

1 **Supplementary Information for**

2 **Directly measuring Fe(III)-catalyzed SO₂ oxidation rate in single levitated**
3 **droplets**

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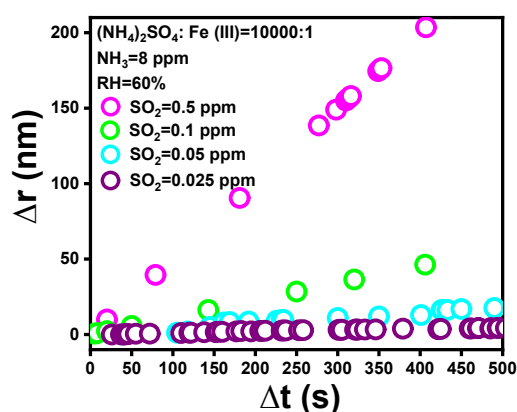
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9 **This document includes:**

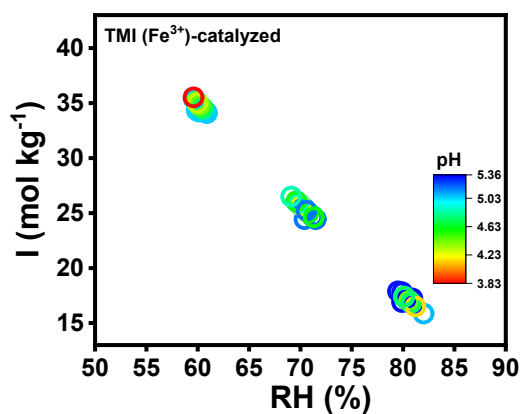
10 Figure S1 to S2

11 Table S1 to S2



12

13 **Figure S1. The reaction-induced hygroscopic growth of microdroplets.** The SO_2
 14 concentration was 25, 50, 100, and 500 ppb. The ambient RH was 60%; Droplet pH
 15 was buffered at 5.0 with 8 ppm NH_3 gas. The molar fraction of Fe (III) was maintained
 16 at 0.01%. At high SO_2 concentration, the droplets grew at faster speeds, indicating faster
 17 reactions.



18

19 **Figure S2. The negative relationship between droplet ionic strength I and ambient**
 20 **relative humidity conditions.** These datasets were calculated with E-AIM model III,
 21 with inputs of droplet composition and ambient conditions matching our experiments.
 22

Table S1. Experimental conditions and droplet properties at initial state

Experiment number	Droplet seed*	RH (%)	SO ₂ (ppm)	NH ₃ (ppm)	r0 (μm)	Aging time (h)	[S(VI)] (M)	pH	I (molal)	Fe (III) molar fraction
1	AS	60.9	1.000	8.00	6.580	0	6.067	5.05	34.09	0.0001
2	AS	60.8	0.500	8.00	6.807	0	6.076	5.05	34.20	0.0001
3	AS	60.8	0.100	8.00	4.849	0	6.076	5.05	34.20	0.0001
4	AS	60.7	0.100	8.00	5.502	0	6.085	5.05	34.29	0.0001
5	AS	60.6	0.050	8.00	5.390	0	6.094	5.05	34.39	0.0001
6	AS	59.9	0.025	8.00	5.149	0	6.155	5.03	35.10	0.0001
7	AS	59.8	0.025	8.00	5.177	0	6.164	5.04	35.20	0.0001
8	AS	60.1	0.025	8.00	4.488	0	6.137	5.04	34.23	0.0001
9	AS	59.9	0.025	8.00	5.148	0	6.155	5.04	35.10	0.0001
