

Supporting information

Quality control and quality assurance (QC/QA)

In order to alleviate contamination from previous samples, canisters had to be repeatedly cleaned using humidified zero air before sampling. In order to inspect the cleanliness and vacuum in canisters, the pre-cleaned canisters, after storing for 24 h, were analyzed using GC-MS according to the same analytical procedures used to analyze the field samples. The canisters without any contamination were used for sampling. No obvious disturbances due to improperly activities (such as smoking, spray fumes, etc.) of sample collectors were present during the sampling events. Daily calibration of the GC-MSD/FID was performed using 2 ppbv standard mixtures to ensure the consistency and sensitivity of the system. The deviation in standard values was within $\pm 20\%$. Abnormal QC/QA data with extremely high or low responses was recalibrated until the deviation was within the acceptable range.

PMF model

In brief, PMF is a diagnostic method involving multivariate analysis, and involves decomposing the integrated sample data (VOCs in this study) into two matrices, namely the source profiles and the source contributions (Jaars et al., 2018). The method takes advantage of other observation data, such as wind direction and speed. Detailed information on the application of PMF can be referred to the publications cited above and to the PMF 5.0 user manual (U.S. EPA, 2014).

Based on the chemical mass balance between the input concentrations of VOCs and the chemical profiles, PMF regarded the ambient data x_{ij} , namely the concentration of j^{th} constituent in i^{th} sample, as the gross values contributed by p sources according to Eq. (1).

$$X_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (1)$$

where g_{ik} stands for the contribution of k^{th} factor in the i^{th} sample, while f_{kj} is the load of j^{th} compound in the k^{th} source and the e_{ij} is the relevant residual.

In order to avoid negative source contributions, a penalty function was adopted

for constraints. Each data point can be individually weighed in the model, while the samples with lots of missing values were excluded.

Based on the algorithm for uncertainties (U), expressed as Q values, the stability of running results was assessed according to Eq. (2).

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left[\frac{X_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right]^2 \quad (2)$$

where u_{ij} stands for the uncertainty of the j^{th} compound in i^{th} sample.

In the PMF model, uncertainty is a function to evaluate the deviations in sampling and analysis procedure (Paatero, 2007). The uncertainty (U) can be calculated based on Eq. (3) (Polissar et al., 1998).

$$U = \begin{cases} \sqrt{(EF \times \text{concentration})^2 + (MDL)^2} & (\text{conc.} > \text{MDL}) \\ \frac{5}{6} MDL & (\text{conc} \leq \text{MDL}) \end{cases} \quad (3)$$

where EF signifies the error fraction that equals 100 times of the percentage uncertainty.

Table S1. Detailed information on the calibration curve for 57VOCs and their MIR

NO.	Species	R ²	MDL (pptv)	RSD	MIR		NO.	Species	R ²	MDL (pptv)	RSD	MIR
1	Ethane	0.9998	6.8	5%	0.28	Alkene	29	Ethylene	0.9997	12.5	10%	9
2	Propane	0.9998	2.8	2%	0.49		30	Propylene	0.9998	6.2	5%	11.66
3	Isobutane	0.9998	3.2	3%	1.23		31	Trans-2-butene	1	3.6	6%	15.16
4	n-Butane	0.9998	6.7	5%	1.15		32	1-Butene	0.9995	7.8	6%	9.73
5	Cyclopentane	0.9971	8.5	7%	0.09		33	Cis-2-butene	0.9997	6.8	6%	14.24
6	Isopentane	0.9999	5.5	4%	0.93		34	1,3-butadiene	0.9874	13.5	8%	
7	n-Pentane	0.9999	6.6	5%	0.88		35	1-Pentene	0.9764	6.3	5%	7.21
8	2,2-Dimethyl-butane	0.9963	5.4	4%	1.17		36	Trans-2-pentene	0.9964	10.1	7%	10.56
9	2,3-Dimethylbutane	0.9966	7.6	6%	0.97		37	Isoprene	0.9966	7.7	6%	10.61
10	2-Methylpentane	0.9958	8	6%	1.5		38	Cis-2-pentene	0.9965	8.6	7%	10.38
11	3-Methylpentane	0.9967	5.4	4%	1.8	Alkyne	39	1-Hexene	0.9961	11.4	9%	4.4
12	n-Hexane	0.9967	7.3	6%	1.24		40	Acetylene	0.9996	7.1	5%	0.95
13	2,4-Dimethylpentane	0.9972	9.6	7%	1.55		41	Benzene	0.9975	6.5	5%	0.72
14	Methyl-cyclopentane	0.9974	5.8	5%	2.19		42	Toluene	0.9963	4.3	4%	4
15	2-Methyl-hexane	0.9968	8.6	7%	1.19		43	Ethyl-benzene	0.9955	4.8	4%	3.04
16	Cyclohexane	0.9958	7.7	6%	1.25		44	m,p-Xylene ^a	0.9969	12.5	5%	7.8
17	2,3-Dimethyl-pentane	0.9969	6.2	5%	1.34		45	o-Xylene	0.9954	5.2	4%	7.64
18	3-Methyl-hexane	0.9946	8.8	7%	1.61		46	Styrene	0.9961	10.6	8%	1.73
19	2,2,4-Trimethyl-pentane	0.9975	7.1	6%	1.26		47	Isopropylbenzene	0.9947	4.3	4%	2.52
20	n-Heptane	0.9974	9	7%	1.07	Aromatic	48	n-Propylbenzene	0.9929	1.6	1%	2.03
21	Methyl-cyclohexane	0.9972	5.8	5%	1.7		49	m-Ethyltoluene	0.991	7.3	6%	7.39
22	2,3,4-Trimethyl-pentane	0.9976	5.7	5%	1.03		50	p-Ethyltoluene	0.9994	8.4	7%	4.44
23	2-Methyl-heptane	0.9971	7	6%	1.07		51	1,3,5-Trimethyl-benzene	0.9994	6.1	5%	11.76
24	3-Methyl-heptane	0.9974	6.7	5%	1.24		52	o-Ethyltoluene	0.9995	4.3	4%	5.59
25	n-Octane	0.9973	7.6	6%	0.9		53	1,2,4-Trimethylbenzene	0.9983	9.7	8%	8.87
26	n-Nonane	0.9963	3.4	3%	0.78		54	1,2,3-Trimethylbenzene	0.9927	9.7	8%	11.97
27	n-Decane	0.9935	7.8	6%	0.68		55	m-Diethylbenzene	0.9967	5.2	4%	7.1
28	n-Undecane	0.9919	7.5	6%	0.61		56	p-Diethylbenzene	0.995	4.2	3%	4.43

