**Manuscript # acp-2016-1032**

**Responses to Reviewer #3**

This study quantified source contributions of black carbon (BC) mass concentrations, trans-Pacific transport of BC, and direct radiative forcing of BC from seven regions in China using the Community Earth System Model with a source-tagging technique. The authors showed that BC concentrations were dominated by local emissions for regions with high emissions (e.g., North China, South China), whereas non-local emissions were important for regions with low emissions (e.g., Northwest China, Tibetan Plateau). They also showed that emissions from China and other regions were equally important for the BC outflow from East Asia and that emissions from China would be important for air quality in western United States. The annual mean direct radiative forcing of BC in China in their simulations was 2.3 W $m^{-2}$, and the contribution from emissions in China was estimated to be 66%.

The purpose of this study is interesting and the results obtained in this study are important to understand BC behavior in the atmosphere over East Asia. I think the authors should describe several points (shown below) more clearly, but overall the manuscript is written well and is suitable for the publication of this journal.

Major comments:

(1) Importance of BC in air quality problems
The authors sometimes use BC as an indicator of pollution (or air quality) in China (Lines 39-40, Lines 101-102, Lines 429-431, and Lines 571-572). However, I think it is questionable whether the concentrations and/or source contributions of BC can be used to represent those of total aerosols. Inorganic and organic species are dominant in China during polluted days, and spatial/temporal variations and source contributions of these species are largely different from those of BC because spatial distributions of emissions (e.g., BC v.s. SO2) and formation processes (primary v.s. secondary) are considerably different. For example, Matsui et al. (2009) showed that primary aerosols around Beijing were controlled by emissions within 100 km around Beijing within the preceding 24 h, while emissions as far as 500 km and within the preceding 3 days were found to affect secondary aerosols. Therefore, it is not always correct to extend the results of BC (e.g., source contributions) to the discussions on pollution and air quality because inorganic and organic species could have larger contributions from non-local emissions than BC. Please consider this point and describe the limitation of using BC results only in the discussions of air quality problems. In addition, please show the

percentage of BC mass to total mass (PM2.5) in China in the manuscript.
Response:

We agree with the referee that by no means BC can represent the total $PM_{2.5}$ in air quality problems, which is not the focus of this study. We have added a paragraph in the discussion section to describe the limitation of using BC as a tracer for air pollution, as "In this study, BC is used as an indicator of pollution (or air quality) in China. Although BC is often co-emitted with other species, such as primary organic matter, organic gases and sulfuric gases, source-receptor relationship of BC may not fully represent that of total aerosols. The contribution of BC to total near-surface $PM_{2.5}$ concentrations averaged over China is less than 10%. Other aerosols, such as sulfate, are dominant in China during polluted days. The spatio-temporal variations and source contributions of these species are largely different from those of BC because spatial distributions of emissions (e.g., $SO_2$) and formation processes can be considerably different. For example, Matsui et al. (2009) showed that primary aerosols around Beijing were determined by emissions within 100 km around Beijing within the preceding 24 hours, while emissions as far as 500 km and within the preceding 3 days were found to affect secondary aerosols in Beijing. Thus, the secondary aerosols could have larger contributions from non-local emissions than BC. BC concentrations are highest in winter over China due to higher emissions, while sulfate concentrations reach maximum in summer when the strong sunlight and high temperature favor the sulfate formation. Therefore, knowing the accurate source attributions of air pollution in China requires source tagging for more aerosol species, such as sulfate."

