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Structural changes of CAST soot and atmospheric aerosol samples during two thermal-optical analysis protocols (EUSAAR2, NIOSH870)

Theresa Haller (1), Eva Sommer (1), Hinrich Grothe (2), and Regina Hitzenberger (1)

(1) University of Vienna, Faculty of Physics, Vienna, 1090, Austria , (2) Vienna University of Technology, Institute of Materials Chemistry, Vienna, 1060, Austria

Elemental Carbon (EC), Black Carbon (BC) and Organic Carbon (OC) contribute a large amount to atmospheric aerosols. Due to their significant influence on climate and health, a reliable measurement of these components is essential. Nevertheless, their correct determination is not trivial since results of different measurement techniques show 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).
EC and OC are usually measured with thermal-optical techniques, where the sample is heated stepwise, first in an inert (He) atmosphere, then in an oxidizing (He+O_2) atmosphere. The pyrolysis of the sample during the heating procedure is traced with a laser transmission/reflection signal and the (theoretical) amount of pyrolyzed carbon (PC) is attributed to OC in the subsequent evaluation. Despite this optical correction, the pyrolyzation of OC can lead to uncertainties in the OC/EC split (Cheng et al., 2012). Especially Brown Carbon (BrC) and other water soluble organic carbon (WSOC) have a high tendency to pyrolyze and therefore bias the OC/EC split. Several metal salts in the atmospheric aerosol can influence the pyrolyzation of OC and/or catalyze the evolution of EC (Wang et al., 2010). This highly complex chemical and physical situation is not fully investigated and understood. In the present study we try to shed more light on the processes, which occur during the thermal-optical heating procedure, particularly the structural reorganisations of the carbonaceous material. Since C-C bonding types are Raman sensitive, we use Raman Spectroscopy to determine the degree of structural ordering (i.e. the amount of aromatic rings in the material) at different temperature levels of two different thermal-optical heating protocols (EUSAAR2, Cavalli et al., 2010 and NIOSH870, Birch and Cary, 1996) performed in an EC/OC analyzer (Sunset Instruments). The EUSAAR2 protocol was originally designed for European aerosol samples with the intention to minimize charring. In this study we compare the charring behaviour during the NIOSH870 and the EUSAAR2 protocols for both CAST generated soot samples and atmospheric aerosol samples. First measurements show, that the increase of structural ordering is slightly higher, when the sample undergoes the NIOSH870 temperature protocol instead of the EUSAAR2 protocol. First results for the atmospheric aerosol samples show a more complex charring behaviour compared to the CAST generated samples. In addition to the Raman analyses, the investigated samples are analyzed with the Integrating Sphere method (Wonaschütz et al., 2009) for BrC and BC. For the atmospheric aerosol samples back trajectories are calculated with HYSPLIT.