^a *m*-Xylene and *p*-Xylene are co-eluted in the chromatographic separation.

TableS2. The correlation coefficient (R^2) between observed and predicted values for each compound at the four sites

	JK	MEM	YH	GS
propane	0.83	0.91	0.84	0.60
iso-butane	0.77	0.92	0.93	0.79
isopentane	0.71	0.42	0.53	0.84
n-pentane	0.91	0.74	0.65	0.89
3-methylpentane	0.92	0.69	0.93	0.88
methylcyclopentane	0.96	0.83	0.89	0.91
2-methylhaxane	0.98	0.98	0.99	0.99
haxane	0.99	0.93	0.95	0.99
2,3-dimethylpentane	0.97	1.00	0.84	0.99
3-methylhaxane	0.97	0.98	0.99	0.99
2,2,4-trimethylpentane	0.89	0.94	0.72	0.80
heptane	0.98	0.88	0.82	0.79
methylcyclohexane	0.80	0.99	0.52	0.85
n-nonane	0.96	0.97	0.66	0.47
n-Nonane	0.83	0.92	0.20	0.75
n-Decane	0.95	0.95	0.41	0.91
n-Undecane	0.93	0.95	0.78	0.86
Trans-2-butene	0.98	0.97	0.97	0.60
1-Butene	0.94	0.97	0.94	0.68
Isoprene	0.99	0.20	0.96	0.91
Acetylene	0.60	0.88	0.85	0.80
Benzene	0.97	0.86	0.41	0.94
Toluene	0.93	0.86	0.97	0.82
Ethyl-benzene	0.91	0.94	0.99	0.98
m,p-Xylene ^a	0.92	0.99	0.98	0.86
o-Xylene	0.97	0.99	0.97	0.87
Styrene	0.75	0.81	0.88	0.84
1,2,4-Trimethylbenzene	0.84	0.88	0.97	0.92
NO ₂	0.88	0.69	0.79	0.67

Table S3 Error estimation summary results, i.e. BS mapping for the four sites
(a) JK

	LPG/CNG+ga- soline evaporation	solvent use	diesel vapor	coal combustion	petrochemical source	vehicle exhaust	biogenic	traffic related	Unmapped
Boot Factor 1	3	0	3	0	1	1	0	90	2
Boot Factor 2	3	0	20	0	0	58	9	7	3
Boot Factor 3	0	100	0	0	0	0	0	0	0
Boot Factor 4	2	0	5	0	2	0	80	6	5
Boot Factor 5	92	0	4	0	1	1	2	0	0
Boot Factor 6	0	0	100	0	0	0	0	0	0
Boot Factor 7	0	0	1	1	98	0	0	0	0
Boot Factor 8	1	0	0	98	0	0	1	0	0

(b) MEM

	LPG/CNG+gasoline evaporation	solvent use	diesel vapor	coal combustion	petrochemical source	vehicle exhaust	biogenic	Unmapped
Boot Factor 1	94	2	0	1	0	2	0	1
Boot Factor 2	0	5	0	91	0	3	0	1
Boot Factor 3	0	100	0	0	0	0	0	0
Boot Factor 4	0	0	100	0	0	0	0	0
Boot Factor 5	0	1	0	0	0	99	0	0
Boot Factor 6	0	0	0	0	100	0	0	0
Boot Factor 7	2	0	3	1	0	5	87	2

(c) YH

	LPG/CNG+gasoline evaporation	solvent use	diesel vapor	coal combustion	petrochemical source	vehicle exhaust	biogenic	Unmapped
Boot Factor 1	0	0	0	0	0	0	100	0
Boot Factor 2	0	0	0	0	0	100	0	0
Boot Factor 3	1	0	1	93	2	2	0	1
Boot Factor 4	1	0	0	0	99	0	0	0
Boot Factor 5	11	70	1	1	9	5	0	3
Boot Factor 6	1	0	99	0	0	0	0	0
Boot Factor 7	100	0	0	0	0	0	0	0

