

Carbonyl compounds in the roadside environment of Hong Kong

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Abstract

The levels of carbonyl compounds were determined at the roadside urban station at the Hong Kong Polytechnic University (HKPU) campus during January 2002 to February 2002. Nine carbonyl compounds were quantified in this study. Temperature and solar radiation were found to affect the photochemical reactions of the carbonyls. Formaldehyde/acetaldehyde ratio ranged from 1.27 to 1.35. Strong correlations between formaldehyde and acetaldehyde were found, in the time period 1800–2100, which indicated that they were originated from the same sources during this time period. Roadside carbonyl samples were also collected at four other roadside environments during 2001. Kwai Chung (KC) station showed the highest average formaldehyde and acetaldehyde concentrations due to its highest traffic flow, especially for diesel vehicles. High concentration of toluene emitted from gasoline-fueled vehicles was believed to be the cause of high benzaldehyde level at the Central (CT) station through the photochemical oxidation of toluene. The average concentrations of formaldehyde and acetaldehyde in Hong Kong are well within the ranges reported in roadside environments of other urban cities. However, Mexico City in Mexico and Cairo in Egypt had much higher concentration levels of formaldehyde and acetaldehyde than in Hong Kong roadside environment due to the incomplete combustion of different fuel compositions.

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1. Introduction

Carbonyl compounds are the common constituents in the urban atmospheres. They continue to receive scientific and regulatory attention as toxic air contaminants, mutagens, and carcinogens [1]. They are also important precursors to ozone and other toxic products such as peroxyacyl nitrates [2]. Vehicle emissions are a major source of carbonyls in outdoor ambient air of Hong Kong, but information about the nature and magnitude of carbonyl emissions by motor vehicles is limited. Formaldehyde (FA) is the most abundant atmospheric aldehyde, followed by acetaldehyde (AA) in most urban and rural areas [3–5]. Aldehyde is irritating to skin, eyes and respiratory system [6] while formaldehyde is suspected carcinogenic. Also, carbonyl compounds are the major species of organic compounds involved in photochemical air pollution because aldehydes and ketone play important roles as products of photooxidation of gas phase hydrocarbons as a major source of

free radicals under strong solar radiation and high temperature [7–9].

Hong Kong is a densely populated city, with 6.8 millions population crowded in 1100 square kilometer of land. This developed city had 533,000 registered vehicles running on 1943 km of roads in 2004 [10]. Most of the roads are very close to high-rise buildings, making the vehicles exhausts the most important pollution source in such microenvironment. In an effort to monitor the variations of carbonyl compounds in urban areas in Hong Kong, samples were collected in the Hong Kong Polytechnic University (HKPU) monitoring station and five different roadside locations to study the characteristics of vehicular emission for carbonyl compounds.

Limited researches have been conducted for the characteristic of carbonyl compounds in the atmosphere of Hong Kong [5,11]. In our previous study, the samples were collected from the two toxic air pollutants (TAP) monitoring stations (managed by Hong Kong Environmental Protection Department (HKEPD)) as well as HKPU roadside station for 1-year period from April 1999 to April 2000 [5]. There were significant seasonal variations of carbonyls, with higher concentrations in summer than those in winter. The urban concentrations of carbonyls have been

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determined in several developed countries [3,7,8,12]. In a study by Viskari et al. [12] the concentrations of the most important carbonyl compounds (FA and AA) were measured along a two-lane highway during spring, summer and winter in Eastern Finland [12]. From the measurements of FA and AA at different locations, motor vehicle emissions were suggested to be the source of these compounds throughout the year. The diurnal variations showed significant decrease of FA and AA concentrations at night due to the decrease in traffic emissions. Christensen et al. [13] and Slemr et al. [14] suggested that the meteorological factors were also the main factors for the concentrations of carbonyls [13,14]. In Christensen's study it was found that the concentrations of carbonyls do not correlated at nighttime due to the dry deposition of especially FA [13]. Also a pronounced diurnal profile and even higher levels of carbonyl compounds were observed on days with high photochemical activity. Slemr et al. [14] compared the diurnal variation of FA with concentrations of ozone, nitrogen oxides and with meteorological data, and they found that both the transportation and photochemical formation contributed to the variation of FA [14].

The objectives of this study were (1) to focus on the diurnal variations of major carbonyls at the roadside microenvironment to order to determine their impact to the atmosphere of Hong Kong. (2) To investigate and interpret the variations of carbonyl compounds at different roadside microenvironments in Hong Kong.

