



Technical Note

Characteristics and Source Apportionment of Ambient Volatile Organic Compounds in a Science Park in Central Taiwan

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ABSTRACT

Air samples were collected concurrently at four sites using stainless steel canisters in a science park in central Taiwan. The airborne volatile organic compounds (VOCs) were then analyzed using a gas chromatograph and a mass spectrometer (GC/MS). Eighteen volatile organic compounds (C₁–C₈) in six hydrocarbon groups were identified. Measurements reveal that the three dominant species were 2-butanone (8.60 ± 0.98 ppbv), toluene (6.13 ± 1.67 ppbv), and acetone (6.03 ± 2.79 ppbv), while most other species were present at a concentration of below 1.00 ppbv. On average, the most abundant hydrocarbon group was ketones (56.95%), followed by aromatic compounds (27.75%), alkanes (8.33%), fluoroalkanes (3.40%), chloroalkanes (2.47%), and nitrile compounds (1.10%). Principal component analysis (PCA) identified two components PC1 and PC2. Ten species in PC1 and eight species in PC2 had loadings of greater than 0.8, suggesting that the emission sources of PC1 were related to high-tech industries and traffic, and those of PC2 were related to fugitive emissions of organic solvents and refrigerants.

Keywords: Volatile organic compounds; Hazardous air pollutants; Source apportionment; PCA; Science park.

INTRODUCTION

High-tech industries, such as the integrated circuit industry, semiconductor industry, optoelectronic industry and biotech industry, are currently the main economic activities. Along with the growth of high-tech industries, they bring us all kinds of chemicals, including silanes, silicon chemicals, halogenated chemicals, inorganic acids, caustics, and volatile organic compounds (VOCs). Similar situations can be found in other industrial and developing countries (Su *et al.*, 2006; Cai *et al.*, 2010; Leuchner and Rappengluck 2010; Tiwari *et al.*, 2010).

Many works have also demonstrated that oil refineries, chemical and plastic factories and semiconductor manufacturers are major sources of VOCs in the atmosphere (Nikolaou *et al.*, 2002; Khwaja and Narang, 2008; Nian *et al.*, 2008; Cai *et al.*, 2010; Leuchner and Rappengluck, 2010; Yuan *et al.*, 2010; Vega *et al.*, 2011). In addition, many VOC species are also precursors of ground level ozone formation and they are mainly contributed from on-road mobiles and industrial factories as to worsen the air quality and reduce atmosphere visibility (Zhang *et al.*, 2009; Vardoulakis *et al.*, 2011; Lin *et al.*, 2012; Peng *et al.*, 2013).

Studies have shown that toluene to benzene (T/B) ratio of approximately 2 is a good predictor of traffic exhaust (Tang *et al.*, 2008; Civan *et al.*, 2011; Wang *et al.*, 2012); meanwhile, the T/B ratio increases with the emission strength of industrial source (Peng *et al.*, 2013).

US EPA (1990) showed that ambient VOCs attribute to 35–55% of the outdoor air cancer risks in the United States. According to the “Hazardous Air Pollutants (HAPs) List”, published by US-EPA (1996), approximately 70% of 189 HAPs are VOC species. The emissions of these species expose workers or nearby residents to risks associated with mutagens and carcinogens (Liao *et al.*, 2004; Ladou and Bailar, 2007) and should be regulated and controlled carefully.

As part of the implementation of the “Silicon-Island Policy”, Taiwan’s government has established many science parks in Taiwan. However, the impact of the emissions of VOCs, acidic and alkaline gases from the high-technology factories in the environment and human health is a matter of serious public concern. Monitoring programs and emission controls for VOCs and HAPs at the science park are important, but few relevant measurements have been made.

This work is part of an integrated research that comprises three sub-projects to study the emissions of HAPs from the science park in central Taiwan, and their associated health risks. This paper identifies the VOC species and their concentrations in the atmosphere around the science park. Principal component analysis (PCA) method is conducted to identify potential sources of the detected species.

