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Field measurements of dicarboxylic acids: Spatial distribution, seasonal trends and influence of air mass origin

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Short-chain dicarboxylic acids (DCA) are found in tropospheric particles all around the world and usually make up an important fraction of the particulate organic carbon. However, despite a large number of measurements, the factors controlling their occurrence are still poorly understood. Furthermore, most of the literature studies focus on PM10 or PM2.5 concentrations of DCA. However, the processes leading to the observed concentrations of DCA might be different in different particle size classes. Therefore it is important to study also size-resolved concentrations of DCA.

During this work a total of 125 particle samples were taken at 2 rural sites (Goldlauter, Melpitz) and 3 urban sites (2 in Leipzig, 1 in Dresden) in Germany using a 5-stage Berner impactor. The field measurements took place between 2001 and 2005 during different seasons. The dataset was evaluated in terms of spatial distribution of DCA concentrations on a regional scale, the influence of the season and the solar radiation during sampling respectively, and the influence of the origin of the sampled air masses on the DCA concentrations.

The regional distribution was studied by comparing the concentration patterns obtained simultaneously at three sites (rural, urban background and urban street canyon). It was found that on average the DCA show very similar concentrations at all three sites. Local sources do not seem to play an important role in the studied area (Leipzig conurbation). Slightly increased concentrations of oxalic acid were determined in small particles (Dp = 0.05–0.14 μm) in the street canyon. They could, however, not be correlated to the traffic density in the street, which excludes a primary source of DCA by traffic emissions.

Highest concentrations of DCA were regularly measured during summer. A correlation of the concentrations in the 5 size classes of the Berner impactor with the intensity of solar radiation (global radiation) led to high correlation coefficients, especially for small (Dp = 0.05–0.42 μm) and large (Dp = 1.2–10 μm) particles (R up to 0.85). This indicates a secondary source of DCA.

DCA concentrations in accumulation mode particles (Dp = 0.42–1.2 μm) showed a weaker correlation with solar radiation. However, in this size range a significant correlation of DCA concentrations with the residence time of the sampled air masses above continental areas was found (R up to 0.67). The continental residence time was obtained from 5-day backward trajectories, which were calculated and quantitatively analysed for all sampling periods. The correlation was poor or not significant for the smallest (Dp = 0.05–0.14 μm) and larger particles (Dp = 1.2–10 μm). This reflects on the one side the atmospheric lifetime of particles, which is highest in the accumulation mode size range. On the other side the high correlation of DCA concentrations with the continental residence time in accumulation mode particles indicates major continental sources for this compound class.

In summary, the main findings of this study regarding the concentrations of short-chain DCA, i.e. homogenous regional distribution, secondary production, and continental sources, support the idea of DCA being suitable tracers for continental secondary organic aerosol in general.