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Regional and local contributions to ambient non-methane volatile organic compounds at a polluted rural/coastal site in Pearl River Delta, China

H. Guo[a,1], T. Wang[a,*], D.R. Blake[b], I.J. Simpson[b], Y.H. Kwok[a], Y.S. Li[a]

[a]*Department of Civil and Structural Engineering, The Hong Kong Polytechnic University, Hong Kong*
[b]*Department of Chemistry, University of California at Irvine, CA 92697, USA*

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Abstract

Identification of major sources of airborne pollutants and their contribution to pollutant loadings are critical in developing effective pollution control and mitigation strategies. In this study, a comprehensive dataset of non-methane volatile organic compounds (NMVOCs) collected from August 2001 to December 2002 at a polluted rural/coastal site in the Pearl River Delta (PRD) is analyzed to assess the relative contributions of major pollution sources to ambient NMVOC mixing ratios. A unique approach based on emission ratios of individual chemical species was used to classify the bulk air samples in order to apportion regional and local source contributions to the measured mixing ratios. The collected air samples fell into four major groups, including air masses from the inner PRD region and Hong Kong (HK) urban area. To estimate the source apportionment of NMVOCs, a principal component analysis/absolute principal component scores receptor model was applied to the classified data points. The results indicate that the regional and local source contributions to ambient NMVOC levels at the site were significantly different due to the differences in local versus regional energy use and industrial activities. For air masses originating from HK, vehicular emissions accounted for approximately 39% of the total NMVOC levels, followed by industrial emissions (35%), gasoline evaporation (14%) and commercial/domestic liquefied petroleum gas/natural gas use (12%). By contrast, for air masses originating from the PRD the industrial emissions accounted for 43% of the total NMVOC burden, followed by vehicular emissions (32%) and biomass burning (25%). In particular, the higher regional contribution of biomass burning found in this study as compared to existing emission inventories suggests that further efforts are necessary to refine the emission inventories of NMVOCs in the PRD region.
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1. Introduction

The Pearl River Delta (PRD) is situated on the southern China coast and is now home to approximately 40 million inhabitants. It is also a major industrial base in southern China (CH2M, 2002).

*Corresponding author. Tel.: +852 2766 6059; fax: +852 2334 6389.
E-mail address: cetwang@polyu.edu.hk (T. Wang).
[1]Now at Department of Chemistry, University of California at Irvine, CA 92697, USA.

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Due to rapid economic development and increased energy use in the past two decades, the amount of major air pollutants emitted to the atmosphere is believed to be increasing significantly, with evidence for higher ozone levels and lower visibility on a regional scale (Wang et al., 2003a, b, 2005; Wang and Kwok, 2003; Wang, 2003). The frequency of visibility impairment in Hong Kong (HK) showed an increasing trend from 1991 to 2002 and visibility degradation in nearby Shenzhen was even more serious (Wang, 2003). Ground-based measurements in HK indicate that ozone levels in the territory showed a slow rising trend between 1991 and 2002, and the annual average ozone level for urban stations was 50% higher in 2002 than in 1991 (HKEPD, 2002). Non-methane volatile organic compounds (NMVOCs) are significant precursors of ozone and other oxidants in the atmosphere, and photochemical analysis shows that NMVOCs are a limiting factor in the formation of ozone in HK (So and Wang, 2003; Zhang et al., 2004). Therefore, to impose efficient emission restrictions on the most dominant local and regional NMVOC sources and to establish the relative influence of regional versus local sources on the ambient air quality, it is important to understand the chemical composition of NMVOCs in the PRD region, identify major source regions of air pollution, and quantify the relative contribution of each source sector to ambient NMVOC levels.

Detailed hydrocarbon speciation can provide fingerprints for estimating source contributions to the overall NMVOC burden. Previous studies show that NMVOCs have significant variations in their composition for different sources (Mayrsohn and Crabtree, 1976; Nelson and Quigley, 1984; Na and Kim, 2001) and their fingerprints can be used to identify emission sources. For instance, ethylene and acetylene are tracers of vehicle exhaust in urban areas (Mayrsohn and Crabtree, 1976). Butanes and pentanes are known to be the representative compounds of gasoline evaporation (Morikawa et al., 1998), and toluene, ethylbenzene and xylenes indicate solvent usage (Brocco et al., 1997).

The principal component analysis (PCA)/absolute principal component scores (APCS) receptor model represents a source apportionment technique which requires a minimum of inputs regarding source characteristics, but provides quantitative information regarding source profiles. The receptor model calculates the source profiles and source strengths in absolute concentration units using data measured at the receptor site. The model does not require prior knowledge as to the number of active pollution sources and their composition. Although other models such as chemical transport models and chemical balance models can also estimate source contributions, they need detailed source profiles. In Asian countries, particularly in China, such source profiles are unavailable or are highly uncertain. Therefore, for those regions such as the PRD without available source profiles, this receptor model becomes a useful tool for source apportionment. A comprehensive field measurement campaign was carried out at a polluted rural/coastal site in the PRD from August 2001 to December 2002, in order to understand the photochemical smog problem in this region. Using this data set, the causes of a multi-day ozone episode were analyzed (Wang and Kwok, 2003), the temporal variability and emission patterns of pollution plumes during October–December 2001 were characterized (Wang et al., 2003a), and the seasonal profiles and atmospheric processes of trace gases (including NMVOCs) at the study site were overviewed (Wang et al., 2005). C_1-C_5 alkyl-nitrates and the relation to their parent NMVOCs and to ozone were examined in Simpson et al. (2005). Here, we focus on the NMVOC data to estimate relative contributions of major sources to atmospheric NMVOC levels. We first examine the abundance and speciation of NMVOCs, and tracers of anthropogenic and biogenic emissions. The air samples are then classified into local and regional groups based on the ratios of chemical species. Finally, the major local and regional pollution sources are identified and their contributions to ambient NMVOCs are quantified.

