



Supplement of

Driving factors of oxalic acid and enhanced role of gas-phase oxidation under cleaner conditions: insights from 2007–2018 field observations in the Pearl River Delta

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S1. Descriptions of Fisher r -to- z transformation.

The Pearson correlation coefficient r is widely used to quantify the strength and direction of linear relationships between two variables. However, the sampling distribution of r is not normally distributed, especially when the true correlation is far from zero or the sample size is small. To address this issue, Fisher (1921) proposed a transformation of r to a variable z , known as the Fisher r -to- z transformation, defined as:

$$z = \frac{1}{2} \ln \frac{1+r}{1-r} \quad (\text{S1})$$

This transformation converts r into an approximately normal distribution, allowing for more accurate estimation of confidence intervals and hypothesis testing. The standard error of z is given as:

$$SE = 1/\sqrt{n-3} \quad (\text{S2})$$

where n is the sample size. After calculating the confidence interval in the z -space, it can be back-transformed to the original r scale, providing a robust measure of uncertainty for correlation estimates.

Furthermore, the Fisher r -to- z transformation can also be used to test whether two correlation coefficients from independent samples differ significantly. For two correlations r_1 and r_2 with sample sizes n_1 and n_2 , their corresponding z values are calculated as above, and the standard error of the difference is calculated as:

$$SE = 1/\sqrt{\frac{1}{n_1-3} + \frac{1}{n_2-3}} \quad (\text{S3})$$

The difference is then standardized as:

$$Z = \frac{z_1 - z_2}{SE} \quad (\text{S4})$$

A two-tailed p value can be derived from the standard normal distribution to determine whether the difference between r_1 and r_2 is statistically significant. This approach provides a rigorous method for comparing correlation strengths across independent datasets.

Table S1. Concentrations of DCA and C₂ (ng m⁻³) at different types of sampling sites.

Site type	Site	Sample type	Period	DCA	C ₂	Reference
Urban	China, Shangdong	PM _{2.5}	January to February, 2017	1415 ± 899	817 ± 544	(Meng et al., 2020)
		PM _{2.5}	January to February, 2020	369 ± 112	210 ± 88	(Meng et al., 2023)
	China, Xi'an	TSP	2009 summer	1350 ± 247	1291 ± 120	(Wang et al., 2012)
			2009 winter	2053 ± 1097	1887 ± 586	
	China, Beijing	PM _{2.5}	January 2017	1068.7	440.9	(Yu et al., 2021)
			April 2017	601.5	228.2	
			July 2017	1471.7	756	
			October 2017	1046.9	519.4	
	14 cities of China	PM _{2.5}	2003 summer	892 ± 457	513 ± 285	(Ho et al., 2007)
			2003 winter	904 ± 480	558 ± 351	
	Mongolia, Ulaanbaatar	PM _{2.5}	November 2007 to January 2008	536 ± 156	107 ± 28	(Jung et al., 2010)
	India, Raipur	PM _{2.1}	2012-2013 winter	1072 ± 375	545 ± 231	(Deshmukh et al., 2016)
	USA, Fairbanks	PM _{2.5}	June to September, 2009	73.6 ± 70.4	36.6 ± 35.6	(Deshmukh et al., 2018)
Coast	China, Shanghai	PM _{2.5}	May to August 2018	359 ± 277	213 ± 177	(Ding et al., 2021)
	China, Shanghai	PM _{2.5}	December 2018 to January 2019	360 ± 233	199 ± 151	(Du et al., 2022)
	East China Sea	PM _{2.5}	2002 winter	193 ± 164	92.6 ± 94.5	(Zhang et al., 2016)
Mountain	China, MT. Hua	PM ₁₀	2019 winter	638 ± 377	399 ± 261	(Meng et al., 2014)
			2019 summer	744 ± 340	522 ± 261	
	China, MT. Tai	PM _{2.5}	July to August, 2016	354 ± 239	213±162	(Meng et al., 2018)
	Japan, Fuji	TSP	July to August, 2009	308 ± 102	160 ± 37	(Kunwar et al., 2019)
Marine	Bay of Bengal	PM _{2.5}	December 2008 to January 2009	154 ± 84	116 ± 65	(Bikkina et al., 2017)
	North Pacific	TSP	August to September 2008 (MBA)	58 ± 45	26.1 ± 15.9	(Bikkina et al., 2014)
			August to September 2008 (LBA)	15 ± 6	10.3 ± 4.8	

Table S2. Information of PM_{2.5} samples.

Year	Duration	Number of samples
2007	October to November	32
2008	November to December	45
2009	November to December	25
2010	October to December	69
2011	November to December	28
2012	November to December	39
2013	November to December	29
2014	October to November	20
2015	October to November	37
2016	October to November	33
2017	October to December	55
2018	October to December	50

Table S3. Information on target organic compounds and internal standards

	Compounds	Standards	Internal standards
Dicarboxylic acid	Succinic acid (C ₄)	C ₄	Lauric acid-D ₂₃
	Glutaric acid (C ₅)	C ₅	Lauric acid-D ₂₃
	Adipic acid (C ₆)	C ₆	Lauric acid-D ₂₃
	Pimelic acid (C ₇)	C ₇	Lauric acid-D ₂₃
	Suberic acid (C ₈)	C ₈	Lauric acid-D ₂₃
	Azelaic acid (C ₉)	C ₉	Lauric acid-D ₂₃
	Sebacic acid (C ₁₀)	C ₁₀	Lauric acid-D ₂₃
	Phthalic acid (Ph)	Ph	Phthalic acid 3,4,5,6-D ₄
	Terephthalic acid (tPh)	Ph	Phthalic acid 3,4,5,6-D ₄
Hopanes	17 α (H)-22,29,30-trisnorhopane (C _{27α})	C _{27α}	C ₂₄ D ₅₀
	17 α (H),21 β (H)-30-norhopane (C _{29$\alpha\beta$})	C _{29$\alpha\beta$}	C ₂₄ D ₅₀
	17 α (H),21 β (H)-30-hopane (C _{30$\alpha\beta$})	C _{30$\alpha\beta$}	C ₂₄ D ₅₀
	17 α (H),21 β (H)-22R-homohopane (C _{31$\alpha\beta$R})	C _{31$\alpha\beta$R}	C ₂₄ D ₅₀
	17 α (H),21 β (H)-22S-homohopane (C _{31$\alpha\beta$S})	C _{31$\alpha\beta$S}	C ₂₄ D ₅₀
Other	Levoglucosan	Levoglucosan	Levoglucosan- ¹³ C ₆
	Octadecanoic acid	Octadecanoic acid	Hexadecanoic acid-D ₃₁
	Picene	Benzo(ghi)perylene	Perylene-D ₁₂
	Citramalic acid	Citramalic acid	Lauric acid-D ₂₃
	Malic acid	Malic acid	Lauric acid-D ₂₃

Table S4. Response factors (RFs) derived from the annual calibration curves.

