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Responses to Reviewer #2

Yang et al. investigated the BC source attributions (or more specifically, region attributions) in China with a source-tagging technique by employing a global climate model, NCAR's Community Earth System Model. They found out that BC emissions from local (inside China) and non-local (outside China) are both generally important contributions to air quality in different regions of China, BC outflow from East Asia and direct radiative forcings. Overall, this paper is a helpful addition to our community that attempts to improve our understanding of BC source-receptor relationship. This paper generally reads well and is within the scope of ACP. However, before it can be accepted for publication in ACP, I have several comments that need to be properly addressed.

Major comments:

An important part of this study was to quantify the BC source contributions to trans-boundary and trans-pacific transport. In terms of model performance evaluation, this study only validated model simulations with observed BC surface concentrations from CAWNET and AAOD from AERONET over China. We don't know the efficiency of BC outflow from East Asia. In this paper, it obviously missed the evaluations of model simulated vertical profiles of BC against aircraft campaign observations, e.g. A-FORCE and HIPPO, which should be employed to compare with model simulations.

Response:

Thanks for the suggestion. The simulated BC vertical profile in CAM5 has been extensively evaluated in many previous studies. Liu et al. (2012) compared the observed and simulated BC vertical profiles in the tropics, middle latitudes, and high latitudes from six aircraft campaigns: AVE Houston (NASA Houston Aura Validation Experiment), CR-AVE (NASA Costa Rica Aura Validation Experiment), TC4 (Tropical Composition, Cloud and Climate Coupling), CARB (NASA initiative in collaboration with California Air Resources Board), ARCTAS (NASA Arctic Research of the Composition of the Troposphere from Aircraft and Satellite), and ARCPAC (NOAA Aerosol, Radiation, and Cloud Processes affecting Arctic Climate), as well as BC vertical profiles over the Arctic Ocean and the remote Pacific Ocean during the HIPPO (HIAPER Pole-to-Pole Observations) campaign. They found that measured BC mixing ratios showed a strong gradient from the boundary layer to the free troposphere in the tropics, while modeled BC mixing ratios showed a smaller decrease with altitude in the free troposphere, thus overestimating observations above 500 hPa. Compared to HIPPO campaign, the CAM5 model captured the vertical variations of BC mixing ratio reasonably well in the

SH high latitudes and NH and SH mid-latitudes. However, modeled BC showed less vertical reduction in the tropics, thus significantly overestimating BC in the upper troposphere.

Wang et al. (2013) implemented in CAM5 a unified treatment of wet removal and vertical transport of aerosols by convection, which included an explicit secondary activation of aerosols being laterally entrained into convective clouds above cloud base. The comparisons between the new CAM5 simulated vertical profiles of BC mass mixing ratios and the HIPPO and the field campaign aircraft observations showed a substantial improvement in the simulation of BC in mid- and upper troposphere, where the excessive BC was significantly reduced. All of these key model improvements by Wang et al. (2013) have been included in the version of CAM5 being used in the present study. Therefore, we did not duplicate the evaluation of BC vertical profiles with HIPPO observation. We have now revised the description before model evaluation to make it clear, as "The simulations of aerosols, especially BC, using CAM5 have been extensively evaluated against observations including aerosol mass and number concentrations, vertical profiles, aerosol optical properties, aerosol deposition, and cloud-nucleating properties in several previous studies (e.g., Liu et al., 2012, 2016; H. Wang et al., 2013; Ma et al., 2013b; Jiao et al., 2014; Qian et al., 2014; R. Zhang et al., 2015a,b)."

In addition, as the referee suggested, we have added a comparison of the simulated BC vertical profile with A-FORCE measurements over East Asia (see Figure S2). The model successfully reproduces the vertical profile of BC. The bias is relatively small. We have also added a relevant discussion to the revised manuscript, as "Figure S2 compares the observed and simulated vertical profiles of BC concentrations in the East-Asian outflow region. The model successfully reproduces the vertical profile of BC that was measured in March–April 2009 during the A-FORCE field campaign and reported by Oshima et al. (2012)."