10	AS	59.6	0.025	8.00	4.901	0	6.181	5.03	35.41	0.0001
11	AS	60.6	0.025	8.00	6.115	0	6.102	5.05	34.39	0.0001
12	AS	60.4	0.025	8.00	5.910	0	6.111	5.04	34.59	0.0001
13	AS	59.9	0.025	8.00	6.530	0	6.155	5.04	35.10	0.0001
14	AS	60.5	0.025	8.00	6.570	0	6.111	5.04	34.50	0.0001
15	AS	60.7	0.025	8.00	6.016	0	6.085	5.05	34.29	0.0001
16	AS	60.5	0.025	8.00	6.999	0	6.102	5.04	34.25	0.0001
17	AS	71.2	0.025	8.00	5.065	0	5.098	5.19	24.70	0.0001
18	AS	70.4	0.025	8.00	5.197	0	5.179	5.17	24.45	0.0001
19	AS	71.5	0.025	8.00	5.153	0	5.068	5.19	24.45	0.0001
20	AS	70.4	0.025	8.00	4.254	0	5.179	5.17	25.38	0.0001
21	AS	70.2	0.025	8.00	4.711	0	5.199	5.17	25.55	0.0001
22	AS	69.9	0.025	8.00	5.350	0	5.229	5.17	25.81	0.0001
23	AS	70.4	0.025	8.00	4.346	0	5.179	5.17	25.38	0.0001
24	AS	70	0.025	8.00	4.200	0	5.219	5.17	25.73	0.0001
25	AS	71.2	0.025	8.00	6.281	0	5.098	5.19	24.70	0.0001
26	AS	71.2	0.025	8.00	6.699	0	5.098	5.19	24.69	0.0001
27	AS	80.9	0.025	8.00	5.018	0	4.023	5.35	16.99	0.0001
28	AS	80.9	0.025	8.00	5.471	0	4.062	5.34	17.24	0.0001
29	AS	79.9	0.025	8.00	4.968	0	4.010	5.35	16.91	0.0001
30	AS	79.9	0.025	8.00	4.010	0	4.152	5.33	17.80	0.0001
31	AS	79.5	0.025	8.00	4.981	0	4.164	5.34	17.35	0.0001
32	AS	80.4	0.025	8.00	6.570	0	4.049	5.34	17.21	0.0001
33	AS	80.2	0.025	8.00	6.179	0	4.075	5.33	17.88	0.0001
34	AS	80.3	0.025	8.00	4.416	0	4.062	5.34	17.16	0.0001
35	AS+ABS	80.3	0.025	8.00	5.122	0	3.981	5.34	17.24	0.0001
36	AS+ABS	80.5	0.025	8.00	5.060	0	3.849	5.35	17.07	0.0001
37	AS+ABS	80.9	0.025	8.00	5.281	0	4.402	5.35	16.75	0.0001
38	AS+ABS	59	0.025	8.00	4.546	0	6.233	5.03	34.29	0.0001
39	AS+ABS	60.5	0.025	8.00	4.570	0	6.102	5.04	33.81	0.0001
40	AS+ABS	60.4	0.025	8.00	3.880	0	6.111	5.04	34.20	0.0001

