

***Interactive comment on* “Significant seasonal changes in optical properties of brown carbon in the mid-latitude atmosphere” by Heejun Han et al.**

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Reviewer #2

Han and coauthors describe off-line analysis of filter samples collected over four seasons in Seoul, Korea in order to investigate relationships between brown carbon (BrC) and water-soluble organic carbon (WSOC) and how they change throughout the year. Potential sources are also discussed using complementary off-line techniques including high performance liquid chromatography (HPLC), excitation-emission matrix (EEM) analysis and isotope ratio mass spectrometry (IRMS). The seasonal behaviour of the relationship between humic-like substances (HULIS) and WSOC is interesting, with a pronounced increase in the HULIS/WSOC ratio in the winter months. The authors

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propose that this pattern is due to more effective reduction of HULIS mas through photodegradation during atmospheric transport in the summertime when photolysis rates and photochemical activity are higher. The absence of a seasonal pattern for potassium, in particular adds weight to the argument that the HULIS/WSOC ratio seasonality cannot be explained only by changes in biomass burning source contributions throughout the year. Although levoglucosan exhibits higher wintertime values, this tracer can also be degraded through photochemical processing. Overall I find the manuscript to be well written and structured and only have some minor comments:

=> Thank you for your review and valuable comments. All your comments were addressed in the revised manuscript.

In the introduction it is mentioned that “the origin of BrC is often attributed predominantly to HULIS”. It would be helpful to discuss known sources of HULIS here.

=> We added more about the sources of HULIS in the introduction section in the revised version.

Through EEM analysis, the extracted HULIS component agreed temporally with the extracted HULIS concentration as shown in S4. How do the other extracted EEM factors agree temporally with HULIS concentration? If the agreement is strong between all three factors, then it is likely that they are associated with the same source. If C2 is characterized by tryptophan then is vegetative debris a likely source for this factor? Are C1 and C3 interpreted to have the same source but differ only in their EEM characteristics? This section (3.1) could be clearer.

=> Since the other EEM factors are highly influenced by one dominant fluorophore HULIS (C1) as shown in the EEM spectra (Fig. S7 in the text; Fig. 1 below), we cannot reliably characterize C2 component using PARAFAC model. Thus, we only use C1 component in this study. This is mentioned in the text (lines 154–159).

Either in the Introduction or Discussion it would be helpful to discuss the findings here

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in the context of previous on-line measurement studies that have observed similar photodegradation of brown carbon from biomass burning sources during atmospheric summer time transport, for example (Forrister et al., 2015; Selimovic et al., 2019; Healy et al., 2019).

=> Yes, this is mentioned in the introduction section in the revised version.

Define “RU”

=> The detailed definition about “RU” is added in the revised manuscript.

Line 186: What is the likely source of the non-crustal V- oil combustion, shipping? It would be useful to discuss

=> Since vanadium is mostly removed during the refining processes, the use of raw materials such as crude oil and coal can be the source of non-crustal vanadium. This is briefly mentioned in the revised manuscript.

Line 195: Define C3 plant-origin materials

=> We briefly mentioned about C3 plant-origin materials in the revised manuscript.

Line 209-210: The last line of this paragraph is unclear and should be rephrased.

=> Yes, we rephrased this more clearly in the revised version.

Line 244: Remove “followed”

=> removed in the revised version.

[END]

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2019-952>, 2019.

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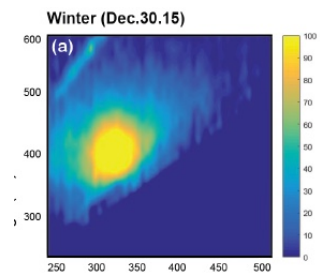


Fig. 1. Fig. S7