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Development and test of a system for the simultaneous measurement of hygroscopic properties and chemical composition of multicomponent aerosol

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Resumo

To understand the role of aerosols in climate change and health hazard is crucial to answer the following questions: where do they come from, what is the composition of the particles, how the composition can be altered by transformation mechanisms and how do aerosols affect every day life conditioning the cloud formation, playing an important role in the radiative budget and reducing the visibility. In this work the focus will be on the interaction of aerosols with water vapour. The changes in hygroscopic behaviour and chemical composition of particles coated with a film of organic material have been investigated.

In order to investigate the effect of organic films on aerosol particles, a Coating Hygroscopicity Tandem Differential Mobility analyzer was set up. The instrument is based on the Hygroscopicity Tandem Differential Mobility Analyzer. In general the TDMA consists of two DMAs and a conditioning unit, but in this work two conditioning units have been involved: a coating device has been used to condense organic vapour on top of an inorganic seed particle and form a film, and the humidifier has been implied to humidify the aerosols to a certain relative humidity which can either be fixed or steadily varied. Moreover, to monitor the chemical composition as well as the growth factor of the particles under examination, the system comprises the simultaneous measurement of the Aerosol Mass Spectrometer along with the Scanning Mobility Particle Sizer (a DMA coupled with a CPC particle counter). The

instrument development comprised the assembly of the Differential Mobility Analyzer used as selection unit to extract a monodisperse aerosol out of a polydisperse aerosol stream as well as the design and the realization of a device to coat the particles with organic vapour and the humidifying unit. The humidifier and the DMA used in SMPS configuration were housed in an insulated frame in order to decrease the temperature fluctuations, and all the possible sources of heat have been placed outside of the frame. The flow scheme for the SMPS implies a closed loop for the sheath-excess cycle along with a second humidifier which assures that the relative humidity of the sheath air has the same relative humidity as the sample aerosol stream.

The system has been used as HTDMA to measure the humidogram for ammonium sulphate in order to calibrate the instrument in accordance with the thermodynamic model prediction based on the paper by Tang and Munkelwitz. Using the Aerosol Chamber as aerosol generator, particles were produced by ozonolyses of terpenes without OH scavenger and the hygroscopic properties were investigated. For Limonene the humidogram measured in this work was compared to the growth factors measured by Virkkula et al. showing a good agreement. The investigation of α -terpinene and myrcene produced growth factors for two terpenes which have not been under examination before. For the α -terpinene experiment, the addition of ammonium sulfate generated externally mixed aerosols. Different growth factors due to the different hygroscopic properties of ammonium sulphate and the oxidation products of α -terpinene were observed and lead to a split in the peaks in the humidogram. During the experiment with oxidation products of myrcene, size dependence of the hygroscopic properties of the particles were investigated. In the humidogram a phase transition was observed which was corroborated by an increase of the width of the monodisperse peak. At the same time the data measured with the AMS exhibit an increase in the carbon to oxygen ratio and thus a possible effect due to aging of the particles.

The coating device was tested using the system in Coating-TDMA configuration, only the coating device in line but no humidifier. The thickness of the layer that the condensation of oleic acid forms on top of ammonium sulphate particles has been measured and linked to the temperature of the water bath that controls the coating device. By interlaced calculation involving the SMPS and the AMS evidences that the organic vapour forms a film on top of the particle were supplied. Hence, by choosing a temperature for the water bath, the thickness of the layer that condenses on the seed particles can be reproducibly controlled. A similar experiment was performed with latex spheres as seed particles in order to supply an inert kernel of known size to the

coating agent. The comparison of the two experiments is in agreement within experimental error. The coating layer has a thickness in the order of 30 nanometres if the coating device has been maintained at 85°C. This value can be considered as an upper limit for studies of atmospheric interest. Experimental evidences from Aerosol Chambers exhibit a layer thickness in the order of 10 to 20 nanometres for seed aerosols coated with oxidation products of terpenes.

The complete Coating-HTDMA system has been used to measure the implications that a film of oleic acid has for the hygroscopic properties of ammonium sulphate, for several layer thickness. The observation of particle shrinking in the coating device due to evaporation at high temperatures raised the question about reference measurements. Thus, in order to provide an unbiased reference for the non-coated particle size, two coating devices have been used: one was filled with the coating agent under examination and the second was maintained empty; and the two devices were kept at the same temperature. With this approach, even if the seed aerosols shrink in the coating device due to evaporation, the reference is not influenced. The discussion in the literature about the possible effects of organic films on the hygroscopic properties of inorganic particles is inconclusive, sometimes an effect of the coating is observed and sometimes the coating does not influence the hygroscopic behaviour of the pure substance. In this work the measurements show that the growth factor of coated ammonium sulphate is sensibly reduced by the condensed organic film with respect to a particle of pure ammonium sulphate of the same size. The analysis of the results has been made using a Zdanovskii-Stokes-Robinson approach which implies that the organic and inorganic part can be treated as two independent subsystems and that the behaviour of the whole particle is given by addition of the two. Under the assumption that both the organic and the inorganic fraction are in thermodynamic equilibrium with the water vapour, the growth factor of the coated particle can be explained dividing the particle in the two components and using the respective growth factors. Hence the effect of an organic film in the reduction of the growth factor is given by the fact that a substantial fraction (up to about 69% for particle of 156 nanometre coated at 85°C) of the particle is formed by the less hygroscopic organic.

The system is set up and has been thoroughly tested, calibrated and used to produce preliminary results of scientific relevance, setting the ground for further investigations.