Data Availability for the paper "Photolytic Radical Persistence due to Anoxia in Viscous Aerosol Particles"

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Figure 1

STXM images used to generate 2-D profiles shown in Fig 1 are indicated below. All .hdf5 file structure can be read following Watts and Raabe[1]. Publically available Matlab software for data analysis can be found in previous work[2,3].

X-ray images prior to UV irradiation at RH = 40%.
Sample_Stack_2017-03-31_057.hdf5
Sample_Stack_2017-03-31_058.hdf5
Sample_Stack_2017-03-31_059.hdf5
Sample_Stack_2017-03-31_060.hdf5
Sample_Stack_2017-03-31_061.hdf5
Sample_Stack_2017-03-31_095.hdf5

X-ray images after UV irradiation at RH = 40%. The time after irradiation stopped is indicated.
Sample_Stack_2017-03-31_105.hdf5, *t* = 13 min
Sample_Stack_2017-03-31_106.hdf5, *t* = 23 min
Sample_Stack_2017-03-31_107.hdf5, *t* = 33 min
Sample_Stack_2017-03-31_108.hdf5, *t* = 42 min
Sample_Stack_2017-03-31_109.hdf5, *t* = 52 min
Sample_Stack_2017-03-31_110.hdf5, *t* = 61 min
Sample_Stack_2017-03-31_111.hdf5, *t* = 71 min
Sample_Stack_2017-03-31_112.hdf5, *t* = 80 min

Sample_Stack_2017-03-31_113.hdf5, t = 92 min

X-ray images prior to UV irradiation at RH = 50%.
Sample_Stack_2017-04-11_186
Sample_Stack_2017-04-11_192
Sample_Stack_2017-04-12_007
Sample_Stack_2017-04-12_014
Sample_Stack_2017-04-12_018
Sample_Stack_2017-04-12_019
Sample_Stack_2017-04-12_024

X-ray images after UV irradiation at RH = 50%.
Sample_Stack_2017-04-12_034.hdf5, t = 8 min
Sample_Stack_2017-04-12_035.hdf5, t = 14 min
Sample_Stack_2017-04-12_039.hdf5, t = 26 min
Sample_Stack_2017-04-12_040.hdf5, t = 32 min
Sample_Stack_2017-04-12_041.hdf5, t = 37 min
Sample_Stack_2017-04-12_047.hdf5, t = 50 min
Sample_Stack_2017-04-12_048.hdf5, t = 58 min
Sample_Stack_2017-04-12_049.hdf5, t = 65 min
Sample_Stack_2017-04-12_053.hdf5, t = 79 min
Sample_Stack_2017-04-12_054.hdf5, t = 88 min
Sample_Stack_2017-04-12_055.hdf5, t = 93 min
Sample_Stack_2017-04-12_056.hdf5, t = 102 min
Sample_Stack_2017-04-12_058.hdf5, t = 119 min
Sample_Stack_2017-04-12_059.hdf5, t = 128 min
Sample_Stack_2017-04-12_060.hdf5, t = 138 min
Sample_Stack_2017-04-12_061.hdf5, t = 146 min
Sample_Stack_2017-04-12_064.hdf5, t = 163 min
Sample_Stack_2017-04-12_065.hdf5, t = 173 min
Sample_Stack_2017-04-12_066.hdf5, t = 184 min
Sample_Stack_2017-04-12_067.hdf5, t = 192 min
Sample_Stack_2017-04-12_073.hdf5, t = 208 min
Sample_Stack_2017-04-12_074.hdf5, t = 217 min

X-ray images prior to UV irradiation at RH = 60%.
Sample_Stack_2017-04-11_080.hdf5
Sample_Stack_2017-04-11_082.hdf5
Sample_Stack_2017-04-11_083.hdf5
Sample_Stack_2017-04-11_097.hdf5

X-ray images after UV irradiation at RH = 60%.
Sample_Stack_2017-04-11_112.hdf5, t = 7 min
Sample_Stack_2017-04-11_113.hdf5, t = 15 min
Sample_Stack_2017-04-11_114.hdf5, t = 23 min

