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Bayesian inference-based estimation of hourly primary and secondary organic carbon in suburban Hong Kong: multi-temporal-scale variations and evolution characteristics during $PM_{2.5}$ episodes

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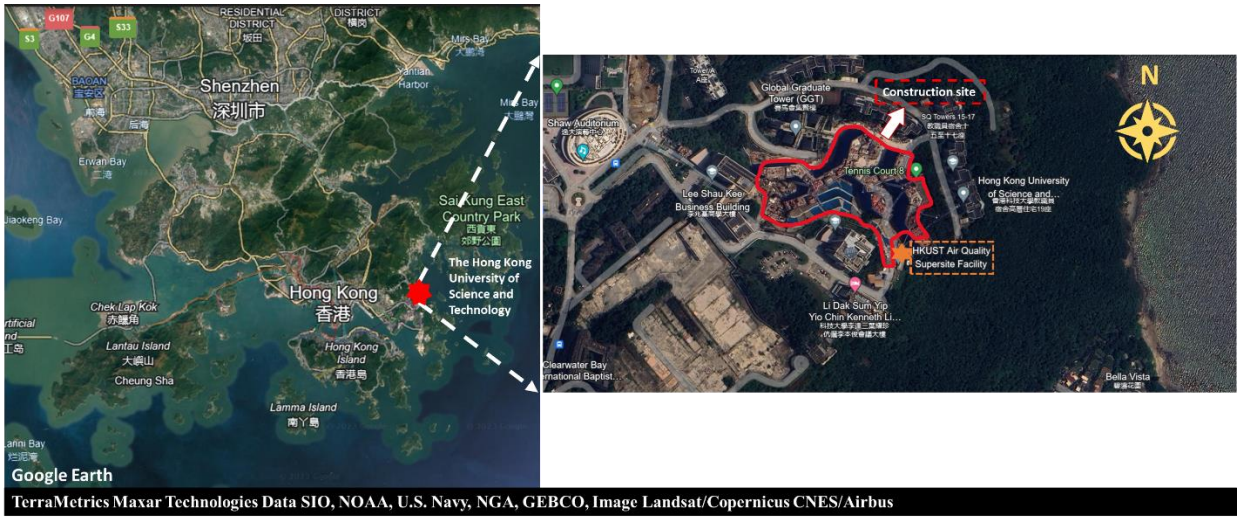

Figure S1. Location of sampling site and the surrounding environment (map data: © Google Earth, TerraMetrics Maxar Technologies Data SIO, NOAA, U.S. Navy, NGA, GEBCO, Image Landsat/Copernicus CNES/Airbus).

S1 The $PM_{2.5}$ correction

The hourly $PM_{2.5}$ concentrations measured by SHARP were biased, evidenced by the measured SHARP $PM_{2.5}$ concentrations consistently falling below the reconstructed speciation data (Figure S2a). The magnitude of bias was determined by comparing the SHARP-measured $PM_{2.5}$ with those 24-h offline filter measurements during the campaign period. The SHARP $PM_{2.5}$ showed similar temporal variations with the offline filter measurements (Figure S2b). Strong correlations were observed between SHARP $PM_{2.5}$ and filter-based $PM_{2.5}$, with R_p values ranging from 0.74 to 0.98. The SHARP $PM_{2.5}$ levels were corrected using the linear relationships shown in Figure S2c.

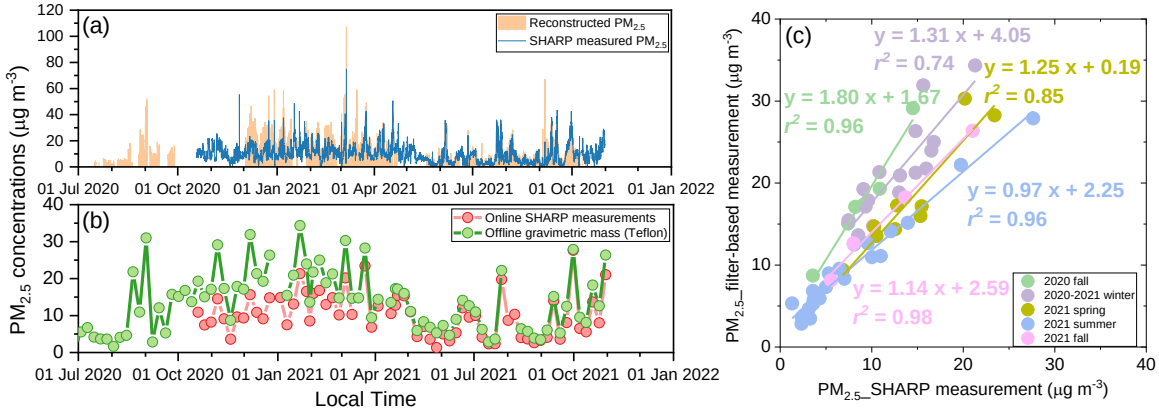

Figure S2. (a) Time series of hourly reconstructed $PM_{2.5}$ and SHARP measured $PM_{2.5}$. (b) Time series of daily SHARP measured $PM_{2.5}$ and offline gravimetric mass (Teflon). (c) Linear relationship between online and offline filter-based $PM_{2.5}$ fata during the campaign period.

S2 Aerosol water content and pH verification

Figure S3 shows the comparisons of predicted and measured HNO_3, NO_3^-, and $\varepsilon(HNO_3)$, and NH_3, NH_4^+ and $\varepsilon(NH_4^+)$ for summer and winter episode cases. The gas-particle phase partitioning of HNO_3, namely $\varepsilon(HNO_3)$, is described as gas-phase HNO_3 concentration divided by the sum of aerosol-phase NO_3 and gas-phase HNO_3. Similarly, the $\varepsilon(NH_4^+)$ is calculated as aerosol-phase NH_4^+ divided by the sum of aerosol-phase NH_4^+ and gas-phase NH_3.