2. Experimental

2.1. Sampling site

The sampling site was located at Tai O, a rural/coastal area in southwest HK (Fig. 1). The study site is located on a hill 80 m above sea level, overlooking the Pearl River Estuary to the west and north, and the South China Sea to the south. Tai O is 32 km away from the HK urban center to the east and approximately the same distance from Macau/Zhuhai to the west. Tai O is at the southern tip of the PRD region. Major anthropogenic sources of NMVOCs in the region are located to the east, north, and southwest. HK, Guangzhou and

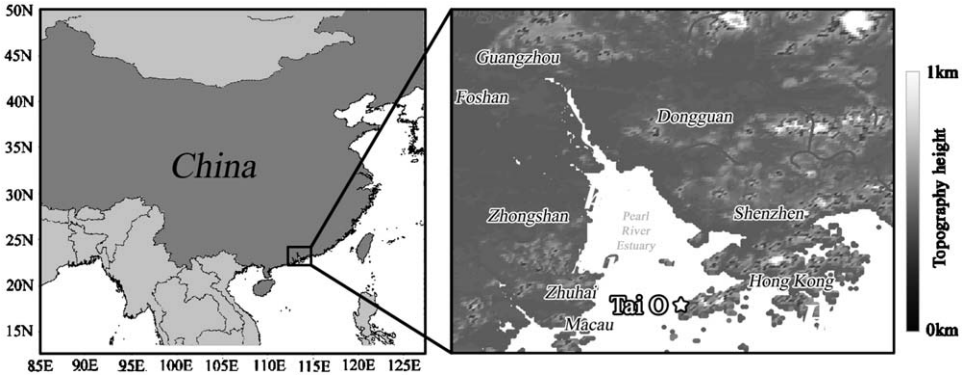

Fig. 1. Map showing the study site, Tai O, and the PRD of China.

Shenzhen are the three largest cities in this region. Local emissions are small due to a sparse population and light traffic at Tai O.

2.2. Sampling and chemical analysis

One hundred and eighty seven air samples were pressurized to about 20 psi into individual 2-L evacuated electro-polished stainless steel canisters using a metal bellows pump. The canisters were cleaned and evacuated at the University of California at Irvine (UCI) before being shipped to Tai O. Details of the preparation and pre-conditioning of the canisters prior to sampling are described in Blake et al. (1994). During sampling the canister bellows valve was slightly opened, allowing about 1 min for the collection of the "integrated" samples. The canisters were then shipped to UCI for chemical analysis. A 6-column multiple GC–MS system was used to identify and quantify the NMVOCs. Detailed descriptions of the chemical analysis and relevant quality assurance/quality control are given by Colman et al. (2001).

Between 1 and 32 air samples were collected every month during the sampling period. The sampling strategy emphasized pollution outflow from the PRD. Thus most of the air samples were acquired between October and December which is the period of strong outflow of pollution from PRD (including HK) and southern China. In contrast, much fewer samples were taken in April–July when air masses are mainly from South China Sea background. Due to the limitation of resources and difficulty in accessing to the site in the evening, nocturnal samples were not taken. Normally one sample was taken in a day, but when an ozone episode was anticipated, 4–7 samples were collected daily on consecutive days until the end of the episode, in order to investigate the relationship between ozone and its precursors. Seventy five percent of the samples were collected in the afternoon and 25% in the mid-morning.

2.3. PCA/APCS receptor model

The chemical analytical technique generated a multivariate database because more than 30 NMVOCs were measured simultaneously in the same air mass. The PCA/APCS model itself generates a hypothesis regarding the number of sources, the source profiles and source apportionment (Swietlicki et al., 1996). The multivariate method is performed by orthogonal transformation method with Varimax rotation and retention of principal components whose eigenvalues are greater than one, followed by multi-linear regression analysis. The main limitations of the multivariate techniques are that they can only identify 5–8 sources and that the dataset must contain a large number of samples, usually no less than 50 (Thurston and Spengler, 1985). Obtaining reliable results also requires the analysis of a variety of NMVOCs, including known source tracers for each major source. When the receptor model is used, the sampled air is assumed to be well mixed, with species from different sources, and the species are relatively stable during the transport from the emission sources to the receptor site. Instances where the receptor model did not appear to correctly attribute individual NMVOCs to specific sources are discussed wherever appropriate below. Details about the PCA/APCS model are given elsewhere (Guo et al., 2004a).

3. Results and discussion

3.1. General characteristics

Table 1 summarizes the average concentrations of 34 NMVOCs measured at Tai O. Large variations in the measured NMVOCs were observed. The total average NMVOC concentration was 25.5 ppbv, in which alkanes accounted for 40%, alkenes 10%, alkynes 11%, aromatics 35% and measured halocarbons (C_2Cl_4 and CH_3Cl) 4%. The most abundantly measured 10 compounds were toluene, ethyne, ethane, propane, ethene, *n*-butane, CH_3Cl, ethylbenzene, benzene and *i*-pentane. These 10 NMVOCs accounted for 76% of the total NMVOCs. In particular, toluene alone accounted for 22% of the average. Though not on the top 10 list, isoprene, which was mainly from local biogenic emissions (Wang et al., 2005), showed highly variable concentrations and could reach as high as 5 ppbv on some days in summer, suggesting its importance in the formation of ozone at the site on these days given its very high reactivity. Table 2 shows a comparison of most abundant NMVOCs in different regions. It should be noted that the mean values of NMVOCs in different regions can vary largely depending on the season the observations cover, the geographical condition (i.e. transport scheme, distance from source regions), sampling strategy, analytical method and so on. Nevertheless, the comparison can still provide information about the composition in different regions. It can be seen that in other regions most of the top 10 species consist of C_2-C_5 alkanes, ethene and ethyne. In addition, toluene and benzene are often included in the top 10 species. The composition of top 10 species is similar from site to site but their ranking and distribution are significantly different. The different ranking and distribution of NMVOCs are controlled by a combination of emission factors, dispersion conditions, chemical mechanisms and site characteristics (Rappenglück and Fabian, 1999; Borbon et al., 2002).

