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Significant seasonal changes in optical properties of brown carbon in the midlatitude atmosphere

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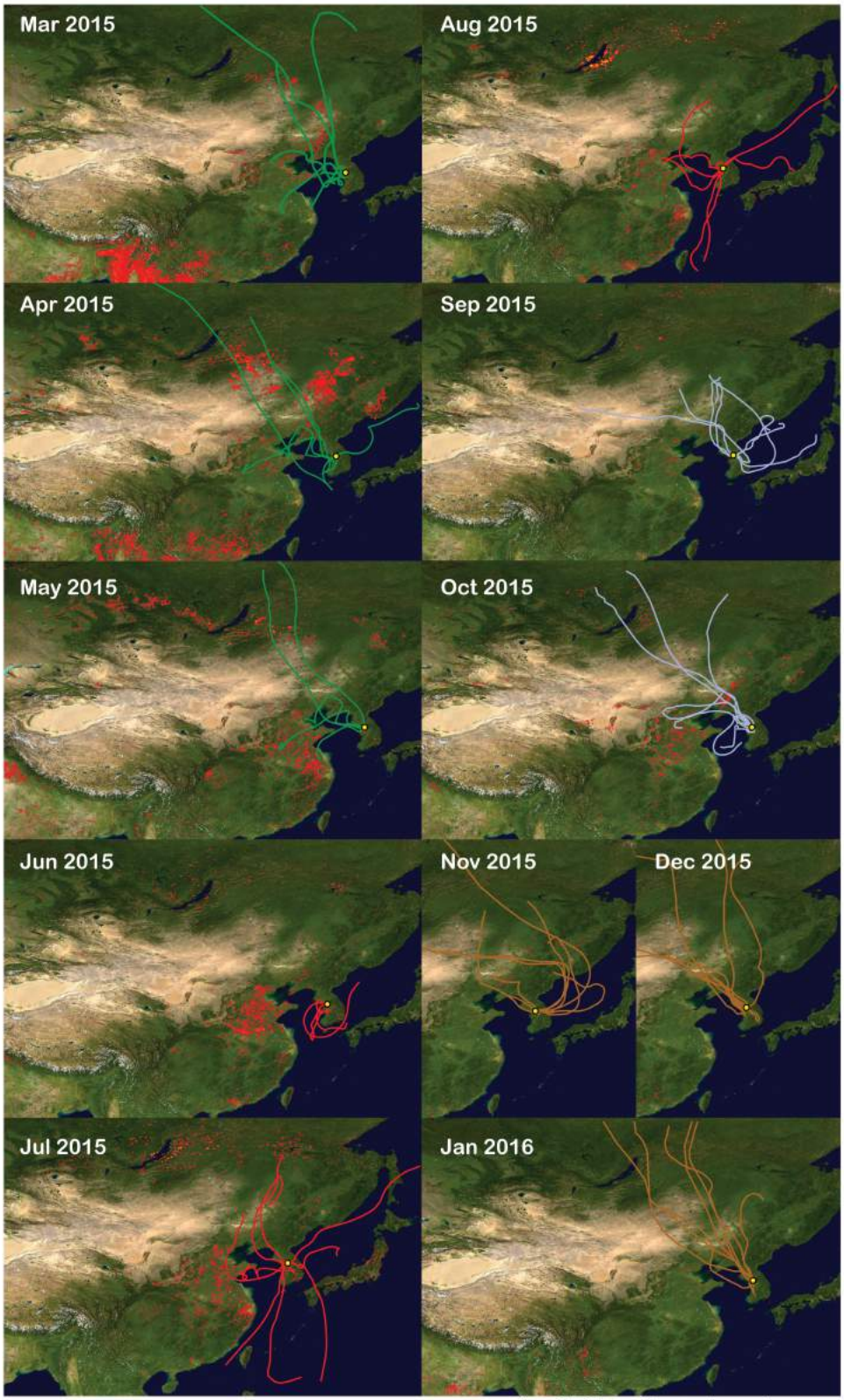

Figure S1: Fire maps obtained by using the moderate resolution imaging spectroradiometer (MODIS) fire location data provided by NASA's fire information for resource management system (FIRMS; https://firms.modaps.eosdis.nasa.gov) combined with the air mass back trajectories around the study site from March 2015 to January 2016.