Experiment number	Droplet seed*	RH (%)	SO ₂ (ppm)	NH ₃ (ppm)	r0 (μm)	Aging time (h)	[S(VI)] (M)	pH	I (molal)	Fe (III) molar fraction
41	AS+ABS	60.3	0.025	8.00	4.312	0	6.120	5.04	34.09	0.0001
42	AS+ABS	60.3	0.025	8.00	4.684	0	6.120	5.04	34.59	0.0001
43	AS+ABS	59.6	0.025	8.00	4.859	0	6.181	5.03	34.20	0.0001
44	AS+ABS	59.7	0.025	8.00	4.778	0	6.172	5.03	34.29	0.0001
45	AS+ABS	59.9	0.025	8.00	4.623	0	6.155	5.04	34.59	0.0001
46	AS+ABS	60	0.025	8.00	4.794	0	6.146	5.04	34.39	0.0001
47	AS+ABS	60	0.025	8.00	4.757	0	6.146	5.04	34.29	0.0001
48	AS+ABS	59.5	0.025	8.00	4.461	0	6.190	5.03	34.20	0.0001
49	AS+ABS	59.5	0.025	8.00	4.036	0	6.190	5.03	35.10	0.0001
50	AS+ABS	59.6	0.025	8.00	7.082	0	6.181	5.03	35.30	0.0001
51	AS+ABS	59.9	0.025	8.00	7.028	0	6.190	5.03	35.10	0.0001
52	AS+ABS	59.6	0.025	8.00	7.091	0	6.181	5.03	35.30	0.0001
53	AS+ABS	60	0.025	8.00	7.078	0	6.146	5.04	34.00	0.0001
54	AS+ABS	59.9	0.025	8.00	6.997	0	6.190	5.03	35.10	0.0001
55	AS+ABS	60	0.025	8.00	6.987	0	6.146	5.04	34.00	0.0001
56	AS+ABS	59.9	0.025	8.00	6.997	0	6.190	5.03	35.10	0.0001
57	AS+ABS	61.2	0.025	8.00	5.258	0	6.041	5.05	33.81	0.00001
58	AS+ABS	60.6	0.025	8.00	5.180	0	6.067	5.05	34.22	0.00001
59	AS+ABS	59.9	0.025	8.00	5.235	0	6.155	5.04	35.10	0.00001
60	AS+ABS	60.2	0.025	8.00	5.116	0	6.129	5.04	34.80	0.00001
61	AS+ABS	59.3	0.025	8.00	5.189	0	6.207	5.03	35.72	0.00001
62	AS+ABS	59.7	0.025	8.00	5.181	0	6.172	5.03	35.31	0.00001
63	AS+ABS	60.1	0.025	8.00	5.175	0	6.137	5.04	34.90	0.00001
64	AS+ABS	59.7	0.025	8.00	6.088	0	6.172	5.03	35.31	0.00001
65	AS+ABS	59.9	0.025	8.00	5.181	0	6.155	5.04	35.10	0.000001
66	AS+ABS	59.9	0.025	8.00	5.309	0	6.155	5.04	35.10	0.000001
67	AS+ABS	60.1	0.025	8.00	5.506	0	6.155	5.04	34.90	0.000001
68	AS+ABS	59.7	0.025	8.00	5.333	0	6.172	5.03	35.31	0.001
69	AS+ABS	59.5	0.025	8.00	5.982	0	6.190	5.03	35.56	0.001
70	AS+ABS	59.7	0.025	8.00	6.048	0	6.172	5.03	35.35	0.001
71	AS+ABS	60.1	0.025	8.00	4.320	0	6.137	5.04	34.95	0.001
72	AS+ABS	59.9	0.025	8.00	5.249	0	6.155	5.04	35.15	0.001
73	AS+ABS	60.4	0.025	8.00	5.158	12	6.111	5.04	34.60	0.0001
74	AS+ABS	59.3	0.025	8.00	5.903	12	6.207	5.03	35.72	0.0001
75	AS+ABS	59.7	0.025	8.00	4.923	12	6.172	5.03	35.31	0.0001
76	AS+ABS	60	0.025	8.00	5.307	12	6.146	5.04	35.01	0.0001
77	AS+ABS	59.9	0.025	8.00	5.254	12	6.155	5.04	35.11	0.0001
78	AS+ABS	60.1	0.025	8.00	5.203	12	6.137	5.04	34.90	0.0001
79	AS+ABS	60.7	0.025	8.00	6.407	36	6.085	5.05	34.30	0.0001
80	AS+ABS	59.7	0.025	8.00	5.261	36	6.172	5.03	35.31	0.0001
81	AS+ABS	59.7	0.025	8.00	5.158	36	6.172	5.03	35.31	0.0001