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METHODS

Sampling and Analysis

The science park studied herein is in Houli in Taichung City in central Taiwan (latitude 24.317–24.323 °N, longitude 120.718–120.737 °E) near Feng-yuan city; it has an area of 134.4 hectares. The third and fourth phases of its development began in 2008 to attract semiconductor, optical-electrical and fine machinery factories. During the study period in March 2012, only two plants A and B were successfully operating while others were still under construction. Plant A mainly produced DRAM (Dynamic Random Access Memory), and plant B mainly produced μ -Si tandem solar PV modules. The allowable emissions for VOCs issued by the Environmental Protection Bureau of Taichung City were 18.10 ton per year for Plant A and were 2.58 ton per year for plant B. According to the emission inventory of TEDs-8.1 (2014), the emissions of NMHCs in Houli were about 17 tons per year, and those of traffic sources were about 60.6 tons per year. The concentration of NMHCs at the Houli air-quality station operated by Taiwan-EPA was 450 ppb during the sampling period, indicating that the influence of ambient concentration was primarily due to local traffics.

Airborne VOCs were collected concurrently at four sites S1, S2, S3, and S4 in the science park on March 22 in 2012 (Fig. 1). Site S1 was 960 m northwest of Plant A; S2 was 680 m northeast of Plant A; S3 was 180 m southeast of Plant A, and S4 was 50 m southwest of Plant A. The weather was fairly well during the sampling period. Air temperature was 21.5–22.4°C, pressure was about 1002.8 hPa, and relative humidity was 57–62%. A southerly wind prevailed at speed of 0.9–1.4 m s⁻¹.

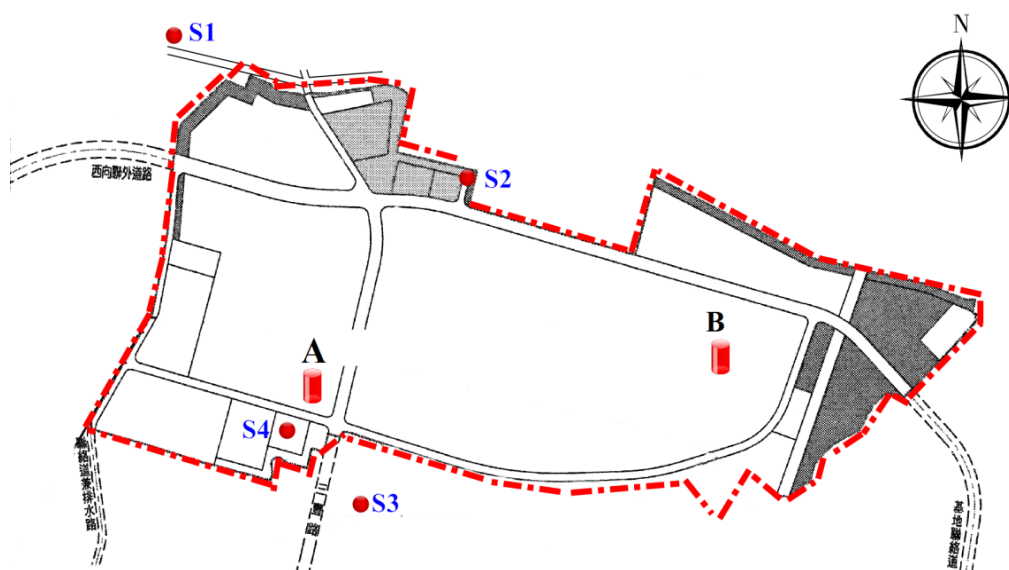
In compliance with the procedures of Taiwan-EPA NIEA-A715.14B, which are equivalent to those of the Compendium Method TO-15 (US-EPA, 1999), air samples

were collected for 2 hr at a flow rate of 40 mL min⁻¹ using 6 L stainless steel canisters (Silonite Summa Style, EN TECH, USA). Before sampling, all canisters were cleaned, moisturized, and checked for leaks to ensure a vacuum pressure of less than 10⁻² mm Hg. The air samples were then analyzed using a gas chromatograph (GC, Agilent 6890N) and a mass spectrometer (MS, Agilent 5973N). The GC oven temperature was set to 40°C initially, rising to 50°C after 2 min, and then increasing at 8 °C min⁻¹ to 230°C, which temperature was held for 10 min.

