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Characterization of nitrous acid and its potential effects on secondary pollution in the warm season in Beijing urban areas

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Ozone photolysis (P_{OH-O3}) was also one production pathway of OH radical, and this was calculated and compared to $P_{OH-HONO}$.

$$P_{OH-HONO} = J(HONO)[HONO] \quad (S1)$$

$$P_{OH-O3} = 2J(O^1D)[O_3]\Phi_{OH} \quad (S2)$$

$$\Phi_{OH} = \frac{k_{O^1D+H_2O}[H_2O]}{k_{O^1D+H_2O}[H_2O]+k_{O^1D+O_2}[O_2]+k_{O^1D+N_2}[N_2]} \quad (S3)$$

$$[H_2O] = RH \times \frac{P_{H_2O}}{P} \times N_{air} \quad (S4)$$

$$P_{H_2O} = 1013.25 \times EXP\left[13.3185 \times \left(1-\frac{375.15}{273.15+T}\right) - 1.97 \times \left(1-\frac{375.15}{273.15+T}\right)^2 - 0.6445 \times \left(1-\frac{375.15}{273.15+T}\right)^3 - 0.1299 \times \left(1-\frac{375.15}{273.15+T}\right)^4\right] \quad (S5)$$

$$[O_2] = 0.20946 \times N_{air} \quad (S6)$$

$$[N_2] = 0.78084 \times N_{air} \quad (S7)$$

$$N_{air} = \frac{P \times N_A}{R \times (273.15+T)} \quad (S8)$$

where [] were the concentration of the substances, Φ was the OH radical yield in the O^1D reaction pathway, $k_{O^1D+H_2O}$ was 2.2×10^{-10} cm^3 $mole^{-1}$ s^{-1}, $k_{O^1D+O_2}$ was 4.0×10^{-11} cm^3 $mole^{-1}$ s^{-1}, $k_{O^1D+N_2}$ was 2.6×10^{-11} cm^3 $mole^{-1}$ s^{-1}(Atkinson et al., 1997), P was the atmospheric pressure, P_{H_2O} was the partial pressure of water vapor (Seinfeld and Pandis, 2006), N_{air} was the number of molecules of air per unit volume, N_A was 6.022×10^{23} $mole^{-1}$ mol^{-1}, and R was 8.314 J mol^{-1} K^{-1}.

Table S1. The average and monthly average value of T, RH, WS.

Meteorological Parameters		T (°C)	RH (%)	WS (m/sec)
average		23	56	1.2
monthly average	June	28	34	1.7
	July	27	71	1.2
	August	26	59	1.2
	September	22	65	1.0
	October	13	50	0.94

Table S2. Overview on HONO field observations performed in Beijing since 2000.

Date	Site Property	HONO (ppbV)		$HONO/NO_2$	Emission Factor	Reference
May 16th-25th, 2000 Jun. 24th- Jul. 4th, 2000 Sep.7th-11th, 2000 Dec. 18th-28th, 2000	Urban site (Peking University) 39.54°N,116.23°E	(mean) 1.8 1.6 1.4 1.5	(min-max) 1.1-3.1 0.9-2.1 1.1-1.5 0.8-2.6	-	---	(Hu et al., 2002)
Jul.-Aug., 2002 Jul.-Aug., 2003	Urban site (Peking University) 39.54°N,116.23°E	(mean) 1.9	(standard deviation) 1.3	-	-	(Wu et al., 2009)
Jan.23nd-Feb.14th., 2007 Aug.2nd-Aug.31st., 2007	Urban site (Peking University) 39.99°N,116.28°E	(mean) 1.0 1.5	(min-max) 0.30-2.7 0.44-2.9	0.03 0.05	0.0065	(Spataro et al., 2013)
Jul.2008-Apr.2009	Urban site (Institute of Atmospheric Physics of the Chinese Academy of Sciences, IAPCAS) 39.98°N,116.38°E	(mean) 0.19 (Spring) 0.18 (Summer) 0.46 (Fall) 0.48 (Winter)	(10th -90th perc.) 0.04-0.45 0.01-0.40 0.05-1.14 0.04-1.04	0.015 (Spring) 0.008 (Summer) 0.020 (Fall) 0.015 (Winter)	-	(Hendrick et al., 2014)
Oct.28th-Nov.3rd, 2014	Urban site (Institute of Chemistry, Chinese Academy of Sciences, ICCAS) 39.99°N,116.32°E	(mean) 1.45	(min-max) 0.54-2.77	0.039	0.0065	(Tong et al., 2015)