The partitioning ratios for summer and winter episode cases were as follows: $\varepsilon(NH_4^+) = 49 \pm 18\%$ and $61 \pm 17\%$ for summer and winter, $\varepsilon(HNO_3) = 28 \pm 9.2\%$ and $17 \pm 8.0\%$ for summer and winter, respectively. Good correlations were observed between the predicted and measured gas-phase HNO_3, aerosol-phase NO_3^- and $\varepsilon(HNO_3)$, with R_p ranging from 0.56-0.86. Despite the good correlations, ISOROPPIA-II predicted gas-phase HNO_3 was systematically higher than measured value, with regression slopes of larger than 2.0. This could be attributed to the underestimate of HNO_3 measured by MARGA system (Makkonen et al., 2014). Regarding the aerosol-phase NO_3^-, the ISOROPPIA-II underpredicted the values with regression slope of 0.62 and 0.82 in summer and winter, respectively. This leads to a regression slope of higher than 1 comparing predicted to measured $\varepsilon(HNO_3)$. In contrast, predicted versus measured gas-phase NH_3, aerosol-phase NH_4^+ and $\varepsilon(NH_4^+)$ is close to 1:1 and highly correlated (R_p: 0.77-0.96). The overall good correlations between the predicted and measured values of NH_4^+, NO_3^-, gas-phase NH_3 and $\varepsilon(NH_4^+)$ suggest the precision and consistent utility of thermodynamic equilibrium model in the quantification of $PM_{2.5}$ aerosol water content and acidity.

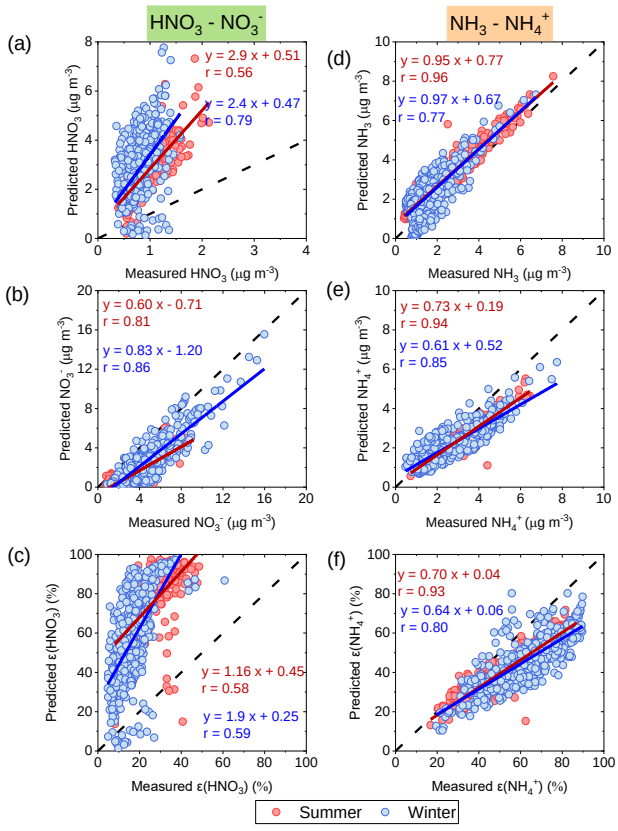

Figure S3. Comparisons of (a, b, c) predicted and measured HNO_3, NO_3^-, and $\varepsilon(NO_3^-)$; and (d, e, f) NH_3, NH_4^+, and $\varepsilon(NH_4^+)$ for summer and winter episode cases.

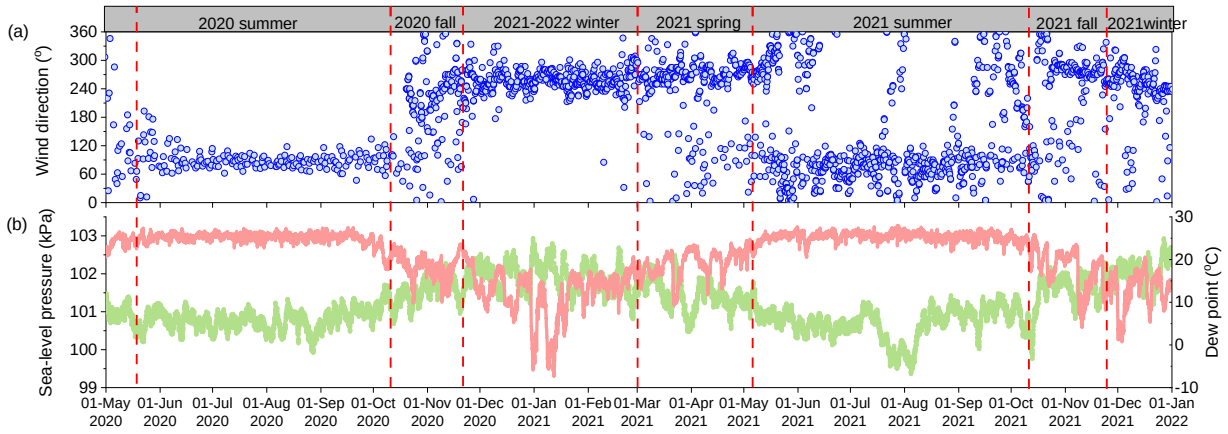

Figure S4. Time series of (a) upper wind direction at 15000 ± 1000 m height, and (b) sea-level pressure and dew point at Hong Kong Observatory station from 1 May 2021 to 31 December 2021.

