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Supplement of

Constraining black carbon aerosol over Asia using OMI aerosol absorption optical depth and the adjoint of GEOS-Chem

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Figure caption.

Figure S1. Comparisons of MISR AOD and GEOS-Chem dust AOD for April 2006. (a) MISR AOD; (b) GEOS-Chem dust AOD with standard dust scheme; (c) GEOS-Chem dust AOD with new PSD scheme; (d) GEOS-Chem dust AOD with standard dust scheme minus MISR AOD; (d) GEOS-Chem dust AOD with new PSD scheme minus MISR AOD.

Figure S2. Optimized total emissions using inversion approaches based on (a) scaling factors and (b) emissions.

Figure S3. WRF-Chem surface BC concentrations for April 2006 based on INTEX-B (a) prior (b) posterior emissions; and their (c) absolute differences and (d) relative differences.

According to the last reviewer's comments, here we summarized the reviewers' comments and our replies in the supplementary, as these will not be published in the discussion.

Reviews' major comments (paraphrased):

1. OMAERUV AAOD product of cloud contamination and aerosol layer height. How do you deal with clear-sky bias? Cloud contamination? It is not proper to interpolate the observed AAOD to the model aerosol profile; this is not a linear relationship and you cannot use linear interpolation in this case. You somehow linearly interpolate the OMI AAOD to your simulated (GEOS-Chem) aerosol height; you do not show that this is (1) even possible (there is likely a non-linear relationship between aerosol height and AAOD in the OMAERUV algorithm) and (2) you show huge differences between the AAOD obtained with the actual observed profile and the made-up one using your model profile, so (3) why do you do this at all? Why would you assimilate this "data"?

2. Inversion method. The real test of this new emissions inventory is if it can improve simulated AAOD and surface BC concentrations in another model that has different aerosol processes and mapping between aerosol mass and optical properties. Otherwise this is a bit of a chicken and egg problem. If your simulated AAOD is to low, and you correct this through assimilation of OMI AAOD, then of course you would improve you surface BC concentrations if they were too low prior to assimilation. It would be more interesting if, after developing such a new BC emissions inventory, you showed that it improved BC concentrations in another model to avoid the circularity inherent in the current study (you assimilate AAOD -> new emissions -> better surface BC concentrations).

3. Brown carbon impacts. Method is likely affected by brown carbon, particularly in this region of the globe.

4. Optimized results. If your simulated AAOD is too low, and you correct this through assimilation of OMI AAOD, then of course you would improve you surface BC concentrations if they were too low prior to assimilation. For all the effort, there does not, at most sites, appear to be much of an improvement in the posteriori compared to the prior (MEIC). This does not provide confidence that the assimilation achieved its goals, especially in October.

The authors' replies (paraphrased):

1. We have expanded discussion of cloud contamination and we find that the role of the

aerosol layer height is relatively small.

About cloud contamination, the available data counts of observed pixels in Fig. 6 and Fig. 7 already account for such aspects of data quality. The major factor affecting the quality of aerosol products is sub-pixel could contamination, while AAOD is less affected by cloud contamination. The final quality flag parameter in the OMAERUV level 2 files is a quality assurance (QA) flag that indicates the level of confidence on the retrieved parameters with regard to the interference of sub-pixel size cloud presence. Best retrievals, i.e., minimally affected by sub-pixel cloud contamination, have a QA flag of 0 and are deemed suitable for scientific use [Ahn et al., 2014]. In this study, only the most reliable retrievals minimally affected by sub-pixel cloud contamination are used.

We agree it is important to consider the nonlinear aspect of this relationship. We have thus evaluated, quantitatively, the magnitude of the nonlinearities, and thus the magnitude of the error introduced through our linear interpolation. As the retrievals are provided for multiple aerosol layer heights, we can use the values for two adjacent heights to estimate the AAOD at the third via linear interpolation, and compare this to the retrieval's actual AAOD based on the third layer height. This likely represents the maximum possible linear interpolation error (since in practice we interpolate to heights that are between two layer heights). We find that there is less than 30% error in linearly interpolating AAOD corresponding to a specific aerosol layer height from the AAODs corresponding to two other aerosol layer heights.

