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Supplement of

Intercomparison of O₃ formation and radical chemistry in the past decade at a suburban site in Hong Kong

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Text S1. Determinations of the locally-produced and regionally-transported O₃ and discussions on the uncertainties.

As an observation based mode, PBM-MCM has been widely used to simulate the in-situ O₃ production (Lam et al., 2013; Ling et al., 2014; Lyu et al., 2017b; Y. Wang et al., 2017). Therefore, the O₃ simulated by PBM-MCM can be regarded as the locally-produced O₃, and the differences between the observed and simulated O₃ were taken as the regionally-transported O₃. However, it should be noted that the observed concentrations of O₃ precursors could be partially built up by regional transport. For example, under the northwest winds, the average mixing ratios of CO (693.9±25.5 ppbv), ethyne (2.15±0.22 ppbv), ethane (2.31±0.25 ppbv), propane (2.97±0.51 ppbv) and toluene (2.42±0.52 ppbv) were the highest among all the wind sectors. Since the PBM-MCM was constrained by the observed concentrations of O₃ precursors, the share of regionally-transported O₃ precursors in the observations made contributions to the simulated O₃, which in fact represented a kind of regional transport. Therefore, the locally produced O₃ were to some extent overestimated in this way. Conversely, the regionally-transported O₃ were underestimated. However, it is difficult to accurately quantify the contributions of regional transport to O₃ precursors at the site, and also due to the non-linear relationships between O₃ and its precursors, we did not quantitatively evaluate the overestimation of the locally-produced O₃ and the underestimation of the regionally-transported O₃.

Text S2. Set-up of the simulation scenarios

The base scenario (scenario A) was established to simulate the local production of O₃, with the observed concentrations of air pollutants (excluding O₃) as model inputs. The observed O₃ was not input because the simulated O₃ would be constrained to the observed values with the outputs exactly the same as the inputs otherwise. The scenario B was established to simulate

O₃ under the assumption that a source of VOCs was totally removed. Namely, the VOCs emitted from a specific source were subtracted from the observed VOCs when allocating the model inputs. In this study, six sources of VOCs were identified (see section 3.4.1). Therefore, 6 sub-scenarios were included in scenario B, because the VOCs emitted from the individual sources were subtracted one by one. In this approach, the differences in simulated O₃ between scenario A and scenario B were the contributions of individual VOC sources to the local O₃ production.

Text S3. Uncertainties in HONO concentrations adopted in model simulations and the subsequent uncertainties in the contributions of HONO to OH formation and loss rates.

To evaluate the uncertainties of adopting the HONO concentrations in 2011 in the model, we calculated the average diurnal cycles of HONO in the 2007, 2013 and 2016 sampling campaigns (Figure S5), according to the diurnal patterns of HONO/NO_x ratios determined at the same site (Xu et al., 2015). It was found that by adopting the values in 2011, the HONO concentrations were underestimated by 14.9% and 11.6% in 2007 and 2013, respectively, but were overestimated by 10.4% 2016. Further, the sensitivity tests indicated that the maximum underestimation (overestimation) of the total OH production rate was 2.3±2.3% (21.6±5.2%) in 2007, 5.8±1.3% (3.4±1.0%) in 2013 and 5.7±1.3% (3.4±0.9%) in 2016. It should be noted that the maximum overestimation of the total OH production rate in 2007 (21.6±5.2%) occurred at 07:00 LT when the OH recycling was weak. During 08:00 – 19:00 LT, both the underestimation and overestimation of the simulated total OH production rates were less than 3%. Therefore, it was concluded that the simulated OH formation and loss rates were not largely biased by adopting the measured HONO at TC in 2011 in all the simulations.

Besides, the HONO concentrations calculated from the HONO/NO_x ratios and NO_x concentrations also had certain uncertainties. Thus, we did not use the calculated HONO

concentrations to constrain the model. In fact, the consistent input of the diurnal cycle of HONO concentrations in the three sampling campaigns enabled us to look into the changes of O₃ and radical photochemistry induced by the other factors, such as VOCs, NO_x and meteorological conditions.

Table S1. Summary of the representative studies regarding O₃ pollution in Hong Kong.

