Supplementary Material

Chemical Insights, Explicit Chemistry and Yields of Secondary Organic Aerosol from OH Radical Oxidation of Methylglyoxal and Glyoxal in the Aqueous Phase

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The supporting information contains 11 pages with following information: the chemical model (Table S1); the simulated concentration of dissolved oxygen during an experiment (Fig. S1); atmospheric CSTR simulated SOA yields (Fig. S2); oligomer distributions for simulated SOA yields (Fig. S3).

Table S1. Reactions and rate/equilibrium constants used in the full kinetic model of unified glyoxal/methylglyoxal + OH

	Reactions	**Rate constants (M^{1-n} s^{-1})**	**Ref**
1	$H_2O_2 \rightarrow 2OH$	1.1e-4×Trans[a]	T, e
2	$OH + H_2O_2 \rightarrow HO_2 + H_2O$	2.7e7	T
3	$HO_2 + H_2O_2 \rightarrow OH + H_2O + O_2$	3.7	T
4	$2\ HO_2 \rightarrow H_2O_2 + O_2$	8.3e5	T
5	$OH + HO_2 \rightarrow H_2O + O_2$	7.1e9	T
6	$HO_2 + O_2^- + H^+ \rightarrow H_2O_2 + O_2$	1e8	T
7	$2OH \rightarrow H_2O_2$	5.5e9	T
8	$OH + O_2^- \rightarrow OH^- + O_2$	1e10	T
9	$O_{2g} \leftrightarrow O_2$	K_{eq} = 1.3e-3 k_r = 5.3e2	T, W
10	$CO_{2g} \leftrightarrow CO_2$	K_{eq} = 3.4e-2 k_r = 5.3e2	T, W
11	$CO_2 \leftrightarrow H^+ + HCO_3^-$	K_{eq} = 4.3e-7 k_r = 5.6e4	T
12	$HCO_3^- \rightarrow H^+ + CO_3^{-2}$	K_{eq} = 4.69e-11 k_r = 5.0e10	T
13	$CO_2^- + O_2 \rightarrow O_2^- + CO_2$	2.4e9	T
14	$HCO_3^- + OH \rightarrow CO_3^- + H_2O$	1e7	T
15	$CO_3^- + O_2^- \rightarrow CO_3^{-2} + O_2$	6.5e8	T
16	$CO_3^- + HCO_2^- \rightarrow HCO_3^- + CO_2^-$	1.5e5	T
17	$CO_3^- + H_2O_2 \rightarrow HCO_3^- + HO_2$	8e5	T
18	GCOLAC + OH → GCOLAC* + H2O	6.0e8	T
19	GCOLAC* + O_2 → GCOLACOO*	1e6	G, L’
20	GCOLACOO* → GLYAC + HO_2	5e1	C
21	2GCOLACOO* → 2GCOLACO* + O_2	3e8*0.95	L’, e
22	2GCOLACOO* → GLYAC + OXLAC + O_2	3e8*0.05	L’, e
23	GCOLACO* → HCO_2H + CO_2	I	Gi, e
24	GCOLACO* → GLYAC*	1e7	Gi, e
25	GCOLAC ↔ H^+ + GCOLAC$^-$	K_{eq} = 1.48e-4 k_r = 2.0e10	T
26	GCOLAC$^-$ + OH → GCOLAC*$^-$ + H_2O	6.0e8	T
27	GCOLAC*$^-$ + O_2 → GCOLACOO*$^-$	1e6	G, L’
28	GCOLACOO*$^-$ → GLYAC$^-$ + HO_2	5e1	C
29	2GCOLACROO*$^-$ → 2GCOLACO*$^-$ + O_2	3e8×0.95	L’, e
30	2 GCOLACROO*$^-$ → GLYAC$^-$ + OXLAC$^-$+ O_2	3e8×0.05	L’, e
31	GCOLACO*$^-$ → HCO_2H + CO_2^-	I	Gi, e
32	GCOLACO*$^-$→ GLYAC*$^-$	1e7	Gi, e
33	GLY + OH → GLY* + H_2O	1.1e9	T
34	GLY* + O_2 → GLYOO*	1e6	G, L’
35	GLYOO* → GLYAC + HO_2	5e1	C
36	2GLYOO* → 2*CHOHOH + 2CO_2 + O_2 + 2H_2O	3e8	L’
37	*CHOHOH + O_2 → HCO_2H + HO_2	5e6	G, L’
38	GLYAC + OH → GLYAC* + H_2O	3.62e8	T
39	GLYAC* + O_2 → GLYACOO*	1e6	G, L’
40	GLYACOO* → OXLAC + HO_2	5e1	C