	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
Succinic acid	1.79	3.66	0.90	0.93	0.93	1.47	2.11	1.47	1.47	1.65	2.41	1.52
Glutaric acid	2.88	4.07	3.31	2.30	2.30	2.93	2.70	2.93	0.81	3.95	5.53	2.38
Adipic acid	2.64	3.87	2.82	2.81	2.81	3.70	3.66	3.76	1.54	5.86	3.76	3.24
Pimelic acid	2.16	2.53	2.30	2.19	2.19	3.97	3.90	3.97	3.96	5.78	3.76	4.54
Suberic acid	2.23	2.47	2.70	2.68	2.68	4.6	3.06	4.60	5.41	7.36	4.39	6.45
Azelaic acid	2.03	2.03	2.42	2.41	2.41	3.99	3.12	3.98	5.07	6.37	3.44	6.87
Sebacic acid	3.13	3.27	4.31	4.29	4.29	5.79	5.54	5.79	5.79	4.83	4.93	4.45
Phthalic acid	1.01	1.07	0.86	0.66	0.98	1.92	1.86	1.93	1.88	1.88	1.98	1.91
Terephthalic acid	1.01	1.07	0.86	0.66	0.98	1.92	1.86	1.93	1.88	1.88	1.98	1.91
17 α (H)-22,29,30-trisnorhopane	0.79	0.79	0.70	0.79	0.55	0.83	0.99	0.82	0.91	0.53	0.67	0.76
17 α (H),21 β (H)-30-norhopane	0.73	0.73	0.63	0.73	0.53	0.74	0.87	0.75	0.78	0.46	0.62	0.78
17 α (H),21 β (H)-30-hopane	0.78	0.78	0.74	0.78	0.52	0.78	0.88	0.76	0.42	0.70	0.59	0.97
17 α (H),21 β (H)-22R-homohopane	1.26	1.26	0.72	1.26	0.45	1.11	1.50	1.12	0.42	0.73	0.91	1.75
17 α (H),21 β (H)-22S-homohopane	1.55	1.55	0.72	1.55	1.33	1.25	1.90	1.19	0.42	1.83	1.01	1.79
Levoglucosan	1.35	1.30	1.22	0.92	1.04	0.93	0.97	0.93	0.97	0.96	0.95	0.96
Octadecanoic acid	0.50	0.56	0.66	0.68	0.96	0.89	1.29	0.89	1.22	0.71	0.89	1.09
Picene	0.84	0.80	0.84	0.77	0.78	0.66	0.80	0.66	0.79	1.11	0.81	1.06
Citramalic acid	1.35	1.95	1.30	3.70	3.56	2.60	2.10	2.59	2.63	2.07	2.04	2.09
Malic acid	1.46	1.46	1.16	0.95	0.91	0.87	0.86	1.02	1.15	0.85	0.76	1.05

Table S5. Meteorological parameters, PM2.5 main components, organic molecular tracers, diacids, pH, and ALWC in the PRD (2007–2018).

