Response to Anonymous Referee # 3

(Note: Reviewer comments are listed in grey, and responses to reviewer comments are in black. Pasted text from the new version of the paper is in italics.)

The authors incorporate the impact of microphysical processes on the wet deposition of black carbon in the CAM5 global climate model. With this parameterization, they carry out a systematic evaluation of the importance of various microphysical processes on the distribution and radiative forcing of black carbon. Global distributions of black carbon remain highly uncertain, and this study provides a novel and substantial contribution towards the understanding a key piece of this complex problem. The paper is well-structured and the presentation is clear. I recommend minor revisions.

We greatly appreciate the reviewer's thorough and constructive review. We believe revising the paper according to the reviewer's comments has considerably improved the paper. We have merged all of the suggestions into the revised manuscript. Please see our response to each comment below:

I have two main comments on how the conclusions can be better supported:

1. Figure 2: authors have two full years of usable simulation data (2009-2010) and one partial year (2011). Do convection scavenging, aerosol activation, ice nucleation, evaporation, and below cloud scavenging dominate when the other years are considered?

   Thanks for bringing up this important issue. We have extended our simulation to the end of 2011. Following the reviewer's comment, we plot the zonal mean fractional changes averaged in year 2010 and 2011. The figure below is very similar to figure 2, which shows the changes for year 2009. Simulations in year 2010 and 2011 also indicate that convection scavenging, aerosol activation, ice nucleation, evaporation, and below cloud scavenging dominate BC vertical distribution.

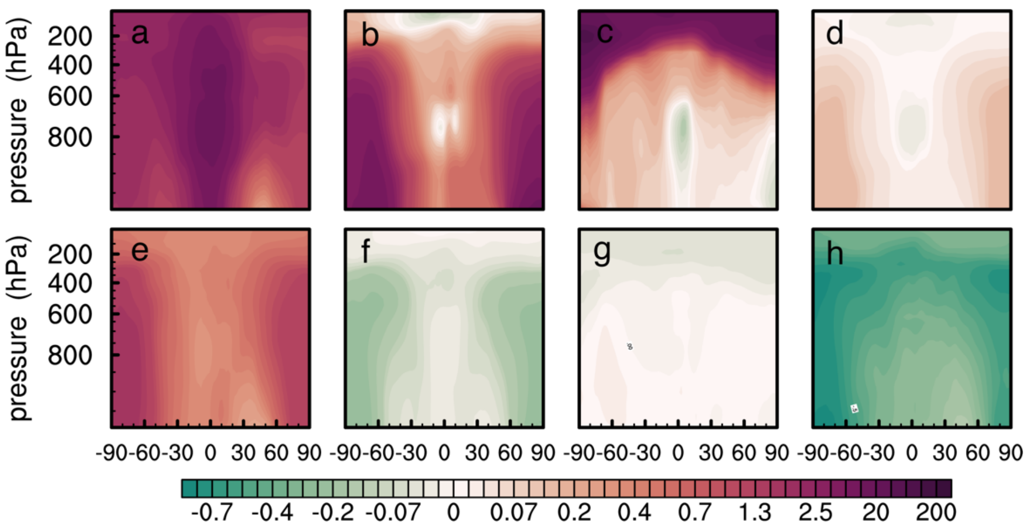

Figure. Zonal mean fractional changes (unitless) in BC concentrations averaged in year 2010 and year 2011 relative to BASE in eight sensitivity simulations when the influence of one cloud process on BC is

turned off. The sensitivity simulations are described in section 2.3, including (a) NO CONVECTION (no convection scavenging), (b) NO CCN (no cloud activation), (c) NO IN (no ice nucleation), (d) NO RIMING (no riming), (e) NO BELOW CLOUD (no below cloud scavenging), (f) NO BERGERON (no Bergeron process), (g) NO CLOUD EVAP (no evaporation of cloud water/ice sedimentation), and (h) NO PRECIP EVAP (no evaporation of rain/snow).

2. Figure 5: The color bar saturates too quickly, making it hard to compare between simulations. In particular, it appears that convective scavenging (a) and cloud activation (b) are on par in the mid-latitudes, rather than the claim that the former dominates. Also, it is difficult to tell the changes in vertical profile, which is relevant for the section on radiative forcing.

Thanks for this great suggestion. Following the reviewer's comment, we have modified the color table in Figure 5 to more clearly demonstrate the results. As the reviewer mentioned, the effects of convective scavenging and activation are on par in the mid-latitudes, however, the influence of NO CONVECTION on global burden is the largest among all sensitivity simulations. Therefore, we conclude that convection dominates BC burden. We also have plotted Figure 6 to show the influences of different cloud processes on BC vertical distribution.