	Suburban Site (Lake yanqi campus of University of Chinese Academy of Sciences, UCAS) 40.4°N, 116.6°E	(mean) 0.74	(min-max) 0.18-1.23	0.088		
Feb.22nd-Mar.2nd, 2014	Urban site (ICCAS) 39.99°N,116.32°E	-	(min-max) 0.28-0.34	-	0.0065	(Hou et al., 2016)
Dec.12th-Dec.22nd, 2015	Urban site (ICCAS) 39.99°N,116.32°E	(mean) 0.86	-	0.052		(Tong et al., 2016)
	Suburban Site (Lake yanqi campus of UCAS) 40.4°N, 116.6°E	(mean) 0.52	-	0.08		
Sep.22nd-Oct.21th,2015 Jan.3rd-Jan.27th,2016 Apr.1st-May14th,2016 Jun.20th-Jul.25th,2016	Urban site (Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences) 40.0078°N, 116.33°E	(mean) 2.3 (Fall) 1.1 (Winter) 1.1 (Spring) 1.4 (Summer)	(standard deviation) 1.8 0.89 0.95 0.90	0.070±0.033 0.046±0.024 0.041±0.023 0.079±0.014	-	(Wang et al., 2017)
Dec. 16th-23nd, 2016	Urban site (ICCAS) 39.99°N,116.32°E	(mean) 3.5	(min-max) 0.3-11	-	0.013	(Zhang et al., 2019)
Apr. 14th-28th, 2017	Urban site (ICCAS) 39.99°N,116.32°E	(mean) 1.2	(min-max) 0.02-6.4	-	0.008	(Lin et al., 2022)
Dec. 15th,2017-Jan. 4th, 2018	Urban site (ICCAS) 39.99°N,116.32°E	(mean) 1.2	(standard deviation) 1.2	-	0.0051-0.0081	(Zhang et al., 2022a)
May 7th-30th, 2017 Jan. 15th-30th, 2018	Urban site (Chinese Research Academy of Environmental Sciences) (CRAES) 40°04′N, 116°42′E	(mean) 1.3 (Summer) 1.0 (Winter)	(standard deviation) (max) 0.94 (6.7) 1.3 (9.6)	0.072±0.052 (Summer) 0.041±0.026 (Winter)	0.008	(Gu et al., 2022)
Apr.-May, 2016 Jul.-Aug., 2017 Oct.-Nov., 2017 Dec. 2017- Feb., 2018	Urban site the Institute of Urban Meteorological 39°56′N,116°17′E	(mean) 1.7 (Spring) 1.8 (Summer) 2.2 (Autumn) 0.89 (Winter)	-	-	-	(Su et al., 2021)
Aug. 18th-Sep. 16th, 2018	Suburban Site Qingyuan campus of Beijing Institute of Petrochemical Technology (BIPT)	(mean) 0.38	(min-max) (standard deviation) 0.01-1.9 (0.35)	-	0.0085	(Xuan et al., 2023)