41	$2GLYACOO^* \rightarrow 2CO_2 + 2COOH$	3e8	L’
42	$^*COOH + O_2 \rightarrow CO_2 + HO_2$	5e6	G, L’
43	$GLYAC \leftrightarrow H^+ + GLYAC^-$	K_{eq} = 3.47e-4 K_r = 2.0e10	T
44	$GLYAC^- + OH \rightarrow GLYAC^{*-} + H_2O$	1.28e7	T
45	$GLYAC^- + OH \rightarrow GLYAC^* + OH^-$	2.9e9	T
46	$GLYAC^{*-} + O_2 \rightarrow GLYACOO^{*-}$	1e6	G, L’
47	$GLYACOO^{*-} \rightarrow OXLAC^- + HO_2$	1e2	C, L’
48	$2GLYACOO^{*-} \rightarrow 2CO_2^- + 2^*COOH$	3e8	L’
49	$MGLY + OH \rightarrow MGLY^* + H_2O$	7.0e8×0.92	T
50	$MGLY + OH \rightarrow {}^*MGLY + H_2O$	7.0e8×0.08	T
51	$MGLY^* + O_2 \rightarrow MGLYOO^*$	1e6	G, L’
52	$MGLYOO^* \rightarrow PYRAC + HO_2$	5e1	C
53	$2MGLYOO^* \rightarrow 2CO_2 + 2CH_3CO_2H + O_2$	3e8	L’
54	$^*MGLY + O_2 \rightarrow {}^*OOMGLY$	1e6	G, L’
55	$2^*OOMGLY \rightarrow 2^*OMGLY + O_2$	3e8×0.95	L’, e
56	$2^*OOMGLY \rightarrow HOMGLY + OMGLY + O_2$	3e8×0.05	L’, e
57	$^*OMGLY \rightarrow HCHO + GLY^*$	I	Gi, e
58	$^*OMGLY \rightarrow {}^*HOMGLY$	1e7	Gi, e
59	$HOMGLY + OH \rightarrow {}^*HOMGLY + H_2O$	4.10e7	M
60	$^*HOMGLY + O_2 \rightarrow {}^*OOHOMGLY$	1e6	G, L’
61	$^*OOHOMGLY \rightarrow OMGLY + HO_2$	5e1	C
62	$OMGLY + OH \rightarrow {}^*OMGLY + H_2O$	6.17e9	M
63	$^*OMGLY + O_2 \rightarrow {}^*OOOMGLY$	5e1	C
64	$GLY^* + {}^*CHOHOH \rightarrow C3D$	1.3e9	G, L’
65	$2GLY^* \rightarrow C4D$	1.3e9	G, L’
66	$GLY^* + {}^*COOH \rightarrow C3D$	1.3e9	G, L’
67	$GLYAC^* + {}^*COOH \rightarrow C3D$	1.3e9	G, L’
68	$GLYAC^* + {}^*CHOHOH \rightarrow C3D$	1.3e9	G, L’
69	$2GLYAC^* \rightarrow C4D$	1.3e9	G, L’
70	$GLYAC^* + GLY^* \rightarrow C4D$	1.3e9	G, L’
71	$GLYAC^{*-} + GLY^* \rightarrow C4D$	1.3e9	G, L’
72	$GLYAC^{*-} + GLYAC^* \rightarrow C4D$	1.3e9	G, L’
73	$2GLYAC^{*-} \rightarrow C4D$	1.3e9	G, L’
74	$GLYAC^{*-} + {}^*COOH \rightarrow C3D$	1.3e9	G, L’
75	$GLYAC^{*-} + {}^*CHOHOH \rightarrow C3D$	1.3e9	G, L’
76	$GLYCOL^{*1} + {}^*CHOHOH \rightarrow C3D$	1.3e9	G, L’
77	$GLYCOL^{*1} + GLY^* \rightarrow C4D$	1.3e9	G, L’
78	$GLYCOL^{*1} + {}^*COOH \rightarrow C3D$	1.3e9	G, L’
79	$GLYCOL^{*1} + GLYAC^* \rightarrow C4D$	1.3e9	G, L’
80	$GLYCOL^{*1} + GLYAC^{*-} \rightarrow C4D$	1.3e9	G, L’
81	$GLYCOL^{*2} + {}^*CHOHOH \rightarrow C3D$	1.3e9	G, L’
82	$GLYCOL^{*2} + GLY^* \rightarrow C4D$	1.3e9	G, L’
83	$GLYCOL^{*2} + {}^*COOH \rightarrow C3D$	1.3e9	G, L’
84	$GLYCOL^{*2} + GLYAC^* \rightarrow C4D$	1.3e9	G, L’
85	$GCOLAC^* + {}^*CHOHOH \rightarrow C3D$	1.3e9	G, L’
86	$GCOLAC^* + GLY^* \rightarrow C4D$	1.3e9	G, L’
87	$GCOLAC^* + {}^*COOH \rightarrow C3D$	1.3e9	G, L’
88	$GCOLAC^* + GLYAC^* \rightarrow C4D$	1.3e9	G, L’
89	$GCOLAC^* + GLYAC^{*-} \rightarrow C4D$	1.3e9	G, L’

