Response to the Reviewer #1

The authors presented black carbon (BC) observations from 2009 to 2019 at the Fukue island located downwind of China and inferred China's BC emission trends from these observations. They used the chemical transport model WRF-CMAQ to estimate the meteorological effects on BC concentrations, used the backward trajectory model HYSPLIT to attribute observed BC trends to emission source regions, and finally identified rapidly decreasing BC emissions from China, which are broadly consistent with the up-to-date bottom-up emission inventories. The comprehensive analysis that integrates several models and datasets gives a strong, convincing conclusion. This paper is well written and deserves publication after several issues addressed.

We appreciate the reviewer's careful reading and positive comments on our manuscript. Detailed point-by-point responses are given below.

Major comments:

1) The key to this method is estimating emission correction factors, assuming that all of the differences between observed and modeled BC are attributed to the errors in surface BC emissions. This assumption has some problems since both models and observations have their own uncertainties. Without considering these uncertainties in the estimation of the emission correction factors, the authors tend to overestimate the uncertainties in BC emission inventories and thus tend to overestimate the correction factors. Although I believe that China's BC emissions have been declining since 2010, the authors need to justify the methodology they used and to acknowledge that the method still has large uncertainties in the emission correction factors. And if I understand correctly, the WRF-CMAQ model used here has no modules simulating BC wet deposition, which can cause <10% of BC loss even under an APT less than 1 mm. The current manuscript lacks a detailed discussion on the deficiencies of the model as well as the uncertainties in the observations.

We agree that the uncertainties of observations and models are important and will clarify this point in the revised manuscript:

"Therefore the only factor that the model failed to replicate the observation<u>, except for their uncertainties,</u> was the emission trend." (Page 5, Lines 33-34, in the track change document)

And the methodologies and results of the revised estimation of systematic and random uncertainties in the observations and models will be clarified:

" From the average and a standard deviation of the monthly MAAP/COSMOS ratios, systematic and random uncertainties were estimated as ± 14% and ± 17%, respectively (± 22% in total)." (Page 4, Lines 8-9)

" We estimated the model uncertainties from meteorology to be ~± 16%, in simulating surface BC concentrations under conditions with negligible wet deposition, considering both horizontal and vertical inhomogeneities in the model and the spread of multi model simulations of CO over the East China Sea (Kong et al., 2019). The uncertainty was assumed to be contributed equally from systematic and random terms (± 12 %)." (Page 5, Lines 25-28)

Then, the uncertainties in the absolute emission correction factors (E(y)/REAS2.1(2008)) and their trends will be clearly mentioned as follows:
"Overall uncertainty in the estimated E(y)/REAS2.1(2008) values was estimated to be ± 27%, including those random and systematic from both model and observation (see Sect. 2). On the other hand, the uncertainty in its trend was estimated to be ± 21%, as influenced only by random uncertainties." (Page 9, Lines 9-11)
In Fig. 7a, the uncertainty range (a band with pale red color) will be expanded accordingly, to cover the overall uncertainty.

Also, the fact that the WRF/CMAQ model included wet deposition will be clarified. The effect of the wet deposition on the used data set with the adopted low-APT criteria (<1 mm) in this study will be clarified, using the ΔBC/ΔCO ratio (Kanaya et al., 2016), for both observations and model simulations:

" The median ΔBC/ΔCO ratio for the observational data with APT < 1 mm (*N* = 26423) was only 1.9 % lower than that for data with APT = 0 mm (*N* = 18907), suggesting insignificant influence." (Page 4, Lines 33- Page 5, Line 2)
" Wet deposition was represented with the cloud_acm_ae5 module. Similarly to the observational data, the modeled ΔBC/ΔCO ratio decreased with APT; the modeled median ΔBC/ΔCO ratio for data with APT < 1 mm (*N* = 26737) was 3.7 % lower than that for data with APT = 0 mm (*N* = 19197). The removal in the model appeared stronger than the observational trend (1.9 %) but the error introduced due to the wet deposition representation was estimated to be small (−2 %) when using the adopted criteria (APT < 1 mm)." (Page 5, Lines 14-18)

2) In order to compare with bottom-up emission inventories, the authors scaled China's emissions from REAS2.1 with the emission correction factors for several large regions (Fig. S3). The footprint map of BC observations mainly covers central and eastern China (Fig. 5), while the emission correction factors are applied over the whole of China. I am wondering how many China's provinces and their BC emissions can be well observed by the station at Fukue Island. And the uncertainty range of the red curve in Fig. 7a should be larger than the current estimates after considering the representation errors of emission correction factors for the whole of China.