Experiment number	Droplet seed*	RH (%)	SO ₂ (ppm)	NH ₃ (ppm)	r0 (μm)	Aging time (h)	[S(VI)] (M)	pH	I (molal)	Fe (III) molar fraction
82	AS+ABS	59.6	0.025	8.00	5.183	36	6.181	5.03	35.42	0.0001
83	AS+ABS	59.7	0.025	8.00	5.061	36	6.172	5.03	35.31	0.0001
84	AS+ABS	59.7	0.025	8.00	5.115	48	6.172	5.03	35.31	0.0001
85	AS+ABS	59.4	0.025	8.00	5.284	48	6.198	5.03	35.62	0.0001
86	AS+ABS	59.6	0.025	8.00	5.018	48	6.181	5.03	35.42	0.0001
87	AS+ABS	60.7	0.025	8.00	5.306	48	6.085	5.05	34.30	0.0001
88	AS+ABS	60.9	0.025	8.00	5.372	48	6.067	5.05	34.10	0.0001

24 *AS denotes ammonium sulfate; ABS, ammonium bisulfate

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Table S2. Droplet hygroscopic growth rate and sulfate formation rate

Experiment number	Reaction Product	dr/dt (nm s^{-1}) *	$R_{S(VI)}$ ($10^{-15} \text{ mol s}^{-1}$)	Γ_{SO_2} (10^{-3})	$R_{S(VI)}/P_{SO_2}/A$ ($10^{-16} \text{ mol s}^{-1} \text{ ppm}^{-1} \text{ um}^{-2}$)
1	AS	0.939±0.0002	3.100±0.076	1.769±0.0435	0.057±0.0014
2	AS	0.552±0.0002	1.953±0.0683	2.083±0.0728	0.067±0.0023
3	AS	0.114±0.0004	0.205±0.001	2.151±0.0079	0.069±0.0003
4	AS	0.103±0.0004	0.238±0.001	2.151±0.0079	0.063±0.0002
5	AS	0.021±0.0003	0.046±0.001	0.776±0.0110	0.025±0.0003
6	AS	0.013±0.0002	0.0267±0.000	0.994±0.0123	0.031±0.0004
7	AS	0.013±0.0000	0.0262±0.000	0.965±0.0034	0.031±0.0001
8	AS	0.0133±0.0002	0.0207±0.000	1.014±0.0144	0.033±0.0005
9	AS	0.013±0.0002	0.027±0.0003	0.994±0.0123	0.032±0.0004
10	AS	0.011±0.0001	0.021±0.0001	0.860±0.0040	0.028±0.0001
11	AS	0.017±0.0002	0.047±0.0005	1.251±0.0131	0.040±0.0004
12	AS	0.010±0.0002	0.028±0.0004	0.782±0.0119	0.025±0.0004
13	AS	0.012±0.0001	0.040±0.0004	0.925±0.0102	0.030±0.0003
14	AS	0.015±0.0002	0.050±0.0006	1.134±0.0139	0.037±0.0004
15	AS	0.0153±0.0001	0.042±0.0014	1.153±0.0381	0.037±0.0012
16	AS	0.010±0.0001	0.038±0.0005	0.769±0.0093	0.025±0.0003
17	AS	0.010±0.0001	0.017±0.0002	0.658±0.0063	0.061±0.0006
18	AS	0.016±0.0003	0.028±0.0005	1.016±0.0188	0.050±0.0023
19	AS	0.007±0.0001	0.011±0.0001	0.414±0.0050	0.039±0.0006
20	AS	0.007±0.0000	0.009±0.0001	0.483±0.0031	0.019±0.0004
21	AS	0.006±0.0001	0.008±0.0001	0.352±0.0059	0.018±0.0002
22	AS	0.006±0.0001	0.011±0.0002	0.386±0.0076	0.013±0.0004
23	AS	0.011±0.0001	0.013±0.0001	0.688±0.0059	0.004±0.0004
24	AS	0.006±0.0001	0.007±0.0001	0.383±0.0074	0.021±0.0002
25	AS	0.006±0.0001	0.032±0.0013	0.348±0.0071	0.033±0.0006
26	AS	0.011±0.0004	0.014±0.0003	0.707±0.0279	0.013±0.0002
27	AS	0.011±0.0003	0.013±0.0003	0.530±0.0136	0.016±0.0001
28	AS	0.013±0.0002	0.020±0.0003	0.673±0.0100	0.011±0.0002
29	AS	0.016±0.0002	0.019±0.0003	0.777±0.0122	0.012±0.0001
30	AS	0.009±0.0004	0.007±0.0003	0.449±0.0186	0.022±0.0002
31	AS	0.016±0.0007	0.017±0.0002	0.451±0.0047	0.012±0.0002
32	AS	0.013±0.0002	0.011±0.0003	0.540±0.0147	0.011±0.0002
33	AS	0.009±0.0001	0.014±0.0009	0.554±0.0360	0.023±0.0009
34	AS	0.011±0.0003	0.028±0.0005	0.639±0.0110	0.005±0.0001
35	AS	0.008±0.0006	0.010±0.0008	0.394±0.0315	0.013±0.0010
36	AS	0.009±0.0005	0.012±0.0006	0.446±0.0238	0.014±0.0008
37	AS	0.008±0.0001	0.011±0.0001	0.401±0.0049	0.013±0.0002
38	AS	0.016±0.0001	0.0261±0.0001	1.247±0.0052	0.040±0.0002
39	AS	0.014±0.0001	0.0227±0.0002	1.077±0.0086	0.035±0.0003
40	AS	0.013±0.0003	0.0148±0.0038	0.972±0.2475	0.031±0.0080