Six-point calibrations were performed for each species, yielding a linear regression with a coefficient of determination, R², above 0.995. Due to limited spaces, calibration curves or control charts for various VOC species were not presented. The detection limit (DL) for each species was implemented following US-EPA Method TO-15 65 component mix (RESTEK cat. No. 34436). Table 1 shows that the DLs for 18 species identified in this work ranged from 0.17 to 0.38 ppbv, with recovery efficiencies in 96.2–103.9%.

Principal Component Analysis

Source apportionment of VOC species was performed using principal component analysis (PCA), based on the varimax orthogonal rotation method. Data entry and analysis were done using SPSS statistical package version 17. The main purpose of the PCA is to describe the covariance relationships among many variables in terms of just a few underlying common components, which hopefully explain the most (typically 80 to 90%) of the total variance in the observed data (Johnson and Wichern, 2007). A component that is identified from PCA, typically with an eigenvalue of greater than one, exhibits a pattern of variation in response to the input parameters (which are the concentrations of pollutants herein). The correlation between the concentration of a particular pollutant and a component increases with its



A: semiconductor company; B: optoelectronics company.

Fig. 1. Four sampling sites in the science park.

Table 1. Results of quality assurance and quality control of 18 VOC species identified.

Group/Species	Recovery efficiency (%)	DL (ppbv)
<i>Alkanes</i>		
Propane (C3)	103.6	0.36
n-Pentane (C5)	96.2	0.19
Hexane (C6)	99.6	0.24
Heptane (C7)	100.0	0.23
<i>Ketones</i>		
Acetone (C3)	98.9	0.29
2-Butanone (C4)	103.9	0.23
4-Methyl-2-Pentanone (C6)	100.0	0.17
<i>Aromatic Compounds</i>		
Benzene (C6)	99.8	0.26
Toluene (C7)	98.8	0.23
Ethylbenzene (C8)	99.5	0.23
(p,m)-Xylene (C8)	98.5	0.35
o-Xylene (C8)	99.1	0.25
<i>Fluoroalkanes</i>		
Chlorodifluoromethane (C1)	99.9	0.25
Dichlorodifluoromethane (C1)	100.5	0.25
Trichlorofluoromethane (C1)	99.9	0.19
<i>Chloroalkanes</i>		
Chloromethane (C1)	100.7	0.30
Methylene Chloride (C1)	100.2	0.20
<i>Nitrile compounds</i>		
Acetonitrile (C2)	101.9	0.27

loading. The source characteristics of a component can then be inferred from the dominant pollutants in existing source profiles or the emission inventory (Derwent *et al.*, 1995; Ho *et al.*, 2002).

RESULTS AND DISCUSSION

Characteristics of Atmospheric VOCs around Science Park

Fig. 2 plots the concentrations of 18 VOC species at four sampling sites. Site S1 exhibited the highest concentrations of 2-butanone (10.30 ppbv) and toluene (8.20 ppbv), while site S2 had the highest concentration of acetone (8.50 ppbv). The concentration profiles at the four sites were clearly similar in that 2-butanone (8.60 ± 0.98 ppbv), toluene (6.13 ± 1.67 ppbv), and acetone (6.03 ± 2.79 ppbv) were the three dominant species, and the other species were minor with concentrations of less than 1.00 ppbv. These three species are commonly utilized as solvents or cleaning liquids in semiconductor plants (Pethrick and Rankin, 1998; Chan and Lai, 2010; Tsai *et al.*, 2011).

