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Atmospheric Chemistry and Physics Discussions

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 #1

General comments:

This paper presents the results of stationary measurements of biomass burning aerosol in South Korea over the course of multiple seasons. The authors identify biomass burning as a major contributor to light-absorbing components of the aerosol, and assert that UV-radiation can explain the lower absorptivity of summer vs winter samples, with irradiation of winter samples resulting in properties similar to observed summer samples. The paper is generally well-written and clear, and the figures are appropriate for the claims being made. In some cases, the language used is too conclusive, and some

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places need clarifying, as requested below.

=> Thank you for your valuable comment. All your comments were carefully taken into account in the revised version.

Specific comments:

Abstract line 10: seasonal changes in sources and sinks of HULIS and WSOC (what was directly measured rather than approximated). The next sentence then relates the HULIS to total BrC.

=> We measured both HULIS (water-soluble BrC) and MeOH-soluble BrC for all samples (Fig. 2b and Fig. S4b). For the UV radiation experiments, we only measured HULIS as an index of BrC. Thus, we will keep the wording unchanged.

Abstract line 17: this was supported by laboratory UV radiation experiments (confirm is too strong of language)

=> We changed the word "confirmed" to "supported."

Lines 23–24: Black carbon aerosol should not be included in the light absorbing organic aerosol category, as black carbon is primarily soot (inorganic carbon)

=> We changed “light-absorbing organic aerosols” to “light-absorbing carbonaceous aerosols.”

Lines 92–93: Since the K fraction and WSON are determined using parameterizations instead of direct measurements, “estimated” is a more accurate term than “calculated.” For the K/Al_crust parameterization, please provide the value used in addition to the reference from where it came.

=> We changed the word “calculated” to “estimated.” We also provide the value used for the parameterization, in addition to the reference in the revised version.

Line 95: The sea spray fraction is calculated assuming a certain ratio of Na to Cl,

(1.4486:1) – how does this ratio compare to the measured Cl/Na ratio? Since the Cl and Na is assumed to come entirely from seawater, this assumption can be verified.

=> Here, 1.4486 is the ratio of the concentration of all elements except Cl to the Na concentration in seawater (Maenhaut et al., 2007). The mass ratio of Cl/Na is 1.79 for seawater. The measured Cl/Na mass ratio ranged from 0.01 to 1.78. Since the maximum sea-salt concentration in our samples was about 0.1% of seawater, the highest value (1.78) verifies that our assumption is valid. Since this is commonly referred to and sea spray fraction is negligible in this study, we do not explain this in the text.

Line 98: Similarly, provide the crustal V/A; parameterization used, and replace "calculated" with "estimated."

=> We provided the value and changed the word "calculated" to "estimated."

Line 201: While Pearson's r-squared value of 0.5 does represent a decent correlation in atmospheric data, by definition this means that 50% of the variation in HULIS is attributable to levoglucosan, not a majority, so the phrase "was the major source" might be slightly misleading.

=> We changed the word "major" to "significant."

Line 228: Similarly, although the negative correlation with r-squared of 0.5 indicates a strong relationship between UV radiation and HULIS, it only explains half of the variance in HULIS, and the system is more complex than it sounds from the current language.

=> Yes, we agree. Thus, we just state similar trends for UV and HULIS. Then, we state that "these results are in agreement with the following laboratory results" instead of linking UV and HULIS at this stage.

Line 242: the phrase "in the winter samples with high HULIS concentrations" leads one to conclude that there were winter samples with low HULIS as well as high HULIS. If, as I assume, the authors simply want to reinforce the idea that winter had higher

HULIS, then perhaps this would be clearer, “in the winter samples (characterized by higher average HULIS concentrations), while no significant changes occurred in the summer samples (characterized by low average HULIS concentrations)”

=> changed as suggested in the revised version.

Line 253–254: Please rephrase this sentence so that it is easier to understand. I don’t quite understand what the authors are saying here.

=> We argue that the trend of “light-absorbing property” is similar to that of “fluorescence property.” We clarified this sentence in the revised version.

Line 264: Given this interesting result (that irradiated winter samples look like summer samples” I wonder if there is more to be done with predicting summer properties from winter observations. It seems harder to back-calculate the properties of the aerosol emitted in summer (when still fresh), but I assume it would be similar to the winter samples. This interesting point deserves more attention, and I wonder if any previous studies have come to this conclusion, or a similar conclusion.

=> Thank you for your insightful comment. Although we cannot back-calculate the summer properties based on unknown UV radiation intensity, exposure time, and other atmospheric conditions, we assume a similar initial property based on other tracers including non-crustal K, air mass back trajectories (Fig. S1), and light absorption property in this study.

Line 271: The use of word “confirmed” here to describe how the laboratory studies can explain the observed differences in fluorescence is too strong. The studies certainly support the hypothesis, and they go a long way toward suggesting the mechanism, but given that other processes in the atmosphere (oxidative, or possibly aqueous aging) could also result in the same bleaching, the language should be softened until there are many further studies, or until signatures of bleaching can be tied specifically to the mechanism studied here.

=> We changed the word “confirmed” to “supported.”

Technical correction:

Line 21: “and THE global climate system”

Line 28: remove plural on “light absorbing aerosols”

Line 40–41: remove space between number and percent symbol in all instances

Line 153: “originates”

Line 204: omit “the” before “BrC”

Line 244: there must be an extra word in this sentence “exhibited followed corresponding”

Line 248: remove plural on “carbon contents”

Line 251: omit “a” before “photo-labile”

Line 267: omit “the” before “different seasons”

=> We made all above technical corrections as suggested.

[END]

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