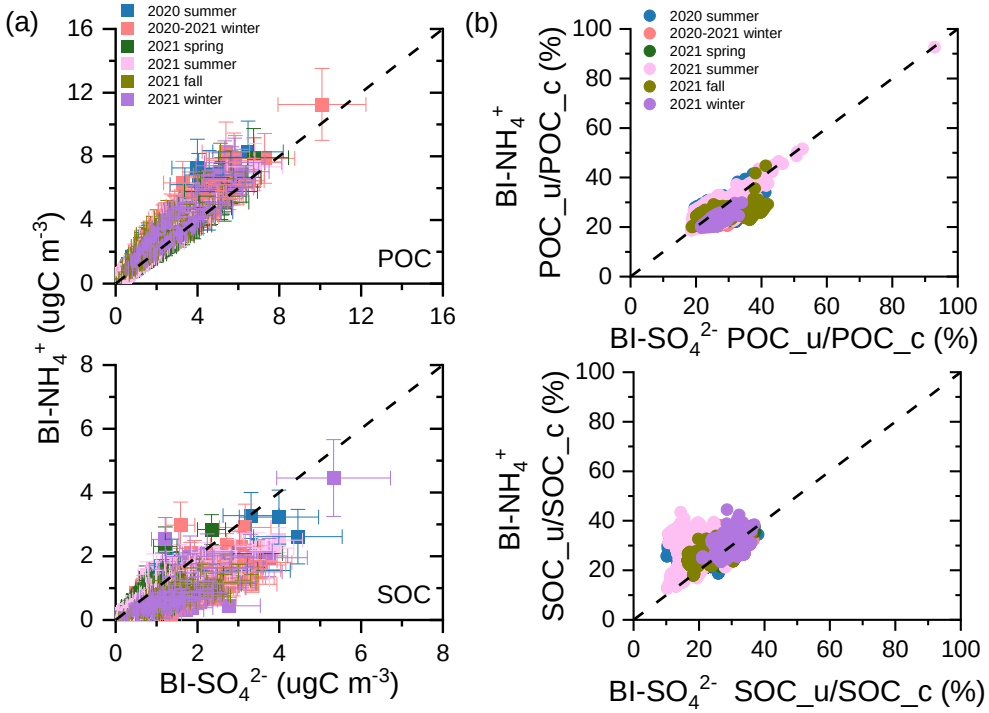

Figure S5. Comparison of (a) absolute concentration and absolute uncertainties (represented in error bar), and (b) relative uncertainty of POC and SOC between BI with sulfate and ammonium as tracers for SOC.

Table S1. Model performance evaluation metrics (Bayesian Inference Criterion, BIC)[a] for individual seasons using sulfate, nitrate and ammonium as tracers for SOC.

Tracers / Period	SO_4^{2-}	NO_3^-	NH_4^+
2020 summer	**2247**	3150	2713
2020-2021 winter	**6266**	6768	7065
2021 spring	**10525**	10731	13450
2021 summer	15951	16491	**14511**
2021 fall	**2614**	3793	4117
2021 winter	**2445**	2449	2600
Entire	**40047**	43382	44456

[a]Note: bold numbers are the minimum value, represents the most optimal model performance.

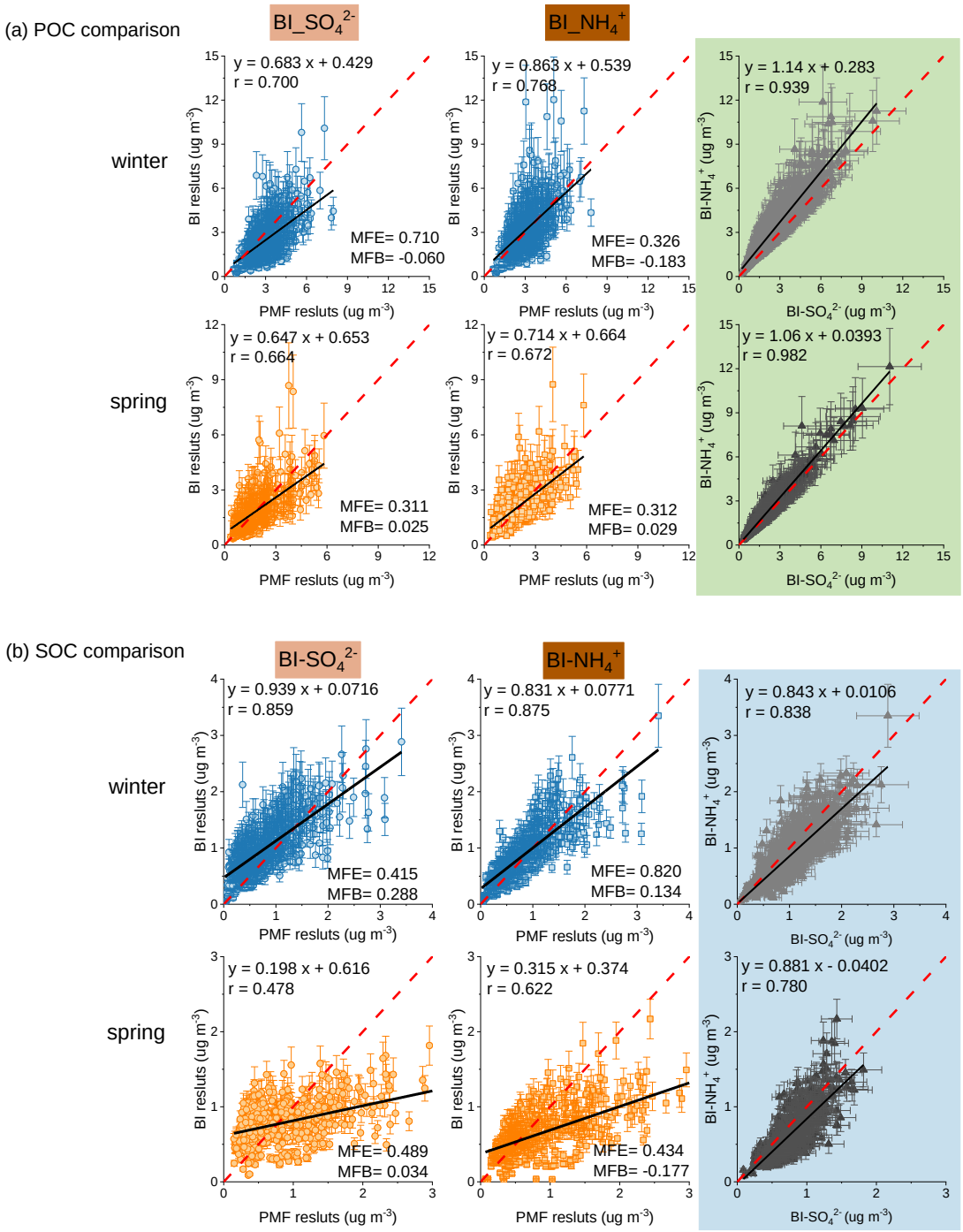

Figure S6. (a) POC and (b) SOC estimations for the HKUST winter (24 November 2020 – 28 February 2021) and spring (1 March -2 May 2021) data. Results comparison between PMF and BI with sulfate and ammonium as tracers for SOC.