(2) Treatment of optical property and CCN activity of BC (Lines 151-169)
I could not find the description on the treatment of optical property (well-mixed, core- shell, or others) and CCN activity (conversion from hydrophobic to hydrophilic BC) of BC in the MAM3 model. I assume that well-mixed optical treatment is used to calculate BC absorption and that all BC particles are treated as hydrophilic BC in MAM3. Please describe the treatment of optical property and CCN activity of BC in the manuscript, and add some description on the potential impact (uncertainty) of these treatments on the estimation of BC concentrations, trans-Pacific transport of BC, AAOD, and direct radiative forcing of BC and their source contributions.
Response:

Thanks for the suggestion. We have added more information regarding the treatment of BC in the model to the Methods section and also added a paragraph discussing the potential influence in the Methods and Conclusions and Discussions section, shown as below:

Aerosol optical properties for each mode are parameterized according to Ghan and Zaveri (2007). Refractive indices for aerosols are taken from the OPAC (optical properties for aerosols and clouds) software package (Koepke and Schult, 1998), but for BC at solar wavelengths the values are updated

from Bond and Bergstrom (2006). In MAM3, the aging process of BC is neglected by assuming the immediate mixing of BC with other aerosol species.

BC aging in the atmosphere is important for BC concentration and its optical properties, which transforms BC from hydrophobic aggregates to hydrophilic particles coated with soluble materials. He et al. (2015, 2016a) found that BC optical properties varied by a factor of two or more due to different coating structures during BC aging process based on their theoretical and experimental intercomparison. Oshima et al. (2009) and He et al. (2016b) pointed out that the use of various microphysical BC aging schemes could significantly improve simulations of BC concentrations compared to the simplified aging parameterizations. Liu et al. (2012) also reported that the wet removal rate of BC simulated in standard CAM5 is 60% higher than AeroCom multi-model mean due to the rapid or instantaneous aging of BC. H. Wang et al. (2013) showed that the explicit treatment of BC aging process with slow aging assumptions in CAM5 could significantly increase BC lifetime and the efficiency of BC long-range transport. In the three-mode aerosol module (MAM3) of CAM5 used in this study, the aging process of BC is neglected by assuming the immediate internal mixing of BC with other aerosol species in the same mode. This assumption could lead to an overestimation of wet removal of BC and, therefore, an underestimation of BC concentrations, absorption optical depth (Fig. 3) and direct radiative forcing. In addition, the internally-mixed optical treatment in CAM5 could also cause bias in BC absorption calculation. However, H. Wang et al. (2014) examined source-receptor relationships for BC under the different BC aging assumptions and found that the quantitative source attributions varied slightly while the qualitative source-receptor relationships still hold. Therefore, although the magnitude of simulated BC and its optical properties could be underestimated due to the instantaneous aging of BC and uncertainty in coating structures, we expect that the aging treatment in MAM3 of CAM5 should not influence the qualitative source attributions examined in this study.

Other comments:

(3) Line 70
Please describe the reason of the faster regional removal.
Response:

Revised as “BC in East Asia has a shorter lifetime than the global mean value due to a faster regional removal (H. Wang et al., 2014), likely associated with a strong precipitation scavenging near sources and along the transport pathways over the Pacific Ocean.”

(4) Lines 168-169
Please clarify the definition of the direct radiative forcing of BC. Is this

calculated from the difference of two radiative transfer calculations with and without BC for the clear-sky condition?

Response:

Revised as “Direct radiative forcing of BC is calculated as the difference in the top-of-the-atmosphere net radiative fluxes with and without BC for the all-sky condition following Ghan (2013).”

(5) Lines 182-204

Please show the difference of BC emission fluxes between the emission inventory used in this study and other emission inventories (e.g., INTEX-B, HTAP). The values are shown later (at Lines 534-538), but I think it is better to show them here. In addition, please add some comments on the impact of larger values of BC emissions in this study on the estimation of source contributions of BC. Can you add the values of BC emissions from outside China (e.g., India, Southeast Asia, Japan, Korea) to Figure 1b?

Response:

We have added Table S1 to compare the anthropogenic emissions used in this study with emissions from some previous studies. The anthropogenic emissions of BC in China in 2010–2014 are larger than those used in the previous studies for earlier years, partly as a result of the rapid increasing trend of BC in China during recent years. The higher emissions likely lead to higher concentrations and direct radiative forcing, and source contributions of BC in China, compared to the values reported in these studies. We have added these descriptions in the Methods section.

