Thank you for your review and comments concerning our manuscript entitled "Significant seasonal change in optical properties by atmospheric humic-like substances (HULIS) in water-soluble organic carbon aerosols". We have concerned all your comments carefully, and our specific actions against each comment are as below.

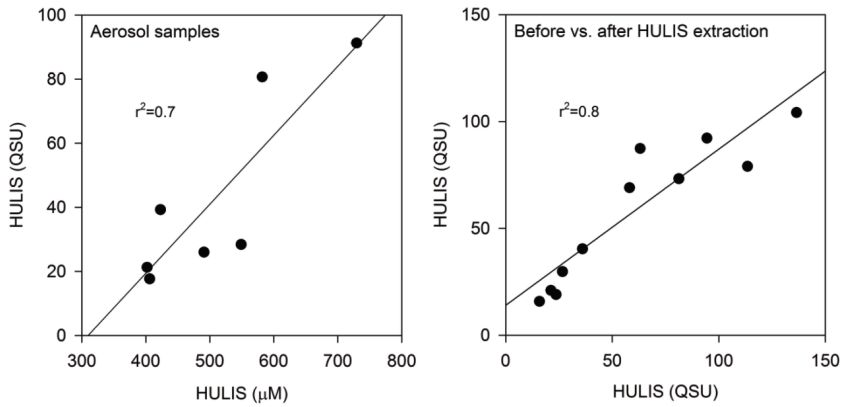
(1) The optical properties of WSOC in urban aerosols have been intensively studied in previous work (Baduel and Jaffrezo, 2010; Hecobian et al., 2010; Mladnov et al., 2011a). The other related studies cited in this work (Mladenov et al., 2011b; Fu et al., 2015; Xie et al., 2016) investigated the sources and optical properties of WSOC in alpine and artic areas, respectively, which are of general interest. The measurement techniques applied in this work are similar as those in Fu et al. (2015) and Xie et al. (2016). However, due to the lack of source tracers, the explanation of WSOC sources is ambiguous.

→ Yes, the optical properties and sources of WSOC have been studied extensively in many previous studies. We also measured the stable carbon isotope ratios and inorganic ion species including K^+, SO_4^{2-}, Na^+, Cl^-, Ca^{2+}, NH_4^+, and NO_X to determine individual sources of WSOC, although tracers such as levoglucosan and vanadium are not available. However, we all know that none of previous papers, including this work, clearly identify all different sources of WSOC.

The originality of our study is the dominance of sink in determining the seasonal variations of HULIS in WSOC. Regardless of different sources (either identified or unidentified) over all seasons, the seasonal variations of HULIS in WSOC or HULIS/WSOC were clearly dependent on photo-degradation. This was verified by statistically for measured samples (Figure 4b) and further confirmed by laboratory experiments (Figure 5). Our finding of this sink-dependent variation of HULIS in WSOC, which has been neglected so far, is a very important breakthrough in aerosol sciences.

(2) This paper focused on the seasonal change in optical properties of WSOC based on EEM analysis. So I would suggest the title to be changed to "Significant seasonal change in optical properties (and/or sources) of water soluble organic aerosol in urban Seoul", or something else similar. In this work, the separation of atmospheric humic-like substance (HULIS) is mainly based on EEM results, but not chemical separation after extraction. In some previous studies (e.g., Baduel and Jaffrezo, 2010; Lin et al., 2010), HULIS in aerosols were extracted from other organic components using column separation.

→ Yes, we changed the title as suggested in revised paper. Yes, we also conducted the separation experiments (using DEAE resin) as a preliminary test (results are shown below). The HULIS EEM results before extraction were in good agreement with the HULIS carbon amounts and HULIS EEM results after extraction. Now the figures are added in supplementary information of revised manuscript.



(3) Page 6, lines 25-28. “The WSOC concentration showed seasonal variation from . . ..This result is consistent with seasonal trends observed by Xie et al. at high elevation remote site (Xie et al., 2016).” In Xie et al. (2016), the WSOC concentration exhibited maxima in the summer and minima in winter, in contrast to the results reported in the current work. The higher WSOC concentration in summer in Xie et al. (2016) is primarily due to SOA formation. Please explain the differences.