(d) GS

	LPG/CNG+gasoline evaporation	solvent use	diesel vapor	coal combustion	petrochemical source	vehicle exhaust	biogenic
Boot Factor 1	0	0	0	1	0	99	0
Boot Factor 2	0	100	0	0	0	0	0
Boot Factor 3	96	0	3	1	0	0	0
Boot Factor 4	0	0	100	0	0	0	0
Boot Factor 5	3	0	3	2	0	1	90
Boot Factor 6	3	0	11	84	0	1	0
Boot Factor 7	0	0	1	0	96	1	0

TableS4. The ratio of Q/Q(exp) at factor size ranged from 3-9 at the four sites

Number of factors	JK	MEM	YH	GS
3	2.78	1.76	2.29	2.68
4	2.46	1.36	1.95	2.14
5	2.15	1.07	1.62	1.87
6	1.81	0.91	1.40	1.58
7	1.65	0.77	1.30	1.41
8	1.54	0.68	1.24	1.29
9	1.46	0.62	1.18	1.18

Table S5. The average mixing ratios of SO₂, NO_x, CO and O₃ during May to September

	SO ₂ (ppb)	NO _x (ppb)	CO(ppm)	O ₃ (ppb)
May	9.55	35.76	0.70	86.73
June	5.49	31.76	0.71	99.32
July	2.52	20.62	0.69	76.27
August	2.98	25.48	0.89	76.92
September	5.72	52.60	0.96	61.31

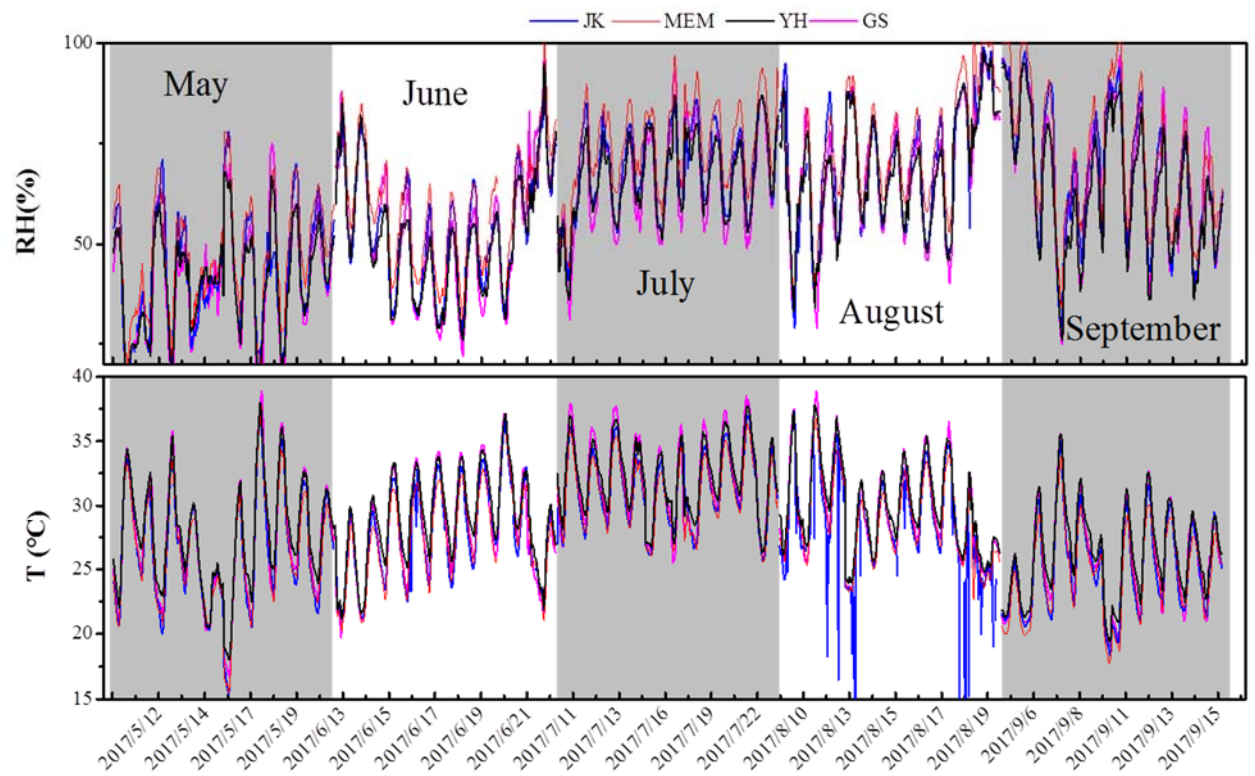


Fig. S1 the temperature and Relative humidity at each site during sampling period

