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Heterogeneous OH oxidation of saturated organic matter in single component and internally mixed aerosol particles: evidence for significant volatilization

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Heterogeneous oxidation of particulate organic matter by the OH radical is a potentially important aging process that can affect particle hygroscopicity, cloud nucleating ability and health effects. This process has also been postulated to be a potentially significant source of volatile organic carbon in the upper troposphere with implications for HOx sources in that region. We present results from laboratory experiments utilizing both an online and a novel impaction-based chemical ionization mass spectrometry method to monitor aerosol composition as a function of OH exposure together with a scanning mobility particle sizing (SMPS) instrument to monitor particle size changes upon oxidation. Pure palmitic acid particles and NaCl particles coated with palmitic acid were used as surrogates for solid or viscous fresh primary organic particles and internal mixtures of aqueous inorganic and organic material. The mean size of pure palmitic acid particles was varied to help elucidate the mechanism. A reaction probability of ˜ 0.2 - 0.4 best explains the loss of palmitic acid from the internally mixed particles, and formic acid is a dominant reaction product from this system. For the pure palmitic acid particles, the observed loss of palmitic acid signal and particle size change is best represented by a surface-only oxidation model containing an OH reaction probability of unity, a surface renewal rate similar to the OH-surface collision frequency, and a particle mass loss rate due to volatilization equal to the palmitic acid loss rate. This degree of volatilization by OH oxidation is similar to that observed on self-assembled monolayers and suggests organic aerosols that are solid or viscous may be important sources of VOC in the upper troposphere.