Answers to Reviewer #2 :

In this reviewer's opinion, the conclusions are oversimplified. The Authors state that the nature of HULIS is mainly a function of the period of the year, while it is largely independent of the geographical location. However, a clear seasonal cycle is provided only for the site of Grenoble, while for the other sites HULIS data are available only for one of the two seasons (warm or cold). Table 1 shows that the UV absorbance of the wintertime samples in Grenoble are similar to those measured in the same periods at the northern French sites, and that the absorbance of Grenoble summertime samples are similar to those of HULIS in southern French cities in the summer. However, no seasonal cycle has been shown to occur for cities other than Grenoble. Therefore, a common phenomenology of HULIS over France is only hypothetical. It is difficult to assess the actual causes of the observed changes in HULIS absorbance in Grenoble without supporting information from meteorological data, back-trajectories and transport patterns, proxies for the various sources, etc.. In the absence of supporting data, the discussion about the sources of HULIS is too speculative and the quality of the paper may not meet the standards of Atmos Chem Phys.

Answer: We agree with the reviewer that in its current form, our paper oversimplified some aspects of our results. This was intended in order to keep the paper short and thus stress the main result: there exists a seasonal cycle in optical properties of HULIS in urban environments, observed in Grenoble, and suspected in other French cities. This, to our knowledge, has never been clearly stated in any previous study: the only published seasonal comparisons on UV-Vis absorption between summer and winter are those of (Duarte et al., 2005) and (Krivácsy et al., 2008), that only deal with a very small number of samples.
We agree that the common phenomenology we claim is more hypothetical than actually proven by our data, and will clearly say so in a revised version of the manuscript. Yet, as we answered to reviewer #1, there is circumstantial evidence to sustain our hypothesis. Our conclusion holds if "summer cities" and "winter cities" have similar sources all along the year, whose seasonal behaviour would then be responsible for our observations. In a revised version of the paper, we will add correlation data on HULIS vs K^{+}_{ndust} in our "winter sites". This extremely clear correlation ($r^2 = 0,87$ in Grenoble), inexistent in summer, implies the dominance of biomass burning as the source for HULIS in winter. As biomass burning in cities in winter is related to residential heating, it is not expected to operate in summer, so that we feel very confident that the observed modification of HULIS optical properties on Grenoble will also be observed for our "winter sites".
This, of course does not imply that the optical properties of HULIS in summer at our "winter sites" should be the same as observed in Grenoble. We might conclude so if we show that on our "summer sites", HULIS are linked to the same unique source, and that this source cannot be ruled out in our "winter sites". We have now correlated HULIS in summer to oxalic acid, which points to a common source in summer. In the same time, we can also show that EC is not correlated to HULIS nor oxalic acid in our "summer sites", excluding automotive exhausts as this common source. From here, the most plausible source for both HULIS and oxalic is secondary processes, from biogenic and/or anthropogenic VOCs. Moreover, as we show that HULIS have similar optical properties in summer across 3 very different French cities (Marseille is heavily influenced by petrochemical and other industrial activities, Grenoble and Toulouse much less so) we feel that our hypothesis is very sound. This discussion will be added in a revised version of the paper.

Wintertime and summertime HULIS are discriminated based on the UV absorbance at 254 nm, and the Authors must acknowledge that their analysis exploits only a very simple spectroscopic parameter, and that taking into account measurements performed at different wavelengths or using high-resolution spectroscopic techniques may lead to a more complex classification of the samples. Some studies have shown that the UV absorbance at 254 nm is not specific for aromatic compounds and can be associated to aliphatic polymeric materials (Guzman et al., J. Phys. Chem. A, 2006, 110, 3619 – 3626). Clearly, without recording the absorbance over the whole spectral range, any interpretation about the underlying chemical mechanisms, like changes in aromaticity or in the substituents of the aromatic rings, must be considered with caution.

Answer: We agree with the reviewer that considering only the aborbtivity @ 254nm is a crude way to treat our spectroscopic data, although it had the merit of simplicity. We will include in a revised version of the manuscript data at several other wavelength (namely 272 nm, 280 nm, 300 nm and 365 nm) as well a typical spectra of winter and summer samples, although for the mixture of compounds that constitutes HULIS, spectra are essentially featureless, unlike the spectra of individual species shown in (Guzman et al., 2006).
Results are essentially unchanged, with an observed decrease of UV absorptivity from winter to summer. Knowledge of HULIS source in winter (biomass burning) then helps in tentatively attributing the increased absorptivity in th UV range to aromatic systems that have been shown to be common in such biomass burning aerosol.

The title of the paper suggests that specific features of HULIS for the urban environment will be presented. On the contrary, the Authors conclude that the chemistry of HULIS can be explained by regional scale processes, even in very big cities (Paris). Is there any rural site (except Chamonix, whose location cannot be considered representative for the whole France) to compare with? If there is no evidence for specific characteristics of HULIS related to their occurrence in the urban atmosphere, I suggest to refocus the title to the actual results presented in the paper.

We do not have data on a rural site to compare with yet. Moreover, Chamonix cannot be considered a rural site, as (especially in winter) it has been showed to be actually typical of a source area.
In order to refocus on our results we propose as a new title : "Seasonal variations of concentrations and optical properties of water soluble HULIS collected in urban environments"

Minor comments:

- Paragraph 3.3.2. The Authors quote the literature on SOA polymers, but I suggest to focus on the studies really presenting data about aromatic or UV-absorbing compounds.
The paragraph has been rewritten with a lighter emphasis on SOA polymers.

- Table 1. The classification of the samples collected in Grenoble in "March, April, October" and in "Mid-April and September" into cold season and mid-season is not clear.
The classification will be made according to months

- I contest that the slope of the line of wintertime HULIS in Figure 1 can be simply explained by mixing biomass burning and fossil fuel combustion aerosols in the same proportions at all locations (end of section 3.3.1). The same emission inventory cannot be assumed for all sites.
The mixing argument made at the time the paper was written because it could not a priori be ruled out, although it implied the unlikely fact that all cities had a similar emission inventory. Since we have investigated the correlation between HULIS vs K^{+}_{ndust} , indicating clearly the origin of HULIS, the mixing argument will be suppressed and the discussion rewritten accordingly.

- I suggest to group the samples shown in Figure 2 according to location not to season.
The figure has been modified so that location also appears together with seasons

Duarte, R. M., Pio, C. A. and Duarte, A. C.: Spectroscopic study of the water-soluble organic matter isolated from atmospheric aerosols collected under different atmospheric conditions, Analytica Chimica Acta, 530(1), 7–14, 2005.

Guzman, M. I., Colussi, A. J. and Hoffmann, M. R.: Photoinduced Oligomerization of Aqueous Pyruvic Acid, The Journal of Physical Chemistry A, 110(10), 3619-3626, doi:10.1021/jp056097z, 2006.

Krivácsy, Z., Kiss, G., Ceburnis, D., Jennings, G., Maenhaut, W., Salma, I. and Shooter, D.: Study of water-soluble atmospheric humic matter in urban and marine environments, Atmos. Res., 87(1), 1-12, doi:10.1016/j.atmosres.2007.04.005, 2008.