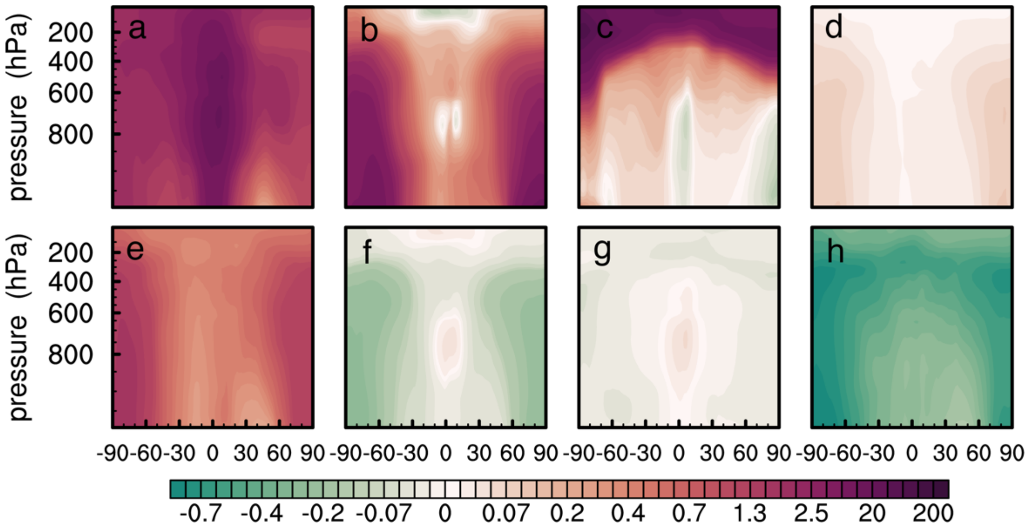

Figure 5. Annual zonal mean fractional changes (unitless) for year 2009 in BC concentrations relative to BASE in eight sensitivity simulations when the influence of one cloud process on BC is turned off. The sensitivity simulations are described in section 2.3, including (a) NO CONVECTION (no convection scavenging), (b) NO CCN (no cloud activation), (c) NO IN (no ice nucleation), (d) NO RIMING (no riming), (e) NO BELOW CLOUD (no below cloud scavenging), (f) NO BERGERON (no Bergeron process), (g) NO CLOUD EVAP (no evaporation of cloud water/ice sedimentation), and (h) NO PRECIP EVAP (no evaporation of rain/snow).

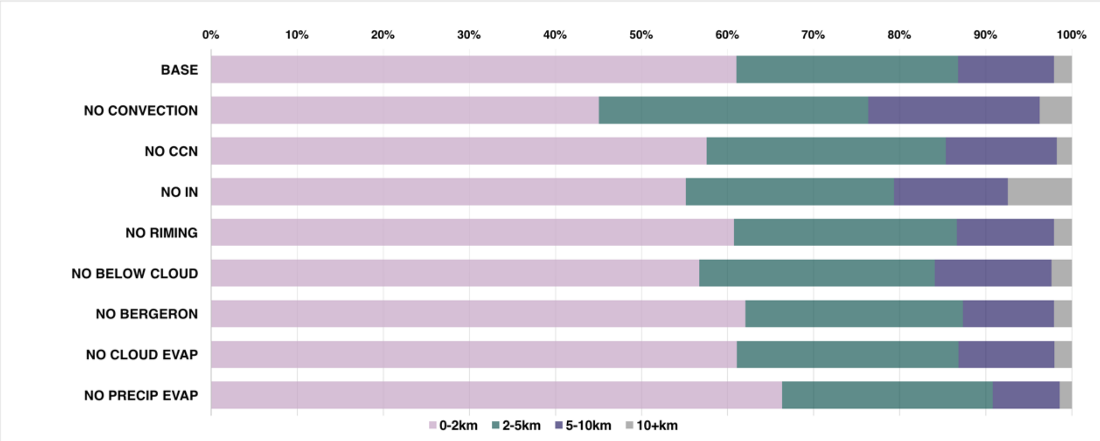

Figure 6. Fraction of global BC burden at four altitude bands for year 2009 in the BASE simulation using our new wet removal scheme described in section 2.2, and sensitivity simulations when the influence of each cloud process on BC is turned off. The sensitivity simulations are described in section 2.3, including NO CONVECTION (no convection scavenging), NO CCN (no cloud activation), NO IN (no ice nucleation), NO RIMING (no riming), NO BELOW CLOUD (no below cloud scavenging), NO BERGERON (no Bergeron process), NO CLOUD EVAP (no evaporation of cloud water/ice sedimentation), NO PRECIP EVAP (no evaporation of rain/snow).

