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Characteristics and sources of non-methane hydrocarbons in background atmospheres of eastern, southwestern, and southern China

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[1] Non-methane hydrocarbons (NMHCs) play very important roles in atmospheric chemistry, yet little is known about their abundance in the atmosphere of China, especially in the background atmospheres. Measurements of C_2-C_{10} NMHCs were concurrently carried out at two remote background sites and one rural background site in April and May 2004 in this study: Tengchong Mountain (TM) in Yunnan province on the border of southwestern China; Jianfeng Mountain (JM) on the southwest coast of Hainan Island, southern China; and Lin'an (LA) in Zhejiang province in eastern China. Additional samples were collected at an urban site, Tengyue Town (TT, about 10 km from TM), to obtain local urban NMHC profiles. NMHC levels in these background sites were compared with other rural and remote sites in Asia and other global background stations. In the urban site of TT, local biofuel combustion was the major contributor to the high levels of ethene, ethyne, and ethane. In LA, the major sources of NMHCs were the emissions from fossil fuel and biofuel combustion and from industrial solvent use. Compared with another study conducted in 2001, toluene levels had increased by 75% in 2004 in this study, whereas the levels of other anthropogenic hydrocarbons remained at the same levels. Local industrial emissions and transports from the Yangtze River Delta were the major sources of toluene, ethylbenzene, and xylenes in LA. While in TM, the major sources were regional biofuel combustions and biomass burning emissions from Southeast Asia. Long-range transport of air masses from Southeast Asia was the major source of NMHCs in JM in spring. JM showed the lowest levels of most hydrocarbons among the three background sites. Several hydrocarbon molar ratios, such as benzene/toluene and ethane/propane, were used to characterize the signatures of air masses from different regions.

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1. Introduction

[2] With the rapid economic growth and fast industrialization and urbanization, China had experienced unprecedented achievements in the improvement of the living standards of its citizens and the alleviation of public poverty. Meanwhile, the excess use of resources, dramatic changes of land use pattern and rapid increase of motor vehicles and industries resulted in severe degradation of air quality in many urban and rural regions in recent decades [*Luo et al.*, 2000; *He et al.*, 2002; *Tang et al.*, 2007a]. From 1980 to 2004, the number of motor vehicles in China increased from 1.8 millions to 26.9 millions, and the total energy consumption increased from 600 to 2030 million tons of standard coal equivalent (SCE), of which about 70% was from coal [*National Bureau of Statistic*, 2006]. The air pollution in many large cities had changed from the conventional coal combustion type to the mixed coal combustion/motor vehicle emission type, and even to the vehicle emission dominant type, because of the explosively increasing vehicle number and the extensive use of coal as an energy source [*He et al.*, 2002; *Barletta et al.*, 2005; *Song et al.*, 2007].

[3] As a secondary air pollutant, ozone is a good indicator of air quality, and hence has been monitored in various rural and urban sites for decades in China. Ozone levels had increased twofold in the urban area of Hong Kong from 1980s to 2000 [*Chan et al.*, 2003]. In the capital city of

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China, Beijing, a recent study revealed that ozone levels had increased by approximately 2% per year from 1995 to 2005 in the lower troposphere [*Ding et al.*, 2008]. In a remote World Meteorological Organization (WMO) Baseline Station at Tibetan Plateau, Waliguan Observatory, northwest China, a slightly increasing trend (0.23 ppbv per year) was observed for surface ozone from 1994 to 2005 [*Deliger and Zhao*, 2007]. Elevated ozone concentrations were also observed in rural and remote sites of eastern China [*Luo et al.*, 2000; *Wang et al.*, 2006].

[4] Non-methane hydrocarbons (NMHCs) are important ozone precursors. Their reactions with the hydroxyl radical (OH) produce many oxygenic compounds, and result in the formation of ozone and other secondary air pollutants in the atmosphere [*Poisson et al.*, 2000]. Because of their important role in the air quality and the formation of ozone, the World Meteorological Organization Global Atmospheric Watch (WMO/GAW) programme commenced to measure NMHCs and other reactive compounds in the global network [*WMO*, 2007]. Several campaigns had been carried out at the WMO background stations, such as Mauna Loa Observatory, Hawaii [*Greenberg et al.*, 1996]; Cape Grim, northwest Tasmania [*Lewis et al.*, 2001]; Alert, Canada [*Gautrois et al.*, 2003], to better understand the atmospheric processes of NMHCs. Atmospheric NMHCs have also been extensively investigated in various rural and remote regions of North America and Europe [*Jobson et al.*, 1994; *Hagerman et al.*, 1997; *Klemp et al.*, 1997; *Kang et al.*, 2001; *Swanson et al.*, 2003]. However studies in China are scarce and sporadic, unmatched with its economy, geography and population size. Several field measurements have been performed to explore the distributions and sources of NMHCs in China recently. A study in a rural site of the Yangtze Rive Delta (YRD) showed that vehicular emissions, biofuel combustion, biomass burning and industrial emissions were the major contributors to hydrocarbons and other trace gases [*Guo et al.*, 2004]. In industrial, industrial-urban, and industrial-suburban areas of the Pearl River Delta (PRD), industrial emissions greatly influenced the ambient levels of NMHCs [*L. Y. Chan et al.*, 2006]. In the urban site of Guangzhou and a rural site of PRD, the composition of NMHCs had changed greatly from 2001 to 2005 because of the drastic social-economical developments in this region [*Tang et al.*, 2008].

[5] Most of these studies were carried out at the economically developed regions of China (such as YRD and PRD). Few NMHC data are available in the background sites of China. *Xu et al.* [1996] carried out a pilot study on NMHCs at three WMO/GAW stations in China (i.e., Waliguan baseline station, Longfengshan background station and Lin'an background station) from August 1994 to April 1995. Later between October 2003 and July 2004, a field study was conducted at Lin'an background station and Shangdianzi background station, which also are WMO/GAW stations, using the adsorbent tube method, to explore the behavior of NMHCs in these background sites [*J. Tang et al.*, 2007]. In this study, we collected canister samples at three background sites of eastern, southwestern, and southern China, to better understand the atmospheric composition of NMHCs and its implications on atmospheric chemistry in the background sites, and to explore the factors that influence the level of these trace gases, These data are crucial to the understanding of the impacts of the fast regional social-economic developments on the regional air quality.

2. Experiment

2.1. Sampling Site Description

[6] Ambient air samples were collected at two remote background sites and one rural background site of China from April to May 2004. Additional air samples were also collected in Tengyue Town to obtain the urban NMHC profiles. Figure 1 shows the locations of these three background sites and Table 1 shows the characteristics of these background and urban sites.

2.1.1. Lin'an (LA)

[7] Lin'an is a rural background site in the YRD. The sampling site is located in the Baseline Air Pollution Monitoring Station of China in Lin'an, one of the three regional background stations of WMO in China. This site represents the regional background atmosphere in the YRD. The general surroundings of this Lin'an monitoring station include the Lin'an Township (with an urban population of 70 thousands in 20 km^2 area), 10 km to the south; Hangzhou (with an urban population of 4.1 millions in 3100 km^2 area), the capital city of Zhejiang province, 53 km to the east; and Shanghai metropolis (with an urban population of 13.7 millions in 5300 km^2 area), 210 km to the northeast. To the west are the less developed and less populous mountainous regions. Near the station, several small villages are within 2 km range. Air samples were collected on the rooftop of a two-story building in the station. Information about this site can also be found in *Cheung and Wang* [2001] and *Wang et al.* [2004].

2.1.2. Tengchong Mountain (TM)

[8] Tengchong Mountain is a remote site on the eastern border of Yunnan province. It is located about 60 km to the east of the Burma border and 750 km to the west of Kunming (with an urban population of 3 millions in 225 km^2 area), the capital city of Yunnan province. The closest town, Tengyue Town is about 10 km to the south. There are several small villages surrounding the sampling site. Air samples were collected on the mountaintop. At the mountainside, there is an observatory, Tengchong station, which is a station of the Global Climate Observing System (GCOS), WMO. The details about this site can be found in the work of *C. Y. Chan et al.* [2006].

2.1.3. Jianfeng Mountain (JM)

[9] Jianfeng Mountain is situated at the southwest of Hainan Island, south China, and is a remote background site. It is about 20 km from the coast of Beibu Gulf (Gulf of Tonkin) to the west; 120 km from Sanya, the second largest city of Hainan Province, to the northeast; and 315 km from Haikou, the capital city of Hainan Province, to the southeast. It is mountainous to the north and east. This sampling site is surrounded by tropical rain forest with a total area of 475 km^2. It locates in a National Field Station for the tropical rain forest. Air samples were collected on the rooftop of a two-story building in this station. There are several hills around the sampling site with elevation above 1000 meters. The details about this site can be found in the work of *Tang et al.* [2007b].

Figure 1. Map showing the sampling sites.

2.1.4. Tengyue Town (TT)

[10] To better understand the impacts of local urban emissions on the air quality of the remote background sites, one city was selected to collect air samples. Tengyue Town (TT) is the capital city of Tengchong County and is the closest city to TM. The total population of TT is 108 thousands, and the urban population is 44 thousands in 14 km^2 areas. It is a typical small size town in rural regions of China with a small number of industrial enterprises. Air samples were collected on the rooftop of a five-story building in the urban center.

2.2. Sampling Procedure

[11] Air samples were collected by 2-L electropolished stainless steel canisters provided by the Research Center for Environmental Changes (RCEC), Academia Sinica, Taiwan. All canisters were humidified and evacuated prior to sample, as described by the standard method TO-15 [*U.S. EPA*, 1999]. About 50 canisters were transported to each station around 1 April 2004. The second batch of about 50 canisters was dispatched to each station around 10 May. Air samples were collected from 08:00 to 23:00 local time at an interval of three hours in each day to monitor the diurnal trend of NMHCs at these background sites. Approximately 6–8 days' diurnal samples were collected at each site in each month, and the other samples were taken around 12:00–14:00. Twelve canister samples were collected in TT. Air samples were taken around 14:30 from 18–24 May 2004, and five additional samples were taken at 8:00, 11:00,

Table 1. Characteristics of the Sampling Sites

Site	Abbreviation	Location	Altitude[a]	Sampling Period	Site Description
Jianfeng Mountain	JM	18°40′N, 108°49′E	820	9–24 Apr, 12–21 May 2004	Remote site, in a tropical rain forest
Tengchong Mountain	TM	24°57′N, 98°29′E	1960	8–17 Apr, 18–24 May 2004	Remote site, in mountainous area with small villages around
Lin'an Station	LA	30°30′N, 119°75′E	130	2 Apr to 20 May 2004	Rural site, in mountainous area with small villages around
Tengyue Town	TT	25°01′N, 98°30′E	1660	18–24 May 2004	Urban site, on rooftop of a five-story building in urban center

[a]Unit: m, above sea level.

