

Referee report on ACP-2022-483 - Concurrent photochemical whitening and darkening of ambient brown carbon

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The manuscript by Li et al. titled “Concurrent photochemical whitening and darkening of brown carbon” describes the behavior of primary and secondary brown carbon (BrC) from field observation in a sub-urban site near Beijing, China. The total aerosol absorption is apportioned between black carbon (BC), primary BrC and secondary BrC. Traffic and biomass burning are identified as main sources of primary BrC and nitrogen-containing oxygenated organic aerosol are identified as the main source of secondary BrC. A reduction is observed in primary BrC absorbance/total absorbance during the day time with a simultaneous increase in secondary BrC absorbance/total absorbance. Their main finding that there is field evidence of concurrent whitening and darkening of BrC due to photochemical processes, is primarily based on this observed diurnal variation.

While there is some improvement from the previous version with the inclusion of other possible explanations for the observations and in acknowledging the uncertainties in the used method, it is questionable whether the authors have provided sufficient evidence that justify their main findings are indeed evidence of whitening and darkening of BrC, rather than formation and degradation of BrC.

Main Comments

1. In Figure 4b), authors have shown the reduction in absorption coefficient of primary BrC/POA (i.e. the MAC of POA) as evidence for the whitening of primary BrC. While there are many uncertainties and other possibilities for this, it is offering some level of evidence for the “whitening” (a better term may be degradation) of primary BrC. However, the same is not shown for secondary BrC. The term “darkening” indicates that the formed secondary BrC increased their absorbance. The increase in secondary BrC abs/total abs only indicates that the absorbance from secondary BrC increased during this time. Due to the very obvious formation of SOA from photochemical processes will indeed result in this. However, it does not indicate that there was a “darkening” process. What does the diurnal variation of MAC of SOA look like? Are you able to see an increase in MACSOA? As OA factors from PMS and BrC absorbance (and primary and secondary BrC absorbance) is available, is it possible to investigate the MAC of the individual OA factor? Do their diurnal variations offer additional information of the “whitening and darkening” effect.
2. Lines 269-271-While the overall AAE of total absorbance may be highly influenced by BC absorbance, the AAE of total BrC, primary BrC and secondary BrC can be individually calculated as the total absorbance has already been apportioned.
3. Line 291-293 – The meaning is very unclear
4. Line 22 and line 316- “fraction of total absorbance of secondary BrC” is unclear. Do you mean fraction of absorbance of secondary BrC to total absorbance?
5. Lines 327-336 - As the primary focus of the paper is the “whitening and darkening” of BrC, a more detailed analysis with further evidence is required to make a strong conclusion on this. The 4. Conclusions section does not even include this main point and does not offer any atmospheric implication of the major observations from this manuscript.