Table 1
Average concentration of carbon monoxide (ppbv) and 34 NMVOCs (pptv) measured at Tai O

	Average	Standard deviation	Range
Carbon monoxide	525	323	82–2007
Ethane	2122	990	375–5047
Propane	2048	2159	18–12,995
n-Butane	1638	2132	6–12,792
i-Butane	804	928	5–6047
n-Pentane	448	650	ND–5594
i-Pentane	804	1441	13–17,247
n-Hexane	503	674	ND–4741
2,2-Dimethylbutane	38	52	ND–396
2,3-Dimethylbutane	135	207	ND–1318
2-Methylpentane	466	581	ND–4143
3-Methylpentane	410	453	ND–134
n-Heptane	333	500	ND–4359
n-Octane	58	58	ND–426
2,2,4-Trimethylpentane	136	1184	ND–15,937
n-Nonane	77	86	ND–594
n-Decane	99	147	ND–1474
Ethene	1674	1685	33–10,530
Propene	223	296	11–2178
i-Butene	110	172	ND–1050
1-Butene	69	101	ND–737
1,3-butadiene	29	55	ND–429
Isoprene	427	727	ND–5352
Ethyne	2772	1996	79–11,746
Benzene	869	921	17–10,318
Toluene	5670	7128	11–48,982
o-Xylene	375	566	ND–4330
m-Xylene	568	1135	ND–9125
p-Xylene	402	731	ND–5594
Ethylbenzene	874	1225	ND–8119
1,3,5-Trimethylbenzene	59	77	ND–536
2-Ethyltoluene	39	52	ND–408
1,2,4-Trimethylbenzene	153	218	ND–1772
Tetrachloroethene	154	181	ND–1094
Methyl chloride	884	181	604–1546

ND: non-detectable.

3.2. Indicators of air masses from the inner PRD and HK metropolitan area

Air masses arriving at the study site are expected to fall into several major groups: (1) urban plumes from HK when wind directions are easterly and northeasterly; (2) polluted air masses from the inner PRD cities in the events of northerly and northeasterly winds; (3) a mixture of (1) and (2) when the winds are light and variable; (4) inflow of South China Sea background air.

Back trajectories and/or local winds are widely used to trace the origin and transport pathway of an arriving air mass. However, due to the complex boundary-layer flow caused by sea–land breezes and the effect of topography in the study area, we adopt here a chemical approach for the segregation of data, which is based on the ratio of chemical species. This kind of classification was performed previously by our group (Wang et al., 2003a) by examining the ratios of CO to NO_y (total reactive nitrogen). The inner PRD had higher CO/NO_y and than that in

Table 2
The most abundant NMVOCs in different regions (ppbv)

Rural/coastal site, Rörvik, Sweden[a]		Rural site, Robinson, IL, USA[b]		Rural site, Linan, Eastern China[c]		Rural/coastal site, Tai O, Hong Kong[d]	
Ethane	3.00	Propane	4.00	Ethane	3.11	Toluene	5.67
Propane	1.60	Ethane	3.00	Ethyne	2.67	Ethyne	2.77
Ethyne	1.10	*n*-Butane	2.12	Ethene	2.09	Ethane	2.12
n-Butane	0.87	Ethene	1.00	Toluene	1.96	Propane	2.05
Ethene	0.65	*i*-Pentane	1.00	Methyl chloride	1.32	Ethene	1.67
i-Butane	0.48	*n*-Pentane	0.70	Propane	1.28	*n*-Butane	1.64
i-Pentane	0.46	*i*-Butane	0.61	Benzene	1.11	Methyl chloride	0.88
n-Pentane	0.24	Ethyne	0.50	*n*-Butane	0.51	Ethylbenzene	0.87
Benzene	N/A	Propene	0.50	*i*-Butane	0.48	Benzene	0.87
Toluene	N/A	Toluene	0.30	*i*-Pentane	0.44	*i*-Pentane	0.80

N/A: not available.
[a]Mowrer and Lindskog (1991).
[b]Sexton and Westberg (1984).
[c]Guo et al. (2004b).
[d]This study.

HK, which has been attributed to incomplete combustion of coal and biofuels in the PRD and the dominance of low CO-emitting diesel vehicles in HK (Kok et al., 1997). In the present study, we chose propane and CO as a pair because they have relatively low loss rates against reaction with OH, with a lifetime 1–2 months and 1–4 months for propane and CO, respectively, and are inefficiently removed by wet removal processes. Considering relatively short distance from sources to the sampling site, the effect of atmospheric mixing is insignificant. Thus changes in their ratio should largely reflect the differences in source emissions.

Fig. 2a shows the scatter plot for CO versus propane for the sampling period. The data points in the plot fell into four groups. Points on the upper-left side contained high propane mixing ratios and low CO, giving a very large propane/CO slope (8.6 pptv/ppbv). In contrast, data points on the lower-right side had high CO mixing ratios and low propane, and thus a much smaller slope (2.2). The third group was composed of points with moderately high CO and propane, with a slope in between the above two groups (4.6). The fourth group was in the lower-left corner with CO mixing ratios less than 125 ppbv, representing the South China Sea air. Similarly, plots of other NMVOCs versus CO also showed both high- and low-NMVOC/CO slopes. The different propane/CO slopes can be interpreted as signatures of urban plumes originating from inner PRD cities ($\Delta\text{propane}/\Delta\text{CO} = 0-4$), from HK ($\Delta\text{propane}/\Delta\text{CO} = 5-300$), and a mixture of both ($\Delta\text{propane}/\Delta\text{CO} = 4-5$). The large difference in the propane/CO ratio between HK and inner PRD cities could be due to the combination of heavy usage of LPG-fuelled taxis, widespread domestic use of LPG, plus a large number of diesel vehicles in HK, and high CO production in low-temperature combustion processes in inner PRD cities. While the CO level in inner PRD (and other parts of eastern China) is higher, propane in urban HK could be more abundant. The latter can be supported by the following data. Among the 500,000 registered vehicles in HK, 17,000 taxis are using LPG as fuel (HKTD, 2005). In addition, LPG is widely used in domestic households such as cooking and heating in HK. Evaporation and leakage of LPG may contribute to the elevated propane mixing ratios in HK urban air. By contrast, only about 3000–4000 taxis and buses in inner PRD region were modified to LPG-fuelled vehicles out of 5.3 million vehicles in 2001 (Industry Canada, 2002; The Asia Foundation and Civic Exchange, 2001). Moreover, the total number of LPG-fuelled vehicles in the PRD region remains the same since the PRD terminated the replacement of LPG-fuelled vehicles in 2000. Thus, LPG fuel contributes relatively less to the ambient propane concentrations in inner PRD.

