

(1) The main assertion of this work is that the seasonal variation of HULIS in the urban environment of Seoul was driven by the photo-induced degradation, and therefore photochemical degradation is an important sink that was previously overlooked. The authors arrived at this conclusion based on (1) a statistically significant negative correlation ($r^2 = 0.5$, $p < 0.001$) was observed between fluorescence intensity of HULIS and UV radiation (Fig. 4 b); and (2) the fluorescence intensity of aerosol water extracts decreased by 52% for a winter sample (Fig. 5a) and no change for a summer sample (Fig. 5b) after 6-week UV irradiation in laboratory experiments. Neither of the two observations could convincingly lead to the main assertion. As for the first observation, correlation does not equate cause-and-effect relationship. Incomplete combustion (e.g., biomass burning, coal combustion) and secondary formation are known to be significant sources of HULIS (e.g., Lin et al, 2010; Kuang et al., 2015). The seasonal variation in HULIS in Seoul (i.e. higher concentrations in the winter and lower in the summer) could well be a result of enhanced HULIS sources in the winter as combustion-related sources were more prominent in the region while in the summer the clean marine air would dilute the overall PM levels and WSOC level as well. While the second observation indicates that the fluorescing component in WSOC in the winter sample photo-degrades, this is not evidence to indicate that the lower HULIS level in the summer is due to photo-degradation. In addition, the photo-degradation experiment was only conducted on two samples and this sample size is too small to draw a conclusion about the degradation rate of HULIS. Without quantitative data on typical photo-degradation kinetics, the claim of photochemical degradation as an important sink remains unsubstantiated.

→ Thank you for your valuable comments. Since your comment points out the same problem with the reviewer 1, 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.

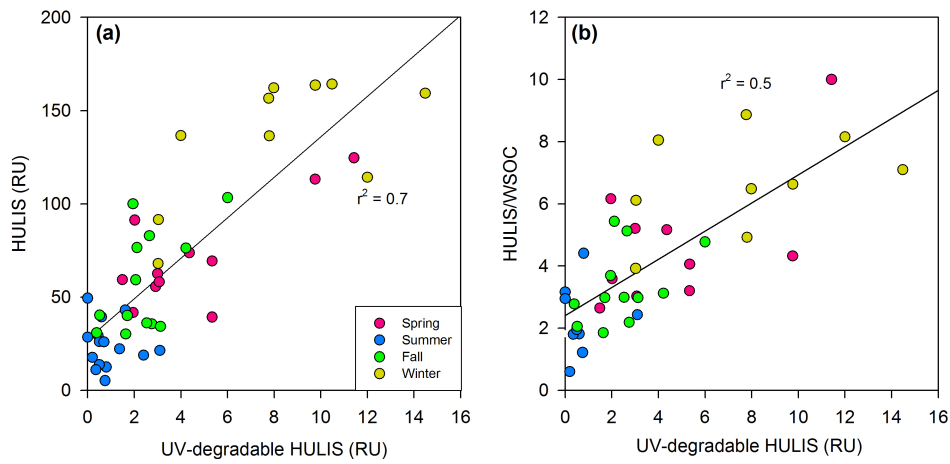


Figure a. The correlations between **(a)** UV-degradable HULIS and HULIS, and between **(b)** UV-degradable HULIS and HULIS/WSOC.

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