Oct. 25th-Dec. 7th, 2018	Urban site (ICCAS) 39.99°N,116.32°E IAPCAS 39.98°N,116.38°E	(mean) 2.5	(standard deviation) 1.6		0.0097	(Zhang et al., 2023b)
May 25th-Jul.15th, 2018 Nov. 26th, 2018-Jan. 15th, 2019	IAPCAS 39.98°N,116.38°E	(mean) 1.3 (Summer) 1.1 (Winter)	(min-max) (standard deviation) 0.53-2.4 (0.44) 0.02-3.2 (0.68)	6.8±1.4% (Summer) 5.1±2.7% (Winter)	0.0078	(Liu et al., 2021)
Dec. 22nd, 2018-Jan. 23rd, 2019	Urban site (ICCAS) 39.99°N,116.32°E	(mean) 0.98	(min-max) (standard deviation) 0.01-4.8 (0.85)	-	0.0051-0.0081	(Zhang et al., 2022b)
Jun. 13th-Jul.4th, 2019	Urban site (CRAES) 40°04´N, 116°42´E	(mean) 0.44	(min-max) (standard deviation) 0.10-1.4 (0.24)	-	0.003,0.0065,0.008	(Li et al., 2021)
Jan. 22th-Feb. 28th, 2018; Dec. 1st, 2018-Feb. 28th, 2019; Dec. 1st, 2019-Feb. 28th, 2020; Dec. 1st, 2020-Feb. 28th, 2021;	Urban site, (West Campus of Beijing University of Chemical Technology) 39.95°N,116.31°E	(mean) 0.66 (2018) 1.4 (2019) 0.95 (2020) 1.3 (2021)	-	0.038(2018) 0.052 (2019) 0.042 (2020) 0.067 (2021)	0.0079	(Lian et al., 2022)
Oct. 1st-Oct. 31st,2019	Urban site (ICCAS) 39.99°N,116.32°E	(mean) 0.99	-	-	-	(Jia et al., 2023)
Mar. 1st-30th, 2021	Urban site (ICCAS) 39.99°N,116.32°E	(mean) 1.5	(standard deviation) 1.1	0.07	0.008	(Zhang et al., 2023a)
Jun.18th-Oct.25th, 2021	Urban site (CRAES) 40°04´N, 116°42´E	(mean) 1.3 (June) 1.3 (July) 1.0 (August) 0.96 (September) 0.89 (October)	(min-max) 0.14-4.4 0.11-4.8 0.052-4.8 0.078-5.2 0.066-5.1	0.052	0.017	This work

Table S3. The measured JNO_2, estimated JO^1D, JHONO, OH concentration, averaged production/loss rates and proportion of daytime (10:00-15:00 LT) HONO in each month during the observation period.

averaged value	June	July	August	September	October
$J(NO_2)$ (s^{-1})	4.0×10^{-3}	3.4×10^{-3}	3.8×10^{-3}	3.0×10^{-3}	2.8×10^{-3}
$J(O^1D)$ (s^{-1})	1.5×10^{-5}	1.6×10^{-5}	1.3×10^{-5}	1.0×10^{-5}	0.66×10^{-5}
J(HONO) (s^{-1})	8.2×10^{-4}	9.1×10^{-4}	8.1×10^{-4}	8.0×10^{-4}	6.7×10^{-4}
OH (mole/cm^3)	6.4×10^{6}	7.0×10^{6}	5.6×10^{6}	4.5×10^{6}	3.0×10^{6}
L_{phot} (ppb/hr)	2.7	3.1	3.9	2.4	1.1
L_{dep} (ppb/hr)	0.26	0.27	0.22	0.26	0.13
$L_{OH+HONO}$ (ppb/hr)	0.13	0.14	0.15	0.085	0.029
P_{OH+NO} (ppb/hr)	0.64	1.5	1.5	0.81	0.68
P_{emis} (ppb/hr)	0.17	0.19	0.16	0.21	0.33
$P_{unknown}$ (ppb/hr)	2.3	1.8	2.7	1.7	0.29
$P_{unknown}/NO_2$ (hr^{-1})	0.32	0.4	0.61	0.25	0.025

Table S4. Correlations of $P_{unknown}$ against various parameters during the observation period.