Across the four sites, on average ketones were the most abundant compounds (56.95%), followed by aromatic compounds (27.75%); and these two groups altogether contributed 84.70% of TVOC (= total concentration of 18 individual species); see Table 2. The other groups were alkanes (8.33%), fluoroalkanes (3.40%), chloroalkanes (2.47%), and nitrile compounds (1.10%). Notably, 11 species superscripted with “a” in Table 1 are characterized as HAPs by the US-EPA (1996); the percentages of these 11 HAPs in TVOC were 66.26% at S1, 59.87% at S2,

87.15% at S3, and 66.61% at S4.

The contours of TVOCs with the corresponding wind rose during the survey period is plotted in Fig. 3. TVOC was the highest at S1 (35.09 ppbv), followed by S2 (30.03 ppbv), S4 (29.23 ppbv), and S3 (14.32 ppbv). Since a southerly wind prevailed with a speed of 0.9–1.4 m s⁻¹, the downwind (northeast) sites S1 and S2 exhibited higher TVOCs than the upwind (south or southwest) sites S3 and S4.

Ratio of Toluene to Benzene (T/B Ratio)

Toluene and benzene are organic solvents commonly used in semiconductor processes (Park *et al.*, 2011; Park and Yeo, 2013), but they are also present in vehicle emissions. The percentages of benzene, toluene and hexane in TVOC at S1, S2, S3, and S4 ranged in 0.00–1.42%, 19.64–25.14%, and 0.00–1.45%, respectively, indicating that toluene was pre-dominant in this work. Table 3 compares the T/B ratios found in various studies, which were 14.8–18.9 here, consistent with other works. For example, the T/B ratio was 14.3 at the Lin-Yuan Industrial Park in Kaohsiung, 14.3 at Guangzhou (industrial) in China, and 11.3 at Bursa industrial city in Turkey. Civan *et al.* (2011) found that T/B > 10 is typically associated with an industrialized city and local sources. Table 3 also reveals that the T/B ratio was 5.2 at Tsuen Wan in Hong Kong and 7.0 at Urayasu city in Japan. Guo *et al.* (2004) and Civan *et al.* (2011) found that T/B < 10 is typically associated with a non-industrialized city where the main pollution source is traffic exhaust. Table 3 reveals that the measured T/B ratios were 2.5 to 3.5 on various road-sides or highways, where vehicle exhausts were the primary sources.