90	GCOLAC* + GLYCOL*^{1} $\rightarrow$ C4D	1.3e9	G, L’
91	GCOLAC* + GLYCOL*^{2} $\rightarrow$ C4D	1.3e9	G, L’
92	GCOLAC* + GCOLAC* $\rightarrow$ C4D	1.3e9	G, L’
93	GCOLAC*$^{-}$ + *CHOHOH $\rightarrow$ C3D	1.3e9	G, L’
94	GCOLAC*$^{-}$ + GLY* $\rightarrow$ C4D	1.3e9	G, L’
95	GCOLAC*$^{-}$ + *COOH $\rightarrow$ C3D	1.3e9	G, L’
96	GCOLAC*$^{-}$ + GLYAC* $\rightarrow$ C4D	1.3e9	G, L’
97	GCOLAC*$^{-}$ + GLYAC*$^{-}$ $\rightarrow$ C4D	1.3e9	G, L’
98	GCOLAC*$^{-}$ + GLYAC*$^{-}$ $\rightarrow$ C4D	1.3e9	G, L’
99	GCOLAC*$^{-}$ + GLYCOL*^{1} $\rightarrow$ C4D	1.3e9	G, L’
100	GCOLAC*$^{-}$ + GLYCOL*^{2} $\rightarrow$ C4D	1.3e9	G, L’
101	GCOLAC*$^{-}$ + GCOLAC* $\rightarrow$ C4D	1.3e9	G, L’
102	2 GCOLAC*$^{-}$ $\rightarrow$ C4D	1.3e9	G, L’
103	2MGLY* $\rightarrow$ C6D	1.3e9	G, L’
104	MGLY* + *CHOHOH $\rightarrow$ C4D	1.3e9	G, L’
105	MGLY* + GLY* $\rightarrow$ C5D	1.3e9	G, L’
106	MGLY* + *COOH $\rightarrow$ C4D	1.3e9	G, L’
107	MGLY* + GLYAC* $\rightarrow$ C5D	1.3e9	G, L’
108	MGLY* + GLYAC*$^{-}$ $\rightarrow$ C5D	1.3e9	G, L’
109	MGLY* + GLYCOL*^{1} $\rightarrow$ C5D	1.3e9	G, L’
110	MGLY* + GLYCOL*^{2} $\rightarrow$ C5D	1.3e9	G, L’
111	MGLY* + GCOLAC* $\rightarrow$ C5D	1.3e9	G, L’
112	MGLY* + GCOLAC*$^{-}$ $\rightarrow$ C5D	1.3e9	G, L’
113	MGLY* + CH_3CO* $\rightarrow$ C5D	1.3e9	G, L’
114	MGLY* + *HOPYRAC $\rightarrow$ C6D	1.3e9	G, L’
115	2*HOPYRAC $\rightarrow$ C6D	1.3e9	G, L’
116	MGLY* + *HOPYRAC^{-} $\rightarrow$ C6D	1.3e9	G, L’
117	*HOPYRAC^{-} + *HOPYRAC^{-} $\rightarrow$ C6D	1.3e9	G, L’
118	*HOPYRAC + *HOPYRAC^{-} $\rightarrow$ C6D	1.3e9	G, L’
119	CH_3CO* + *HOPYRAC $\rightarrow$ C6D	1.3e9	G, L’
120	CH_3CO* + *HOPYRAC^{-} $\rightarrow$ C6D	1.3e9	G, L’
121	2LA* $\rightarrow$ C6D	1.3e9	G, L’
122	LA* + MGLY* $\rightarrow$ C6D	1.3e9	G, L’
123	LA* + *CHOHOH $\rightarrow$ C4D	1.3e9	G, L’
124	LA* + GLY* $\rightarrow$ C5D	1.3e9	G, L’
125	LA* + *COOH $\rightarrow$ C4D	1.3e9	G, L’
126	LA* + GLYAC* $\rightarrow$ C5D	1.3e9	G, L’
127	LA* + GLYAC*$^{-}$ $\rightarrow$ C5D	1.3e9	G, L’
128	LA* + GLYCOL*^{1} $\rightarrow$ C5D	1.3e9	G, L’
129	LA* + GLYCOL*^{2} $\rightarrow$ C5D	1.3e9	G, L’
130	LA* + GCOLAC* $\rightarrow$ C5D	1.3e9	G, L’
131	LA* + GCOLAC*$^{-}$ $\rightarrow$ C5D	1.3e9	G, L’
132	LA* + CH_3CO* $\rightarrow$ C5D	1.3e9	G, L’
133	2CH_3CO* $\rightarrow$ C4D	1.3e9	G, L’
134	LA* + *HOPYRAC $\rightarrow$ C6D	1.3e9	G, L’
135	LA* + *HOPYRAC^{-} $\rightarrow$ C6D	1.3e9	G, L’
136	OXLAC + OH $\rightarrow$ COOH + CO_2 + H_2O	1.4e6	T
137	OXLAC $\leftrightarrow$ H^{+} + OXLAC^{-}	K_{eq} = 5.67e-2 k_r = 5.0e10	T
138	OXLAC^{-} + OH $\rightarrow$ COOH + CO_2^{-} + H_2O	2.0e7	T, L’