We also revised Fig. 1 to include BC emissions from outside China. Emissions at regional scale are summarized here instead of Country level because the model revolution is a bit course to characterize emissions by countries. Total BC emissions from neighboring regions including rest of East Asia (REA, with China excluded), South Asia (SAS), Southeast Asia (SEA), and Russia/Belarussia/Ukraine (RBU) are shown in Figure 1c. These source regions outside China are consistent with source regions defined in the second phase of Hemispheric Transport of Air Pollution (HTAP2). South Asia and Southeast Asia have relatively high emissions. They may dominate the contribution to concentrations and direct radiative forcing of BC in China, especially southern and western China, from foreign sources through long-range transport. We have also added these in the Methods section. We did not compare the emissions outside China with other studies, which is beyond the scope of this study. However, the comparison of CEDS emissions with other emission inventories can be found in Hoesly et al. (2017), which includes detailed information of the CEDS emissions and will be submit very soon.

We also added a paragraph about uncertainty in BC emissions in the Conclusions and discussions section, as “Uncertainty in China BC emissions has been estimated as –43% to 93% by Lu et al. (2011), –50% to 164% by Qin

and Xie (2012), ±176% by Kurokawa et al. (2013), and –28 to 126% by Zhao et al. (2013). The BC emissions estimates used here for China in 2010 are 40% higher than those of Zhao et al. (2013) and Lu et al. (2011) and 30% higher than Klimont et al. (2016), in large part due to a higher estimate of BC emissions from coal coke production. Emissions from coke production are particularly uncertain given that "there are no measurements for PM2.5 and BC emissions" (Huo et al. 2012) available to guide inventory estimates. Total rest of the world emissions other than China, which appear to be a major contributor to burdens over western regions, are within 1% of those from Klimont et al. (2016)."

**Table S1.** Comparison of CEDS annual mean anthropogenic BC emissions in China with those used in other studies

| | Year | Anthropogenic emission in China (Gg/yr) |
|---|---|---|
| CEDS (Hoesly et al., 2016; this study) | 2010–2014 | 2467 |
| MIX (Li et al., 2017) | 2010 | 1765 |
| HTAP V2.2 (Janssens- Maenhout et al., 2015) | 2010 | 1741 |
| Lu et al. (2011) | 2010 | 1751 |
| Qin and Xie (2012) | 2009 | 1764 |
| Wang et al. (2012) | 2007 | 1879 |
| INTEX-B (Zhang et al., 2009) | 2006 | 1811 |