	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
I. Meteorological parameters												
Temperature (°C) ^a	22.2 ± 2.1	17.2 ± 2.9	17.0 ± 3.1	19.7 ± 3.2	19.9 ± 3.8	22.1 ± 1.4	20.9 ± 1.1	20.2 ± 4.4	25.1 ± 2.4	23.8 ± 3.8	21.3 ± 3.2	22.4 ± 2.9
Relative humidity (%) ^a	57 ± 11	47 ± 12	67 ± 13	64 ± 11	57 ± 11	61 ± 7	50 ± 13	57 ± 14	63 ± 8	67 ± 7	56 ± 13	63 ± 12
Solar radiation (W m ⁻²) ^a	161.3 ± 41.3	156.5 ± 28.3	135.2 ± 36.3	141.8 ± 51.4	134.7 ± 36.9	101.2 ± 40.4	125.2 ± 46.2	95.3 ± 49.1	149.0 ± 36.2	122.4 ± 44.0	128.9 ± 53.1	122.2 ± 39.9
Boundary layer height (m) ^b	624 ± 124	572 ± 141	633 ± 105	635 ± 183	555 ± 167	550 ± 121	515 ± 88	586 ± 146	591 ± 115	577 ± 165	540 ± 142	532 ± 107
II. Molecular tracers (ng m⁻³)												
Levoglucosan	203 ± 77	422 ± 301	333 ± 259	264 ± 170	208 ± 152	48 ± 43	257 ± 140	205 ± 131	110 ± 66	92 ± 44	143 ± 63	63 ± 33
Hopanes	2.81 ± 1.05	6.20 ± 3.30	3.38 ± 2.57	2.69 ± 2.36	2.61 ± 1.53	0.86 ± 0.65	1.09 ± 0.64	1.11 ± 0.59	0.70 ± 0.39	1.08 ± 0.74	0.86 ± 0.47	0.71 ± 0.30
Octadecanoic acid	38.5 ± 8.1	41.8 ± 29.6	30.8 ± 11.1	28.4 ± 17.3	24.3 ± 14.2	23.9 ± 10.1	53.1 ± 20.8	26.8 ± 13.8	23.6 ± 10.6	16.3 ± 15.0	14.6 ± 7.8	17.9 ± 14.7
Picene	0.13 ± 0.07	0.29 ± 0.25	0.19 ± 0.15	0.21 ± 0.17	0.29 ± 0.20	0.19 ± 0.10	0.22 ± 0.11	0.18 ± 0.11	0.17 ± 0.08	0.24 ± 0.11	0.26 ± 0.14	0.12 ± 0.07
Terephthalic acid (tPh)	32.9 ± 23.6	58.9 ± 30.5	22.7 ± 12.0	34.4 ± 17.6	26.1 ± 35.6	39.1 ± 30.3	103.1 ± 59.5	62.3 ± 31.3	52.8 ± 29.1	51.8 ± 43.6	31.3 ± 16.4	17.4 ± 8.7
Phthalic acid (Ph)	51.9 ± 14.9	36.9 ± 10.9	23.6 ± 12.1	24.1 ± 10.0	18.3 ± 12.1	22.4 ± 12.1	47.6 ± 18.1	53.3 ± 21.0	31.1 ± 15.7	28.3 ± 16.6	20.5 ± 8.0	16.7 ± 5.7
2,3-dihydroxy-4-oxopentanoic acid (DHOPA)	1.85 ± 1.35	2.30 ± 2.50	1.15 ± 1.25	2.10 ± 1.70	2.11 ± 2.16	0.44 ± 0.41	2.15 ± 1.59	2.64 ± 2.37	2.42 ± 2.53	1.21 ± 1.02	2.07 ± 1.57	1.05 ± 0.88
Malic acid	24.2 ± 19.4	14.1 ± 17.8	2.5 ± 2.7	9.4 ± 7.3	15.6 ± 18.9	4.57 ± 3.4	14.3 ± 12.9	17.9 ± 16.8	27.9 ± 21.7	10.7 ± 8.3	15.2 ± 10.0	5.9 ± 4.9
III. Aliphatic Diacids (ng m⁻³)												
Oxalic acid (C ₂)	692 ± 243	460 ± 171	NA	386 ± 168	357 ± 161	317 ± 191	526 ± 274	432 ± 200	468 ± 205	361 ± 208	404 ± 208	274 ± 114
Succinic acid (C ₄)	106.8 ± 51.5	47.3 ± 61.3	29.8 ± 34.1	20.5 ± 12.9	18.4 ± 14.6	13.4 ± 10.7	34.8 ± 20.4	41.0 ± 25.3	23.8 ± 16.7	30.0 ± 22.0	22.1 ± 13.8	10.9 ± 5.8
Glutaric acid (C ₅)	24.9 ± 11.2	6.6 ± 9.4	5.8 ± 5.3	6.2 ± 3.7	5.1 ± 3.5	1.9 ± 1.7	9.6 ± 5.1	9.8 ± 5.6	1.4 ± 1.3	9.7 ± 7.9	6.3 ± 4.6	3.5 ± 1.9
Adipic acid (C ₆)	8.7 ± 3.6	4.3 ± 4.5	3.8 ± 2.7	4.8 ± 2.9	3.8 ± 2.6	2.3 ± 1.8	9.9 ± 4.4	7.2 ± 3.5	2.2 ± 1.3	5.5 ± 2.9	4.8 ± 2.8	4.1 ± 2.4
Pimelic acid (C ₇)	2.0 ± 0.5	1.5 ± 1.3	1.3 ± 0.9	0.9 ± 0.5	1.0 ± 0.7	1.1 ± 0.6	2.8 ± 1.7	2.2 ± 1.0	1.5 ± 0.8	1.9 ± 1.5	1.2 ± 0.5	1.1 ± 0.4
Suberic acid (C ₈)	2.7 ± 0.5	2.0 ± 1.5	3.4 ± 2.4	1.5 ± 2.8	1.8 ± 1.3	1.9 ± 1.0	5.1 ± 2.8	3.1 ± 1.7	2.4 ± 0.9	3.4 ± 2.2	1.7 ± 0.8	2.4 ± 0.9

Table S5. (continued)

	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
Azelaic acid (C ₉)	8.5 ± 1.8	8.6 ± 6.1	18.7 ± 13.2	6.7 ± 3.7	7.3 ± 5.5	9.6 ± 4.8	27.3 ± 14.3	16.8 ± 8.6	11.6 ± 4.9	17.8 ± 10.9	7.5 ± 3.1	11.4 ± 4.0
Sebacic acid (C ₁₀)	1.1 ± 0.2	1.1 ± 0.8	2.8 ± 2.3	1.1 ± 0.8	1.1 ± 0.9	1.7 ± 0.8	3.6 ± 2.1	2.2 ± 1.3	2.0 ± 0.8	2.9 ± 2.1	1.2 ± 0.6	1.5 ± 0.5
Subtotal	864 ± 283	532 ± 202	NA	427 ± 156	396 ± 181	352 ± 194	610 ± 305	527 ± 173	529 ± 227	416 ± 238	452 ± 226	307 ± 122
IV. Other species												
pH ^a	1.51 ± 1.07	2.60 ± 0.71	1.94 ± 0.29	1.97 ± 1.00	2.54 ± 0.37	2.55 ± 0.43	2.69 ± 0.42	2.29 ± 0.33	2.13 ± 0.33	2.05 ± 0.46	2.60 ± 0.45	2.66 ± 0.37
ALWC (µg m ⁻³) ^a	20.6 ± 10.0	11.3 ± 7.9	28.8 ± 11.4	22.0 ± 10.8	19.3 ± 9.9	13.5 ± 5.6	12.0 ± 7.5	10.8 ± 5.4	12.5 ± 6.2	12.0 ± 7.3	9.9 ± 6.5	11.0 ± 6.7
O _x (µg m ⁻³)	113 ± 31	136 ± 29	123 ± 39	119 ± 29	113 ± 26	NA	NA	125 ± 24	128 ± 46	100 ± 43	127 ± 44	114 ± 32

^a These data were reported by our previous study (He et al., 2025). ^b The boundary layer height (BLH) data used in this study were obtained from the ERA5 reanalysis dataset provided by the European Centre for Medium-Range Weather Forecasts (ECMWF) via the Copernicus Climate Data Store (CDS, <https://cds.climate.copernicus.eu/datasets/>). The dataset has a spatial resolution of 0.25° × 0.25° and an hourly temporal resolution. BLH represents the height of the planetary boundary layer, defined as the atmospheric layer affected by surface friction and turbulence. ‘NA’ means the data are not available in this study.

Table S6. Meteorological parameters, PM2.5 main components, organic molecular tracers, diacids, pH, and ALWC in the PRD (IT0-IT4).