We agree that the low sensitivity to the emissions from inland areas increases the uncertainty. In the revised manuscript, after stating that total BC emissions for China and its decadal trend were estimated "on a best effort basis" (page 9, lines 26) in this study, a detailed analysis on this point will be provided as follows (page 10, lines 7-17):

One caveat in this analysis would be that the emissions from inland areas (e.g., Sichuan) produced less observational signal at Fukue than that from the CEC, while the footprint (Fig. 5) covered inland areas to 100° E well. We estimated 36% of BC emissions from China, particularly those from south or west areas including Sichuan, Guizhou, and Guangdong provinces (pale colors in Fig. S6), might have resulted in only 5% of signal at Fukue, when the pseudo signal synthesized by multiplying the footprint with the emission rates was analyzed. Thus any emission trends in such areas with less signal weight might be easily overlooked. A broken line in Fig. 7a represents a case when emission from such areas is assumed unchanged during the study period. Even in such a case, the trend was clearly negative and was within the range of overall uncertainty. We assumed that this scenario provided the possible weakest negative trend, considering that Zhang et al. (2019) reported negative trends in BC concentrations in those provinces similarly to CEC. Obviously reliable BC measurements at other locations with larger footprint over the areas are required to improve the analysis in the future. Here, nonetheless, we were able to conclude a decadal decreasing trend in the Chinese BC emissions from a long-term observation at a single site of Fukue.

Figure 7a and the supplementary figure (Figure S6 after revision) will be revised accordingly.

Minor comments:

1) Line 3 on Page 4. “minimize the gaps related to failure of individual instruments”
Please clarify how many data points are missing from individual instruments. Are the BC trend estimates affected by the missing observation data?

We will mention that the gap periods were 9673 and 10974 h (11 and 12 %) for COSMOS and MAAP, respectively (Page 3, Lines 4-5). The impact of the gaps on the overall trend was negligible.

2) Line 7 on Page 4. Please clarify why the uncertainty of the BC observations is estimated at 12%.

As mentioned earlier the statement on the revised estimation of the observational uncertainties will be revised as follows:
" From the average and a standard deviation of the monthly MAAP/COSMOS ratios, systematic and random uncertainties were estimated as ± 14% and ± 17%, respectively (± 22% in total)." (Page 4, Lines 8-9)

3) Line 3 on Page 5. “representation of wet deposition in the model was not important in this study” I do not think so, because air masses without significant influence from wet deposition can still cause <10% of BC loss (Line 24 on Page 4).

The effect of the wet deposition on the used data set with the adopted low-APT criteria (<1 mm) for this study will be clarified, using the ΔBC/ΔCO ratio (Kanaya et al., 2016), for both observations and model simulations:

" The median ΔBC/ΔCO ratio for the observational data with APT < 1 mm (N = 26423) was only 1.9 % lower than that for data with APT = 0 mm (N = 18907), suggesting insignificant influence." (Page 4, Lines 33- Page 5, Line 2)

" Wet deposition was represented with the cloud_acm_ae5 module. Similarly to the observational data, the modeled ΔBC/ΔCO ratio decreased with APT; the modeled median ΔBC/ΔCO ratio for data with APT < 1 mm (N = 26737) was 3.7 % lower than that for data with APT = 0 mm (N = 19197). The removal in the model appeared stronger than the observational trend (1.9 %) but the error introduced due to the wet deposition representation was estimated to be small (−2 %) when using the adopted criteria (APT < 1 mm)." (Page 5, Lines 14-18)

4) Line 9 on Page 5. "Hourly outputs at the nearest grid were used for analysis" The nearest grid is the grid cell where the Fukue Island is located, right?

Yes. We will state that hourly outputs at a grid including the Fukue site were used for analysis. (Page 5, Lines 29-30)

We thank the reviewer again for the productive comments.

References

Kanaya, Y., Pan, X., Miyakawa, T., Komazaki, Y., Taketani, F., Uno, I., and Kondo, Y.: Long-term observations of black carbon mass concentrations at Fukue Island, western Japan, during 2009–2015: constraining wet removal rates and emission strengths from East Asia, Atmos. Chem. Phys., 16, 10689–10705, https://doi.org/10.5194/acp-16-10689-2016, 2016.