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ScienceDirect
Atmospheric Environment 42 (2008) 3780–3794
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Implications of changing urban and rural emissions on non-methane hydrocarbons in the Pearl River Delta region of China

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Received 17 April 2007; received in revised form 23 November 2007; accepted 21 December 2007

Abstract

Guangzhou (GZ) is one of the highly industrialized and economically vibrant cities in China, yet it remains relatively understudied in terms of its air quality, which has become severely degraded. In this study, extensive air sampling campaigns had been conducted at GZ urban sites and in Dinghu Mountain (DM), a rural site, in the Pearl River Delta (PRD) during the spring of 2001 and 2005. Additionally, roadside and tunnel samples were collected in GZ in 2000 and 2005. Later, exhaust samples from liquefied petroleum gas (LPG)- and gasoline-fueled taxis were collected in 2006. All samples were analyzed for C_2–C_{10} non-methane hydrocarbons (NMHCs). NMHC profiles showed significant differences in the exhaust samples between gasoline- and LPG-fueled taxis. Propane (47%) was the dominant hydrocarbon in the exhaust of the LPG-fueled taxis, while ethene (35%) was the dominant one in that of gasoline-fueled taxis. The use of LPG-fueled buses and taxis since 2003 and the leakage from these LPG-fueled vehicles were the major factors for the much higher level of propane in GZ urban area in 2005 compared to 2001. The mixing ratios of toluene, ethylbenzene, *m*/*p*-xylene and *o*-xylene decreased at the GZ and DM sites between 2001 and 2005, especially for toluene in GZ, despite the sharp increase in the number of registered motor vehicles in GZ. This phenomenon was driven in part by the closure of polluting industries as well as the upgrading of the road network in urban GZ and in part by the implementation of more stringent emission standards for polluting industries and motor vehicles in the PRD region.

Keywords: NMHCs; LPG; PRD; Industrial emissions; Guangzhou

1. Introduction

The Pearl River Delta (PRD) region is located on the southern coast of China (Fig. 1) and is one of the highly populated, urbanized and industrialized

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1352-2310/$ - see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2007.12.069

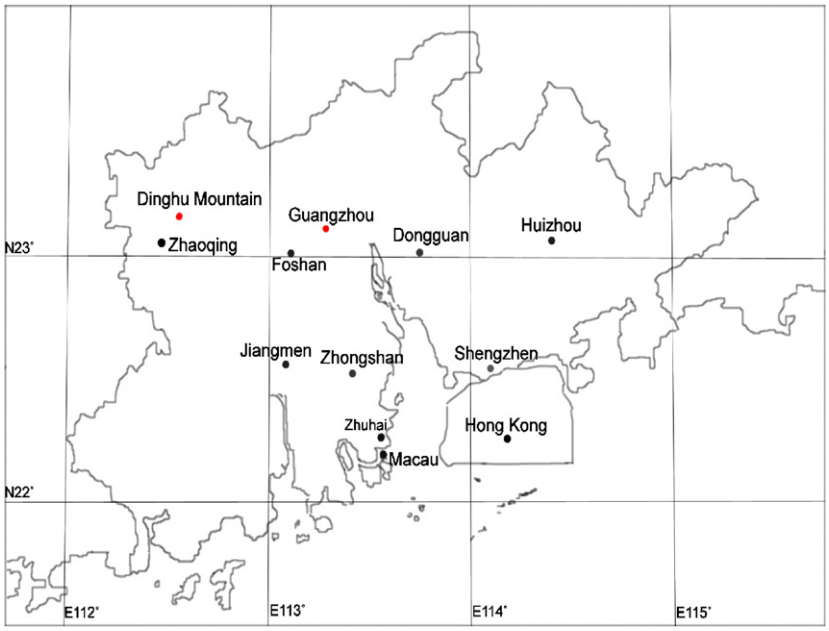

Fig. 1. Map of the Pearl River Delta showing the sampling sites.

regions in China. Several important cities, such as Guangzhou (GZ), Shenzhen, Dongguan and Zhuhai are located there. The PRD accounted for about 70% of the gross domestic product (GDP) and 50% of the population of Guangdong Province in 2000 (Guangdong Statistics Bureau, 2001). Economic achievements in the PRD have been significant since the establishment of the open door policy and economic reforms in 1978. Between 1980 and 2000, the GDP in the PRD increased at an average rate of 17%, whereas the nationwide increase was 10% and that for Guangdong Province was 14% (Guangdong Statistics Bureau, 2001). However, the rapid urbanization and industrialization has resulted in a significant rise in pollutant emissions and therefore a severe degradation of air quality (Wang et al., 2003). Elevated levels of volatile organic compounds (VOCs), especially aromatic hydrocarbons, have been found at roadsides in Hong Kong and the PRD cities (Chan et al., 2002; Wang et al., 2002; Zhao et al., 2004). Ozone episodes, with strongly enhanced concentrations of other air pollutants, are frequently observed in Hong Kong, located downwind from the PRD (Chan and Chan, 2000; Wang et al., 2006).

Non-methane hydrocarbons (NMHCs) are important ozone precursors. Photochemical reactions of NMHCs with the hydroxyl radical (OH) and nitrate radical produce various pollutants such as carbonyls, carbon monoxide, ozone and secondary organic aerosols (Odum et al., 1997; Poisson et al., 2000). In urban areas of the PRD, ozone formation is limited by VOCs, with the abundance of NMHCs (a subset of VOCs) determining the magnitude of ozone formation (Wang X.M. et al., 2005). Previous investigations of NMHCs at a rural site in Hong Kong revealed that vehicular and industrial emissions from the PRD were important sources of NMHCs at that site (Guo et al., 2006; Wang T. et al., 2005). However, few NMHC studies have been undertaken in GZ or other PRD cities (Wang et al., 2004; Chan et al., 2006).

Of particular interest, liquefied petroleum gas (LPG) is widely used in Guangdong Province as a clean energy fuel, and LPG-fueled buses and taxis was promoted by the GZ municipal government, with the intention of improving air quality (Guangzhou Yearbook Committee, 2004). The effect of LPG usage on urban air quality had been studied in many large metropolises (Blake and Rowland, 1995; Chen et al., 2001; Gamas et al., 2000; Schifter et al., 2000). For example, Gamas et al. (2000) found that LPG leakage from domestic usage was a leading contributor to air pollution in Mexico City. However, to date, limited studies on the effect of LPG usage on air quality in China have been conducted. Tsai et al. (2006) reported increased levels of propane, *n*-butane, and *i*-butane in tunnel samples due to the

increasing number of LPG-fueled vehicles in Hong Kong; Zhao and Melaina (2006) stated that simple conversion from gasoline to LPG and compressed natural gas (CNG) does not significantly help in emissions reduction.

This present study investigates characteristics of NMHCs at one rural site (Dinghu Mountain—DM) and two urban sites of a major PRD city (GZ) during the spring of 2001 and 2005. The aim of the study is to determine the effects of economic, industrial and urban changes on the NMHC profiles in rural and urban environments. This study is the first of its kind in the PRD at our knowledge.