2. Sampling and analysis

2.1. Sampling sites

To observe the diurnal variations of carbonyl concentrations, samples were collected at the roadside HKPU station (~1 m

away from the traffic road). The station is adjacent to Hong Chong Road, which leads to the Cross Harbour Tunnel with very high traffic volume of more than 170,000 vehicles per day. To analysis of the effects of traffic volume on the carbonyl levels, samples were collected at 3-hours intervals from 09:00 to 21:00 during January 2002 to February 2002. A total of six sets (24 samples) of data were collected. The samples were divided into four periods: the morning period (09:00–12:00) with high traffic activity with minor photochemical reactions; the noon period (12:00–15:00) with high photochemical activity and the time for the most intense vertical mixing; the afternoon period (15:00–18:00) again high traffic activity with photochemical reactions; and the evening period (18:00–21:00) showing the result of photochemical reactions of the day. In addition, four other roadside monitoring locations were used for the carbonyl compounds monitoring with various traffic densities and vehicle types. These were Central (CT), Kwai Chung (KC), Shatin (ST) and the Chinese University (CU). The location and brief descriptions of sampling locations are shown in Fig. 1 and Table 1. At these sites, three 2-h samples were collected at each site during summer (July to August) and winter (November)

2.2. Sampling and analysis

Carbonyls were collected by drawing air through a cartridge impregnated with acidified 2,4-dinitrophenylhydrazine (Water Corporation, Sep-Pak DNPH-silica), which is very reactive with carbonyls. An ozone scrubber was connected before the DNPH-silica cartridge to prevent interference from ozone. Flow rate of 1–1.2 L/min was used for 2 or 3 h roadside sampling (as measured with BIOS DryCal DC-Lite medium flow meter before and after each sampling). No breakthrough was

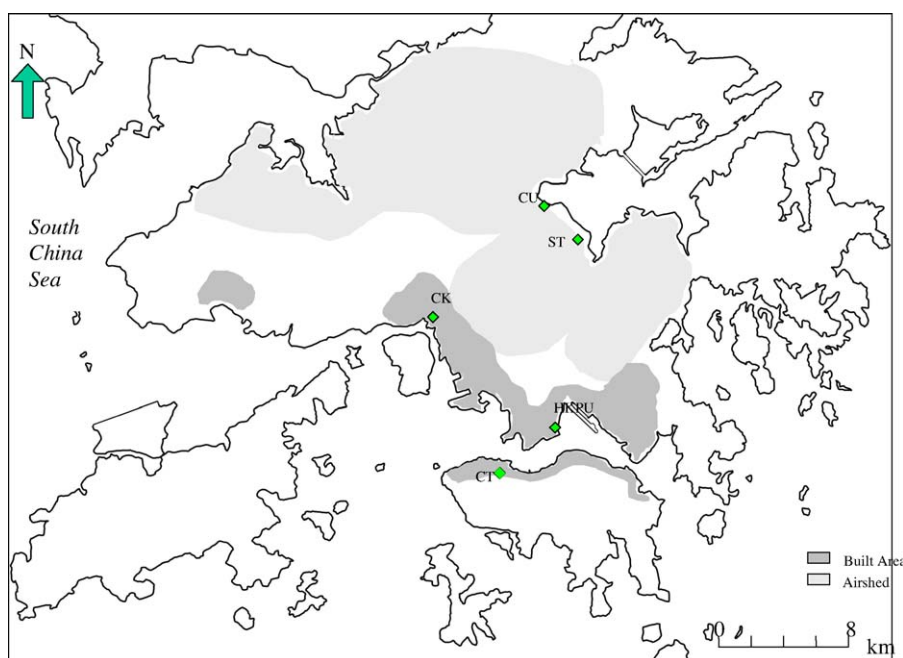


Fig. 1. Location of monitoring sites: the Hong Kong Polytechnic University Campus (HKPU), Central (CT), Kwai Chung (KC), Shatin (ST) and the Chinese University of Hong Kong (CU).