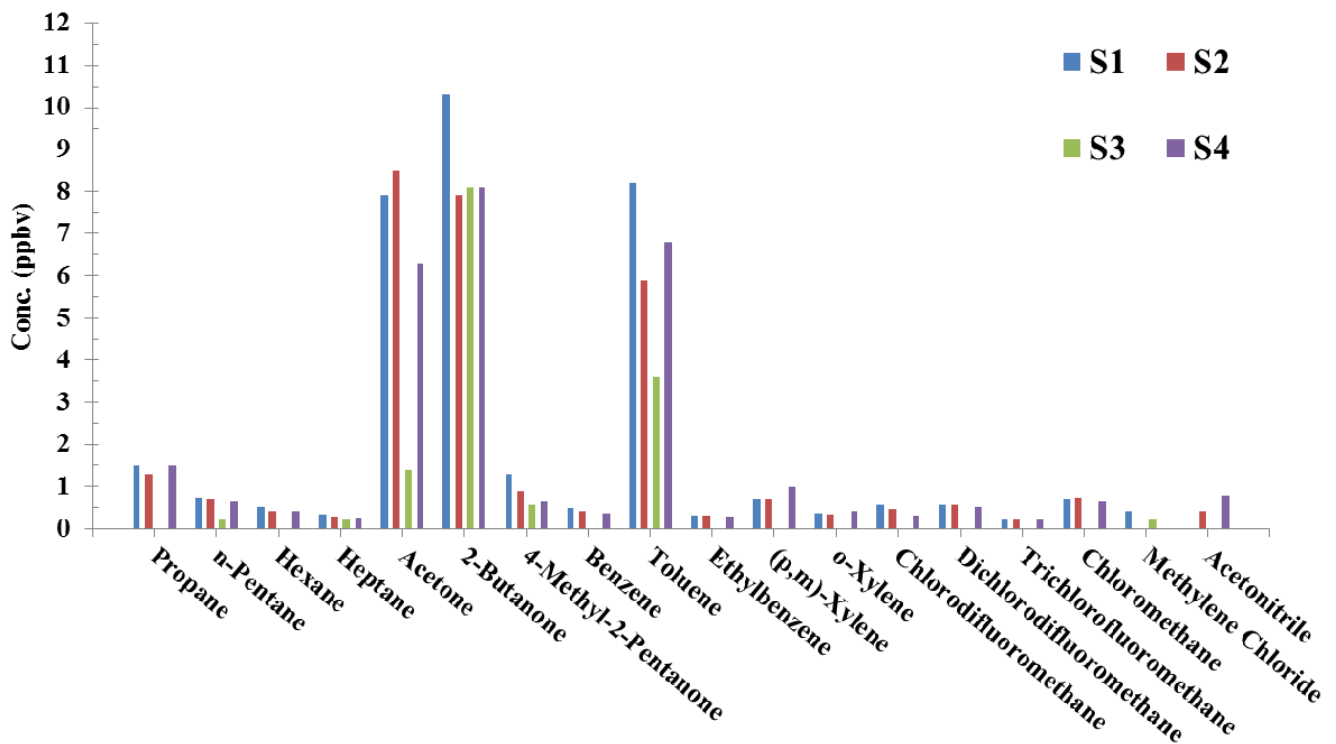


Fig. 2. Concentration profiles of 18 VOC species at four sampling sites.

Table 2. Average concentrations (ppbv) of 18 VOC species at four sites.

Group/Species	Sample site				Avg \pm S.D. (ppbv)	Group contribution (%)
	S1	S2	S3	S4		
<i>Alkanes</i>						
Propane (C3)	1.50	1.30	N.D	1.50	1.08 \pm 0.63	8.33
n-Pentane (C5)	0.74	0.69	0.20	0.63	0.57 \pm 0.21	
Hexane (C6)	0.51	0.42	N.D	0.42	0.34 \pm 0.20	
Heptane (C7)	0.34	0.29	0.24	0.27	0.29 \pm 0.04	
<i>Ketones</i>						
Acetone (C3)	7.90	8.50	1.40	6.30	6.03 \pm 2.79	56.95
2-Butanone (C4)	10.30	7.90	8.10	8.10	8.60 \pm 0.98	
4-Methyl-2-Pentanone (C6)	1.30	0.90	0.56	0.63	0.85 \pm 0.29	
<i>Aromatic Compounds</i>						
Benzene (C6)	0.50	0.40	N.D	0.36	0.32 \pm 0.19	27.75
Toluene (C7)	8.20	5.90	3.60	6.80	6.13 \pm 1.67	
Ethylbenzene (C8)	0.30	0.30	N.D	0.29	0.22 \pm 0.13	
(p,m)-Xylene (C8)	0.68	0.70	N.D	1.00	0.60 \pm 0.37	
o-Xylene (C8)	0.37	0.34	N.D	0.42	0.28 \pm 0.13	
<i>Fluoroalkanes</i>						
Chlorodifluoromethane (C1)	0.56	0.46	N.D	0.32	0.34 \pm 0.21	3.40
Dichlorodifluoromethane (C1)	0.57	0.57	N.D	0.52	0.42 \pm 0.24	
Trichlorodifluoromethane (C1)	0.23	0.24	N.D	0.22	0.17 \pm 0.10	
<i>Chloroalkanes</i>						
Chloromethane (C1)	0.68	0.72	N.D	0.65	0.51 \pm 0.30	2.47
Methylene Chloride (C1)	0.41	N.D	0.22	N.D	0.16 \pm 0.17	
<i>Nitrile compounds</i>						
Acetonitrile (C2)	N.D	0.40	N.D	0.80	0.30 \pm 0.33	1.10
TVOC	35.09	30.03	14.32	29.23	27.17 \pm 7.75	100%

