Responses to comments by Referee 1

General comments: This study attempted to investigate the role of cloud on the formation of brown carbon. A comprehensive and valuable dataset was collected, including the light-absorption properties of the cloud droplet residual, the cloud interstitial and cloud-free particles, the light-absorption and fluorescence properties of water-soluble organic carbon in the collected cloud water and PM2.5 samples, and the concentration of water-soluble ions. The presented data further indicate the formation of secondary BrC during cloud processing and a considerable contribution of water-insoluble BrC to total BrC light-absorption. Such results improve our understanding on the optical properties and secondary formation of BrC in cloud, and thus merit publication in ACP. Here are some minor issues that need to be addressed.

Reply: Thanks for the reviewer's positive comments.

Main comments:

Experiment section: why was PM2.5 inlet applied to rule out the cloud interstitial particles? Discussions should be provided on the possible uncertainty that may be introduced.

Reply: Thanks for the comments. It is assumed that the activated particles would grow to cloud droplets with median size at around 10 μm in the present study. This is reasonable since such a size distribution pattern have been previously observed at various regions such as at Mt. Tai (Li et al., 2017). In such case, the size of cloud droplets with size lower than 2.5 μm would be limited, and thus particles with size lower than 2.5 μm in cloud are regarded as cloud interstitial particles. We note that possible uncertainty could be introduced due to such an approximation. It would not lead to ambiguous conclusions since cloud residual particles were mainly focused.

The limitation has been added in the revised manuscript as "*It should be noted that the $PM_{2.5}$ inlet may introduce possible uncertainty for the collection of cloud interstitial particles due to the interference of cloud droplets, although the size distribution of cloud droplets were mainly concentrated on 6-9 μm at mountain sites (Li et al., 2017). However, it would not be the case when cloud residual particles were mainly focused in the present study.*"

References

Li, J., Wang, X., Chen, J., Zhu, C., Li, W., Li, C., Liu, L., Xu, C., Wen, L., Xue, L., Wang, W., Ding, A. and Herrmann, H.: Chemical composition and droplet size distribution of cloud at the summit of Mount Tai, China, Atmos. Chem. Phys., 17(16), 9885–9896, doi:10.5194/acp-17-9885-2017, 2017.

"The contribution of water-insoluble BrC to the light-absorption is estimated to be ~75% for the cloud INT particles and ~48% for the cloud RES particles on average, based on these differences (Fig. 3)." It is interesting to know that water-insoluble BrC contributes to such a high fraction of BrC in the cloud INT particles and the cloud RES particles. I wonder if some of this insoluble fraction is secondary origin.

Reply: Thanks for the comment. We further analyzed the correlation between the light-absorption of water-insoluble organic carbon ($Abs_{370,WIOC}$, as the difference of $Abs_{370,\ total\ BrC}$ and $Abs_{370,WSOC}$) with SNA concentration. There is a positive correlation for the cloud INT particles ($r = 0.80$, $p < 0.01$) but no correlation for the cloud RES particles ($r = -0.15$, $p = 0.38$), as shown in the figure below. This result might indicate possible secondary origin for the water-insoluble BrC in the cloud INT particles, yet there is no evidence for that in the cloud RES particles, which needs further investigations.

