Supporting Information

Compositional Evolution of Particle Phase Reaction Products and Water in the Heterogeneous OH Oxidation of Aqueous Organic Droplets

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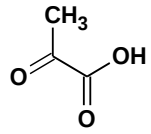
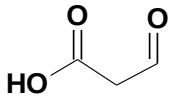
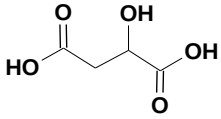
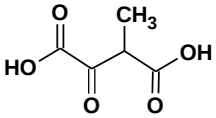
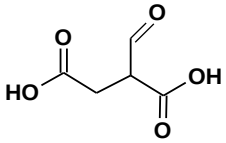
Table S1. Experimental data obtained from the hygroscopicity measurement.

Relative Humidity (%)	Refractive Index (650nm)	Refractive Index (589nm)	Mass fraction of solute, *mfs*
87.0	1.411	1.417	0.595
83.4	1.418	1.424	0.642
72.8	1.433	1.439	0.743
68.0	1.440	1.446	0.791
65.6	1.444	1.450	0.818
63.0	1.446	1.452	0.831
85.8	1.415	1.421	0.622
83.2	1.421	1.427	0.662
80.8	1.425	1.431	0.689
78.3	1.428	1.434	0.710
75.8	1.432	1.438	0.737
73.4	1.435	1.441	0.757
71.0	1.438	1.444	0.777
68.5	1.439	1.445	0.784
66.1	1.442	1.447	0.801
63.7	1.444	1.449	0.814
61.3	1.446	1.452	0.831
58.9	1.447	1.453	0.838
56.5	1.450	1.456	0.858
54.2	1.451	1.457	0.865
51.8	1.453	1.459	0.879
49.4	1.455	1.461	0.892
47.1	1.456	1.462	0.899
44.5	1.458	1.463	0.909
41.8	1.459	1.464	0.916
39.0	1.460	1.466	0.926
36.2	1.463	1.469	0.946
33.4	1.464	1.470	0.953
30.7	1.465	1.471	0.960
28.0	1.467	1.473	0.973
25.3	1.468	1.474	0.980

Table S2. Minor reaction products observed under the heterogeneous OH oxidation of aqueous methylsuccinic acid droplet. The relative abundance is reported at the maximum OH exposure (1.47×10^{12} molecule cm^{-3} s).

Chemical Formula	Molecular Weight	Relative Abundance (%)	Proposed Chemical Structure	
$C_3H_4O_3$	87	1.1	 1st generation fragmentation product (Scheme 1, Path A)	 1st generation fragmentation product (Scheme 1, Path C)
$C_3H_4O_4$	103	1.3	Possible 2nd generation product	
$C_4H_4O_4$	115	1.1	Possible 2nd generation product	
$C_4H_6O_4$	117	1.1	Possible 2nd generation product	
$C_4H_6O_5$	133	1.8	 1st generation fragmentation product (Scheme 1, Path C)	
$C_5H_6O_5$	145	1.1	 1st generation ketone functionalization product (Scheme 1, Path B)	 1st generation ketone functionalization product (Scheme 1, Path C)
$C_5H_8O_6$	163	1.0	Possible 2nd generation product	

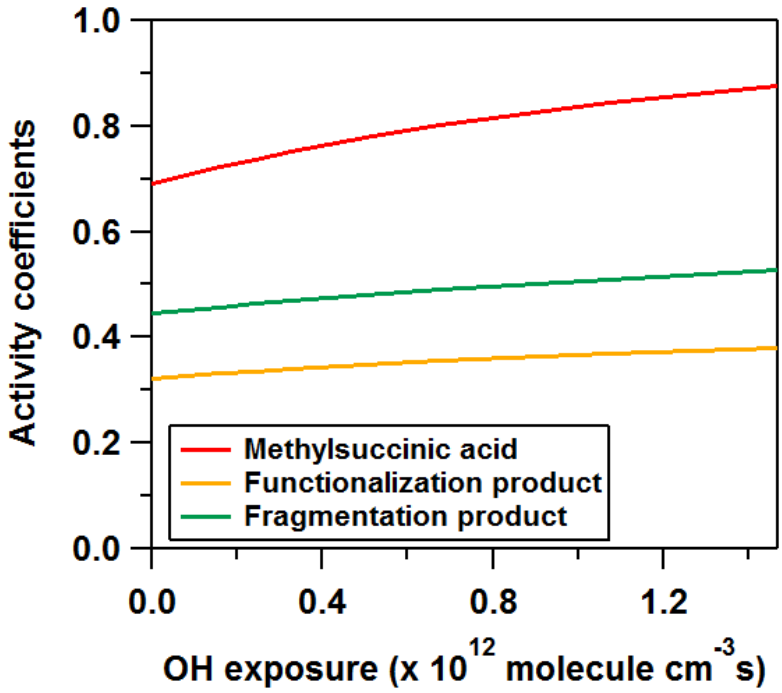


Figure S1. Simulated change of activity coefficients (γ_i) of parent ($C_5H_8O_4$), major functionalization ($C_5H_8O_5$) and major fragmentation ($C_4H_6O_3$) products (listed in Scheme 2) estimated by the AIOMFAC model. It is noted that no reaction products are formed prior to oxidation (i.e. OH exposure of 0 molecule cm^{-3} s). The results show the simulated activity coefficients of the two reaction products once the oxidation is initiated.