Reference	Site	Measurement period	Nature of monitoring	Target	Main conclusions
Cheng et al, 2010	Wanqingsha (WQS), Guangdong and Tung Chung (TC), Hong Kong	Oct-Dec 2007	Suburban	O ₃	O ₃ formation was limited by VOCs at both sites. Carbonyls played important roles in photochemistry.
Cheng et al, 2013	TC, HK	Sep 2007 and Sep 2008	Suburban	O ₃ , VOCs	Major sources of VOCs in HK included consumer products, paint and printing solvent, road transport, and industrial, commercial, domestic and off-road transport.
Ding et al, 2004	PRD	Sep 2001	Large area	O ₃	O ₃ pollution events in PRD were close associated with sea-land breezes and tropical cyclones.
Guo et al, 2011	WQS and TC	Oct-Dec 2007	Suburban	VOCs	Solvent use, vehicular emissions, biomass burning, LPG usage and gasoline evaporation dominated the sources of VOCs in PRD.
Guo et al, 2013a	Tsuen Wan (TW) and Tai Mao Shan (TMS), HK	Sep-Nov 2010	TW: Urban TMS: Mountainous	O ₃	Less NO titration, vertical transport, valley breeze and regional transport caused higher O ₃ at the mountainous site.
Huang et al, 2005	HK	1999-2003	Large area	O ₃	Tropical cyclones, continental anticyclones and troughs were conducive to O ₃ pollution events in Hong Kong.
Lam et al, 2005	TC, HK	Aug 1999	Suburban	O ₃ , VOCs	Local thermal circulation under calm synoptic conditions trapped air pollutants, resulting in O ₃ enhancements in HK.
Ling et al, 2013	TC, HK	Sep-Nov 2010	Suburban	O ₃	High O ₃ in HK was a combined effect of both local formation and regional transport
Ling et al, 2014	TW and TMS, HK	Sep-Nov 2010	TW: Urban TMS: Mountainous	O ₃	Different O ₃ production and destruction pathways at two sites. More aged air masses at the mountainous site favored O ₃ formation.
Lyu et al, 2016a	Multiple, HK	Sep 2013 and Sep 2014	Urban Suburban Rural	VOCs	VOCs emitted from LPG-fueled vehicles significantly decreased at urban roadside sites. O ₃ formation was limited by VOCs regardless of locations, while VOCs and NO _x co-limited O ₃ formation in rural areas.
Lyu et al, 2016b	Mong Kok (MK), HK	Jun 2011-May 2014	Roadside	O ₃ , VOCs	Replacing catalytic converters in LPG-fueled vehicles led to substantial reductions of VOCs and NO _x emissions.
Lyu et al, 2017a	TC, HK	2005-2013	Suburban	VOCs	VOCs emitted from solvent usage and diesel exhaust decreased in Hong Kong from 2005 to 2013.

Ou et al, 2015	TC, HK	2005-2013	Suburban	VOCs	Vehicular exhaust, gasoline evaporation and LPG usage, consumer product and printing, architectural paints, and biogenic emissions were identified as the sources of VOCs in the study area.
So and Wang, 2003	Multiple, HK	Jun 1999-May 2000	Urban Suburban Rural	O ₃	Air quality in rural areas was frequently influenced by regional air masses, while was predominantly affected by local emissions in urban areas.
Y. Wang et al, 2017	TC, HK	2005-2014	Suburban	O ₃	Locally produced autumn O ₃ decreased in Hong Kong, which was reversed by regionally-transported O ₃ between 2005 and 2013.
Y. Wang et al, 2018	Wan Shan Island (WSI), GD	Aug-Nov 2013	Rural	O ₃	O ₃ formation switched from the NO _x -limited regime on low O ₃ days to VOC-limited regime on high O ₃ days over South China Sea.
Xu et al, 2008	Linan, Zhejiang	Aug 1991-Jul 2006	Rural	O ₃	Monthly highest 5% of O ₃ increased from 1991 to 2006, with enhanced variability, likely due to the increased NO _x emissions.
Xue et al, 2014a	TC, HK	Sep-Nov of 2002-2013	Suburban	O ₃	Increase of regionally-transported O ₃ offset the decrease of locally-produced O ₃ and resulted in the increase of observed O ₃ in the autumn in HK during 2002-2013.
Zhang et al, 2007	Multiple, HK	Oct-Dec 2002	Urban Suburban Rural	O ₃	50-100% of O ₃ enhancement during O ₃ episodes in HK was explained by local photochemical formation, following the oxidation of anthropogenic VOCs.

Table S1. Summary of the sampling periods, VOC sampling dates and number of samples in the 2007, 2013 and 2016 sampling campaigns.

Year	Sampling period	Month	Date	Number of NMHC samples	Number of OVOC samples
2007	23 Oct.–1 Dec.	Oct.	26-27	96	28
		Nov.	13, 15-17, 23		
		Dec.	1		
2013	11 Aug.–22 Nov.	Sept.	11-12, 26	146	124
		Oct.	2-4, 9-10, 17, 22-25, 30-31		
		Nov.	15, 19-21		
2016	25 Sept. – 29 Nov.	Sept.	26	172	123
		Oct.	14-17, 23, 31		
		Nov.	1-7,14-18		
Total				414	275

Table S3. Instruments, analysis techniques, detection limits and time resolutions for the real-

time measurements of trace gases at the TC air quality monitoring station.