Parameters	June (N = 42)		July (N = 82)		August (N = 103)		Summer (N= 227)		September (N = 102)		October (N = 64)		Autumn (N= 166)	
	R	P	R	P	R	P	R	P	R	P	R	P	R	P
NO_2	**0.42**	0.0061	**0.42**	1.0E-4	**-0.3**	0.002	**0.19**	0.0046	-0.05	0.62	0.08	0.53	**-0.21**	0.0068
JNO_2	-0.18	0.50	0.038	0.074	-0.076	0.63	**-0.044**	0.026	0.21	0.24	-0.13	0.34	**0.23**	5.4E-4
$PM_{2.5}$	**0.66**	2.1E-6	**0.49**	2.8E-6	0.06	0.54	**0.30**	5.6E-6	**-0.20**	0.042	0.16	0.22	-0.088	0.26
RH	**0.63**	7E-6	0.04	0.74	**-0.38**	9.4E-5	0.05	0.46	**-0.23**	0.021	0.11	0.39	-0.067	0.39
NO_3^-	**0.39**	0.011	**0.48**	4.5E-6	-0.17	0.082	**0.21**	0.0012	-0.16	0.10	0.16	0.21	-0.083	0.29
OC	**0.81**	1.1E-10	**0.44**	2.9E-5	0.13	0.19	**0.28**	2.4E-5	-0.17	0.095	0.17	0.19	0.017	0.83
JNO_2*RH	**0.71**	0.0042	0.15	0.45	**-0.36**	8.0E-4	0.039	0.31	-0.024	0.69	-0.025	0.78	**0.20**	0.0058
NO_2*$PM_{2.5}$	**0.57**	8.3E-5	**0.50**	2.0E-6	-0.1	0.32	**0.28**	2.4E-5	-0.19	0.056	0.034	0.79	**-0.17**	0.028
NO_2*OC	**0.57**	9.2E-5	**0.45**	2.3E-5	-0.058	0.55	**0.26**	5.8E-5	-0.15	0.15	0.062	0.63	**-0.17**	0.026
NO_2*EC	0.22	0.16	**0.32**	0.0035	-0.12	0.22	**0.15**	0.022	-0.14	0.16	0.11	0.40	**-0.16**	0.043
NO_2*JNO_2*$PM_{2.5}$	**0.62**	1.0E-5	**0.32**	0.0038	-0.12	0.24	**0.24**	3.0E-4	-0.11	0.27	-0.06	0.65	-0.11	0.16
NO_2*JNO_2*RH*$PM_{2.5}$	**0.60**	3.2E-5	**0.33**	0.0022	**-0.18**	0.068	**0.22**	8.9E-4	-0.12	0.22	-0.049	0.70	-0.092	0.24
NO_2*JNO_2*OC	**0.58**	5.9E-5	**0.21**	0.059	-0.12	0.25	**0.19**	0.0037	-0.031	0.76	-0.1	0.44	-0.013	0.87
NO_2*JNO_2*RH*OC	**0.62**	1.4E-5	**0.26**	0.021	**-0.18**	0.063	**0.20**	0.0030	-0.07	0.47	-0.04	0.74	0.008	0.92
NO_2*JNO_2*EC	**0.36**	0.019	**0.22**	0.044	-0.15	0.14	0.095	0.16	-0.11	0.29	-0.1	0.45	-0.12	0.13
NO_2*JNO_2*RH*EC	**0.41**	0.007	**0.22**	0.047	-0.19	0.061	0.088	0.19	-0.13	0.20	-0.07	0.58	-0.10	0.19
JNO_2*NO_3^-	**0.48**	0.001	**0.29**	0.0083	**-0.20**	0.044	**0.18**	0.0082	-0.09	0.37	0.032	0.80	0.005	0.95
JNO_2*RH*NO_3^-	**0.50**	7.2E-4	**0.30**	0.0058	**-0.24**	0.015	**0.16**	0.017	-0.10	0.31	0.006	0.96	0.047	0.55
JNO_2*NO_3*SO_4^{2-}	0.26	0.1	**0.32**	0.0031	-0.14	0.15	0.10	0.12	-0.16	0.12	0.13	0.30	-0.023	0.77
JNO_2*RH*NO_3*SO_4^{2-}	0.23	0.14	**0.33**	0.0028	-0.17	0.08	0.092	0.17	-0.15	0.12	0.16	0.22	0.0069	0.93
JNO_2*NO_3*Cl^-	0.16	0.31	**0.46**	1.3E-5	**-0.26**	0.0087	**0.17**	0.011	-0.12	0.22	0.093	0.46	-0.074	0.35
JNO_2*RH*NO_3*Cl^-	0.14	0.37	**0.44**	4.1E-5	**-0.27**	0.0053	-0.029	0.67	-0.11	0.28	0.075	0.56	-0.058	0.46
JNO_2*NO_3^-*OC	**0.55**	1.5E-4	**0.26**	0.019	-0.05	0.58	**0.19**	0.0039	-0.14	0.17	0.045	0.72	-0.05	0.52
JNO_2*RH*NO_3*OC	**0.52**	4.3E-4	**0.29**	0.009	-0.23	0.019	**0.18**	0.0064	-0.11	0.27	0.027	0.83	-0.020	0.80