	IT0	IT1	IT2	IT3	IT4
	<i>N</i> =129	<i>N</i> =144	<i>N</i> =72	<i>N</i> =84	<i>N</i> =33
I. Meteorological parameters					
Temperature (°C)	20.2 ± 2.9	21.5 ± 3.6	21.6 ± 3.4	22.8 ± 3.1	20.8 ± 4.8
Relative humidity (%)	56 ± 12.4	56 ± 13	62 ± 10	67 ± 9	66 ± 7
Solar radiation (W m ⁻²)	148.0 ± 43.9	145.6 ± 42.6	118.0 ± 46	115.5 ± 43.4	112.0 ± 50.5
Boundary layer height (m)	578 ± 159	578 ± 134	613 ± 167	583 ± 142	626 ± 154
II. Molecular tracers (ng m⁻³)					
Levoglucosan	333 ± 225	194 ± 131	114 ± 79	96 ± 74	63 ± 34
Hopanes	3.4 ± 2.6	2.0 ± 1.6	1.3 ± 1.9	0.88 ± 0.70	0.54 ± 0.30
Octadecanoic acid	37.5 ± 21.0	28.4 ± 17.2	22.3 ± 14.8	17.3 ± 8.7	11.3 ± 0.93
Picene	0.26 ± 0.20	0.22 ± 0.15	0.18 ± 0.11	0.17 ± 0.10	0.10 ± 0.04
Terephthalic acid	50.0 ± 46.8	48.9 ± 30.7	32.1 ± 31.3	27.9 ± 27.1	14.5 ± 12.4
Phthalic acid	40.3 ± 17.8	29.2 ± 16.0	22.7 ± 10.2	19.6 ± 10.1	14.1 ± 8.8
DHOPA	2.52 ± 2.28	2.27 ± 2.07	1.42 ± 1.06	1.05 ± 1.01	0.78 ± 0.43
Malic acid	19.0 ± 19.0	16.6 ± 16.4	9.6 ± 8.3	7.4 ± 6.1	3.9 ± 2.3
III. Aliphatic Diacids (ng m⁻³)					
Oxalic acid (C ₂)	619 ± 290	483 ± 200	329 ± 158	293 ± 125	189 ± 102
Succinic acid (C ₄)	55.0 ± 49.5	29.3 ± 28.5	18.5 ± 14.2	16.7 ± 12.7	12.9 ± 12.1
Glutaric acid (C ₅)	12.5 ± 10.5	6.4 ± 5.9	4.8 ± 2.7	4.2 ± 4.2	4.5 ± 5.6
Adipic acid (C ₆)	7.1 ± 4.2	4.9 ± 3.4	4.0 ± 2.7	3.4 ± 2.5	2.9 ± 2.6
Pimelic acid (C ₇)	1.9 ± 1.3	1.4 ± 0.8	1.1 ± 0.7	1.1 ± 0.9	0.7 ± 0.5
Suberic acid (C ₈)	3.0 ± 2.2	2.5 ± 1.5	2.2 ± 1.3	2.0 ± 1.3	1.4 ± 1.0
Azelaic acid (C ₉)	13.5 ± 12.3	11.9 ± 8.3	10.4 ± 7.0	9.6 ± 6.1	6.7 ± 3.8
Sebacic acid (C ₁₀)	2.0 ± 1.8	1.7 ± 1.2	1.6 ± 1.3	1.5 ± 1.1	1.0 ± 0.9
Subtotal	734 ± 337	540 ± 218	358 ± 163	325 ± 135	208 ± 67
IV. Other species					
pH	2.04 ± 0.96	2.40 ± 0.61	2.48 ± 0.43	2.36 ± 0.58	2.11 ± 0.71
ALWC (μg m ⁻³)	20.9 ± 11.0	15.1 ± 9.9	13.1 ± 6.9	13.1 ± 8.0	7.2 ± 3.0
O _x (μg m ⁻³)	136.7 ± 31.7	134.9 ± 34.4	111.9 ± 27.1	98.5 ± 25.0	72.7 ± 19.1

Table S7. Correlations between C₂ and various factors under different pollution levels.

	IT0	IT1	IT2	IT3	IT4
Levogluconan	0.17 (-0.05, 0.37)	-0.03 (-0.22, 0.16)	-0.10 (-0.36, 0.16)	0.01 (-0.23, 0.26)	-0.29 (-0.61, 0.11)
Hopanes	-0.04 (-0.25, 1.08)	-0.21 (-0.38, -0.01) *	-0.05 (-0.31, 0.22)	0.29 (0.05, 0.49) *	0.41 (-0.01, 0.70) *
Octadecanoic acid	0.54 (0.36, 0.68) **	-0.03 (-0.22, 0.16)	0.01 (-0.26, 0.27)	-0.03 (-0.26, 0.22)	0.17 (-0.23, 0.52)
Picene	0.06 (-0.17, 0.28)	-0.28 (-0.46, -0.07) *	-0.18 (-0.45, 0.12)	0.08 (-0.25, 0.39)	0.02 (-0.62, 0.64)
Terephthalic acid	0.40 (0.20, 0.57) **	0.23 (0.04, 0.40) *	0.43 (0.19, 0.62) **	0.34 (0.11, 0.54) *	0.41 (0.04, 0.69) *
Phthalic acid	0.63 (0.47, 0.74) **	0.28 (0.01, 0.45) **	0.44 (0.20, 0.63) **	0.34 (0.11, 0.54) **	0.31 (0.01, 0.54) **
DHOPA	0.19 (-0.13, 0.30) *	0.49 (0.29, 0.60) **	0.45 (0.21, 0.64) **	0.42 (0.20, 0.61) **	0.32 (-0.01, 0.65) **
Malic acid	0.33 (0.13, 0.52) *	0.53 (0.38, 0.66) **	0.66 (0.48, 0.77) **	0.69 (0.44, 0.75) **	0.72 (0.45, 0.87) **
O_x	0.28 (0.05, 0.48) *	0.54 (0.37, 0.68) **	0.56 (0.25, 0.70) **	0.51 (0.42, 0.75) **	0.68 (0.39, 0.84) **
J(O1D)	0.366 (0.15, 0.53) **	0.17 (-0.03, 0.36)	0.33 (0.05, 0.56) *	0.13 (-0.12, 0.37)	-0.09 (-0.49, 0.34)
J(NO2)	0.29 (0.08, 0.48) **	0.14 (-0.07, 0.33)	0.49 (0.24, 0.68) **	0.22 (-0.03, 0.45)	0.02 (-0.40, 0.44)
Sulfate	0.49 (0.28, 0.62) **	0.29 (0.12, 0.46) **	0.60 (0.43, 0.74) **	0.42 (0.21, 0.59) **	0.55 (0.24, 0.76) **
ALWC	0.48 (0.31, 0.65) **	0.36 (0.19, 0.50) **	0.32 (0.09, 0.53) **	0.30 (0.08, 0.49) **	0.15 (-0.01, 0.31)
pH	-0.19 (-0.39, 0.03)	-0.15 (-0.32, 0.03)	-0.38 (-0.57, -0.16) **	-0.01 (-0.24, 0.22)	-0.19 (-0.54, 0.21)
Temperature	0.24 (0.02, 0.43) *	0.42 (0.27, 0.56) **	0.50 (0.30, 0.67) **	0.40 (0.19, 0.58) **	0.63 (0.35, 0.81) **
RH	0.15 (-0.06, 0.36)	0.28 (0.11, 0.44) **	-0.03 (-0.21, 0.26)	-0.03 (-0.19, 0.26)	-0.03 (-0.39, 0.33)
SR	-0.01 (-0.23, 0.21)	0.13 (-0.06, 0.30)	0.43 (0.21, 0.61) **	0.42 (0.21, 0.59) **	0.53 (0.22, 0.75) **

The values in brackets indicate the 95% confidence intervals (CIs) of the correlation coefficients. One, two asterisks denote p values less than 0.05, 0.01, respectively. No asterisk denotes the correlations are not statistically significant.