Species	Instrument	Analysis technique	Detection limit	Time resolution
SO ₂	API 100E	UV fluorescence	0.4 ppb	1 sec
CO	API 300	Non-dispersive infra-red absorption with gas filter correlation	<0.050 ppm	1 sec
NO-NO ₂ -NO _x	API 200A	Chemiluminescence	0.4 ppb	1 sec
O ₃	API 400	UV absorption	0.6 ppb	1 sec

Table S4. Summary of limits of detection (LoDs), precisions and accuracies of the GC-MSD/FID/ECD systems used for NMHCs analyses of whole air samples collected in 2007, 2013 and 2016 (Simpson et al., 2010; Y. Wang et al., 2018).

Institution	Year	Limit of Detection	Precision	Accuracy
UCI	2007	3-10 pptv	3%	5%
GIG	2013	3-57 pptv	2-5%	5%
HKPolyU	2016	3-10 pptv	5%	5%

Table S5. Statistics of VOC mixing ratios measured in 2007, 2013 and 2016. The 95% C.I., Max. and S.D. denote the 95% confidence interval, maximum and standard deviation of the mixing ratios of VOC species, respectively (Unit: pptv).

VOC species	2007			2013			2016		
	Mean±95%CI	Max.	S.D.	Mean±95%CI	Max.	S.D.	Mean±95%CI	Max.	S.D.
Ethane	1587.6±37.3	1994.6	186.7	1918.9±140.1	5700.4	1123.4	1729.1±85.3	3951.2	655.6
Ethene	1388.7±53.5	2284.7	267.3	1074.9±94.3	4529.8	751.9	886.8±79.1	4335.5	605.3
Ethyne	1939.3±105.3	3855.4	526.3	1563.4±94.8	3339.0	760.0	1399.2±83.1	4254.7	637.5
Propane	2826.2±94.1	3889.6	472.0	1619.1±110.4	5826.3	883.4	2308.4±224.2	13269.8	1715.9
Propene	325.2±34.0	1036.7	169.9	296.1±20.3	1052.3	162.7	192.3±13.8	773.6	105.5
<i>i</i> -Butane	790.9±40.7	1269.3	203.6	1556.4±115.5	5778.5	922.7	901.5±67.5	3229.9	516.6
<i>n</i> -Butane	1562.4±124.3	3360.3	621.5	1402.6±95.8	5353.7	767.8	1403.8±116.3	5001.2	893.8
1- <i>i</i> -Butene	275.1±71.2	2945.2	356.1	240.7±70.6	5944.6	566.2	98.0±9.4	715.8	71.7
trans-2-Butene	22.5±6.0	213.4	29.7	31.2±6.7	387.1	42.5	12.1±2.9	184.1	18.3
cis-2-Butene	24.4±6.0	201.9	29.7	25.2±5.0	275.2	32.4	9.3±2.8	178.5	17.5
3-methyl-1-Butene	12.0±2.4	76.6	11.9	35.6±16.0	332.0	67.2	8.7±5.5	207.0	26.6

2-methyl-1-Butene	16.4±3.3	89.0	16.3	47.4±25.3	1074.0	146.6	17.4±11.4	765.6	73.4
cis-2-Pentene	28.6±5.1	130.4	25.4	34.5±7.9	273.9	46.8	5.5±3.4	130.1	16.4
2-methyl-2-Butene	4.6±1.0	25.8	4.8	37.4±14.6	558.0	84.3	10.2±3.6	139.3	17.5
Butyne	41.0±6.3	171.2	31.5	40.5±12.1	406.7	58.6	18.5±2.8	142.2	18.5
<i>i</i> -Pentane	1809.6±339.6	7793.6	1697.8	827.5±63.3	4646.9	504.0	466.3±33.2	1681.2	254.1
<i>n</i> -Pentane	351.9±63.7	2271.2	318.7	50.1±8.6	416.2	59.9	265.1±25.1	1104.0	192.5
1-Pentene	20.4±5.7	244.3	28.2	50.1±8.6	416.2	59.9	15.6±2.9	182.5	20.3
trans-2-Pentene	15.3±4.1	182.0	20.8	23.6±7.7	302.1	41.4	9.8±4.9	189.6	24.1
2-Methylpentane	268.8±68.7	2253.0	343.6	257.1±29.6	1643.9	237.9	195.5±27.1	1171.6	204.9
<i>n</i> -Hexane	1785.2±547.3	17974.4	2736.1	930.6±147.7	9829.0	1182.2	163.4±23.8	1426.7	182.1
<i>n</i> -Heptane	113.8±39.3	1196.3	196.7	100.2±12.8	877.3	102.4	65.9±5.8	290.7	44.7
<i>n</i> -Octane	42.4±9.6	245.2	48.1	47.5±8.9	739.3	70.4	34.0±3.9	188.0	30.4
2,2-Dimethylbutane	18.7±2.2	64.8	11.3	52.7±8.7	719.6	65.0	20.2±2.0	91.9	14.2
2,3-Dimethylbutane	31.2±17.1	822.5	84.5	68.8±7.1	389.3	55.9	46.3±7.3	311.9	50.0