Table 1
Correlations (*R*) of carbonyl compounds during 18:00–21:00 sampling period

	1	2	3	4	5	6	7	8	9	10
Formaldehyde (1)										
Acetaldehyde (2)	0.97	1.00								
Acetone (3)	0.53	0.42	1.00							
Propionaldehyde (4)	0.92	0.92	0.60	1.00						
Cronaldehyde (5)	0.90	0.89	0.17	0.80	1.00					
Methacrolein (6)	0.46	0.32	0.69	0.31	0.16	1.00				
Buyraldehyde (7)	0.94	0.96	0.23	0.82	0.94	0.26	1.00			
Benzaldehyde (8)	-0.37	-0.36	0.24	-0.01	-0.47	-0.12	-0.50	1.00		
Valeraldehyde (9)	0.26	0.19	0.00	0.00	0.28	0.52	0.21	-0.56	1.00	
Hexaldehyde (10)	0.28	0.44	-0.29	0.22	0.30	-0.31	0.52	-0.44	-0.25	1.00

found within this flow rate. All cartridges were sealed with Teflon caps immediately, then wrapped with aluminum foil and stored in a refrigerator at 4 °C after sampling. The formed 2,4-dinitrophenylhydrazones were eluted with 5 ml of acetonitrile and 20 μ l were analyzed by high-performance liquid chromatography (HPLC) with a Waters Breeze™ HPLC System equipped with a 1525 binary HPLC pumps, a Nava-Pak C18 reverse phase analytical column (Waters), and a 2487 dual λ absorbance detector. The derivatives were detected at 360 nm. The operating conditions and overall analytic protocol have been described in detail [5]. All analysis procedures met the requirements of the USEPA Method TO-11 [15].

2.3. Quality assurance/quality control

Identification and quantification of carbonyl compounds were based on retention times and peak areas of the corresponding calibration standards. The instrument was calibrated using four standard concentrations covering the concentration of interest. Cartridge collection efficiency was determined with 2 cartridges in series; over 96% of carbonyl compounds were found in the first cartridge. The relative percent deviations (RPDs) of replicates were within 15% and the RPD of co-located samples were below 25%. Replicate analysis was performed for 10% of the samples randomly selected from previously analyzed groups for precision checking. The precision of replicate analysis was within $\pm 10\%$ for concentrations of 1 μ g/ml and within $\pm 20\%$ for concentrations near 0.5 μ g/ml.

3. Results and discussion

3.1. Temporal variation

An intensive sampling period started from 09:00 to 21:00, and a total of four 3-h samples were collected each day at the HKPU roadside station in January and February 2002. Traffic volumes of different fuel type vehicles (gasoline, diesel and LPG) were determined for the different time period (Fig. 2). Over 50% of traffic volume pass through this station were gasoline-fueled. Even though the traffic volume peaks at 15:00–18:00 evening peak hours, the variations of the traffic volume were not significant. Small variations were observed in the numbers of diesel and

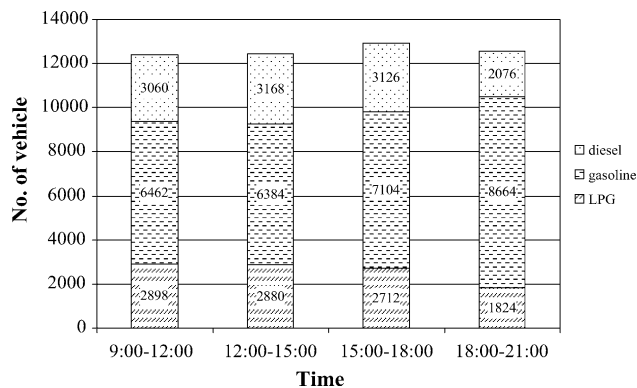


Fig. 2. Traffic volume counts in different time periods in HKPU roadside station.

LPG fueled vehicles during the morning and afternoon periods but both decreased at 18:00–21:00. Yet the number of gasoline-fueled vehicles greatly increased during this period.

FA is the most abundant atmospheric aldehyde followed by AA at the HKPU roadside environment during the intensive winter sampling period. The average FA concentration was 4.7 μ g/m³, ranged from 3.0 to 9.9 μ g/m³; and the average AA concentration was 3.6 μ g/m³, ranged from 2.2 to 7.2 μ g/m³. Fig. 3 shows the diurnal variations of FA and AA. FA and AA concentrations were the highest during the morning peak (09:00–12:00), caused by the vehicular emissions in the morning traffic peak hours. And due to traffic congestion starting

