermore, the emissions inventories used do not include the cited one from Cohen and Wang, which is larger in terms of magnitude from all of the others used. Why was this inventory also not used?
Reply: The inventories from Cohen [2014] and Cohen and Wang [2014] were not published when we started the work. While it is reasonable to suggest that we consider this recent work in our discussion, any expectation that we would have somehow used these very recent results in our work is not. Currently, the GFED inventory is the only biomass burning data that has been implemented in the standard GEOS-Chem model. If the authors of the papers mentioned by the anonymous reviewer wish to make their inventory available to the GEOS-Chem community, they are encouraged to do so.

Further, we do agree that uncertainties in biomass burning emissions could impact our results. We used differences between GFEDv2 and GFEDv3 as a proxy for quantifying this impact. Figure 12 in our original manuscript shows how uncertainties in biomass burning impact our constraints on anthropogenic BC sources (see section 5.1). As shown here, the impacts are largely 2nd order (mostly less than 25%), and do thus find they do not constitute a “major problem.”

Specific comments:
1. Bond et al. 2013 is an assessment paper, not a piece of original research. As such, using it as a primary source in most instances is inappropriate. Better would be to find the underlying paper which made the claim and cite that instead.
Reply: That is good point. The original references have been updated.

2. p28397: Again, this is a critical mistake. OMI measured AAOD, due to the fact that it is at 388nm and based on other values from the UV, is not just measuring BC and dust,

but is actually a composite of these and other species. For example, even sulfate absorbs at those wavelengths. If this was taken into account, then please clearly state so. If not, then the results of this work are likely in error and should be repeated from scratch, also considering this factor.
Reply: Please see the response to Major comment 2.

3. Equation 3 is incomplete. In the UV absorption is also from sulfate and other particles.
Reply: In GEOS-Chem, the absorption is mainly from these three particles that contribute to the total AAOD. We do agree that sulfate and other organic aerosols would also be light absorbing particles that contribute to the total AAOD. But the contributions are very small part (less then 3%), similar to that which we have quantified from the OC contributions.

4. P28398 how is the AAOD computed in the model? You state the observed is e observed * model-BC / model-all. But how the model-all is computed is not mentioned anywhere. Is it a single moment, binned, two-moment, etc. method? Is it mixed internally, externally, core-shell, etc.? This will lead to dramatically different results in each case. This means he rest of the equations are not useful in this section as well, including (7) and (8).
Reply: The aerosol optical depth at 400 nm is calculated online assuming log-normal size distributions of externally mixed aerosols and is a function of the local relative humidity to account for hygroscopic growth [Martin et al., 2003]. The AAOD of each aerosol species is derived by [Cohen and Wang 2014; Cohen 2014]

$$AAOD=AOD^{*}\ (1-SSA).$$

The AAOD of model-all is computed by Eq. 3 in revised manuscript, which is the sum of BC, OC and dust AAOD.

The GEOS-Chem result is single moment, and the aerosol treatment is external mixing.

5. Figures 13, 14, and 16 clearly show that the end results are still grossly underperforming, especially outside of northern China.
Reply: There are indeed persistent biases after optimization, which may due to one or more of the several factors that we discuss the last section, such as constraints from the prior inventory. This is also mentioned in the abstract,

“Low biases in BC concentrations are improved or corrected in most eastern and central sites over China after optimization, while the constrained model still underestimates concentrations in Indian sites in both April and October, possibly as a consequence of low prior emission”

6. Figure 17 does not match with observations of the extreme burning season from 2006 at the AERONET sites, or as given by Cohen 2014.
Reply: Figure 17 shows the spatial distributions of optimized surface BC concentrations using INTEX-B and MEIC_SEAC^{4}RS inventories overlaid with situ measurements of surface BC concentrations at 20 sites. They are not the AAOD results, and can not be directly compared with the AAOD results of AERONET sites and Cohen 2014.

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