Table 2. Precision, Measurement Detection Limit (MDL) and Response Factor RSD of Calibration Curves for the Measured Species

Group	Species	Precision[a] (%)	MDL[b] (ppbv)	Response Factor RSD[c] (%)
Alkanes	ethane	0.7	0.039	4.0
	propane	0.5	0.031	8.4
	i-butane	0.8	0.017	5.5
	n-butane	0.6	0.021	4.4
	i-pentane	1.8	0.014	2.3
	n-pentane	1.8	0.008	11.0
	2,2-dimthylbutane	1.9	0.014	4.5
	cyclopentane	3.5	0.009	5.4
	2-methylpentane	1.3	0.008	6.3
	3-methylpentane	1.9	0.007	1.8
	n-hexane	3.7	0.006	4.9
	2,4-dimethylpentane	1.5	0.004	2.1
	methylcyclopentane	1.2	0.009	3.2
	cyclohexane	1.5	0.006	4.0
	2-methylhexane	1.9	0.006	8.9
	2,3-dimethylpentane	1.8	0.009	2.9
	3-methylhexane	1.7	0.006	3.7
	2,2,4-trimethylpentane	2.8	0.009	4.7
	n-heptane	1.9	0.010	12.2
	methylcyclohexane	1.8	0.005	3.8
	2,3,4-trimethylpentane	1.6	0.006	4.0
	2-methylheptane	1.5	0.004	2.9
	3-methylheptane	2.4	0.005	2.6
	n-octane	0.9	0.006	2.4
	n-nonane	1.9	0.006	5.6
Alkenes	ethene	0.6	0.041	6.5
	propene	0.5	0.031	2.5
	trans-2-butene	1.0	0.013	3.0
	1-butene	0.8	0.017	2.3
	i-butene	0.7	0.034	5.1
	cis-2-butene	0.5	0.011	4.2
	3-methyl-1-butene	1.4	0.006	1.8
	1-pentene	1.9	0.020	4.3
	isoprene	1.4	0.013	3.0
	trans-2-pentene	1.3	0.010	9.9
	cis-2-pentene	2.6	0.006	10.1
	2-methyl-2-butene	1.8	0.006	2.9
	cyclopentene	2.7	0.006	3.5
	4-methyl-1-pentene	1.2	0.013	5.3
	2-methyl-1-pentene	4.0	0.004	9.1
	trans-2-hexene	5.0	0.023	2.7
	cis-2-hexene	3.8	0.009	5.7
	alpha-pinene	1.3	0.009	5.4
Alkyne	ethyne	1.0	0.057	9.4
Aromatics	benzene	1.6	0.014	9.4
	toluene	1.6	0.009	6.2
	styrene	1.3	0.006	5.2
	ethylbenzene	1.4	0.006	1.5
	m/p-xylene	1.9	0.009	11.0
	o-xylene	1.1	0.004	2.8
	i-propylbenzene	1.5	0.004	2.3
	n-propylbenzene	1.4	0.004	3.1
	1,3,5-trimethylbenzene	2.0	0.003	3.1
	1,2,4-trimethylbenzene	1.5	0.006	2.0

[a]Reproducibility for measured species evaluated by repeated injection of a standard mixture at around 1 ppbv (n = 7).
[b]Each aliquot of 210 mL drawn from the canister for analysis.
[c]Injecting various concentrations of a gas standard mixture containing target species with concentrations in the range of 0.05 to 15 ppbv.

14:00, 17:00 and 20:00 on 23 May 2004. A stainless steel passive capillary tube was attached to the inlet of each canister to collect a 1-hour integrated air sample. After sampling, all canisters were returned to the RCEC and were analyzed within one month.

2.3. Chemical Analysis

[12] Air samples were analyzed by RCEC with an automated GC/MS/FID (Varian CP-3800 and Saturn 2200 ion trap MS) system using dual columns and dual detectors to simultaneously analyze both low- and high-boiling-point NMHCs with each injection. The analytical system is the upgrade of the one as described by *Chang et al.* [2003]. The fundamental framework and analytical procedures between them are the same, and details can refer to the work of *Chang et al.* [2003]. Here we only describe the features of the upgraded system briefly. The PLOT column connected to a FID was responsible for separation and detection of C_2-C_4 compounds, and the DB-1 column was connected to the ion trap MS for the separation and detection of C_4-C_{11} compounds. Each aliquot of 210 mL was drawn from the canister to the cryogenic trap packed with fine glass beads cooled at $-170°C$ for preconcentration. During injection, the trap was resistively heated up to 85°C within seconds, and a stream of ultrahigh purity helium (99.9999%) was used to flush the trapped hydrocarbons onto the columns. Ultrahigh purity helium used as carrier gas as well as carefully selecting characteristic ion of each compound with the least interference in MS spectrum as quantitative ion are quite beneficial to decrease baseline noise and hence lower detection limits and improve precision. Two standard gas cylinders (68 C_2-C_{11} NMHCs, Scott Marrin Inc., USA; 57 C_2-C_{12} NMHCs, Spectra Gases Inc., USA) were employed for concentration calibration and quality control purposes. Calibration curve was made by injecting various concentrations of gas standard mixture with concentrations in the range of 0.05 to 15 ppbv. Table 2 shows the precision, measurement detection limit (MDL) and response factor RSD of calibration curves for the measured species. The precisions (1σ) and detection limits of the upgraded method for most species are 0.5–2% and below 10 pptv, respectively.

3. Results and Discussion

3.1. Characteristics of Total and Dominant NMHC Species

[13] A total of 56 Photochemical Assessment Monitoring Stations (PAMS) targeted compounds were identified and quantified. At the three background sites, mixing ratios of some hydrocarbons were very low and some even below the detection limit. Hence only the common and abundant species in the atmosphere are presented. The statistics of these selected hydrocarbons at the four sites are shown in Table 3. Selected NMHC data at JM was published by *Tang et al.* [2007b]. Here the entire data set is presented for comparison. The total mixing ratios of all hydrocarbons (total NMHC, in ppbv) measured and quantified are also shown in Table 3. Although some hydrocarbons were not listed in Table 3, their mixing ratios were added in their corresponding hydrocarbon groups and also the total NMHC. The listed C_2-C_7 hydrocarbons accounted for approximately 90% of the total NMHC in background sites and 80% in the urban site. The mixing ratios of total NMHC in ascending order are: JM, TM, LA and TT. Ethane, ethyne, ethene, propane and benzene were among the top 10 hydrocarbons on the mixing ratio scale.

[14] Among the three background sites, JM shows the highest levels of isoprene but the lowest levels of other

Table 3. Statistics of Selected Hydrocarbon Mixing Ratios at the Four Sites (in Parts Per Billion by Volume)

	JM (n = 91)[a]					TM (n = 78)					LA (n = 83)					TT (n = 12)				
Hydrocarbon	Mean	SD	Median	Min	Max	Mean	SD	Median	Min	Max	Mean	SD	Median	Min	Max	Mean	SD	Median	Min	Max
Ethane	0.86	0.45	0.74	0.38	3.20	1.40	0.33	1.38	0.71	2.20	2.63	0.81	2.50	1.29	8.32	2.31	0.86	2.00	1.36	3.76
Propane	0.17	0.16	0.13	0.02	1.26	0.23	0.07	0.22	0.07	0.41	1.31	1.01	1.06	0.37	7.98	0.73	0.42	0.56	0.36	1.72
i-Butane	0.05	0.09	0.03	BDL[b]	0.80	0.03	0.01	0.03	0.01	0.08	0.51	0.46	0.37	0.08	2.85	0.54	0.39	0.40	0.17	1.59
n-Butane	0.10	0.18	0.06	0.02	1.46	0.06	0.03	0.06	0.02	0.20	0.73	0.77	0.52	BDL	5.31	0.48	0.26	0.42	0.17	0.97
i-Pentane	0.20	0.18	0.16	BDL	1.22	0.15	0.12	0.13	0.03	0.76	0.46	0.28	0.38	BDL	1.33	1.15	0.56	1.08	0.54	2.36
n-Pentane	0.15	0.15	0.12	0.06	1.44	0.12	0.04	0.11	0.06	0.25	0.29	0.24	0.25	0.09	2.02	0.49	0.20	0.49	0.27	0.89
Ethene	0.38	0.32	0.32	0.14	2.14	0.87	0.43	0.73	0.33	2.28	1.73	1.17	1.52	0.37	8.45	4.73	3.12	3.16	1.23	9.74
Propene	0.11	0.08	0.10	0.05	0.77	0.17	0.07	0.15	0.09	0.50	0.33	0.27	0.29	0.09	2.11	0.86	0.56	0.56	0.22	1.89
Isoprene	0.55	0.52	0.32	0.02	2.19	0.08	0.16	0.05	0.01	1.32	0.40	0.39	0.27	0.02	1.88	0.13	0.06	0.10	0.06	0.25
α-Pinene	0.05	0.03	0.05	BDL	0.12	0.05	0.04	0.04	0.02	0.18	0.17	0.12	0.14	0.01	0.74	0.13	0.10	0.10	0.05	0.40
Ethyne	0.39	0.35	0.31	0.04	2.97	0.78	0.31	0.76	0.05	1.41	2.18	0.93	1.90	0.79	4.54	4.35	2.19	3.55	1.42	7.45
Benzene	0.13	0.09	0.11	0.05	0.79	0.24	0.07	0.23	0.10	0.40	0.75	0.37	0.67	0.24	2.41	1.13	0.59	0.92	0.45	2.12
Toluene	0.17	0.44	0.09	0.05	4.16	0.16	0.10	0.13	0.05	0.54	2.64	3.00	1.90	0.22	16.07	1.08	0.78	0.85	0.37	2.86
Ethylbenzene	0.02	0.02	0.02	BDL	0.12	0.03	0.04	0.02	0.01	0.30	0.33	0.26	0.27	0.03	1.47	0.27	0.16	0.25	0.08	0.68
m/*p*-Xylene	0.04	0.03	0.03	BDL	0.19	0.06	0.10	0.04	0.02	0.79	0.32	0.26	0.25	0.04	1.33	0.68	0.38	0.63	0.24	1.61
o-Xylene	BDL	BDL	BDL	BDL	0.07	0.03	0.05	0.01	0.01	0.41	0.15	0.15	0.11	0.02	1.01	0.24	0.13	0.21	0.09	0.54
Alkanes	1.64	0.92	1.39	0.60	7.11	2.09	0.47	1.98	1.07	3.22	7.02	3.46	6.17	2.42	23.03	7.58	3.26	7.18	3.86	12.59
Alkenes[c]	0.68	0.44	0.58	0.31	3.30	1.25	0.53	1.08	0.63	3.18	2.44	1.72	2.23	0.59	13.42	7.56	4.60	5.42	2.36	15.78
Alkynes	0.39	0.35	0.31	0.04	2.97	0.78	0.31	0.76	0.05	1.41	2.18	0.93	1.90	0.79	4.54	4.35	2.19	3.55	1.42	7.45
Aromatics	0.40	0.51	0.31	0.13	4.63	0.62	0.49	0.53	0.25	3.83	4.40	3.99	3.51	0.62	23.66	3.75	2.03	3.40	1.69	8.44
Biogenics	0.59	0.54	0.35	0.01	2.28	0.12	0.15	0.09	0.02	1.36	0.55	0.46	0.46	0.01	2.14	0.25	0.15	0.20	0.13	0.58
Total NMHC	3.70	1.85	3.55	1.56	15.08	4.87	1.28	4.69	2.56	7.55	16.59	8.21	15.32	7.24	48.15	23.50	11.78	19.47	9.95	44.73

[a]The total number of samples.
[b]BDL, below detection limit.
[c]Isoprene and α-pinene not included in alkenes but in biogenics.