139	$OXLAC^- \leftrightarrow H^+ + OXLAC^{-2}$	K_{eq} = 5.42e-5 k_r = 5e10	T
140	$OXLAC^{-2} + OH \rightarrow *COOH + CO_2^- + OH^-$	4.0e7	T, L’
141	$LA + OH \rightarrow LA* + H_2O$	4.3e8	H
142	$LA* + O_2 \rightarrow LAOO*$	1e6	G, L’
143	$LAROO* \rightarrow PYRAC + HO_2$	5e1	C
144	$LA \leftrightarrow LA^- + H^+$	K_{eq} = 1.38e-4 k_r = 5.0e10	E&C
145	$LA^- + OH \rightarrow LA*^- + H_2O$	3e8	B
146	$LA*^- + O_2 \rightarrow LAOO*^-$	1e6	G, L’
147	$LAOO*^- \rightarrow PYRAC^- + HO_2$	5e1	C
148	$PYRAC + OH \rightarrow PYRAC* + H_2O$	6.0e7×0.85	T
149	$PYRAC + OH \rightarrow CH_3CO* + CO_2 + H_2O$	6.0e7×0.15	T
150	$CH_3CO* + O_2 \rightarrow CH_3C(O)OO*$	1e6	G, L’
151	$CH3C(O)OO* \rightarrow CH_3CO_2H + HO_2$	5e1	C
152	$2CH3C(O)OO* \rightarrow 2CH_3C(O)O* + O_2$	3e8	L’
153	$CH_3C(O)O* \rightarrow CO_2 + HCHO$	1e7	Gi
154	$PYRAC* + O_2 \rightarrow PYRACOO*$	1e6	G, L’
144	$2PYRACOO* \rightarrow 2PYRACO* + O_2$	3e8×0.95	L’, e
145	$2PYRACOO* \rightarrow HOPYRAC + OPYRAC + O_2$	3e8×0.15	L’, e
146	$PYRACO* \rightarrow HCHO + GLYAC*$	I	Gi, e
147	$PYRACO* \rightarrow *HOPYRAC$	1e7	Gi, e
148	$HOPYRAC + OH \rightarrow *HOPYRAC + H_2O$	3.6e8	H
149	$*HOPYRAC + O_2 \rightarrow *OOHOPYRAC$	1e6	G, L’
150	$*OOHOPYRAC \rightarrow OPYRAC + HO_2$	5e1	C
151	$OPYRAC + OH \rightarrow *OPYRAC + H_2O$	5e7	e
152	$*OPYRAC + O_2 \rightarrow *OO(O)PYRAC$	1e6	G, L’
153	$*OO(O)PYRAC \rightarrow MOXLAC + HO_2$	5e1	C
154	$PYRAC \leftrightarrow PYRAC^- + H^+$	K_{eq} = 3.2e-3 k_r = 2e10	T
155	$PYRAC^- + OH \rightarrow PYRAC*^- + H_2O$	6.0e7×0.95	T
156	$PYRAC^- + OH \rightarrow CH3CO* + CO_2 + OH^-$	6.0e7×0.05	T
157	$PYRAC*^- + O_2 \rightarrow PYRACOO*^-$	5e1	C
158	$2PYRACOO*^- \rightarrow 2PYRACO*^- + O_2$	3e8×0.95	L’, e
159	$2PYRACOO*^- \rightarrow HOPYRAC^- + OPYRAC^- + O_2$	3e8×0.05	L’, e
160	$PYRACO*^- \rightarrow HCHO + GLYAC*^- + O_2$	I	Gi, e
161	$PYRACO*^- \rightarrow *HOPYRAC^-$	1e7	Gi, e
162	$HOPYRAC \leftrightarrow HOPYRAC^- + H^+$	K_{eq} = 3.2e-3 k_r = 2e10	e
163	$OPYRAC \leftrightarrow OPYRAC^- + H^+$	K_{eq} = 3.2e-3 k_r = 2e10	e
164	$HOPYRAC^- + OH \rightarrow *HOPYRAC^- + H_2O$	2.6e9	H
165	$*HOPYRAC^- + O_2 \rightarrow *OOHOPYRAC^-$	1e6	G, L’
166	$*OOHOPYRAC^- \rightarrow OPYRAC^- + HO_2$	5e1	C
167	$OPYRAC^- + OH \rightarrow *OPYRAC^- + H_2O$	5e7	M
168	$*OPYRAC^- + O2 \rightarrow *OO(O)PYRAC^-$	1e6	G, L’
169	$*OO(O)PYRAC^- \rightarrow MOXLAC^- + HO_2$	5e1	C
170	$MOXLAC + OH \rightarrow GLYAC* + CO_2 + H_2O$	5.7e7	Gl
171	$MOXLAC^- + OH \rightarrow GLYAC*^- + CO_2 + H_2O$	7.85e7	e
172	$MOXLAC^{-2} + OH \rightarrow GLYAC*^- + CO_2 + OH^-$	1.0e8	H