2. Methodology

2.1. Sampling

Pre-evacuated 2-L stainless steel electropolished canisters, each equipped with a bellows valve, were provided by the University of California, Irvine (UCI) for sampling. Between 3 and 19 March 2001, and 16 and 21 April 2005, 39 and 30 rural whole air samples were, respectively, collected at a forest site on a hill top in Dinghushan National Natural Reserve, DM (~85 km west of GZ and 18 km northeast of Zhaoqing; 23.10°N, 112.32°E; Fig. 1). This mountain site is a station of the "Man and the Biosphere Programme", United Nations Educational, Scientific and Cultural Organization (UNESCO). It is surrounded by counties that are less developed than elsewhere in the PRD. Nearby there is a small town, Guicheng, with a population of 20,000, and a highway about 3 km to the south. Between 3 and 26 March 2001, 43 urban air samples were collected from the rooftop of a nine-story building located in the Liwan district in GZ. Liwan is an old city center and the sampling site was surrounded by traffic-congested roads, residential buildings, restaurants and schools. In 2005, the Liwan site was not available during the sampling periods. Another site—Dongshan was selected. The two sites were both surrounded by congested traffic and residential buildings. The selected sampling buildings at both sites were higher than their nearby buildings, thus made it possible to collect well mixed and representative air samples. The two sites were about 5 km away from each other. Between 16 and 21 April 2005, 30 urban air samples were collected in the Dongshan District of GZ. The sampling site was located on the rooftop of a 21-story building.

Nineteen samples were collected within a 3-day period at both sites in 2001 to study the diurnal variations of the NMHCs. The remaining samples were collected at noon (mostly at 12:00 in GZ and 13:00 in DM, both in local time). On average, the 2001 sampling canisters were filled in approximately 2 min. All ambient samples in 2005 were collected between 08:00 and 21:00 local time at intervals of 3 h. A flow-controlling device was used to collect 1-h integrated samples. The rural mountaintop and urban rooftop sampling sites were chosen to help collect well mixed and representative air samples.

Roadside and tunnel samples were also collected in GZ in 2000 and 2005 to get a snapshot of NMHC profiles from vehicular emissions. On 6 February 2000, two samples were collected at a roundabout on Xingang Road, Haizhu District, one at 09:00 and the other at 10:00, and two additional samples were collected from the Zhujiang (Pearl River) Tunnel at 10:30 and 15:30 on the same day. Xingang Road has very heavy traffic and the Zhujiang tunnel links the two urban areas of Liwan and Fangcun districts. On 13 August 2000, one sample was collected at a roundabout of Xingang Road at 09:30, and another was collected at a roundabout of Tianhe Road, a traffic-congested road, at noon. Six samples were also collected on 8 September 2005, between 07:00 and 20:00 at the Xingang roadside. All the roadside and tunnel samples were 1-min integrated samples.

Two exhaust samples were collected directly next to the exhaust pipes of LPG-fueled taxis and three exhaust samples from gasoline-fueled taxis at idling state. Each canister was used to collect exhaust from three taxis. A pressure gauge with a flow controller was used to regulate the sampling volume from each taxi. All the taxis selected were newly registered cars (around 1 year old).

2.2. Chemical analysis

All the roadside, tunnel and exhaust samples were analyzed at the University of California, Irvine (UCI), as were the 2001 rural and urban samples. The 2005 rural and urban samples were analyzed by the Research Center for Environmental Changes (RCEC), Academia Sinica, Taiwan. Details of the analytical procedures employed in this study can be found in Colman et al. (2001) and Doezema (2004) for UCI and in Chang et al. (2003) for RCEC. The UCI samples were analyzed for 45 C_2–C_{10} NMHCs. For the RCEC samples, the detection limit for the 54 C_2–C_{10} hydrocarbons is 15 pptv, and the

measurement precision is 5%. For the UCI samples, the detection limit for the C_2–C_{10} hydrocarbons is 5 pptv, and the measurement precision varies by compounds and by mixing ratio. For example, the precision is 1% (or 1.5 pptv, whichever is larger) for alkanes and alkynes, and 3% (or 3 pptv, whichever is larger) for alkenes and aromatics.

To test the consistency of data from the two laboratories, an interlab comparison experiment was carried out. Eight canisters were filled with identical content of NMHC gas mixtures (an expired commercial NMHC standard mixture). Four were sent to UCI and the other four to RCEC for chemical analysis. The results are discussed below.

3. Results and discussion

3.1. NMHCs in GZ and DM in 2001

Table 1 summarizes the average mixing ratios and ranges of NMHCs in GZ and DM in 2001 together with the Spearman's correlation coefficients (*R*s) of NMHCs with ethyne and *i*-pentane. Most hydrocarbons showed higher levels in the GZ urban site than in the DM rural site, especially toluene (about four times higher in GZ than in DM), ethene and ethyne (about three times higher in GZ than in DM). The total mixing ratio of hydrocarbons measured was approximately four times higher in GZ than in DM. This coincided with the fact that there were more NMHC emission sources in the urban site than in the rural site. Toluene was the most abundant hydrocarbon both in GZ (20% of total NMHC) and DM (15% of total NMHC), and ethyne, ethene, propane, ethane and *n*-butane were the next most abundant hydrocarbons in GZ and DM. Alkanes were the dominant hydrocarbon groups, and accounted 40% of total NMHC in DM and 35% in GZ. The major sources of alkanes in urban area included vehicular emissions, gasoline evaporation, LPG and natural gas (NG) leakage (Blake et al., 1995). Aromatic hydrocarbons accounted for about one-third of total NMHC both in GZ and DM. The major sources of aromatic hydrocarbons included gasoline evaporation, solvent evaporation and diffuse emissions and vehicular emissions (Heeb et al., 2000).

The mixing ratios of most hydrocarbons in GZ fell in the ranges of those measured in other 43 Chinese cities except for toluene (Barletta et al., 2005). The average mixing ratio of toluene in GZ (14.1 ppbv) exceeded the maximum mixing ratio of toluene (11.2 ppbv) in the other 43 Chinese cites. High levels of toluene were also found in PRD cities in previous studies, like Hong Kong (37.0 ppbv, Lee et al., 2002), GZ (24.4 ppbv, Wang et al., 2002) and Macao (28.4 ppbv, Wang et al., 2002) and in a northeast Chinese city, Changchun (21.3 ppbv, Liu et al., 2000). The elevated toluene levels were attributed to the industrial activities and vehicular emissions (Lee et al., 2002, Chan et al., 2006). The high levels of ethyne and ethene in GZ indicates that there existed substantial combustion sources, in particular vehicular exhaust. The levels of toluene (14.1 ppbv) were also higher in GZ than in Beijing, the capital city of China (3.03 ppbv in Beijing, Song et al., 2007). Other hydrocarbons also showed higher levels in GZ than in Beijing, except for *i*-pentane (4.11 ppbv in Beijing) and *n*-pentane (1.7 ppbv in Beijing). The major sources of VOCs in Beijing were gasoline-related emission (52%), petrochemicals (20%) and LPG leakage (11%) (Song et al., 2007).

The levels of most hydrocarbons in the rural DM site were higher than most rural sites in China, such as Jianfeng Mountain in Hainan Island (Tang et al., 2007a), Lin'an in the Yangtze River Delta (Guo et al., 2004), Hok Tsui (Wang et al., 2003) in Hong Kong, but lower than Tai O in Hong Kong, when air masses passed through the PRD urban region (Wang T. et al., 2005). The levels of toluene (6.01 ppbv) and propane (2.17 ppbv) in Tai O were much higher than those in DM (3.36 and 1.59 ppbv, respectively). In another study carried out in summer 2000 in industrial, industrial-urban and industrial-suburban atmospheres of the PRD, the levels of toluene were 13.5 ± 11.8, 11.5 ± 11.6 and 7.3 ± 5.4 ppbv and the levels of propane were 3.7 ± 2.5, 2.5 ± 1.4 and 2.1 ± 1.7 ppbv, respectively (Chan et al., 2006). The high levels of toluene and propane in DM and other PRD sites were due to the widespread industrial activities in the PRD region.