Table S8. Significance (p values) of the difference between correlation coefficients in different categories (C₂-ALWC).

	IT0	IT1	IT2	IT3
IT1	0.25			
IT2	0.21	0.76		
IT3	0.15	0.64	0.89	
IT4	< 0.05	0.22	0.37	0.43

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Table S9. Significance (p values) of the difference between correlation coefficients in different categories (C₂-O_x).

	IT0	IT1	IT2	IT3
IT1	< 0.01			
IT2	< 0.05	0.84		
IT3	< 0.01	0.47	0.65	
IT4	< 0.01	0.22	0.33	0.55

Table S10. Impact factor (IF, %) of individual factor under different pollution conditions

	IT0	IT1	IT2	IT3	IT4
Levogluconan	3 ± 3	2 ± 3	2 ± 1	1 ± 1	1 ± 0
Hopanes	2 ± 2	3 ± 2	3 ± 2	3 ± 1	2 ± 1
Octadecanoic acid	3 ± 2	1 ± 1	1 ± 1	1 ± 1	1 ± 0
Picene	1 ± 1	1 ± 1	1 ± 1	1 ± 1	0 ± 0
Terephthalic acid	6 ± 6	7 ± 6	8 ± 5	9 ± 8	6 ± 3
O _x	28 ± 15	37 ± 13	35 ± 11	36 ± 12	44 ± 3
J(O ¹ D)	8 ± 5	10 ± 7	6 ± 3	8 ± 6	9 ± 7
J(NO ₂)	1 ± 1	2 ± 1	1 ± 1	1 ± 1	2 ± 2
ALWC	8 ± 6	7 ± 6	6 ± 3	7 ± 5	3 ± 2
pH	6 ± 7	3 ± 2	3 ± 3	3 ± 5	1 ± 1
SO ₄ ²⁻	28 ± 10	20 ± 8	28 ± 8	24 ± 9	25 ± 6
Temp	5 ± 4	5 ± 3	4 ± 3	3 ± 1	3 ± 2
RH	1 ± 1	1 ± 1	1 ± 1	0 ± 0	0 ± 0
SR	2 ± 2	2 ± 2	2 ± 2	2 ± 3	1 ± 1

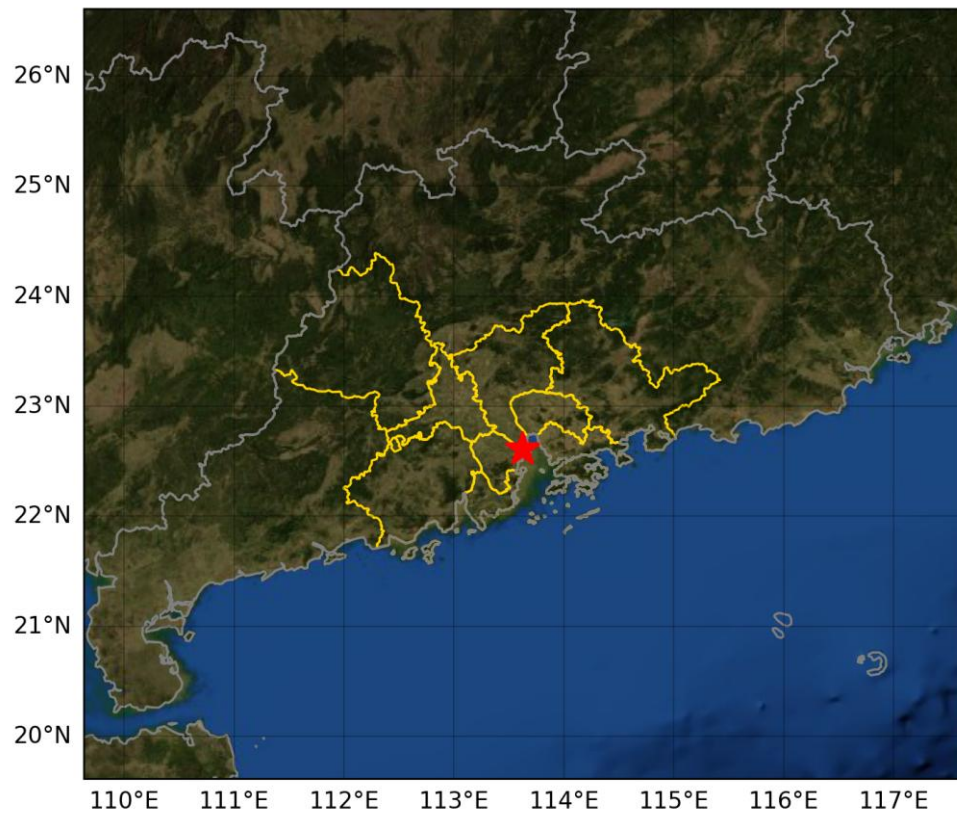


Figure S1. PRD region (golden line) consists of Guangzhou, Shenzhen, Zhuhai, Dongguan, Foshan, Huizhou, Zhongshan, Zhaoqing, Jiangmen, Hong Kong, and Macao. The measurement station (red star) is located in the central area of the PRD. The map background is based on NASA Blue Marble imagery. NASA (public domain).

