

(1) As mentioned by the author in the replier, the originality of this work is to demonstrate that photo-degradation is the primary sink of WSOC, which determines the seasonality of ambient WSOC in urban Seoul. Also, the authors thought the dominance of photo-degradation on seasonal variation of WSOC and HULIS had been neglected so far, and should be a very important breakthrough in aerosol science.

However, due to the lack of evidence, it is very hard to be convinced that the low WSOC concentration and fluorescence intensity of HULIS in summer is dominated by photo-degradation. The statistical correlation (Fig. 4b) could not be used as direct evidence. Like ambient temperature, UV radiation rate is a meteorological parameter, it always reaches maxima in summer and minima in winter. If negative correlation was also observed between HULIS fluorescence and temperature (very possible), can we say the gas/particle partitioning is the dominant mechanism leading to the seasonal variation?

Here are other possibilities for the seasonal variations: the high temperature in warm periods can evaporate more volatile WSOC and HULIS components into gas phase; as mentioned in the original comments, source type and region changes might also lead to that seasonal variation, we can clearly identify the source region changes from Figure 1. Air masses are mainly from continental areas (China, Mongolia and Russia) in spring, fall and winter; marine areas are the origin of summer time aerosols.

Without quantifying the impact of these factors, the major conclusion (photo-degradation dominates seasonal variation of WSOC) of this study is not valid. At least, the authors need to estimate the relative contribution of marine and continental influences during the sampling periods.

The photochemical degradation experiment cannot be used to rule out the existence of gas/particle partitioning and source region changes. It can neither help to quantify the relative contribution of photo-degradation on WSOC and HULIS in summer.

(2) In the authors' replies to my previous comments, a few studies were cited to show the consistent seasonal variation of WSOC between the current work and other urban areas. Hecobian et al. (2010) observed high WSOC in both winter and summer, dominated by biomass burning and SOA formation respectively. The low light absorption of WSOC in summer is due to the weak source strength of biomass burning. Jo et al (2016) is a modeling study BrC absorption, which might not be appropriate for comparison. In that study, the low BrC absorption in summer is also attributed to the missing of biomass burning. If the seasonal variation of WSOC and HULIS in those two studies (Southeastern US) and the current study are dominated by the same mechanism, then biomass burning contributions need to be quantified in cold seasons. The biomass burning source in winter China is very strong, which might have an influence on the seasonal variation of WSOC and HULIS fluorescence in this study. Cheng et al. (2013) investigated the seasonal variation of WSOC and other organic compounds in an inland city of China (Xi'an), a totally different study site than the current work. Xi'an is much less impacted by marine aerosols than Seoul. In Cheng et al. (2013), the higher concentrations of WSOC in winter is mainly due to the stronger emissions from household combustion of coal and open burning of waste material. Huang et al. (2012) investigated the spatial and seasonal changes of WSOC and other PM_{2.5} components in Pearl Delta River in China, a similar sampling site as the current work. They inferred that the lower WSOC level in summer is explained by clean marine air masses from the south China sea and the washing effect of frequent rainfall. All the above cited studies conclude that source type or region change dominate the seasonal variation of WSOC in PM_{2.5}. It is very hard to believe that the influence of source type and region change is minor compared to photo-degradation without direct evidence. Therefore, due to the lack of evidence, the result and discussion of this work cannot support the conclusion. In addition,

the measurements and experiments conducted in this work cannot guarantee novelty. Again, this work is not recommended for publication on Atmospheric Chemistry and Physics.

→ Thank you for your valuable comments. Since your comment points out the same problem with the reviewer 2, our responses and actions are the same as below.

Yes, we agree that the source and sink terms cannot be distinguished by the correlation analyses. If there are no significant source inputs in summer, low concentrations of photo-resistant HULIS can be observed in summer, while high photo-degradable HULIS concentrations can be observed in winter. However, photo-degradation experiments showed that the winter HULIS concentrations could be decreased to the summer HULIS concentrations if the samples are exposed to the enhanced summer UV. This indicates that UV degradation can be also the major cause considering the residence times (~10 days) of aerosols in the atmosphere. Therefore, in the revised version, we focus on the characteristics of HULIS for different seasons although we open questions for the cause of the seasonal variations of HULIS.

In order to quantify UV-degradable HULIS, in the revised version, we added more UV degradation analyses (n=50), expanded from the two. All aerosol filter samples for the experiment were duplicated or triplicated to rule out experimental bias. The sharp decreases in UV-degradable HULIS from winter to summer (correlating well to either HULIS or HULIS/WSOC) clearly characterizes that there is significant seasonal variations in UV-degradable HULIS. The figures will be added in supplementary information of revised manuscript.

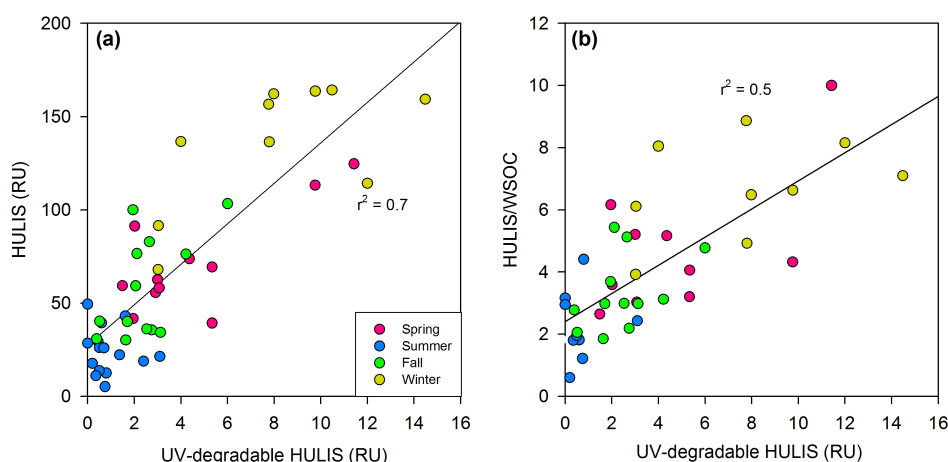


Figure a. The correlations between (a) UV-degradable HULIS and HULIS, and between (b) UV-degradable HULIS and HULIS/WSOC.

We believe that this finding alone is very important for understanding HULIS quantity and quality in the atmosphere.