Sample_Stack_2017-04-11_115.hdf5, *t* = 29 min
Sample_Stack_2017-04-11_117.hdf5, *t* = 43 min
Sample_Stack_2017-04-11_118.hdf5, *t* = 61 min
Sample_Stack_2017-04-11_120.hdf5, *t* = 72 min
Sample_Stack_2017-04-11_122.hdf5, *t* = 87 min
Sample_Stack_2017-04-11_123.hdf5, *t* = 97 min
Sample_Stack_2017-04-11_124.hdf5, *t* = 107 min
Sample_Stack_2017-04-11_126.hdf5, *t* = 112 min

Figure 2

A data list for EDB experiments is below included in Fig. 2A. The error is r_0 is estimated from a change in the refractive index.

***RH* = 16.5 ± 2 %, r_0 = 7.94 ± 0.794 µm**

t / min	*r*(*t*)/r_0	+Δ *r*(*t*)/r_0	-Δ *r*(*t*)/r_0
0	1.00E+00	2.01E-05	2.01E-05
30	9.98E-01	2.88E-04	3.42E-04
60	9.98E-01	3.14E-04	3.79E-04
90	9.98E-01	3.53E-04	4.25E-04
120	9.97E-01	3.94E-04	4.74E-04
150	9.97E-01	4.43E-04	5.29E-04
180	9.97E-01	4.71E-04	5.63E-04
210	9.97E-01	4.82E-04	5.83E-04
240	9.96E-01	5.37E-04	6.43E-04
270	9.96E-01	5.47E-04	6.61E-04
300	9.96E-01	5.98E-04	7.17E-04
330	9.96E-01	6.11E-04	7.37E-04

***RH* = 28.5 ± 2 %, r_0 = 7.90 ± 0.790 µm**

t / min	*r*(*t*)/r_0	+Δ *r*(*t*)/r_0	-Δ *r*(*t*)/r_0
0	1.00E+00	2.01E-05	2.01E-05
30	9.99E-01	1.38E-04	1.47E-04
60	9.99E-01	1.75E-04	1.92E-04
90	9.99E-01	2.40E-04	2.73E-04
120	9.98E-01	2.91E-04	3.35E-04
150	9.98E-01	3.43E-04	3.99E-04
180	9.98E-01	3.96E-04	4.63E-04
210	9.97E-01	4.65E-04	5.46E-04
240	9.97E-01	5.26E-04	6.19E-04
270	9.96E-01	5.95E-04	7.03E-04
300	9.96E-01	6.55E-04	7.76E-04
330	9.95E-01	7.14E-04	8.48E-04

RH = 40.0 ± 2 %, r_0 = 9.76 ± 0.976 µm

t / min	$r(t)/r_0$	$+\Delta\, r(t)/r_0$	$-\Delta\, r(t)/r_0$
0	1.00E+00	1.98E-05	1.98E-05
30	9.96E-01	5.51E-04	6.66E-04
60	9.95E-01	7.39E-04	8.92E-04
90	9.93E-01	9.82E-04	1.19E-03
120	9.92E-01	1.21E-03	1.46E-03
150	9.90E-01	1.46E-03	1.76E-03
180	9.88E-01	1.72E-03	2.08E-03
210	9.86E-01	1.99E-03	2.41E-03
240	9.84E-01	2.27E-03	2.74E-03
270	9.82E-01	2.57E-03	3.11E-03
300	9.80E-01	2.90E-03	3.51E-03
330	9.78E-01	3.24E-03	3.92E-03

RH = 47.0 ± 2 %, r_0 = 9.37 ± 0.937 µm

t / min	$r(t)/r_0$	$+\Delta\, r(t)/r_0$	$-\Delta\, r(t)/r_0$
0	1.00E+00	1.95E-05	1.95E-05
30	9.95E-01	6.67E-04	8.07E-04
60	9.93E-01	9.84E-04	1.19E-03
90	9.90E-01	1.39E-03	1.67E-03
120	9.87E-01	1.89E-03	2.28E-03
150	9.84E-01	2.38E-03	2.87E-03
180	9.80E-01	2.96E-03	3.58E-03
210	9.75E-01	3.62E-03	4.39E-03
240	9.71E-01	4.31E-03	5.23E-03
270	9.65E-01	5.13E-03	6.24E-03
300	9.59E-01	6.05E-03	7.37E-03
330	9.53E-01	7.05E-03	8.62E-03