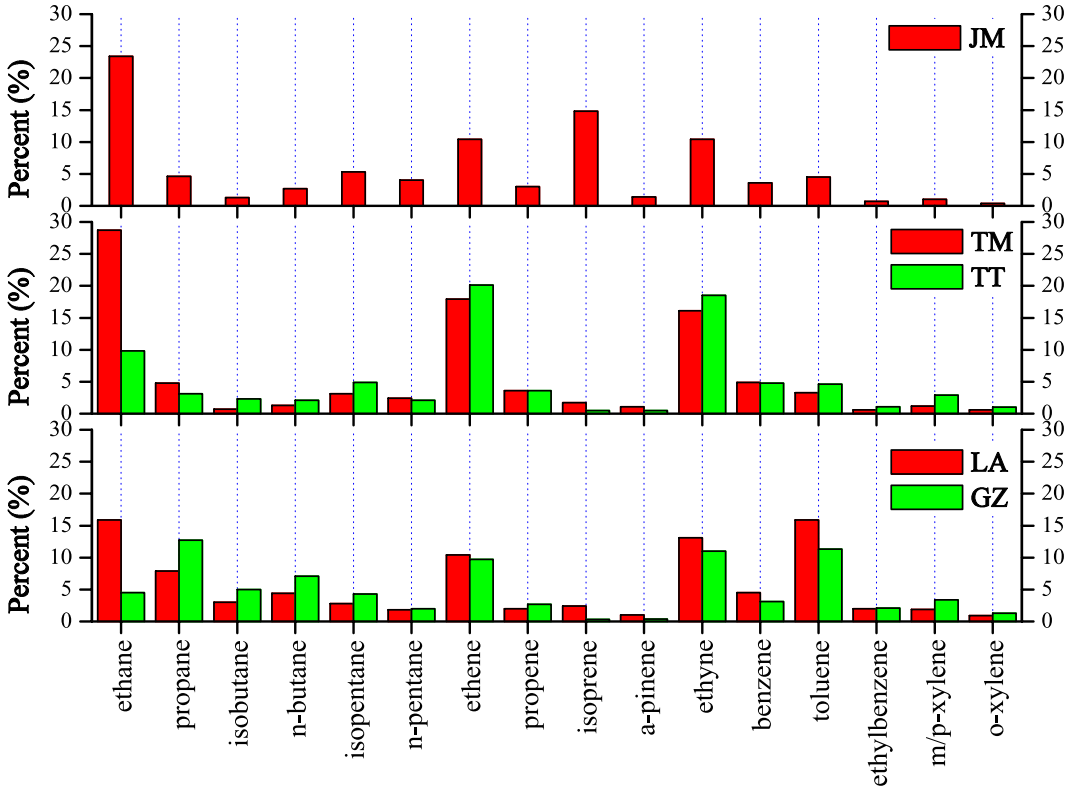

Figure 2. Distributions of hydrocarbons at the four sites and compared with Guangzhou (based on mixing ratio scale).

hydrocarbons. LA shows the highest levels of most hydrocarbons, except for isoprene. The NMHC compositions were calculated at the three background sites and the urban site, by dividing the mean mixing ratios of certain hydrocarbons by the mixing ratio of total NMHC in percentage (Figure 2). The hydrocarbon data collected in Guangzhou (GZ, the capital city of Guangdong Province) in April 2005 were cited in this study for comparison [*Tang et al.*, 2007a]. The air samples in GZ were collected at the rooftop of a 21-story building, located in the urban center. These samples represented well mixed urban air masses. The samples in GZ were analyzed by the RCEC using the same method as this study. The profiles of NMHCs in these sites varied noticeably. Ethane, ethene and ethyne were among the top 5 abundant hydrocarbons at the three background sites and TT. While in GZ, propane, toluene, ethyne, ethene and *n*-butane were the top five ones. At JM, isoprene was the second largest contributor to the total NMHC (15%) after ethane (23%). While at LA, toluene and ethane were the largest contributors (16%). The distributions of hydrocarbons in LA and GZ were similar except for a higher ethane percentage in LA and a higher propane percentage in Guangzhou. This indicates that the two stations shared similar emission sources for most of hydrocarbons. The distributions of hydrocarbons at TM and TT were very similar except for ethane, which was approximately three times higher at TM (29%) than at TT (9%). TT also showed slightly higher concentrations of ethene, ethyne, *n*-butane, *i*-pentane and toluene than those at TM. Between the two urban sites, TT and GZ, the distributions of hydrocarbons were different notably, indicating the sources of hydrocarbons in these two sites varied remarkably.

[15] Table 3 also shows the mixing ratios of hydrocarbon groups at different sites. Isoprene and α-pinene were categorized into biogenics, as their major sources in rural and forest sites are biogenic emissions, although we noted that anthropogenic emissions of isoprene were also reported in some urban and rural environments [*Reimann et al.*, 2000; *L. Y. Chan et al.*, 2006; *Durana et al.*, 2006]. Alkanes were the dominant groups at all sampling sites, with similar percentages to the total NMHC at the three background sites (41–44%), but were the lowest at TT (32%). The highest levels of alkenes and alkynes (i.e., ethyne) were found at TT. The highest levels of aromatics were found at LA, even higher than those at the urban TT area. The variations of the fractional contributions are attributed to the sources of hydrocarbons at different sites, and information about NMHC emission sources at each site will be discussed in the following section.

3.2. Comparison With Other Studies in Rural and Remote Sites

[16] Sporadic data collected in Asia are available only recently, and they were mostly collected in Hong Kong and Japan [*Sharma et al.*, 2000a, 2000b, 2000c; *Kato et al.*, 2001, 2004; *Wang et al.*, 2003, 2005; *Guo et al.*, 2007]. Table 4 shows the average mixing ratios of hydrocarbons at various rural and remote sites in Asia. The mixing ratios of hydrocarbons at different sites are influenced by many factors, such as sampling period, geographical location, source emission strength, proximity to the source region,

Table 4. Comparison of Average Mixing Ratios of Hydrocarbons at Various Rural or Remote Sites in Asia (in Parts Per Billion by Volume)

Hydrocarbon	Chichi-jima[a] Japan Aug 1997–1998	Oki[b] Japan Jul–Aug 1998	Nagarkot[c] Nepal Nov 1998	Happo[d] Japan Spring 1999	Okinawa[e−1] Japan Nov–Dec 2000	Okinawa[e−2] Japan Nov–Dec 2000	Lin'an[f] China Mar 2001	Hok Tsui[g] Hong Kong Feb–Apr 2001	Heng Chuen[h] South Taiwan Oct 2002	Tai O[i−1] Hong Kong Aug 2001–Dec 2002	Tai O[i−2] Hong Kong Aug 2001–Dec 2002	Tap Mun[j] Hong Kong Sept 02–Aug 03	Mt. Abu[k] India April 2002	JM This Study	TM This Study	LA This Study
Ethane	1.58	0.72	1.10	1.74	1.90	1.14	3.11	2.37	2.3	0.47	2.22	1.79	1.33	0.86	1.40	2.63
Propane	0.54	0.16	0.25	0.61	0.75	0.41	1.20	0.81	0.9	0.09	2.17	0.86	0.44	0.17	0.23	1.31
i-Butane	0.17	0.05	0.13	0.13	0.14	0.07	0.39	0.22	0.1	0.03	0.85	0.34	0.24	0.05	0.03	0.51
n-Butane	0.10	0.08	0.32	0.22	0.23	0.12	0.43	0.33	0.4	0.05	1.74	0.59	0.29	0.10	0.06	0.73
i-Pentane	0.05	0.06	0.06	0.11	0.10	0.05	0.33		1.3	0.06	0.85	0.37		0.20	0.15	0.46
n-Pentane	0.08	0.04	0.06	0.08	0.08	0.04	0.13	0.08	0.3	0.02	0.48	0.17		0.15	0.12	0.29
Ethene	0.17	0.26	0.69	0.21	0.37	0.16	1.61	0.50	1.5	0.14	1.78	0.86	0.30	0.38	0.87	1.73
Propene		0.11	0.14	0.06	0.06	0.04	0.28	0.06	0.7	0.06	0.24	0.14	0.16	0.11	0.17	0.33
Isoprene	0.08	0.73					0.07		0.5	0.84	0.37	0.33		0.55	0.08	0.40
Ethyne	0.45	0.20	0.52	0.70	0.84	0.39	2.39	1.40	0.4	0.14	2.94	1.36	0.88	0.39	0.78	2.18
Benzene	0.12				0.19	0.08	0.80	0.49	0.3	0.05	0.92	0.40		0.13	0.24	0.75
Toluene	0.08				0.11	0.09	1.50	0.54	0.9	0.07	6.01	1.03		0.17	0.16	2.64
Ethylbenzene							0.19	0.06	0.2	0.02	0.93	0.12		0.02	0.03	0.33
m/p-Xylene							0.49		0.3	0.66	1.03	0.16		0.04	0.06	0.32
o-Xylene							0.19		0.3	0.03	0.40	0.06		BDL	0.03	0.15

[a]*Kato et al.* [2001].
[b]*Sharma et al.* [2000a].
[c]*Sharma et al.* [2000b].
[d]*Sharma et al.* [2000c].
[e]*Kato et al.* [2004]; 1, air masses from Chinese coast area; 2, air masses from east-north directions.
[f]*Guo et al.* [2004].
[g]*Wang et al.* [2003].
[h]*Chang et al.* [2005].
[i]*Wang et al.* [2005]; 1, inflow of South China Sea; 2, outflow of regional air pollution.
[j]*Guo et al.* [2007].
[k]*Sahu and Lal* [2006].

meteorological condition, and others. Direct comparison of hydrocarbon levels at different sites should thus be prudently interpreted. Notwithstanding the limitations, the relative distributions of NMHCs at different sites provided a clue to their source patterns and their regional air pollution impacts.

[17] Among these rural and remote sites in Asia, the lowest mixing ratios of anthropogenic hydrocarbons were found at Tai O, a coastal station in Hong Kong, where air masses were originated from marine boundary layer [*Wang et al.*, 2005]. However this site was also greatly influenced by regional air pollution. The highest mixing ratios of anthropogenic hydrocarbons (e.g., toluene, ethyne, propane and *n*-butane) were observed in the air mass from regional pollution plumes (Table 4). Very low levels of hydrocarbons were also found at a remote site in Oki Island, Japan, where most air masses were originated from the Pacific Ocean [*Sharma et al.*, 2000a]. High levels of NMHCs were found at other rural sites strongly influenced by nearby urban or local emissions, such as Hok Tsui [*Wang et al.*, 2003] and Lin'an [*Guo et al.*, 2004].

