Comments on "**Identifying Better Indicators of Aerosol Wet Scavenging During Long-Range Transport**" by Hilario et al.

General

This paper describes the results from the data analyses of the wet scavenging of black carbon (BC) aerosols. The authors attempted to seek possible good indicators to describe the wet removal of BC during transport. In previous studies, the precipitation amount accumulated along backward trajectories (APT) has been analyzed as one of the indicators. Beside this, the authors suggested that the other several indicators related to the precipitation and humidity along the backward trajectories can well account for the variation of the degree of the removal of BC which is defined as the enhancement of BC to CO (ΔBC/ΔCO ratio). The major results and discussion in this study meet the scope of Atmospheric Measurement Technology. Despite the significance of this study, there are several important issues to be addressed before accepting the manuscript. Please consider the following comments and necessary revisions of the data analyses and the descriptions in the manuscript.

Major comments

1. Calculations of the enhancement ratio of BC to CO (ΔBC/ΔCO)

The serious mistake is the choice of the enhancement ratio "ΔBC/ΔCO" for the quantitative investigations of wet scavenging of BC. This is because (1) the degree of the removal depends on the emission ratio of BC to CO ($ER_{BC2CO} \equiv \Delta BC/\Delta CO$ at the emission), (2) the background levels of BC and CO can vary with the air mass origins, and (3) ΔBC/ΔCO can vary with the air mass mixing during the long-range transport. As to (1), authors also stated this point in section 4 (Limitations). Authors can take care of this point by analyzing the ER_{BC2CO} from the observation data sets. In many previous studies using APT, this kind of adequate data preparations were conducted. For this purpose, the observed air masses need to be separated according to the air mass origins and/or emission sources as shown in Hilario et al. (2021), and then the variability of ER_{BC2CO} (not ΔBC/ΔCO–APT relationship) during the CAMP^{2}Ex campaign needed to be analyzed. The authors must justify the important and critical assumption that the variability is enough small among the air mass origins and/or emission sources to use ΔBC/ΔCO as a unified indicator for the removal of BC. Separation of air mass types can lead to decrease the number of data to be analyzed, as discussed in section 4. However, if the authors do not show that the observation-based ER_{BC2CO} did not largely vary among the different air mass origins/emission sources in the study region, this is not a factor of the methodological limitations but it is just one of large error sources in the data analyses and the following interpretations. Please justify this assumption. If it were not for the validity, the data sets could not be suitable for the validation of authors' proposed method. As to (2), authors determined the background levels of BC and CO by analyzing the potential temperature profiles based on some previous studies. Currently, only the seasonal transition of background levels is considered by separating the periods to be analyzed. Are there any possibilities that the background levels vary depending on the air mass origins? As to (3), this effect depends on the time scale of the transport. The current manuscript is lacking in this information. Also regarding (1) and (2), it is needed to describe and discuss the observed feature of BC in the CAMP^{2}Ex campaign such as the relationship between the observed enhancements of BC and CO concentrations and backward trajectories (e.g., air mass origins) and typical transport time from the source regions. The former can affect the variabilities of ER_{BC2CO}. The latter can provide the insight into the adequate integration time for calculating the APT. Kanaya et al. (2016) indeed set 3 days for the integration time to calculate the APT by considering typical transport time from the source regions (e.g., central China) to the observation site (remote island in western Japan).

2. The criteria to evaluate the performance of the predictors

In section 2.5, authors stated "We use R because we are more interested in ~". The performance of the combination of the predictors and fitted functions was evaluated by comparing the Pearson correlation coefficients (R) of the correlations to account for the variations of the observed $\Delta BC/\Delta CO$. Therefore, the accuracy of the predication (i.e., slope and WAD) was not weighted in this study, resulting in the inaccurate performance of almost all the predictions that seriously overestimate the observed values of $\Delta BC/\Delta CO$ especially for their low value ranges (positive values of the WAD and intercepts). To me as a potential reader of this paper, this fact suggests that the approach proposed in this study is not always better than the previous works. The APT approach used in the previous studies showed the better performance to predict the $\Delta BC/\Delta CO$ or transport efficiency ($TE_{BC} = (\Delta BC/\Delta CO)/ER_{BC2CO}$) using the long-term averaged data sets (e.g., Kanaya et al., 2016; Choi et al., 2020). Although the correlation of the TE_{BC} and APT was not so good, based on the binned average data sets of TE_{BC}-APT relationship, the decreasing tendency of the TE_{BC} during the transport was successfully predicted by the APT in their studies. In this study, the APT-based prediction skills were not fully described. The predictions with the lower R should be discussed for the fair and comprehensive evaluations of the accuracy of all the predictions tried in this study.