Ethyne was widely used as a tracer for combustion related sources, and *i*-pentane as a tracer for gasoline evaporation sources (Barletta et al., 2005). Most hydrocarbons highly correlated with ethyne and *i*-pentane, indicating that vehicular exhaust and gasoline evaporation would be the major sources of these hydrocarbons at both sites.

3.2. Variation of the NMHC profiles between 2001 and 2005

The NMHC data collected in 2005 in GZ and DM were published in another manuscript by Tang

Table 1
Average mixing ratios and ranges of NMHCs (ppbv) and Spearman's correlation coefficients (Rs) of NMHCs with ethyne and *i*-pentane in GZ and DM 2001

Hydrocarbon	DM 2001 ($n = 39$[a])				GZ 2001 ($n = 43$)			
	Mixing ratio		Correlation		Mixing ratio		Correlation	
	Mean	Range	Ethyne	*i*-Pentane	Mean	Range	Ethyne	*i*-Pentane
Ethane	3.18	1.61–7.28	0.58	0.43	4.06	1.70–8.69	0.38	0.24
Ethene	2.18	0.32–6.54	0.92	0.90	7.37	2.73–18.85	0.90	0.83
Ethyne	2.86	0.75–7.91	1	0.90	8.83	3.49–25.23	1	0.94
Propane	1.59	0.41–3.68	0.83	0.90	5.39	2.09–13.71	0.92	0.87
Propene	0.40	0.08–2.00	0.79	0.85	1.79	0.47–5.16	0.77	0.71
i-Butane	0.58	0.12–1.62	0.83	0.94	3.11	0.77–8.22	0.93	0.91
n-Butane	0.88	0.14–2.68	0.87	0.96	4.60	1.30–13.04	0.92	0.90
1-Butene	0.11	0.02–0.39	0.81	0.87	0.60	BDL–1.74	0.69	0.67
i-Butene	0.14	0.03–0.39	0.60	0.57	0.85	0.19–2.26	0.47	0.47
Trans-2-butene	0.03	BDL[b]–0.12	0.54	0.67	0.54	0.13–2.06	0.77	0.79
Cis-2-butene	0.03	BDL–0.11	0.66	0.78	0.43	0.14–1.59	0.83	0.84
i-Pentane	0.50	0.07–1.69	0.90	1	3.00	0.54–9.05	0.94	1
n-Pentane	0.27	0.03–0.91	0.90	0.97	0.99	0.24–3.13	0.95	0.97
1,3-Butadiene	0.04	BDL–0.18	0.62	0.70	0.30	0.07–1.02	0.86	0.86
Propyne	0.02	BDL–0.06	0.76	0.71	0.10	BDL–0.33	0.91	0.90
1-Pentene	0.46	BDL–1.35	−0.08	−0.01	0.33	0.08–1.09	0.21	0.24
Isoprene	0.41	0.05–1.40	−0.33	−0.33	0.26	BDL–0.74	0.75	0.74
Trans-2-pentene	0.03	BDL–0.14	0.52	0.59	0.42	0.11–1.48	0.86	0.90
Cis-2-pentene	0.02	BDL–0.09	0.34	0.30	0.45	0.08–1.78	0.75	0.78
2,2-Dimethylbutane	0.02	BDL–0.06	0.82	0.83	0.10	BDL–0.31	0.93	0.92
Cyclopentane	0.04	0.01–0.14	0.67	0.73	0.15	BDL–0.43	0.92	0.95
2,3-Dimethylbutane	0.03	BDL–0.11	0.84	0.92	0.28	BDL–1.92	0.89	0.91
2-Methylpentane	0.26	0.01–1.84	0.88	0.93	1.28	0.20–3.80	0.92	0.90
3-Methylpentane	0.25	0.07–0.75	0.89	0.95	1.02	0.28–2.88	0.91	0.91
n-Hexane	0.19	0.03–0.75	0.94	0.93	1.13	0.31–4.27	0.81	0.74
Methylcyclopentane	0.08	0.01–0.30	0.93	0.97	0.54	0.10–1.59	0.94	0.95
Cyclohexane	0.08	BDL–0.25	0.67	0.65	0.31	0.07–2.02	0.77	0.72
2,2,4-Trimethylpentane	0.07	0.01–0.30	0.31	0.36	0.24	0.06–2.26	0.44	0.40
n-Heptane	0.42	0.04–1.13	0.41	0.44	1.22	0.34–4.80	0.74	0.72
2,3,4-Trimethylpentane	0.05	BDL–0.72	−0.07	−0.06	0.05	0.01–0.33	0.26	0.27
n-Octane	0.04	BDL–0.11	0.84	0.93	0.21	0.07–0.58	0.92	0.90
Benzene	0.85	0.17–2.66	0.94	0.90	2.80	0.86–6.51	0.92	0.92
Toluene	3.36	0.40–13.39	0.74	0.82	14.09	2.37–45.32	0.87	0.81
Ethylbenzene	0.56	0.04–2.62	0.85	0.96	2.21	0.26–7.59	0.88	0.85
m-Xylene	0.71	0.05–2.02	0.93	0.93	3.47	0.41–15.49	0.92	0.92
p-Xylene	0.52	0.03–1.48	0.84	0.87	1.69	0.04–8.23	0.80	0.82
o-Xylene	0.64	0.04–7.77	0.80	0.92	2.63	0.34–10.49	0.91	0.90
n-Nonane	0.07	0.01–0.74	0.67	0.77	0.22	0.06–0.66	0.84	0.85
Isopropylbenzene	0.04	BDL–0.40	0.37	0.40	0.06	0.01–0.16	0.79	0.84
Propylbenzene	0.04	BDL–0.42	0.69	0.75	0.09	0.02–0.26	0.88	0.91
3-Ethlytoluene	0.10	0.01–1.40	0.52	0.69	0.31	0.07–1.01	0.91	0.93
4-Ethyltoluene	0.09	0.01–0.84	0.54	0.59	0.17	0.04–0.52	0.88	0.92
1,3,5-Trimethylbenzene	0.11	0.01–2.01	0.51	0.54	0.22	0.06–0.75	0.88	0.90
2-Ethyltoluene	0.03	BDL–0.54	0.63	0.58	0.09	0.01–0.34	0.90	0.92
1,2,4-Trimethylbenzene	0.21	0.02–3.84	0.58	0.73	0.52	0.11–1.63	0.90	0.91
Total NMHC	21.69	7.66–56.68			77.69	27.38–203.00		

[a] n = number of samples.
[b] BDL = below detection limit.

et al. (2007b). As hydrocarbons showed large diurnal variations, here we only present the data collected between 11:00 and 12:00 in 2005 (six samples in each site) with those in 12:00 in 2001 (22 samples in GZ and five samples in DM). There are many factors that would influence the levels of NMHCs in different environments and in different time spans. Hence, it is more difficult to provide exact reasons for the variations of hydrocarbon concentrations using such limited data, especially to those from two different urban sites in GZ. Some of the discussions in this study are thus presumptions and yet it is to the best of our understanding of this region, and relevant references are used whenever applicable. The comparison of NMHC profiles in Liwan (GZ) in 2001 with those in Dongshan (GZ) in 2005 only provides a clue to the change of emission sources and emission levels in this region. Nevertheless, the trend in NMHC profile change has been established. In this paper, we also address the factors causing the rapid emission changes and their effects on the NMHC profiles.

