Structural changes of combustion aerosol samples during a thermal-optical measurement protocol

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Keywords: EC/BC, combustion aerosol, thermo-optical, Raman, structure
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Elemental Carbon (EC), Black Carbon (BC) and Organic Carbon (OC) contribute a large amount to atmospheric aerosols. Due to climate and health effects, there is an essential need for reliable measurements of these components. Despite intensive investigations, there are still discrepancies between data obtained with different methods. Especially during winter, the discrepancies increase with a coincident increase of Brown Carbon (BrC) caused by wood combustion. (Reisinger et al. 2008, Hitzenberger et al. 2006, Wonaschütz et al. 2009).

The thermal-optical method, based on a two-phase thermal analysis, is a common tool for EC/OC determination. An optical laser correction should take into account the effect of pyrolysis. Nevertheless this effect leads to uncertainties in the OC/EC split (Cheng et al. 2012)

In this study structural analysis of the pyrolysed material was performed with Raman micro-spectroscopy. Combustion aerosol from a miniCAST soot generator, deposited on quartz fibre filters, was analysed with a dual-optics ECOC analyser (Sunset Instruments Inc.). Filter punches were heated for Raman measurements. The obtained data were analysed according to Sadezky et al. (2005) and compared to the thermal-optical measurements of EC/OC and to optical measurements of BC/BrC with the integrating sphere method (Wonaschütz et al. 2009).

The Raman spectroscopy analysis sheds light on structural changes caused by heating. By applying a five curve fitting method developed for soot spectra (Sadezky et al. 2005) it was possible to interpret the degree of graphitisation and the amount of organic and amorphous carbon in the sample (Table 1)

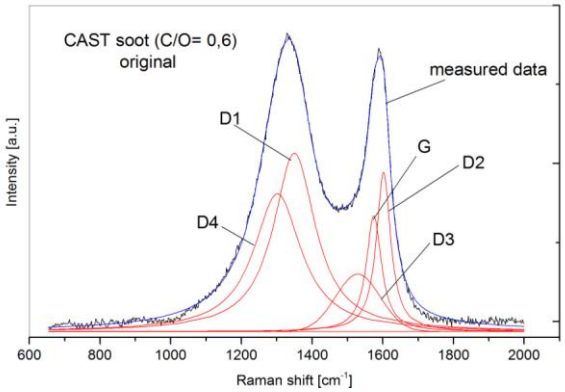

Figure 1 Raman spectrum of a CAST soot sample before heating

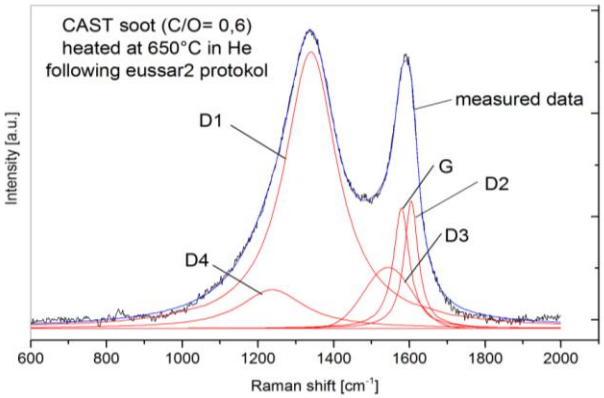

Figure 2 Raman spectrum of the same CAST soot sample after heating

Sample	G/D1	D1/D4
orig. Sample	0.243	1.155
heated Sample	0.137	5.972

Table 1 Ratios of the fitted peak areas

First results from the analysis show the bands of amorphous and organic carbon (D4, D3), graphitic lattice (G, D2) and graphitic defects (D1) (Fig 1 and 2). While the relationship of the G, D2 and D3 bands stays rather constant, significant differences appear in the D1/D4 ratios before and after heating, indicating a change in the degree of graphitisation (Table 1)

This measuring procedure is applied to mixtures of soot and BrC standards, wood combustion aerosol and atmospheric aerosol, respectively, and compared to EC/OC as well as BC/BrC analyses.

Acknowledgement: This work is supported by the FWF (Austrian Science Fund) grant P26040.

Cheng Y. et al (2012) Atmospheric Environment, 61 453-462.
Hitzenberger R. et al. (2006) Environmental Science and Technology, 40 6377-6383.
Reisinger P. et al. (2008) Environmental Science and Technology, 42 884-889.
Sadezky A. (2005) Carbon, 43 1731-1742.
Wonaschütz A. et al. (2009) Environmental Science and Technology, 43 1141-1146.