Geophysical Research Abstracts
Vol. 15, EGU2013-4185, 2013
EGU General Assembly 2013

Functionalised carboxylic acids in atmospheric particles: An annual cycle revealing seasonal trends and possible sources

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Carboxylic acids represent a major fraction of the water soluble organic carbon (WSOC) in atmospheric particles. Among the particle phase carboxylic acids, straight-chain monocarboxylic acids (MCA) and dicarboxylic acids (DCA) with 2-10 carbon atoms have extensively been studied in the past. However, only a few studies exist dealing with functionalised carboxylic acids, i.e. having additional hydroxyl-, oxo- or nitro-groups. Regarding atmospheric chemistry, these functionalised carboxylic acids are of particular interest as they are supposed to be formed during atmospheric oxidation processes, e.g. through radical reactions. Therefore they can provide insights into the tropospheric multiphase chemistry.
During this work 28 carboxylic acids (4 functionalised aliphatic MCAs, 5 aromatic MCAs, 3 nitroaromatic MCAs, 6 aliphatic DCAs, 6 functionalised aliphatic DCAs, 4 aromatic DCAs) were quantitatively determined in 256 filter samples taken at the rural research station Melpitz (Saxony, Germany) with a PM10 Digitel DHA-80 filter sampler. All samples were taken in 2010 covering a whole annual cycle. The resulting dataset was examined for a possible seasonal dependency of the acid concentrations. Furthermore the influence of the air mass origin on the acid concentrations was studied based on a simple two-sector classification (western or eastern sector) using a back trajectory analysis.
Regarding the annual average, adipic acid was found to be the most abundant compound with a mean concentration of 7.8 ng m-3 followed by 4-oxopimelic acid with 6.1 ng m-3. The sum of all acid concentrations showed two maxima during the seasonal cycle; one in summer and one in winter, whereas the highest overall acid concentrations were found in summer. In general the target acids could be divided into two different groups, where one group has its maximum concentration in summer and the other group during winter. The first group contains all investigated aliphatic mono- and dicarboxylic acids. The high concentrations in summer could lead to the conclusion that these acids are mostly formed during photochemical processes in the atmosphere. However, the concentrations in autumn were often exceeded by the ones in winter. Therefore probably other sources beside photochemical processes have to be considered. The second group consists of aromatic compounds. Because of the high concentrations in winter it can be concluded that photochemical formation plays a minor role and primary emission sources e.g., wood combustion are likely.
Further evidence in determining sources of the carboxylic acids could be obtained from the air mass origin. In general, air masses transported from East have a more anthropogenic influence than the air mass inflow from West. For all aromatic carboxylic acids higher concentrations were determined during eastern inflow, indicating anthropogenic sources. This presumption is supported by high correlations with the elemental carbon (EC). Regarding the aliphatic carboxylic there is one group with higher concentrations when the air mass is transported from West and one with higher concentrations when air mass is transported from East.
In summary the findings of this study reveal a clear difference in the seasonal trends of the single target acids indicating a variety of different sources.