The interlab comparison results are presented in Table 2. Both laboratories show good analytical precisions (very low value of relative standard deviation (RSD), Table 2) for most hydrocarbons, but large differences were observed for several hydrocarbons with mixing ratios lower than 0.5 ppbv. The average absolute difference between mean mixing ratios obtained by the two laboratories was 16%. So, the two datasets were comparable. The sampling periods took place about a month earlier in 2001 compared to 2005 (Section 2), which raises the question of whether the difference in wind directions could have had an effect on the NMHC levels. However, local wind roses show that the prevailing wind directions were from the southeast in GZ and from the northeast in Zhaoqing in both sampling periods (Fig. 2).

The statistic of some hydrocarbons with mixing ratio higher than 0.5 ppbv in GZ is listed in Table 3. Fig. 3 compares the percentage distribution of these hydrocarbons at the two sites for 2001 and 2005, which was calculated by dividing the average mixing ratio of each hydrocarbon by the average mixing ratio of total NMHC. It can be seen from Fig. 3 and Table 3 that there are both similarities and differences in NMHC levels and distributions between the urban and rural sites. Toluene, ethyne, ethene, ethane and propane were the dominant hydrocarbons. The mixing ratios of toluene, ethylbenzene, *m*/*p*-xylene and *o*-xylene decreased at both sites between 2001 and 2005, especially toluene in GZ. The mixing ratios of propane increased at GZ, but showed nearly identical mixing ratios at DM between 2001 and 2005. The mixing ratios of total NMHC also decreased slightly at both sites between 2001 and 2005.

Fig. 3 also shows the percentage distributions of hydrocarbon from gasoline- and LPG-fueled taxis. Propane (47%), ethene (18%) and *n*-butane (11%) are the most abundant hydrocarbons in the exhaust of LPG-fueled taxi, while ethene (35%), ethyne (14%) and ethane (9%) are the dominant hydrocarbons in the exhaust of gasoline-fueled taxis. *i*-Pentane was used as a tracer for gasoline evaporation, and was the major component in the gasoline used in PRD (Tsai et al., 2006). In this study, *i*-pentane is also a major component in the exhaust of gasoline-fueled taxi (8%). However, its percentages in exhaust of LPG-fueled taxis are negligible.

3.3. Impacts of road network development and vehicle emission control on NMHC levels

Fig. 4 shows the average daily usage of LPG and gasoline in Guangdong Province and the number of motor vehicles in GZ and Guangdong Province between 1998 and 2004 (Guangdong Statistics Bureau, 1999–2005; Guangzhou Municipal Statistics Bureau, 1999–2005). With the rapid urbanization and economic development in recent years, the number of motor vehicles had soared both in GZ and throughout Guangdong Province, by about 125% between 1998 and 2004 (Guangdong Statistics Bureau, 1999–2005). Similarly, the motor vehicle fleet has increased by about 80% in Zhaoqing, from 29,000 in 2000 to 52,000 in 2004 (Zhaoqing Municipal Statistics Bureau, 2005). However, the increase of NMHC emissions from vehicular exhaust (indicated by the levels of ethene, propene, ethyne and benzene) did not increase as sharp as the vehicle numbers and the consumed fossil fuels (Table 3). The reasons for these observations are discussed below.

The implementation of the EURO-I emission standard for motor vehicles began in GZ in October 2000, with EURO-II following in 2005. Vehicles registered after 2000 were required to be equipped with electric injection technology and three-way catalytic converters, significantly reducing hydrocarbon emissions when older vehicles were replaced by new ones. In addition, the road conditions in

Table 2
Results of the NMHC inter-comparison for the RCEC and UCI laboratories

Hydrocarbon	RCEC			UCI			Difference (%)[a]
	Mean (ppbv)	S.D.	RSD (%)	Mean (ppbv)	S.D.	RSD (%)	
Ethane	5.642	0.048	0.8	5.384	0.016	0.3	−4.8
Propane	3.438	0.033	1.0	3.124	0.013	0.4	−10.0
Isobutene	0.837	0.021	2.5	0.810	0.006	0.7	−3.4
n-Butane	3.170	0.027	0.9	2.529	0.007	0.3	−25.4
Isopentane	3.699	0.022	0.6	4.364	0.008	0.2	15.2
n-Pentane	1.584	0.027	1.7	1.708	0.005	0.3	7.3
2,2-Dimthylbutane	0.204	0.006	3.0	0.269	0.005	1.9	24.2
Cyclopentane	0.185	0.005	2.6	0.166	0.003	1.7	−11.3
2-Methylpentane	1.353	0.031	2.3	1.702	0.020	1.2	20.5
3-Methylpentane	0.917	0.018	1.9	0.999	0.006	0.6	8.2
n-Hexane	0.860	0.026	3.1	1.089	0.006	0.5	21.0
Cyclohexane	0.363	0.017	4.7	0.416	0.003	0.8	12.8
2-Methylhexane	0.602	0.013	2.2	0.742	0.005	0.6	18.9
2,3-Dimethylpentane	0.480	0.008	1.6	0.537	0.003	0.6	10.7
3-Methylhexane	0.586	0.010	1.6	0.747	0.017	2.2	21.5
2,2,4-Trimethylpentane	0.591	0.007	1.1	0.816	0.007	0.9	27.5
n-Heptane	0.316	0.007	2.2	0.355	0.005	1.5	11.1
Methylcyclohexane	0.255	0.004	1.5	0.187	0.003	1.8	−36.3
2,3,4-Trimethylpentane	0.221	0.005	2.3	0.274	0.004	1.6	19.3
2-Methylheptane	0.163	0.003	1.6	0.223	0.005	2.2	26.9
3-Methylheptane	0.169	0.007	3.9	0.179	0.003	1.7	5.9
n-Octane	0.118	0.004	3.2	0.141	0.001	0.9	16.0
n-Nonane	0.044	0.002	3.9	0.085	0.002	1.8	49.0
Ethane	33.987	0.100	0.3	33.201	0.107	0.3	−2.4
Propene	8.550	0.050	0.6	10.018	0.034	0.3	14.7
Trans-2-butene	0.582	0.007	1.3	0.581	0.003	0.5	−0.3
1-Butene	1.340	0.025	1.9	1.221	0.006	0.5	−9.8
Isobutene	5.128	0.027	0.5	5.805	0.022	0.4	11.7
Cis-2-butene	0.577	0.012	2.0	0.570	0.002	0.4	−1.1
3-Methyl-1-butene	0.122	0.003	2.3	0.118	0.003	2.2	−3.7
1-Pentene	0.171	0.003	1.9	0.127	0.001	0.6	−34.8
Isoprene	0.029	0.001	4.4	0.026	0.000	0.0	−10.5
Trans-2-pentene	0.269	0.003	1.3	0.248	0.001	0.5	−8.9
Cis-2-pentene	0.136	0.004	3.0	0.137	0.002	1.6	1.0
2-Methyl-2-butene	0.253	0.006	2.5	0.262	0.003	1.0	3.2
2-Methyl-1-pentene	0.085	0.004	4.7	0.308	0.002	0.6	72.4
Ethyne	22.269	0.139	0.6	19.967	0.006	0.4	−11.5
Benzene	2.776	0.030	1.1	3.133	0.011	0.3	11.4
Toluene	5.206	0.046	0.9	5.925	0.009	0.4	12.1
Ethylbenzene	0.986	0.007	0.7	0.921	0.007	1.6	−7.0
m/*p*-Xylene	2.941	0.037	1.3	3.363	0.013	0.4	12.5
o-Xylene	1.093	0.011	1.0	1.214	0.010	0.7	10.0
Isopropylbenzene	0.075	0.001	1.5	0.050	0.015	2.6	−51.8
n-Propylbenzene	0.130	0.001	0.9	0.134	0.009	2.3	3.4
1,3,5-Trimethylbenzene	0.196	0.003	1.6	0.199	0.016	1.4	1.7
1,2,4-Trimethylbenzene	0.620	0.004	0.7	0.716	0.007	1.4	13.4

[a]Difference = (data from UCI−data from RCEC)/(data from UCI).

urban sites improved in 2005. Vehicle speeds were generally higher in 2005, resulting in a more even and smooth traffic flow than in 2000. This in turn would result in lower hydrocarbon emissions from vehicles and improve roadside air quality.