3-Methylpentane	1911.5±423.0	10292.0	2114.7	172.0±20.5	1260.3	164.4	131.8±18.9	1025.8	144.7
Cyclohexane	66.1±29.1	905.4	145.7	84.1±8.7	601.5	70.3	68.1±8.0	414.4	54.3
2-Methylhexane	49.0±16.4	557.0	82.1	85.7±9.7	429.6	77.5	205.8±19.2	492.8	89.9
3-Methylhexane	172.8±53.5	1651.2	266.3	171.5±19.4	1171.5	155.6	43.7±5.4	271.3	37.0
<i>n</i> -Nonane	29.1±6.4	226.8	32.2	47.6±8.1	621.6	64.4	39.9±7.3	477.2	56.4
<i>n</i> -Decane	457.3±138.9	3680.5	694.4	72.2±15.7	1198.3	126.3	34.0±3.9	188.0	30.4
<i>n</i> -Undecane	71.9±13.6	319.7	65.9	90.8±18.1	1367.5	145.4	--	--	--
Isoprene	270.6±53.0	868.5	264.9	417.0±44.4	1561.0	343.4	409.1±52.5	3122.0	402.0
α -Pinene	261.8±191.5	6810.0	957.3	29.9±4.4	321.5	34.2	21.1±2.9	218.0	22.2
β -Pinene	30.3±10.1	306.4	50.6	53.8±7.3	314.5	57.5	9.9±0.7	32.0	5.4
Benzene	567.0±99.7	3761.2	498.5	493.1±33.2	2308.0	266.3	257.3±14.5	658.2	110.3
Toluene	2878.9±845.3	26556.8	4225.8	1445.3±149.2	6494.2	1196.5	1169.5±229.5	13376.0	1601.8
Ethylbenzene	358.8±63.2	1597.7	316.3	389.1±52.3	4274.0	419.5	193.6±20.7	1350.2	159.1
<i>m/p</i> -Xylene	411.1±82.1	1789.7	410.7	491.8±109.7	10168.0	879.8	299.4±28.4	1600.8	217.9

<i>o</i> -Xylene	168.1±30.0	773.5	150.3	231.3±34.1	2082.0	274.1	151.7±13.7	891.2	105.0
1,3,5-Trimethylbenzene	43.8±8.8	323.5	44.3	38.6±8.4	680.3	67.5	11.6±1.6	125.5	12.7
1,2,4-Trimethylbenzene	177.3±40.1	1527.5	200.6	143.3±25.1	1492.3	201.7	39.6±5.6	336.7	42.9
1,2,3-Trimethylbenzene	64.7±23.8	940.9	117.8	63.1±11.6	809.7	93.4	16.7±3.5	320.3	27.2
<i>i</i> -Propylbenzene	7.4±1.2	38.0	6.3	16.5±1.8	111.1	14.2	7.5±0.7	63.0	5.8
<i>n</i> -Propylbenzene	16.4±2.6	68.9	13.0	34.3±4.3	305.2	34.5	12.4±2.0	188.6	15.3
3-Ethyltoluene	34.6±7.2	212.4	36.1	74.0±10.6	653.0	85.5	24.7±2.9	171.4	22.4
4-Ethyltoluene	18.1±3.4	114.4	17.1	45.4±6.1	429.8	48.7	16.6±2.4	220.2	18.3
2-Ethyltoluene	23.2±4.3	143.0	21.5	40.4±6.7	476.3	53.9	14.0±2.1	186.1	16.3
Formaldehyde	8608.3±1298.0	30244.3	6352.3	2729.0±146.5	6403.5	1123.8	2673.3±283.1	13633.0	2171.5
Acetaldehyde	9853.8±1732.6	46380.0	8198.0	1395.9±81.1	3694.8	622.2	779.4±92.1	3126.0	649.4
Acetone	12786.4±2712.9	75590.0	13131.5	5156.2±421.3	22921.5	3231.7	4995.6±460.6	13370.9	3469.7
Propionaldehyde	--	--	--	325.0±38.2	1593.2	247.0	1547.9±547.2	3071.6	1081.3

Table S6. Comparison of meteorological conditions in the autumns of 2007, 2013 and 2016 (HKO, 2017).