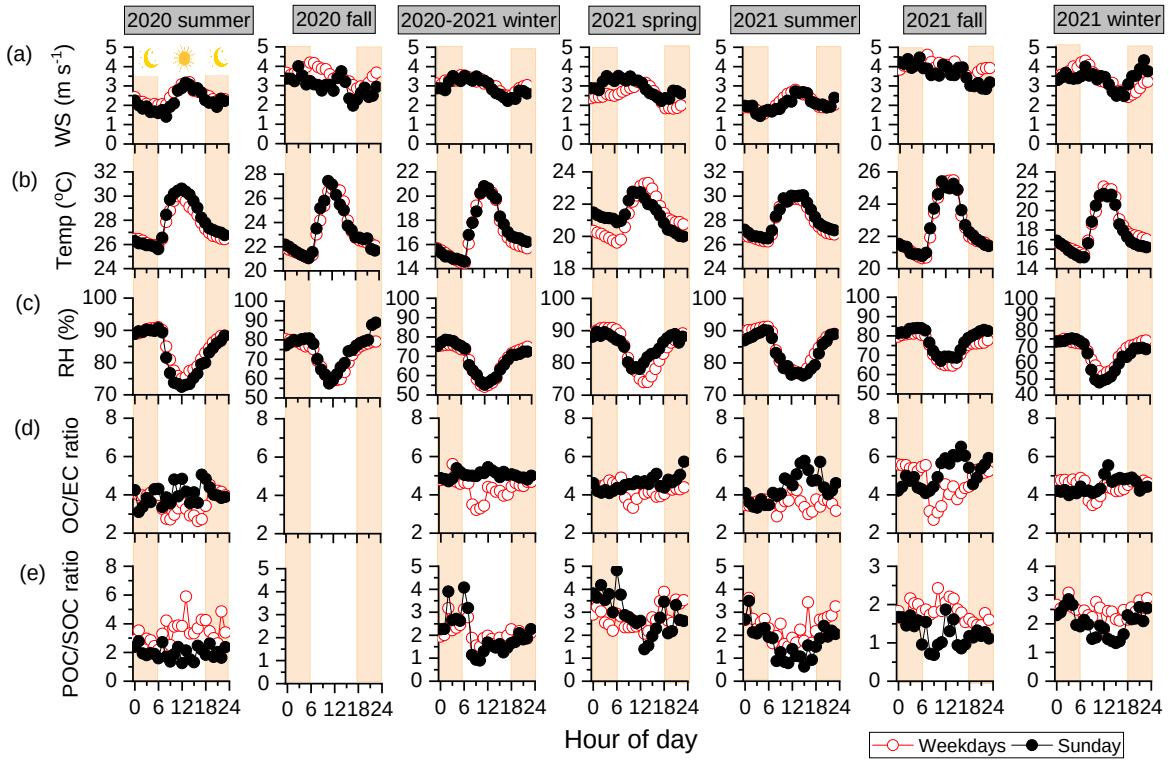

Figure S7. Diurnal variations of (a) wind speed, (b) temperature, (c) RH, and (d) OC/EC ratio and (e) POC/SOC ratio over the entire measurement period. The circles represent the hourly data averaged over weekdays (Monday -Saturday, red) and Sunday (black). The yellow shades represent nighttime periods.

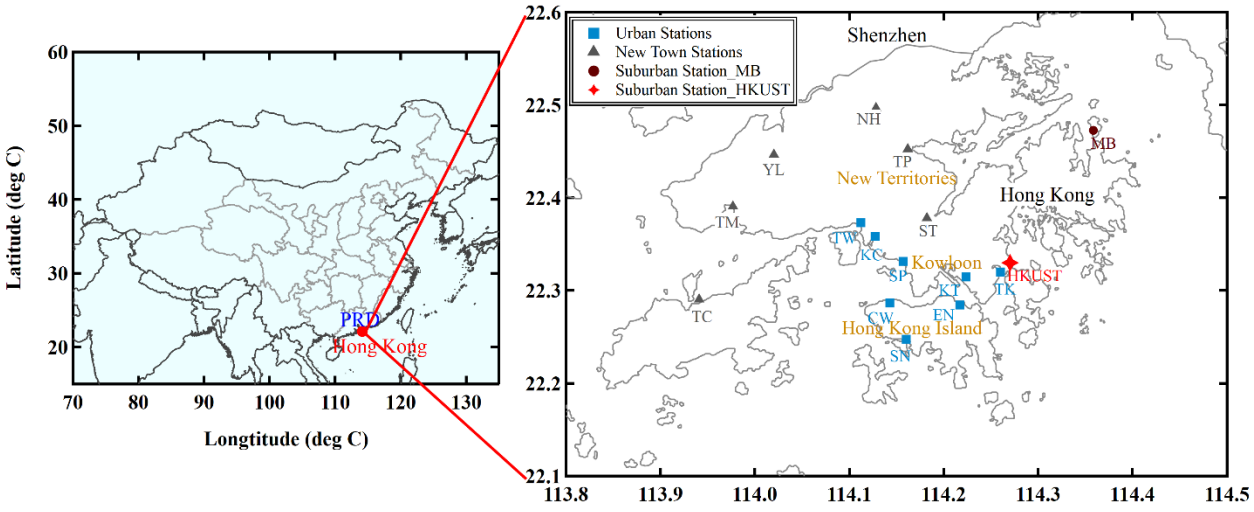

Figure S8. Geographical locations of the 15 HKEPD general air quality monitoring stations and the HKUST supersite.

