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Structural changes of a combustion aerosol standard (CAST) soot during a thermal-optical measurement procedure

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Elemental Carbon (EC), Black Carbon (BC) and Organic Carbon (OC) are important components of the atmospheric aerosol because of their climatic and health effects. However, measurements show method specific differences by factors up to nine especially in the presence of Brown Carbon (BrC) (e.g. Reisinger et al. 2008, Hitzenberger et al. 2006, Wonaschütz et al. 2009). Hence a better understanding of the physico-chemical basis of these differences is needed.

During thermo-optical analyses of EC and OC, samples are heated first in an inert (He), then in an oxidizing (He+O_2) atmosphere. The pyrolysis of the samples during the inert mode is corrected for optically, but uncertainties in the OC/EC split remain (Cheng et al. 2012). The structural reorganizations in the sample caused by the pyrolization during the heating process are not yet entirely understood.

In the present study, structural reorganizations of samples produced by a miniCAST soot generator during the heating process were investigated using Raman microscopy, transmission electron microscopy (TEM) and UV-VIS spectroscopy. The integrating sphere method (Wonaschütz et al. 2009) was used to analyse the BrC content of the samples.

A miniCAST soot generator (Jing Ltd) was operated under different combustion conditions from oxygen rich to oxygen poor fuel mixtures in order to generate samples with different amounts of BrC. The original samples were analysed with a dual-optics ECOC analyser (Sunset Instruments Inc.) which was also used for the preparation of the heated samples. The heated samples were analysed with Raman microscopy which is sensitive to different bonding types in carbonaceous materials. The Raman spectra show a change at heating to 870°C in the He atmosphere for samples with a high BrC content which indicates an increased amount of polyaromatic rings in the sample (Ferrari and Robertson 2000) i.e. an increase of order in the carbonaceous material.

The TEM measurements confirm the interpretation of the Raman spectra and give additional information about the arrangement of the aromatic rings.