RH = 47.5 ± 2 %, r_0 = 9.90 ± 0.990 µm

t / min	$r(t)/r_0$	$+\Delta\, r(t)/r_0$	$-\Delta\, r(t)/r_0$
0	1.00E+00	2.04E-05	2.04E-05
30	9.95E-01	6.68E-04	8.08E-04
60	9.93E-01	9.96E-04	1.20E-03
90	9.90E-01	1.42E-03	1.71E-03
120	9.87E-01	1.89E-03	2.28E-03
150	9.84E-01	2.38E-03	2.88E-03
180	9.80E-01	2.94E-03	3.55E-03
210	9.76E-01	3.55E-03	4.30E-03
240	9.71E-01	4.24E-03	5.15E-03
270	9.66E-01	5.00E-03	6.08E-03
300	9.61E-01	5.84E-03	7.11E-03
330	9.54E-01	6.77E-03	8.27E-03

***RH* = 50.0 ± 2 %, r_0 = 7.98 ± 0.798 μm**

***t* / min**	***r*(*t*)/r_0**	**+Δ *r(t)*/r_0**	**-Δ *r(t)*/r_0**
0	1.00E+00	8.99E-05	8.99E-05
30	9.95E-01	7.63E-04	9.21E-04
60	9.91E-01	1.28E-03	1.54E-03
90	9.86E-01	1.97E-03	2.38E-03
120	9.81E-01	2.77E-03	3.34E-03
150	9.74E-01	3.79E-03	4.59E-03
180	9.66E-01	4.95E-03	6.01E-03
210	9.57E-01	6.38E-03	7.78E-03
240	9.45E-01	8.18E-03	1.00E-02
270	9.31E-01	1.05E-02	1.29E-02
300	9.10E-01	1.38E-02	1.72E-02
330	8.76E-01	1.96E-02	2.49E-02

The raw data files indicating the intensity map of Mie resonance spectra, from which the decrease in particle radius is derived in EDB experiments are given in the files below. Each file is a matrix of tab-separated relative scattering intensities over time, where rows are 30 s time increments. There are 1024 columns determining the wavelength channels in bins indicated in the data file, MieResonance_WavelengthBins.dat, with units of nanometers. Analysis procedures can be found in previous work[4].

MieResonance_RH165.dat
MieResonance_RH285.dat
MieResonance_RH40.dat
MieResonance_RH47.dat
MieResonance_RH475.dat
MieResonance_RH50.dat

A data list for CWFT experiments in Fig. 2B and Fig. S4 is tabulated below. The M_r error is ± 8% on its value. The film thickness error is ± 18% on its value. The *j* error is ± 10% on its value. The O_2 partial pressure error is ± 3% on its value. The *RH* error is ± 2%. Analysis and validation procedures followed previous work[5,6].

