Seasonal Variation and Source Apportionment of Atmospheric Carbonyl Compounds in Urban Kaohsiung, Taiwan

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ABSTRACT

The concentrations of 18 atmospheric carbonyls species were measured at Nan-Chie and Hsiung-Kong sites in Kaohsiung City, Taiwan, during the summer and winter of 2006. Formaldehyde and acetaldehyde were the most abundant carbonyls with respective annual mean concentrations of 17.99 μg/m^3 and 13.69 μg/m^3 at Nan-Chie, and 21.47 μg/m^3 and 16.68 μg/m^3 at Hsiung-Kong; altogether the two species accounted for approximately 56–57% of total carbonyls. In summer, the total concentrations of carbonyls were 74.06 μg/m^3 and 89.99 μg/m^3 at Nan-Chie and Hsiung-Kong, respectively. In winter, the concentrations were 37.14 μg/m^3 and 46.50 μg/m^3 at Nan-Chie and Hsiung-Kong, respectively. Measured results indicated the predominance of photolysis and photooxidation reactions of aldehydes in summer. In this study, receptor models using principal component analysis (PCA) and absolute principal component scores (APCS) suggest that the primary pollution sources at Nan-Chie in the summer were secondary emissions/vehicle exhausts (gasoline engines)/stationary emissions (food industry), stationary emissions (petrochemical)/waste treatment and restaurant emissions; the primary pollution sources at Nan-Chie in winter were vehicle exhausts (gasoline engines)/stationary emissions (petrochemical) and restaurant emissions. At Hsiung-Kong, the primary pollution sources in summer were secondary emissions/vehicle exhausts (gasoline engines and diesel engines)/stationary emissions (metal assembly), restaurant emissions and others; the primary pollution sources in winter were vehicle exhausts (gasoline engines)/restaurant emissions and vehicle exhausts (diesel engines)/stationary emissions (metal assembly).

Keywords: Carbonyl compounds; Seasonal variation; Receptor model; PCA; APCS.

INTRODUCTION

Carbonyl compounds (carbonyls), which have adverse health effects on humans (Carlier *et al*., 1986; Zhang *et al*., 1994; Hauptmann *et al*., 2004; Cerón *et al*., 2007), are emitted directly from primary sources, including exhaust gases of motor vehicles, and by the incomplete combustion of hydrocarbon fuels in industrial machinery and industrial processes (Cavalcante *et al*., 2006; Liu *et al*., 2006; Seco *et al*., 2007). Atmospheric photooxidation is an important secondary source of carbonyls, and involves a reaction of ozone with organic compounds that are associated with air pollution. Aldehydes and ketones are important products of the photooxidation of gas-phase hydrocarbons in sunlight (Hoekman *et al*., 1992; Baez *et al*., 1995; Grosjean *et al*., 1996; Katsoyiannis *et al*., 2008).

Kaohsiung City (22°38'N, 120°17'E) is a heavily industrialized and densely populated harbor city in southern Taiwan, with approximately 1.49 million inhabitants, an area of 153.6 km^2, and approximately 1,281,000 registered vehicles (380,000 cars, 886,000 motorcycles, and 15,000 trucks). Significantly, dense traffic, and intensive industrial and commercial development have caused Kaohsiung city and the surrounding Kaohsiung and Pingtung counties to have the poorest air quality in Taiwan. The quality of the air in southern Taiwan, is lowest from the late fall to the middle of spring, either because of increased ground-level concentrations of PM or ozone that is associated with unfavorable meteorological conditions (Chen *et al*., 2004).

This study presents measurements of the concentrations of 18 carbonyls species at two Kaohsiung sites in summer and winter. The seasonal variations of the atmospheric carbonyls levels were investigated. Receptor models using principal component analysis (PCA) and absolute principal component scores (APCS) were applied to estimate seasonal variations in source contributions.

EXPERIMENTAL PROCEDURES

Sampling Sites and Periods

The two sampling sites, Nan-Chie and Hsiung-Kong,

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shown in Fig. 1, are located in the northern and central parts of Kaohsiung City, respectively. As shown in Fig. 1, the Kaohsiung areas are six large industrial complexes, two in Kaohsiung City and four in Kaohsiung County. The distance between the Nan-Chie and Hsiung-Kong sites is approximately 20 km. The sampling period was two seasons (include summer and winter), from August 2006 to December 2006. Samples were obtained on three days in summer and on three days in winter. Taiwan's EPA (Environmental Protection Administration) has set up air-quality monitoring stations at the two sites. Therefore, hourly air quality and meteorological data, including temperature and winds, are available. Table 1 presents the meteorological conditions at the sampling sites, including temperature, wind speed, wind direction, period of sunshine and air pressure.
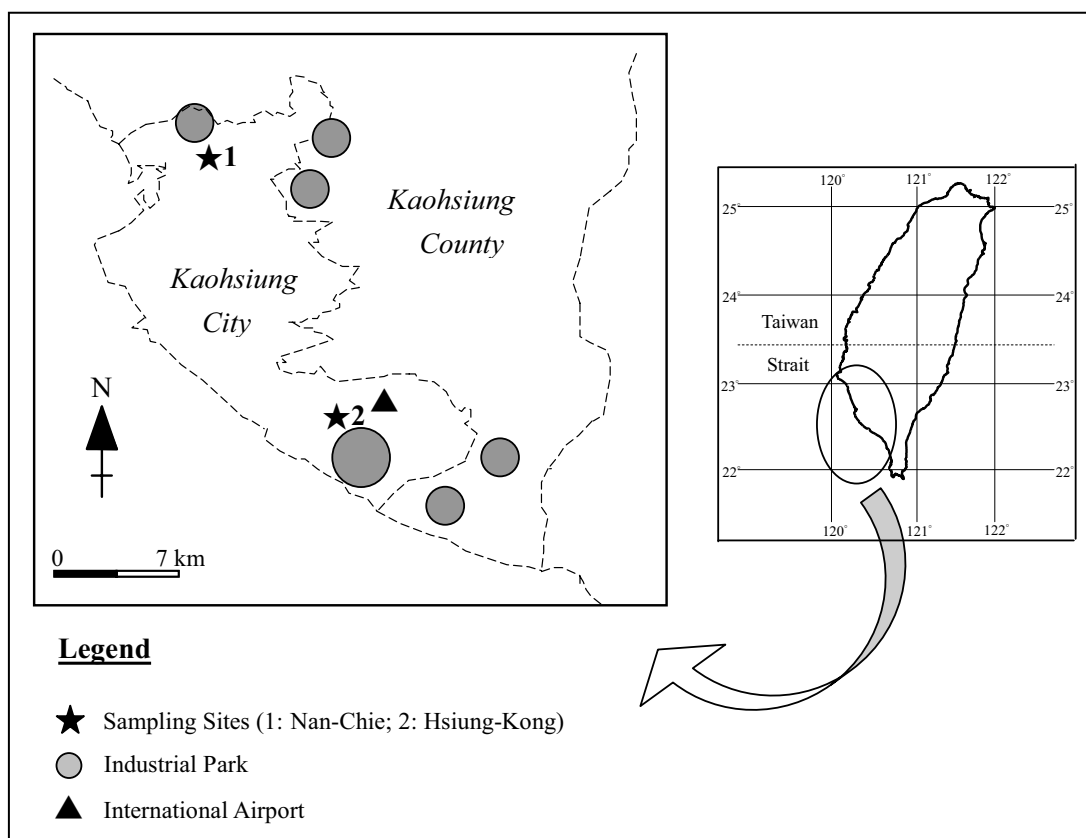

Fig. 1. Location of two monitoring sites in Kaohsiung City.

Table 1. Meteorological conditions at Nan-Chie and Hsiung-Kong sites in Kaohsiung City in 2006.