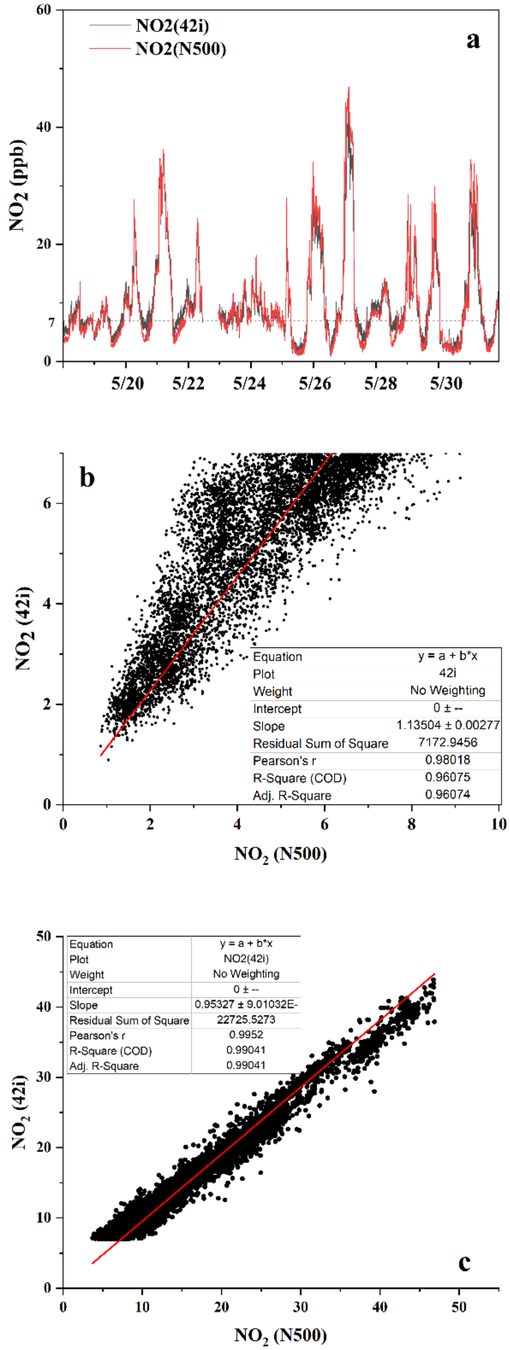

Figure S1. (a) The comparison plot of NO_2 data during the observation period (2024.0519-2024.0531, the Teledyne API Model N500 CAPS NOx analyzer and the Thermo Scientific 42i analyzer); (b) Comparison of the corresponding data between the two devices when the 42i device measures NO_2 concentrations below 7 ppb; (c) Comparison of corresponding data between the two devices when 42i device measures NO2 concentrations above 7 ppb.

Figure S2. Temporal trends of hourly average RH, T, WD (wind direction), WS (wind speed), and JNO_2 during the measurement.