Using the classification based on the propane/CO ratios, the relationship of NO_y and CO in corresponding air masses was then plotted (Fig. 2b). The CO to NO_y ratios in local (HK), mixed and regional (inner PRD) were 6.0, 10.2 and 14.2 ppbv/ppbv, respectively. These values are comparable to those

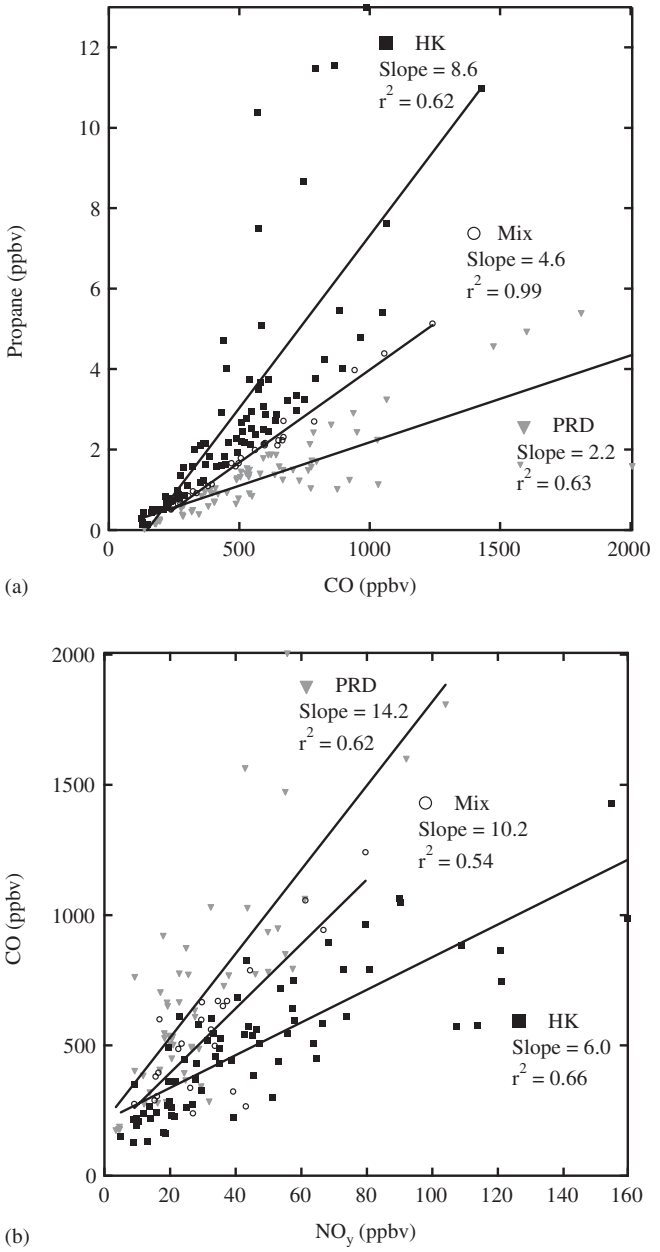

Fig. 2. Scatter plots for CO versus propane (a), and NO_y versus CO (b). Here, filled squares represent HK air masses, gray triangles represent PRD air masses and open circles represent mixed air masses.

obtained by Wang et al. (2003a) (~6, 9.2, ~21 ppbv/ppbv for HK, mixed and inner PRD, respectively), Lind and Kok (1999) (3.3 ppbv/ppbv for urban HK plumes and 14 ppbv/ppbv for plumes from inner PRD city) and those derived from emission inventories developed by Streets et al. (2003).(An overall ratio of 14 ppbv/ppbv for inner PRD and 1.4 ppbv/ppbv for HK. For transportation sector, the ratio is 20 ppbv/ppbv for inner PRD and 3.4 ppbv/ppbv for HK.) These results suggest that the segregation of air masses at Tai O using propane/CO ratios is reasonable. A comparison with the segregation result using local winds at Tai O indicates that the chemical approach better captured the characteristics of air masses from HK and the inner PRD.

To examine the chemical composition of air masses designated as local HK and inner PRD

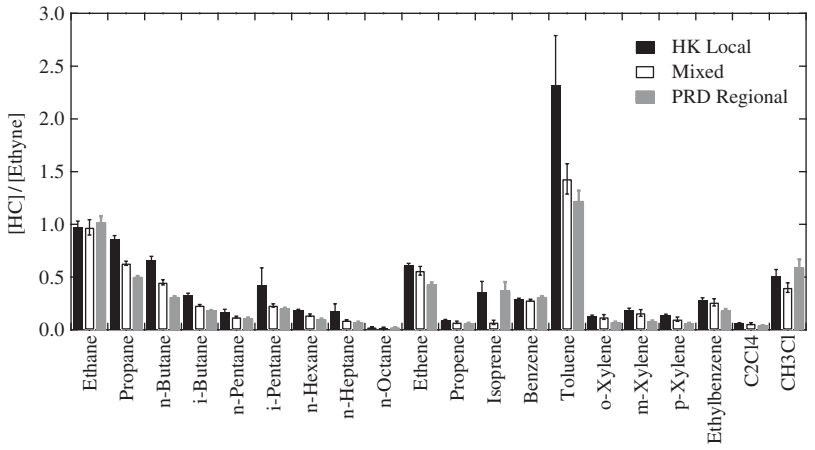

Fig. 3. Comparison of NMVOC distributions in HK (local) and the inner PRD (regional).

cities, the ratios of major individual NMVOCs to ethyne are shown for the local, mixed and regional groups (Fig. 3). Since ethyne is emitted from vehicular exhaust in urban areas and has a relatively low photochemical reactivity compared to most of the other trace gases listed in Fig. 3 (lifetime of 2–3 weeks), the ratio is suitable to understand the compositions of NMVOCs in different regions and the impact of vehicular exhausts on the NMVOC concentrations. The results indicate that the propane/ethyne and *n*-butane/ethyne ratios in HK (0.85 ± 0.04 and 0.66 ± 0.04, respectively) were significantly higher than those in the inner PRD (0.50 ± 0.02 and 0.31 ± 0.02, respectively). Statistical analyses show that there was a significant difference for propane and *n*-butane between HK and the inner PRD ($p < 0.001$) but not for ethyne ($p > 0.05$), indicating the enrichment of propane and *n*-butane in HK. We also found that HK had a much higher toluene/ethyne ratio than the inner PRD. This is due to the use of toluene-rich fuels or solvents in HK, as the toluene level in HK is significantly higher than in the inner PRD ($p < 0.01$). This is consistent with results obtained during a previous study in HK (Sin et al., 2000).