[18] JM is a clean remote site with the mixing ratios of all hydrocarbons under 1 ppbv, and the average mixing ratios of most anthropogenic hydrocarbons (for example, ethane, propane, ethene and ethyne) were lower than those found at other rural or remote sites in Asia, except at Tai O, where air masses originated from the South China Sea [*Wang et al.*, 2005], and Oki Island (Table 4). At JM, very low mixing ratios of hydrocarbons were also observed where air masses originated from the western Pacific Ocean. The average mixing ratios of ethane (0.45 ppbv), propane (0.05 ppbv), ethene (0.15 ppbv) and ethyne (0.06 ppbv) in these samples were very close to the hydrocarbon levels observed at Tai O (0.47, 0.09, 0.14, and 0.14 ppbv, respectively) in the air masses from marine boundary layer [*Tang et al.*, 2007b; *Wang et al.*, 2005]. It indicates that JM is a suitable remote site for collecting background air sample in south China.

[19] Compared with the global background atmosphere stations, the levels of ethane and ethyne at JM (0.86 and 0.39 ppbv, respectively) were close to those in the free troposphere at Mauna Loa Observatory in the mid Pacific in spring (1.00 and 0.22 ppbv, respectively [*Greenberg et al.*, 1996]). However the levels of anthropogenic alkanes (ethane, propane, butanes, and pentanes) at JM were significantly higher than those in the unpolluted Southern Ocean boundary layer air measured at Cape Grim from January to February 1999, and in clear air of Northern Hemisphere at Mace Head, Eire from July to August 1999 [*Lewis et al.*, 2001]. The levels of light hydrocarbons (ethyne, propane, butanes, and benzene) at JM were within the range of hydrocarbons measured in summer and winter in the lower polar troposphere at Alert station, Canada [*Gautrois et al.*, 2003]. It indicates that although JM is a remote background site, it is still influenced by the anthropogenic emission sources.

[20] TM is a remote mountainous site, and the mixing ratios of some anthropogenic hydrocarbons (e.g., ethane, ethene, ethyne and benzene) at TM are higher than those at JM but lower than those at LA. Comparing to another nearby rural site, Nagarkot in Nepal [*Sharma et al.*, 2000b], which has altitude (2015 m asl) and latitude (27.5°N) similar to those of TM (1960 m asl and 24.6°N, respectively), the mean mixing ratios of ethane, ethene and ethyne are higher at TM than at Nagarkot (Table 4). Higher levels of ethene than ethyne at these two rural sites indicate that there are strong local or fresh emission sources of ethene. At Mt. Abu (24.6°N, 72.7°E, 1680 m asl) in India, the levels of ethane (1.33 ppbv) and ethyne (0.78 ppbv) in April 2002, were close to those (1.40 and 0.94 ppbv, respectively) at TM. However the level of ethene (0.30 ppbv) at Mt. Abu was much lower than that at TM (0.87 ppbv). Mt. Abu is a remote site in India and the major sources of NMHCs were long-range transport from distant source regions [*Sahu and Lal*, 2006].

[21] Among the rural and remote sites in Asia, LA showed the second highest mixing ratios of toluene and ethylbenzene after Tai O (Table 4). Compared with the data collected in March 2001, by *Guo et al.* [2004], the mixing ratio of toluene had increased by 75% at Lin'an in 2004 in this study. The high levels of toluene and ethene at LA indicate that there were fresh or local emission sources nearby. The levels of most anthropogenic hydrocarbons (e.g., ethane, ethene, ethyne, and benzene) at LA were close to those observed at Tai O, Hong Kong, which was influenced by fresh PRD pollution plume. It implies that LA was influenced by nearby urban and industrial emissions.

3.3. Source Patterns of Hydrocarbons at Different Sites

[22] The JM site is located in the remote tropical rain forest with few households nearby, and is far away from urban or industrial areas, while TM and LA are surrounded by small villages and are located near to their capital cities. TM is about 10 km from Tengyue Town and LA is about 10 km from Lin'an Township. It is expected that TM and LA are more impacted by local anthropogenic emissions and should have higher levels of anthropogenic hydrocarbons than those at JM. Table 5 shows some social-economic statistical data of these three rural regions [*National Bureau of Statistics*, 2005a]. JM is under the administration of Ledong County. Most inhabitants lived in rural areas in these three regions (80% at Ledong County, 90% at Tengchong County and 90% at Lin'an City, Table 5) compared to 60% of the national population living in rural region in China in 2004 [*National Bureau of Statistics*, 2005b]. The three background sites represent different types of rural regions in China. Agriculture is the pillar sector of economy, and industry contributes little to local economy in Ledong County (Table 5). Lin'an City is located in the western part of YRD region. The number of industrial enterprises and their contributions to the GDP are at least one order of magnitude larger at Lin'an City than at the other two regions (Table 5). This was also reflected by the much higher levels of toluene at LA than at the other two sites.

3.3.1. LA

[23] Ethyne had been widely used as a tracer for combustion (both biofuel and fossil fuel combustion) emissions [*Choi et al.*, 2003; *Barletta et al.*, 2005; *Lee et al.*, 2006]. To assess the impact of combustion emissions on the ambient NMHC level, the average mixing ratios of hydrocarbons to ethyne were calculated (Figure 3). The data collected from March to June in 2001 by *Guo et al.* [2004] at the same site were compared with our data. The hydrocarbon/ethyne ratios matched very well for most hydrocarbons between the two studies, except for a remarkable increase in toluene/

Table 5. Comparisons of Tengchong County, Ledong County, and Lin'an City

Item	Unit	Ledong County[a]	Tengchong County	Lin'an City
Area	km^2	2747	5845	3124
Population	ten thousands	48	62	52
Population in rural	ten thousands	40	57	45
Value added of the primary industry	million Yuan	1479	807	1501
Value added of the secondary industry	million Yuan	107	556	7562
Total grain output	ton	171,184	248,358	93,139
Number of industry above designated size[b]	unit	14	19	428
Gross industry output value for the industry above designated size	million Yuan	68	393	13,980

[a]Jianfeng Mountain under the administration of Ledong County.
[b]Industry above the designated size, all state-owned enterprises and non-state-owned enterprises with annual sales revenue over 5 million Yuan (source: *National Bureau of Statistics* [2005a]).

ethyne ratio in 2004. The average mixing ratio of toluene increased from 1.50 ppbv in 2001 to 2.64 ppbv in 2004, while the levels of other hydrocarbons (such as ethene, ethyne, and benzene) were similar between the two studies. It indicates that the source patterns of NMHCs were similar between 2001 and 2004, except for increasing emission strengths of toluene from industrial sources. In the study of 2001 at LA, about 70% of emissions of the total volatile organic compounds were attributed to a combination of vehicular emissions and biofuel burning, and about 7% to gasoline evaporation and solvent emissions [*Guo et al.*, 2004]. In this study, the combustion of fossil fuel and biofuel might still be important sources for NMHCs, but the industrial emission played a more important role, as indicated by the increasing levels of toluene in 2004.

[24] Backward air trajectories were calculated for the sampling periods each day at LA to determine the source regions of NMHCs using the National Oceanic and Atmospheric Administration (NOAA) HYSPLIT Model [*Roph*, 2003]. During the study period, there are two major types of pathways for air masses to arrive at LA. One originated from the lower troposphere of northwestern or southern China, then passed over the less developed regions, and arrived at LA from the west (Type W). The other originated from the lower troposphere of northeastern China, then passed over the YRD region (especially Shanghai metropolis), and arrived at LA from the northeast (Type NE). Figure 4 shows the typical backward air trajectories of these two types. The levels (average plus standard deviation) of hydrocarbons from these two paths are compared in Table 6. Most hydrocarbons (except for isoprene) showed higher levels in air masses Type NE than in Type W. In Type W, air masses passed over the rural and mountainous regions, where there was more vegetation covered than that in the YRD region. It is reasonable that high isoprene levels were observed in Type W than in Type NE. In Type NE, air masses passed over the YRD region, where there were more urban and industrial emission sources and the anthropogenic

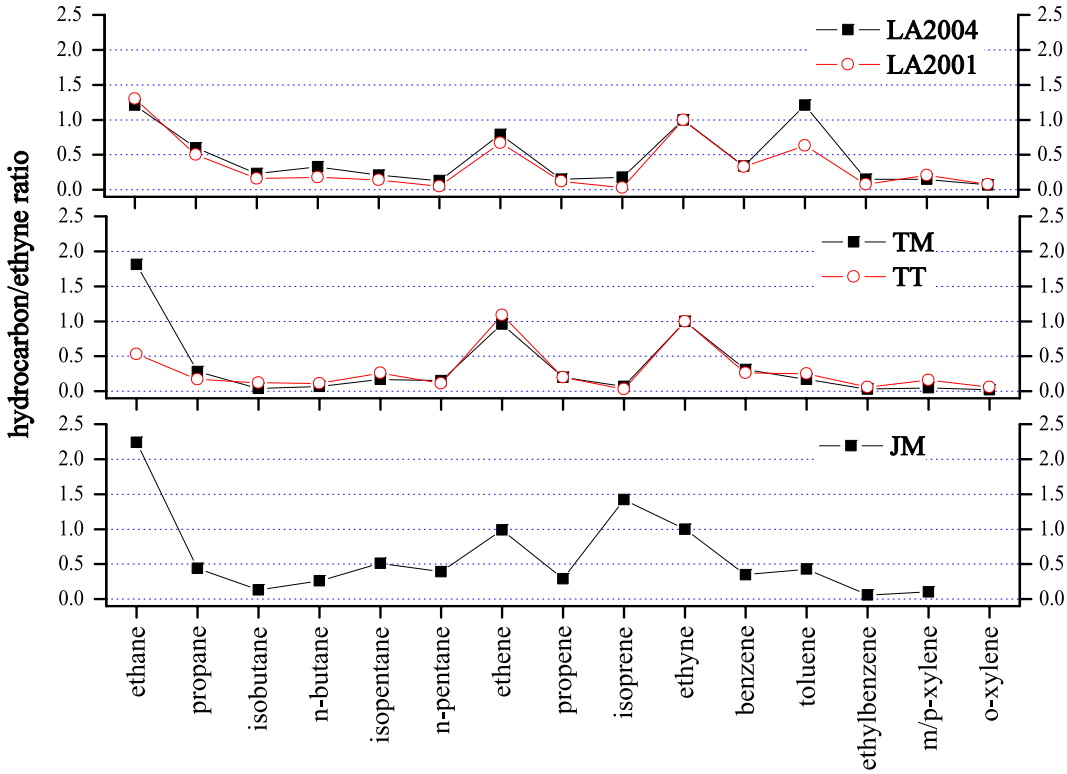

Figure 3. Hydrocarbon/Ethyne ratios at the four sites and compared with data at LA in spring 2001.

