Response to Referee #1

In this study, the authors examined evaluate the roles of aerosol-radiation interaction (ARI), aerosol-cloud interaction (ACI), black carbon (BC) and none BC (non-BC) aerosols in the formation and maintenance of the heavy fog event in early 15 December 2013 in the Yangtze River Delta (YRD) region in eastern China using WRF-Chem model. They found that ARI dominates this fog-haze episode while the effects of ACI are negligible. BC plays a more important role in fog formation than non-BC aerosols, inducing temperature contrast over land and sea and transports moister air to the YRD region. The topic of this study is interesting and the manuscript is well written. I would suggest publishing after addressing my comments below.

Response:

We are appreciated the comments, which are helpful to further improve the article.

My main concern is the method to distinguish the role of BC and non-BC. First, the authors turned off ARI and ARI+ACI to separate the roles of ARI and ACI, which is reasonable. Then the authors compare the simulation with ARI+ACI turned off and with anthropogenic BC emission removed to separate to roles of non-BC and BC, which could be inappropriate. These roles were quantified using different methods and comparing them could lead to an apple and orange comparison. Without BC emission, the internal mixing state could be changed, the impact of BC on vegetation could be changed, chemistry on aerosol surface could be changed, besides the BC-radiation and BC-cloud interactions. To separate the role of BC and non-BC, I would suggest the authors do another parallel simulation with all aerosol emissions turned off. Or at least discuss the biases of the results.

Response:

We have conducted four parallel numerical experiments in this study. At first, the control experiment (EXP_CTL) is conducted with complete emission and with ARI+ACI turned on. Then, anthropogenic BC emission was removed from the model with both ARI+ACI still turned on (EXP_NOBC). After that, we conducted the experiments that turned off both ARI and ACI (EXP_NOAER), which has neither ARI nor ACI from the BC and non-BC aerosols at all. In EXP_NOAER, we shut off the aerosol impact on the radiation transfer and cloud as well as the wet removal and cloud chemistry, which make this experiment equivalent to a parallel experiment that remove all aerosols regarding the interacting between aerosols and fog formation. So, the effect of BC can be calculated as the EXP_CTL - EXP_NOBC, the effect of non-BC is EXP_NOBC - EXP_NOAER. This method well separated the role of ARI and ACI, and distinguished the role of BC from the non-BC with less computational cost. And the role of BC was later proved be an important finding in this article, which was to enhance the moisture advection mainly through its radiative effect during the fog-haze event in the early December 2013 in the YRD region. To clarify this, we will add more description about the experimental design in section 2.2. In EXP_CTL, the BC and other aerosol are assumed to be internally mixed within each bin. Without BC emission, in EXP_NOBC, the particle aerosol would absorb much less shortwave radiation, since BC is the largest absorption particle in WRF (https://ruc.noaa.gov/wrf/wrf-chem/tutorial2018.htm, Page 11). This change due to BC's removal is mostly accountable for the cooling of the ambient atmosphere where high loadings of BC locate, thus indicating the heating effect of BC's role in this study. Not only that, the change to the mixing state of BC and other aerosols can also leads to the significant change to the optical property of the aerosol particle, which is not discussed in this paper. While, other changes, such as the change to the surface chemistry and the vegetation can also be important influencing factors. They are also not discussed in this work, since they do not affect the main mechanism for the advection-radiation fog that we investigated. However, this argument provides us some insight to the BC roles on affecting the fog in a more generally way, which needs more attention in the future work.

Minor comments:
Page 1 Line 24: 'heave' -> 'heavy'

Response:

Accepted. We will correct it in the revision.

Page 2 Line 15: In addition to impact on PBL, recent study also found BC can change land-sea thermal contrast and weaken the East Asian winter monsoon (Lou et al., 2019), which mechanism is very similar to this study, as well as large-scale circulations (Yang et al., 2019).

Lou, S., Y. Yang, H. Wang, S. J. Smith, Y. Qian, and P. J. Rasch, Black carbon amplifies haze over the North China Plain by weakening the East Asian winter monsoon, Geophys. Res. Lett., 46, 452–460, doi:10.1029/2018GL080941, 2019.

Yang, Y., S. J. Smith, H. Wang, C. M. Mills, and P. J. Rasch, Variability and timescales in the climate response to black carbon emissions, Atmos. Chem. Phys., 19, 2405-2420, doi:10.5194/acp-19-2405-2019, 2019.

Response:

Indeed, these references are quite related to this work. We will discusses and add these two new studies in the introduction and discussion part.