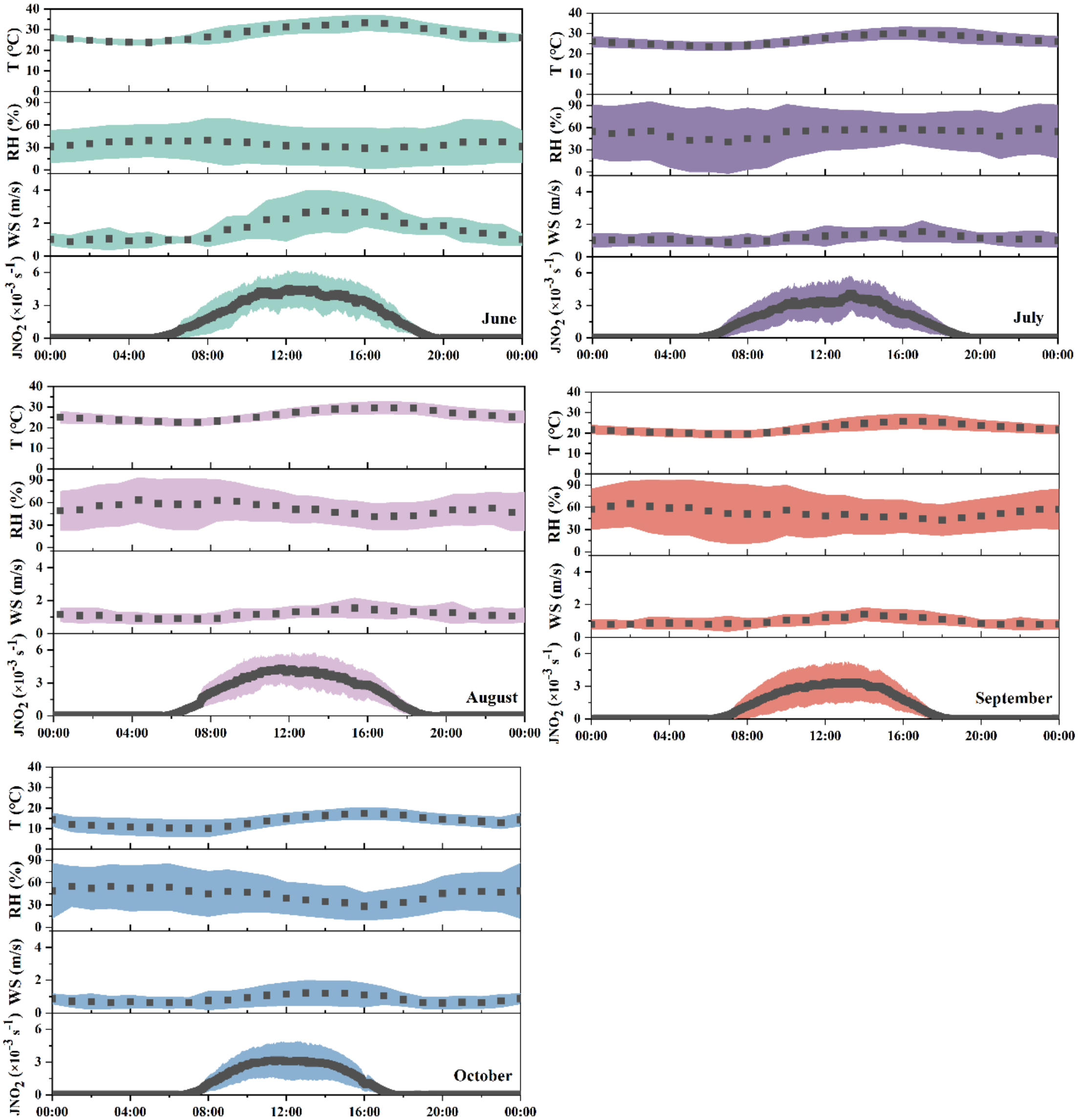

Figure S3. Daily averaged variation of several meteorology data during the observation.

