Geophysical Research Abstracts
Vol. 16, EGU2014-12402, 2014
EGU General Assembly 2014

Identification of light absorbing oligomers from glyoxal and methylglyoxal aqueous processing: a comparative study at the molecular level

Emanuela Finessi (1), Jacqueline Hamilton (1), Andrew Rickard (1,3), Maria Baeza-Romero (2), Robert Healy (4), Salvatore Peppe (5), Tom Adams (6), Mark Daniels (6), Stephen Ball (6), Iain Goodall (6), Paul Monks (6), Esther Borras (7), and Amalia Munoz (7)

(1) Wolfson Atmospheric Chemistry Laboratories, Department of Chemistry, University of York, York, YO10 5DD, UK (emanuela.finessi@york.ac.uk), (2) Escuela Ingeniería Industrial Toledo, Universidad Castilla la Mancha, Toledo 45071, Spain, (3) National Centre for Atmospheric Science, University of York, York, YO10 5DD, UK, (4) Southern Ontario Centre for Atmospheric Aerosol Research, University of Toronto, Canada, (5) School of Earth and Environment, University of Leeds, Leeds, LS2 9JT, UK, (6) Department of Chemistry, University of Leicester, Leicester, LE1 7RH, UK, (7) EUPHORE Laboratories, Instituto Universitario CEAM-UMH, 46980, Valencia, Spain

Numerous studies point to the reactive uptake of gaseous low molecular weight carbonyls onto atmospheric waters (clouds/fog droplets and wet aerosols) as an important SOA formation route not yet included in current models. However, the evaluation of these processes is challenging because water provides a medium for a complex array of reactions to take place such as self-oligomerization, aldol condensation and Maillard-type browning reactions in the presence of ammonium salts. In addition to adding to SOA mass, aqueous chemistry products have been shown to include light absorbing, surface-active and high molecular weight oligomeric species, and can therefore affect climatically relevant aerosol properties such as light absorption and hygroscopicity. Glyoxal (GLY) and methylglyoxal (MGLY) are the gaseous carbonyls that have perhaps received the most attention to date owing to their ubiquity, abundance and reactivity in water, with the majority of studies focussing on bulk physical properties. However, very little is known at the molecular level, in particular for MGLY, and the relative potential of these species as aqueous SOA precursors in ambient air is still unclear. We have conducted experiments with both laboratory solutions and chamber-generated particles to simulate the aqueous processing of GLY and MGLY with ammonium sulphate (AS) under typical atmospheric conditions and investigated their respective aging products. Both high performance liquid chromatography coupled with UV-Vis detection and ion trap mass spectrometry (HPLC-DAD-MSn) and high resolution mass spectrometry (FTICRMS) have been used for molecular identification purposes. Comprehensive gas chromatography with nitrogen chemiluminescence detection (GCxGC-NCD) has been applied for the first time to these systems, revealing a surprisingly high number of nitrogen-containing organics (ONs), with a large extent of polarities. GCxGC-NCD proved to be a valuable tool to determine overall amount and rates of formation of the light absorbing species. Data obtained from laboratory solutions were merged with those from SOA generated in chamber experiments conducted at the European PhotoReactor (EUPHORE) from the uptake of gas-phase GLY and MGLY onto AS seeds. While in general the results confirm previous studies in ranking MGLY as more effective than GLY in brown carbon formation, the link between overall optical properties and the identified molecular species is reported here for the first time for both systems.