

Interactive comment on “Investigation of structural changes of atmospheric aerosol samples during two thermal-optical measurement procedures (EUSAAR2, NIOSH870)” by Theresa Haller et al.

Anonymous Referee #2

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This study investigated the structural changes of aerosol while heating of atmospheric aerosol samples during thermal-optical measurements. Two different thermal-optical measurement protocols (EUSAAR2, NIOSH870) were investigated. Charring of organic carbon is an important issue to investigate and can impact the thermal-optical measurements. The authors used Raman spectroscopy to track structural changes upon heating the atmospheric aerosol samples. They also used ion chromatography and integrating sphere measurements to derive ionic compositions and light absorbing fraction of carbonaceous material. They used different peak ratios derived from Ra-

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man spectra as an indicator to probe structural changes. The authors observed that D/G peak ratios of the Raman spectra was higher for sample heated via NIOSH870, suggesting higher degree of structural ordering. Samples heated via EUSAAR2 protocol exhibited complex behavior of structural changes. Investigation of washed sample with low water-soluble organic carbon and inorganic salts showed no structural ordering and darkening of sample. Overall, the authors suggested that structural ordering of sample may not be responsible for darkening of sample.

I think the research topic is relevant and important for the community. However, the overall presentation of the manuscript bit convoluted and it can be structured better way so that it will easier for readers to follow. For example, discussion of structural changes for unwashed (original) and washed sample should be discussed together. Perhaps a comparison of Fig 2 and Fig. 10, maybe just compare for one temperature would be useful for readers. Some of discussions need to elaborate. When you state about some observation and I suggest elaborating what does that mean. After revision of the manuscript, I think it will be suitable for publication.

Specific comments:

In the abstract the authors highlighted that structural ordering may not be responsible for darkening of sample, I suggest the authors to put the hypothesis here, what might be the possible cause for this.

I was wondering if the pre-edge of the spectra was considered or normalized while comparing the spectra? It was not clear to me how the different “Raman categories” were assigned? I suggest the authors to provide quantitative peak ratios to define more robust categories.

It was not clear to me how the authors relate the structural changes with the changes in the transmission laser signal. Having a quantified variable structural change (may be using the peak ratios) and compare with the transmission laser signal will be useful. I see the Fig 7 discussed the BrC/LAC ratios with qualitative structural changes. A

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quantified comparison would be useful here too.

Comparison of washed and unwashed samples are interesting. Just wondering if authors investigated any standard water-soluble organic carbon to investigate the laser signal. Observation of higher initial laser signal of washed samples compared to unwashed samples need to be discussed in detail.

I was wondering about the sensitivities of the Raman peaks here. How confident authors are regarding the graphitization of carbon. Some of the previous studies showed changes in graphitic structure upon heating black carbon particles using high resolution TEM imaging.

Why the early and fast category of Raman categories are noisier compared to others?

The increase of laser signal above 650 degree C maybe due to the decomposition of the dark intermediate OC products without graphitic structures. Please elaborate the discussion about the decomposition of the dark intermediate OC products.

The authors stated that EUSAAR2 produced less pyrolyzed carbon. Please add some discussion here.

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