Interactive comment on "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" *by* Yugo Kanaya et al.

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Response to the Reviewer #2:

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. Detailed responses to the comments are given below.

Comment 1) Wet deposition of BC includes both in-cloud and below-cloud scavenging. In-cloud scavenging is where BC gets into cloud droplets and below-cloud scavenging is the washout of BC by falling rain droplets. While the occurrence of the latter is always associated with precipitation, the former may not because not all clouds precipitate.

Both scavenging processes are expected to happen during the relative long course of transport BC from its source regions to the Fukue Island sampling site. The authors use precipitation data as a proxy of BC wet deposition, thus neglecting the role of incloud scavenging which may be potentially important. This bias needs to be corrected.

Answer:

The in-cloud scavenging that occurs without precipitation was partly discussed in the previous manuscript. First part was in section 3.2.1.2, where we discussed that critical selection of data regarding RH, BC mass concentration, and traveling time increased the $\Delta BC/\Delta CO$ in the cases without precipitation, and the second part was with Figure 9, where we showed the overall tendency of all $\Delta BC/\Delta CO$ ratios (i.e., irrespective of zero and non-zero APT) against maximum RH. Upon comment by the reviewer, we strengthened the analysis by (1) studying dependence on maximum RH by selecting data without precipitation, and (2) by a sensitivity test in which we re-evaluated wet removal rates after correcting for the possible in-cloud removal that was not associated with precipitation. For (1), we found that when data with zero APT were only used (red triangles of Fig. 1 below, to be used as Fig. 9 in the revised manuscript), the overall decreasing trend became weak; from the difference between the highest and lowest 3 bins, potential loss of BC by cloud processes without precipitation was estimated to be 16% at maximum. The estimation is consistent with the 13–25% increase that occurred with the $(\Delta BC/\Delta CO)_{APT=0}$ by the critical choice of BC concentrations, maximum RH, and traveling time, as discussed in Sect. 3.2.1.2. These analyses suggest that BC loss is mainly associated with precipitation, and is less influenced by clouds. For (2), when 16% upshift in the $(\Delta BC/\Delta CO)_{APT=0}$ was considered to correct for the potential BC loss due to in-cloud scavenging without precipitation, a stretched exponential fitting yielded A1 and A2 values of 0.249 and 0.450, respectively, and the APT values to reduce TE to 1/2 and 1/e became 9.8 ± 1.8 mm and 22.4 ± 5.1 mm. As the uncertainty was large, we will mention this analysis as a sensitivity study in the revised manuscript. The APT values to reduce TE to 1/2 was more sensitive (changed

from 15.0 to 9.8 mm) than that to reduce TE to 1/e (changed from 25.5 to 22.4 mm). Therefore the latter value (25.5 mm), considered to be more robust, will be mentioned in Abstract and conclusion, instead of the former value (15.0 mm).

Comment 2) Fig 1 and 3 (source regions of BC sampled at Fukue): It is not clear to me how the different regions are determined. I doubt that observations from a single site are able to offer unambitious information of such refined source regions. The back trajectories (Fig 3) appear to overlap between different clusters. The authors need to demonstrate BC data at Fukue can distinguish those source regions; if not some of the regions should be lumped together. In addition, the authors seem to claim that the BC observations at Fukue can even distinguish emissions between coasts and in-land of the continent (pg 10, line 14-15), which is not plausible given the distance of this site from the continent and large spatial extension of the back trajectories.

Answer:

We agree with the reviewer that fully clear separation was not possible. However, we demonstrate here that reasonable separation is possible. Regarding the important areas II and III, mainly discussed in the manuscript, as the air mass types were defined by the segmented borderlines that the trajectories crossed for the first time, no overlap was found in the regions near the coast. On the other hand, the overlap becomes somewhat significant for the inland areas. When weighted by the geographical distribution of the BC emission strength, however, we found the effect from other regions was estimated to be <25% in total. Therefore main information (>75%) is still from the defined region. Regarding area V, influence from other regions was mentioned and thus area V' was defined in the previous manuscript. We did not intend to "distinguish" emissions between coastal and in-land areas. There we meant that the signal "weight" from the coastal area was larger than from in-land, because of the shorter distance to the coastal area from the observational site.

Comment 3) Section 3.3 and Figure 12, trend of BC masses by source region: BC data

with nonzero APT should not be used to constrain emissions. Wet scavenging is efficient to remove all BC during transport and thus non-zero APT air masses sampled at Fukue have lost all the source signatures of BC and contain only background. Although the authors apply an empirical correction to infer the BC loss due to precipitation, that correction is obviously subject to high uncertainty which is difficult to constrain. The authors should use only a subset of observations which are determined to have minimal influence of wet deposition when constraining emissions.

Answer:

We reexamined the trend of data with zero APT and found that it was associated with large uncertainty. Therefore we remove this part and Figure 12 from the revised manuscript.

We again thank the reviewer for the important suggestions.
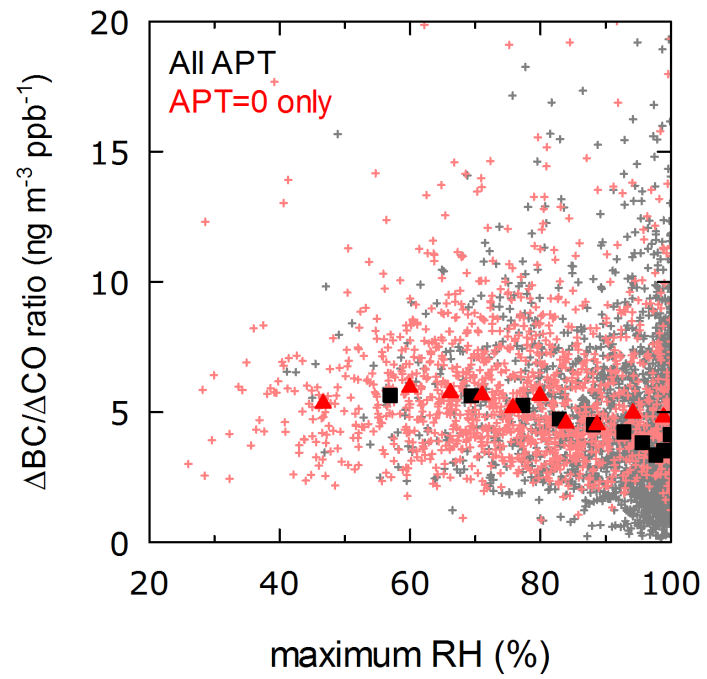
Fig. 1. Observed ΔBC/ΔCO ratios as function of maximum RH (%) that air mass experienced in 72 hours before arrival at Fukue Island. All data (gray/black) and datawith APT=0 (red) are shown separately.