Date	Temperature (°C)	Wind speed (m/s)	Wind direction	Period of Sunshine (h)	Relative humidity (%)	Air pressure (mb)
(a) *Nan-Chie site*						
August 1, 2006	30.2	1.97	WNW	7.8	68	1006.0
August 2, 2006	29.9	2.04	W	7.7	66	1005.4
August 3, 2006	28.0	1.82	NNW	1.5	70	1007.8
December 18, 2006	16.9	1.38	ENE	7.7	60	1023.4
December 19, 2006	19.2	1.84	NW	5.7	62	1022.2
December 20, 2006	20.1	2.40	NW	5.2	71	1019.9
(b) *Hsiung-Kong site*						
August 1, 2006	31.0	2.00	W	7.8	66	1006.0
August 2, 2006	31.2	2.53	WSW	7.7	65	1005.4
August 3, 2006	29.6	2.38	SW	1.5	69	1007.8
December 18, 2006	18.6	1.65	W	7.7	55	1023.4
December 19, 2006	20.8	1.78	NE	5.7	56	1022.2
December 20, 2006	21.7	2.16	WNW	5.2	66	1019.9

Analysis

The concentrations of 18 carbonyls were measured in ambient air. They were formaldehyde, acetaldehyde, acetone, propionaldehyde, crotonaldehyde, butyraldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, *o*-tolualdehyde, *m*-tolualdehyde, *p*-tolualdehyde, hexaldehyde, cyclohexanone, heptaldehyde, octaldehyde, nonaldehyde, and decylaldehyde. Carbonyls in air were sampled for 3 h at a flow rate of 0.8 L/min using a silica cartridge that was impregnated with acidified 2,4-dinitrophenylhydrazine (Waters Sep-Pak DNPH–silica), which is very reactive toward carbonyls. The flow rate through the cartridges was measured using a rotameter before and after each sampling period. The rotameter was calibrated in the laboratory against a soap bubble flow meter. An ozone scrubber was connected before the DNPH–silica cartridge to prevent interference by ozone. The sampled cartridges were eluted slowly with 5 mL of acetonitrile (ACN) into a 5 mL volumetric flask, and stored under refrigerating conditions until use. The eluted samples were stable at 4°C for up to one month. A 10 μL aliquot was injected into the High Performance Liquid Chromatograph (HPLC) system through an auto-sampler.

Calibrations were conducted using standard solutions of five concentrations for carbonyls species, all with a coefficient of determination, R^2, above 0.999. The calibration standard was run daily to ensure that instruments were stable. In series, cartridge collection efficiency was determined using two cartridges, analyzed cartridge collection efficiency exceeded 99% for carbonyls found in the first cartridge. Second set of elution tests indicated the complete recovery of all of the carbonyls. Relative percentage differences (RPDs) for duplicate analysis were less than 5%. Method detection limits (MDLs) were determined by performing seven replicate analyses of the working standards at the lowest concentration. For sample volumes of 144 L, the MDLs of DNPH derivatives for carbonyls were found to be in the range 0.38 to 1.38 μg, with accuracies from 93.9 to 104.1%. Laboratory blank samples were prepared and analyzed; all data were corrected with reference to a blank. Recovery efficiencies of 97.8 to 114.3% were achieved.

PCA/APCS RECEPTOR MODELS

In this study, principal component analysis (PCA) was applied to identify potential sources of atmospheric carbonyls. PCA is frequently used in data reduction to identify a small number of factors that explain most of the variance observed in a larger number of manifest variables. The factors were extracted using PCA, which involves varimax orthogonal rotation to determine the eigenvalues of the variance matrix of original variables; usually, factors with eigenvalues > 1 are chosen. Once a factor is determined by PCA, it consists of patterns of variation of the factor loadings of input parameters. The correlation between a specific pollutant and a factor increases with the factor loading. The characteristics of a factor can then be inferred from the dominant pollutants, such as VOCs, PAHs and carbonyls (Derwent *et al*., 1995; Ho *et al*., 2002a; Johnson and Wichern, 2002; Chang *et al*., 2009; Wang *et al*., 2010). Notably, the factor loadings in PCA only provide the relative influences of individual pollution sources on specified pollutants.

Absolute principal component scores (APCS) were used to assess contributions of individual sources to specified pollutants (Thurston and Spengler, 1985; Swietlicki *et al*., 1996; Guo *et al*., 2004a, 2004b; Karar and Gupta, 2007; Chen *et al*., 2008; Duan *et al*., 2008; Wang *et al*., 2010). In the application of APCS, the concentration of each pollutant at each sample site was normalized with respect to its mean and standard deviation. In PCA, the new standardized variables, with zero means and unit variances, are expressed as a linear combination of pollution sources and pollutants in source profiles, which can then be determined by varimax orthogonal rotation to determine the coefficients and therefore the absolute scores (or contributions) of the pollution sources. Like all other multivariate receptor models, the PCA and APCS models need adequate source profiles and degrees of freedom enable regressions to be performed. These receptor models may not be able to separate sources that are similar. The problem of collinearity can be solved by combining similar sources of single category (Ho *et al*., 2002a; Guo *et al*., 2004b; Karar and Gupta, 2007; Chen *et al*., 2008), to help identify possible sources.

RESULTS AND DISCUSSION

Characteristics of Carbonyl Compounds in Summer and Winter

In this study, concentrations of 18 carbonyls were measured at the Nan-Chie and Hsiung-Kong sites in Kaohsiung city. Fig. 2 plots the seasonal variation of carbonyls at Nan-Chie and Hsiung-Kong sites. The average concentrations of formaldehyde and acetaldehyde at both sites were significantly high in summer. The concentration of total carbonyls at Nan-Chie was 74.06 μg/m³ in the summer and 37.14 μg/m³ in the winter, and that at Hsiung-Kong was 89.99 μg/m³ in the summer and 46.50 μg/m³ in the winter, due to the fact that summer has stronger photochemical activities than in winter. Photochemical activity and temperature are important in the creation of carbonyls at the sampling site during the summer (Cerón *et al*., 2007). Atmospheric photooxidation is an important secondary source of carbonyls, which it produces photochemically at a higher level in summer than in winter, direct vehicular emissions were the principal source of carbonyls in the winter (Possanzini *et al*., 1996; Ho *et al*., 2002b). The low concentrations of these carbonyls in the summer were due to the photolysis of these carbonyls to form hydroxyl radicals (Anderson *et al*., 1996; Christensen *et al*., 2000).

Table 2 presents a statistical summary of the carbonyls measured throughout the study period (August to December 2006). At the Nan-Chie site, the average concentration of formaldehyde was 17.99 μg/m³, and that of acetaldehyde was 13.69 μg/m³; the two species combined accounts for

approximately 57% of total carbonyls. The values at the Hsiung-Kong site were similar to those at Nan-Chie, the average concentration of formaldehyde was 21.47 μg/m^3, and that of acetaldehyde was 16.68 μg/m^3; the two species combined accounts for approximately 56% of total carbonyls. Thus, formaldehyde and acetaldehyde are the most abundant carbonyls at both sites. The third most abundant carbonyls at Nan-Chie and Hsiung-Kong were propionaldehyde (2.83 μg/m^3) and *m*-tolualdehyde (4.77 μg/m^3), respectively.

The difference between the major carbonyl species at the two sites was related to the different sources of