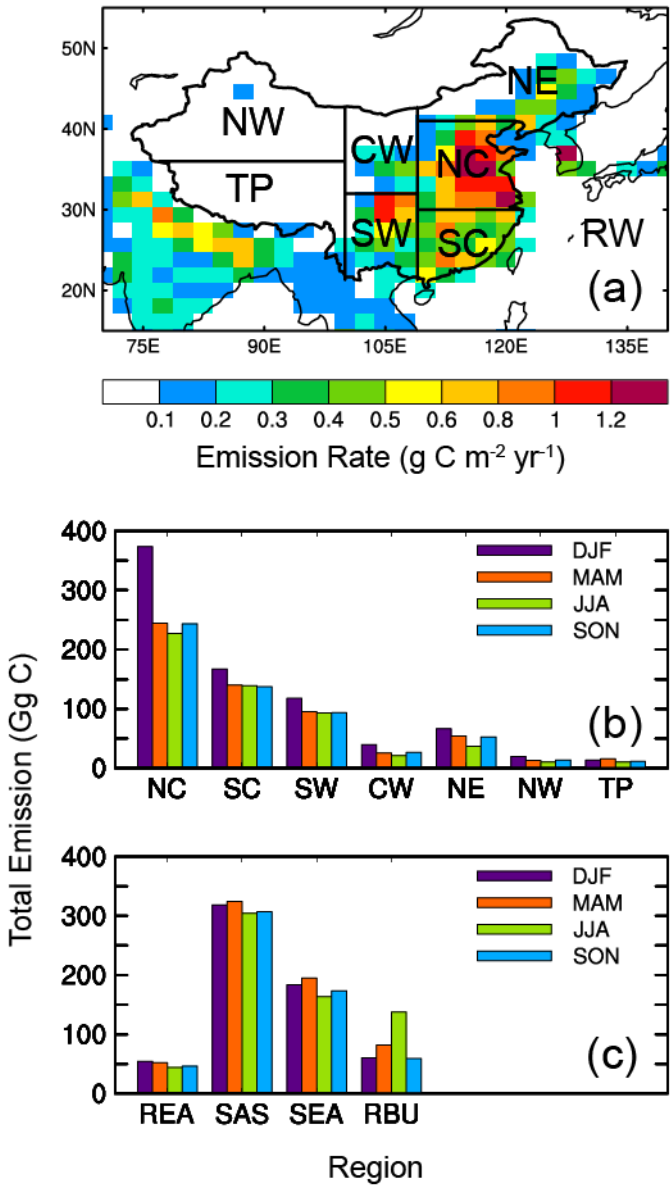


**Figure 1.** (a) Spatial distribution of annual mean total emissions (anthropogenic plus biomass burning, units: g C $m^{-2}$ $yr^{-1}$) of black carbon (BC) averaged over 2010–2014. The geographical BC source regions are selected as North China (NC, 109°E–east boundary, 30°–41°N), South China (SC, 109°E–east boundary, south boundary–30°N), Southwest China (SW, 100°–109°N, south boundary–32°N), Central-West China (CW, 100°–109°N, 32°N–north boundary), Northeast China (NE, 109°E–east boundary, 41°N–north boundary), Northwest China (NW, west boundary–100°E, 36°N–north boundary), and Tibetan Plateau (TP, west boundary–100°E, south boundary–36°N) in China and regions outside of China (RW, rest of the world). (b) Seasonal mean total emissions (units: Gg C, Gg = $10^9$g) of BC from the seven BC source regions in China and (c) emissions from rest of East Asia (REA, with China excluded), South Asia (SAS), Southeast Asia (SEA), and Russia/Belarussia/Ukraine (RBU).

(6) Lines 261-263

You can show the contributions from outside China quantitatively from the tagged simulation results.

Response:

We show the quantitative contributions from outside China in the Results section. It is not appropriate to directly compare the contribution to surface concentrations and that to column burden. Therefore, we decide to remove this sentence to avoid duplication and potential misunderstanding.

(7) Line 281

Please describe the reason of BC underestimation by up to a factor of 20.

Response:

We have discussed possible causes of this underestimation of BC in the following part of the Results section and in the Discussion section as well:

"Note that the model largely underestimates BC concentrations over China, compared to the observation, which has also been reported in many previous studies using different models and different emission inventories (e.g., Liu et al., 2012; Fu et al., 2012; Huang et al., 2013; H. Wang et al., 2013; Q. Wang et al., 2014; R. Wang et al., 2014; Li et al., 2016). One possible reason is that in situ measurements are point observations, while the model does not treat the subgrid variability of aerosols and assumes aerosols are uniformly distributed over the grid cell. R. Wang et al. (2014) found a reduction of negative bias (from −88% to −35%) in the modeled surface BC concentrations when using high-resolution emissions and modeling at 0.5° X 0.7° resolution. They find, however, that modeling over the North China Plain at an even higher resolution of 0.1°, further reduced the surface concentration bias there from 29% to 8%. This result indicates that the siting of observational stations can result in an artificial bias when comparing with relatively coarse model results. Further investigation of this siting/resolution bias is warranted, including investigation on whether this type of bias might extend, presumably to a lesser extent, also to AAOD measurements.

