

Answers to Reviewer 1 :

There are several significant findings mentioned in the conclusions that are not highlighted in the abstract (e.g., HULIS_ws comprise up to 43% of WSOC). If added to the abstract, future works are more likely to cite the relevant findings of this paper.

Answer: This will be done.

The authors suggest that the trends are seasonal and not necessarily a function of location (P21571 L18). Yet only one city (Grenoble) supports this. The reviewer is concerned that data set is too limited for this conclusion. For a statistically sound argument, more data points (sites) are required to show minimal effects of location/source on the composition of HULIS_ws. Figure 1 and text suggest that the measured absorbance is influenced by the contributions from different sources (e.g., biomass burning and vehicle emissions). Do some French cities have larger ambient anthropogenic sources or biomass burning? If so, do the grouping of cities in Figure 1 and 2 reflect this?

Answer: We agree that we do not have so many data points (sites) and that having complete seasonal cycles in different cities would be preferable. Our findings have actually lead us to plan for a soon to be started multi site seasonal investigation of HULIS that should be included in a future publication. At this stage, as the reviewer stresses, it would be safer to treat our affirmation of a geographically independent seasonality of HULIS structure as a hypothesis. The text will be modified in that direction. Yet, it should be stressed that our data represent the first complete seasonal cycle on HULIS absorptivities and that the measured values compare favourably with the few data published by (Duarte et al., 2005), who show comparable values of $18-22 \text{ L.g}^{-1} \cdot \text{cm}^{-1}$ in summer, increasing to $25-30 \text{ L.g}^{-1} \cdot \text{cm}^{-1}$ in autumn. Although they use a different HULIS isolation scheme, this difference should be minimal in summer, which strengthen our hypothesis. Another study that can be compared to ours is that of (Krivácsy et al., 2008), although the units used for absorptivity are not so clear. This lead us to add to our discussion a paragraph on absorbance ratios at 2 different wavelength, that shows similar trends between Grenoble and Auckland.

Another point made by the reviewer concerns seasonal sources of HULIS and our grouping of cities. If we understand correctly the reviewer's concerns, he fears that our conclusion only holds only if "summer cities" and "winter cities" have similar sources all along the year, whose seasonal behaviour would then be responsible for our observations. The main currently assumed source for HULIS in winter (Feczko et al., 2007) is biomass burning. This is further reinforced by (Schmidl et al., 2008) who show that HULIS amount for ~20% of PM10 emitted by biomass burning and the numerous studies that show the high prevalence of biomass burning in most cities. Anyway, we show on our "winter sites" a clear correlation between HULIS and $\text{K}_{\text{ndust}}^{+}$, which is generally considered as a tracers for biomass burning. This correlation will be included in the revised version of our paper. 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. Yet, as we show that HULIS have similar optical properties in summer across 3 very different French cities, we feel that our hypothesis is very sound. This discussion will be included in the revised version of out paper.

P21564. Are inorganics present in samples? Will they not also affect the absorbance at 250nm? The contribution of inorganic may affect solution pH and HULISws samples (Shapiro et al., 2009). This issue is not addressed.

Answer: Our isolation scheme has been showed to efficiently exclude common anions and especially nitrate from the HULIS extract (Baduel et al., 2009), so those potentially UV absorbing species will not interfere on the measured spectra. This is now stressed in the text. Another concern raised by the Shapiro et al paper could be that reactions between HULIS components occur in our high concentration $\text{Na}^{+}, \text{Cl}^{-}$ extract that could modify the overall absorption spectrum. Although this cannot be ruled out, we have no knowledge of such reactions having ever been reported; in (Nozière et al., 2009), $\text{Na}^{+}\text{Cl}^{-}$ is actually used as a comparison point to make the case of reactions occurring in other salt solutions. Moreover, our spectra are measured online, so that HULIS only are 2 minutes in contact with the saline solution, which would make for an extremely fast reaction.

P21562. HULIS_ws is not defined in the abstract. : It now is defined
P21562 L22. Asa-Awuku et al, 2008 also investigate the properties of WSOC HULIS
from biomass burning sources. : Citation added

P21566 L18. “. . .aromatic structures show strong absorption at this [250 nm] wavelength.”
Please cite reference.
Several references will be added, coming from literature on fulvic material in natural waters

P21566 L22. What is resp.? respectively? Response? Not clear. :
resp is respectively. It is now written in full

P21566 L11. DOC? Dissolved organic carbon? Or is this TOC? Total organic carbon.
Abbreviation is not defined in text.
DOC is Dissolved Organic Carbon. Has been confused with TOC. The text now states TOC

P21570 L18. Remove “kind of.”
Done

P21571 L11. What is meant by “has been realised?”
The sentence now reads : “Organic aerosol concentrations have been monitored in six different French cities during winter and summer.”

Baduel, C., Voisin, D. and Jaffrezo, J. L.: Comparison of analytical methods for Humic Like Substances (HULIS) measurements in atmospheric particles, *Atmos. Chem. Phys.*, 9(16), 5949-5962, 2009.

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Feczko, T., Puxbaum, H., Kasper-Giebl, A., Handler, M., Limbeck, A., Gelencsér, A., Pio, C., Preunkert, S. and Legrand, M.: Determination of water and alkaline extractable atmospheric humic-like substances with the TU Vienna HULIS analyzer in samples from six background sites in Europe, *J. Geophys. Res.*, 112(D23), D23S10, doi:10.1029/2006JD008331, 2007.

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.

Nozière, B., Dziedzic, P. and Córdoba, A.: Common inorganic ions are efficient catalysts for organic reactions in atmospheric aerosols and other natural environments, *Atmospheric Chemistry and Physics Discussions*, 9(1), 1–21, 2009.

Schmidl, C., Marr, I. L., Caseiro, A., Kotianová, P., Berner, A., Bauer, H., Kasper-Giebl, A. and Puxbaum, H.: Chemical characterisation of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions, *Atmospheric Environment*, 42(1), 126–141, 2008.