I encourage the authors to frame their discussion on the direct radiative forcing (DRF) in the context of the major factors known to affect the direct radiative effect (e.g. in Equation 6.1 of Bond et al. (2013)): emissions, lifetime, absorption cross-section, and absorption efficiency. In particular, recent work suggests that there has been both an underestimate in emissions (e.g. Cohen and Wang (2014)) and overestimate in lifetime, and that the two factors act to cancel each other (Hodnebrog, Myhre, and Samset 2014). I agree that wet deposition is an important piece of constraining DRF; my concern is that a reader may walk away thinking that it is the only factor. In the introduction, the authors may want to comment on the relative roles of transport vs removal, as in the introduction of Q. Wang et al. (2014).

Excellent point. I agree that our study only consider wet removal as a factor contribute to uncertainty of DRF, which may mislead our reader. We reframe the beginning of section 5 as below :

"*Emissions, lifetime, absorption cross-section, and absorption efficiency can all affect BC DRF (Bond et al., 2013). The total impacts of uncertainties in these processes on BC DRF estimates is complex. For instance, recent studies suggest that there has been both an underestimate in emissions (e.g.Cohen and Wang (2014)) and an overestimate in lifetime, and that the two factors act to cancel each other (Hodnebrog et al., 2014). In this study, we only focus on how cloud processes influence BC DRF via altering BC wet removal*."

Meanwhile, we also added a paragraph in the introduction to compare the relative importance of emission, transport, dry deposition, and wet removal in controlling BC concentrations:

"The inter-model discrepancies and disagreement between models and measurements reflect uncertainties in emissions, transport, dry deposition, and wet scavenging of BC simulation. The uncertainties in BC concentrations over source regions are mainly contributed by errors in emission inventories. Fu et al. (2012) and Leibensperger et al. (2012) suggest that emission inventories lead to normalized mean bias of less than 2 against observations over source regions. Using inert 222Rn as a tracer, previous studies show that pollution transport in three dimensional models is fairly well constrained with observations; seasonality and magnitude of profiles 222Rn vertical profiles are captured by the models (Jacob et al., 1997;Stockwell and Chipperfield, 1999). Dry and wet deposition are the sinks of BC. Previous literature suggests that global total wet deposition is 3-6 times larger than dry deposition (Jurado et al., 2008;Huang et al., 2010;Zhang et al., 2015). In the remote troposphere, wet scavenging is considered as the most important source of BC simulation uncertainties (Koch et al., 2009;Schwarz et al., 2010;Croft et al., 2010;Liu et al., 2011;Wang et al., 2014). "

Specific comments

1. p1, line 26: I don't see significance tests, perhaps rephrase as 'largest impact'

   Thanks for this comment. We have changed the phrase:

   "*Convective scavenging is found to have the largest impact on BC concentrations at mid-altitudes over the tropics and even globally.*"

2. p1, line 29: do you mean "convection scavenging mainly increases the fraction of column BC below 5 km"?

   Thanks. Based on Figure 6, convection scavenging greatly influences the fraction of BC at all altitude bands. We have change the sentence to make this point clearer:

   "*As for BC vertical distributions, convective scavenging greatly influences BC fractions at different altitudes.*"

3. NO BERGERON and NO PRECIP EVAP are misspelt as NO BEGERON and NO PERCIP EVAP in some cases (eg p19 line 7, p21 line 21, Figure 7)

   Thanks for catching this mistake. We have modified the sentences and Figure 7, as shown below:

   "*Therefore, NO BERGERON decreases BC concentrations in the Arctic relative to BASE (Fig. 5(f)).*"