	2007		2013		2016	
	Mean \pm 95% C.I.	Max.	Mean \pm 95% C.I.	Max.	Mean \pm 95% C.I.	Max.
Temperature ($^{\circ}$ C)	23.5 \pm 0.4	30.0	25.2 \pm 0.3	33.9	25.0 \pm 0.3	32.7
Relative humidity (%)	64.4 \pm 1.4	80.0	58.6 \pm 1.3	83.1	77.1 \pm 0.7	94.5
Solar radiation (W m^{-2})	190.3 \pm 37.7	788.9	159.2 \pm 21.3	869.1	119.8 \pm 18.2	829.3
Pressure (hPa)	1016.9 \pm 0.4	1024.3	1015.0 \pm 0.3	1023.1	1012.3 \pm 0.3	1020.2
Wind speed at the sampling site (m s^{-1})	2.3 \pm 0.2	5.3	1.0 \pm 0.1	3.2	0.9 \pm 0.1	4.3
Wind speed at HKIA* (m s^{-1})	4.7 \pm 0.3	10.8	4.8 \pm 0.4	11.7	4.5 \pm 0.3	12.4

*HKIA: Hong Kong International Airport.

Table S7. Number of O₃ episode days identified under the tropical cyclone, continental anticyclone and low pressure trough in the autumns of 2007, 2013 and 2016.

Year	Total No. of Episode	Tropical cyclone	Continental anti-cyclone	Low Pressure trough
2007	15*	8 (4 typhoons)	8	1
2013	16	11 (5 typhoons)	5	0
2016	5	4 (3 typhoons)	0	1

*Two O₃ episode days were under the combined influence of tropical cyclone and continental anticyclone.

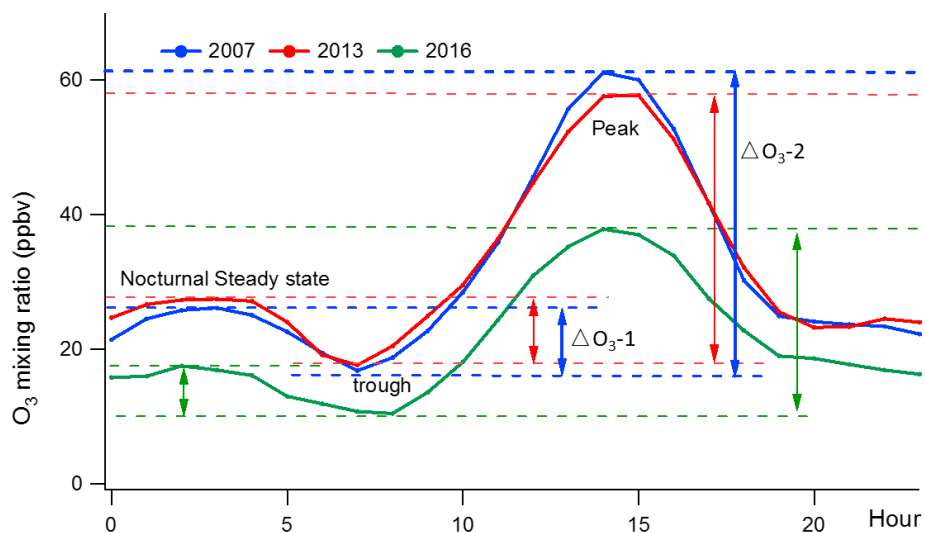


Figure S1. Average diurnal cycles of O₃ in the 2007, 2013 and 2016 sampling campaigns. ΔO_3-1 : O₃ decrease in the early morning driven by NO titration; ΔO_3-2 : photochemically formed O₃ in the daytime (diurnal cycle of O₃ in 2007 as an example).