	M_r	**Thickness / cm**	***j* / s^{-1}**	**O_2 Partial Pressure / torr**	**RH / %**	**P_{HO2} / cm^{-2} min^{-1}**	**ΔP_{HO2} / cm^{-2} min^{-1}**	**Maximum HO_2 1st order loss rate / s^{-1}**	**Maximum HO_2 turnover rate / M s^{-1}**
Mole	0.010	1.05	6.37E-02	146	39.8	7.41E+10	4.02E+10	5.22E+02	5.74E-02
Ratio	0.031	1.07	6.37E-02	146	39.8	2.56E+11	6.34E+10	4.03E+02	1.35E-01
Dependence	0.050	1.08	6.37E-02	146	38.0	2.32E+11	6.24E+10	4.72E+02	1.46E-01
	0.082	1.12	6.37E-02	146	38.0	3.58E+11	6.32E+10	3.61E+02	1.37E-01
	0.100	1.14	6.37E-02	146	38.0	6.16E+11	5.16E+10	2.14E+02	1.26E-01
	0.113	1.15	6.37E-02	146	37.7	6.35E+11	5.08E+10	1.98E+02	1.19E-01
Film	0.078	0.18	6.37E-02	146	37.1	8.45E+10	2.83E+10	1.43E+03	1.39E-01
Thickness	0.078	0.53	6.37E-02	146	37.2	1.06E+11	3.06E+10	1.06E+03	1.29E-01
Dependence	0.078	0.89	6.37E-02	146	36.9	1.32E+11	3.18E+10	8.40E+02	1.38E-01
	0.078	1.24	6.37E-02	146	36.5	1.32E+11	3.18E+10	7.96E+02	1.40E-01
	0.078	1.78	6.37E-02	146	37.5	1.18E+11	3.13E+10	1.04E+03	1.41E-01

Light	0.078	1.24	9.10E-03	146	36.5	5.53E+10	2.28E+10	1.46E+03	9.55E-02
Dependence	0.078	1.24	2.73E-02	146	36.5	1.05E+11	3.04E+10	1.00E+03	1.54E-01
	0.078	1.24	4.55E-02	146	36.5	1.19E+11	3.13E+10	9.13E+02	1.56E-01
	0.078	1.24	6.37E-02	146	36.5	1.32E+11	3.18E+10	7.96E+02	1.40E-01
	0.078	1.24	0.00E+00	146	36.5	0.00E+00	2.01E+10	0.00E+00	0.00E+00
Oxygen	0.078	1.56	6.37E-02	146	33.6	1.16E+11	3.12E+10	5.51E+02	1.43E-01
Dependence	0.078	1.56	6.37E-02	293	33.8	1.17E+11	3.12E+10	1.12E+03	2.80E-01
	0.078	1.56	6.37E-02	439	33.8	1.18E+11	3.13E+10	1.61E+03	3.84E-01
	0.078	1.56	6.37E-02	585	33.8	1.31E+11	3.18E+10	1.74E+03	4.12E-01
	0.078	1.56	6.37E-02	731	33.8	1.17E+11	3.13E+10	2.26E+03	4.32E-01
	0.078	1.56	6.37E-02	0	33.8	0.00E+00	2.01E+10	0.00E+00	0.00E+00

The PRAD model input parameter for the HO_2 self-reaction rate, k_{SR}, for the CWFT experiments is listed in the table below, along with the maximum HO_2 concentration, $[HO_2]_{max}$, predicted in any single layer. All other entries are identical in the table above.

	M_r	Thickness / cm	j / s^{-1}	O_2 Partial Pressure / torr	RH / %	k_{SR} / M^{-1} s^{-1}	$[HO_2]_{max}$ / M
Mole	0.010	1.05	6.37E-02	146	39.8	2.85E+08	1.83E-06
Ratio	0.031	1.07	6.37E-02	146	39.8	7.21E+07	5.59E-06
Dependence	0.050	1.08	6.37E-02	146	38.0	9.15E+07	5.16E-06
	0.082	1.12	6.37E-02	146	38.0	5.69E+07	6.34E-06
	0.100	1.14	6.37E-02	146	38.0	2.18E+07	9.84E-06
	0.113	1.15	6.37E-02	146	37.7	1.99E+07	9.99E-06
Film	0.078	0.18	6.37E-02	146	37.1	8.85E+08	1.62E-06
Thickness	0.078	0.53	6.37E-02	146	37.2	5.27E+08	2.02E-06
Dependence	0.078	0.89	6.37E-02	146	36.9	3.08E+08	2.73E-06
	0.078	1.24	6.37E-02	146	36.5	2.71E+08	2.93E-06
	0.078	1.78	6.37E-02	146	37.5	4.61E+08	2.26E-06
Light	0.078	1.24	9.10E-03	146	36.5	1.34E+09	1.09E-06
Dependence	0.078	1.24	2.73E-02	146	36.5	3.93E+08	2.55E-06
	0.078	1.24	4.55E-02	146	36.5	3.21E+08	2.84E-06
	0.078	1.24	6.37E-02	146	36.5	2.71E+08	2.93E-06
	0.078	1.24	0.00E+00	146	36.5	-	-
Oxygen	0.078	1.56	6.37E-02	146	33.6	1.27E+08	4.33E-06
Dependence	0.078	1.56	6.37E-02	293	33.8	2.71E+08	4.15E-06
	0.078	1.56	6.37E-02	439	33.8	4.03E+08	3.99E-06
	0.078	1.56	6.37E-02	585	33.8	4.41E+08	3.95E-06
	0.078	1.56	6.37E-02	731	33.8	7.07E+08	3.19E-06
	0.078	1.56	6.37E-02	0	33.8	-	-