atmospheric carbonyls. Motor vehicle exhaust is expected to be the most important source of carbonyls in cities, which are the key compounds in the photochemical generation of air pollution (Carlier *et al.*, 1986). The overall mean concentrations of total carbonyls at Nan-Chie (55.60 μg/m^3) were approximately 22.8% lower than at Hsiung-Kong (68.25 μg/m^3), consistent with the fact that the mean traffic volume (including motorcycles, gasoline cars, and diesel trucks) at Hsiung-Kong (11,067 vehicles per day) is about 60% higher than that at Nan-Chie (6,611 vehicles per day) (Lai *et al.*, 2005).
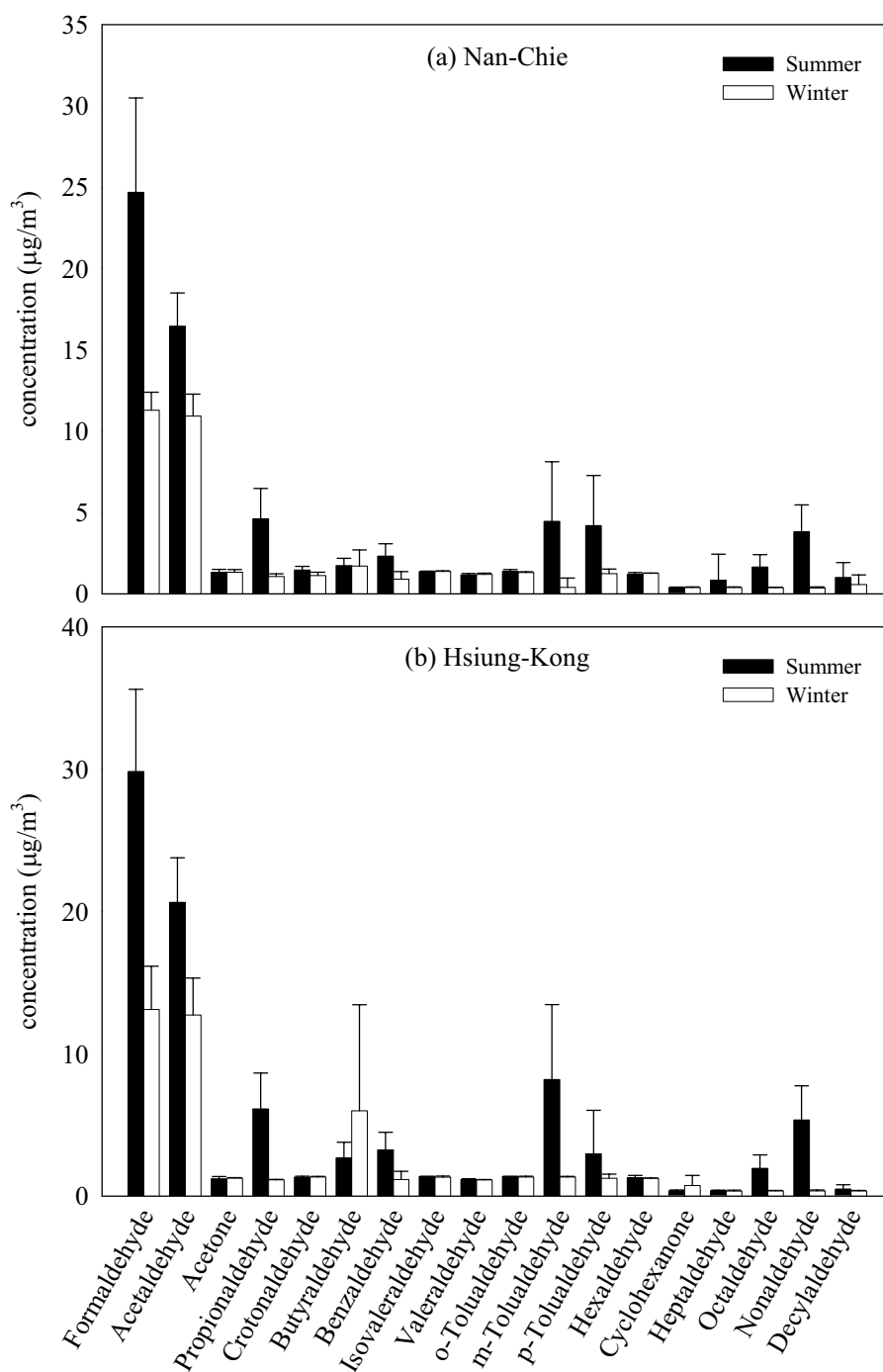

Fig. 2. Concentrations of carbonyl compounds at Nan-Chie and Hsiung-Kong sites in summer and winter of 2006.

Table 2. Statistical summary of carbonyl compounds at Nan-Chie and Hsiung-Kong sites in 2006 (unit: $\mu g/m^3$).

Compounds	Nan-Chie			Hsiung-Kong		
	Mean ± SD	Maximun	Minimun	Mean ± SD	Maximun	Minimun
Formaldehyde	17.99 ± 9.48	35.86	10.08	21.47 ± 11.82	39.40	9.94
Acetaldehyde	13.69 ± 3.90	19.53	8.36	16.68 ± 5.61	26.90	9.08
Acetone	1.33 ± 0.01	1.67	1.10	1.24 ± 0.03	1.30	0.69
Propionaldehyde	2.83 ± 2.51	7.29	0.69	3.63 ± 3.53	8.77	1.10
Crotonaldehyde	1.29 ± 0.24	1.95	0.61	1.34 ± 0.01	1.45	1.24
Butyraldehyde	1.72 ± 0.02	3.47	0.35	4.34 ± 2.34	27.78	0.69
Benzaldehyde	1.60 ± 1.00	3.47	0.35	2.20± 1.48	4.51	0.00
Isovaleraldehyde	1.37 ± 0.01	1.45	1.30	1.35 ± 0.04	1.42	1.20
Valeraldehyde	1.18 ± 0.01	1.36	1.10	1.14 ± 0.02	1.32	1.10
o-Tolualdehyde	1.34 ± 0.06	1.60	1.13	1.36 ± 0.03	1.45	1.28
m-Tolualdehyde	2.42 ± 2.87	11.46	0.00	4.77 ± 4.84	14.47	1.28
p-Tolualdehyde	2.71 ± 2.09	8.68	0.35	2.11 ± 1.22	10.09	0.35
Hexaldehyde	1.23 ± 0.04	1.30	1.04	1.27 ± 0.03	1.75	1.04
Cyclohexanone	0.38 ± 0.01	0.47	0.29	0.55 ± 0.26	2.43	0.18
Heptaldehyde	0.61 ± 0.32	5.90	0.31	0.36 ± 0.03	0.45	0.20
Octaldehyde	1.02 ± 0.90	2.78	0.00	1.15 ± 1.13	3.07	0.30
Nonaldehyde	2.09 ± 2.45	5.56	0.21	2.85 ± 3.53	8.68	0.24
Decylaldehyde	0.79 ± 0.31	2.78	0.35	0.43 ± 0.10	1.32	0.29
Total	55.60 ± 26.10	94.22	33.00	68.25 ± 30.75	110.24	35.04

Ratios of Concentrations of Carbonyl Compounds

Table 3 presents the ratios of formaldehyde/acetaldehyde (C_1/C_2) and acetaldehyde/propionaldehyde (C_2/C_3) thus obtained in this study and those obtained in other (Zhang *et al*., 1994; Possanzini *et al*., 1996; Slemr and Junkermann, 1996; Andreini *et al*., 2000; Grosjean *et al*., 2002; Ho *et al*., 2002b; Bakeas *et al*., 2003; Villanueva-Fierro *et al*., 2004; Moussa *et al*., 2006; Rubio *et al*., 2006; Pal *et al*., 2008). C_1/C_2 ratios usually vary from one to two in an urban area to about ten in a rural area (Shepson *et al*., 1991; Possanzini *et al*., 1996). A high C_1/C_2 ratio may reflect the local generation of natural reactive hydrocarbons, whose oxidation yields more formaldehyde than acetaldehyde (Lloyd *et al*., 1983). The higher C_1/C_2 ratio in summer (1.47) in this study may be related to the fact that the lifetime of formaldehyde exceeds that of acetaldehyde with respect to photolysis and reactions with hydroxyl radicals, NO_3 radicals, and O_3 (Atkinson, 2000). The higher C_1/C_2 ratio in summer (1.47) than in winter (1.03) herein matched findings in Rome (Possanzini *et al*., 1996), eastern Finland (Viskari *et al*., 2000), and South Korea (Pal *et al*., 2008). This difference is further explained by the experimental estimation of the secondary conversion of alkenes to formaldehyde and acetaldehyde of 0.39 and 0.18 (0.23 and 0.15) in summer (in winter), respectively (Asthuller, 1993).

The C_2/C_3 ratio should be used as indicators of the anthropogenic origin of ambient carbonyls, since propionaldehyde is believed to be associated only with anthropogenic emissions (Anderson *et al*., 1996). Thus, this ratio is typically found to be high in rural air, but low in urban air (Feng *et al*., 2005). C_2/C_3 ratios are much lower in summer (3.46) than in winter (10.77). They are persistent, despite the fact that the lifetime of acetaldehyde

exceeds that of propionaldehyde with respect to photolysis and reactions with hydroxyl radicals, NO_x radicals, and O_3 (Atkinson, 2000). Notably, the large photochemical production of acetaldehyde at high temperatures and strong solar radiation may be counterbalanced by its rapid loss due to photolysis and reactions with hydroxyl radicals (Christensen *et al*., 2000; Sin *et al*., 2001).