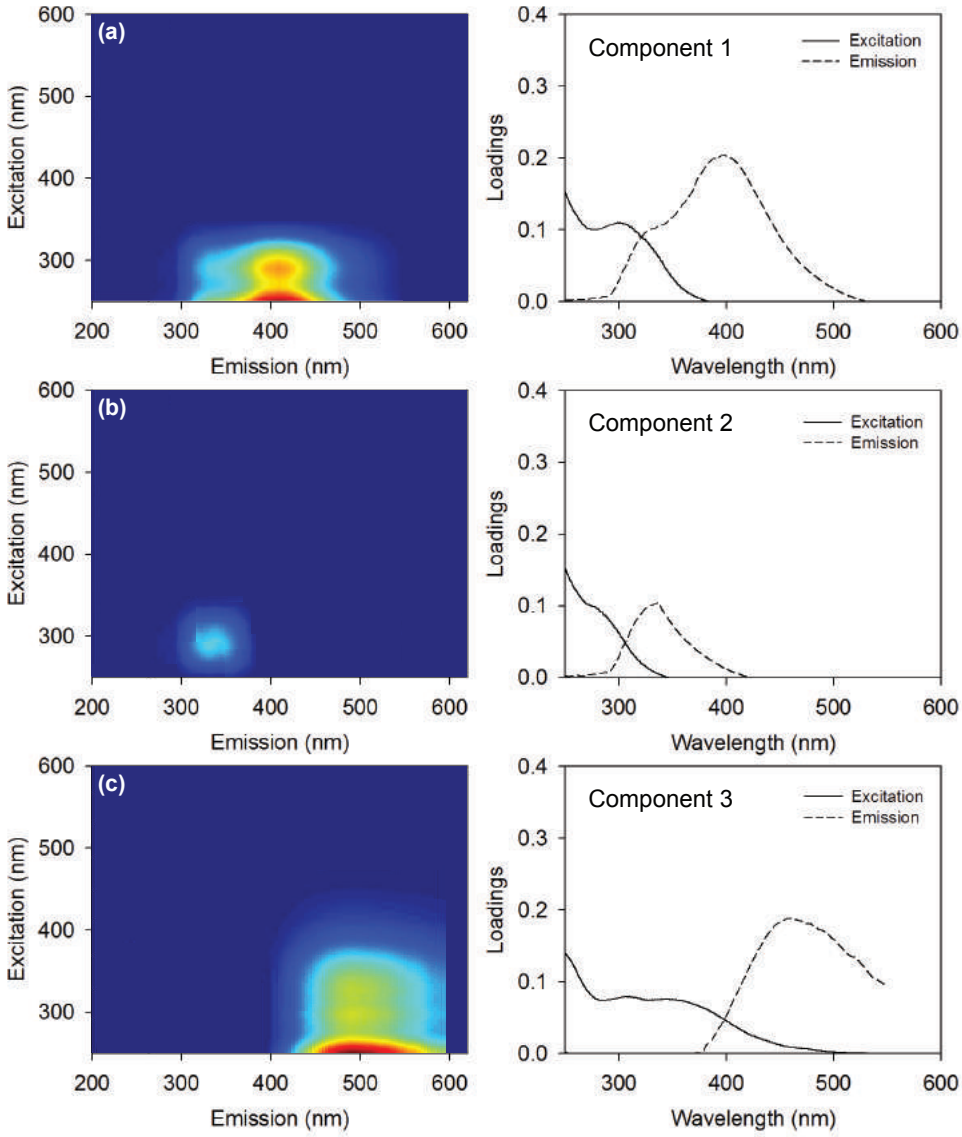

Figure S2: Contour plots of fluorescence EEM spectra and excitation-emission loadings of (a) humic-like (C1), (b) protein-like (tryptophan) (C2), and (c) humic-like (C3) components identified from the HULIS samples collected from March 2015 to January 2016 in Seoul.