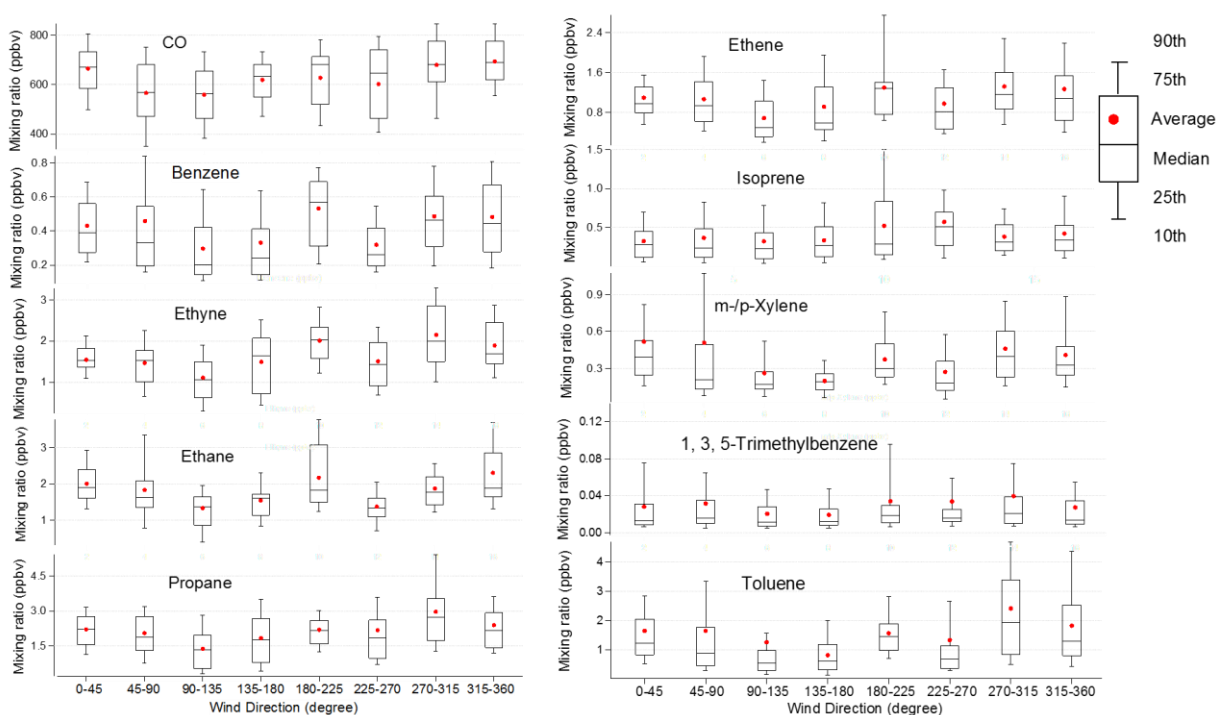


Figure S2. Average mixing ratios of some O₃ precursors in different wind sectors at TC in the three sampling campaigns.

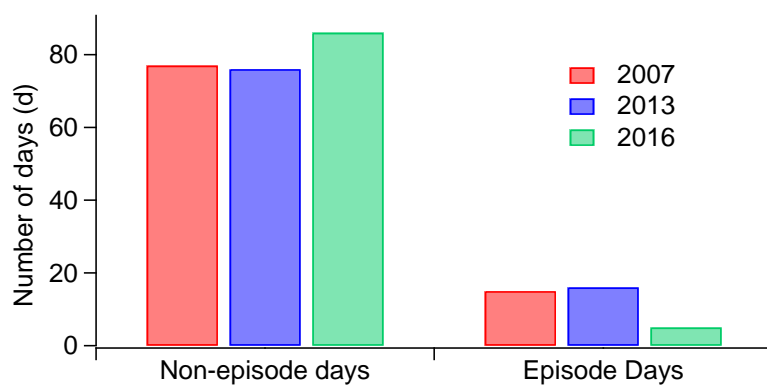


Figure S3. Number of O₃ episode days and non-O₃ episode days in the autumns of 2007, 2013 and 2016.