173	MOXLAC $\leftrightarrow$ $MOXLAC^-$ + H^+	K_{eq} = 3.16e-3 k_r = 5e10	H
174	$MOXLAC^-$ $\leftrightarrow$ $MOXLAC^{-2}$ + H^+	K_{eq} = 1.5e-2 k_r = 5e10	V
175	CH_3CO_2H + OH $\rightarrow$ $*CH_2CO_2H$ + H_2O	1.36e7	T
176	CH_3CO_2H + OH $\rightarrow$ CO_2 + HCHO + HO_2 + H_2O	2.40e6	T
177	$*CH_2CO_2H$ + O_2 $\rightarrow$ $*OOCH_2CO_2H$	1e6	G, L’
178	$2*OOCH_2CO_2H$ $\rightarrow$ $2*OCH_2CO_2H$ + O_2	3e8*0.95	L’, e
179	$2*OOCH_2CO_2H$ $\rightarrow$ GLYAC + GCOLAC + O_2	3e8*0.05	L’, e
180	$*OCH_2CO_2H$ $\rightarrow$ $2CO_2$ + 2HCHO	I	Gi, e
181	$*OCH_2CO_2H$ $\rightarrow$ GCOLAC*	1e7	Gi, e
182	CH_3CO_2H $\leftrightarrow$ $CH_3CO_2^-$ + H^+	K_{eq} = 1.75e-5 k_r = 5.0e10	T
183	$CH_3CO_2^-$ + OH $\rightarrow$ $*CH_2CO_2^-$ + H_2O	7.23e7	T
184	$CH_2CO_2^-$ + OH $\rightarrow$ CO_2 + HCHO + HO_2 + OH^-	1.28e7	T
185	$*CH_2CO_2^-$ + O_2 $\rightarrow$ $*OOCH_2CO_2^-$	1e6	G, L’
186	$2*OOCH_2CO_2n1$ $\rightarrow$ $2*OCH_2CO_2$ + O_2	3e8×0.95	L’, e
187	$2*OOCH_2CO_2^-$ $\rightarrow$ $GLYAC^-$ +$GCOLAC^-$+O2	3e8×0.05	L’, e
188	$*OCH_2CO_2^-$ $\rightarrow$ $2CO_2^-$ + 2HCHO	I	Gi, e
189	$*OCH_2CO_2^-$ $\rightarrow$ $GCOLAC*^-$	1e7	Gi, e
190	H_2O $\leftrightarrow$ H^+ + OH^-	K_{eq} = 1.0e-14 k_r = 1.4e11	T
191	HO_2 $\leftrightarrow$ H^+ + O_2^-	K_{eq} = 1.6e-5 k_r = 5.0e10	T
192	HCO_2H + OH $\rightarrow$ *COOH + H_2O	1e8	T
193	HCO_2^- + OH $\rightarrow$ CO_2^- + H_2O	2.4e9	T
194	HCO_2H $\leftrightarrow$ H^+ + HCO_2^-	K_{eq} = 1.77e-4 k_r = 5.0e10	T
195	GLYAC + H_2O_2 $\rightarrow$ HCO_2H + CO_2 + H_2O	0.3	T
196	PYRAC + H_2O_2 $\rightarrow$ CH_2CO_2H + H_2O + CO_2	0.11	T
197	$PYRAC^-$ + H_2O_2 $\rightarrow$ $CH_2CO_2^-$ + H_2O + CO_2	0.11	T
198	MOXLAC + H_2O_2 $\rightarrow$ OXLAC + CO_2 + H_2O	0.5	T
199	$MOXLAC^-$ + H_2O_2 $\rightarrow$ $OXLAC^-$ + CO_2 + H_2O	0.5	T
200	HCO_2H + OH $\rightarrow$ COOH + H_2O	1e8	T
201	HCO_2^- + OH $\rightarrow$ CO_2^- + H_2O	2.4e9	T
202	HCO_2H $\leftrightarrow$ H^+ + HCO_2^-	K_{eq} = 1.77e-4 k_r = 5.0e10	T
203	2*CHOHOH $\rightarrow$ GLY	1.3e9	G, L’
204	*CHOHOH + *COOH $\rightarrow$ GLYAC	1.3e9	G, L’
205	2*COOH $\rightarrow$ OXLAC	1.3e9	G, L’
206	C3D $\leftrightarrow$ MA + H2O	K_{eq} = 1e5 k_r = 1e-8	L’
207	2*COOH $\rightarrow$ OXLAC	1.3e9	G, L’
208	CO_2^- + *COOH $\rightarrow$ $OXLAC^-$	1.3e9	G, L’
209	$2CO_2^-$ $\rightarrow$ $OXLAC^{-2}$	1.3e9	G, L’