Page 4 Line 26: Please clarify that how did the authors turn off ARI and ACI in the model. What variables they excluded or fixed in model?

Response:

According to user guide of WRF-Chem (https://ruc.noaa.gov/wrf/wrf-chem/Users_guide.pdf) page 26 - 27, both of ARI and ACI can be turned on and off by configuring the namelist.input. The ARI is turned off by setting aer_ra_feedback = 0. The ACI is further turned off by setting the chem_opt to 201 (mosaic without aq), wet_scav_onoff = 0 (wet scavenging off), cldchem_onoff = 0 (cloud chemistry), and progn = 0 (turns off prognostic cloud droplet number), then the aerosol would have no impact on the cloud/fog droplet. The effect of the ACI calculated this way represented the ACI effect from the fog-haze event, which is reasonable to be insignificant compared with the ARI under heavy polluted condition.

Page 5 Line 4: Please provide the sites or data reservoir where the authors got all these data.

Response:

Here list the dataset used in this work. To make it more clear, we will add them in the acknowledgments.

1. NCDC dataset was obtain from https://gis.ncdc.noaa.gov/maps/ncei/cdo/hourly
2. The vertical profile sounding was retrieved from http://weather.uwyo.edu/upperair/sounding.html
3. The CNEMC dataset was gather from http://106.37.208.233:20035/ since 2013.

As for the sensible heat, shortwave radiation flux and BC concentration observation, they were retrieved from SORPES at NJU, Xianlin Campus, which we've like to provide to those who are interesting on the subject.

Figure 4: It should be days 2,5,6,"9".

Response:

Accepted. Figure 4 will be corrected in the revised manuscript.

Page 6 Line 22: underestimated during the occurrence of fog in Nanjing "in the model".

Response:

Accepted. We will revise this sentence.

Page 6 Line 26: a possible enhancement of the formation of secondary aerosols through aqueous phase or heterogeneous reactions "in the real world".

Response:

Accepted.

Page 7 Line 23-28: I am confused that why temperature over the sea was less sensitive to the reduction of incoming solar radiation but sensitive to aerosol warming effect? Both of them are *changes in radiative fluxes. Large heat storage capacity only implies that ocean is insensitive to heat or cooling effects (changes in radiative fluxes) compared to land. Instead, Lou et al. (2019) found that BC-induced heating over Bohai Sea, Yellow Sea, and East China Sea evaporated low cloud and increased high cloud, leading to larger warming over oceans east of China mainland.*

Response:

Since the clean atmosphere only absorbs a small fragment of the incoming solar radiation. The near surface layer, being close to the land/sea surface, is mostly warmed up during daytime by taking the sensible or latent heat that emitted from the surface. This process essentially transfers the solar energy into the internal energy of the underlying surface, and then pass it to the air near the surface. However, the water body has a large heat storage capacity than the land. That means the fact that the heating of the near-surface atmosphere would be slower over the water body (which is actually the main cause for the sea breeze). Thus, when the aerosols reduce the incoming solar radiation, the aerosol cooling effect on the near surface atmosphere is correspondingly less sensitive over the sea than that over the land. With the same amount of aerosol concentration increased over both land and sea, the near surface air temperature would have less reduction over the sea than the land surface. Part of the intercepted solar energy is absorbed by the particle like BC, if present, and warm the atmosphere directly. This heating effect from absorbing aerosol can be more efficient than taking the heat from the underlying water body during the daytime. Therefore, over the sea, the near surface temperature is generally more sensitive to the heating effect than the cooling effect of the aerosol. In our case, the BC's distribution over the sea surface and corresponding heating region matched well (Figure 10 and Figure S4), suggesting the heating effect from BC increase the air temperature in the near surface over the offshore region in the East China sea. Similar to our study, Wang et al. (2015) found as large as +0.3 °C feedback from aerosol heating effect over Gulf of Mexico and Atlantic Ocean and – 0.3 °C feedback over eastern U.S. during July 2006. As for the change to the cloud, the simulation result from both EXP_CTL (Figure R1a and b) show that the cloud cover over the eastern China is low before the formation of fog on 7 December. After, the BC is excluded from the model, less cloud cover condition merely changes (Figure R1c and d). And below or above the 850 hPa, the cloud cover or its change due to BC is much smaller. On the other hand, if BC change the cloud cover over the sea which lead to the heating over the surface, there would be a corresponding positive change in the Downward Shortwave Radiation (DSW). However, there're only negative change in the DSW (Figure R2e) in the heating region over the eastern China Sea (Figure R2b). By comparisons, impact from Total or non-BC indicate the increase of temperature over the East China Sea was mostly result from BC effect (Figure R2d and f). Therefore, in this study, we conclude the warming over the East China Sea is the direct result from the BC's heating effect over the sea for the early December 2013. This leads to the enhancement of moisture advection and consequently the increase of the moisture in YRD and adjacent offshore region in the near surface (Figure R2i) as discussed in the article.