The mean C_1/C_2 value obtained from all data in this study was 1.30, which may indicate strong emissions from urban areas. The mean C_2/C_3 value in this study was 4.70, suggesting that the local participation of anthropogenic hydrocarbons was important in the production of carbonyls at the sampling site. The results were similar to those obtained in Rome (Possanzini *et al*., 1996).

Source Identification and Apportionment

The PCA/APCS receptor models were applied to identify and evaluate potential emission sources of carbonyls and their contribution to carbonyls at both sites in the summer and winter in Kaohsiung. Table 4 presents PCA/APCS results for the 18 carbonyls species at the Nan-Chie site in summer. The three primary factors (sources) were: secondary emissions/vehicle exhausts (gasoline engines)/stationary emissions (food industry), stationary emissions (petrochemical)/waste treatment and restaurant emissions. The characteristics of the three sources at the Nan-Chie site were as follows:

FC1: The dominant species are formaldehyde and acetaldehyde, which are related to secondary emissions (photochemical reaction) (Christensen *et al*., 2000), formaldehyde, benzaldehyde and tolualdehyde are related to gasoline engines emissions (Kean *et al*., 2001), and acetaldehyde and isovaleraldehyde are related to food industry emissions (Kim *et al*., 2008).

Table 3. Concentration ratios of carbonyl compounds in worldwide cities.

Location	C_1/C_2	C_2/C_3	References
Kaohsiung city, Taiwan			This study
All data	1.30	4.70	
Summer	1.47	3.46	
Winter	1.03	10.77	
Rome, Italy	1.25	3.92	Possanzini *et al.* (1996)
Hong Kong, China	2.21	8.75	Ho *et al.* (2002b)
Athens, Greece	1.00	7.62	Bakeas *et al.* (2003)
Beirut, Lebanon	1.63	1.60	Moussa *et al.* (2006)
Milan, Italy	1.44	5.02	Andreini *et al.* (2000)
New Mexico	2.35	0.53	Villanueva-Fierro *et al.* (2004)
New Jersey, USA	3.24	1.58	Zhang *et al.* (1994)
Rio de Janeiro, Brazil	1.04	9.45	Grosjean *et al.* (2002)
Santiago, Chile	0.88	4.94	Rubio *et al.* (2006)
Schauinsland, Germany	0.97	0.20	Slemr and Junkermann (1996)
South Korea	0.99	0.53	Pal *et al.* (2008)

Table 4. PCA/APCS analysis of carbonyl compounds at Nan-Chie site in summer of 2006.

Compounds	Factor			Source (average ± standard error) (%)			
	FC1	FC2	FC3	FC1	FC2	FC3	R^2
Formaldehyde	**0.98**	0.26	−0.15	76.9 ± 1.5	20.2 ± 4.9	3.1 ± 0.1	0.86
Acetaldehyde	**0.96**	0.24	0.08	88.3 ± 1.2	8.5 ± 0.4	−1.7 ± 0.2	0.80
Acetone	0.36	**0.93**	0.22	5.2 ± 0.5	104.4 ± 12.7	−12.1 ± 3.2	0.82
Propionaldehyde	0.41	**0.91**	0.15	5.9 ± 3.1	98.2 ± 0.8	−1.2 ± 4.6	0.90
Crotonaldehyde	0.58	0.42	0.11	36.2 ± 0.3	34.8 ± 9.6	17.0 ± 1.8	0.81
Butyraldehyde	0.44	**0.88**	0.05	20.5 ± 5.3	87.3 ± 3.1	−2.2 ± 0.2	0.88
Benzaldehyde	**0.88**	0.42	0.18	115.6 ± 24.4	6.0 ± 1.9	−4.9 ± 5.4	0.92
Isovaleraldehyde	**0.91**	−0.25	0.06	87.4 ± 15.7	−6.0 ± 7.9	3.4 ± 9.9	0.89
Valeraldehyde	0.38	**0.73**	0.12	19.6 ± 2.8	77.7 ± 20.3	10.9 ± 3.0	0.87
o-Tolualdehyde	**0.88**	0.27	−0.05	73.4 ± 2.6	17.6 ± 6.8	10.3 ± 5.2	0.90
m-Tolualdehyde	**0.94**	0.26	0.02	85.5 ± 2.7	15.5 ± 16.3	−5.9 ± 8.8	0.93
p-Tolualdehyde	**0.92**	0.18	0.15	73.6 ± 7.4	18.0 ± 3.5	4.6 ± 4.1	0.87
Hexaldehyde	0.58	**0.80**	0.06	27.4 ± 3.6	70.6 ± 2.8	5.8 ± 1.7	0.88
Cyclohexanone	0.62	0.28	0.15	45.6 ± 6.8	33.4 ± 2.6	17.3 ± 2.2	0.91
Heptaldehyde	0.38	−0.16	**0.82**	13.6 ± 6.3	−5.9 ± 4.8	89.0 ± 13.7	0.95
Octaldehyde	0.34	0.14	**0.81**	4.4 ± 4.8	2.5 ± 1.5	105.2 ± 14.7	0.90
Nonaldehyde	0.26	−0.18	**0.84**	2.0 ± 0.2	6.9 ± 1.3	89.0 ± 3.5	0.82
Decylaldehyde	0.41	0.26	**0.73**	20.6 ± 4.9	5.9 ± 2.9	75.5 ± 2.6	0.85
Eigenvalue	8.21	4.57	2.76				
% Total variance	45.6	25.4	15.3				
Cumulative %	45.6	71.0	86.3				
Source	Secondary emissions /vehicle exhausts (gasoline engines)/ stationary emissions (food industry)	Stationary emissions (petrochemical)/ waste treatment	Restaurant emissions				

Notes. Only factor loadings > 0.1 are listed and > 0.7 appear in **bold**.

FC2: The dominant species are acetone, propionaldehyde and hexaldehyde. These species are related to petrochemical industry emissions (Dincer *et al.*, 2006). Acetone and butyraldehyde are found not only in emissions from petrochemical industry emissions but also in waste treatment emissions (Kim *et al.*, 2008).

FC3: The dominant species are in restaurant emissions are heptaldehyde, octaldehyde, nonaldehyde and decylaldehyde

(Chiang *et al.*, 1997; Shimoda *et al.*, 1997; Schauer *et al.*, 2002).

Three factors were identified at the Nan-Chie site in winter (Table 5). FC1: high loadings of formaldehyde, benzaldehyde and tolualdehyde are related to gasoline engines emissions, whereas acetone, propionaldehyde and hexaldehyde are related to petrochemical industry emissions. FC2: high loadings of acetaldehyde and isovaleraldehyde are related to food industry emissions (Kim *et al.*, 2008), whereas acetaldehyde, crotonaldehyde and cyclohexanone are found in diesel engines emissions (Schauer *et al.*, 1999; Peng *et al.*, 2008; Lin *et al.*, 2009). FC3: high loadings of heptaldehyde, octaldehyde, nonaldehyde and decylaldehyde are related to restaurant emissions.

At Hsiung-Kong, three primary factors in summer (Table 6); were secondary emissions/vehicle exhausts (gasoline and diesel engines)/stationary emissions (metal assembly); restaurant emissions; and others. Notably, the high loadings of carbonyls in FC1 and FC2 species are similar to those at Nan-Chie. However, isovaleraldehyde and valeraldehyde were difficult to identify from FC3. However, acetaldehyde, acetone and butyraldehyde were associated with metal assembly industry (Kim *et al.*, 2008). In winter, the following only two factors at Hsiung-Koang (Table 7); the only two factors were vehicle exhausts (gasoline engines)/restaurant emission and vehicle exhausts

(diesel engines)/stationary emissions (metal assembly). All dominant FC1 and FC2 species were similar to those at Nan-Chie.