[a]Trans = Transmittance = $10^{-18.4 \times 0.80 \times [H_2O_2]}$; * = radical (e.g., glyoxal* = glyoxal radical); $*^n$ = radical type n (e.g., $GLYCOLAC*^1$ = glycolic acid radical type 1); O* (or *O) = alkoxy radical ; OO* (or *OO) = peroxy radical; CnD = C_n dimer or carboxylic acid oligomer (e.g., C4D = C_4 dimer or C_4 carboxylic acid oligomer); X_g = X in the gas phase (e.g., O_{2g} = O_2 in the gas phase); MGLY = methylglyoxal, PYRAC = pyruvic acid, GLYAC = glyoxylic acid, GLYCOL = glycolaldehyde, GLYCOLAC = glycolic acid, LA = lactic acid, MOXLAC = mesoxalic acid, OXLAC = oxalic acid; n = n^{th} order; K_{eq} = the equilibrium

constant (M), k_r = the reverse rate constant for corresponding K_{eq}., Thus, the forward rate constant can be calculated by $K_{eq} \times k_r$; (g) = in the gas phase; I (= the decomposition rate constant from alkoxy radicals) = 5e6 s^{-1} for ~10 μM acetic acid/methylglyoxal, 8e6 s^{-1} for ~10^2 μM acetic acid/methylglyoxal, and 2e7 s^{-1} for ~10^3 μM acetic acid/ 3.2e7 s^{-1} for ~10^3 μM methylglyoxal.

Reference
T = Tan et al., 2009, 2010 and 2012
G = Guzman et al., JPCA, 2006
C = Carter et al., JPC, 1979
H = Herrmann et al., AE, 2005
E = Ervens et al., PCCP, 2003
M = Monod et al., AE, 2005, 2008
L = Lim et al.,EST, 2005
L' = Lim et al., ACP, 2010
W = Warneck, PCCP, 1999
E&C = Eyal and Canari, Ind. Eng. Chem. Res., 1995
B = Buxton et al., JPCRD, 1988
Gi = Gilbert et al., 1976 and 1981
V = Volgger et al., J. Chrom. A, 1997
e = Estimation by fitting