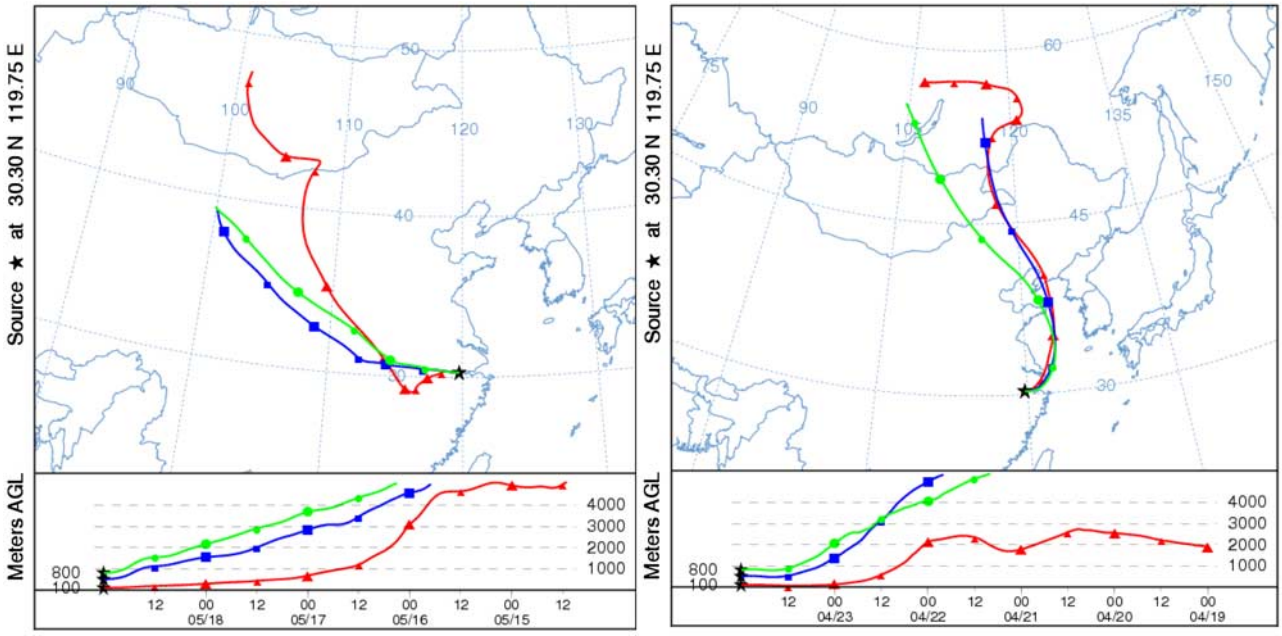

Figure 4. Typical backward air trajectories at LA from (left) west and (right) northeast.

hydrocarbons (such as propane, ethene, ethyne and toulene) emitted from this region were uplifted, and transported to LA.

[25] Toluene is widely used as an industrial solvent and elevated levels of toluene were reported in the atmosphere over the industrial PRD region [*L. Y. Chan et al.*, 2006]. High levels of toluene were observed in LA in both air mass groups (1.66 ± 1.54 ppbv in Type W and 2.78 ± 2.91 ppbv in Type NE). Figure 5 shows the scatterplots of several hydrocarbons versus toluene at LA. Toluene correlated well with ethylbenzene, *m*/*p*-xylene and *o*-xylene, but poorly with *i*-pentane, ethyne, and benzene. It indicates that toluene, ethylbenzene, *m*/*p*-xylene, and *o*-xylene (TEXs) shared common sources, most likely industrial emissions. The poor correlation of toluene with *i*-pentane and ethyne implies that gasoline evaporation and vehicular exhausts contribute less to the high levels of toluene, as *i*-pentane was widely used as a tracer for gasoline evaporation and ethyne for vehicular emissions [*Barletta et al.*, 2005]. The wind rose of toluene at LA demonstrates that air masses from southwestern and south directions carried abundant toluene (up to 16 ppbv) (Figure 6). Lin'an Township is about 10 km away from the sampling site to the south. Hence industrial emission from Lin'an Township was expected to be the major source of TEXs. Similar phenomenon was also reported by *J. Tang et al.* [2007], which stated that 60% of high concentration NMHCs appear in the air masses from SSW to WSW directions. The higher levels of TEXs in air masses Type NE than in Type W indicate that contributions from the YRD also played important roles, besides the local Lin'an downtown emissions.

3.3.2. TM and TT

[26] TM is a site affected more by local emissions than JM and less by industrial emissions than LA, as there are very few industrial enterprises nearby (Table 5). This is demonstrated by the average mixing ratios of ethene and ethyne at TM being nearly twice higher than at JM, but about one order of magnitude lower of toluene mixing ratios at TM than at LA (Table 3). The profile of hydrocarbon/ethyne ratio at TM is very similar to that at TT, except for a higher ethane/ethyne ratio (1.8) than at TT (0.56). TM is a rural site and is more distant to the source region than TT and this resulted in higher ethane/ethyne ratios at TM than at TT. The well matched other hydrocarbon/ethyne ratios indicate that these hydrocarbons shared common or similar sources. In the atmosphere of TT, very high levels of ethene (4.73 ppbv) and ethyne (4.35 ppbv) were observed. Ethene, ethyne, ethane and benzene were among the top five hydrocarbons at TT, and they accounted for about half of total NMHC.

[27] The scatterplots of ethyne with ethane, *i*-pentane, ethene, propene, ethyne and benzene are presented in Figure 7. Guangzhou is one of the most urbanized and motorized megacities in south China, and vehicular and industrial emissions were the major sources of atmospheric NMHCs in this city [*Tang et al.*, 2007a]. The correlations of ethyne with other hydrocarbons in GZ were plotted for comparison (Figure 7). In TT, ethyne correlated well with ethane, ethene, propane, propene, and benzene, but poorly with *i*-pentane. In GZ, ethyne correlated well with ethene, propane, propene, benzene, but poorly with ethane. The slopes of propane, *i*-pentane and ethene to ethyne were also different between TT and GZ, indicating that traffic related emissions (i.e., vehicular emission, gasoline evaporation) were not the major sources of these hydrocarbons in TT. The slope of ethene to ethyne correlations in TT (1.40) was higher than that in GZ (0.88). It indicates that ethene and ethyne have different emission sources from those in GZ, most likely be the local biofuel combustion. The slopes of propane and *i*-pentane to ethyne correlation in TT were lower than those in GZ, indicating they originated from different sources. In GZ, the high levels of propane were leakage from LPG-fueled vehicle emission [*Tang et al.*, 2007a]. While in TT, there were no LPG-fueled vehicles in use. In GZ, traffic related emissions were the major sources of ethyne and *i*-pentane. While in TT, ethyne had other

Table 6. Comparison of Average Mixing Ratios of Selected Species in Different Air Masses Groups at Three Sites (in Parts Per Billion by Volume)

	Lin'an		Tengchong Mountain			Jianfeng Mountain		
Hydrocarbon	Type W (n = 28)	Type W (n = 53)	Type West-April (n = 38)	Type SWC-May (n = 23)	Type-West-May (n = 17)	Type SCS (n = 32)	Type SEA (n = 25)	Type WP (n = 27)
Ethane	2.29 ± 0.47	2.80 ± 0.91	1.43 ± 0.31	1.44 ± 0.44	1.29 ± 0.11	0.78 ± 0.39	1.11 ± 0.28	0.57 ± 0.10
Propane	0.77 ± 0.37	1.59 ± 1.13	0.25 ± 0.07	0.23 ± 0.27	0.19 ± 0.03	0.14 ± 0.10	0.22 ± 0.09	0.09 ± 0.03
i-Butane	0.24 ± 0.20	0.64 ± 0.50	0.03 ± 0.02	0.03 ± 0.02	0.03 ± 0.01	0.06 ± 0.15	0.05 ± 0.03	0.03 ± 0.01
n-Butane	0.36 ± 0.30	0.92 ± 0.88	0.08 ± 0.04	0.05 ± 0.02	0.04 ± 0.01	0.11 ± 0.25	0.11 ± 0.17	0.05 ± 0.03
i-Pentane	0.24 ± 0.11	0.57 ± 0.27	0.11 ± 0.12	0.19 ± 0.10	0.19 ± 0.10	0.09 ± 0.05	0.17 ± 0.10	0.23 ± 0.10
n-Pentane	0.17 ± 0.10	0.36 ± 0.26	0.10 ± 0.03	0.14 ± 0.04	0.14 ± 0.04	0.10 ± 0.02	0.14 ± 0.06	0.15 ± 0.04
Ethene	1.14 ± 0.63	2.01 ± 1.28	0.90 ± 0.44	0.98 ± 0.42	0.65 ± 0.34	0.43 ± 0.38	0.40 ± 0.17	0.25 ± 0.10
Propene	0.24 ± 0.15	0.37 ± 0.30	0.18 ± 0.08	0.20 ± 0.05	0.12 ± 0.04	0.13 ± 0.12	0.11 ± 0.03	0.09 ± 0.02
Isoprene	0.72 ± 0.48	0.22 ± 0.18	0.10 ± 0.26	0.06 ± 0.03	0.09 ± 0.07	0.77 ± 0.65	0.36 ± 0.36	0.53 ± 0.43
Ethyne	1.57 ± 0.60	2.48 ± 0.92	0.84 ± 0.27	0.79 ± 0.36	0.65 ± 0.29	0.34 ± 0.16	0.50 ± 0.22	0.20 ± 0.09
Benzene	0.49 ± 0.18	0.88 ± 0.37	0.25 ± 0.06	0.24 ± 0.09	0.22 ± 0.07	0.11 ± 0.05	0.17 ± 0.06	0.09 ± 0.03
Toluene	1.66 ± 1.54	2.78 ± 2.91	0.15 ± 0.11	0.16 ± 0.08	0.18 ± 0.09	0.11 ± 0.12	0.12 ± 0.04	0.11 ± 0.05
Ethylbenzene	0.23 ± 0.22	0.35 ± 0.22	0.04 ± 0.05	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	0.03 ± 0.02
m/p-Xylene	0.24 ± 0.23	0.34 ± 0.24	0.09 ± 0.14	0.04 ± 0.01	0.03 ± 0.02	0.04 ± 0.02	0.03 ± 0.03	0.03 ± 0.02
o-Xylene	0.11 ± 0.11	0.16 ± 0.15	0.04 ± 0.07	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.01 ± 0.01	0.01 ± 0.01
Total NMHC	11.69 ± 3.47	18.61 ± 8.50	5.06 ± 1.26	5.06 ± 1.35	4.22 ± 1.07	3.60 ± 1.46	3.83 ± 0.76	3.07 ± 1.66

important contributors than traffic related emissions. This resulted in the lower slope of *i*-pentane to ethyne in TT than in GZ.