3.3. Sources of NMVOCs in inner PRD and HK

In this section, we discuss source identifications of NMVOCs from the inner PRD and HK, respectively. A PCA was performed using the classified PRD and HK data. Before applying PCA to the datasets, sensitivity analyses were conducted. Detailed information about the sensitivity analyses can be found elsewhere (Guo et al., 2004a, b). In general, suspected outliers were taken out from the dataset one at a time until a stable PCA solution was achieved, in the sense that the random exclusion of further samples had very little effect on the solution and on the interpretation of the principal components as actual physical/chemical sources (Swietlicki et al., 1996). Three out of 82 local samples and two of the 68 regional samples were removed. In addition, 21 species out of the 78 measured NMVOCs were chosen for PCA/APCS analysis since they were the most abundant measured trace compounds and are typical tracers of various emission sources. Four and three factors were extracted from the classified data for HK and PRD, respectively (Tables 3 and 4).

In the HK air-mass group, four factors were identified which explained 69%, 8%, 7% and 5% of the data variance (Table 3). The species in F1 poorly correlated with the combustion tracer CO suggesting that F1 was not related to vehicular emissions. In F1, high factor loadings of propane and butanes were found, but the loadings were still lower than those in F3, indicating the LPG and natural gas (NG) sources of these species (Na and Kim, 2001). High factor loadings of *n*-hexane, *n*-heptane, *n*-octane and BTEX (benzene, toluene, ethylbenzene and xylenes) in F1 represent the solvent use (Borbon et al., 2002). It should be noted that BTEX could be emitted from either vehicular exhaust or solvent use, and this can be evaluated using their correlation with the combustion tracer CO. We also found that the industrial/urban tracer C_2Cl_4 (Blake et al., 1997) had a high factor loading in F1. Furthermore, ethene and ethyne were also correlated with F1, despite higher loadings in F2. In addition to

Table 3
PCA results for NMVOCs measured in air masses originating from HK

	Factor			
	F1	F2	F3	F4
Carbon monoxide		0.72		
Ethane		0.84		
Propane	0.50		0.72	
n-Butane	0.51		0.76	
i-Butane	0.56		0.71	
n-Pentane				0.77
i-Pentane				0.98
n-Hexane	0.78			
n-Heptane	0.76			
n-Octane	0.64			
Ethene	0.53	0.64		
Propene			0.61	
Ethyne	0.58	0.69		
Benzene	0.69	0.55		
Toluene	0.56			0.77
o-Xylene	0.86			
m-Xylene	0.81			
p-Xylene	0.84			
Ethylbenzene	0.89			
Tetrachloroethene	0.79			
Methyl chloride				
Initial eigenvalue	15.18	1.86	1.44	1.03
% of variance	68.99	8.48	6.53	4.67
Cumulative %	68.99	77.46	83.99	88.66
Sources	Industrial emissions	Vehicular emissions	Commercial/domestic LPG/NG use	Gasoline evaporation

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Only factor loadings with an absolute value $\geqslant 0.50$ listed. Only factors with eigenvalue $\geqslant 1.00$ shown.

emission from combustion sources, ethene is a widely used synthetic chemical and is used to produce the plastics, polythene, polychloroethene, and polyvinyl chloride, whereas ethyne is used in the manufacture of the synthetic rubber neoprene, and in oxyacetylene welding and cutting. The possible sources included a petroleum storage base to the northeast of the site. They might also originate from industrial processes in Shenzhen, a large city in mainland China which is upwind of HK (Fig. 1). Based on the above analysis, F1 is most likely associated with industrial emissions.

The pollutants associated with F2 were predominantly of vehicular origins because CO, ethane, ethene, ethyne and benzene had high factor loadings in F2, and these gases are mainly emitted from vehicular exhaust (Nelson and Quigley, 1984; Wadden et al., 1986).

In F3, high factor loadings were found for propane, *n*/*i*-butanes and propene. These compounds are mainly released from the use of LPG or NG (Na and Kim, 2001). It is noteworthy that there was also a LPG/NG source in F1. This implies that two different LPG/NG sources were extracted from the HK dataset. The source in F1 had a similar emission pattern to industrial sources of solvent use. Thus, the LPG/NG use in F3 was probably related to commercial and domestic usage.

n/*i*-Pentanes were found to highly correlate with toluene in F4. *n*/*i*-Pentanes are known to be tracers of gasoline evaporation (Morikawa et al., 1998). Aromatics are also emitted by gasoline evaporation. Thus, gasoline evaporation is the main contributor to F4.

It is noteworthy that the factor loading of the biomass burning tracer CH_3Cl was below 0.5 for all four factors, indicating that biomass burning is not a major source in HK. This is consistent with highly urbanized nature of the city.

Table 4
PCA results for NMVOCs measured in air masses originating from the inner PRD

	Factor		
	F1	F2	F3
Carbon monoxide	0.60		
Ethane	0.58		0.55
Propane	0.73	0.52	
n-Butane	0.74	0.56	
i-Butane	0.75	0.57	
n-Pentane	0.86		
i-Pentane	0.77	0.55	
n-Hexane		0.85	
n-Heptane		0.85	
n-Octane	0.61	0.65	
Ethene	0.86		
Propene	0.92		
Ethyne	0.57	0.66	
Benzene	0.66	0.57	
Toluene	0.54	0.82	
o-Xylene	0.85		
m-Xylene	0.90		
p-Xylene	0.83		
Ethylbenzene		0.90	
Tetrachloroethene		0.86	
Methyl chloride			0.89
Initial eigenvalue	15.55	1.83	1.26
% of variance	74.04	8.72	6.01
Cumulative %	74.04	82.76	88.77
Sources	Vehicular emissions	Industrial emissions	Biomass burning

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Only factor loadings with an absolute value $\geqslant 0.50$ listed. Only factors with eigenvalue $\geqslant 1.00$ shown.