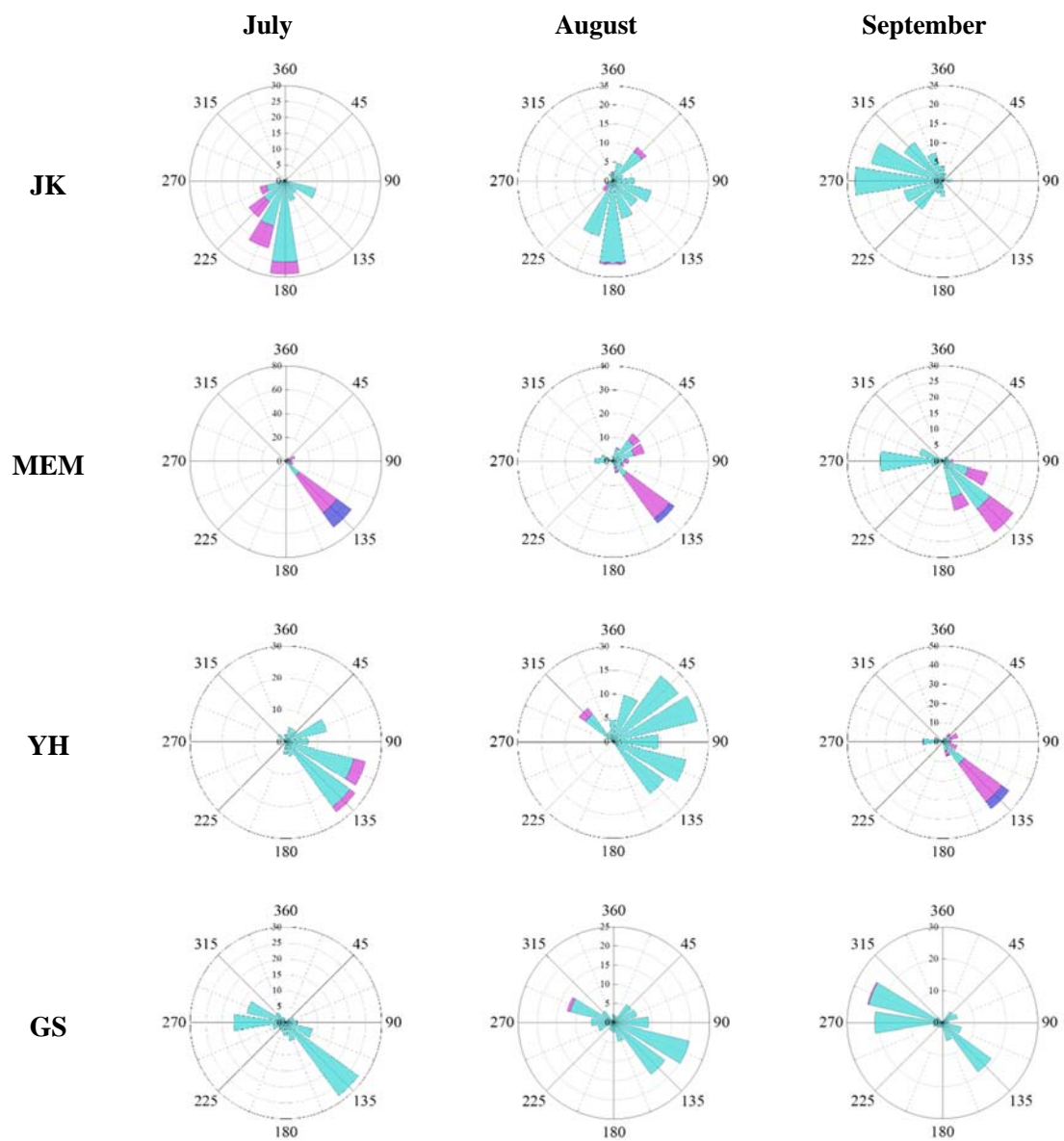


Fig. S2 The wind distribution at each site in July, August and September

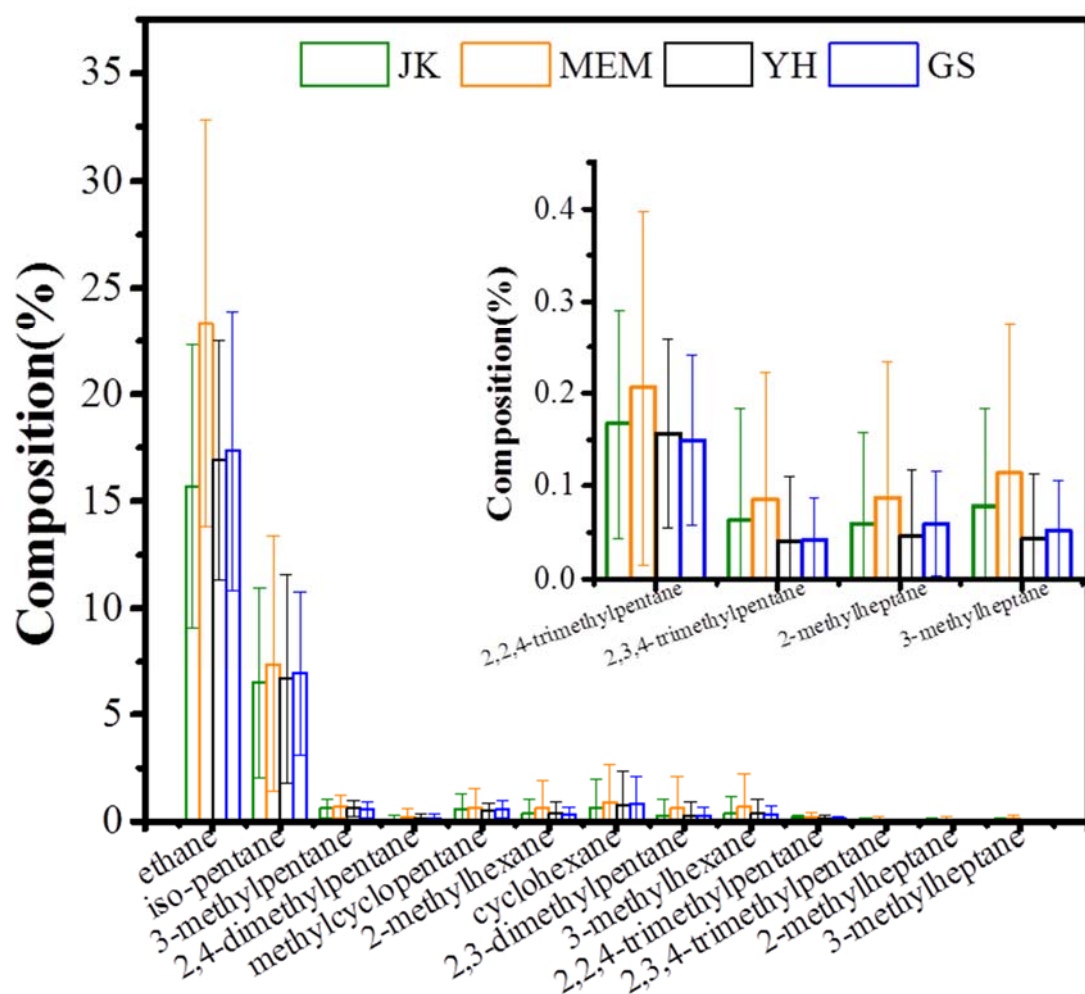


Fig. S3 Compositions of ethane, iso-pentane