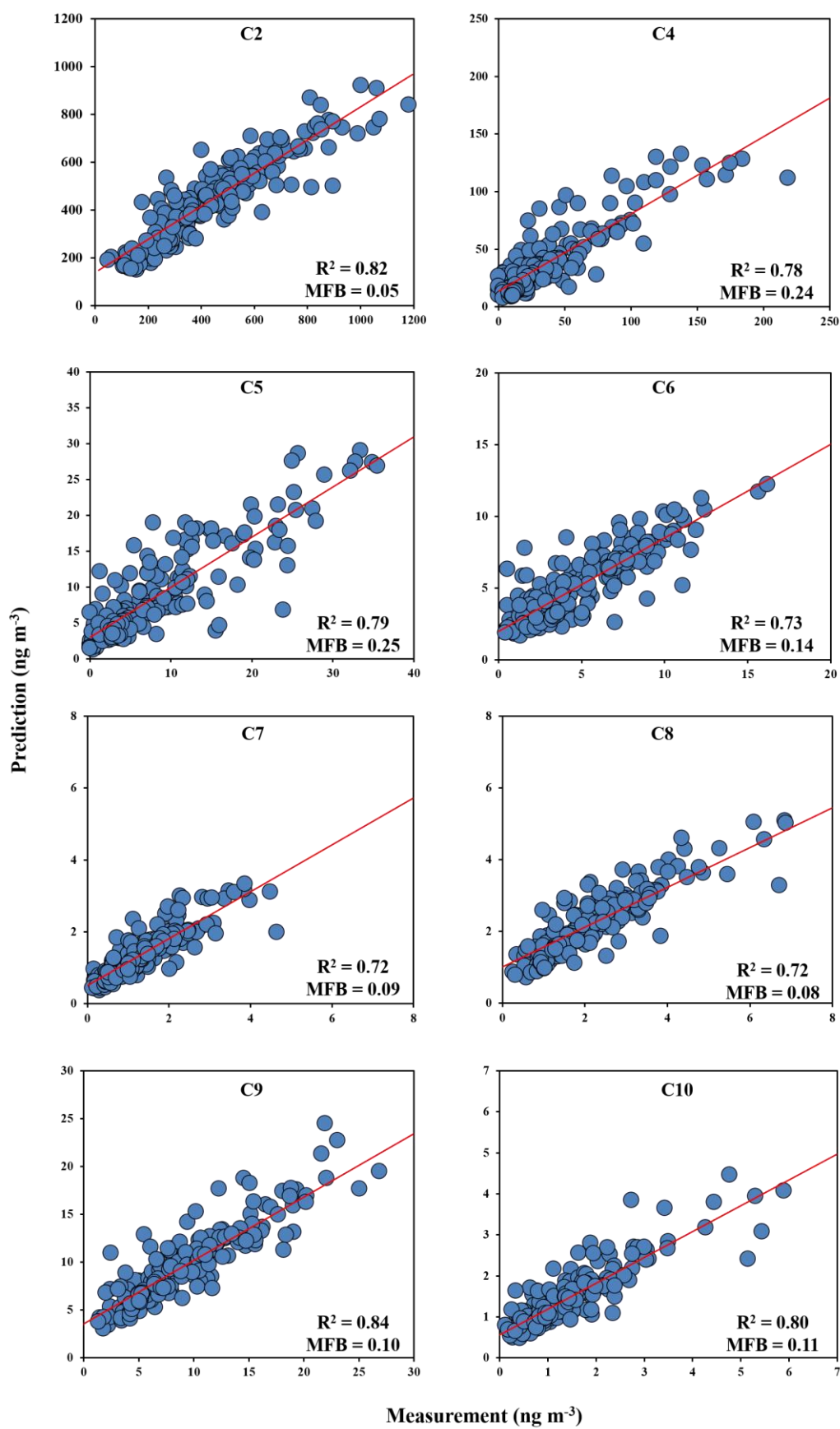


Figure S2. Observations and simulations of DCA.

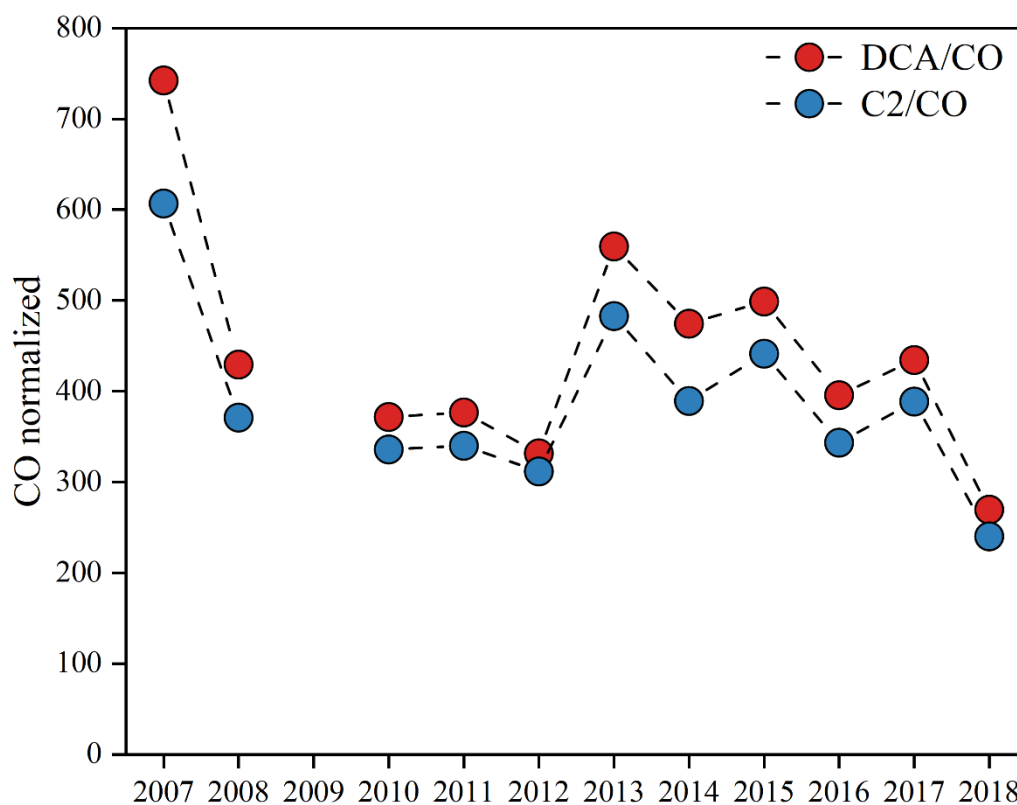
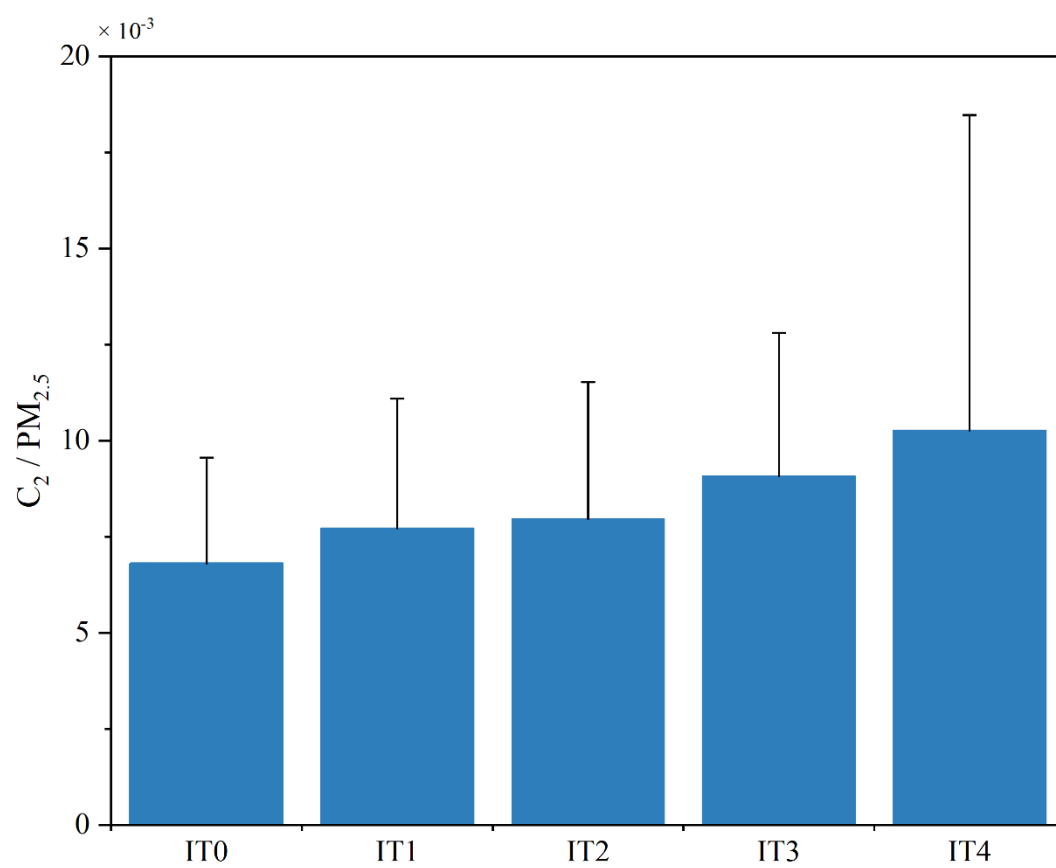