Having both recognized and quantified the error in our approximation, we believe it to be an acceptable approximation, also given that this source of error is small compared to uncertainties present elsewhere in the inversion caused by resolution and uncertain prior emissions. Further, we would like to reiterate the motivation for producing an AAOD estimate wherein the assumed aerosol layer height is consistent with the aerosol layer height used in the assimilation model. The retrieval "Final AAOD" products (OMI_Final) are interpolated values using the aerosol layer height value given by the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) climatology. First, such climatologies may not correspond to the exact time and location of the AAOD being assimilated in our case. Second, upon further investigation we learned that there are many cases for which the aerosol layer height is not available from CALIOP climatology, in which case the OMI aerosol height is obtained based on a climatology of GOCART model simulated aerosol heights, not observed values from CALIOP.

We prefer to use aerosol layer heights from a consistent, known source, i.e. from GEOS-Chem, for the sake of clarity and precision even if this sacrifices accuracy. 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. Nevertheless, we repeated the inversion using the native OMI layer heights, and found that the inversion results changed by less than 30%

in April, and by less than 10% in October. This is now mentioned explicitly in the third to last paragraph of the conclusions. (3). We prefer to use our own, known aerosol layer heights for the sake of consistency, such that we can avoid having to parse out the aspects of the inversion that may be due to assimilation of CALIOP or GOCART climatologies of aerosol layer heights rather than OMI AAODs.

Our goal is to have a consistent vertical treatment for both the retrieved and the modeled AAOD. 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]."

2. There is nothing circular about the use of independent data to evaluate the results of inverse modeling. It is also very rare to find studies in which such evaluation is performed using a model other than the one used for the inversion itself (although one could easily find dozens of examples to the contrary). However, to investigate this further we have implemented the optimized INTEX-B BC emissions in the WRF-Chem model and compared these simulations to those using the prior INTEX-B emissions (See Figure S.3 in the supplemental). Low biases of simulated surface BC concentrations still persist over broad areas when using our optimized emissions in WRF-Chem. However, these biases have been significantly improved (by a factor of 1.5 to 2) over the major source regions, compared to WRF-Chem simulations with the prior emissions. We thus believe our emissions constraints are not exclusively an artifact of GEOS-Chem model error. Moreover, our approach is not entirely circular, as AAOD is governed by a different balance of processes in the model than surface BC mass concentrations, and thus the latter provide an independent check of the inversion quality. However, some aspects of the model transport error would be present in both comparisons.

3. We agree with this point, and thus acknowledge that the absence of brown carbon in GEOS-Chem means that our results should be interpreted as constraints on absorbing primary carbonaceous aerosol emissions. It is important to realize that BC from most emission sources contains not only elemental and organic fractions [Chow et al., 2009], but also non-soot OC, i.e., brown carbon, that has a significant absorbing component at short wavelengths comparable to elemental carbon absorption [Jacobson, 1999; Kirchstetter et al., 2004; Andreae and Gelencser, 2006; Hoffer et al., 2006; Magi et al., 2009]. However, absorbing aerosols in GEOS-Chem only include BC, OC and dust,

while brown carbon has not yet been taken into account. 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 are present since the brown carbon constitutes 28% on average of the total absorption at 440 nm [Bahadur et al., 2012]. This likely resulted in overestimation of BC emissions after optimization in the areas where brown carbon and other absorbing aerosols were present in the observed AAOD.

4. We disagree that such agreement is guaranteed. If the initial model's AAOD was too low owing to reasons other than emissions magnitude (particle aging, optical assumptions, mixing state treatment, vertical distribution) then it would be possible to imagine that assimilation of AAOD would lead to worse agreement with independent observations, such as surface BC mass concentrations, that are governed by a different balance of factors (e.g., boundary layer height, model resolution), if not in sign then in location and magnitude. The comparison of the optimized model to this independent dataset is thus of critical value.