Further reasons that could contribute to this bias are emission underestimation or inaccurate aerosol processes in the model. Given that the differences between modeled and observed AAOD over eastern China are relatively small (–18%), we conclude that, given current evidence, the total amount of atmospheric BC in these simulations is reasonable at least in this sub-region.

Over eastern China, the BC concentrations are dominated by local emissions in this study, with local contribution of 64–93%. The underestimation of simulated BC concentrations over eastern China is more likely due to either underestimation of local emissions, too much aerosol removal within these regions, or resolution bias between observations and model grids. Over western China, 22–76% of the BC originates from emissions outside China.

Thus biases of simulated BC concentrations could also come from underestimation of emissions outside China and or too much removal of BC during long-range transport. Satellite data are a promising method to validate modeling and emissions inventories, given that they do not depend on the location of observing stations, providing more uniform spatial coverage. A comparison of modeled AAOD and satellite aerosol index (AI) provides an indication that the modeled burden in western China is underestimated, although the role of dust needs to be better characterized."

(8) Line 343

I cannot find large sources of BC in Northwest China in Figure 1a. Does the description here mean that there may be large sources of BC which are not considered in the emission inventory? Can you show the contribution of BC and dust to AAOD (in model) over this region? I think dust is dominant over this region.

Response:

Yes, underestimation of emissions over Northwest China could be part of the story. In the source attribution analysis, we found that emissions from outside China also contribute substantially to BC concentrations in Northwest China. Therefore the underestimation in BC concentrations could also come from low biases in emissions outside China or too much removal during along the transport pathways in the model.

Dust AAOD is indeed dominant over Northwest China (Fig. A). Potential biases in dust simulation could also lead to the difference in AAOD between model and observation. That is why we noted the potential bias from dust simulation, as "However, it is difficult to draw a firm conclusion, given the likely differential role of dust that also contributes to AAOD and AI, biases in modeling dust, and possible biases in the satellite derived AI values." And "A comparison of modeled AAOD and satellite aerosol index (AI) provides an indication that the modeled burden in western China is underestimated, although the role of dust needs to be better characterized."

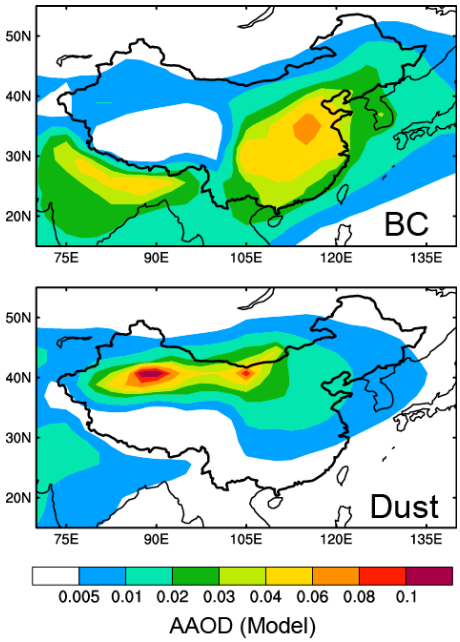


Figure A. Simulated annual mean AAOD of BC and dust.

(9) Lines 667-669

Related to the comment (2), is an internally-mixed treatment used in the calculations of AAOD? If so, AAOD should be lower (underestimated more) when more realistic BC mixing state treatment is used in the optical calculations.

Response:

CAM5 assumes internal mixing between BC and other components in the same mode, but external mixing between modes in the calculations of optical properties of the mixture of aerosol particles. Compared to the more sophisticated shell-core treatment, the current treatment might underestimate the absorption of aerosol. The aerosol mixing state along with the representation of size distribution is still quite uncertain. There is no strong evidence suggesting which one is more realistic, so we prefer not to draw such a conclusion here.

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