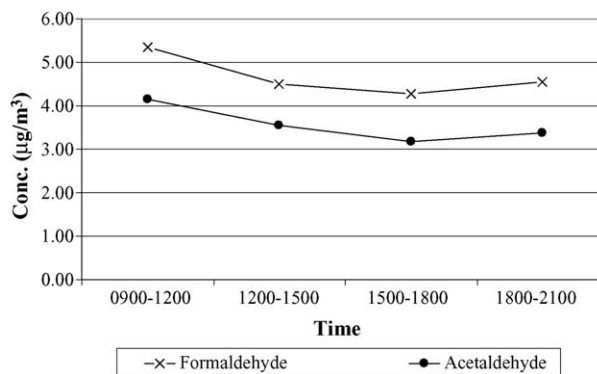


Fig. 3. Diurnal variations of formaldehyde and acetaldehyde in HKPU roadside station.

from 08:00 around the tunnel area in the morning, more pollutants were emitted at idle mode. These in turn accumulated and resulted in high pollutant levels. Temperature increased from 10:00 and led to vertical mixing processes, and pollutants levels decreased because of the higher mixing height during the afternoon. Also the traffic is no longer congested and all the vehicles were in the hot-stabilized driving modes, and the emission from vehicles were reduced. Moreover, FA and AA degrade rapidly as they have a half-life in terms of hours, principally due to reactions with hydroxyl radicals and by photolysis. Their concentration levels clearly decreased after several hours of sun light, which corresponds to the period of 12:00–18:00. After 18:00, pollutant levels increased slightly due to the increased vehicular emissions during the evening peak hours, and zero solar radiation and decreased ambient temperature reduced both the dilution as well as degradation processes.

3.2. The concentration ratios and correlations of FA and AA

The FA/AA concentration ratios were measured during different time periods. It is used as indicators of anthropogenic origin for ambient carbonyls. The ratios in this study show small variations with the range from 1.27 to 1.35. It should be pointed out that FA/AA ratios usually vary from 1 to 2 in urban areas and about 10 in rural areas [3] in many studies, as it is the case also in our study. However, some studies reported that the ratios often show large variations and caution should be exercised when using these [16].

The correlations between FA and AA at HKPU roadside station during different time periods were quantified. Strong correlations between FA and AA ($R > 0.80$) were obtained. This value is even higher in the time period of 18:00–21:00 ($R = 0.97$), which imply that these two organic compounds were coming from the same sources (vehicular exhaust). Possanzini et al. [3] concluded that vehicular exhaust was the primary source of FA and AA in winter, while both vehicular exhaust and photochemical reactions being the major sources in summer [3]. In his study, high correlations between most carbonyl compounds were found during 18:00–21:00 (Table 1), which was the time period with the lowest solar radiation and in turn the least photochemical reactions, making vehicular exhaust the major source. Lower correlations were found for time periods 12:00–15:00 and 15:00–18:00. Since vertical mixing as well as the reaction rates (both photochemical generation and photochemical degradation) of each carbonyl under high solar radiation are different, therefore the variations of the concentrations of carbonyls in daytime with high solar radiation are significant.

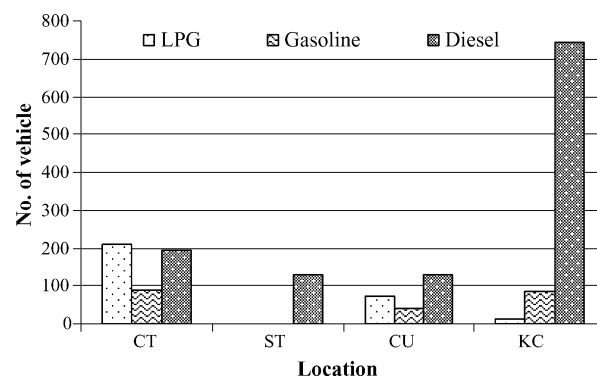


Fig. 4. Hourly traffic volumes in different roadside sampling sites.

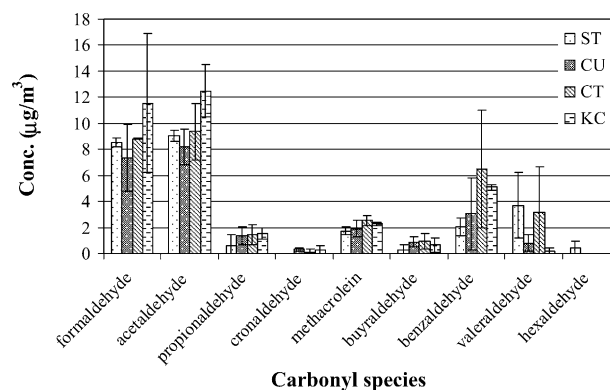


Fig. 5. The concentrations of carbonyl compounds determined at the four roadside sampling locations.