In the inner PRD air-mass group, three factors were extracted which explained 74%, 9% and 6% of the data variance (Table 4). High factor loadings of CO, ethene, propene, ethyne and BTX indicate the dominance of vehicular emissions in F1 (Nelson and Quigley, 1984; Wadden et al., 1986). Different chemical compositions between vehicles from HK and the inner PRD indicate that different vehicle source profiles exist in these two areas. This is true due to higher fraction of LPG/diesel cars in HK as stated in Section 3.2.

n-Hexane, *n*-heptane, *n*-octane, BTE and C_2Cl_4 were correlated in F2 indicating solvent usage (Borbon et al., 2002; ARB (Air Resources Board), 1991). In addition, C_3-C_4 alkanes and *i*-pentane were associated with F2 (though to a weaker degree than for F1). As mentioned above, C_3-C_5 alkanes are known to be emitted from the petrol-related industry, i.e. gasoline evaporation and generation of LPG/NG. The combustion tracer ethyne, also used in the manufacture of the synthetic rubber neoprene, had a high factor loading as well. Therefore, F2 is dominated by industrial sources with influence from LPG/NG emissions.

CH_3Cl had a high factor loading in F3. Since CH_3Cl is mostly emitted from biomass burning in the countryside in China, and activities associated with biomass burning were frequently observed during our site visits in the region, we concluded that biomass burning dominated F3. This can be further confirmed by the high factor loading of ethane as it is also emitted from biomass burning (Rudolph, 1995).

Sources of NMVOCs in HK differ from those in the inner PRD. Although industrial and vehicular emissions were important sources in HK and PRD, biomass burning made contributions to the atmosphere in the PRD but in HK. Furthermore, LPG/NG usage in HK could more significantly influence the abundance of NMVOCs at the receptor site.

3.4. Source apportionments of NMVOCs in HK and inner PRD

Source apportionments of NMVOCs from HK and the PRD were achieved by running the receptor model on the extracted components in Tables 3 and 4, respectively. The relative contributions of individual sources to each NMVOC were quantified using the software package of SPSS. The results are presented in Tables 5 and 6, respectively. In APCS, source contribution estimates can be negative (Miller et al., 2002). In this study, the number of factors was chosen such that negative values in the APCS source profiles, if any, were relatively small.

Table 5 shows that for air masses from HK, vehicular emissions made important contributions to most of the NMVOCs in the atmosphere. In particular, 33–62% of BTEX, 24–82% of C_2-C_8 alkanes, 66% of ethene and 69% of ethyne levels were attributed to vehicular emissions. On the other hand, 31–48% of C_6-C_8 alkanes, 28–51% of BTEX, 26–31% of C_2-C_4 alkanes and 57% of C_2Cl_4 were accounted for by industrial sources in HK. We noted that 15% of C_2Cl_4 was assigned to LPG/NG usage and 23% to vehicular emission although the industrial/urban tracer C_2Cl_4 had a poor correlation with these two sources. This is probably due to the fact that relatively long-lived C_2Cl_4 was also mixed with other urban sources during its transport to the receptor site. We also found that around 24% of C_2-C_4 alkanes were due

Table 6
Source apportionment of NMVOCs for air masses originating from the inner PRD

Compound	F1	F2	F3	R^2
Ethane	21±5	12±2	67±5	0.86
Propane	31±6	28±5	41±3	0.90
n-Butane	35±7	34±5	32±2	0.91
i-Butane	35±7	34±5	31±2	0.95
n-Pentane	56±11	32±5	12±1	0.89
i-Pentane	38±7	34±5	28±2	0.94
n-Hexane	18±4	50±9	32±2	0.92
n-Heptane	21±4	59±10	20±1	0.88
n-Octane	26±5	34±6	40±3	0.88
Ethene	39±8	21±4	40±3	0.96
Propene	56±12	4±1	40±3	0.90
Ethyne	21±4	31±5	48±3	0.95
Benzene	30±6	33±5	37±2	0.87
Toluene	29±6	55±9	16±1	0.97
o-Xylene	58±13	32±6	10±1	0.86
m-Xylene	66±15	28±5	6±0.4	0.91
p-Xylene	74±18	53±11	-27±2	0.95
Ethylbenzene	25±6	74±13	1±0.1	0.96
Tetrachloroethene	7±1	59±10	34±2	0.79
Methyl chloride	2±0.4	8±1	90±6	0.88
Sources	Vehicular emissions	Industrial emissions	Biomass burning	

Table 5
Source apportionment of NMVOCs from air masses originating from HK (average percentage ± standard error percentage)

Compound	F1	F2	F3	F4	R^2
Ethane	14±6	82±14	4±3	0.1±0.1	0.82
Propane	26±12	47±8	23±16	4±4	0.94
n-Butane	29±13	42±7	25±18	5±4	0.95
i-Butane	31±14	40±7	23±17	6 ±5	0.94
n-Pentane	26±12	37±6	15±11	22±21	0.96
i-Pentane	7±3	24±4	4±3	65±64	0.95
n-Hexane	44±20	34±6	15±11	7±7	0.93
n-Heptane	48±22	31±5	10±7	11±11	0.81
n-Octane	31±14	50±9	10±7	9±9	0.83
Ethene	21±9	66±11	11±8	2±2	0.93
Propene	23±10	57±10	17±12	3±3	0.84
Ethyne	22±10	69±12	7±5	2±2	0.93
Benzene	28±12	62±11	7±5	4±3	0.92
Toluene	36±16	34±6	7±5	23±22	0.97
o-Xylene	46±21	43±7	7±5	4±4	0.90
m-Xylene	47±21	33±6	14±10	6±5	0.91
p-Xylene	50±23	34±6	13±10	3±2	0.90
Ethylbenzene	51±23	35±6	10±7	4±4	0.95
Tetrachloroethene	57±26	23±4	15±11	5±5	0.78
Sources	Industrial emissions	Vehicular emissions	LPG/NG use	Gasoline evaporation	

to the use of LPG/NG, and 65% of *i*-pentane was explained by gasoline evaporation. In Table 5, R^2 represents the correlation between measured concentrations and model-derived values. Most of the NMVOCs had a R^2 greater than 0.80, indicating a good fit between observed and calculated concentrations.