[28] In the period of 6–10 May 2004, at the same sampling site in Tengchong Mountain, *C. Y. Chan et al.* [2006] reported that enhanced levels of ozone, carbon monoxide (CO) and particulate matters were detected in the air masses from the Southeast Asian subcontinent, where active biomass burnings occurred. However we did not collect canister samples in these days because of the limitation of resources. The impacts of biomass burning in the neighboring Southeast Asian subcontinent on TM were investigated with the help of backward trajectory analysis and satellite images of fire counts.

[29] For the air masses arrived at TM during the study period, two types of trajectories were observed. In April, the major pathways were originated from the Bay of Bengal and traveled across Burma (Type West-April). While in May, there were two types of pathways: one from southwestern China in the period of 18–21 May (Type SWC-May) and the other from Burma (Type West-May) in the period of 22–24 May, the same route as Type West-April. Figure 8 shows typical pathways of the two groups. Table 6 compares mixing ratios of selected hydrocarbons in different air mass groups. Although the routes of air masses differed remarkably between Type West-April and Type SWC-May, the mixing ratios of most hydrocarbons were quite close. While the routes of air masses between Type West-April and Type West-May were similar, the mixing ratios of hydrocarbon were much lower in air mass Type West-May. Fire count map (http://maps.geog.umd.edu/products.asp) was used to identify the biomass burning activities in Southeast Asian region. Figure 9 shows the fire count maps in April and May during the sampling periods. In April, intensive biomass burning activities were detected at the Southeast Asian subcontinent. While in May, only a few fire points were detected. In the routes of air masses in April, there were some biomass burning activities. The air pollutants from the biomass burning emissions were uplifted and transported to TM, resulting in the high NMHC levels in Type West-April. The NMHC levels were nearly identical between air masses Type West-April and Type SWC-May. It indicates that emissions from SE Asian biomass burning and from TM local biofuel combustion contributed equally to the local NMHC levels.

[30] High levels of ethene and ethyne were observed both in the air masses from the Southeast Asian subcontinents and from southwestern China. The major sources of ethyne in the atmosphere are incomplete combustion emissions, such as biomass burning and fossil fuel combustion [*Choi et al.*, 2003; *Lee et al.*, 2006]. In urban areas, its major source was vehicular emission and ethyne was commonly used as a tracer for vehicular emissions [*Henry et al.*, 1994; *Barletta et al.*, 2005]. While in the rural regions of China, biofuel combustion was its major source. It was estimated that 50% of ethane and ethene in China were from biofuel combustion emission [*Phadnis and Carmichael*, 2000]. In another study, 45% of ethyne and ethane, and 50% of ethene were attributed to biofuel combustion in East Asia [*Streets et al.*, 2003]. Ethene, ethane and ethyne were the three dominant hydrocarbons in the plume of biomass/biofuel combustion [*Tsai et al.*, 2003; *Woo et al.*, 2003]. Ethene and ethane

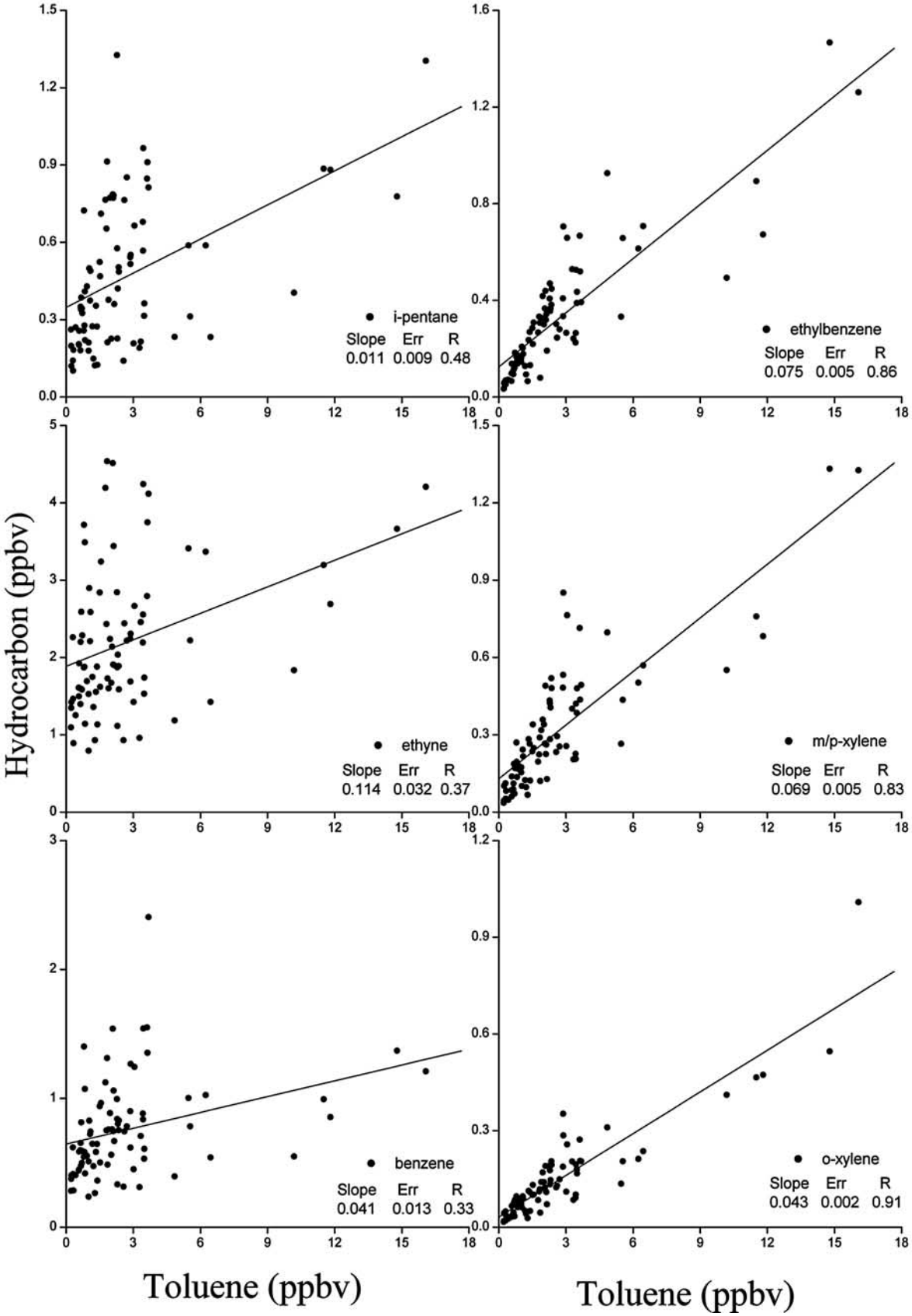

Figure 5. Scatterplots of several hydrocarbons versus toluene at LA.

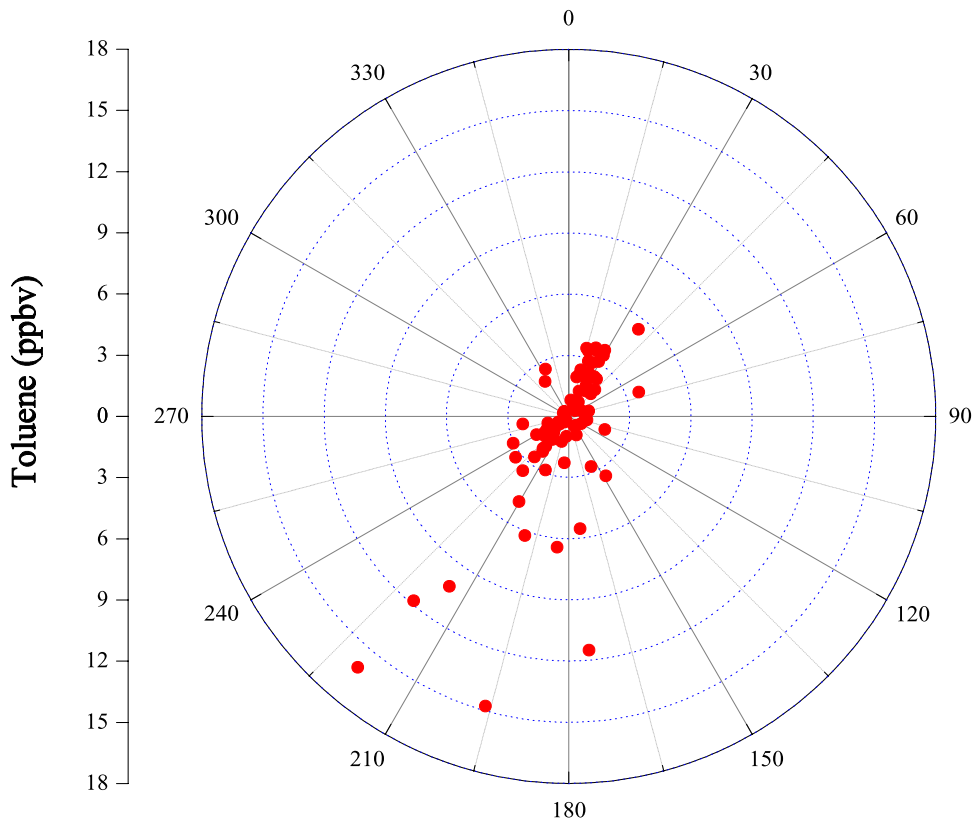

Figure 6. Wind rose of toluene at LA.

accounted for about 35% and 40% of the total NMHC in the atmosphere of TM and TT, respectively. In China, it has been reported that biomass accounted for about two thirds of the total energy used by rural households [*Jiang and O'Neill*, 2004]. As a less developed province in China, the major energy sources in rural households are firewood (41%), coal (38%) and straw (12%) in Yunnan province [*Li et al.*, 2005]. Hence biofuel combustions by local peasant households in TM region play very important roles on the high levels of ethene and ethyne. There is only one paved road from TT to TM, and few vehicles ran daily on this mountainous and remote region. It is reasonable to say that vehicular emission is not an important contributor to the levels of NMHCs. This was also confirmed by the poor correlation of ethene and ethyne with *i*-pentane (R^2 = 0.14 for both species, not shown).

3.3.3. JM

[31] In the previous study by *Tang et al.* [2007b], the major air masses at JM in April and May 2004 were categorized into three types of trajectories: originated from South China Sea (Type SCS); originated from Indian Ocean or South China Sea and passed over Southeast Asia (Type SEA); originated from Western Pacific and passed over Philippines (Type WP). In this study, the entire data set is presented and compared with the other two background sites (Table 6). The highest levels of most anthropogenic hydrocarbons were observed in Type SEA and the lowest in Type WP. Influenced by the nearby Southeast Asia, levels of most anthropogenic hydrocarbon in air masses from the South China Sea were also higher than in air masses from Western Pacific. Hence the major source of anthropogenic hydrocarbons at this remote site was long-range transport from distant source regions, such as Southeast Asian subcontinent.