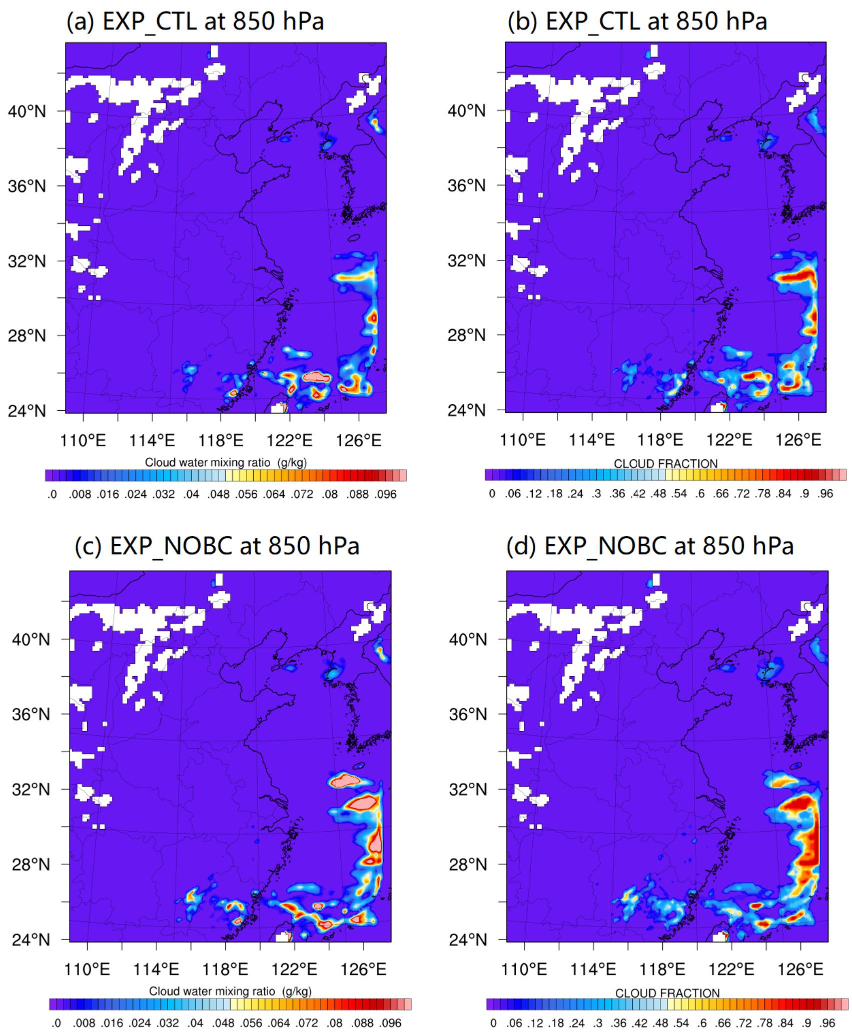

Figure R1. The mean cloud water mixing ratio and cloud fraction at 850 hPa over the eastern China during LST 8:00-17:00 6 December 2013. (a) and (b) are results from EXP_CTL, while (c) and (d) are result from EXP_NOBC.