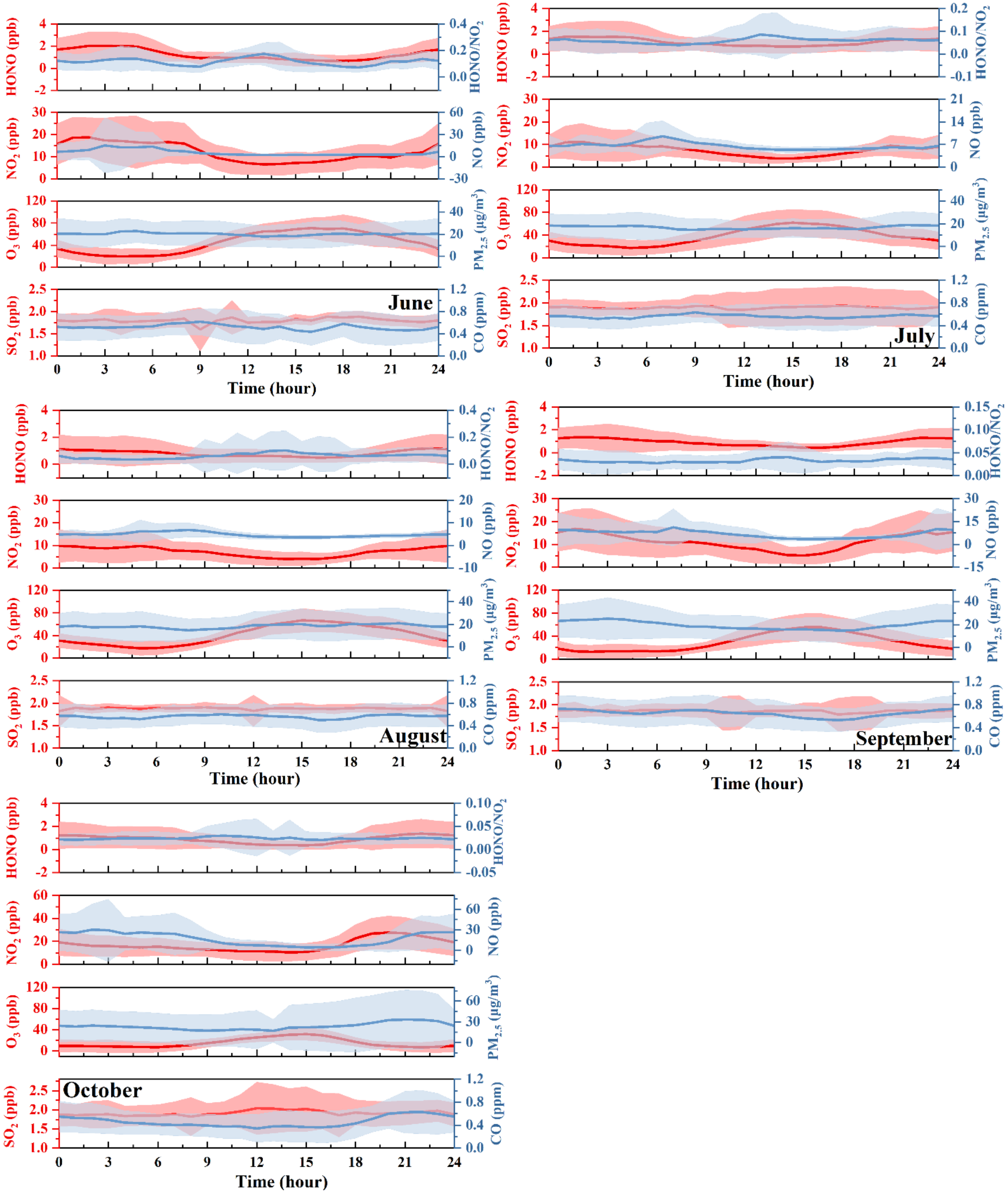

Figure S4. Daily averaged values of several parameters during the observation. The shaded areas represent the standard deviation of the corresponding pollutant concentration.

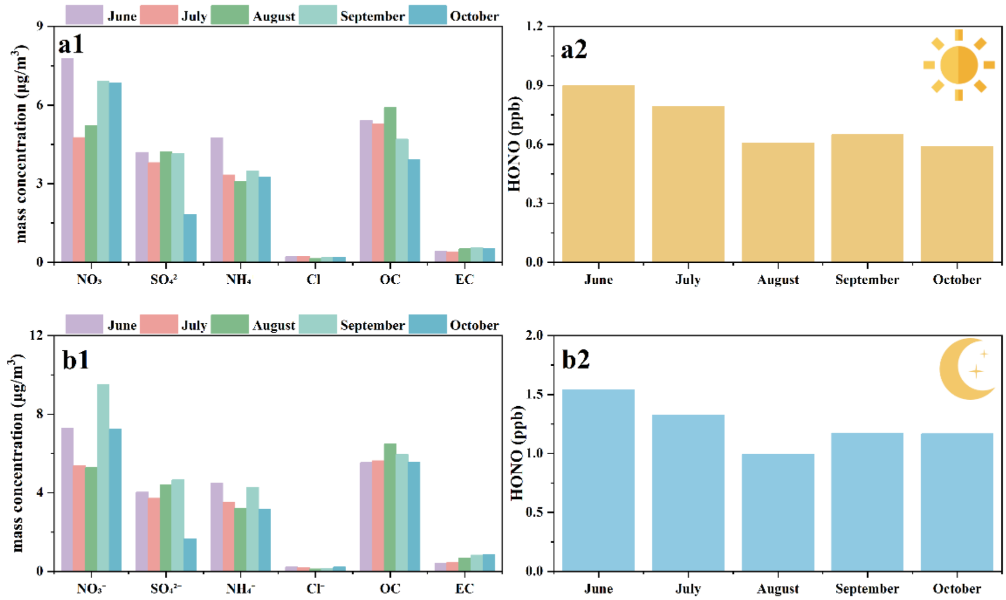

Figure S5. Distributions of NO_3^-, SO_4^{2-}, NH_4^+, Cl^-, OC, EC, and HONO mean concentrations under different months. The upper panel was the daytime average value (7:00-18:00), and the bottom panel was the nighttime average value (19:00-6:00)

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