and C7-C8 alkanes at JK, MEM, YH and GS

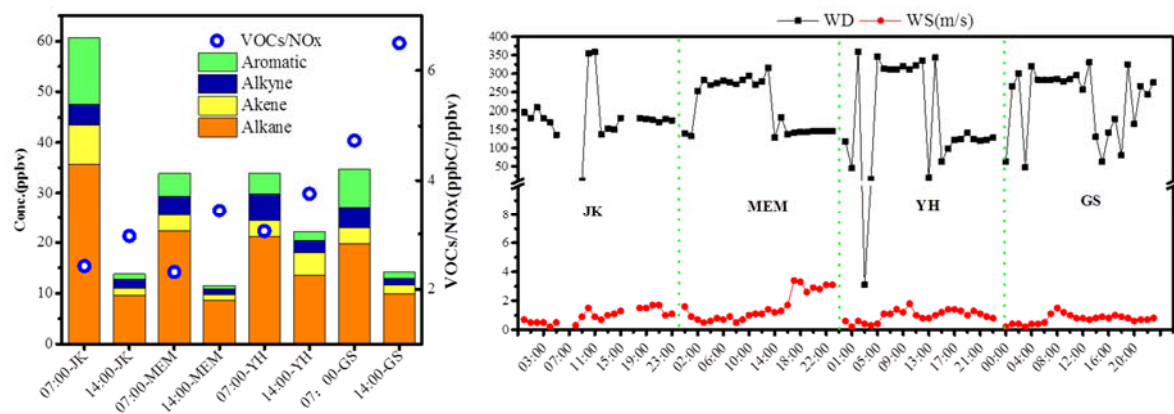


Fig.S4 Temporal variation of compositions, VOCs/NOx, wind direction and wind speed on 10th of August 2017