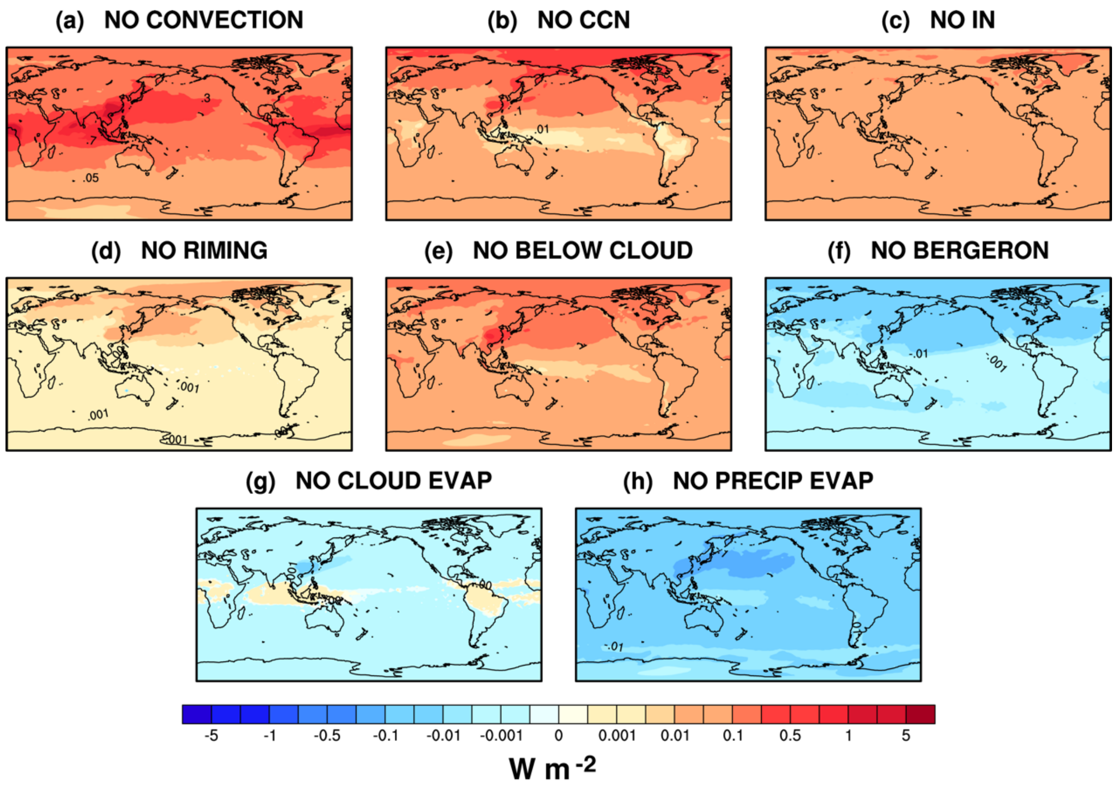

Figure 7. Change in global radiative forcing of BC estimated by sensitivity simulations relative to BASE for year 2009. The sensitivity simulations are described in section 2.3, including (a) NO CONVECTION (no convection scavenging), (b) NO CCN (no cloud activation), (c) NO IN (no ice nucleation), (d) NO RIMING (no riming), (e) NO BELOW CLOUD (no below cloud scavenging), (f) NO BERGERON (no Bergeron process), (g) NO CLOUD EVAP (no evaporation of cloud water/ice sedimentation), and (h) NO PRECIP EVAP (no evaporation of rain/snow).

4. p6, line 6: 'more accurately simulates' -> as compared to? 1

Thanks for pointing this out. The shallow convection that we used was compared to Hack et al. (1994) shallow convection scheme in CAM3 and CAM4. We revised the sentence:

"*Shallow convection is treated with a parameterization developed by Park and Bretherton (2009) that computes vertical velocity and fractional area of convection, and more accurately simulates spatial distribution of shallow column activity, as compared to Hack (1994) shallow convection scheme in CAM3 and CAM4.*"

5. p9, line 17: 'we turn off the impact of each cloud process on BC' -> I assume you mean that the changes in cloud processes do not affect the climate. Would be good to make clear.

Good suggestion. We have added the following sentence in section 2.3 to make it clear.

"Note that changes in cloud processes of sensitivity simulations do not affect the climate and there is no radiative feedback on the climate system from bulk BC tracers in this study."

6. p14, line 6: do you mean 1.9 kg/s?

Thanks for pointing out this mistake. We revised the number:

"*Similarly, sublimation of ice crystal sedimentation from the upper level converts* BC_{ice} *to* BC_{philic}*, at rate of 1.9 kg/s.*"

7. please be consistent in use of abbreviations (e.g. fig in p15 vs Fig in p16, figure in p18 line 24 vs Figure in p18 line 1)

Thanks. We have unified the use of abbreviations in the article.

References:

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, Journal of Geophysical Research: Atmospheres, 118, 5380-5552, 10.1002/jgrd.50171, 2013.

Cohen, J. B., and Wang, C.: Estimating global black carbon emissions using a top-down Kalman Filter approach, Journal of Geophysical Research: Atmospheres, 119, 307-323, doi:10.1002/2013JD019912, 2014.

