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Investigations of Surface Chemistry on Carbonaceous Particles

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The relevance of carbonaceous particles in atmospheric science is increasing. Reasons for that are their contribution to the urban aerosol, their toxicological importance and their impact on the global radiation balance. Particle emissions like soot or secondary organic aerosols like HULIS (humic like substances) are currently a major topic in atmospheric research. Physical chemistry provides surface analytical methods for these particles [1,2,3,4], to understand their relevance as part of the atmospheric gas phase chemistry.

For these surface investigations Diffuse-Reflectance-Infrared-Fourier-Transform-Spectroscopy (DRIFTS), Temperature-Programmed-Surface-Reaction-Spectroscopy using a mass spectrometer (TPSR-MS), Temperature-Programmed-Desorption-Mass-Spectroscopy (TPD-MS) and Electron-Paramagnetic-Resonance (EPR) Spectroscopy were applied. All of these methods were used to characterize humic substances and soot. DRIFTS provides detailed information about formation and transformation of functional surface groups monitoring the characteristic vibration modes. TPD-MS shows the thermal stability of these groups and TPSR-MS follows up the reaction behaviour of these particles with gases of atmospheric relevance. Radicals on the surface could be studied by EPR Spectroscopy. The combination of these four spectroscopic methods allowed a detailed understanding of heterogeneous reactions with carbonaceous surfaces at a molecular level. Investigations were done on i) the heterogeneous reaction with nitrogen oxides and ii) with reactive halogen compounds. Stability and reactivity of new formed functional groups could be assigned. Single heterogeneous reaction mechanism could be clarified.

[1] Muckenhuber H., Grothe H., The heterogeneous reaction between soot and NO₂

at elevated temperature, Carbon 44 (2006) 546.

[2] Sadezky A., Muckenhuber H., Grothe H., Niessner R., Pöschl U., Raman microspectroscopy of soot and related carbonaceous materials: Spectral analysis and structural information, Carbon 43 (2005) 1731.

[3] Muckenhuber H., Grothe H., The reaction between soot and NO_2 – investigation of functional groups using TPD-MS, Topics in Catalysis 30/31 (2004) 287.

[4] Muckenhuber H., Grothe H., A DRIFTS study of the heterogeneous reaction of NO_2 with carbonaceous materials at elevated temperature, Carbon 45 (2007) 321.