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Heterogeneous oxidation of saturated organic aerosols by hydroxyl radicals: uptake kinetics, condensed-phase products, and particle size change

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Abstract. The kinetics and reaction mechanism for the heterogeneous oxidation of saturated organic aerosols by gas-phase OH radicals were investigated under NO_x-free conditions. The reaction of 150 nm diameter Bis(2-ethylhexyl) sebacate (BES) particles with OH was studied as a proxy for chemical aging of atmospheric aerosols containing saturated organic matter. An aerosol reactor flow tube combined with an Aerodyne time-of-flight aerosol mass spectrometer (ToF-AMS) and scanning mobility particle sizer (SMPS) was used to study this system. Hydroxyl radicals were produced by 254 nm photolysis of O₃ in the presence of water vapour. The kinetics of the heterogeneous oxidation of the BES particles was studied by monitoring the loss of a mass fragment of BES with the ToF-AMS as a function of OH exposure. We measured an initial OH uptake coefficient of $\gamma_0 = 1.3 (\pm 0.4)$, confirming that this reaction is highly efficient. The density of BES particles increased by up to 20% of the original BES particle density at the highest OH exposure studied, consistent with the particle becoming more oxidized. Electrospray ionization mass spectrometry analysis showed that the major particle-phase reaction products are multifunctional carbonyls and alcohols with higher molecular weights than the starting material. Volatilization of oxidation products accounted for a maximum of 17% decrease of the particle volume at the highest OH exposure studied. Tropospheric organic aerosols will become more oxidized from heterogeneous photochemical oxidation, which may affect not only their physical and chemical properties, but also their hygroscopicity and cloud nucleation activity.

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