Figure S3. The concentrations of DCA and C₂ normalized by carbon monoxide (CO, ppm). Due to the lack of in situ CO measurements at the sampling site, monthly CO data were obtained from the Copernicus Atmosphere Monitoring Service (CAMS) global reanalysis product (EAC4), provided by the European Centre for Medium-Range Weather Forecasts (ECMWF, <https://cds.climate.copernicus.eu/datasets>). The dataset has a horizontal resolution of approximately $0.75^{\circ} \times 0.75^{\circ}$.

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40 Figure S4. The ratio of C_2 to $PM_{2.5}$ under different pollution conditions.

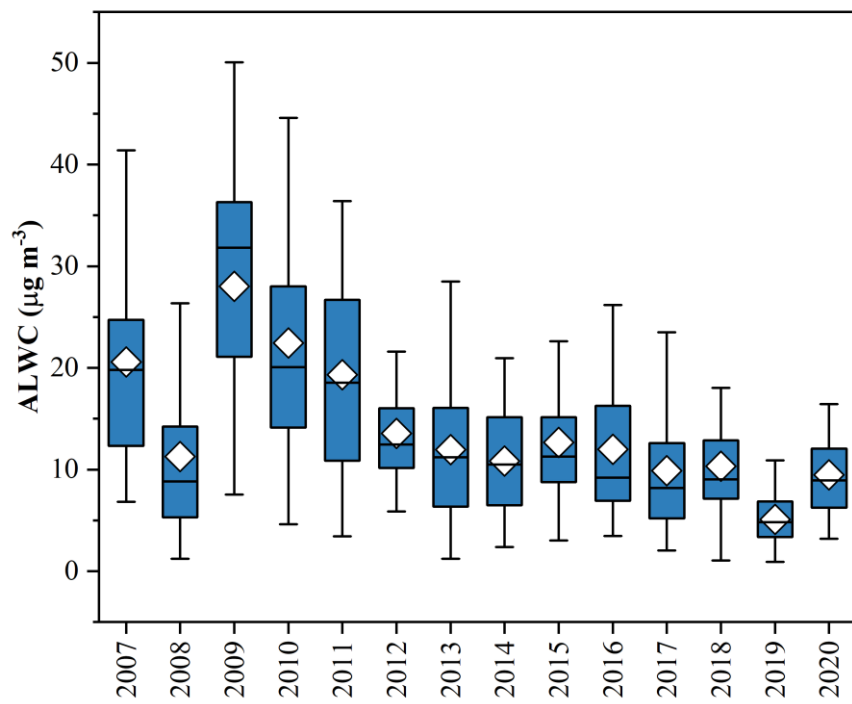


Figure S5. The variations in ALWC from 2007 to 2018 in the PRD (He et al., 2025).