Note:

1. Superscript "a" denotes to those species that are HAPs specified by the US-EPA (1996).
2. N.D. (not detected).

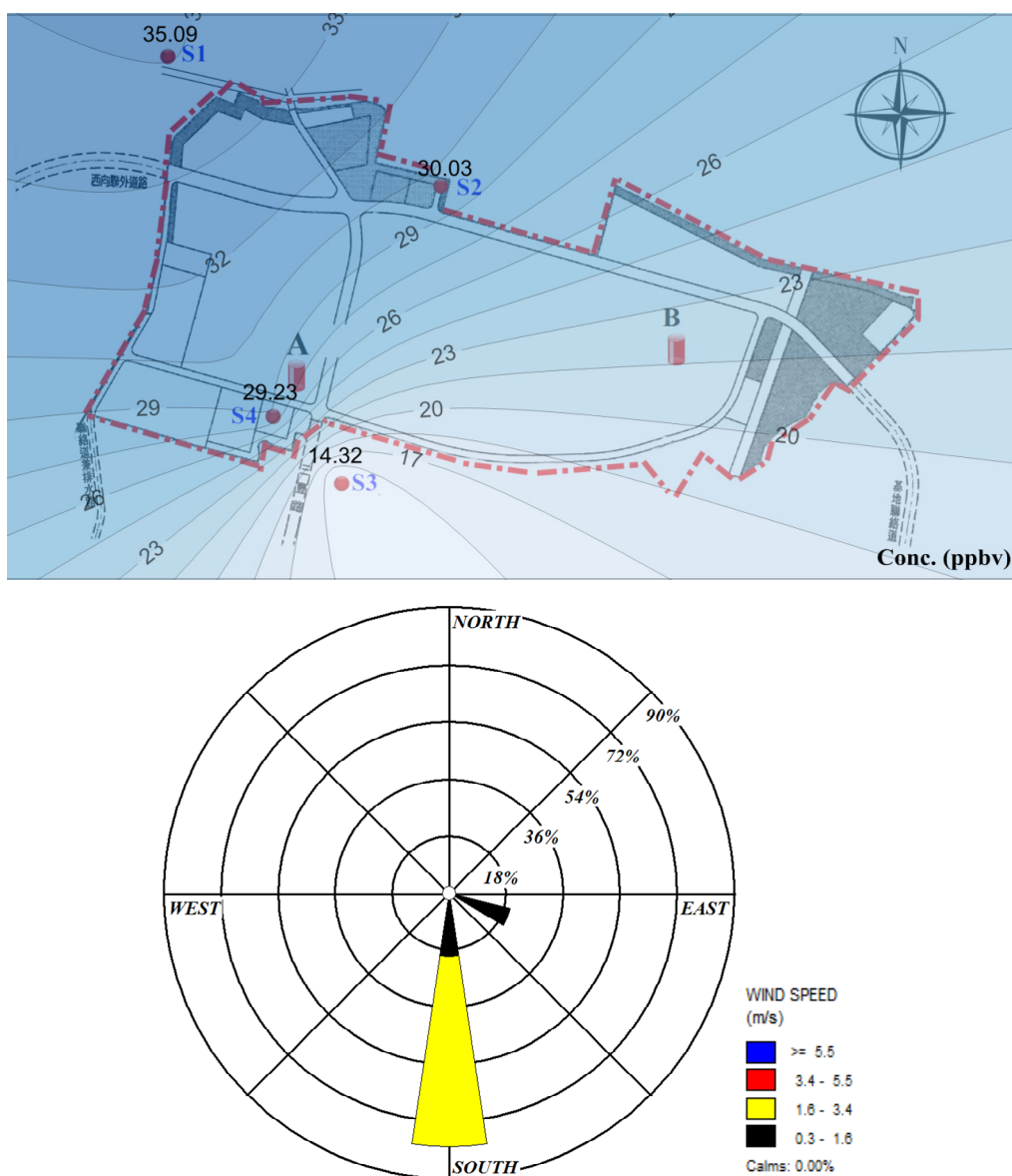


Fig. 3. Plots of TVOC contours (ppbv) with the corresponding wind rose.