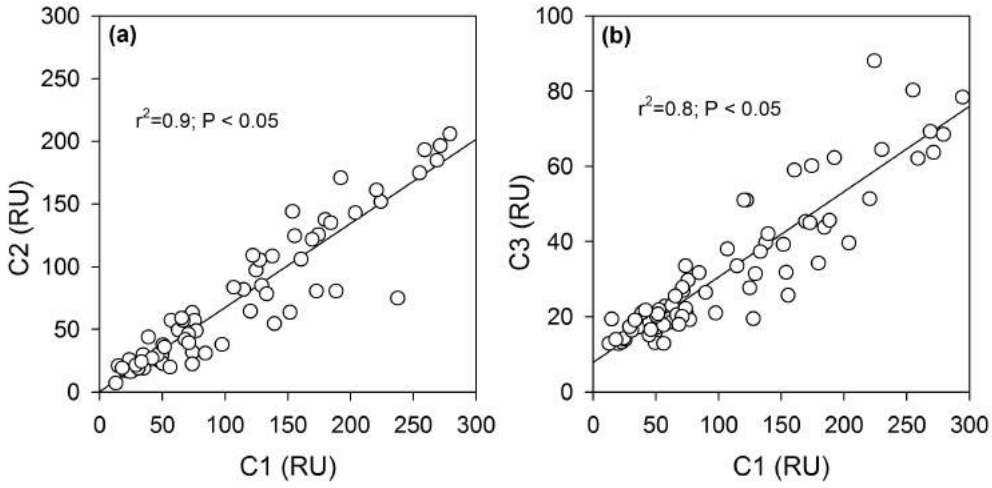

Figure S3: Plots of the fluorescence intensity of (a) C1 (HULIS) versus C2 and (b) C1 versus C3.

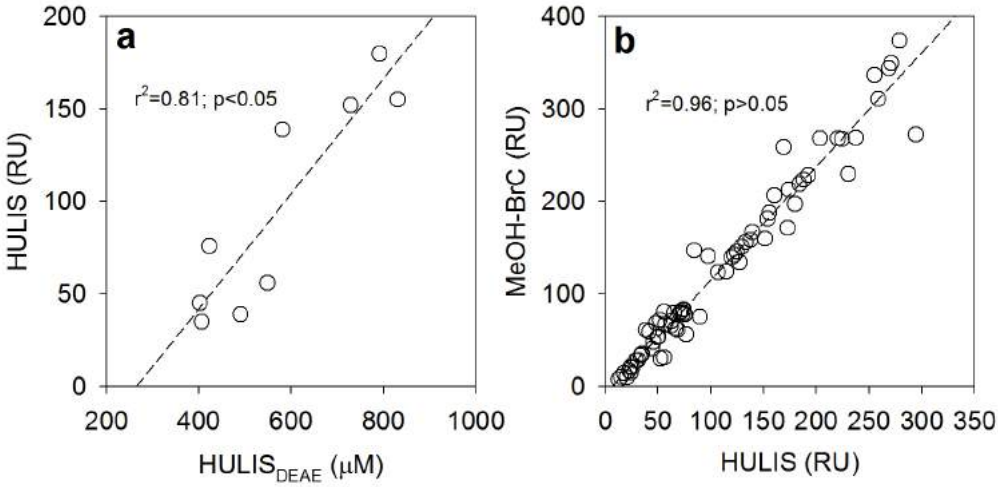

Figure S4: Plots of the (a) fluorescence intensity of HULIS versus DEAE column extracted HULIS concentration and the (b) fluorescence intensity of HULIS versus MeOH-BrC.