Experiment number	Reaction Product	dr/dt (nm s ⁻¹) *	R _{S(VI)} (10 ⁻¹⁵ mol s ⁻¹)	Γ _{SO₂} (10 ⁻³)	R _{S(VI)} /P _{SO₂} /A (10 ⁻¹⁶ mol s ⁻¹ ppm ⁻¹ um ⁻²)
41	AS	0.013±0.0001	0.0189±0.0001	1.004±0.0059	0.032±0.0002
42	AS	0.013±0.0002	0.0214±0.0004	0.966±0.0170	0.031±0.0005
43	AS	0.009±0.0005	0.0165±0.0010	0.691±0.0412	0.022±0.0013
44	AS	0.011±0.0001	0.0202±0.0002	0.874±0.0065	0.028±0.0002
45	AS	0.011±0.0001	0.0188±0.0001	0.872±0.0049	0.028±0.0002
46	AS	0.019±0.0001	0.0330±0.0001	1.420±0.0057	0.046±0.0002
47	AS	0.012±0.0002	0.0203±0.0004	0.886±0.0182	0.029±0.0006
48	AS	0.014±0.0001	0.0221±0.0002	1.100±0.0093	0.035±0.0003
49	AS	0.012±0.0008	0.0151±0.0010	0.915±0.0612	0.029±0.0020
50	AS	0.015±0.0001	0.0592±0.0006	1.167±0.0112	0.038±0.0004
51	AS	0.010±0.0005	0.0384±0.0017	0.769±0.0347	0.025±0.0011
52	AS	0.014±0.0001	0.0535±0.0001	1.052±0.0027	0.034±0.0001
53	AS	0.014±0.0004	0.0534±0.0016	1.054±0.0311	0.034±0.0010
54	AS	0.011±0.0004	0.0427±0.0017	0.861±0.0338	0.028±0.0011
55	AS	0.012±0.0001	0.0437±0.0003	0.886±0.0058	0.029±0.0002
56	AS	0.016±0.0003	0.0602±0.0011	1.215±0.0224	0.039±0.0007
57	AS	0.011±0.0001	0.0227±0.0002	0.810±0.0060	0.026±0.0002
58	AS	0.011±0.0001	0.0219±0.0001	0.807±0.0052	0.026±0.0002
59	AS	0.016±0.0001	0.0339±0.0001	1.223±0.0047	0.039±0.0001
60	AS	0.012±0.0001	0.0250±0.0001	0.944±0.0043	0.030±0.0001
61	AS	0.010±0.0001	0.0210±0.0001	0.771±0.0043	0.025±0.0001
62	AS	0.011±0.0002	0.0237±0.0004	0.874±0.0130	0.028±0.0004
63	AS	0.009±0.0003	0.0192±0.0005	0.709±0.0196	0.023±0.0006
64	AS	0.012±0.0002	0.0354±0.0007	0.943±0.0175	0.030±0.0006
65	AS	0.011±0.0001	0.0235±0.0001	0.864±0.0048	0.028±0.0002
66	AS	0.015±0.0001	0.0336±0.0002	1.178±0.0068	0.038±0.0002
67	AS	0.011±0.0002	0.0256±0.0005	0.833±0.0155	0.027±0.0005
68	AS	0.013±0.0002	0.0296±0.0004	1.027±0.0155	0.033±0.0005
69	AS	0.013±0.0002	0.0362±0.0004	1.000±0.0119	0.032±0.0004
70	AS	0.014±0.0001	0.0389±0.0004	1.051±0.0096	0.034±0.0003
71	AS	0.015±0.0001	0.0214±0.0001	1.136±0.0064	0.037±0.0002
72	AS	0.014±0.0002	0.0305±0.0005	1.093±0.0180	0.035±0.0006
73	AS	0.006±0.0004	0.0118±0.0007	0.440±0.0267	0.014±0.0009
74	AS	0.005±0.0002	0.0139±0.0004	0.393±0.0124	0.013±0.0004
75	AS	0.006±0.0001	0.0111±0.0002	0.452±0.0080	0.015±0.0003
76	AS	0.004±0.0002	0.0091±0.0005	0.321±0.0182	0.010±0.0006
77	AS	0.005±0.0002	0.0105±0.0003	0.375±0.0121	0.012±0.0004
78	AS	0.005±0.0002	0.0094±0.0004	0.343±0.0153	0.011±0.0005
79	AS	0.003±0.0003	0.0094±0.0008	0.227±0.0202	0.007±0.0007
80	AS	0.003±0.0002	0.0073±0.0004	0.261±0.0127	0.008±0.0004
81	AS	0.003±0.0001	0.0068±0.0003	0.252±0.0114	0.008±0.0004
82	AS	0.004±0.0002	0.0075±0.0003	0.276±0.0126	0.009±0.0004

Experiment number	Reaction Product	dr/dt (nm s⁻¹) *	R_{S(VI)} (10⁻¹⁵ mol s⁻¹)	Γ_{SO₂} (10⁻³)	R_{S(VI)}/P_{SO₂}/A (10⁻¹⁶ mol s⁻¹ ppm⁻¹ μm⁻²)
83	AS	0.003±0.0002	0.0058±0.0003	0.222±0.0122	0.007±0.0004
84	AS	0.003±0.0001	0.0055±0.0002	0.207±0.0078	0.007±0.0003
85	AS	0.003±0.0003	0.0054±0.0008	0.193±0.0267	0.006±0.0009
86	AS	0.002±0.0001	0.0043±0.0001	0.169±0.0041	0.005±0.0001
87	AS	0.003±0.0001	0.0055±0.0002	0.194±0.0082	0.006±0.0003
88	AS	0.002±0.0001	0.0051±0.0002	0.173±0.0072	0.006±0.0002

27 * Error represents the 95% confidence interval values of dr_p/dt , which were obtained by linear fitting $r_p(t)$ data.