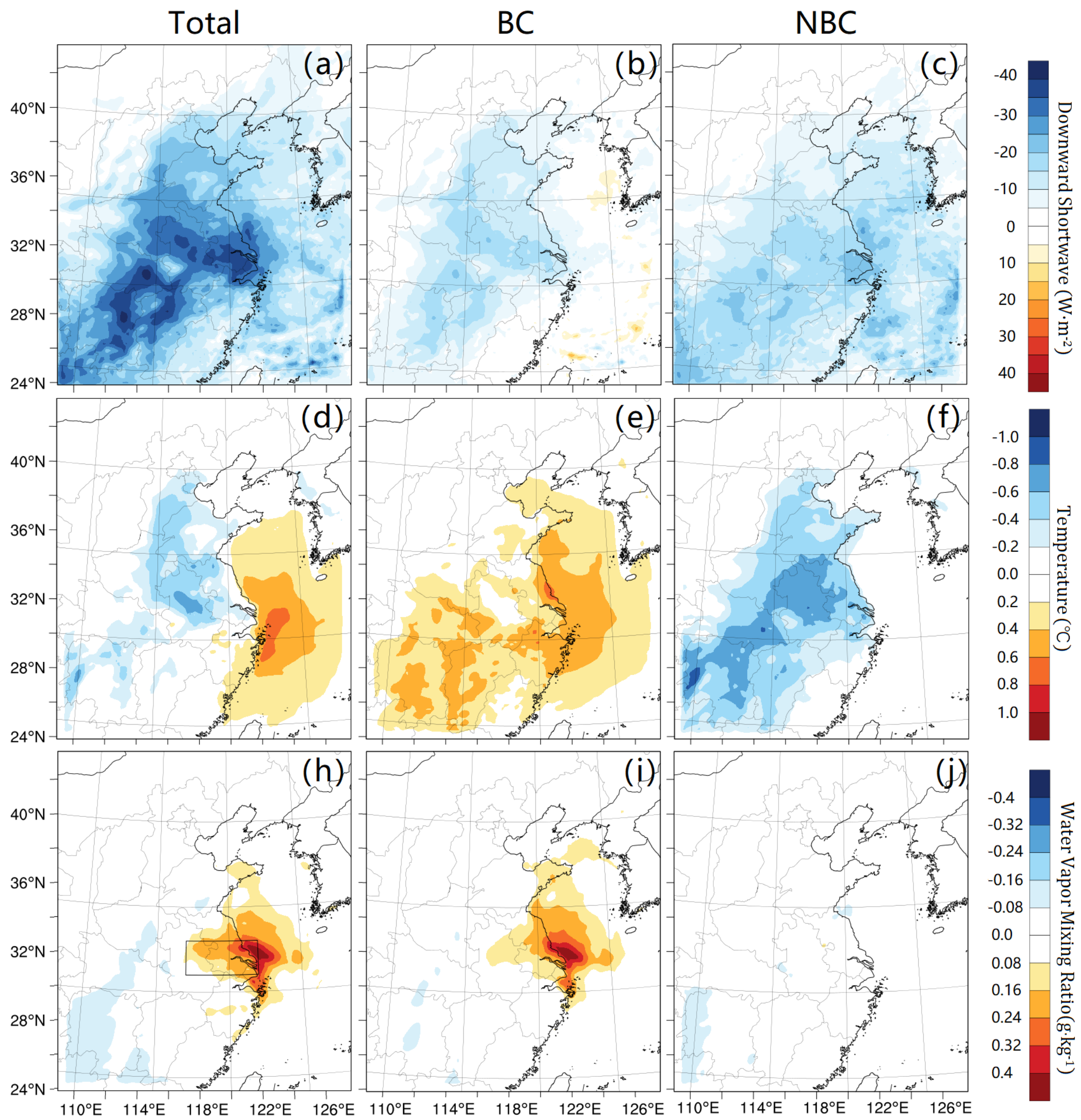

Figure R2. Average changes of DSR (upper), Temperature in the near surface (middle) and water vapor mixing ratio in the near surface (lower) induced by total effect of aerosols (left), effect of BC (middle) and effect of non-BC aerosols (right) in the daytime (08:00 – 17:00 LST) during 1-10 December 2013.

Page 8 Line 30: The BC impact on surface temperature depends on vertical location of BC and also depends on models.

Response:

Accepted. We will add more explanations to the response of near-surface atmospheric temperature to the radiative effect of BC being different to that of non-BC aerosols. And the statement of the uncertainties in simulating the BC impact on the near surface temperature in numerical modellings will be also included.

Page 9 Line 2: What does "other forcings" mean.

<u>Response</u>:

Here, the "other forcings" is referred to the other cause of the RH increase rather than the decrease air temperature induced by local aerosol cooling effect. Here, it was indicated that the moisture advection from outside the YRD region might be the cause of the large increase in RH over the YRD region on 6 December 2013. We will change "other forcing" to "other factors such as moisture advection" in order to avoid ambiguous indications.

References:

Wang, K., Zhang, Y., Yahya, K., Wu, S. Y., and Grell, G.: Implementation and initial application of new chemistry-aerosol options in WRF/Chem for simulating secondary organic aerosols and aerosol indirect effects for regional air quality, Atmos. Environ., 115, 716-732, doi: 10.1016/j.atmosenv.2014.12.007, 2015.