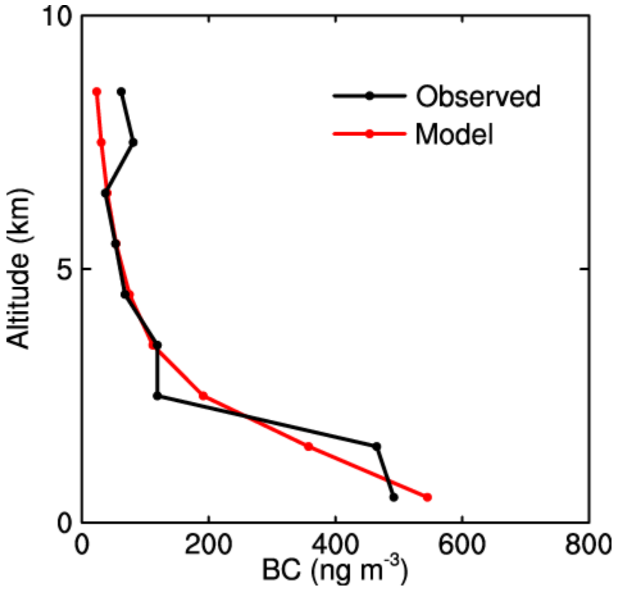

Figure S2. Observed and simulated mean vertical profiles of BC concentrations in the East-Asian outflow region. The observed BC profile is from the A-FORCE field campaign conducted over the Yellow Sea, the East China Sea, and the western Pacific Ocean in March–April 2009 (Oshima et al., 2012).

Other minor comments:

Line 124: the reference Hoesly et al., 2016 is missing in the reference list.

Response:

The paper is still in preparation. A draft can be made available to referees upon request.

Line 162: A brief description of dry/wet deposition scheme for BC in CAM is lacking here, especially the wet scavenging and how it is improved following H. Wang et al. (2013).

Response:

Aerosol dry deposition velocities are calculated using the Zhang et al. (2001) parameterization. The wet deposition of aerosols in our CAM5 model includes in-cloud wet removal (i.e., activation of interstitial aerosols to cloud-borne particles followed by precipitation scavenging) and below-cloud wet removal (i.e., capture of interstitial aerosol particles by falling precipitation particles) for both stratiform and convective clouds. Aerosol activation is calculated with the parameterization of Abdul-Razzak and Ghan (2000) for stratiform cloud throughout the column and convective cloud at cloud base, while the secondary activation above convective cloud base has a simpler treatment with an assumed maximum supersaturation in convective updrafts (H. Wang et al., 2013). The unified treatment for convective transport and aerosol wet removal along with the explicit aerosol activation above convective cloud base was developed by H. Wang et al. (2013) and included in the CAM5 version being used in this study. As discussed in the response to the major comment, this implementation reduces the excessive BC aloft and better simulates observed BC concentrations in the mid- to upper-troposphere.

We have now added these descriptions to the Methods section.

Line 334-336: This sentence should be corrected as “AI derived from Total Ozone Mapping Spectrometer (TOMS) measurements also shows similar pattern as simulate AAOD (Fig. S2).”

Response:

Corrected.

Line 339-344: What's the assumption here? Why the ratio of AAOD to AI should be the same between western and eastern China? What is the role of dust here in assisting the speculation?

Response:

Based on the comparison of AAOD between the model simulation and AERONET, we found that the model reproduced well the observed AAOD over eastern China. Therefore, we assume that the ratio of modeled AAOD and satellite AI (indicator for absorbing aerosols) is correct over eastern China. The AAOD/AI over eastern China is much larger than western China, suggesting that the ratio AAOD/AI is lower than the true value and AAOD or BC burden is likely underestimated in the model. Both AAOD and AI represent absorbing aerosols, the ratio of AAOD to AI may be different but similar between different regions in China. The difference in the ratio between eastern and western China is quite large, suggesting the existence of a significant bias. This is consistent with the contrast of biases in near-surface BC concentrations between the two regions (shown in Fig. 3). However, both BC and dust can contribute to AAOD and AI. Potential biases in the modeled dust could also lead to the inconsistence of AAOD/AI between eastern and western China.