To solve the congested traffic situations in urban areas and to promote the development of the suburban and rural areas, a large number of highways were built in the PRD. During the government's 10th 5-year plan (2000–2005), about

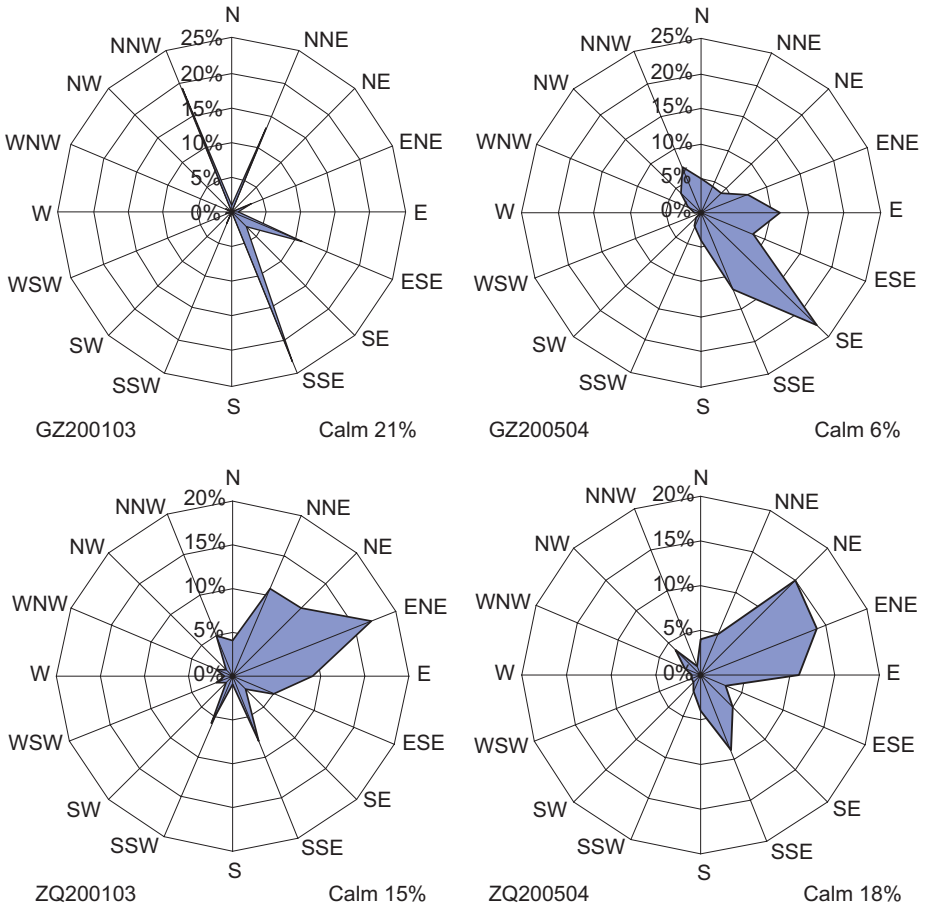

Fig. 2. Wind roses at Guangzhou (GZ) and Zhaoqing (ZQ) in March 2001 and April 2005.

2000 km of highways were built, and the highway in Guangdong Province reached 3100 km. During the 11th 5-year plan, 25,000 km of roads, including 2000 km of highways, are to be built in Guangdong Province. This will further increase road traffic and will encourage the spread of industrial activity to less developed regions.

Although only six roadside and tunnel samples and six roadside samples were collected in 2000 and 2005, respectively, these limited data supported our hypothesis that tighter emission standards and improved roadways helped to limit the increase in NMHC emissions from vehicles relative to the increase in vehicular fleet. For example, Xingang Road was widened from three lanes in both directions in 2000 to four lanes in 2005, and several overpasses and connections were built after 2000 to improve the traffic jams and deadlock situations, which were common in 2000. These changes are expected to have contributed to the significant decrease of most roadside and tunnel hydrocarbon levels in 2005 compared to 2000, although we also note that the 2000 samples were collected at a roundabout, where vehicle speeds were slower than for the 2005 roadside samples, and therefore emissions are expected to be higher.

3.4. Impact from LPG emission and leakage

The consumption of LPG sharply increased from 2000 to 2004 in Guangdong Province (Fig. 4; Guangdong Statistics Bureau, 1999–2005). In GZ, approximately 700,000 tons of LPG were consumed in 2004, of which 450,000 tons were for residential use. While in 2000, it was 190,000 and 120,000 tons, respectively (Guangzhou Municipal Statistics Bureau, 2000, 2005). The rapid increase in the use of LPG is the likely cause of the significant increase of propane observed in GZ. To mitigate the air pollution problem from vehicular exhaust, the GZ municipal government began promoting the use of LPG-fueled buses and taxis in 2003. By the end of that year, there

Table 3
Comparison of mixing ratios of selected hydrocarbons in ambient air of GZ and DM between 2001 and 2005 and roadside sample of GZ between 2000 and 2005

Hydrocarbon	Mixing ratio (ppbv)					
	GZ2001 ($n = 22$)	GZ2005 ($n = 6$)	DM2001 ($n = 5$)	DM2005 ($n = 6$)	Rd & Tun 2000 ($n = 6$)	Roadside 2005 ($n = 6$)
Ethane	4.19±1.37	3.41±0.69	4.07±1.81	2.73±0.25	10.56±4.13	3.63±0.83
Propane	5.90±2.84	8.94±2.65	2.09±0.95	1.91±0.69	11.42±6.16	29.85±12.31
Isobutene	3.55±1.99	3.96±1.11	0.73±0.45	0.81±0.38	11.41±6.30	5.72±1.27
n-Butane	5.21±3.02	4.61±1.28	1.18±0.76	1.05±0.58	15.43±10.24	10.71±2.59
Isopentane	3.37±2.01	3.47±0.75	0.68±0.44	1.16±0.50	47.04±30.17	3.73±1.06
n-Pentane	1.11±0.68	1.61±0.61	0.37±0.33	0.66±0.40	10.55±6.35	1.63±0.45
2-Methylpentane	1.50±0.90	1.53±0.46	0.28±0.19	0.41±0.17	8.06±0.10	1.22±0.39
3-Methylpentane	1.19±0.65	1.03±0.32	0.33±0.19	0.26±0.12	4.56±0.15	0.79±0.24
n-Hexane	1.30±0.79	1.49±0.59	0.26±0.18	0.42±0.19	6.30±3.72	0.66±0.27
Methylcyclopentane	0.64±0.40	0.62±0.17	0.12±0.08	0.14±0.07	N.A.[a]	N.A.
2,3,4-Trimethylpentane	1.40±1.17	1.26±0.63	0.05±0.04	0.01±0.01	N.A.	N.A.
Ethane	8.01±4.26	7.35±1.58	3.09±1.50	2.58±0.79	82.20±34.00	12.67±5.29
Propene	1.92±1.22	2.24±0.77	0.80±0.74	0.56±0.24	22.07±8.44	2.91±1.28
1-Butene	0.55±0.35	0.42±0.21	0.20±0.15	0.11±0.06	8.78±5.56	0.85±0.28
Isobutene	0.68±0.43	0.73±0.29	0.14±0.09	0.25±0.06	7.35±3.05	1.11±0.50
Trans-2-butene	0.78±0.53	0.73±0.27	0.06±0.05	0.06±0.02	10.44±6.80	0.73±0.22
Ethyne	10.08±4.99	7.65±1.30	3.35±1.20	4.06±0.99	94.28±43.49	10.20±5.14
Benzene	3.35±1.52	2.87±0.52	0.98±0.58	1.63±0.86	18.10±7.43	2.07±0.52
Toluene	16.62±0.49	11.70±4.55	4.95±4.32	3.96±1.97	25.90±10.50	4.01±2.34
Ethylbenzene	2.76±2.00	1.70±0.82	0.80±0.74	0.54±0.35	3.32±0.24	0.91±0.36
m/*p*-Xylene	6.49±6.23	3.27±1.35	2.31±0.90	0.96±0.75	15.73±1.23	1.92±0.64
o-Xylene	3.31±2.64	1.25±0.52	0.85±0.73	0.37±0.27	8.02±0.97	0.63±0.19
Total NMHC	87.4±47.1	75.3±17.1	28.3±14.1	25.9±9.2		