We agree that the optimized inventory does not eliminate the model prediction error. However, we believe that reporting when, where, and why AAOD assimilation may or may not improve the assimilation is of value to the community. Low biases of surface BC concentrations were improved or corrected at urban sites and eastern rural sites over China in April, with the linear regression slope between model and observed values increasing by more than a factor of four. However, the adjustments were not strong enough in most sites over India in April and October and over China in October. Moreover, the optimization had less impact on the western sites over China and costal sites over India due to the very low prior emissions and the large uncertainties in AAOD retrieval for low aerosol amounts over ocean. Model resolution error was also an important factor contributing to discrepancies of BC concentrations compared to in situ measurements. Results of the inversion were also compared to remote and in situ measurements that were not assimilated. The posterior AAOD were quite comparable to AERONET AAOD observations in April in China; however, large discrepancies remained at the sites over India and Thailand after data assimilation. These residual errors compared to AERONET may be associated with the limited and sparse observations of OMI observed AAOD in these regions, which themselves were not very consistent with the AERONET AAOD. Hiren et al., [2014] also pointed out that much of the observed inconsistency of SSA between OMI and AERONET is found to occur at moderate to lower aerosol loading (AOD 440nm<0.7) for which both inversion techniques might have issues related to signal-to-noise ratio and algorithmic assumptions.

Inversion of OMI AAOD measurements did not always lead to significant improvements in model estimates of BC emissions and sources. However, we did characterize much about this inverse problem in the process that will be of value for future studies that may advance the tools for greater success or for helping other avoid similar pitfalls. The major factor affecting the quality of the OMI aerosol product was sub-pixel cloud contamination due to the relatively large footprint of OMI observations. The availability of observed pixels is quite different in different seasons. These would highly impact the inversion results for different months. On the other hand, the remaining residual error in the simulated AAOD, which was significant in October, particularly in India, may be a consequence of the inverse modeling framework, which had difficulty introducing emissions in locations where the prior emissions were close to zero This downside may be overcome by performing inversions directly for the emissions, rather than emissions scaling factors.

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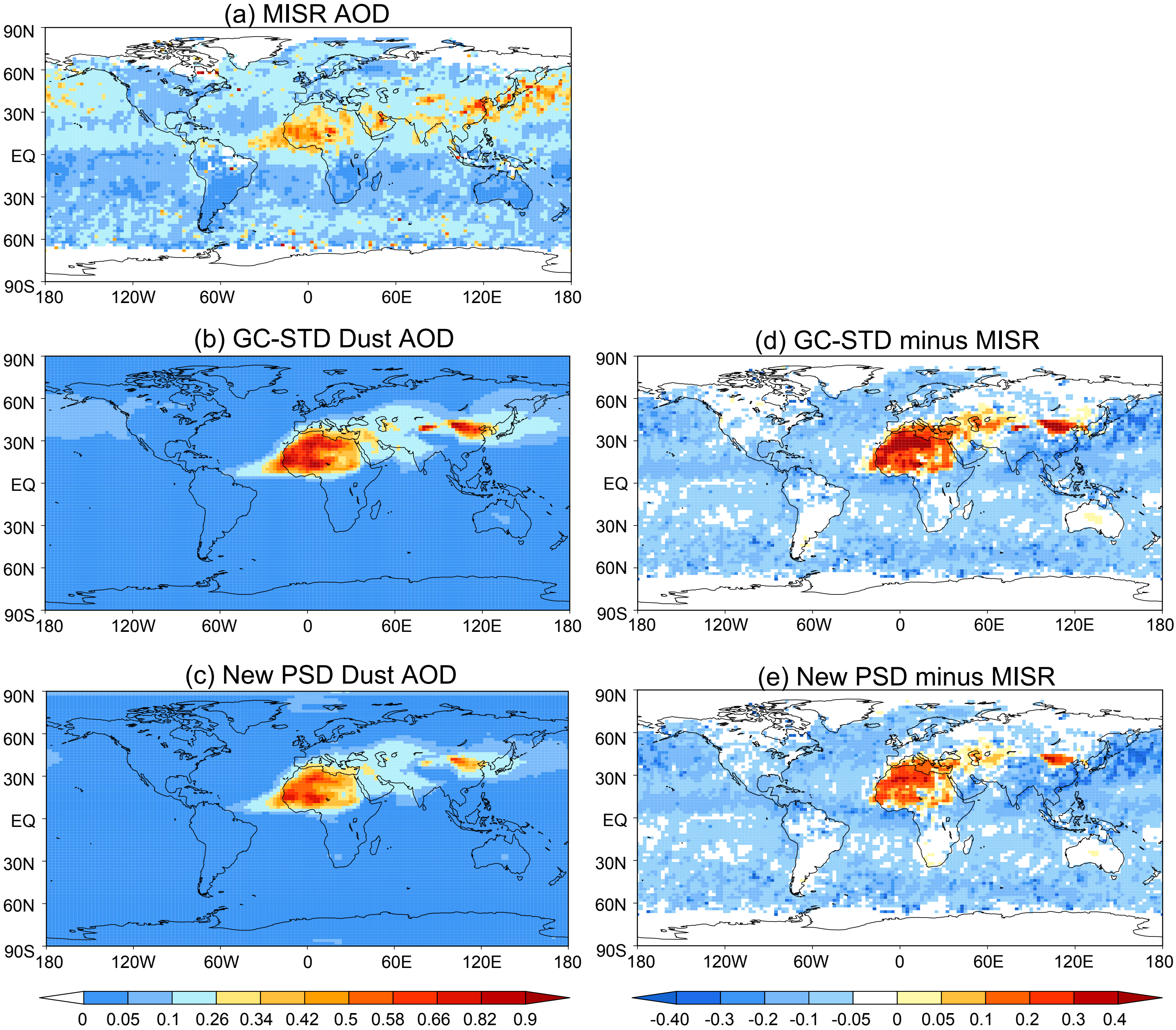