The APCS receptor model was applied to quantify the contributions of the sources. At the Nan-Chie site in the summer, secondary emissions/vehicle exhausts (gasoline engines)/stationary emissions (food industry) were responsible for $44.5 \pm 11.6\%$ of the carbonyls concentration, followed by stationary emissions (petrochemical)/waste treatment ($33.1 \pm 11.7\%$), and restaurant emissions ($22.4 \pm 12.1\%$) (Fig. 3a). In the winter, vehicle exhausts (gasoline engines)/stationary emissions (petrochemical) accounted for most of the carbonyls concentration ($57.2 \pm 12.7\%$), followed by stationary emissions (food industry)/vehicle exhausts (diesel engines) ($22.8 \pm 10.8\%$), and restaurant emissions ($20.0 \pm 12.1\%$) (Fig. 3b). At the Hsiung-Kong site in the summer, secondary emissions/vehicle exhausts (gasoline engines and diesel engines)/stationary emissions (metal assembly) were responsible for most of the carbonyl concentration ($63.9 \pm 13.6\%$), followed by restaurant emissions ($21.1 \pm 11.4\%$), and others ($15.0 \pm 8.5\%$) (Fig. 4a). In the winter, vehicle exhausts (gasoline engines)/ restaurant emissions were responsible for most of the carbonyls concentration ($55.9 \pm 12.8\%$), followed by vehicle exhausts (diesel engines)/stationary emissions (metal assembly) ($44.1 \pm 13.5\%$) (Fig. 4b).

Table 5. PCA/APCS analysis of carbonyl compounds at Nan-Chie site in winter of 2006.

Compounds	Factor			Source (average ± standard error) (%)			
	FC1	FC2	FC3	FC1	FC2	FC3	R^2
Formaldehyde	**0.90**	0.38	–0.03	106.1 ± 5.2	6.4 ± 2.4	–6.2 ± 0.1	0.96
Acetaldehyde	0.46	**0.71**	–0.02	18.7 ± 1.3	78.9 ± 0.3	10.9 ± 1.4	0.89
Acetone	**0.76**	0.53	–0.03	94.9 ± 7.4	4.9 ± 4.4	3.4 ± 0.4	0.94
Propionaldehyde	**0.80**	0.55	0.10	102.1 ± 5.7	2.1 ± 1.7	–2.1 ± 0.7	0.99
Crotonaldehyde	0.41	**0.70**	0.12	5.6 ± 0.2	88.9 ± 6.8	3.7 ± 2.5	0.93
Butyraldehyde	**0.72**	0.42	0.05	79.9 ± 6.9	12.7 ± 4.2	2.3 ± 0.9	0.94
Benzaldehyde	**0.92**	0.20	–0.06	87.0 ± 5.0	20.8 ± 4.0	0.4 ± 0.9	0.66
Isovaleraldehyde	0.44	**0.82**	0.01	15.1 ± 2.9	85.1 ± 26.9	0.5 ± 0.4	0.92
Valeraldehyde	**0.73**	0.43	0.18	78.7 ± 3.5	11.6 ± 2.1	2.5 ± 1.9	0.94
o-Tolualdehyde	**0.92**	0.10	–0.05	90.6 ± 13.9	6.2 ± 1.5	1.6 ± 0.9	0.85
m-Tolualdehyde	**0.98**	0.13	0.08	107.3 ± 5.1	2.2 ± 0.4	–9.3 ± 0.4	0.99
p-Tolualdehyde	**0.90**	0.20	0.04	89.1 ± 3.6	16.5 ± 4.9	–5.8 ± 2.5	0.89
Hexaldehyde	**0.83**	0.46	0.05	72.6 ± 2.0	20.7 ± 2.4	1.8 ± 0.6	0.94
Cyclohexanone	0.58	**0.72**	0.10	26.3 ± 3.4	76.7 ± 8.0	2.8 ± 1.8	0.89
Heptaldehyde	0.46	–0.12	**0.74**	10.1 ± 3.0	–7.8 ± 0.5	93.0 ± 2.8	0.94
Octaldehyde	0.32	–0.04	**0.80**	4.1 ± 1.3	–2.7 ± 1.6	95.5 ± 5.2	0.95
Nonaldehyde	0.40	0.12	**0.70**	35.1 ± 11.1	–10.8 ± 2.8	72.2 ± 8.9	0.96
Decylaldehyde	0.49	0.10	**0.72**	6.6 ± 4.8	–2.2 ± 0.9	92.7 ± 1.2	0.87
Eigenvalue	8.71	3.78	2.28				
% Total variance	49.2	20.2	12.7				
Cumulative %	49.2	69.4	82.1				
Source	Vehicle exhausts (gasoline engines)/ stationary emissions (petrochemical)	Stationary emissions (food industry)/ vehicle exhausts (diesel engines)	Restaurant emissions				

Notes. Only factor loadings > 0.1 are listed and > 0.7 appear in **bold**.

Table 6. PCA/APCS analysis of carbonyl compounds at Hsiung-Kong site in summer of 2006.

Compounds	Factor			Source (average ± standard error) (%)			
	FC1	FC2	FC3	FC1	FC2	FC3	R^2
Formaldehyde	**0.98**	0.18	0.13	99.5 ± 9.3	2.0 ± 0.4	1.1 ± 0.9	0.85
Acetaldehyde	**0.93**	0.22	0.16	95.6 ± 5.0	2.7 ± 0.4	2.3 ± 0.1	0.99
Acetone	**0.92**	0.05	0.19	101.0 ± 9.6	–2.0 ± 1.0	3.7 ± 2.7	0.97
Propionaldehyde	**0.87**	0.15	0.23	106.4 ± 6.6	0.6 ± 0.3	0.9 ± 0.2	0.93
Crotonaldehyde	**0.84**	0.22	0.25	72.2 ± 6.2	8.1 ± 3.8	17.1 ± 6.7	0.94
Butyraldehyde	**0.90**	0.16	0.14	100.2 ± 8.8	3.9 ± 2.7	–2.1 ± 3.0	0.89
Benzaldehyde	**0.80**	0.10	0.28	95.2 ± 4.4	1.5 ± 1.1	4.2 ± 1.2	0.98
Isovaleraldehyde	0.40	0.35	**0.71**	4.1 ± 1.9	3.6 ± 1.9	75.6 ± 10.4	0.85
Valeraldehyde	0.46	0.32	**0.74**	5.5 ± 0.1	–0.1 ± 0.1	97.5 ± 3.8	0.99
o-Tolualdehyde	**0.83**	–0.06	0.30	107.5 ± 13.5	–5.8 ± 2.3	6.5 ± 4.2	0.96
m-Tolualdehyde	**0.84**	0.08	0.43	94.5 ± 3.2	–2.3 ± 1.2	8.2 ± 1.7	0.95
p-Tolualdehyde	**0.79**	0.14	0.34	96.5 ± 16.1	–3.8 ± 1.8	8.2 ± 3.7	0.93
Hexaldehyde	0.56	0.41	0.31	43.8 ± 3.7	28.7 ± 4.1	22.8 ± 12.3	0.84
Cyclohexanone	**0.88**	0.18	0.26	89.3 ± 9.6	1.8 ± 0.8	3.9 ± 0.8	0.85
Heptaldehyde	0.23	**0.76**	0.31	3.2 ± 0.4	86.8 ± 9.4	4.8 ± 1.6	0.96
Octaldehyde	0.35	**0.78**	0.22	4.8 ± 1.4	84.6 ± 0.6	2.2 ± 0.8	0.92
Nonaldehyde	0.36	**0.79**	0.26	11.7 ± 0.2	93.0 ± 9.9	4.3 ± 1.1	0.90
Decylaldehyde	0.33	**0.82**	0.28	19.6 ± 0.5	76.0 ± 10.4	8.6 ± 2.9	0.96
Eigenvalue	9.20	3.24	2.35				
% Total variance	51.1	18	13.1				
Cumulative %	51.1	69.1	82.2				
Source	Secondary emissions/ vehicle exhausts (gasoline engines and diesel engines)/ stationary emissions (metal assembly)	Restaurant emissions	Others				

Notes: Only factor loadings > 0.1 are listed and > 0.7 appear in **bold**.

Table 7. PCA/APCS analysis of carbonyl compounds at Hsiung-Kong site in winter of 2006.