Figure S4

A data list for the experiments in S4 is shown below. We note the data for the green symbols can be found in Dou et al.[7] The error on β is ±0.07. Raw STXM/NEXAFS images available for download for each time stamp are also listed.

Black Open Circles

t / min	β	STXM/NEXAFS Images
4	0.23	Sample_Stack_2017-06-02_051.hdf5
13	0.19	Sample_Stack_2017-06-02_053.hdf5
19	0.34	Sample_Stack_2017-06-02_054.hdf5
34	0.29	Sample_Stack_2017-06-02_056.hdf5
43	0.33	Sample_Stack_2017-06-02_058.hdf5
52	0.30	Sample_Stack_2017-06-02_059.hdf5
58	0.31	Sample_Stack_2017-06-02_061.hdf5
70	0.32	Sample_Stack_2017-06-02_062.hdf5

Black Filled Circles

t / min	β	STXM/NEXAFS Images
185	0.45	Sample_Stack_2017-06-02_101.hdf5
190	0.38	Sample_Stack_2017-06-02_102.hdf5
195	0.38	Sample_Stack_2017-06-02_103.hdf5
210	0.30	Sample_Stack_2017-06-02_106.hdf5
217	0.33	Sample_Stack_2017-06-02_107.hdf5
226	0.32	Sample_Stack_2017-06-02_109.hdf5
236	0.38	Sample_Stack_2017-06-02_111.hdf5
240	0.34	Sample_Stack_2017-06-02_112.hdf5
246	0.34	Sample_Stack_2017-06-02_113.hdf5
256	0.34	Sample_Stack_2017-06-02_115.hdf5
263	0.36	Sample_Stack_2017-06-02_116.hdf5

Red Circles

t / min	β	STXM/NEXAFS Images
6	0.30	Sample_Stack_2017-06-22_161.hdf5
12	0.29	Sample_Stack_2017-06-22_162.hdf5
18	0.34	Sample_Stack_2017-06-22_163.hdf5
24	0.33	Sample_Stack_2017-06-22_164.hdf5
34	0.39	Sample_Stack_2017-06-22_166.hdf5
38	0.38	Sample_Stack_2017-06-22_167.hdf5
43	0.42	Sample_Stack_2017-06-22_168.hdf5
48	0.34	Sample_Stack_2017-06-22_169.hdf5
58	0.40	Sample_Stack_2017-06-22_171.hdf5
64	0.41	Sample_Stack_2017-06-22_172.hdf5
71	0.44	Sample_Stack_2017-06-22_173.hdf5
77	0.41	Sample_Stack_2017-06-22_174.hdf5
87	0.46	Sample_Stack_2017-06-22_176.hdf5
93	0.47	Sample_Stack_2017-06-22_177.hdf5
99	0.46	Sample_Stack_2017-06-22_178.hdf5
105	0.45	Sample_Stack_2017-06-22_179.hdf5
117	0.48	Sample_Stack_2017-06-22_181.hdf5
125	0.44	Sample_Stack_2017-06-22_182.hdf5
130	0.51	Sample_Stack_2017-06-22_183.hdf5

137	0.42	Sample_Stack_2017-06-22_184.hdf5
151	0.52	Sample_Stack_2017-06-22_186.hdf5
157	0.49	Sample_Stack_2017-06-22_187.hdf5
166	0.56	Sample_Stack_2017-06-22_188.hdf5
181	0.53	Sample_Stack_2017-06-22_189.hdf5
198	0.61	Sample_Stack_2017-06-22_191.hdf5

Figure S11

A data list from viscosity experiments is shown below. The η error was ± 10% on its value. The *RH* error was ± 2%. The *wt* error was ± 5% on its value.