3.3.4. Biogenic Emissions

[32] Isoprene and α-pinene were identified and quantified in this study. The two species are major components of biogenic emission in the atmosphere [*Guenther et al.*, 1995]. Compared with isoprene, the levels of α-pinene were very low. Among the three background sites, JM shows the highest isoprene levels, while TM shows the lowest, and LA shows the highest α-pinene levels. Figure 10 shows the diurnal variations of isoprene at the three background sites in May 2004. The higher levels of isoprene at midday than in the morning and evening indicate that that there are strong local sources at JM and LA. Emissions from local vegetation were expected to be the major sources at JM and LA. While at TM, the levels of isoprene were much lower compared with the other two sites. It indicates that there were very limited isoprene-emitting vegetations. An plausible explanation for the low isoprene levels in the mountainous TM region is that, the altitude of the sampling site is nearly 2000 m asl, and the vegetation at this altitude are shrubs and thus emitted less isoprene than at the other two low-altitude sites [*Klinger et al.*, 2002].

3.3.5. Characteristic Ratios in Air Masses From Different Regions

[33] Characteristic ratios of hydrocarbon concentrations were useful tools to differentiate air masses from various sources [*Barletta et al.*, 2005; *Jobson et al.*, 2004; *Tang et al.*, 2007b]. In 43 Chinese cities, the benzene/toluene (B/T)

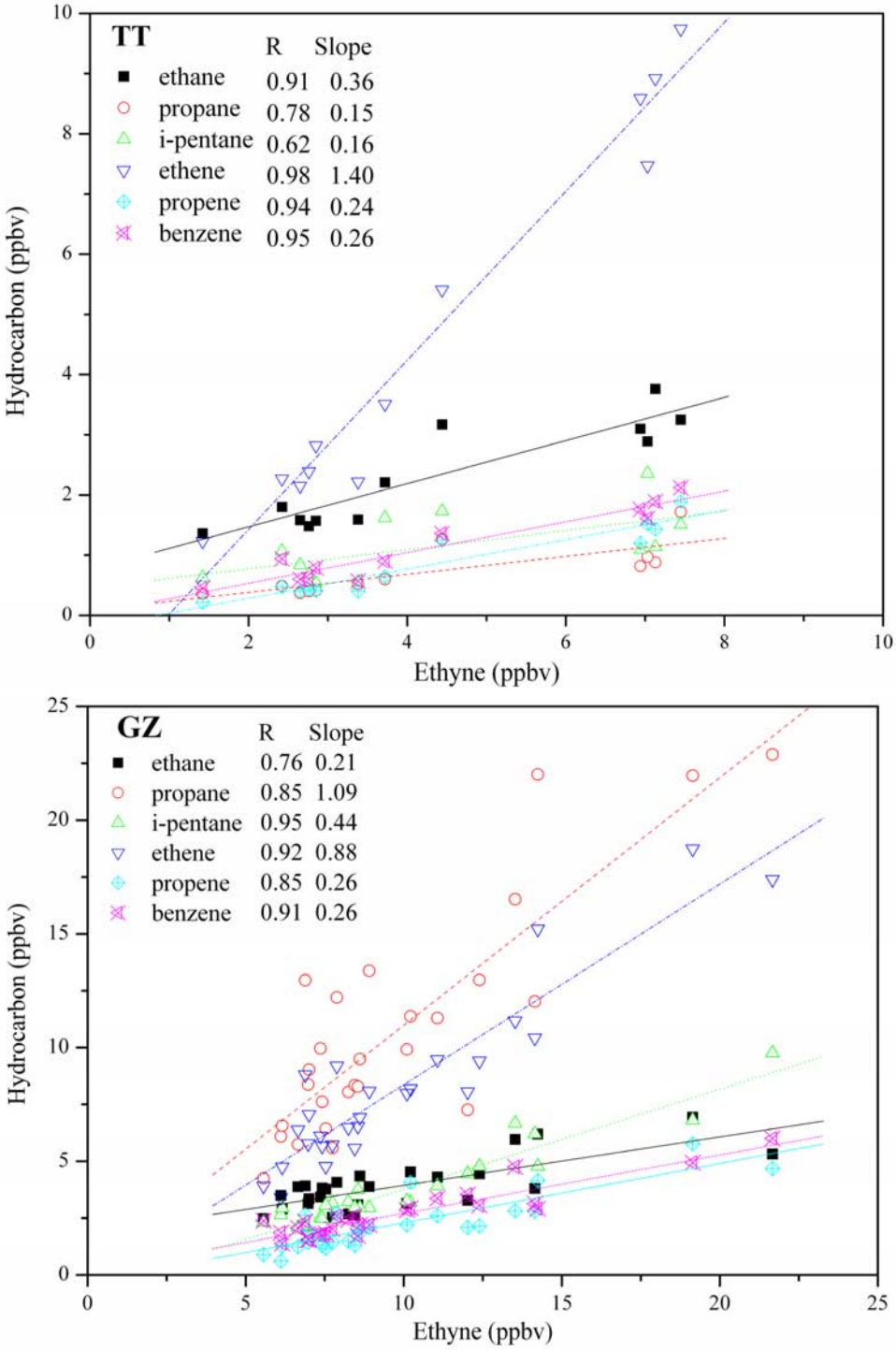

Figure 7. Scatterplots of ethyne with other anthropogenic hydrocarbons at TT and compared with those in Guangzhou.

ratio was ranged from 0.2 to 2.2 and a ratio of 0.7 ppbv/ppbv (0.6 wt/wt) was used to differentiate traffic-related emissions and other sources [*Barletta et al.*, 2005]. In the tunnel and vehicular exhaust samples in Houston, average B/T ratios were found to be 0.50 and 0.62, respectively [*Jobson et al.*, 2004]. In the roadside samples of GZ in 2005, the average B/T ratio was 0.7 [*Tang et al.*, 2008]. In this study, several hydrocarbon molar ratios in different air masses at the four sampling sites were calculated to explore the possible source signatures and the ratios in GZ were also presented for comparison (Table 7). The average B/T ratios were 1.25 and 0.30 in TT and GZ, respectively. In TT, the high emission of benzene relative to toluene (B/T >1) indicates that the main source of benzene is not likely vehicular emission, but most likely biofuel combustion. The low B/T ratio (0.30) in GZ was due to the additional

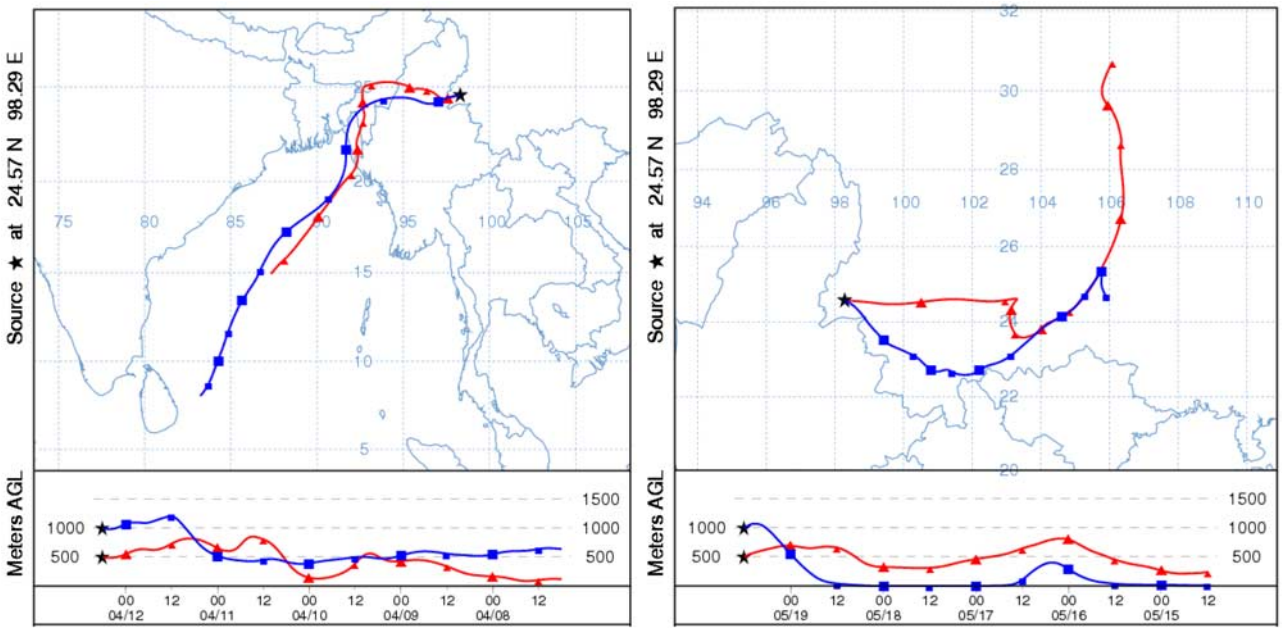

Figure 8. Typical backward air trajectories at TM from (left) east and (right) west.

emission source of toluene from industrial activities, besides the vehicular emissions [*Tang et al.*, 2007]. In TM, the B/T ratios in the air masses Type West-April (2.04 ± 0.84) were higher than in Type SCS-May (1.65 ± 0.64) and Type West-May (1.36 ± 0.51). This was attributed to the biomass burning emissions in Burma in April.

[34] The photochemical lifetimes of *i*-butane and *n*-butane are nearly equal in the atmosphere, and their concentration ratios should be constant after emitted into atmosphere, although the ratios varied greatly among different emission sources. *Parrish et al.* [1998] compared various ambient NMHC data sets in USA and found that *i*/*n*-butane ratios were relatively constant (0.48 ± 0.02) in those studies. A mean ratio of 0.45 ± 0.06 was also observed at six sites of United Kingdom [*Derwent et al.*, 2000]. In this study, *i*/*n*-butane ratios in TT were the highest (1.07 ± 0.21) among the four sites, close to the median one (1.03) in ambient air of La Porte, an outskirt site in Houston, USA [*Jobson et al.*,