[a]N.A., not available.

were 207 and 874 LPG-fueled buses and taxis in service, respectively (Guangzhou Yearbook Committee, 2004). By November 2005, there were approximately 5060 and 9200 LPG-fueled buses and taxis in GZ, respectively, which accounted for 67% and 58% of the total buses and taxis (New Express, 2005). By the end of 2006, all taxis and buses were LPG fueled.

Propane and *n*-butane, which are the major components of LPG (Tsai et al., 2006) and the dominant hydrocarbons in exhaust of LPG-fueled taxis (Fig. 3), are strongly correlated in the GZ, DM, and GZ roadside and tunnel samples, indicating that they originated from common sources (Fig. 5). The slope of propane to *n*-butane in the 2005 roadside samples (3.7) was much higher than those in the roadside and tunnel samples of 2000 (0.6), which is consistent with the introduction of LPG-fueled buses and taxis in 2003.

Propane was the only hydrocarbon in the roadside samples to show an increase rather than a decrease between 2000 and 2005 (Table 3). This suggests that LPG-fueled vehicles emit more propane than *n*-butane in GZ, although the composition of LPG used in GZ is unknown to us. In Hong Kong, the composition of LPG is 46% *n*-butane, 26% propane and 22% *i*-butane (Tsai et al., 2006), although LPG formulations with higher percentages of propane have been reported in other cities (e.g. propane:*n*-butane:*i*-butane = 68:16:12 in Mexico City, Vega et al., 2000; propane:*n*-butane:*i*-butane = 83:11:5 in Taiwan, Chang et al., 2001). In 2001, before the introduction of LPG-fueled buses and taxis, the propane:*n*-butane slopes were nearly the same in GZ (0.97) and at DM (1.07), suggesting a characteristic ratio of propane/*n*-butane in ambient air of approximately 1.0 in these regions. The slopes increase to 1.9 (GZ) and 1.6 (DM) in 2005, which suggests that the introduction of LPG-fueled vehicles affected the ambient levels of propane to a greater extent than *n*-butane. This is supported by the higher percentage of propane than

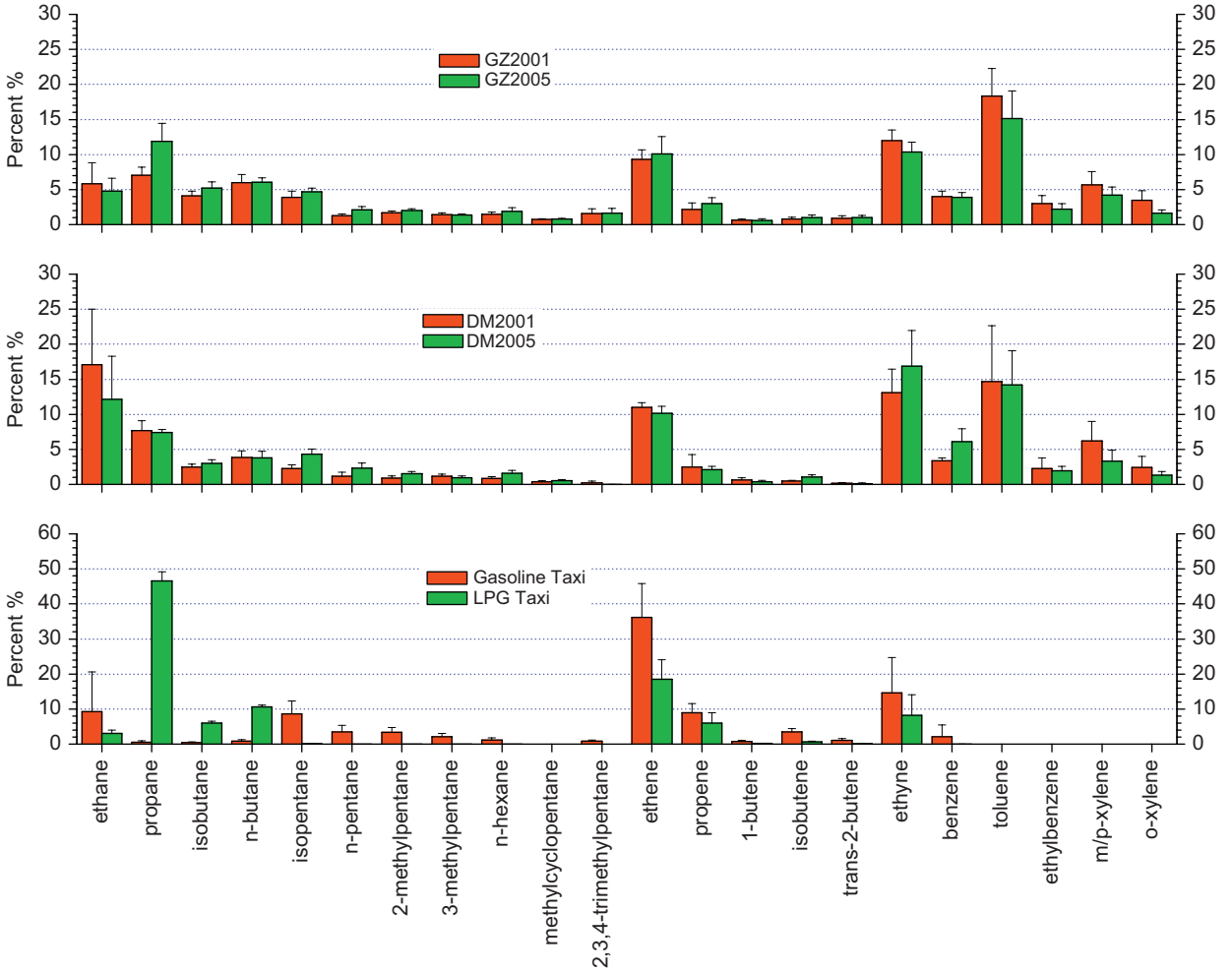

Fig. 3. Distribution of hydrocarbons in ambient air of Guangzhou and Dinghu mountain and exhaust samples of gasoline- and LPG-fueled taxis in Guangzhou.

n-butane in the LPG-fueled taxi exhaust (Fig. 3). The propane vs. *n*-butane slopes for both sites were lower than for the roadside samples in 2005 (3.7), which indicate that *n*-butane has a significant source other than LPG-fueled vehicle emissions, possibly with contribution from gasoline evaporation and vehicular exhaust (Tsai et al., 2006).