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(a) MISR AOD;
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(d) GEOS-Chem dust AOD with standard dust scheme minus MISR AOD;
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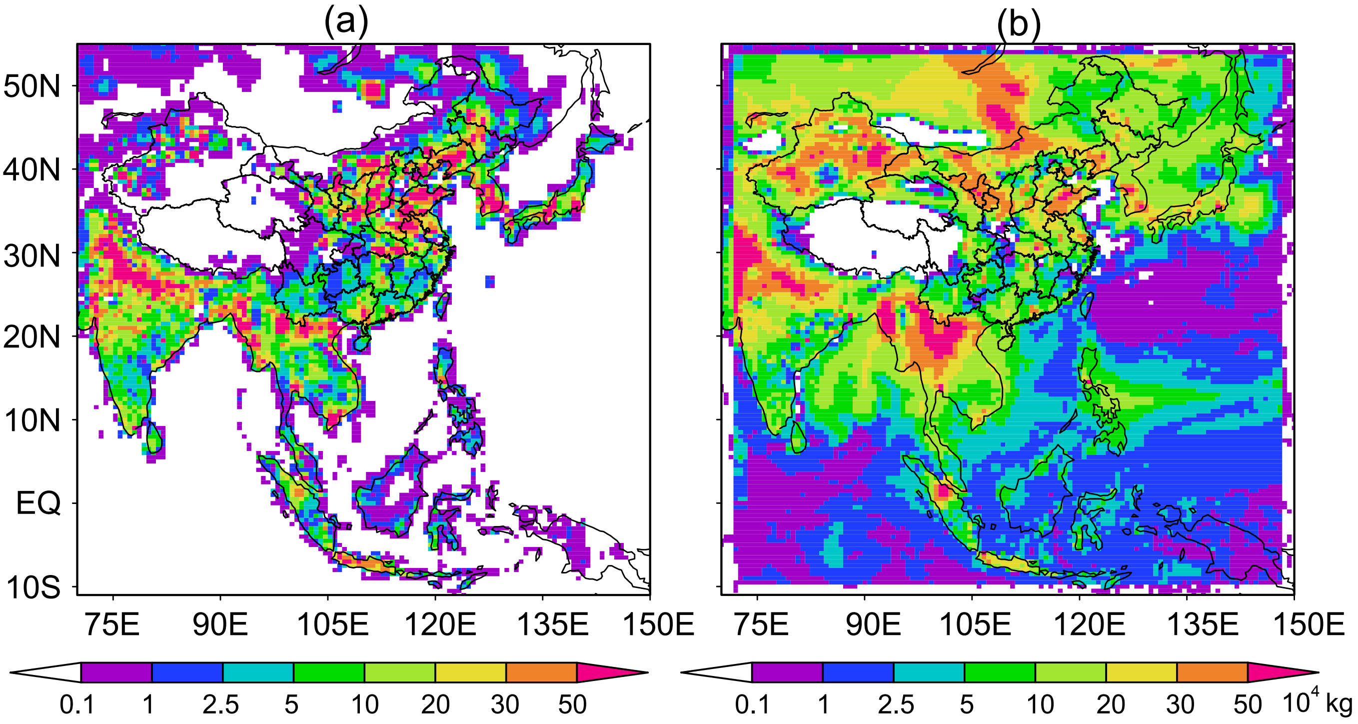

Fig. S2 Optimized total emissions using inversion approaches based on (a) scaling factor and (b) emission directly.

