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Chemical and physical changes of organic particles upon reaction with ozone

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Heterogeneous oxidation reactions are an important process in the aging of the organic fraction in atm
particles, specifically in affecting their hygroscopic properties and their activity as cloud condensatio
Our study provides information on the heterogeneous reactions of coated polystyrene latex (PSL) co
oleic acid layers (2-30 nm) and pure oleic acid droplets with ozone at low relative humidity. The co
phase products of the reacted coated particles are characterized in situ with an aerosol mass spectromete
and a scanning mobility particle sizer (SMPS). Attenuated total reflectance infrared spectroscopy (A
gas chromatography-mass spectrometry (GC-MS), and liquid chromatography-mass spectrometry (
are further used for product identification when studying pure oleic acid droplets.
The major products detected by the AMS are 9-oxononanoic acid (20-35%) and large (>C18) oxy
organics (35-50%) as a result of the reaction of Criegee intermediates with the oleic acid double bond
acid (1-3%) and nonanoic acid (1-3%) are minor products. The condensed-phase products detected
MS and LC-MS for pure oleic acid droplets are 1-nonanal (30%), 9-oxononanoic acid (14%), nonan
(7%), octanoic acid (1%), azelaic acid (6%), and unidentified products. FTIR results show that at least
the unidentified products contain an ester group. Additionally, the mass spectra show that at least son
unidentified products have high molecular weight (>1000 amu), which implicates a polymerization
Moreover, the observed steps of 172 amu (9-oxononanoic acid) and 188 amu (azelaic acid Criegee inter
in the mass spectra of high molecular weight species, provide evidence of these species as chain propa
The reactive uptake coefficient is determined for the coated particles. The apparent uptake coefficient d
with increasing layer thickness and tends towards 10-3. A change in the physical behavior is observed
8-10nm which may represent the diffuso-reactive length. Surface and bulk reactions may be distinguis
surface reaction is found to be much faster than the bulk reaction.
Our results indicate a change in oxidation state upon O3 reaction. An increase in the carbon-normalized
content (z/x) in the average chemical composition of the layer is observed from 0.1 for pure oleic aci
after high ozone exposure and is correlated with an increase in particle density. After reaction, the drop
up water at lower relative humidities compared to the unreacted droplets as shown by the environmen
ning electron microscopy (ESEM) study. The increased hygroscopic response may indicate that the c
aging of atmospheric organic aerosol particles has significant impact on radiative forcing.