3.5. *Factors affecting the levels of toluene, ethylbenzene and xylenes*

In the PRD region, several studies have shown that transportation is the largest contributor to VOC emission (CH2M HILL, 2002; Streets et al., 2006). However, Chan et al. (2006) found that industrial activities also greatly contributed to ambient VOC levels. Their study of NMHCs in industrial, industrial-urban and industrial-suburban environments showed that toluene was the most abundant hydrocarbon, and industrial solvent and vehicular emission were its major sources. Extremely high levels of toluene (2560 ± 560 ppbv) were also reported at the rooftop of a printing factory in the PRD (Tang et al., 2007b).

Toluene was the most abundant hydrocarbon in GZ in both 2001 and 2005. At DM, toluene was the most abundant species in 2001 and the second most abundant in 2005, after ethyne (Table 3). Both the mixing ratio and the percentage of toluene, ethylbenzene, *m*/*p*-xylene and *o*-xylene (TEXs) decreased from 2001 to 2005 in the ambient air of GZ and DM. TEXs have common sources in the atmosphere, such as industrial solvent evaporation and diffuse emissions and gasoline evaporation. In this

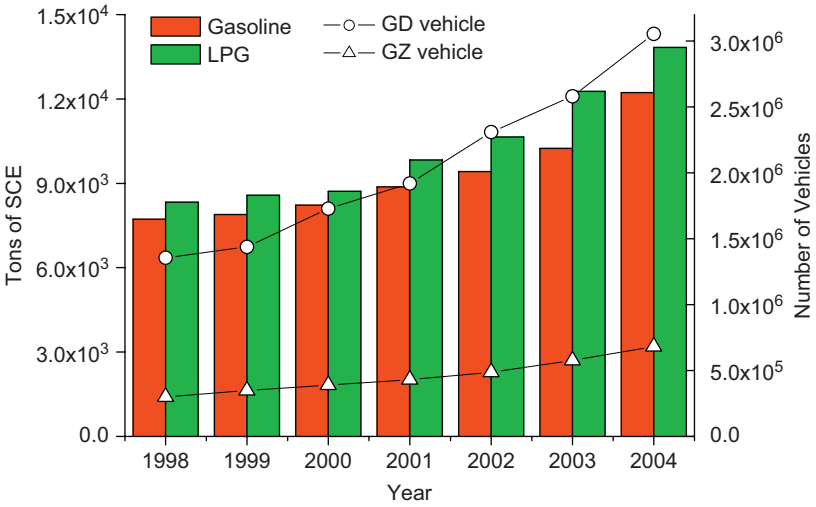

Fig. 4. Average daily usage of LPG and gasoline in Guangdong Province (units: tons of Standard Coal Equivalent) and motor vehicle numbers in Guangdong Province (GD) and Guangzhou (GZ) (*Source*: Guangdong Statistics Bureau, 1999–2005 and Guangzhou Municipal Statistics Bureau, 1999–2005).

study, the correlation of toluene with ethylbenzene and *o*-xylene were plotted for the entire ambient samples (Fig. 6). Toluene highly correlated with ethylbenzene and *o*-xylene at both sites in both years ($R \geqslant 0.87$), indicating they shared common sources.

In previous studies, the toluene to benzene (T/B) concentration ratio was used to infer their sources (Chan et al., 2002; Lee et al., 2002; Barletta et al., 2005; Wang T. et al., 2005). High T/B ratios were found in the Hong Kong urban area ($T/B = 5.0$, Lee et al., 2002) and rural area ($T/B = 5.4$, Wang T. et al., 2005) as compared with other cities (0.7−2.0). Very high T/B ratios were also found in gasoline evaporation samples of Hong Kong (22.1 ± 12.7), GZ (3.7 ± 1.5), Zhuhai (3.9 ± 0.7) and Macau (23.0 ± 1.8), due to the use of high levels of toluene in aromatic-rich unleaded fuels and industrial solvents (Tsai et al., 2006). A previous study in five PRD cities found that the T/B ratio in ambient samples of industrial, industrial-urban and industrial-suburban sites were 6.3 ± 8.0, 6.0 ± 4.7 and 6.9 ± 5.6, respectively (Chan et al., 2006). In this study, the T/B ratios decreased in 2005 compared with 2001 at both sites from 4.9 ± 2.9 to 3.7 ± 1.2 in GZ and from 3.0 ± 1.8 to 2.5 ± 1.0 at DM, while it was the same in the roadside and tunnel samples for 2000 (1.4) and the roadside samples for 2005 (1.4, when one sample of 4.9 was excluded). This indicates that additional source(s) (e.g. industrial solvents) other than motor vehicle emissions also contributed to ambient levels of toluene and benzene in GZ and DM. The higher T/B ratio in GZ than on DM indicates that the emission strength of toluene is greater in GZ than on DM and this is consistent with the existence of more industrial enterprises in GZ.

One hundred and fifty-seven highly polluting factories were forced to close, or relocate, if they did not comply with the emission standards set by the government in the year 2004 (Guangzhou Yearbook Committee, 2005). Additionally, many factories in this region had voluntarily relocated out of GZ in recent years in pursuit of cheaper labor, less stringent pollutant emission standards and lower running costs. Some large factories in GZ had closed down due to high-energy consumption, high pollutant emission, low efficiency, and changes in government policy on large pollutant emitter. For example, the number of large industrial enterprises decreased dramatically from 2002 to 2003 (Fig. 7). Although the number of medium and small-sized industrial enterprises increased between 1999 and 2004, these new enterprises are less polluting than the older ones. Due to the closing or relocation of large industrial enterprises and other control efforts, ambient levels of TEXs in the GZ urban area and DM rural site decreased between 2001 and 2005.

Another important factor reducing the levels of TEXs in the atmosphere should be the implementation of more strict emission standard for motor vehicles. Vehicles registered after 2000 were forced to equip with three-way catalytic converters. A study in