→ Yes, Xie et al. (2016) showed that the WSOC concentrations were maxima in summer and minima in winter due to SOA formation at high elevation sites in the Colorado Rocky Mountains. However, Hecobian et al. (2010) and Jo et al. (2016) showed absorption (water-soluble brown carbon) maxima during the winter and minima during the summer periods over the Southeastern US and in Atlanta, GA. Cheng et al. (2013) and Huang et al., (2012) also showed WSOC maxima in winter and minima in summer seasons in different regions of China. Our results are consistent with these studies, which seem to be more general phenomenon (Hecobian et al., 2010; Jo et al., 2016; Cheng et al., 2013; Huang et al., 2012). As explained earlier, our statistical and laboratory experiment results explain this general variation tends clearly.

In general, SOA formation results in more depleted $\delta^{13}C$ values due to the atmospheric processing (Miyazaki et al., 2012; Bosch et al., 2014). However, our general summer $\delta^{13}C$ values are enriched, indicating the seasonal trend observed in Xie et al. (2016) is unusual.

(4) Page 7 lines 25-30. The authors inferred that the decrease of HULIS fluorescence should be attributed to the increase in UV radiation in summer, supported by the negative correlation shown in Figure 4b. It is well known that the UV radiation always reaches the maxima in warm season and the minima in cold season, and the increase in UV radiation can play a role in the degradation of HULIS in summer. However, the author might ignore the source type or source region changes from cold to warm season, which might impact the HULIS fluorescence more than the photo-oxidation process. As shown in Figure 1, air masses are mainly from ocean area in summer, and from continental areas (China and Mongolia) in other seasons. It is probably that the continental aerosols contain more WSOC and HULIS components than ocean aerosols. The seasonal changes in HULIS and WSOC might be more associated with the variation in source type or region. To identify which factor (photo-oxidation vs. source variation) is the primary reason for the seasonal changes in optical properties of WSOC, the authors need to do more work on source apportionment of WSOC for the sampling period. For example, the 13C fraction of WSOC has been analyzed in this work, so the authors should be able to analyze the relative contribution of continental and marine WSOC based on the method applied in Fu et al. (2015). Then the major WSOC source in cold and warm periods could be known.

→ Yes, the source term is very important. However, as explained earlier, our statistical data from measurements and laboratory experiments show that sink is the dominant term. Our carbon isotope measurements, air-mass trajectories, and other source tracers show that the sources, pathways, and aging are diverse. If these source terms are more important, our UV versus HULIS correlations should show corresponding scatters. Also such source dependent scatters should appear in the laboratory exposure results. We believe that our work can be applied to explain general seasonal variations of HULIS in WSOC, except for some special regions (Xie et al., 2016).

(5) The photochemical degradation experiment can only demonstrate that HULIS could be photoxidized by UV lights, which has already been reported in previous work (Kieber et al., 2007; Zhang et al., 2013). While the oxidation of HULIS in the ambient samples could hardly be reflected. As such, section 3.4 cannot help to rule out the effect of source type and/or region change on the seasonal variations of HULIS in WSOC.

→ Yes, photochemical degradation on CDOM has been well documented in aquatic environment including in rainwater (Kieber et al., 2007) and river waters (Zhang et al., 2013). However, there is no documented work for UV effects on HULIS in ambient aerosols. Furthermore, our work argues that this sink term, which has been neglected so far, is the most important process that explains the seasonal variations of HULIS in the atmosphere. The reviewer think that UV effect is very important and well known, although the source term is more important to explain the seasonal variations of HULIS in WSOC from above comments. However, our data, together with laboratory experiments, clearly show that the source term is minor compared with the sink term in typical temperate atmosphere. We believe that this photo-induced removal mechanism of HULIS provides better understanding of hidden life cycle of light-absorbing organic aerosols in the atmosphere for various source inputs.

Minor comments: 1. In supporting information, Figures S1 and S2 are mistakenly labeled as Figures 1 and 2.

→ Labels are changed in the revised version.

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