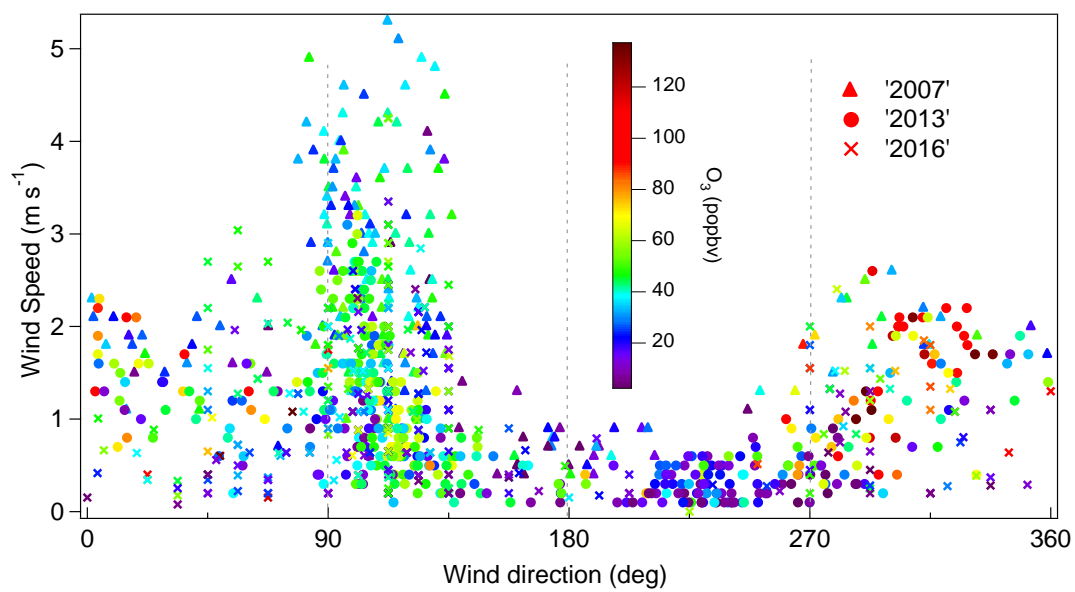


Figure S4. Relationship between the hourly observed O₃ and the wind fields at TC in the three sampling campaigns.

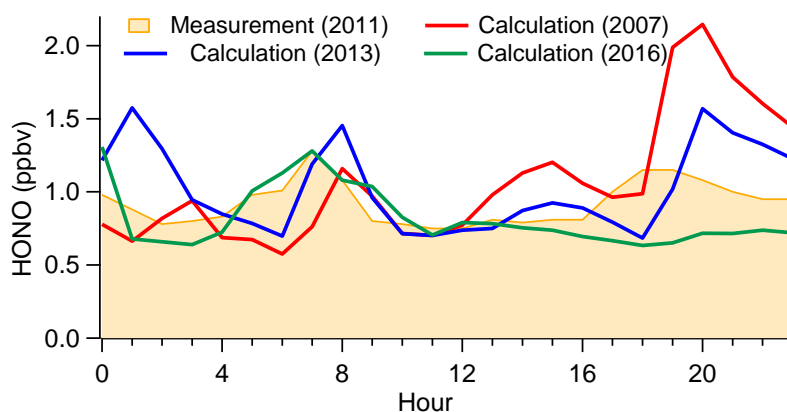


Figure S5. Diurnal cycles of HONO mixing ratios measured at TC in the autumn of 2011 and those calculated from the measured HONO/NO_x ratios and NO_x mixing ratios at the same site in the three sampling campaigns.

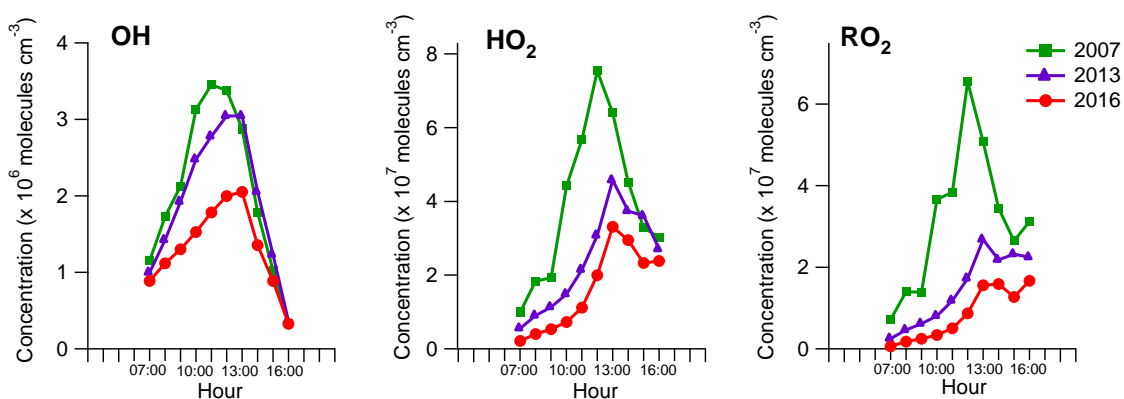


Figure S6. Average diurnal profiles of the simulated OH, HO₂ and RO₂ concentrations on VOC sampling days in 2007, 2013 and 2016. The rebounding of HO₂ and RO₂ concentrations in the late afternoon in 2007 and 2016 were caused by the substantial increases in the concentrations of some VOCs or OVOCs in several samples.