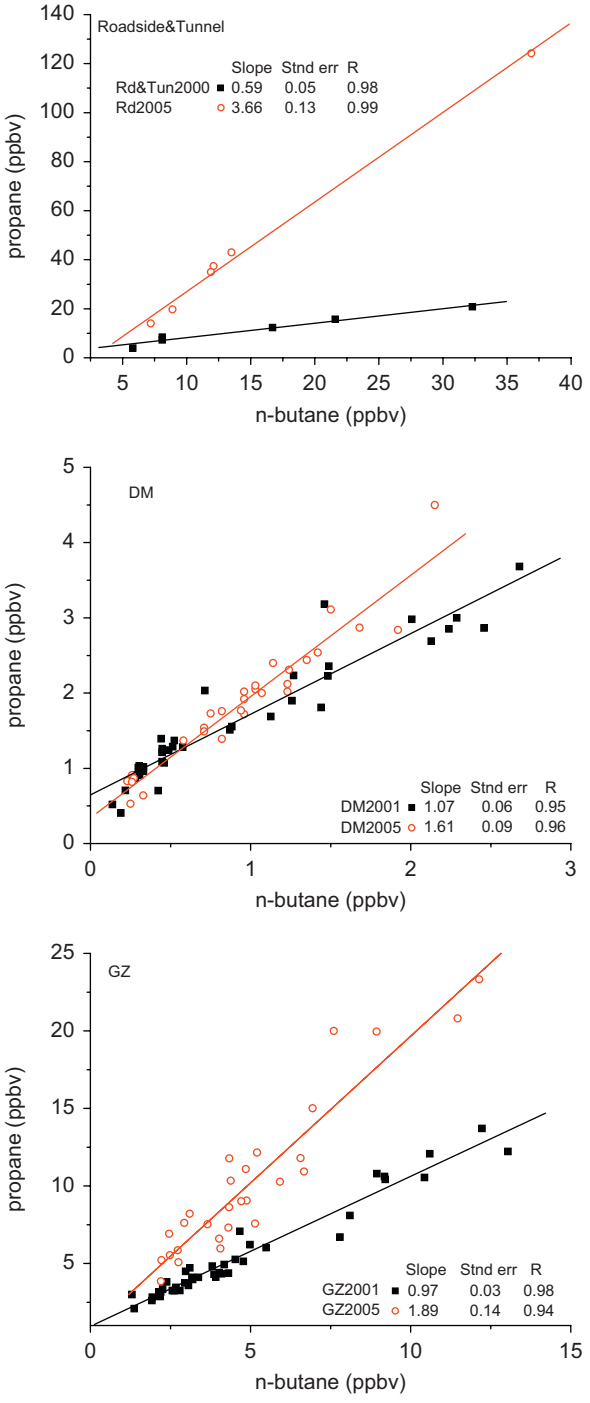

Fig. 5. Correlation of propane with *n*-butane.

a suburban site of Switzerland found that the levels of toluene and C_2-benzenes (ethylbenzene and xylenes) had steady decreased from 1994 to 1998, when the proportion of three-way catalyst passenger cars increased from 60% to 82% (Heeb et al., 2000).

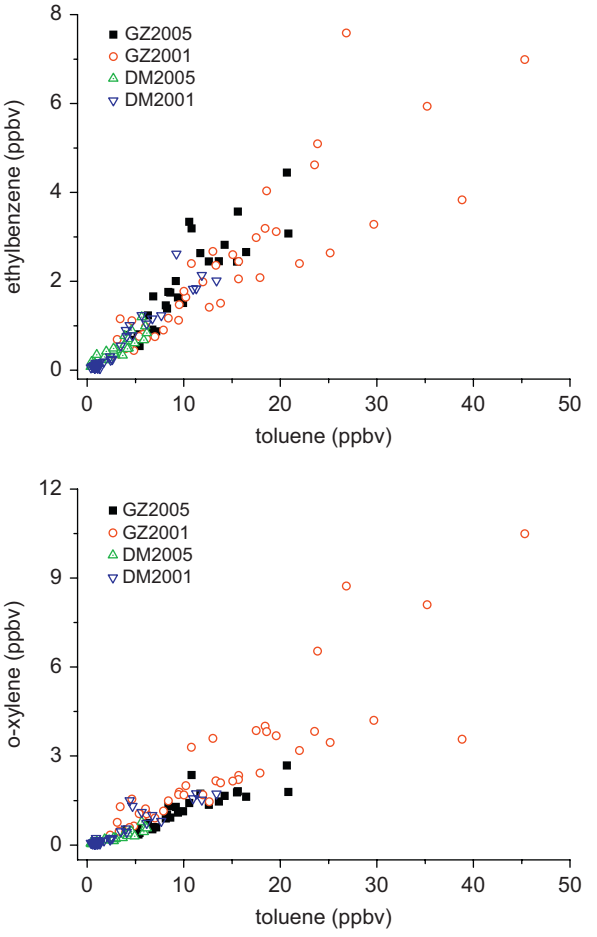

Fig. 6. Correlation of toluene with ethylbenzene and *o*-xylene.

There were no available data about the proportion of vehicles equipped with three-way converter in GZ and DM regions, but the number had certainly increased during the study period. It also can be inferred from the steady increase of new registered vehicle number (Fig. 4).

4. Conclusions

From 2001 to 2005, NMHC profiles varied greatly in both the city of GZ and a rural site, DM. Leakage of LPG is a major source of propane and *n*-butane in GZ and on DM. The increasing use of LPG-fueled buses and taxis, especially in GZ, has significantly affected ambient levels of propane. The main reasons for the decrease of ambient TEXs in GZ and DM should be the relocation and close of some polluting industries especially in GZ, the introduction of LPG-fueled buses and taxis, and the implementation of stricter vehicle and industrial

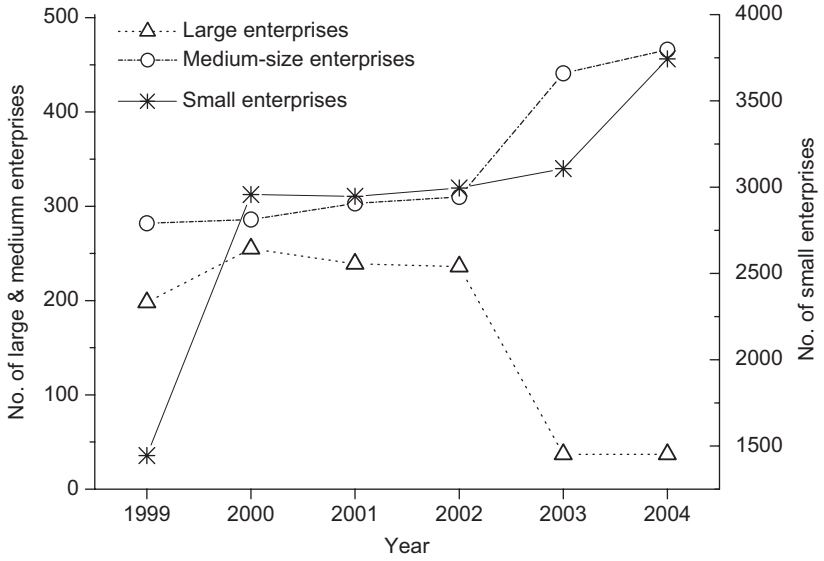

Fig. 7. The number of industrial enterprises designated by size in the Guangzhou urban area (*Source*: Guangzhou Municipal Statistics Bureau, 2000–2005).

emission standards. To better understand the effect of these emission controlling methods and strategies, more studies coupled with long-term monitoring programs should be implemented.

With a fast growing economy and ongoing urbanization, the number of motor vehicles and usage of LPG will continue to increase in GZ and the PRD region. Emissions from vehicular exhaust and leakage of LPG into the atmosphere will further degrade the local and regional air quality. The closing and relocation of polluting factories from developed urban areas to less developed outskirt towns will spread pollution to a larger part of the PRD, while perhaps decreasing industrial pollution in central cities. The observed toluene/benzene reductions in 2005 compared with 2005 at both sites indicate the effectiveness of local/regional emission regulations. However, it is imperative that the PRD government should continue to impose additional emission regulations and modify existing ones to address the continuing emission changes in the PRD so that the air quality will not further deteriorate.

Acknowledgment

This project was funded by the Research Grants Council of Hong Kong (PolyU 5048/02E and PolyU 5131/04E), a Research Grant of The Hong Kong Polytechnic University (A504), and the Comer Foundation. We would like to acknowledge the contribution of all the staff who had participated in the field sampling. The comments of the two anonymous reviewers have helped to improve greatly the quality of this manuscript, and their helps are much appreciated.

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