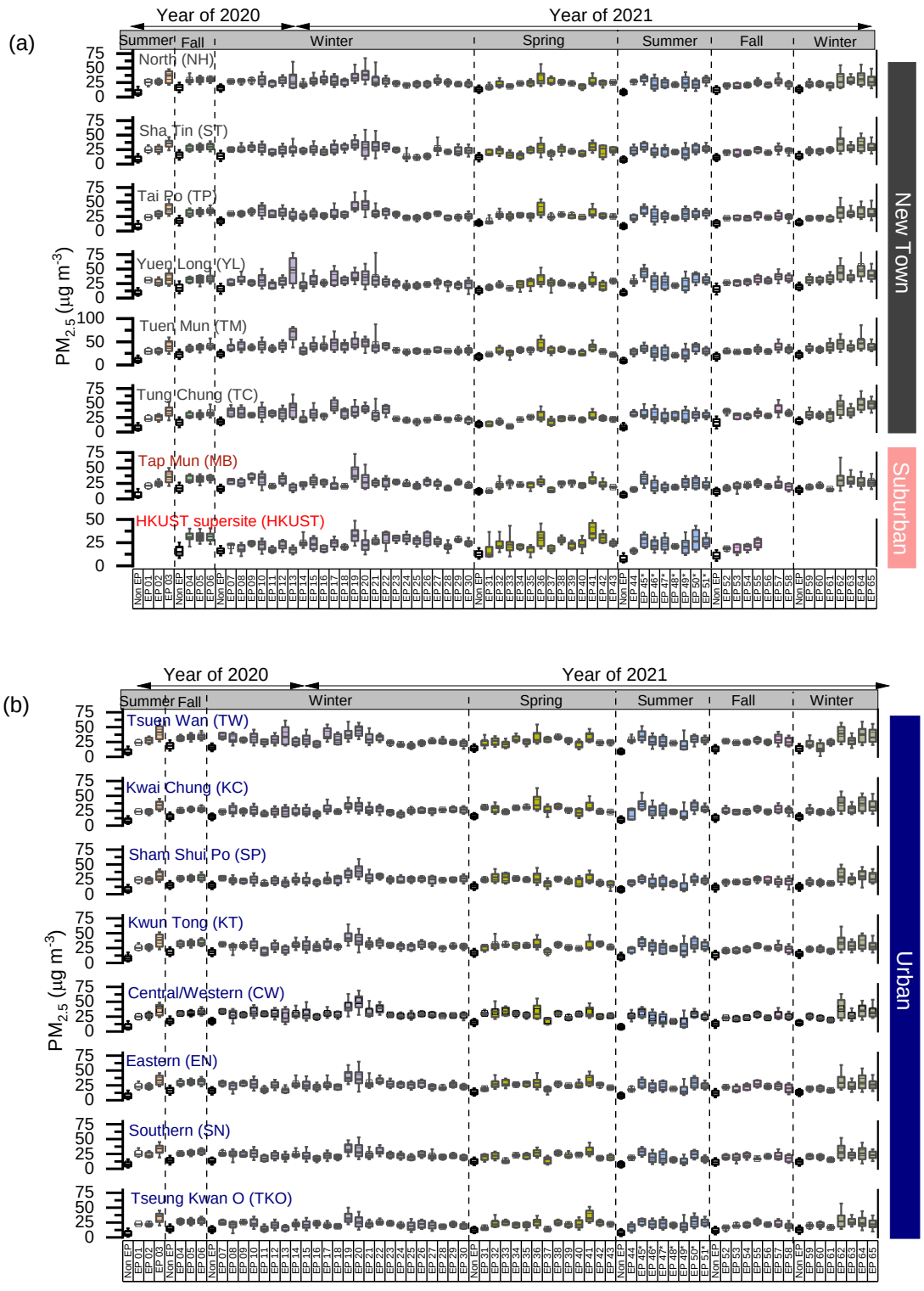

Figure S9. The average concentrations of $PM_{2.5}$ during individual episodes (solid pattern) and those during the non-episode hours in the same season (blank pattern) in the 15 HKEPD general air quality monitoring stations and the HKUST supersite in HK.

Table S2. Statistical summary of $PM_{2.5}$ at 15 Hong Kong EPD general air quality monitoring stations and the HKUST supersite during the 65 episodes and the remaining non-episode hours for the period from 16 July 2020 to 31 December 2021. Meteorological parameters and gas pollutant data are from the HKUST supersite. The NO_x and O_3 at MB station were used as reference during the missing period.

Episode	Season	Time period (date, time)	Duration (h)	Wind speed ($m\ s^{-1}$)	T (^oC)	RH (%)	O_3 (ppb)	NOx (ppb)	Mean $PM_{2.5}$ concentrations			
									City-wide	Min	Max	Max-to-min
EP01	2020 summer	21 Aug, 09:00-22 Aug, 22:00	38	1.71	28.9	78.6	33.4	4.8	24.9	21.1	31.0	1.5
EP02	2020 summer	29 Aug, 10:00-31 Aug, 23:00	38	1.64	29.4	76.2	58.7	5.4	25.7	22.1	30.1	1.4
EP03	2020 summer	01 Sep, 12:00-04 Sep, 23:00	84	1.36	29.5	79.2	60.2	7.1	36.0	30.9	41.7	1.3
Non-EP	2020 summer	16 July, 00:00-7 Oct, 23:00	1841	2.47	27.4	84.5	53.8	4.9	8.50	7.10	10.6	1.5
EP04	2020 fall	29 Oct, 19:00-30 Oct, 20:00	26	2.39	22.6	82.3	38.2	7.9	29.4	25.4	35.2	1.4
EP05	2020 fall	02 Nov, 09:00-04 Nov, 21:00	61	3.91	23.2	65.5	57.8	8.9	30.5	26.7	37.7	1.4
EP06	2020 fall	06 Nov, 11:00-10 Nov, 23:00	109	3.38	24.2	57.1	68.9	11	32.7	28.2	40.6	1.4
Non-EP	2020 fall	08 Oct, 00:00-23 Nov, 23:00	932	3.46	23.2	74.3	52.8	6.6	17.1	14.4	22.8	1.6
EP07	2020-2021 winter	24 Nov, 08:00-19:00	11	3.24	23.5	73.2	48.5	14	29.4	23.2	38.3	1.7
EP08	2020-2021 winter	26 Nov, 10:00-27 Nov, 03:00	18	2.01	23.2	79.1	59.8	8.0	27.7	20.2	38.8	1.9
EP09	2020-2021 winter	02 Dec, 19:00-03 Dec, 11:00	17	5.55	16.5	66.2	28.9	9.2	28.5	22.4	37.3	1.7
EP10	2020-2021 winter	05 Dec, 00:00-14 Dec 00:00	239	2.59	18.6	74.1	42.4	11	30.8	24.2	43.2	1.8
EP11	2020-2021 winter	17 Dec, 11:00-23:00	13	2.90	15.0	66.3	6.60	25	23.0	14.5	35.1	2.4
EP12	2020-2021 winter	19 Dec, 13:00-25 Dec 23:00	155	3.63	16.3	68.5	37.8	8.9	28.3	20.6	41.5	2.0
EP13	2020-2021 winter	27 Dec, 11:00-28 Dec, 11:00	25	1.89	20.5	58.0	61.5	5.1	29.3	15.6	65.8	4.2
EP14	2020-2021 winter	29 Dec, 00:00-30 Dec, 05:00	30	2.67	19.2	73.5	43.0	10	25.5	21.7	32.0	1.5
EP15	2020-2021 winter	03 Jan, 03:00-08 Jan, 23:00	141	3.44	14.4	64.0	45.6	8.1	29.1	22.6	42.2	1.9
EP16	2020-2021 winter	12 Jan, 00:00-23:00	24	5.52	11.4	22.0	40.4	10	25.1	17.2	41.5	2.4
EP17	2020-2021 winter	14 Jan, 05:00-23:00	19	1.81	15.0	45.7	64.7	12	30.5	20.4	47.8	2.3
EP18	2020-2021 winter	15 Jan, 15:00-16 Jan, 12:00	22	1.74	15.9	61.5	78.0	6.6	26.8	18.7	41.2	2.2
EP19	2020-2021 winter	16 Jan, 22:00-19 Jan, 11:00	62	3.57	14.0	56.7	54.6	7.7	39.2	33.1	49.2	1.5
EP20	2020-2021 winter	21 Jan, 22:00-24 Jan, 08:00	59	1.81	20.2	73.1	39.1	14	38.1	27.7	51.5	1.9
EP21	2020-2021 winter	25 Jan, 04:00-27 Jan, 21:00	66	1.76	17.6	78.5	56.2	9.3	30.9	22.7	44.1	1.9
EP22	2020-2021 winter	28 Jan, 08:00-23:00	16	3.16	20.3	55.1	60.0	7.9	31.7	25.2	41.1	1.6
EP23	2020-2021 winter	29 Jan, 21:00-30 Jan, 16:00	20	3.21	14.8	70.1	59.2	4.2	25.8	22.3	31.6	1.4
EP24	2020-2021 winter	02 Feb, 18:00-03 Feb, 11:00	18	3.78	16.3	78.2	57.8	3.7	22.4	12.8	28.5	2.2
EP25	2020-2021 winter	06 Feb, 10:00-08 Feb, 14:00	53	2.03	19.2	73.2	56.3	8.8	22.9	17.5	30.0	1.7
EP26	2020-2021 winter	09 Feb, 01:00-18:00	18	4.29	16.3	82.7	56.8	2.5	24.8	13.7	31.1	2.3
EP27	2020-2021 winter	14 Feb, 17:00-15 Feb, 13:00	21	1.51	20.6	60.9	51.8	9.5	25.9	19.7	32.1	1.6
EP28	2020-2021 winter	16 Feb, 20:00-17 Feb, 07:00	12	2.65	17.3	72.1	49.1	3.8	23.1	17.3	29.9	1.7
EP29	2020-2021 winter	17 Feb, 22:00-18 Feb, 23:00	26	3.06	16.6	68.6	49.5	3.7	24.8	21.7	29.9	1.4
EP30	2020-2021 winter	20 Feb, 12:00-22 Feb, 13:00	50	1.68	18.8	76.9	68.0	5.2	24.1	18.8	31.3	1.7
Non-EP	2020-2021 winter	24 Nov, 00:00-28 Feb, 23:00	1192	3.24	16.6	67.8	43.6	6.9	16.3	13.2	23.4	1.8
EP31	2021 spring	01 Mar, 20:00-02 Mar, 06:00	11	2.32	19.6	89.5	37.2	13	21.4	13.7	30.9	2.3
EP32	2021 spring	04 Mar, 20:00-05 Mar, 23:00	39	1.74	17.8	95.8	31.8	7.3	26.1	17.6	33.5	1.9
EP33	2021 spring	06 Mar, 09:00-20:00	12	1.34	18.9	99.9	34.5	5.5	22.6	9.60	34.1	3.6