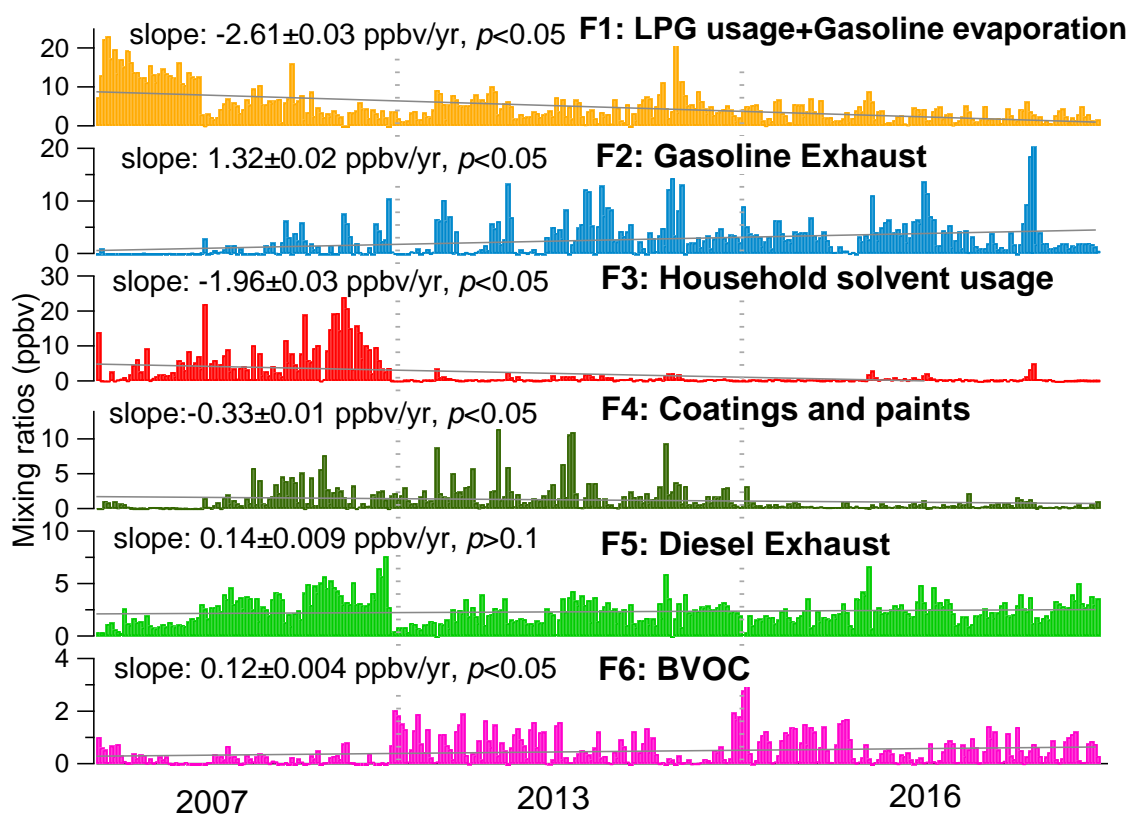


Figure S7. Total mixing ratio of VOCs emitted from each individual source extracted from PMF in the 2007, 2013 and 2016 sampling campaigns. The solid lines represent the linear regressions of the VOC mixing ratios against the sequence number of the samples, with the slope being converted to yearly rates.

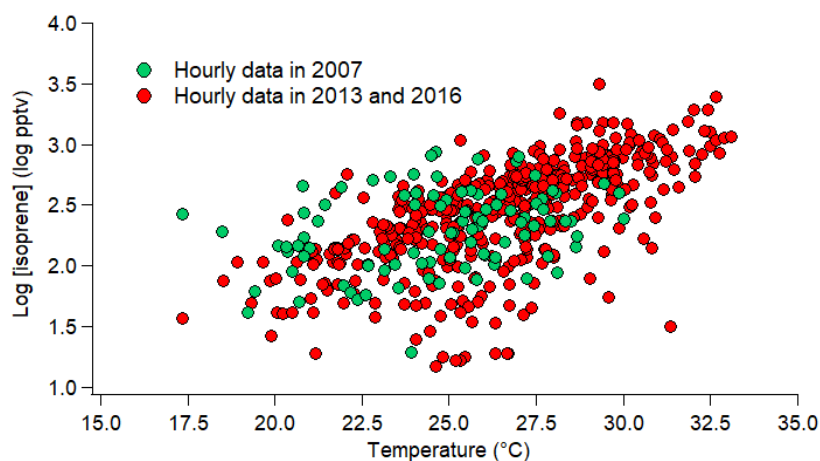


Figure S8. Relationship between the common logarithm of isoprene mixing ratios and temperature.

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