In Table 6, vehicular emissions in the inner PRD accounted for 18–56% of C_2-C_8 alkanes, 39–56% of C_2-C_3 alkenes, 25–74% of BTEX and 21% of ethyne, whereas 34–59% of C_6-C_8 alkanes, 28–74% of BTEX and 59% of C_2Cl_4 were attributed to industrial sources. The rest of the C_2Cl_4 (34%) was observed in the biomass burning sector. Again, although C_2Cl_4 is industrial in origin, this is most likely because of its mixing with biomass burning plumes during transport to the receptor site. The contribution of biomass burning to individual NMVOC levels showed large variations ranging from 0% to 90%, including 67% for ethane, 48% for ethyne and 90% for CH_3Cl. It is surprising that much of the C_2-C_4 alkanes were associated with biomass burning. This contribution is probably overestimated because the biomass burning plumes could mix and co-locate with species from other sources before being sampled. In this case, it is likely that the biomass burning air masses were affected by LPG/NG emissions.

Based on the contributions of each source to individual NMVOC levels, the total NMVOC concentration from each source was derived. Consequently, the contribution of each source to the total NMVOCs in the atmosphere was estimated. Fig. 4 shows the mass contribution of each source to the total NMVOCs in HK urban air and in the inner PRD. The unit of NMVOC concentration was converted to μgm^{-3}. It is assumed that for the factors with mixed sources, the tracers for a source were assumed to be exclusively emitted from this specific source and their contributions were assigned to this source. For example, C_2Cl_4 in F2 and F3 was exclusively allotted to the industrial emissions in Table 3. It is also assumed that contributions from similar sources were summed and represented a single source. For instance, species emitted from different industrial sources in F1 in Table 3 were all treated as industrial sources.

About 39% of the total NMVOCs in HK air masses were attributed to vehicular emissions, whereas industrial emissions accounted for 35% (Fig. 4). Moreover, the contributions of gasoline evaporation and domestic/commercial use of LPG/

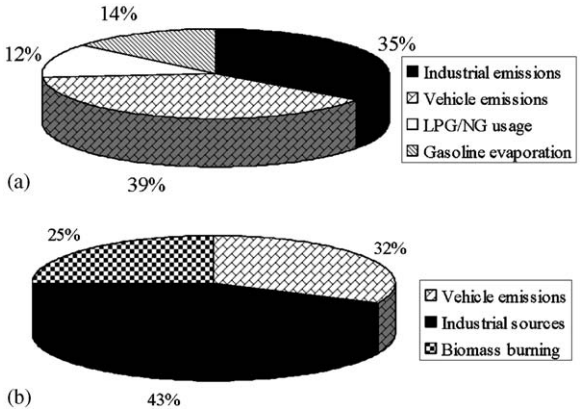

Fig. 4. Source contribution to ambient NMVOCs in HK (a) and PRD (b).

NG were 14% and 12%, respectively. We compared the result from this study with the emission inventory developed by Hong Kong Environmental Protection Department (CH2M, 2002), and with our estimates at two urban sites in HK (Guo et al., 2004a). In the inventory, the contribution of vehicles to NMVOC levels in HK was 25%. In contrast, our estimates at HK urban sites showed that 39–48% of ambient VOCs were from vehicular emissions and 11–19% from LPG/NG use. The present results were higher than the inventory but very close to those based on the HK urban data using the same receptor model, suggesting consistency of the results from the PCA/APCS analysis of two different datasets. The source contributions obtained in this study were also compared with Streets' emission inventory. In the inventory, 22% of NMVOCs were attributed to transport and 47% to industry in HK. Similarly, the Streets' inventory estimate for vehicular emissions was lower than that derived in this study.

The inconsistency between the receptor analysis and emission inventories could reflect inaccuracies in the gross inventories, uncertainty in the receptor model, or bias in the sampling strategies. The inventory was based on emissions data obtained almost exclusively for sources operating in the United States and Europe and may be inappropriate for sources operating in China. Furthermore, problems were found in activity reporting or inaccuracies in fuel characteristics after applying the inventory to both surface and aircraft measurements (Carmichael et al., 2003). Thus, the inventories for many species particularly NMVOCs are

sometimes highly uncertain (Streets et al., 2003; Carmichael et al., 2003).

Sampling strategies may also affect the source apportionment, as the results of any PCA depend on the averaging time period taken for the basic data elements, the number of pollutants and data points included (Derwent et al., 1995). Choice of particular sampling time, sampling duration and sampling frequency may emphasize or exclude specific emission sources (Derwent et al., 1995). As stated in Section 2.2, nocturnal samples were not taken in this study which could bias the source contribution because of the difference in chemistry and transport between daytime and night-time. Moreover, most samples were taken between August and November, during which time open burning of agricultural residues in rural areas is more frequent than in other months.

Under certain circumstances, the receptor model itself may generate uncertainty in the source apportionment. The model may not be able to separate sources that are strongly correlated, such as the industrial emissions in Table 3 in which LPG/NG usage was co-located with solvent use. However, this would not cause a large uncertainty in this study as we treated both sources as industrial emissions. Uncertainty may also be generated if the collected air samples are not well mixed with species from different sources. To maximize the representativeness of air samples, we took 5–7 samples on individual sampling days. If the weekly weather forecast showed similar weather conditions on the consecutive days, one of the days was chosen for air sampling to minimize the daily variation. If only one sample was taken per day, the sampling time was usually at noon when intense daytime convective mixing occurred. In addition, some very short-lived species such as isoprene could hamper the model since they may be lost in photochemical reactions during the transport to the receptor site. In this study, most NMVOCs selected for source apportionment had relatively long lifetimes considering the distance between source regions and the receptor site, and those species with short lifetimes had very low mixing ratios compared to the relatively long-lived species. Thus, we believe the uncertainty caused by the receptor model has been minimized.