Source Apportionment Using PCA

Table 4 shows loadings of 18 VOC species categorized in two components obtained from PCA. The first component PC1 is the main one, which explained the 67.37% of total variance, while the second component PC2 explained the remaining (32.63%); that is, these two factors together explained all the variances (100%).

In PC1, ten species had loadings of greater than 0.8; these included n-pentane, 2-butanone, hexane, 4-methyl-2-pentanone, toluene, heptane, benzene, chlorodifluoromethane, methylene chloride and acetonitrile. This cluster of compounds matched those of solvent profiles used in the high-tech industries, including integrated circuit, semiconductor, optoelectronic, and precision machinery industries (Lu *et al.*, 2006; Chan and Lai, 2010; Tsai *et al.*, 2011; Beyersdorf *et al.*, 2012). The aromatic VOCs (toluene, benzene) together with n-pentane and hexane also suggested

two possible sources of traffic emissions and gasoline vapor evaporations (Wang and Zhao, 2008; Chang *et al.*, 2009; Wang *et al.*, 2010; Civan *et al.*, 2011; Peng *et al.*, 2013; Fanizza *et al.*, 2014).

In PC2, another eight species had factor loadings of greater than 0.8, including propane, acetone, ethylbenzene, o-xylene, (p,m)-xylene, chloromethane, trichlorofluoromethane and dichlorodifluoromethane. Acetone is a widely-used organic solvent in chemical and semiconductor industries (Lu *et al.*, 2006; Tsai *et al.*, 2011; Lan *et al.*, 2014). Meanwhile, trichlorofluoromethane and dichlorodifluoromethane were related to the emissions from refrigerator factories (Huang *et al.*, 2012).

The PCA results discussed above suggested that the possible primary sources of PC1 were related to high-techs (e.g., semiconductor and optical-electrical industries), while the possible secondary sources of PC2 were related to organic

Table 3. Comparisons of toluene to benzene ratios (*T/B*).

	<i>T/B</i>	References
S1 (Science park)	16.4	This study
S2 (Science park)	14.8	This study
S3 (Science park)	-	This study
S4 (Science park)	18.9	This study
Taichung (Science park), Taiwan	13.8	Kuo (2007)
Hsinchu (Science park), Taiwan	13.1	Chiu <i>et al.</i> (2005)
Daliao (Industrial park), Taiwan	31.6	Chiang <i>et al.</i> (2007)
Tzoying (Industrial city), Taiwan	16.2	Chiang <i>et al.</i> (2007)
Guangzhou (Industrial city), China	14.3	Chan <i>et al.</i> (2006)
Lin Yuan (Industrial park), Taiwan	14.3	Liu <i>et al.</i> (2008)
Bursa (Industrial city), Turkey	11.3	Civan <i>et al.</i> (2011)
Dongguan (Industrial city), China	10.4	Chan <i>et al.</i> (2006)
Urayasu (Urban), Japan	7.0	Matsunaga <i>et al.</i> (2010)
Shimizu (Urban), Japan	6.3	Ohura <i>et al.</i> (2006)
Fuji, (Urban), Japan	5.8	Kume <i>et al.</i> (2008)
Tsuen Wan (Urban), Hong Kong	5.2	Guo <i>et al.</i> (2004)
Bursa Roadside, Turkey	2.5	Civan <i>et al.</i> (2011)
Nanjing Roadside, China	2.5	Wang and Zhao (2008)
Beijing Highway, China	2.5	Wang <i>et al.</i> (2012)
Guangzhou Tunnel, China	1.4	Tang <i>et al.</i> (2008)

Table 4. Factor loadings obtained from PCA.