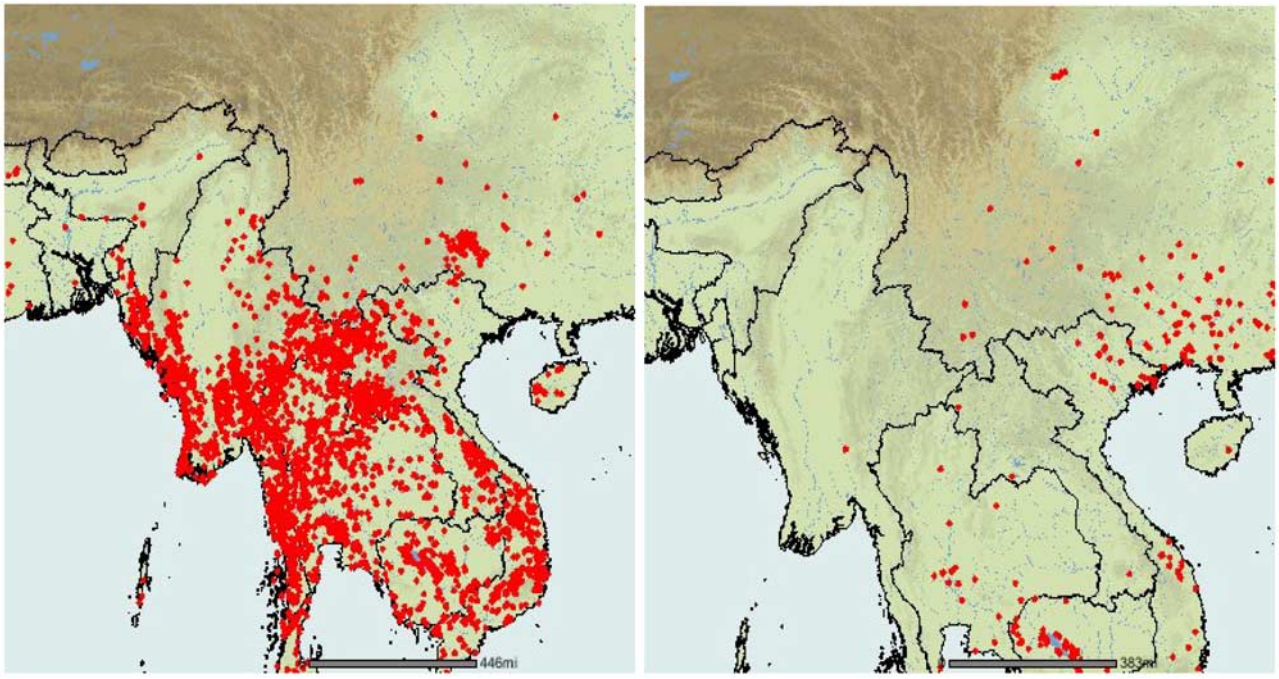

Figure 9. Fire count maps during (left) 8–18 April and (right) 18–25 May.

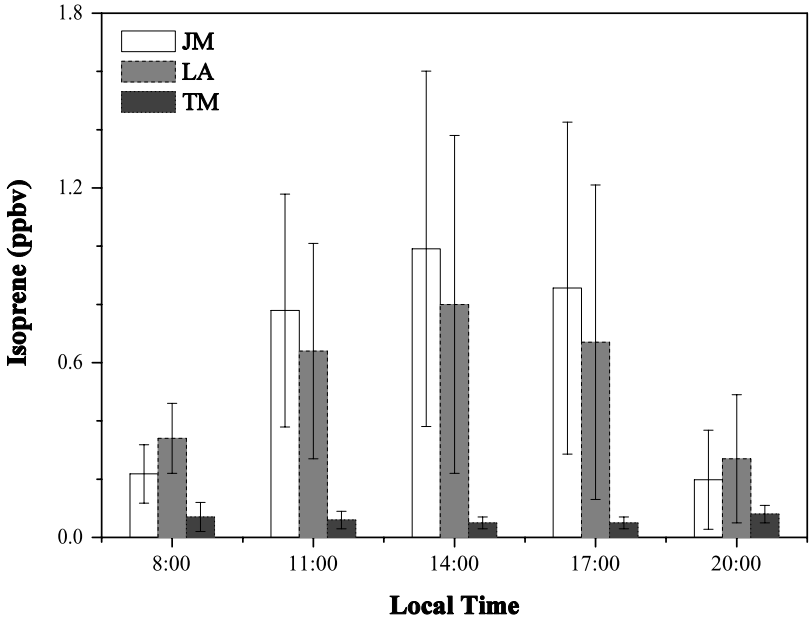

Figure 10. Diurnal variations of isoprene at the three sites.

2004]. In that study, the high *i*-butane to *n*-butane ratio was attributed to the emission from natural gas feed stocks for the petrochemical industry nearby. In TT, there was no petrochemical industry and natural gas was not a popular fuel for energy. Thus natural gas contributed less to the high ratio. Biofuel combustion and biomass burning also might not be the main contributor to the high ratio, as the ratio in TM was 0.57 ± 0.26. The possible source might be the coal combustion in TT. In the air masses (Type West-April) from Southeast Asia where biomass burning occurred, the ratio was 0.46 ± 0.21, a little smaller than those without biomass burning activities (0.65 ± 0.25 and 0.72 ± 0.25 in air masses Type SWC-May and Type West-May, respectively). In JM, the ratios were around 0.5. While in LA, the ratios were around 0.7, close to those in the urban of GZ (0.71 ± 0.06), indicating a contribution from nearby urban/industrial emissions.

[35] Ethane to propane ratio was also widely used to investigate the emission characteristics of various source regions and emission source types [*Carmichael et al.*, 2003; *Tang et al.*, 2007a; *Wang et al.*, 2003, 2005]. Using the TRACE-P data set, *Carmichael et al.* [2003] found that biomass burning in Southeast Asia had a value of 8, biofuel combustion 2.3, and transportation 0.5. In this study, the ratios in GZ were around 0.39, close to the transportation type. In Tengyue Town, ethane/propane ratios were 3.55 ± 0.68, close to the calculated values (2.5–3.0) for Yunnan province based on emission inventory [*Carmichael et al.*, 2003]. The ethane to propane ratios were much higher in TM (around 5.8–6.8) than in TT. High ethane to propane ratios were also found at JM (around 5.3–7.4). These high values may characterize the well mixed and aged air masses in remote background sites of China. In LA, the ethane to propane ratios were 2.10 ± 0.70 in air masses from NE China and 3.28 ± 0.82 in air masses from western China. In a previous study at LA, a mean value of 2.7 for ethane/propane ratio was reported by *Wang et al.* [2004]. The lower values in air masses from NE China than in air masses from western China indicates urban/industrial impacts from YRD region (such as Shanghai metropolis).

4. Summaries

[36] The NMHC levels varied greatly among the three background sites. LA shows the highest levels for most hydrocarbons, and JM shows the lowest levels of most hydrocarbons except for isoprene. Ethane, ethene, and ethyne are the most abundant anthropogenic hydrocarbons at the three sites. Compared with other rural or background sites, the levels of anthropogenic hydrocarbon in JM were very low. The source patterns of NMHCs at the three background sites differed significantly. LA was influenced by the YRD regional urban and industrial emissions and local urban emissions. The level of toluene had increased by 75% in 2004 in our study compared with the study in 2001 at LA, possibly because of the increasing industrial activities in the region. Local industrial emissions played an important role on the high levels of toluene, ethylbenzene, and xylenes at LA. Combustions of fossil fuel and biofuel from local peasant households and biomass burning emission from Southeast Asia were the major sources at TM. While at JM, long-range transport was the major source of most anthropogenic hydrocarbons. Local biogenic emissions were the major sources of isoprene and α-pinene at JM and LA. However the biogenic emission strength was negligible at TM.

[37] Hydrocarbon molar ratios were used to differentiate the possible sources of air masses in the four sampling sites. High B/T ratios in air masses originated from Burma in April indicate a contribution of biomass burning emission in TM. The high ratios of ethane to propane (5–8) feature the well mixed and aged air masses in remote background atmosphere in south China. The low ethane/propane ratio at LA in air masses from NE China implies a contribution from urban/industrial emissions.

[38] Among the three rural and background stations, LA is influenced more by nearby urban and industrial emissions,

Table 7. Hydrocarbon Molar Ratios (in Parts Per Billion by Volume/Parts per Billion by Volume) in Air Masses From Different Regions and Compared With Those in Guangzhou, China

Hydrocarbon Ratio	Lin'an		Tengchong Mountain			Jianfeng Mountain			Tengyue Town	Guangzhou
	Type W (n = 28)	Type NE (n = 53)	Type West-April (n = 38)	Type SWC-May (n = 23)	Type West-May (n = 17)	Type SCS (n = 32)	Type SEA (n = 25)	Type WP (n = 27)	(n = 12)	(n = 28)
Ethane/Propane	3.28 ± 0.82	2.10 ± 0.70	5.78 ± 0.98	6.34 ± 1.50	6.85 ± 1.07	7.17 ± 3.08	5.26 ± 0.95	7.42 ± 4.70	3.55 ± 0.68	0.39 ± 0.11
Ethane/Ethyne	1.58 ± 0.46	1.22 ± 0.36	1.78 ± 0.40	2.06 ± 0.76	2.06 ± 0.54	2.45 ± 0.65	2.46 ± 0.63	3.60 ± 1.95	0.57 ± 0.16	0.42 ± 0.10
i-Butane/*n*-Butane	0.71 ± 0.16	0.73 ± 0.09	0.46 ± 0.21	0.65 ± 0.25	0.72 ± 0.25	0.51 ± 0.19	0.51 ± 0.17	0.56 ± 0.25	1.07 ± 0.21	0.71 ± 0.06
i-Pentane/*n*-Pentane	1.48 ± 0.32	1.67 ± 0.38	1.06 ± 1.13	1.31 ± 0.41	1.31 ± 0.39	0.93 ± 0.36	1.11 ± 0.28	1.43 ± 0.47	2.24 ± 0.22	2.22 ± 0.37
Benzene/Toluene	0.65 ± 0.57	0.50 ± 0.31	2.04 ± 0.84	1.65 ± 0.64	1.36 ± 0.51	1.35 ± 0.46	1.70 ± 0.98	0.89 ± 0.36	1.25 ± 0.40	0.30 ± 0.09
Ethylbenzene/Benzene	0.51 ± 0.54	0.41 ± 0.21	0.18 ± 0.31	0.09 ± 0.04	0.08 ± 0.04	0.10 ± 0.13	0.05 ± 0.06	0.09 ± 0.14	0.27 ± 0.17	0.70 ± 0.30
Ethylbenzene/Toluene	0.17 ± 0.14	0.16 ± 0.05	0.21 ± 0.10	0.14 ± 0.06	0.09 ± 0.05	0.10 ± 0.10	0.09 ± 0.10	0.06 ± 0.09	0.31 ± 0.21	0.19 ± 0.05
Propane/Benzene	1.62 ± 0.61	1.79 ± 0.70	1.06 ± 0.25	1.00 ± 0.26	0.96 ± 0.28	1.18 ± 0.67	1.35 ± 0.27	1.15 ± 0.50	0.64 ± 0.15	4.18 ± 1.28
Ethyne/Benzene	3.22 ± 0.40	2.89 ± 0.48	3.40 ± 0.39	3.16 ± 0.37	2.94 ± 0.78	2.94 ± 0.42	2.92 ± 0.44	2.26 ± 0.74	3.99 ± 0.86	3.77 ± 0.65

and hence is not a very suitable site to reflect the regional air quality in a rapidly urbanizing and industrializing region in eastern China. TM is influenced by regional biofuel combustion and biomass burning emissions from Southeast Asia, and is far away from major urban and industrial center. TM is a good background station to feature regional air quality in the less developed southwest China region. The air quality in JM is influenced by long-range transport from distant source regions. JM is a very appropriate station to monitor regional air quality in south China and a long-term monitoring station for air pollutants should be set up to better understand the air quality in this rapidly developing region.

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