Compounds	Factor		Source (average ± standard error) (%)		
	FC1	FC2	FC1	FC2	R^2
Formaldehyde	**0.85**	0.28	88.1 ± 6.3	20.1 ± 3.7	0.98
Acetaldehyde	0.02	**0.99**	1.5 ± 0.6	104.3 ± 7.9	0.91
Acetone	0.24	**0.82**	6.4 ± 2.3	90.4 ± 7.1	0.86
Propionaldehyde	–0.27	**0.92**	1.8 ± 0.6	110.6 ± 2.8	0.94
Crotonaldehyde	0.12	**0.76**	10.8 ± 1.2	84.3 ± 7.2	0.83
Butyraldehyde	0.14	**0.98**	5.8 ± 3.5	112.3 ± 10.0	0.90
Benzaldehyde	**0.90**	0.13	96.4 ± 11.8	9.9 ± 3.9	0.89
Isovaleraldehyde	0.58	0.40	47.1 ± 2.4	41.9 ± 5.1	0.98
Valeraldehyde	0.56	0.31	46.2 ± 5.0	39.6 ± 7.3	0.87
o-Tolualdehyde	**0.82**	0.21	104.5 ± 0.9	–3.8 ± 0.1	0.99
m-Tolualdehyde	**0.92**	0.15	101.9 ± 14.1	–12.7 ± 9.0	0.99
p-Tolualdehyde	**0.86**	0.10	102.7 ± 6.6	3.3 ± 0.6	0.89
Hexaldehyde	0.52	0.25	43.7 ± 5.4	40.4 ± 3.4	0.90
Cyclohexanone	0.13	**0.79**	11.2 ± 2.8	90.1 ± 12.1	0.89
Heptaldehyde	**0.82**	0.26	85.3 ± 4.3	10.5 ± 2.8	0.96
Octaldehyde	**0.80**	0.28	81.4 ± 3.3	15.9 ± 4.1	0.99
Nonaldehyde	**0.84**	0.23	82.2 ± 2.1	19.8 ± 8.8	0.92
Decylaldehyde	**0.82**	0.21	88.6 ± 3.6	17.5 ± 5.2	0.95

Table 7. *(continued).*

Compounds	Factor		Source (average ± standard error) (%)		
	FC1	FC2	FC1	FC2	R^2
Eigenvalue	7.85	5.13			
% Total variance	43.6	28.5			
Cumulative %	43.6	72.1			
Source	Vehicle exhausts (gasoline engines)/ restaurant emissions	Vehicle exhausts (diesel engines)/stationary emissions (metal assembly)			

Notes. Only factor loadings > 0.1 are listed and > 0.7 appear in **bold**.

(a) Summer
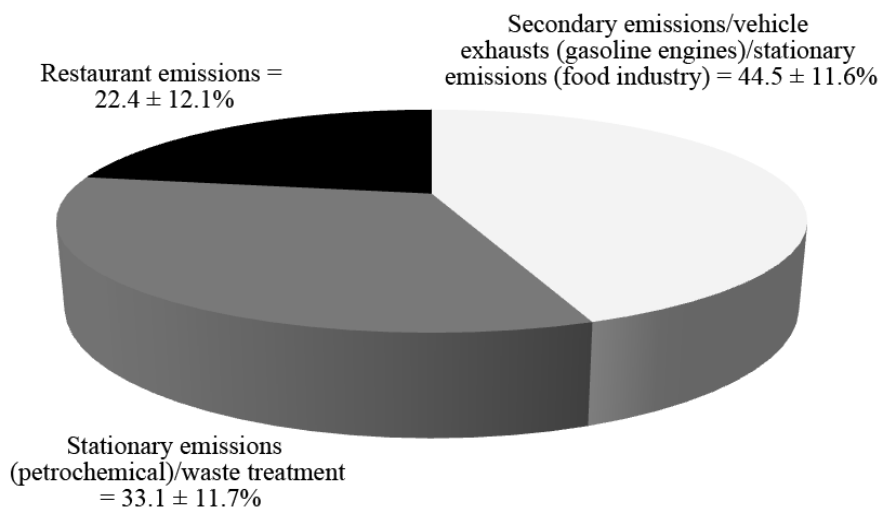
(b) Winter
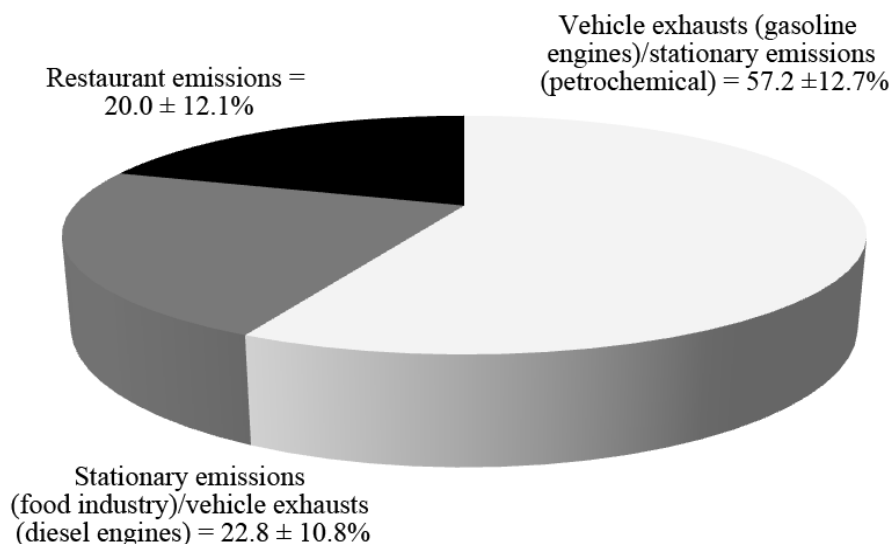
Fig. 3. Average percentage source contributions (% of calculated mass ± SD) to carbonyls of APCS results at Nan-Chie site in (a) summer and (b) winter, 2006.

The APCS results demonstrate that secondary emissions and vehicular emissions are the dominant source of ambient carbonyl in the summer, while vehicle exhausts predominated carbonyls emission in the winter at the two sites. However, other sources, such as stationary emissions and restaurant cooking emissions, are also important.

Notably, the APCS results may vary from case to case, because many factors, including temperature, seasonal variation, wind speed, wind direction, and other pollution sources, affect the contribution of pollution sources to atmospheric carbonyls concentrations.
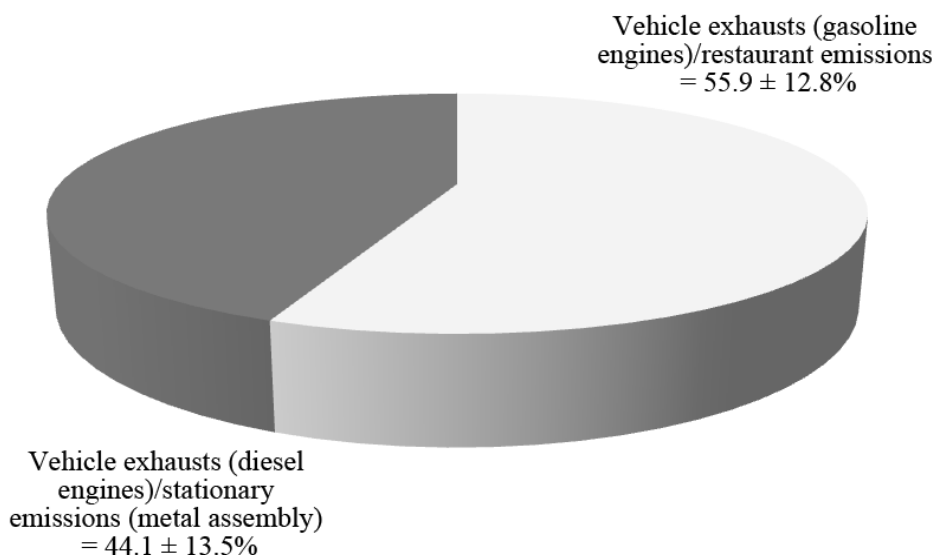
(a) Summer
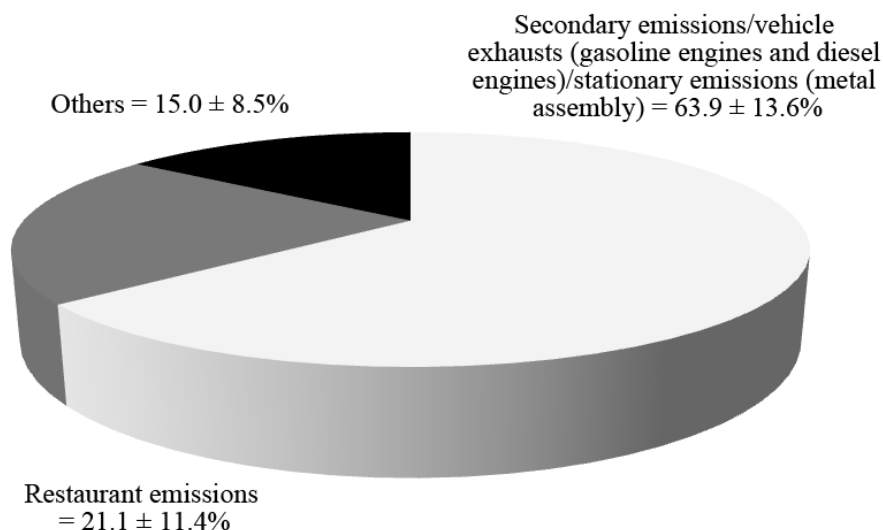
(b) Winter