EP34	2021 spring	14 Mar, 10:00-23:00	14	2.11	19.2	85.3	67.7	5.7	25.4	15.0	32.5	2.2
EP35	2021 spring	21 Mar, 07:00-14:00	8	4.57	20.3	74.0	18.6	12	25.8	20.8	32.5	1.6
EP36	2021 spring	22 Mar, 16:00-25 Mar, 23:00	80	2.60	19.0	64.6	45.7	14	31.7	25.8	41.9	1.6
EP37	2021 spring	04 Apr, 10:00-23:00	14	2.64	21.7	97.5	46.1	5.8	21.1	13.9	30.8	2.2
EP38	2021 spring	05 Apr, 19:00-07 Apr, 21:00	32	2.31	21.3	85.2	67.9	5.4	27.7	23.0	33.9	1.5
EP39	2021 spring	09 Apr, 21:00-10 Apr, 09:00	13	4.07	19.2	76.7	50.6	5.1	24.3	21.2	28.1	1.3
EP40	2021 spring	17 Apr, 10:00-21:00	12	1.80	21.1	93.3	53.4	7.3	22.6	18.0	29.3	1.6
EP41	2021 spring	18 Apr, 11:00-19 Apr, 16:00	30	4.67	20.9	72.2	80.1	5.5	31.6	26.8	39.1	1.5
EP42	2021 spring	23 Apr, 08:00-22:00	15	1.91	28.1	69.0	48.7	32	24.1	17.9	32.1	1.8
EP43	2021 spring	02 May, 00:00-12:00	13	1.26	25.1	78.4	46.0	13	23.0	16.6	29.4	1.8
Non-EP	2021 spring	01 Mar, 00:00-02 May, 23:00	1219	2.52	21.5	85.5	40.9	7.6	14.0	12.0	18.0	1.5
EP44	2021 summer	05 May, 14:00-23:00	10	2.18	26.0	81.5	70.3	19	23.0	15.2	31.0	2.0
EP45	2021 summer	05 June, 14:00-06 June, 23:00	34	1.65	27.2	73.3	81.4	18	32.8	25.3	44.5	1.8
EP46	2021 summer	23 July, 13:00-25 July, 16:00	52	1.50	29.7	78.2	71.3	17	24.0	19.6	29.3	1.5
EP47	2021 summer	27 July, 10:00-28 July, 17:00	32	1.15	30.4	78.6	43.9	19	23.1	18.6	27.2	1.5
EP48	2021 summer	06 Aug, 09:00-07 Aug, 13:00	20	1.49	28.0	87.7	26.5	25	21.1	16.0	26.9	1.7
EP49	2021 summer	11 Sep, 15:00-13 Sep, 18:00	52	1.35	31.0	76.5	51.4	16	21.6	13.4	29.8	2.2
EP50	2021 summer	17 Sep, 08:00-18 Sep, 03:00	20	1.67	28.9	83.5	79.6	7.7	28.8	21.8	36.5	1.7
EP51	2021 summer	27 Sep, 14:00-02 Oct, 03:00	110	1.43	28.7	81.2	55.3	11	26.7	16.1	31.8	2.0
Non-EP	2021 summer	03 May, 00:00-07 Oct, 23:00	3462	2.16	27.8	85.0	28.0	9.9	8.40	6.60	10.2	1.6
EP52	2021 fall	23 Oct, 15:00-23:00	9	3.44	19.8	75.6	20.4	21	22.8	16.0	35.6	2.2
EP53	2021 fall	27 Oct, 12:00-22:00	11	2.40	24.2	79.7	66.1	10	21.9	18.2	28.2	1.5
EP54	2021 fall	28 Oct, 12:00-29 Oct, 21:00	34	3.16	23.8	81.6	66.2	6.6	23.4	19.7	29.1	1.5
EP55	2021 fall	30 Oct, 10:00-01 Nov, 20:00	59	2.95	23.2	79.1	58.5	9.7	27.5	17.2	33.5	1.9
EP56	2021 fall	03 Nov, 10:00-04 Nov, 09:00	24	3.01	23.8	80.5	63.6	6.1	23.6	19.5	30.0	1.5
EP57	2021 fall	15 Nov, 22:00-18 Nov, 04:00	55	2.67	21.7	73.9	39.2	8.7	28.3	20.8	39.8	1.9
EP58	2021 fall	18 Nov, 17:00-20 Nov, 07:00	39	2.15	21.5	79.1	43.1	6.5	24.3	17.4	34.5	2.0
Non-EP	2021 fall	08 Oct, 00:00-24 Nov, 23:00	897	4.23	22.5	74.1	35.1	5.6	13.2	11.3	18.0	1.6
EP59	2021 winter	24 Nov, 14:00-27 Nov, 06:00	65	2.56	19.9	61.8	38.9	10	24.0	19.1	35.1	1.8
EP60	2021 winter	27 Nov, 18:00-01 Dec, 04:00	83	3.82	20.5	61.6	39.9	8.5	23.0	16.6	32.7	2.0
EP61	2021 winter	09 Dec, 14:00-10 Dec, 11:00	21	2.34	18.6	75.8	36.3	7.3	22.4	15.0	39.5	2.6
EP62	2021 winter	11 Dec, 15:00-15 Dec, 02:00	84	2.45	19.4	72.5	44.9	8.4	36.6	30.0	46.4	1.6
EP63	2021 winter	18 Dec, 19:00-20 Dec, 16:00	64	4.43	15.9	58.0	34.6	8.4	29.3	23.3	37.9	1.6
EP64	2021 winter	23 Dec, 19:00-26 Dec, 19:00	73	3.08	17.0	81.2	22.4	11	36.9	26.6	54.4	2.0
EP65	2021 winter	27 Dec, 12:00-31 Dec, 23:00	108	2.72	15.9	75.3	24.5	13	32.7	23.6	48.2	2.0
Non-EP	2021 winter	25 Nov, 00:00-31 Dec, 23:00	414	3.63	18.1	63.2	36.2	7.2	15.0	11.9	21.7	1.8

