Geophysical Research Abstracts
Vol. 17, EGU2015-14404, 2015
EGU General Assembly 2015

Secondary brown carbon – Formation of light-absorbing compounds in atmospheric particulates from selected dicarbonyls and amines

Christopher Kampf (1,2), Alexander Filippi (1), and Thorsten Hoffmann (1)

(1) Johannes Gutenberg University, Institute for Inorganic and Analytical Chemistry, Mainz, Germany (kampfc@uni-mainz.de), (2) Max Planck Institute for Chemistry, Multiphase Chemistry Department, Mainz, Germany

One of the main open questions regarding organic compounds in atmospheric chemistry today is related to the formation of optically-active compounds and the occurrence of so called brown carbon (Andreae and Gelencsér, 2006). While organic compounds in ambient fine particles for decades have been assumed to not absorb solar radiation, thus resulting in a net cooling effect on climate (IPCC, 2007), it is now generally accepted that a continuum of light-absorbing carbonaceous species is present in fine aerosols (Pöschl, 2003).
In this study, light-absorbing compounds from reactions between dicarbonyl compounds, i.e. glyoxal, methylglyoxal, acetylacetone, 2,3-butanedione, 2,5-hexanedione, and glutaraldehyde, and amine species, i.e. ammonia and glycine, were investigated at atmospherically relevant concentrations in bulk solution experiments mimicking atmospheric particulates. Product analyses were performed using UV/Vis spectrophotometry and (ultra) high performance liquid chromatography coupled to diode array detection and ion trap mass spectrometry (HPLC-DAD-ESI-MS/MS), as well as ultra-high resolution (Orbitrap) mass spectrometry (UHPLC-ESI-HRMS/MS).
We demonstrate that light-absorbing compounds are formed from a variety of atmospherically relevant dicarbonyls via particle phase reactions with amine nucleophiles. Single dicarbonyl and mixed dicarbonyl experiments were performed and products were analyzed. The reaction products are suggested to be cyclic nitrogen containing compounds such as imidazoles or dihydropyridines as well as open chain compounds resulting from aldol condensation reactions. Further, the reactive turnover was found to be higher at increasing pH values. The aforementioned processes may be of higher relevance in regions with high aerosol pH, e.g., resulting from high ammonia emissions as for example in northern India (Clarisse et al., 2009).

References

Andreae, M.O., and Gelencsér, A. (2006): Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols. Atmos. Chem. Phys., 6, 3131-3148.

Clarisse, L., Clerbaux, C., Dentener, F., Hurtmans, D., and Coheur, P.F. (2009): Global ammonia distribution derived from infrared satellite observations. Nature Geoscience, 2, 479-483.

Pöschl, U. (2003): Aerosol particle analysis: challanges and progress. Analytical and Bioanalytical Chemistry, 375, 30-32.