3. Calculating the APT and other indicators

Authors might misunderstand the previous studies to apply the APT in their data analyses. As an example of ground-based studies, Kanaya et al. (2016) defined the length of the total backward time to calculate the trajectories (5 days) and integration time to calculate the APT (3 days) by considering the typical source areas (East Asian continent) affecting the observation site (a remote island in western Japan) and the typical meteorological field. Oshima et al. (2012; 2013) analyzed the aircraft observation data sets of BC and CO for uplifted air parcels sampled at the upper atmosphere (3–6 km), and investigated the effect of upward transport of air masses associated with the precipitation. The APT for uplifted air masses were calculated by integrating the precipitation water content from the uplifted location to the sampling point (Oshima et al., 2012). Depending on the definitions of the APT, the sensitivity of the precipitation to the transport efficiency of BC was significantly different from those from ground-based investigations (e.g., Kanaya et al., 2016). What I would like to claim is that possible indicators for the wet removals of BC should be designed by the careful consideration of the actual atmospheric conditions (i.e., meteorology) and the observation types (e.g., ground vs. aircraft).

In this study, the basic characterizations of how the air parcels sampled at the aircraft observatory were transported from where (i.e., transport pathway), and the atmospheric transport time scale from the possible source area/region of BC and CO are critically missing (Referring Hilario et al. (2021) in section 2.1 is insufficient.). In section 3 "Results and discussion part", author should prepare additional subsection to describe the observed features of BC aerosols during CAMP^{2}Ex campaign to clarify the above points (This was also pointed out in the comment 1). Based on the descriptions about the basic data analyses of the BC and CO enhancements, authors should define the proper length of the backward calculations of the trajectories and integration time for calculating various parameters in relation to the removal of BC.

4. Curve-fitting equations

The authors prepared 4 equations in the data analyses. Two of them (Oshima and Kanaya (stretched exponential)) were derived from the previous studies analyzing the TE_{BC}–APT relationship. What was the basis to apply the remaining two? In this study, four types of parameters other than APT were analyzed, however two types of the equations were applied to these. Are there needs to apply and test more equations to these various indicators? Please clarify the reason why the authors decide to select these two equations for the non-APT parameters.

5. **How should we judge as “Better” when the performance to predict the removal of BC from atmosphere is evaluated?**

In relation to the above comments, I strongly suggest not to use “identifying” and “better” in the title of the current manuscript. At least, the proposed approaches are not better than the previous works using the APT. So, the better indicators were not identified yet. More careful discussion based on more careful data analyses is needed to justify it is “better”. Please consider the significant revisions of the data analyses and descriptions in the manuscript.

Minor comments

P5, L146. Please clarify how authors determined the number of k for k-fold cross validation analyses.

P5, L147. “jug” should be “Jug”. What is the version of Jug used in this study?

P7, L232–L234. “We hypothesize ~” High RH condition is also related to in-cloud condition as well as precipitation as suggested. It is well known that BC can be activated to form cloud droplets as CCN. This results in the removal of BC from atmosphere (into hydrometeor). Needs to be revised accordingly.

P20, Table 2. The curve-fitting equation which produces the highest value of R should be added with each predictor listed in Table 2. This will help us to easily find which equation works well with which indicator. Adding typical values of the coefficients in the fitted curves to the list is highly recommended for the clarity of the shape of the determined curves.

P22, Figure 1b. To clarify the percentile blocks, please consider to modify the figure style of the lines between points to the lines between markers especially for the predicted traces.

P25, Figure 4. Same as the comment to Figure 1b.

References

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