	***RH* / %**	***wt***	**η / Pa s**
Sucrose	98.4	0.27	4.26E-03
	96.0	0.41	4.80E-03
	93.9	0.48	3.10E-02
	88.6	0.60	6.20E-02
	81.2	0.70	4.20E-01
	63.7	0.83	5.60E+01
	55.3	0.87	1.15E+03
Citric Acid	95.5	0.30	3.05E-03
	87.2	0.50	8.60E-03
	78.8	0.61	2.50E-02
	75.5	0.65	4.00E-02
	64.0	0.75	2.80E-01
	55.3	0.80	1.35E+00
	55.1	0.80	2.18E+00
	45.5	0.85	1.16E+01
Citric Acid	98.1	0.20	1.21E-03
and Fe^{III}Cit	98.1	0.15	1.20E-03
M_r = 0.05	98.0	0.25	1.79E-03
	93.2	0.30	2.08E-03
	93.2	0.30	2.00E-03
	82.2	0.55	1.63E-02
Citric Acid	98.2	0.20	2.18E-03
and Fe^{III}Cit	98.2	0.20	2.15E-03
M_r = 1.0	97.7	0.30	2.60E-03
	96.2	0.20	2.52E-03
	95.3	0.30	4.10E-03
	95.2	0.30	3.41E-03
	95.0	0.30	2.66E-03
	92.3	0.50	2.82E-02
	89.0	0.50	2.55E-02
	88.7	0.61	3.94E-01
	88.5	0.67	2.01E+00
	84.3	0.71	5.88E+00
water	100.0	1.00	1.00E-03

References:

1 Watts, B. & Raabe, J. A NeXus/HDF5 based file format for STXM. *AIP Conference Proceedings* **1696**, 020042, doi:10.1063/1.4937536 (2016).

2 Moffet, R. C. *et al.* Iron speciation and mixing in single aerosol particles from the Asian continental outflow. *J. Geophys. Res.-Atmos.* **117**, D07204, doi:10.1029/2011JD016746 (2012).

3 Moffet, R. C., Henn, T., Laskin, A. & Gilles, M. K. Automated Chemical Analysis of Internally Mixed Aerosol Particles Using X-ray Spectromicroscopy at the Carbon K-Edge. *Anal. Chem.* **82**, 7906-7914, doi:10.1021/ac1012909 (2010).

4 Dou, J. *et al.* Carbon Dioxide Diffusivity in Single, Levitated Organic Aerosol Particles. *J. Phys. Chem. Lett.* **10**, 4484-4489, doi:10.1021/acs.jpclett.9b01389 (2019).

5 Corral Arroyo, P., Aellig, R., Alpert, P. A., Volkamer, R. & Ammann, M. Halogen activation and radical cycling initiated by imidazole-2-carboxaldehyde photochemistry. *Atmos. Chem. Phys.* **19**, 10817-10828, doi:10.5194/acp-19-10817-2019 (2019).

6 Corral Arroyo, P. *et al.* Particle-Phase Photosensitized Radical Production and Aerosol Aging. *Environ. Sci. Technol.* **52**, 7680-7688, doi:10.1021/acs.est.8b00329 (2018).

7 Dou, J. *et al.* Photochemical degradation of iron(III)-citrate/citric acid aerosol quantified with the combination of three complementary experimental techniques and a kinetic process model. *Atmos. Chem. Phys.* **21**, 315-338, doi:10.5194/acp-21-315-2021 (2021).