Geophysical Research Abstracts,
Vol. 10, EGU2008-A-01930, 2008
SRef-ID: 1607-7962/gra/EGU2008-A-01930
EGU General Assembly 2008

Photochemical cycling of humic-like substances in haze aerosol

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Colored, humic-like substances (HULIS) arising from the biodegradation of organic detritus are widespread in natural surface waters, where they ultimately undergo solar photolysis into small alpha-dicarbonylic species, such as glyoxal, glyoxylic and pyruvic acids. Diversely generated and chemically dissimilar HULIS are also found in the atmospheric aerosol. How are significant levels of colored HULIS produced and sustained in the concentrated aerosol phase under intense solar irradiation? Here, this issue is tackled by investigating the solar photolysis of aqueous pyruvic acid (PA) solutions at concentrations representative of the atmospheric aerosol using UV-absorption, high resolution electrospray mass, and nuclear magnetic resonance spectrometries. Under such conditions, PA is not photodegraded but yields polyfunctional polymers, whose mass and UV-absorption spectra remain unaffected after 3, 8 and 22 h photolysis. Unless diluted, these polymers undergo condensation/polymerization in the post-photolysis period into mass $<$ 700 Da species that absorb in the visible, and are bleached upon resuming irradiation. The re- photolyzed solutions recover the mass and UV-absorption spectra of first photolyzed solutions. Whereas initial pH has no effect on the mechanism of reaction, ammonium bisulfate, a major component of the aerosol, markedly influences these processes. These findings suggest that the chemical identity and concentration levels of complex organic substances in the aerosol are the result of dynamic photochemical processing in the condensed phase.