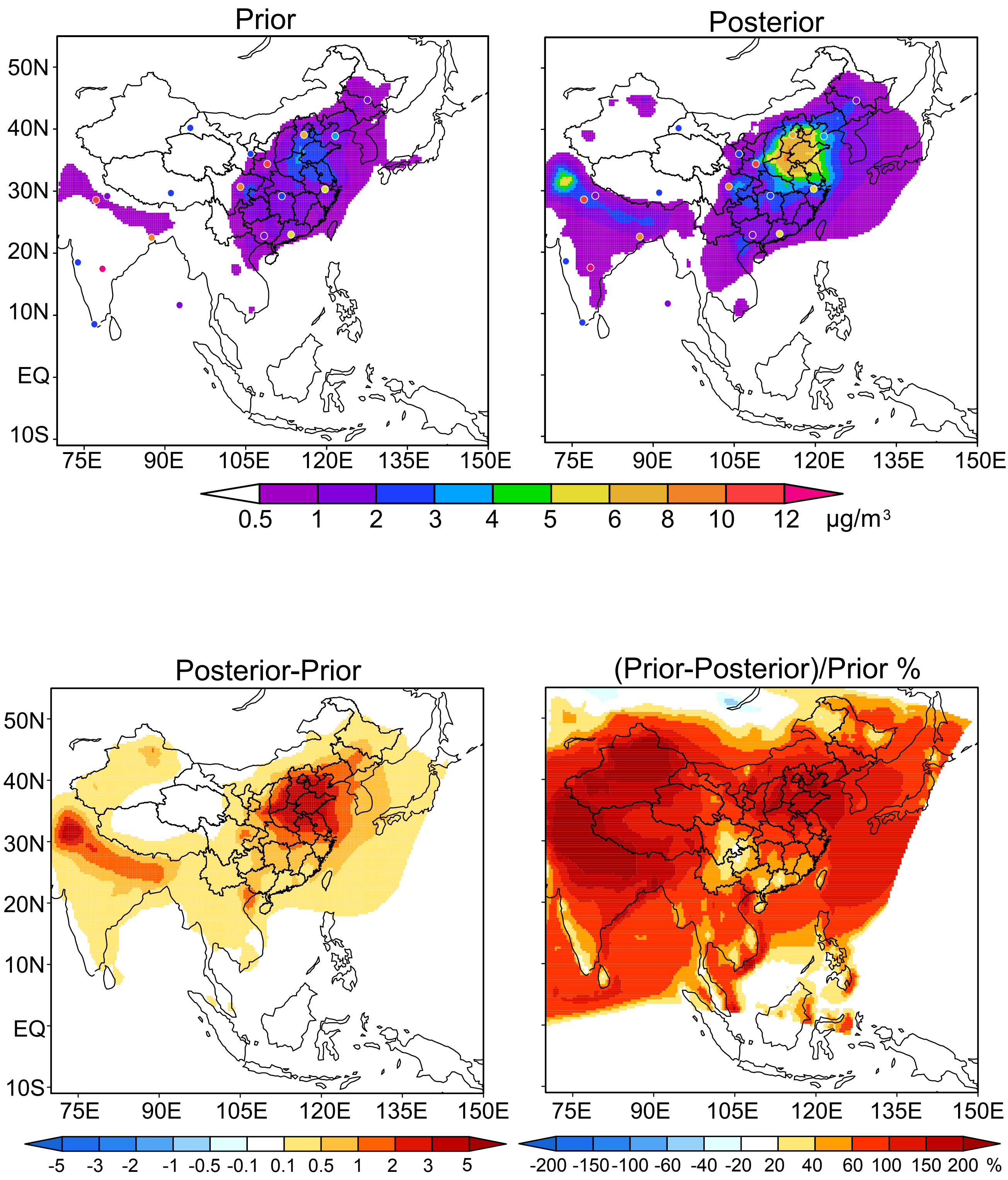

Figure S3. WRF-Chem simulated surface BC concentrations for April 2006 based on (a) prior INTEX-B (b) posterior emissions; and their (c) absolute differences and (d) relative differences.