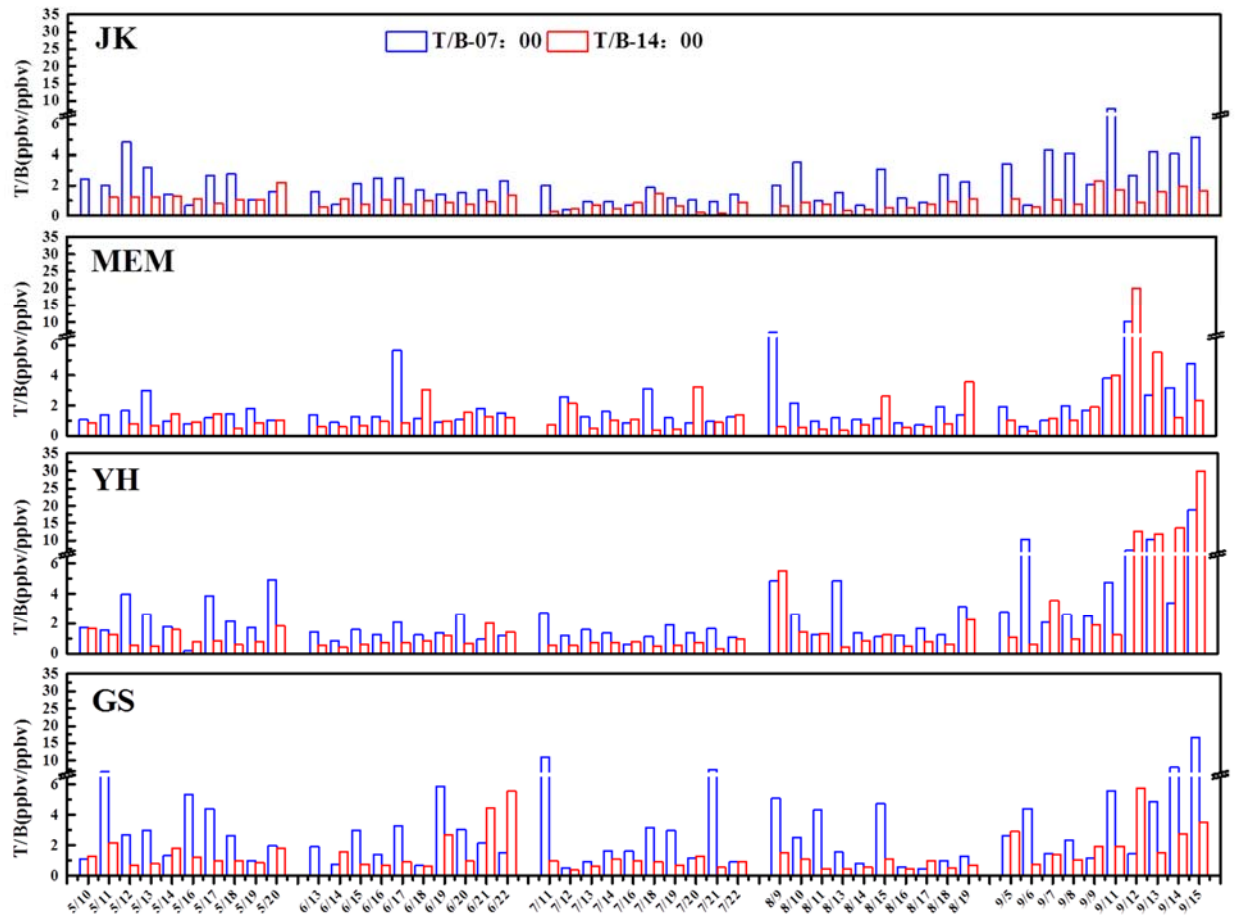
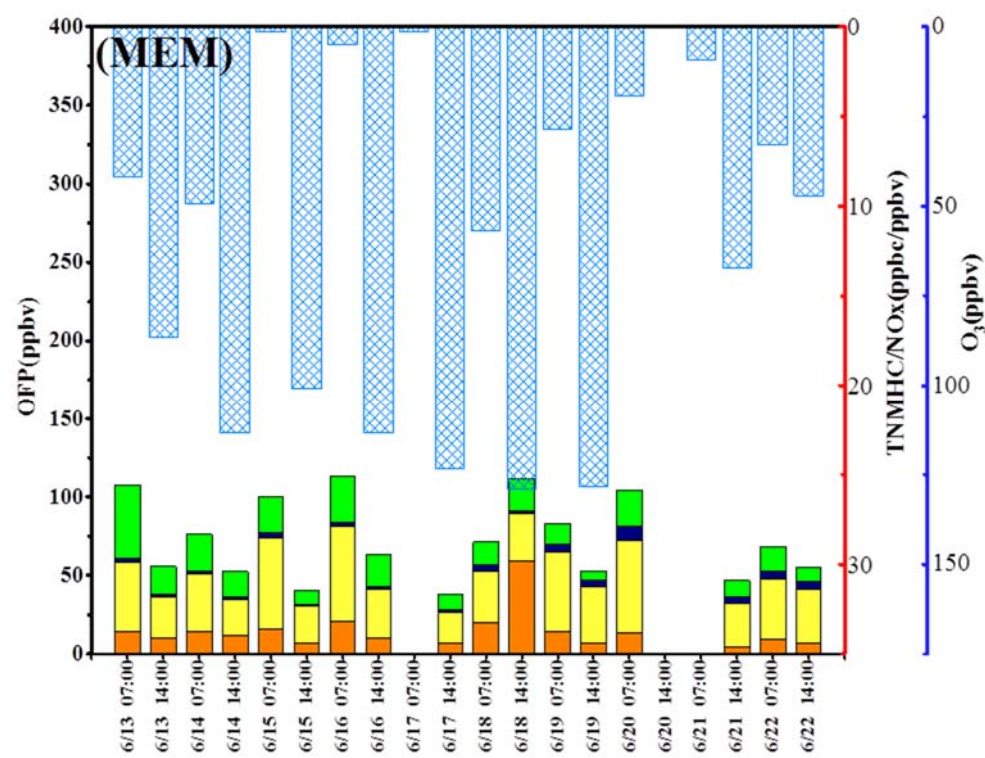