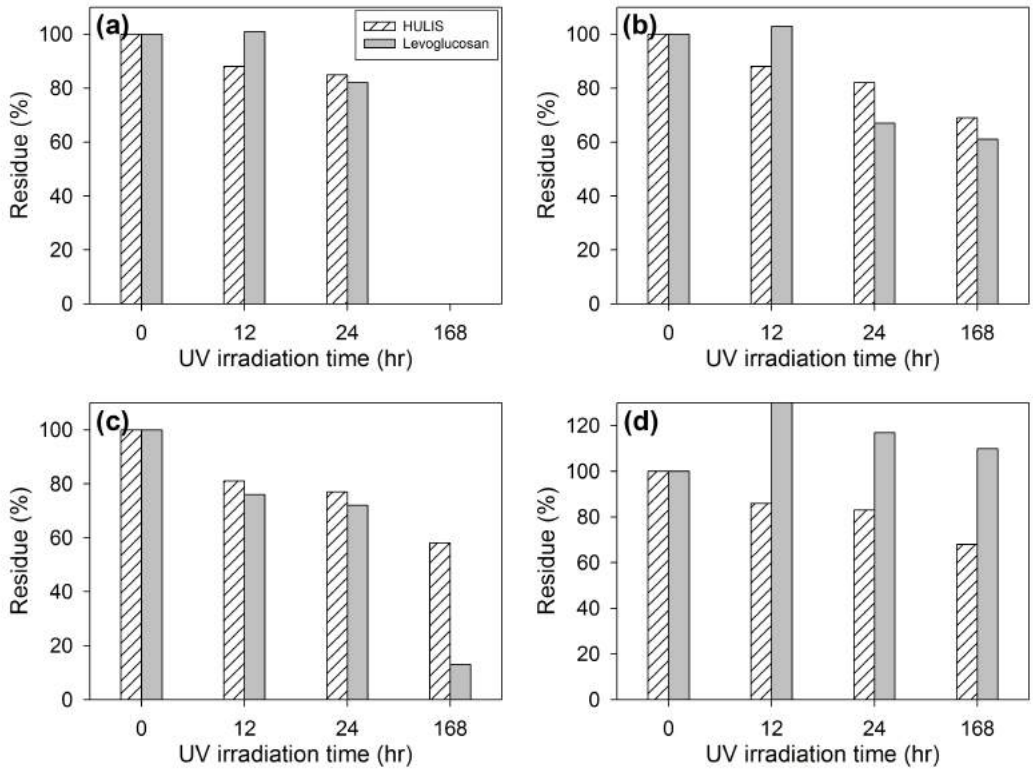

Figure S5: The changes of HULIS and levoglucosan concentrations for four different samples during the UV radiation (7-day) experiment.

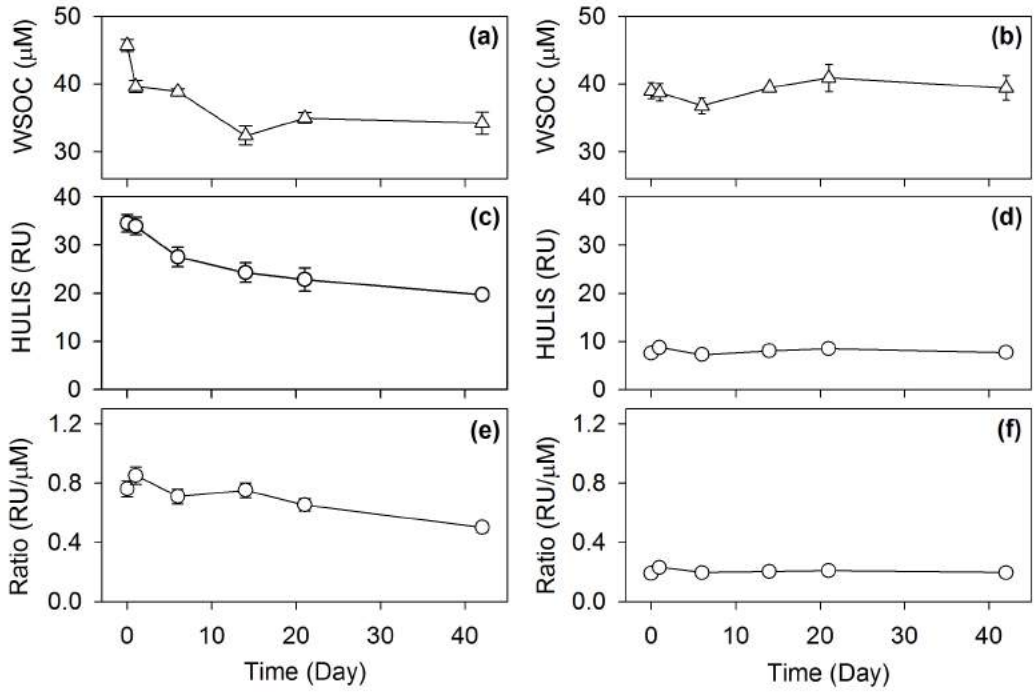

Figure S6: The WSOC concentration of the (a) winter and the (b) summer aerosol samples, fluorescence intensity of HULIS of the (c) winter and the (d) summer aerosol samples, and ratio of fluorescence intensity to WSOC concentration of the (e) winter and the (f) summer aerosol samples during laboratory experiment. Error bars represent the standard deviation for each parameter.