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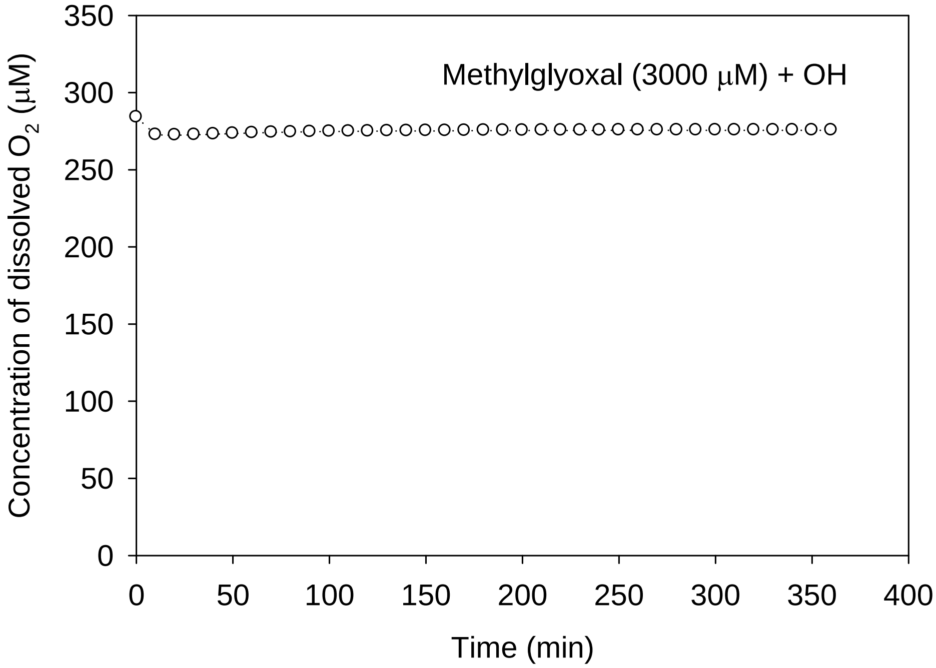

Figure S1. The simulated concentration of dissolved O_2 during the reaction of methylglyoxal (3000 μM) + OH