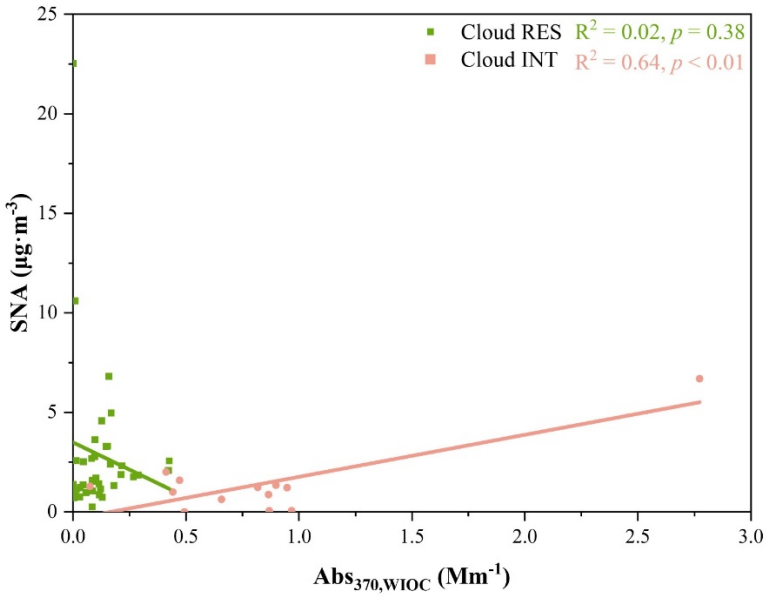

Figure 1 Correlation between $Abs_{370,WIOC}$ with SNA concentration in cloud RES and cloud INT particles

Lines 197: The authors presented correlation analysis between the Abs365 of cloud water and PM2.5 aqueous extract with SNA (sulfate, nitrate, and ammonium) ($r > 0.77$, $p < 0.01$), and NOx ($r > 0.58$, $p < 0.01$), and the result supports the secondary formation of BrC. Why was PM2.5 aqueous extract included in the analysis? Does this result also indicate the significance of secondary production of BrC in PM2.5?

Reply: Thanks for the comments. Correlation analysis between the Abs_{365} of cloud water and $PM_{2.5}$ is compared to show if there are differences between cloud water and $PM_{2.5}$. The results also indicate possible secondary origin of BrC in the $PM_{2.5}$, which is similar to that observed for the cloud water. However, due to the limited size of samples (13 for INT-$PM_{2.5}$ and 19 for FREE-$PM_{2.5}$), the PMF method may introduce large uncertainty and thus cannot be used to estimate the secondary fraction of BrC.

Minor comments:

Line 53 what does "These light-absorption species" refer to?

Reply: The "*These light-absorption species*" refer to the products of the reaction that has been proved to produce secondary BrC in the laboratory. For more accurate expression, this sentence has been rewritten as "*The secondary BrC such as nitrophenols, aromatic carbonyls, imidazole, and organosulfates have also been detected in cloud/fog water*".

Line 134 "(SUVA, m2·g-1,)" error typo.

Reply: Thanks for the comment. The typo has been corrected to "*(SUVA, $m^2 \cdot g^{-1}$)*" in the revised manuscript

Line 156 "As expected, there is a positive correlation between Abs365 and WSOC concentration in cloud water and PM2.5 aqueous extracts ($r > 0.61$, $p < 0.01$)." Does it mean that WSOC in cloud water is mostly from PM2.5?

Reply: We are sorry for the misleading. Actually, this sentence means that Abs_{365} poses a positive correlation with WSOC concentration both in the cloud water or $PM_{2.5}$ aqueous extracts. For accurate expression, this sentence has been revised to "*As expected, there is a positive correlation between Abs_{365} and WSOC concentration in both cloud water and $PM_{2.5}$ aqueous extracts ($r > 0.61$, $p < 0.01$)*" in the revised manuscript.

Line 160 "much lower than those in urban areas (as summarized in Table S1)". I suggest to include the observed values.

Reply: Thanks for the reviewer's kind suggestion. The observed values have been added in the revised manuscript: "*much lower than those in urban areas (3.4-33.9 Mm^{-1}, as summarized in Table S1)*".

Line 197 what does "wet particles" refer to?

Reply: The "*wet particles*" refer to the cloud-free and cloud-INT particles. For better expression, the word "*wet particles*" has been replaced by "*cloud-free and cloud-INT particles*" in the revised manuscript.

Line 208 revise "Consistently, the source and contribution apportionment of BrC" to "the source apportionment of BrC".

Reply: Thanks for the reviewer's kind suggestion. We have replaced the sentence "*Consistently, the source and contribution apportionment of BrC*" with "*The source apportionment of BrC*" in the revised manuscript.