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Reversible and irreversible processing of biogenic olefins on acidic aerosols

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Abstract. Recent evidence has suggested that heterogeneous chemistry of oxygenated hydrocarbons, primarily carbonyls, plays a role in the formation of secondary organic aerosol (SOA); however, evidence is emerging that direct uptake of alkenes on acidic aerosols does occur and can contribute to SOA formation. In the present study, significant uptake of monoterpenes, oxygenated monoterpenes and sesquiterpenes to acidic sulfate aerosols is found under various conditions in a reaction chamber. Proton transfer mass spectrometry is used to quantify the organic gases, while an aerosol mass spectrometer is used to quantify the organic mass uptake and obtain structural information for heterogeneous products. Aerosol mass spectra are consistent with several mechanisms including acid catalyzed olefin hydration, cationic polymerization and organic ether formation, while measurable decreases in the sulfate mass on a per particle basis suggest that the formation of organosulfate compounds is also likely. A portion of the heterogeneous reactions appears to be reversible, consistent with reversible olefin hydration reactions. A slow increase in the organic mass after a fast initial uptake is attributed to irreversible reactions, consistent with polymerization and organosulfate formation. Uptake coefficients (γ) were estimated for a fast initial uptake governed by the mass accommodation coefficient (α) and ranged from 1×10^{-6} – 2.5×10^{-2} . Uptake coefficients for a subsequent slower reactive uptake ranged from 1×10^{-7} – 1×10^{-4} . These processes may potentially lead to a considerable amount of SOA from the various biogenic hydrocarbons under acidic conditions, which can be highly significant for freshly nucleated aerosols, particularly given the large array of atmospheric olefins.

[Final Revised Paper](#) (PDF, 3807 KB) [Supplement](#) (121 KB) [Discussion Paper](#) (ACPD)

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