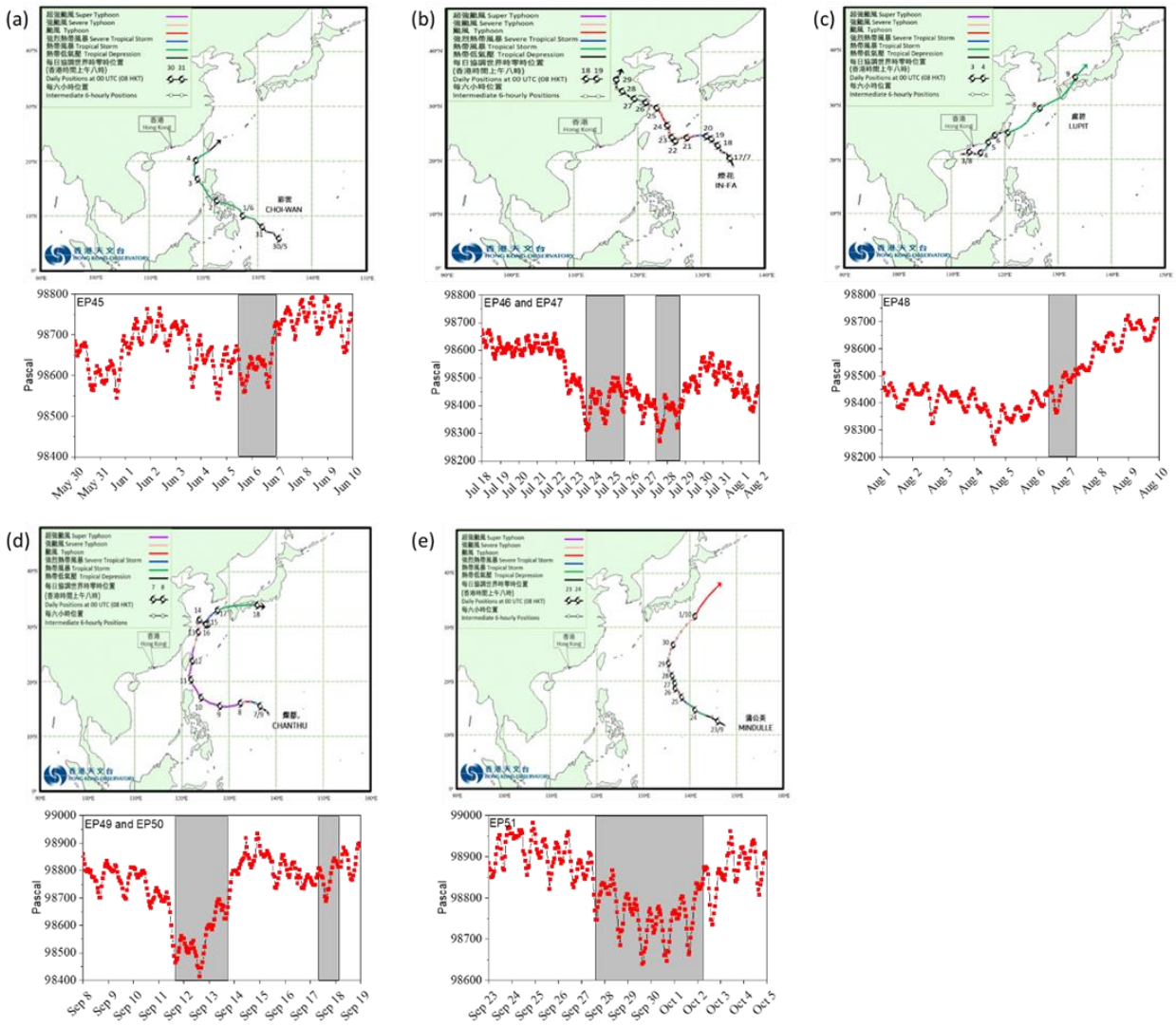

Figure S10. The provisional tropical cyclone tracks of (a) Chaiwan, (b) Infa, (c) Lupit, (d) Chanthu and (e) Mindule together with the corresponding atmospheric pressure (pascal) at the HKUST supersite. The episode period is indicated by the grey area. The tropical cyclone tracks were obtained from Hong Kong Observatory website (https://www.hko.gov.hk/en/publica/tc/tc2021/track.html, lass access: May 2023).