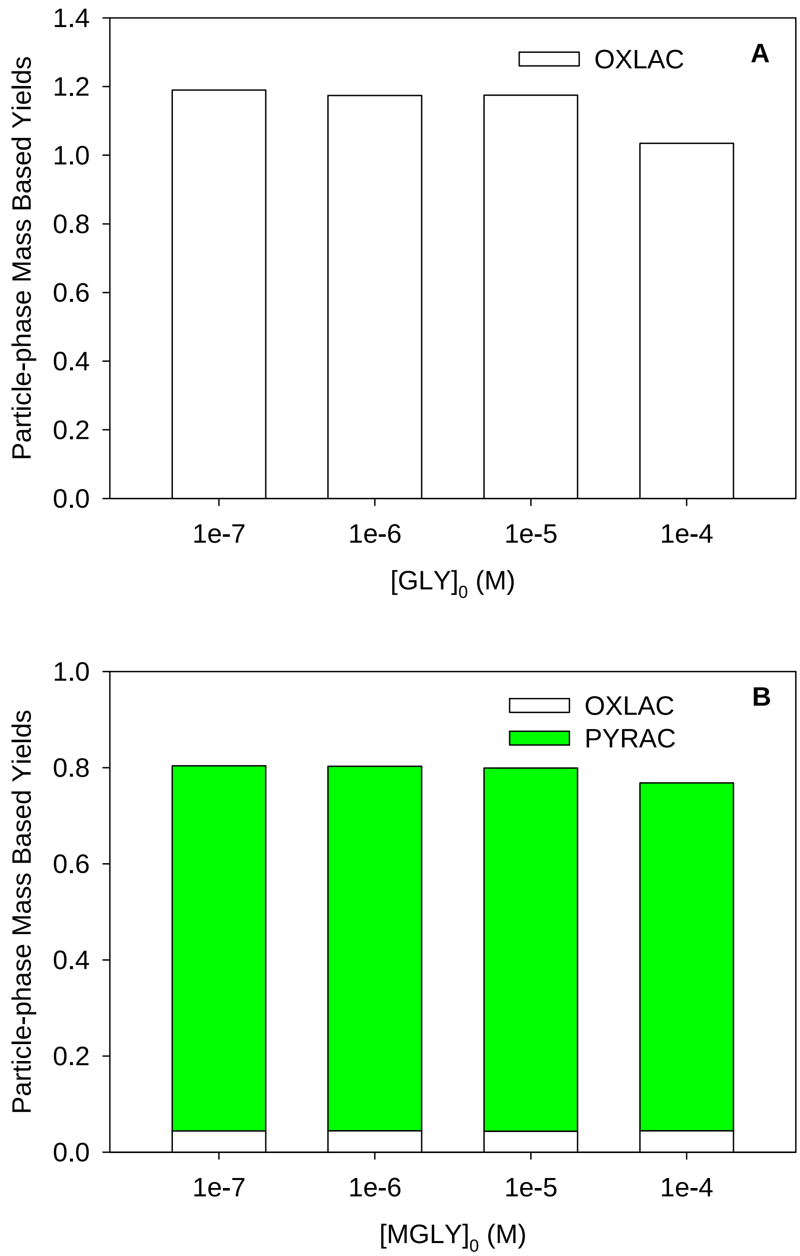

Figure S2. Atmospheric CSTR simulations **(A)** for particle-phase mass yields of oxalate (Y_{OXLAC}) with increasing initial concentrations of glyoxal ($[GLY]_0$) for aqueous-phase OH radical reactions (Y_{OXLAC} = 1.19/(1+1450$[GLY]_0$); Y_{SOA}(GLY)= Y_{OXLAC}), and **(B)** for particle-phase mass yields of oxalate (Y_{OXLAC}) and pyruvate (Y_{PYRAC}) with increasing initial concentrations of methylglyoxal ($[MGLY]_0$) for aqueous-phase OH radical reactions(Y_{PYRAC} = 0.759/(1+495$[MGLY]_0$); Y_{OXLAC} = 0.0439/(1-127$[MGLY]_0$); Y_{SOA}(MGLY) = Y_{PYRAC} + Y_{OXLAC}).

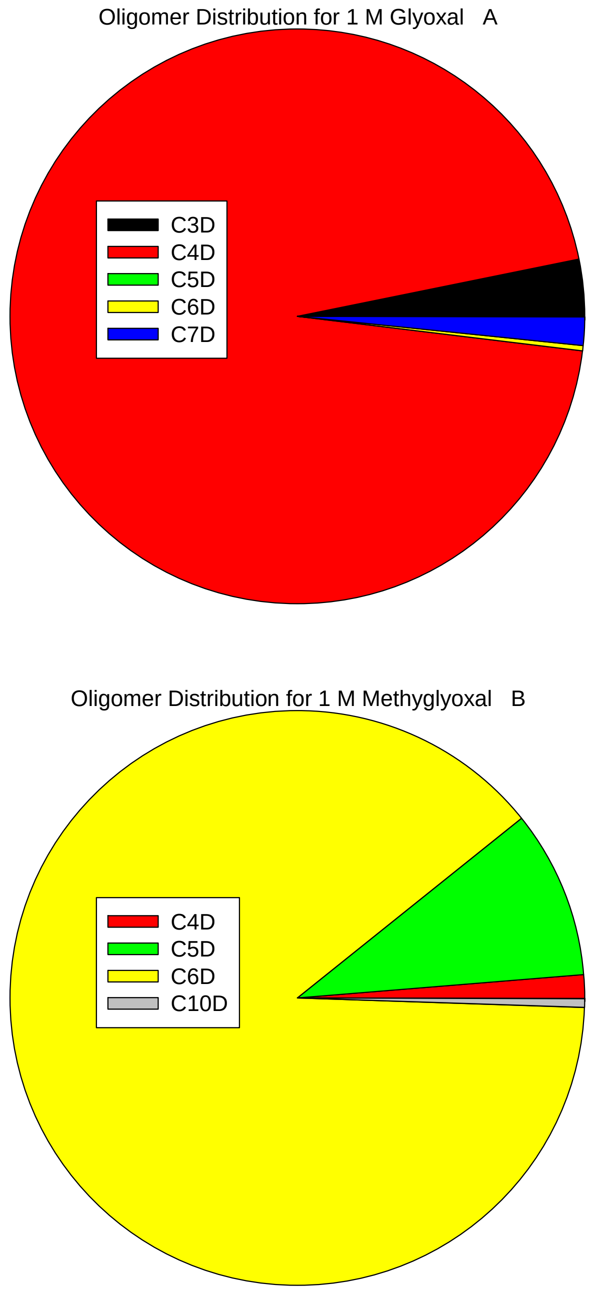

Figure S3. Oligomer distributions for 1 M glyoxal (A) and 1 M methylglyoxal (CnD = C_n dimer or carboxylic acid oligomer)