VOC species	Factor	
	PC1	PC2
Propane		0.866
n-Pentane	0.839	
Hexane	1.000	
Heptane	0.961	
Acetone		0.967
2-Butanone	0.997	
4-Methyl-2-Pentanone	1.000	
Benzene	0.961	
Toluene	0.922	
Ethylbenzene		0.866
(p,m)-Xylene		0.837
o-Xylene		0.990
Chlorodifluoromethane	0.814	
Dichlorodifluoromethane		0.866
Trichlorofluoromethane		1.000
Chloromethane		0.997
Methylene Chloride	0.916	
Acetonitrile	0.866	
Eigenvalue	12.13	5.87
Variance (%)	67.37	32.63
Cumulative (%)	67.37	100.00
Potential Source	High-tech industries/Vehicle exhausts	Organic solvents/Refrigerators

Note: VOCs compounds with |factor loading| ≥ 0.800 listed.

solvents and refrigerators (see Table 4). Table 5 further summarized the possible pollution sources of the 18 VOC species from other studies, which included semiconductor, electronic and machinery factories, vehicles, gas stations, organic solvents, and refrigerants. Comparisons between Tables 4 and 5 indicate that the possible pollution sources suggested here were reasonable.

CONCLUSIONS

This study identified 18 volatile organic compounds (C_1 – C_8) in six hydrocarbon groups at four monitoring sites in a science park in Taichung city, central Taiwan. Eleven of these 18 VOC species are specified as HAPs by the US-EPA (1996). Measurements reveal that the three dominant

Table 5. Potential emission sources of VOCs taken from the literature.

Emission sources	VOC species	References
Vehicles and gasoline vapor	benzene, toluene, xylene, ethylbenzene, propane, n-pentane	Chiu <i>et al.</i> (2005); Chang <i>et al.</i> (2009); Civan <i>et al.</i> (2011); Fanizza <i>et al.</i> (2014); Peng <i>et al.</i> (2013); Wang <i>et al.</i> (2010) and Wang and Zhao (2008)
High-tech industries (semiconductor, optoelectronic and biotech industry), Refrigerants	acetone, hexane, toluene, propane, xylene, 2-butanone, ethylbenzene, 4-methyl-2-pentanone, chlorodifluoromethane, dichlorodifluoromethane,	Beyersdorf <i>et al.</i> (2012); Chan and Lai (2010); Lu <i>et al.</i> (2006) and Tsai <i>et al.</i> (2011)
Circuit manufacturing, Precision machinery, Organic solvents	propane, heptane, acetone, hexane, n-pentane, acetonitrile, methylene chloride	Kuo <i>et al.</i> (2007); Civan <i>et al.</i> (2011); Huang <i>et al.</i> (2012) and Lan <i>et al.</i> (2014)

species were 2-butanone (8.60 ± 0.98 ppbv), toluene (6.13 ± 1.67 ppbv), and acetone (6.03 ± 2.79 ppbv), while most other species were present at concentrations of below 1.00 ppbv. On average, the most abundant hydrocarbon group was ketones (56.95%), followed by aromatic compounds (27.75%), alkanes (8.33%), fluoroalkanes (3.40%), chloroalkanes (2.47%), and nitrile compounds (1.10%). The ratios of toluene to benzene, T/B, were in the range 14.8–18.9, indicating that airborne pollutants were associated with industry and local sources.

Source apportionment using PCA identified two source components. The primary component PC1 explained the 69.77% of total variance, while the second component PC2 explained the rest (30.23%). Ten species in PC1 and eight species in PC2 had loadings of more than 0.8. These results suggest that the emission sources of PC1 were associated with the high-tech industries (e.g., semiconductor and optical-electronics) and vehicle exhausts; and those of PC2 were associated with fugitive emissions of organic solvents and refrigerants.

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