Concerning the source apportionment results for the PRD air mass, approximately 32% of the total NMVOCs were attributed to vehicular emissions, 43% to industrial emissions and 25% to biomass burning (and still as high as 17% if we assume that all C_2-C_4 alkanes were emitted from LPG/NG leakage in F3 in Table 6) (Fig. 4). The vehicular contribution was lower than those in the HKEPD and Streets' inventories (58.5% and 64%, respectively), whereas the estimated biomass burning was much higher than the inventories (3–6% for biomass burning) (CH2M, 2002; Streets et al., 2003). Biomass burning may be not a major source of air pollution within the industrialized PRD, but could be important in rural areas surrounding the PRD during the dry autumn/winter season. To investigate whether the signals of biomass burning in PRD were strong, the time series of factor scores were explored for the extracted biomass burning factor (F3, Table 4) (Fig. 5) and global fire maps (Fig. 6). Since factor scores are related to source contributions, higher factor scores of air samples indicate the higher contribution of biomass burning source in the samples. The highest factor scores were observed in October–November 2001 and September and December 2002 (Fig. 5), suggesting the frequent occurrence of biomass burning in these months. The global fire maps in these months further support our observation. The fire maps were generated on a monthly basis using the standard MODIS MOD14 Fire and Thermal Anomalies Product (Justice et al., 2002). As examples, Fig. 6 shows the fire maps in November 2001 and July 2002. It was found that there were more fires in dry season (November) than in wet season (July) in the PRD and the larger southern China. Since most air samples in this study were collected between September and December, the high contribution of biomass burning is expected. It should be emphasized that although most samples were collected during months with a high occurrence of biomass burning, the contribution of biomass burning was still low (25%) compared to that of vehicular

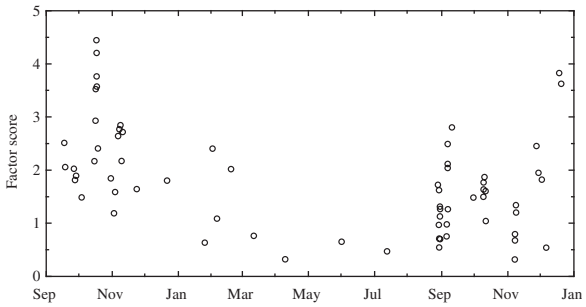

Fig. 5. Time series of factor scores for the biomass burning source from the inner PRD.

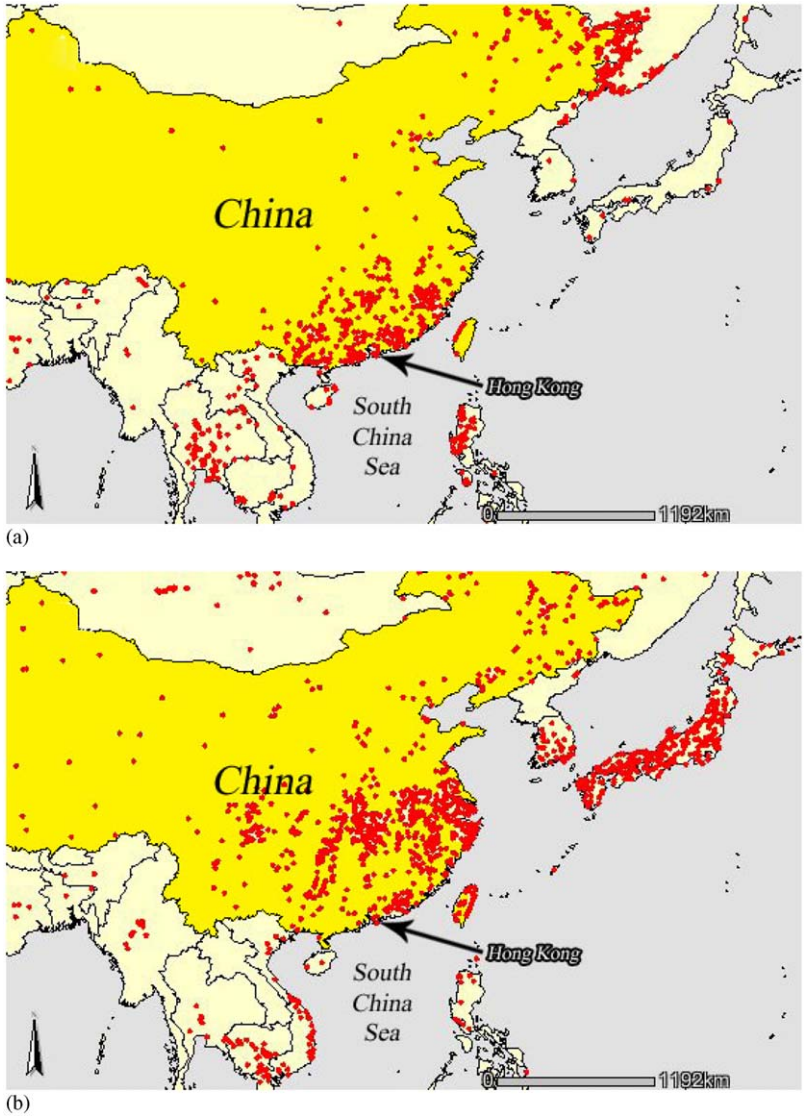

Fig. 6. Global fire maps in November 2001 (a) and July 2002 (b). This is based on the MODIS MOD 14 product.

emissions plus industrial sources (75%). Moreover, most of the reactive species were mainly emitted from these two sources, indicating the predominance of anthropogenic sources in the inner PRD.

4. Summary and conclusion

In this study, we analyzed a comprehensive dataset of NMVOCs collected at a polluted rural/coastal site in southern China. The bulk air samples were classified into inner PRD regional and HK local air masses based on the ratios of propane to carbon monoxide. A PCA/APCS receptor model was then applied to the classified data points to identify the major sources of NMVOCs and to estimate the source contributions to NMVOC mixing ratios. The modeling results indicate that the industrial emissions are the most important sources of NMVOCs in inner PRD region whereas vehicular emissions predominate in HK urban area. In HK, 39% of the total NMVOC in the atmosphere is attributed to vehicular emissions whereas 35% is explained by industrial emissions. In addition, gasoline evaporation and commercial/domestic LPG/NG use account for 14% and 12%, respectively. On the other hand, in the inner PRD region, the industrial emissions explain 43% of the total NMVOC, followed by vehicular emissions

(32%) and biomass burning (25%). The discrepancy between the predicted source contributions in this study and the existing emission inventories suggests the needs of further studies to refine the emission inventories of NMVOCs, using more accurate emission data (emission factors and activity) and to improve the source apportionment results.

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