Fig. 4. Average percentage source contributions (% of calculated mass ± SD) to carbonyls of APCS results at Hsiung-Kong site in (a) summer and (b) winter, 2006.

CONCLUSIONS

The characteristics of 18 atmospheric carbonyls species at northern (Nan-Chie) and southern (Hsiung-Kong) sites in summer and winter of 2006 in Kaohsiung City were studied. The most abundant species in the atmosphere of Kaohsiung were formaldehyde and acetaldehyde, altogether accounting for approximately 56–57% of total carbonyls. The mean concentration of carbonyls at Nan-Chie was 74.06 μg/m^3 in the summer and 37.14 μg/m^3 in the winter, while those at Hsiung-Kong were 89.99 μg/m^3 in the summer and 46.50 μg/m^3 in the winte. The overall mean concentration of carbonyls at Hsiung-Kong (68.25 μg/m^3) is higher than that at Nan-Chie (55.60 μg/m^3), consistent with the fact that the mean traffic volume at Hsiung-Kong is about 60% higher than that at Nan-Chie.

The receptor models of PCA/APCS showed that secondary emissions, at both sites, stationary emissions, vehicle exhausts and restaurant emission were primary sources of atmospheric carbonyls in the summer, and stationary emissions, vehicle exhausts and restaurant emission in the winter season.

REFERENCES

Anderson, L.G., Lanning, J.A., Barrell, R., Miyagishima, J., Jones, R.H. and Wolfe, P. (1996). Sources and Sinks of Formaldehyde and Acetaldehyde: An Analysis of Denver's Ambient Concentration Data. *Atmos. Environ.*

30: 2113–2123.

Andreini, B.P., Baroni, R., Galimberti, E. and Sesana, G. (2000). Aldehydes in the Atmospheric Environment: Evaluation of Human Exposure in the North-west area of Milan. *Microchem. J.* 67: 11–19.

Asthuller, A.P. (1993). Production of Aldehydes as Primary Emissions and from Secondary Atmospheric Reactions of Alkenes and Alkanes during the Night and Early Morning Hours. *Atmos. Environ.* 27A: 21–32.

Atkinson, R. (2000). Atmospheric Chemistry of VOCs and NO_x, *Atmos. Environ.* 34: 2063–2101.

Baez, A.P., Belmont, R. and Padilla, H. (1995). Measurements of Formaldehyde and Acetaldehyde in the Atmosphere of Mexico City. *Environ. Pollut.* 89: 163–167.

Bakeas, E.B., Argyris, D.I. and Siskos, P.A. (2003). Carbonyls Compounds in the Urban Environment of Athens, Greece. *Chemosphere* 52: 805–813.

Carlier, P., Hannachi, H. and Mouvier, G. (1986). The Chemistry of Carbonyl Compounds in the Atmosphere-A Review. *Atmos. Environ.* 20: 2079–2099.

Cavalcante, R.M., Campelo, C.S., Barbosa, M.J., Silveria, E.R., Carvalho, T.V. and Nascimento, R.F. (2006). Determination of Carbonyl Compounds in Air and Cancer Risk Assessment in an Academic Institute in Fortaleza, Brazil. *Atmos. Environ.* 40: 5701–5711.

Cerón, R.M., Cerón, J.G. and Muriel, M. (2007). Diurnal and Seasonal Trends in Carbonyl Levels in a Semi-urban Coastal Site in the Gulf of Campeche, Mexico. *Atmos. Environ.* 41: 63–71.

Chang, C.C., Wang, J.L., Candice Lung, S.C., Liu, S.C. and Shiu, C.J. (2009). Source Characterization of Ozone Precursors by Complementary Approaches of Vehicular Indicator and Principal Component Analysis. *Atmos. Environ.* 43: 1771–1778.

Chen, K.S. Ho, Y.T., Lai, H.C., Tsai, Y.A. and Chen, S.J. (2004). Trends in Concentration of Ground-level Ozone and Meteorological Conditions during High Ozone Episodes in Kao-Ping Airshed, Taiwan. *J. Air Waste Manage. Assoc.* 54: 36–48.

Chen, K.S., Wang, H.K., Peng, Y.P., Wang, W.C., Chen, C.H. and Lai, C.H. (2008). Effects of Open Burning of Rice Straw on Concentrations of Atmospheric Polycyclic Aromatic Hydrocarbons in Central Taiwan. *J. Air Waste Manage. Assoc.* 58: 1318–1327.

Chiang, T.A., Wu, P.F., Wang, L.F., Lee, H., Lee, C.H. and Ko, Y.C. (1997). Mutagenicity and Polycyclic Aromatic Hydrocarbon Content of Fumes from Heatedcooking Oils Produced in Taiwan. *Mutat. Res.* 381: 157–161.

Christensen, C.S. Skov, H. Nielsen, T. and Lohse, C. (2000). Temporal Variation of Carbonyl Compound Concentrations at a Semi-rural Site in Denmark. *Atmos. Environ.* 34: 287–296.

Derwent, R.G., Middleton, D.R., Field, R.A., Goldstone, M.E., Lester, J.N. and Perry, R. (1995). Analysis and Interpretation of Air Quality Data from an Urban Roadside Location in Central London over the Period from July 1991 to July 1992. Atmos. Environ. 29: 923–946.

Dincer, F. and Muezzinoglu, A. (2006). Chemical Characterization of Odors Due to some Industrial and Urban Facilities in Izmir, Turkey. *Atmos. Environ.* 40: 4210–4219.

Duan, J., Tan, T., Yang, L., Wu, S. and Hao, J. (2008). Concentration, Sources and Ozone Formation Potential of Volatile Organic Compounds (VOCs) during Ozone Episode in Beijing. *Atmos. Res.* 88: 25–35.

Feng, Y., Wen, S., Chen, Y., Wang, X., Lu, H., Bi, X., Sheng, G. and Fu, J. (2005). Ambient Levels of Carbonyl Compounds and their Sources in Guangzhou, China. *Atmos. Environ.* 39: 1789–1800.

Grosjean, D., Grosjean, E. and Moreira, L.F.R. (2002). Speciated Ambient Carbonyls in Rio de Janeiro, Brazil. *Environ. Sci. Technol.* 36: 1389–1395.

Grosjean, E. Grojean, D., Fraser, M.P. and Cass, G.R. (1996). Air Quality Model Evaluation Data for Organics. 2. C_1–C_{14} Carbonyls in Los Angeles Air. *Environ. Sci. Technol.* 30: 2687–2703.

Guo, H., Wang, T. and, Louie, P.K.K. (2004a). Source Apportionment of Ambient Non-methane Hydrocarbons in Hong Kong: Application of a Principal Component Analysis/Absolute Principal Compound Scores (PCA/APCS) Receptor Model. *Environ. Pollut.* 129: 489–498.

Guo, H., Wang, T., Simpson, I.J., Blake, D.R., Yu, X.M., Kwok, Y.H. and Li, Y.S. (2004b). Source Contributions to Ambient VOCs and CO at a Rural Site in Eastern China. *Atmos. Environ.* 38: 4551–4560.