Croft, B., Lohmann, U., Martin, R., Stier, P., Wurzler, S., Feichter, J., Hoose, C., Heikkilä, U., Donkelaar, A. v., and Ferrachat, S.: Influences of in-cloud aerosol scavenging parameterizations on aerosol concentrations and wet deposition in ECHAM5-HAM, Atmospheric Chemistry and Physics, 10, 1511-1543, 2010.

Fu, T. M., Cao, J. J., Zhang, X. Y., Lee, S. C., Zhang, Q., Han, Y. M., Qu, W. J., Han, Z., Zhang, R., Wang, Y. X., Chen, D., and Henze, D. K.: Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution, Atmos. Chem. Phys., 12, 2725-2746, 10.5194/acp-12-2725-2012, 2012.

Hack, J. J.: Parameterization of moist convection in the National Center for Atmospheric Research community climate model (CCM2), Journal of Geophysical Research: Atmospheres, 99, 5551-5568, doi:10.1029/93JD03478, 1994.

Hodnebrog, Ø., Myhre, G., and Samset, B. H.: How shorter black carbon lifetime alters its climate effect, Nature Communications, 5, 5065, 10.1038/ncomms6065 https://www.nature.com/articles/ncomms6065#supplementary-information, 2014.

Huang, L., Gong, S. L., Jia, C. Q., and Lavoué, D.: Importance of deposition processes in simulating the seasonality of the Arctic black carbon aerosol, Journal of Geophysical Research: Atmospheres, 115, doi:10.1029/2009JD013478, 2010.

Jacob, D. J., Prather, M. J., Rasch, P. J., Shia, R. L., Balkanski, Y. J., Beagley, S. R., Bergmann, D. J., Blackshear, W., Brown, M., and Chiba, M.: Evaluation and intercomparison of global atmospheric transport models using 222Rn and other short-lived tracers, Journal of Geophysical Research: Atmospheres, 102, 5953-5970, 1997.

Jurado, E., Dachs, J., Duarte, C. M., and Simo, R.: Atmospheric deposition of organic and black carbon to the global oceans, Atmospheric Environment, 42, 7931-7939, 10.1016/j.atmosenv.2008.07.029, 2008.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J., Balkanski, Y., Bauer, S., Berntsen, T., Bond, T. C., and Boucher, O.: Evaluation of black carbon estimations in global aerosol models, Atmospheric Chemistry and Physics, 9, 9001-9026, 2009.
Leibensperger, E. M., Mickley, L. J., Jacob, D. J., Chen, W. T., Seinfeld, J. H., Nenes, A., Adams, P. J., Streets, D. G., Kumar, N., and Rind, D.: Climatic effects of 1950–2050 changes in US anthropogenic aerosols – Part 1: Aerosol trends and radiative forcing, Atmos. Chem. Phys., 12, 3333-3348, 10.5194/acp-12-3333-2012, 2012.
Liu, J., Fan, S., Horowitz, L. W., and Levy, H.: Evaluation of factors controlling long-range transport of black carbon to the Arctic, Journal of Geophysical Research, 116, 10.1029/2010jd015145, 2011.
Park, S., and Bretherton, C. S.: The University of Washington shallow convection and moist turbulence schemes and their impact on climate simulations with the Community Atmosphere Model, Journal of Climate, 22, 3449-3469, 2009.
Schwarz, J., Spackman, J., Gao, R., Watts, L., Stier, P., Schulz, M., Davis, S., Wofsy, S. C., and Fahey, D.: Global-scale black carbon profiles observed in the remote atmosphere and compared to models, Geophysical Research Letters, 37, 2010.
Stockwell, D. Z., and Chipperfield, M. P.: A tropospheric chemical-transport model: Development and validation of the model transport schemes, Quarterly Journal of the Royal Meteorological Society, 125, 1747-1783, 1999.
Wang, Q., Jacob, D. J., Spackman, J. R., Perring, A. E., Schwarz, J. P., Moteki, N., Marais, E. A., Ge, C., Wang, J., and Barrett, S. R. H.: Global budget and radiative forcing of black carbon aerosol: Constraints from pole-to-pole (HIPPO) observations across the Pacific, Journal of Geophysical Research: Atmospheres, 119, 195-206, 10.1002/2013jd020824, 2014.
Zhang, L., Henze, D. K., Grell, G. A., Carmichael, G. R., Bousserez, N., Zhang, Q., Torres, O., Ahn, C., Lu, Z., Cao, J., and Mao, Y.: Constraining black carbon aerosol over Asia using OMI aerosol absorption optical depth and the adjoint of GEOS-Chem, Atmospheric Chemistry and Physics, 15, 10281-10308, 10.5194/acp-15-10281-2015, 2015.