- Bikkina, S., Kawamura, K., and Sarin, M.: Secondary Organic Aerosol Formation over Coastal Ocean: Inferences from Atmospheric Water-Soluble Low Molecular Weight Organic Compounds, *Environ. Sci. Technol.*, 51, 4347-4357, <https://doi.org/10.1021/acs.est.6b05986>, 2017.
- Bikkina, S., Kawamura, K., Miyazaki, Y., and Fu, P. Q.: High abundances of oxalic, azelaic, and glyoxylic acids and methylglyoxal in the open ocean with high biological activity: Implication for secondary OA formation from isoprene, *Geophys. Res. Lett.*, 41, 3649-3657, <https://doi.org/10.1002/2014gl059913>, 2014.
- Deshmukh, D. K., Kawamura, K., and Deb, M. K.: Dicarboxylic acids, ω -oxocarboxylic acids, α -dicarbonyls, WSOC, OC, EC, and inorganic ions in wintertime size-segregated aerosols from central India: Sources and formation processes, *Chemosphere*, 161, 27-42, <https://doi.org/10.1016/j.chemosphere.2016.06.107>, 2016.
- Deshmukh, D. K., Mozammel Haque, M., Kawamura, K., and Kim, Y.: Dicarboxylic acids, oxocarboxylic acids and α -dicarbonyls in fine aerosols over central Alaska: Implications for sources and atmospheric processes, *Atmos. Res.*, 202, 128-139, <https://doi.org/10.1016/j.atmosres.2017.11.003>, 2018.
- Ding, Z., Du, W., Wu, C., Cheng, C., Meng, J., Li, D., Ho, K., Zhang, L., and Wang, G.: Summertime atmospheric dicarboxylic acids and related SOA in the background region of Yangtze River Delta, China: Implications for heterogeneous reaction of oxalic acid with sea salts, *Sci. Total Environ.*, 757, 143741, <https://doi.org/10.1016/j.scitotenv.2020.143741>, 2021.
- Du, W., Ding, Z. J., Lei, Y. L., Zhang, S., Wu, C., Zhang, F., Wang, F. L., Lv, S. J., Liu, X. D., Meng, J. J., and Wang, G. H.: Atmospheric fine particulate dicarboxylic acids and related SOA in winter at the background site of Yangtze River Delta: Implication for the long-distance transport of solid fuels burning, *Atmos. Environ.*, 289, <https://doi.org/10.1016/j.atmosenv.2022.119320>, 2022.
- Fisher, R. A. S.: On the "Probable Error" of a Coefficient of Correlation Deduced from a Small Sample, *Metron*,
- He, Y., Ding, X., He, Q., Zhang, Y., Chen, D., Zhang, T., Yang, K., Wang, J., Cheng, Q., Jiang, H., Wang, Z., Liu, P., Wang, X., and Boy, M.: Long-term Trends in PM_{2.5} Chemical Composition and Its Impact on Aerosol Properties: Field Observations from 2007 to 2020 in Pearl River Delta, South China, *Atmos. Chem. Phys.*, 25, 13729-13745, <https://doi.org/10.5194/acp-25-13729-2025>, 2025.
- Ho, K. F., Cao, J. J., Lee, S. C., Kawamura, K., Zhang, R. J., Chow, J. C., and Watson, J. G.: Dicarboxylic acids, ketocarboxylic acids, and dicarbonyls in the urban atmosphere of China, *J. Geophys. Res.: Atmos.*, 112, <https://doi.org/10.1029/2006JD008011>, 2007.
- Jung, J., Tsatsral, B., Kim, Y. J., and Kawamura, K.: Organic and inorganic aerosol compositions in Ulaanbaatar, Mongolia, during the cold winter of 2007 to 2008: Dicarboxylic acids, ketocarboxylic acids, and α -dicarbonyls, *J. Geophys. Res.: Atmos.*, 115, <https://doi.org/10.1029/2010JD014339>, 2010.
- Kunwar, B., Kawamura, K., Fujiwara, S., Fu, P., Miyazaki, Y., and Pokhrel, A.: Dicarboxylic acids, oxocarboxylic acids and α -dicarbonyls in atmospheric aerosols from Mt. Fuji, Japan: Implication for primary emission versus secondary formation, *Atmos. Res.*, 221, 58-71, <https://doi.org/10.1016/j.atmosres.2019.01.021>, 2019.
- Meng, J., Liu, X., Hou, Z., Yi, Y., Yan, L., Li, Z., Cao, J., Li, J., and Wang, G.: Molecular characteristics and stable carbon isotope compositions of dicarboxylic acids and related compounds in the urban atmosphere of the North China Plain: Implications for aqueous phase formation of SOA during the haze periods, *Sci. Total Environ.*, 705, 135256, <https://doi.org/10.1016/j.scitotenv.2019.135256>, 2020.

Meng, J., Wang, G., Li, J., Cheng, C., Ren, Y., Huang, Y., Cheng, Y., Cao, J., and Zhang, T.: Seasonal characteristics of oxalic acid and related SOA in the free troposphere of Mt. Hua, central China: Implications for sources and formation mechanisms, *Sci. Total Environ.*, 493, 1088-1097, <https://doi.org/10.1016/j.scitotenv.2014.04.086>, 2014.

Meng, J., Wang, Y., Li, Y., Huang, T., Wang, Z., Wang, Y., Chen, M., Hou, Z., Zhou, H., Lu, K., Kawamura, K., and Fu, P.: Measurement Report: Investigation on the sources and formation processes of dicarboxylic acids and related species in urban aerosols before and during the COVID-19 lockdown in Jinan, East China, *Atmos. Chem. Phys.*, 23, 14481-14503, <https://doi.org/10.5194/acp-23-14481-2023>, 2023.

Meng, J. J., Wang, G. H., Hou, Z. F., Liu, X. D., Wei, B. J., Wu, C., Cao, C., Wang, J. Y., Li, J. J., Cao, J. J., Zhang, E. X., Dong, J., Liu, J. Z., Ge, S. S., and Xie, Y. N.: Molecular distribution and stable carbon isotopic compositions of dicarboxylic acids and related SOA from biogenic sources in the summertime atmosphere of Mt. Tai in the North China Plain, *Atmos. Chem. Phys.*, 18, 15069-15086, <https://doi.org/10.5194/acp-18-15069-2018>, 2018.

Wang, G., Kawamura, K., Cheng, C., Li, J., Cao, J., Zhang, R., Zhang, T., Liu, S., and Zhao, Z.: Molecular Distribution and Stable Carbon Isotopic Composition of Dicarboxylic Acids, Ketocarboxylic Acids, and α -Dicarbonyls in Size-Resolved Atmospheric Particles From Xi'an City, China, *Environ. Sci. Technol.*, 46, 4783-4791, <https://doi.org/10.1021/es204322c>, 2012.

Yu, Q., Chen, J., Cheng, S., Qin, W., Zhang, Y., Sun, Y., and Ahmad, M.: Seasonal variation of dicarboxylic acids in PM_{2.5} in Beijing: Implications for the formation and aging processes of secondary organic aerosols, *Sci. Total Environ.*, 763, 142964, <https://doi.org/10.1016/j.scitotenv.2020.142964>, 2021.

Zhang, Y. L., Kawamura, K., Fu, P. Q., Boreddy, S. K. R., Watanabe, T., Hatakeyama, S., Takami, A., and Wang, W.: Aircraft observations of water-soluble dicarboxylic acids in the aerosols over China, *Atmos. Chem. Phys.*, 16, 6407-6419, <https://doi.org/10.5194/acp-16-6407-2016>, 2016.