Hauptmann, M., Lubin, J.H., Stewart, P.A., Hayes, R.B. and Blair, A. (2004). Mortality from Solid Cancers among Workers in Formaldehyde Industries. *Am. J. Epidemiol.* 159: 1117–1130.

Ho, K.F., Lee, S.C. and Chiu, G.M.Y. (2002a). Characterization of Selected Volatile Organic Compounds, Polycyclic Aromatic Hydrocarbons and Carbonyl Compounds at a Roadside Monitoring Station. *Atmos. Environ.* 36: 57–65.

Ho, K.F., Lee, S.C., Louie, P.K.K. and Zou, S.C. (2002b). Seasonal Variation of Carbonyl Compound Concentrations in Urban Area of Hong Kong. *Atmos. Environ.* 36: 1259–1265.

Hoekman, S.K. (1992). Speciated Measurements and Calculated Reactivities of Vehicle Exhaust Emissions from Conventional and Reformulated Gasoline. *Environ. Sci. Technol.* 26: 1206–1216.

Johnson, R. and Wichern, D. (2002). *Applied Multivariate Statistical Analysis*, 5th ed., Prentice Hall: New York.

Karar, K. and Gupta, A.K. (2007). Source Apportionment of PM_{10} at Residential and Industrial Sites of an Urban Region of Kolkata, India. *Atmos. Res.* 84: 30–41.

Katsoyiannis, A., Leva, P. and Kotzias, D. (2008). VOC and Carbonyl Emissions from Carpets: A Comparative Study Using Four Types of Environmental Chambers. *J. Hazard. Mater.* 152: 669–676.

Kean, A.J., Grosjean, E., Grosjean, D. and Harley, R.A. (2001). On-road Measurement of Carbonyls in California Light-duty Vehicle Emissions. *Environ. Sci. Technol.* 35: 4198–4204.

Kim, K.H. Hong, Y.J., Pal, R., Jeon, E.C., Koo, Y.S. and Sunwoo, Y. (2008). Investigation of Carbonyl Compounds in Air from Various Industrial Emission

Sources. *Chemosphere* 70: 807–820.

Lai, C.H., Chen, K.S., Ho, Y.T., Peng, Y.P. and Chou, Y.M. (2005). Receptor Modeling of Source Contributions to Atmospheric Hydrocarbons in Urban Kaohsiung, Taiwan. *Atmos. Environ.* 39: 4543–4559.

Lin, Y.C., Wu, T.Y., Ou-Yang, W.C. and Chen, C.B. (2009). Reducing Emissions of Carbonyl Compounds and Regulated Harmful Matters from a Heavy-duty Diesel Engine Fueled with Paraffinic/Biodiesel Blends at One Low Load Steady-state Condition. *Atmos. Environ.* 43: 2642–2647.

Liu, W., Zhang, J., Zhang, L., Turpin, B.J., Weisel, C.P., Morandi, M.T., Stock, T.H., Colome, S. and Korn, L.R. (2006). Estimating Contributions of Indoor and Outdoor Sources to Indoor Carbonyl Concentrations in Three Urban Areas of the United States. *Atmos. Environ.* 40: 2202–2214.

Lloyd, A.C., Atkinson, R., Lurmann, F.W. and Nitta, B.M. (1983). Modelling Potential Ozone Impacts from Natural Hydrocarbons–I. Development and Testing of a Chemical Mechanism for the NO_x-air Photooxidations of Isoprene and a-pinene under Ambient Conditions. *Atmos. Environ.* 17: 1931–1950.

Moussa, S.G., El-Fadel, M. and Saliba, N.A. (2006). Seasonal, Diurnal and Nocturnal Behaviors of Lower Carbonyl Compounds in the Urban Environment of Beirut, Lebanon. *Atmos. Environ.* 40: 2459–2468.

Pal, R., Kim, K.H., Hong, Y.J. and Jeon, E.C. (2008). The Pollution Status of Atmospheric Carbonyls in a Highly Industrialized Areas. *J. Hazard. Mater.* 153: 1122–1135.

Peng, C.Y., Yang, H.H., Lan, C.H. and Chien, S.M. (2008). Effects of the Biodiesel Blend Fuel on Aldehyde Emissions from Diesel Engine Exhaust. *Atmos. Environ.* 42: 906–915.

Possanzini, M., Di Palo, V., Petricca, M., Fratarcangeli, R. and Brocco, D. (1996). Measurements of Lower Carbonyls in Rome Ambient Air. *Atmos. Environ.* 30: 3757–3764.

Rubio, M.A., Zamorano, N., Lissi, E., Rojas, A., Gutiérrez, L. and Baer, D.V. (2006). Volatile Carbonylic Compounds in Downtown Santiago, Chile. *Chemosphere* 62: 1011–1020.

Schauer, J.J., Kleeman, M.J., Cass, G.R. and Simoneit, B.R.T. (1999). Measurement of Emissions from Air Pollution Sources. 2. C_1 though C_{30} Organic Compounds from Medium Duty Diesel Trucks. *Environ. Sci. Technol.* 33: 1578–1587.

Schauer, J.J., Kleeman, M.J., Cass, G.R. and Simoneit, B.R.T. (2002). Measurement of Emissions from Air Pollution Sources. 4. C_1–C_{27} Organic Compounds from Cooking with Seed Oils. *Environ. Sci. Technol.* 36: 567–575.

Seco, R., Peñuelas, J. and Filellaet, I. (2007). Short-chain Oxygenated VOCs: Emission and Uptake by Plants and Atmospheric Sources, Sinks, and Concentrations. *Atmos. Environ.* 41: 2477–2499.

Shepson, P.B., Hastie, D.R., Schiff, H.I., Polizzi, M., Bottenheim, J.W., Anlauf, K., Mackay, G.I. and Karecki, D.R. (1991). Atmospheric Concentrations and Temporal Variations of C_1–C_3 Carbonyl-compounds at 2 Rural Sites in Central Ontario. *Atmos. Environ.* 25: 2001–2015.

Shimoda, M., Nakada, N., Nakashima, M. and Osajima, Y. (1997). Quantitative Comparison of Volatile Flavor Compounds in Deep-roasted Sesame Seed Oil. *J. Agric. Food. Chem.* 45: 3193–3196.

Sin, D.W.M., Wong, C.Y. and Louie, P.K.K. (2001). Trends of Ambient Carbonyl Compounds in the Urban Environment: Evaluation of Hong Kong. *Atmos. Environ.* 35: 5961–5969.

Slemr, J. and Junkermann, W. (1996). Temporal variations in Formaldehyde, Acetaldehyde, and Acetone and Budget of Formaldehyde at a Rural Site in Southern Germany. *Atmos. Environ.* 21: 3667–3676.

Swietlicki, E., Puri, S., Hansson, H.C. and Edner, H. (1996). Urban Air Pollution Source Apportionment Using a Combination of Aerosol and Gas Monitoring Techniques. *Atmos. Environ.* 30: 2795–2809.

Thurston, G.D. and Spengler, J.D. (1985). A Quantitative Assessment of Source Contributions to Inhalable Particulate Matter Pollution in Metropolitan Boston. *Atmos. Environ.* 19: 9–25.

Villanueva-Fierro, I., Popp, C.J. and Martin, R.S. (2004). Biogenic Emissions and Ambient Concentrations of Hydrocarbons, Carbonyl Compounds and Organic Acids from Ponderosa Pine and Cottonwood Trees at Rural and Forested Sites in Central New Mexico. *Atmos. Environ.* 38: 249–260.

Viskari, E.L., Vartianinen, M. and Pasanen, P. (2000). Seasonal and Diurnal Variation in Formaldehyde and Acetaldehyde Concentrations along a Highway in Eastern Finland. *Atmos. Environ.* 34: 917–923.

Wang, H.K., Huang, C.H., Chen, K.S., Peng, Y.P. and Lai, C.H. (2010). Characteristics and Sources of Carbonyl Compounds in the Atmosphere in Kaohsiung City, Southern Taiwan. *J. Hazard. Mater.* 179: 1115–1121.

Zhang, J.F., He, Q.C. and Lioy, P.J. (1994). Characteristics of Aldehydes: Concentrations, Source, and Exposures for Indoor and Outdoor Residential Microenvironments. *Environ. Sci. Technol.* 28: 146–152.