We have revised the description to make it clear, as "If we assume that the simulated AAOD do not have large biases over eastern China based on the evaluation against observations shown above (Fig. 3b and Table S3), then this difference hints a possible underestimation of BC column burden in the model over the western regions. However, it is difficult to draw a firm conclusion, given the likely differential role of dust in eastern vs western China. This differential likely also contributes to AAOD biases in modeling dust and may also impact biases in the satellite derived AI values."

Line 424-426: I think BC emissions from SC are also important for the column burdens over continental China in some seasons (e.g. JJA and SON), which needs to be outlined as well.

Response:

Thanks for the suggestion. We have now added the SC contribution to column burden, as "Column burdens of BC averaged over continental China mainly originate from emissions in North China, South China and outside China, with relative contributions ranging from 31–42%, 16–24% and 14–31%, respectively."

Line 443: "Figure S4a" should be replaced with "Fig. S5a".

Response:

Corrected.

Line 443-463: It is helpful for the authors to make supplemental plots showing the anomalous winds during polluted days that favor the accumulation of pollutants over each region.

Response:

We did show in Fig. 8 the anomalous winds at 850 mb between polluted and normal days for each region during winter.

Line 505-509: Why the authors only choose the latitude range along longitude 150°E, not a domain covering East China Sea and West Pacific to quantitatively assess the BC contributions from China and outside China, similar to that impact over West United States?

Response:

The outflow of aerosols, defined as the column-integrated aerosol flux or concentration along a vertical cross-section, is used to characterize the export of BC from East Asia. This calculation of outflow follows previous studies and thus is comparable to the results in these studies (Hadley et al., 2007; Matsui et al., 2013; Yang et al., 2015). In addition, using a region around 150°E does not change the values significantly (e.g., contribution from China changes from 53% for the outflow at150°E to 54% for an average over 145–155°E). We don't mean to assess the contributions from China and outside China to air quality over this region.

Line 509-510: I get lost here. It is not clear to me that 58% contribution from China emissions is for outflow or something else. Authors need to clarify this.

Response:

Clarified as "The yearly contribution from emissions from China to outflow from East Asia in this study is 58%, similar to the contribution of 61% in Matsui et al. (2013) calculated based on eastward BC mass flux using WRF-CMAQ model with INTEX-B missions."

Line 531-538: I think the authors should list a table to compare your results with other studies, including annual BC emission budgets, burden, lifetime, DRF and DRF efficiency.

Response:

Thanks the suggestion. We have added Table S5 to compare these values with previous studies.

We have also added a discussion of this comparison in the manuscript, as "The total DRF of BC averaged over continental China simulated in this study is 2.27 W m^{-2}, larger than 0.64–1.55 W m^{-2} in previous studies (Wu et al., 2008; Zhuang et al., 2011; Li et al., 2016), probably due to the different emissions in the time periods of study, as shown in Table S5." And "The annual mean and regional mean DRF efficiency in China is 0.91 W m^{-2} Tg^{-1}, within the range of 0.41–1.55 W m^{-2} Tg^{-1} from the previous studies (Table S5)."

Table S5. Comparison of the simulated annual mean emission, burden, DRF and DRF efficiency in China in this study with the values reported in three previous studies.

Reference	Model	Year	Emission in China (Gg yr^{-1})	Burden (mg m^{-2})	DRF (Wm^{-2})	DRF efficiency (W m^{-2} Tg^{-1})
Wu et al. (2008)	RegCM3	2000	1005	0.55–1.42	0.64–1.55	0.64–1.55
Zhuang et al. (2011)	RegCCMS	2006	1811	1.12	0.75	0.41
Li et al. (2016)	GEOS-Chem	2010	1840		1.22	0.66
This study	CESM	2010–2014	2497	1.45	2.27	0.91

Line 654-655: Other modeling studies also found model low bias over China using CAWNET, e.g. Huang et al., 2013; Wang et al., 2014, which can be referenced here.

