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Interactive comment on “Constraining black carbon aerosol over Southeast Asia using OMI aerosol absorption optical depth and the adjoint of GEOS-Chem” by L. Zhang et al.

Anonymous Referee #2

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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-

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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.

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, Korea, and Japan is far different from that of very Southern Greater China and ASEAN. Furthermore, the Indian sub-continent is also significantly different.

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.

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.

4. The carbonaceous aerosol scheme used in GEOS-CHEM, which UNDERPINS this

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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. These show that the lifetime of BC and OC are significantly different in these regions of the world due to the strong non-linear chemistry and physics. Additionally, multiple measurement studies have been 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.

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?

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.

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

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scratch, also considering this factor.

3. Equation 3 is incomplete. In the UV absorption is also from sulfate and other particles.

4. P28398 how is the AAOD computed in the model? You state the observed is $e_{\text{observed}} * \text{model-BC} / \text{model-all}$. 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 the rest of the equations are not useful in this section as well, including (7) and (8).

5. Figures 13, 14, and 16 clearly show that the end results are still grossly underperforming, especially outside of northern China.

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.

Reference: [1] Jason Blake Cohen (2014) Quantifying the occurrence and magnitude of the Southeast Asian fire climatology. Environ. Res. Lett. 9 114018 doi:10.1088/1748-9326/9/11/114018

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