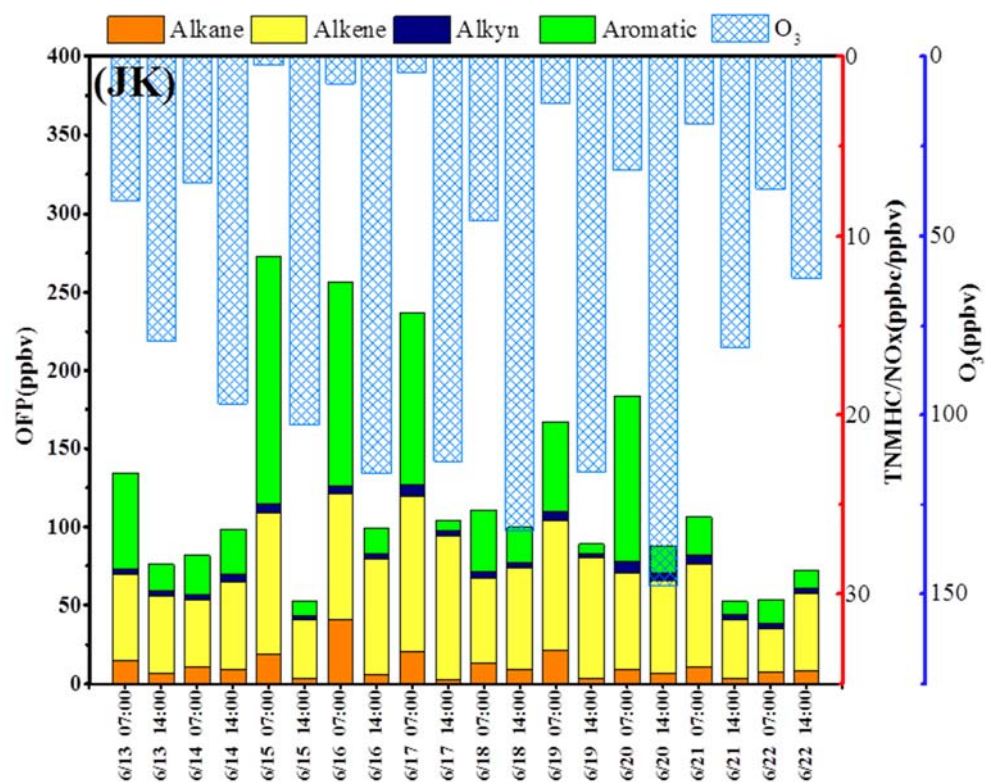