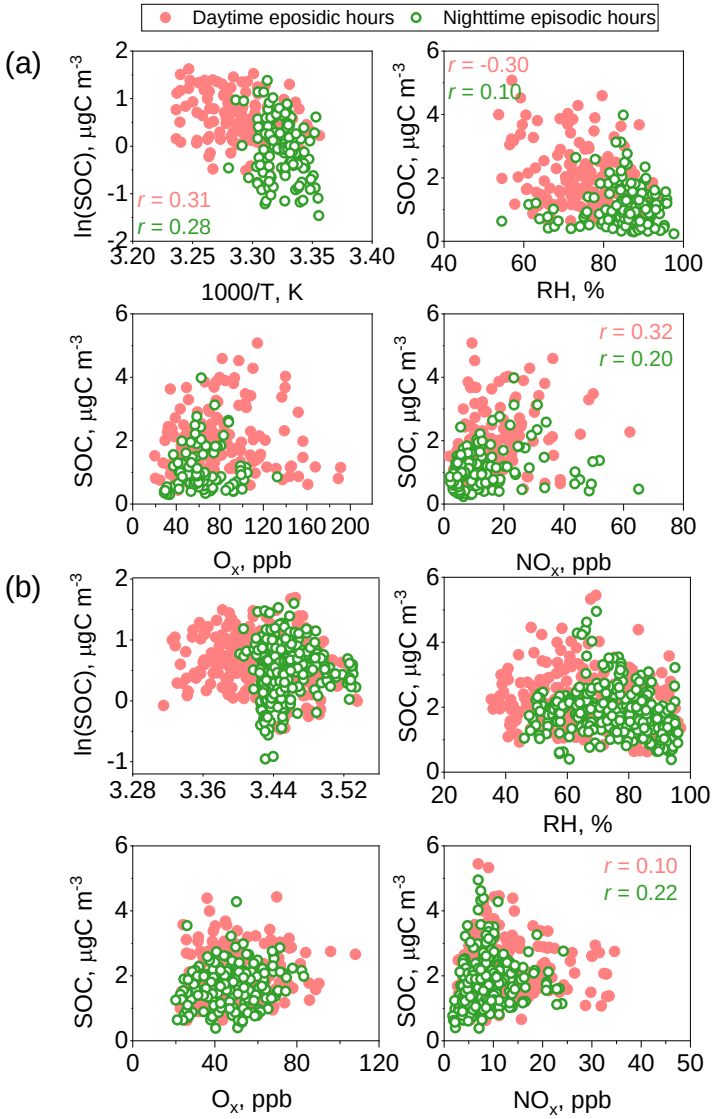

Figure S11. Scatter plot of SOC with meteorological parameters (i.e., temperature and RH), and oxidants (i.e., O_x and NO_x) during the episodic hours in (a) summer typhoon episodes and (b) winter haze episodes. The solid red circle represents daytime hours, blank green circle represents nighttime hours.

Table S3. Statistical summary of aerosol water content and [H^+] during the summer typhoon episodes and winter haze episodes during the daytime and nighttime, respectively.

Episodes	Day/night	AWC (μg m^{-3})	[H^+] (mol m^{-3})
Summer typhoon episodes	Day	12.5 ± 8.34	1.70 ± 1.20 E-10
Summer typhoon episodes	Night	23.1 ± 23.4	1.49 ± 1.00 E-10
Winter haze episodes	Day	6.36 ± 6.60	7.51 ± 1.80 E-11
Winter haze episodes	Night	8.86 ± 8.47	1.54 ± 9.72 E-10

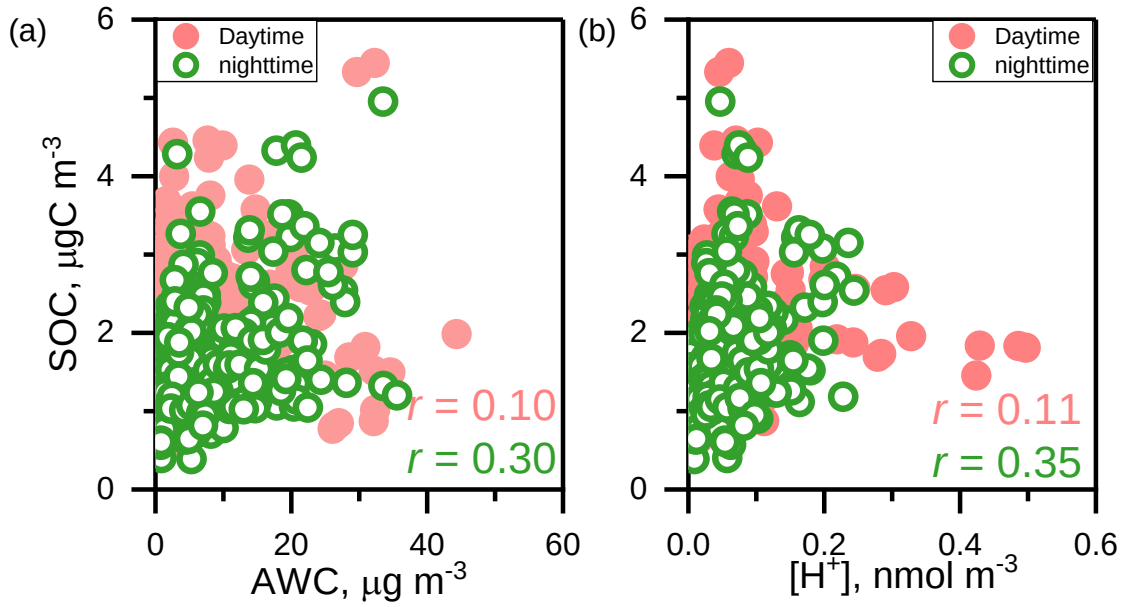

Figure S12. Scatter plot of SOC with (a) aerosol water contents (AWC) and (b) [H^+] during the winter haze episodes. The solid red circle represents daytime hours, blank green circle represents nighttime hours.