Huang, Y., S. Wu, M. K. Dubey, and N. H. F. French, Impact of aging mechanism on model simulated carbonaceous aerosols, Atmos. Chem. Phys., 13, 6329–6343, doi:10.5194/acp-13- 6329-2013, 2013.

Wang, Q., D.J. Jacob, J.R Spackman, A.E. Perring, J.P. Schwarz, N. Moteki, E.A. Marais, C. Ge, J. Wang and S.R.H. Barrett, Global budget and radiative forcing of black carbon aerosol: constraints from pole-to-pole (HIPPO) observations across the Pacific, J. Geophys. Res., 119, 195- 206, 2014.

Response:

Added.

Line 669: “and” is missing between “modeled” and “observed”.

Response:

Added.

References:

Zhang, L. M., Gong S. L., Padro J. and Barrie L.: A size-segregated particle dry deposition scheme for an atmospheric aerosol module, Atmos. Environ., 35, 549-560, doi:10.1016/S1352-2310(00)00326-5 ,2001.

Abdul-Razzak, H., and Ghan S. J.: A parameterization of aerosol activation: 2. Multiple aerosol types, J. Geophys. Res., 105, 6837–6844, doi:10.1029/1999JD901161, 2000.

Wang, H., R. C. Easter, P. J. Rasch, M. Wang, X. Liu, S. J. Ghan, Y. Qian, J.-H. Yoon, P.-L. Ma, and V. Vinoj (2013), Sensitivity of remote aerosol distributions to representation of cloud-aerosol interactions in a global climate model, Geosci. Model Dev., 6, 765–782, doi:10.5194/gmd-6-765-2013.

Hadley, O. L., V. Ramanathan, G. R. Carmichael, Y. Tang, C. E. Corrigan, G.

C. Roberts, and G. S. Mauger (2007), Trans-Pacific transport of black carbon and fine aerosols (D < 2.5 μm) into North America, J. Geophys. Res., 112, D05309, doi:10.1029/2006JD007632.

Matsui, H., M. Koike, Y. Kondo, N. Oshima, N. Moteki, Y. Kanaya, A. Takami, and M. Irwin (2013), Seasonal variations of Asian black carbon outflow to the Pacific: Contribution from anthropogenic sources in China and biomass burning sources in Siberia and Southeast Asia, J. Geophys. Res. Atmos., 118, 9948–9967, doi:10.1002/jgrd.50702.

Yang Y., H. Liao, and S. Lou (2015), Decadal trend and interannual variation of outflow of aerosols from East Asia: Roles of variations in meteorological parameters and emissions, Atmos. Environ., 100, 141-153, doi:10.1016/j.atmosenv.2014.11.004.

Oshima, N., Y. Kondo, Moteki N., Takegawa N., Koike M., Kita K., Matsui H., Kajino M., Nakamura H., Jung J. S., and Kim Y. J.: Wet removal of black carbon in Asian outflow: Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign, J. Geophys. Res., 117, D03204, doi:10.1029/2011JD016552, 2012.

Huang, Y., S. Wu, M. K. Dubey, and N. H. F. French, Impact of aging mechanism on model simulated carbonaceous aerosols, Atmos. Chem. Phys., 13, 6329–6343, doi:10.5194/acp-13- 6329-2013, 2013.

Wang, Q., D.J. Jacob, J.R Spackman, A.E. Perring, J.P. Schwarz, N. Moteki, E.A. Marais, C. Ge, J. Wang and S.R.H. Barrett, Global budget and radiative forcing of black carbon aerosol: constraints from pole-to-pole (HIPPO) observations across the Pacific, J. Geophys. Res., 119, 195- 206, 2014.