Fig.S5 Time series of T/B ratios for each site during May-September, 2017



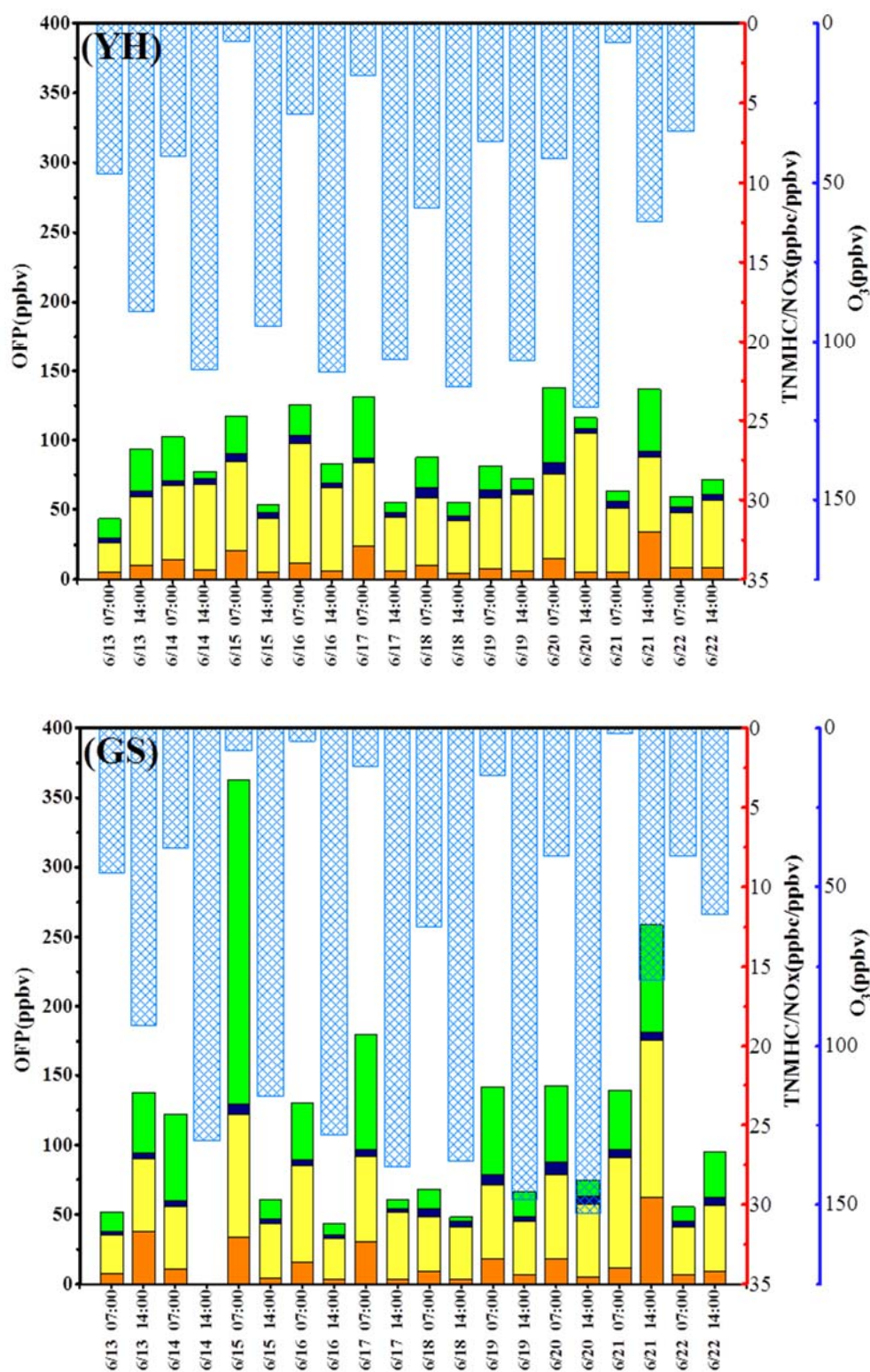


Fig.S6 Spatio-temporal variations in meteorological factors, OFP of each organic group, and mixing ratios of O₃ in June.

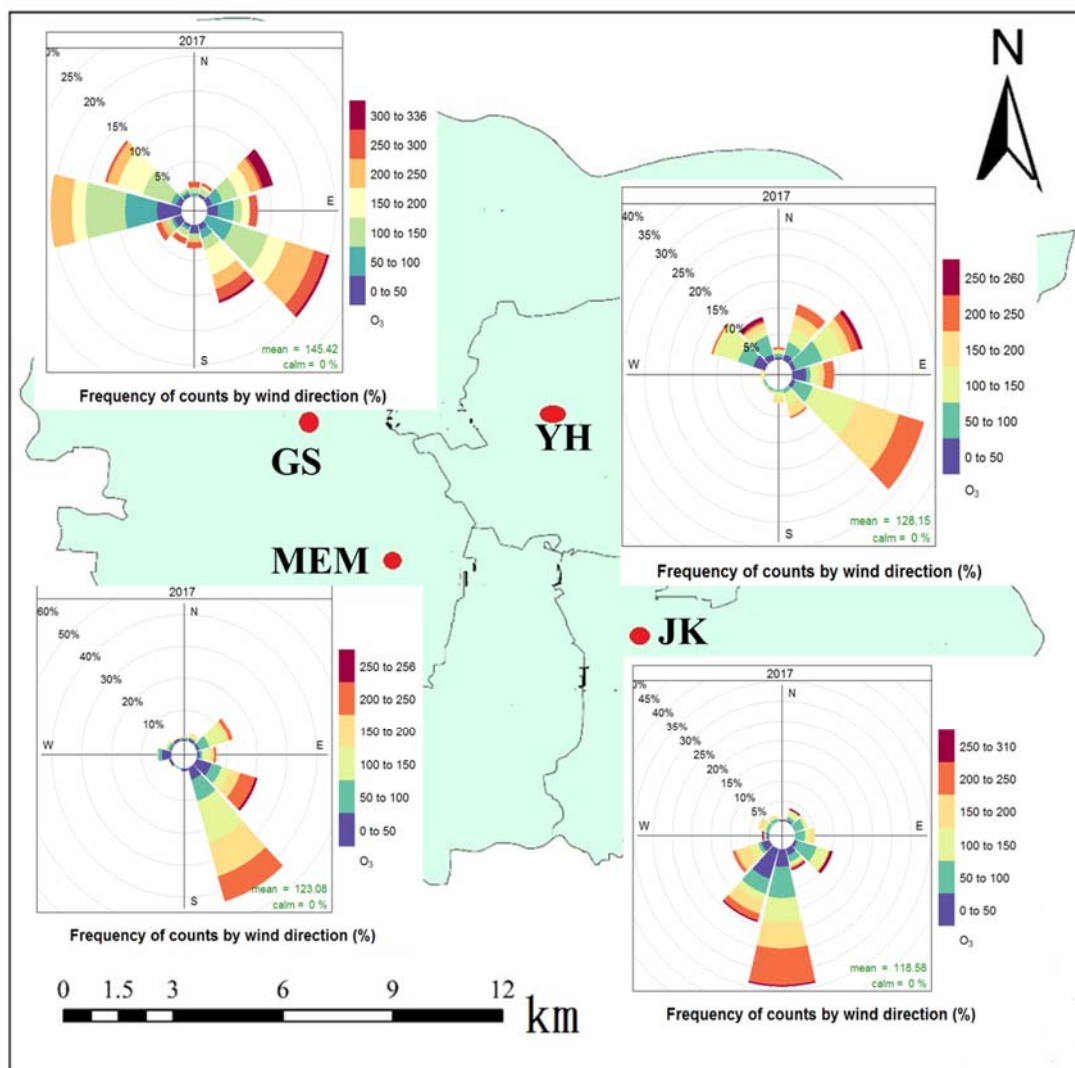


Fig.S7 Relationship among O₃(μ g m⁻³), wind direction and wind speed (m s⁻¹) during sampling period in June, 2017