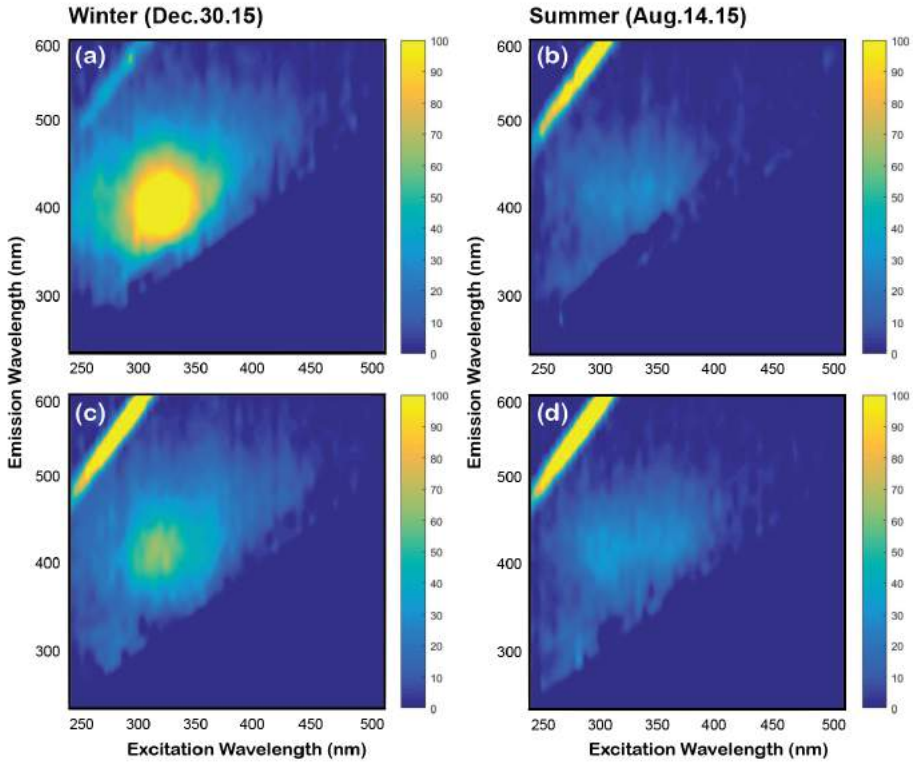

Figure S7: Contour plots of fluorescence EEM spectra of non-irradiated initial samples collected in the (a) winter (Dec/30/15) and (b) summer (Aug/14/15), and UV irradiated (6 weeks) samples collected in the (c) winter (Dec/30/15) and (d) summer (Aug/14/15).

Table 1: Spectral characteristics of the fluorescence components identified by PARAFAC model for the atmospheric HULIS.

Component	Ex/Em wavelengths [nm]	Peak	Description and origin
C1	305/416	M	Terrestrial humic-like component (Stedmon and Markager, 2005); HULIS (Pöhlker et al., 2012)
C2	290/340	T	Protein-like (tryptophan-like) component derived from autochthonous processes (Stedmon and Markager, 2005; Pöhlker et al., 2012)
C3	365/484	C	Terrestrial humic-like component (Stedmon and Markager, 2005)

References

Stedmon, C. A. and Markager, S.: Resolving the variability in dissolved organic matter fluorescence in a temperate estuary and its catchment using PARAFAC analysis, Limnol. Oceanogr., 50, 686–697, 2005.

Pöhlker, C., Huffman, J. A., Pöschl, U.: Autofluorescence of atmospheric bioaerosols – fluorescent biomolecules and potential interferences, Atmos. Meas. Tech., 5, 37–71, 2012.