3.3. Spatial variations among different roadside microenvironments

Samples were collected at four roadside locations (Central (CT), Kwai Chung (KC), Shatin (ST) and The Chinese University of Hong Kong (CU)). These sites show different land usage and traffic flow, and have different site configurations (Table 2). The hourly traffic volumes in different roadside sampling sites are shown in Fig. 4. As shown in Fig. 5, KC had the highest concentrations of FA and AA, with average concentrations of 11.55 and 12.46 $\mu\text{g}/\text{m}^3$, respectively (the two compounds contribute 70% of total determined carbonyls). In other tunnel studies, the emission rates of FA and AA from diesel vehicles are generally higher than from gasoline vehicles [17]. KC had the highest traffic volume among the four sampling locations, with hourly flow rate of around 850 vehicles, and over 85% of traffic passing this station were heavy duty vehicles using diesel fuels. This is

Table 2
Brief summary of the roadside sampling locations

Locations	Area type	Abbreviation	Sampling period (h)	No. of samples	Sampling time
HKPU	Commercial area	HKPU	3	24 (Winter)	9:00–21:00
Central	Commercial area, near bus and taxi station	CT	2	3 (Summer), 3 (winter)	Afternoon
Kwai Chung	Industrial area, near container terminal	KC	2	3 (Summer), 3 (winter)	Afternoon
Shatin	Commercial and residential area, near bus station	ST	2	3 (Summer), 3 (winter)	Afternoon
Chinese University	Residential area, near bus and taxi station	CU	2	3 (Summer), 3 (winter)	Afternoon

Table 3
Mean concentrations ($\mu\text{g}/\text{m}^3$) of formaldehyde and acetaldehyde at roadside sites in other countries

Location	Sampling time	HCHO	CH ₃ CHO	Reference
Roadside sites				
Ealing, West London, UK	1991–1992	18.8 ± 29.3, 26.6 ± 37.4	4.6 ± 5.9, 2.9 ± 0.6	[4]
Wood Green, North London, UK	1991–1992	4.9 ± 3.3, 9.2 ± 5.3	3.5 ± 3.3, 4.3 ± 2.6	[4]
Rio de Janeiro, Brazil	December 1998 to January 2001	18.3 ± 21.1	26.3 ± 25.6	[19]
Eastern Finland	May, July, January	13.2 ± 16.5	9.0 ± 8.5	[12]
HKPU, Hong Kong	January to February 2002	4.7	3.6	Present study
CT, Hong Kong	Summer, 2001	8.8	10.9	Present study
	Winter, 2001	8.8	7.8	
CU, Hong Kong	Summer, 2001	9.1	9.2	Present study
	Winter, 2001	5.6	7.2	
KC, Hong Kong	Summer, 2001	16.7	13.9	Present study
	Winter, 2001	6.4	11.0	
ST, Hong Kong	Summer, 2001	8.3	8.7	Present study
	Winter, 2001	8.8	9.3	
Urban sites				
Central/Western, Hong Kong	Winter, 1999–2000	4.3	2.3	[11]
Tsuen Wan, Hong Kong	Winter, 1999–2000	5.1	2.6	[11]
Denver, USA	December 1987 to October 1991	2.82–4.81	1.78–2.95	[20]
Rome, Italy	January to March 1995	13.7	8.2	[3]
Mexico City, Mexico	March to May 1995	43.5	28.6	[7]
Grenoble, France	May 1995	3.1–22	3.6–18	[21]
Cairo, Egypt	March–August 1999	40	–	[22]

one of the reasons for the high concentrations of FA and AA at KC. Atmospheric photooxidation was an important secondary source of FA and AA at this station, especially in the hot summers. As it is an open area solar radiation is high, increased the rate of photooxidation and in turn the concentrations of FA and AA at this station were especially high.

The average concentrations of FA, AA, and benzaldehyde measured at the CT were 8.81, 9.37, and 6.48 $\mu\text{g}/\text{m}^3$, respectively. CT had the second highest traffic volume, with around 500 vehicles an hour. The introduction of unleaded fuel has led to the change of fuel composition and significantly increased the levels of aromatic compounds. Toluene is one of the most popular compounds that added to unleaded fuels in order to increase the octane index during fuel reformulation. However, the increased aromatics, olefins and other organic compounds in fuels used in vehicles, which are not fitted with catalytic converters have been shown to increase emissions of aromatic VOCs [18]. Benzaldehyde (about 20% of the total determined carbonyls) at the CT was the highest among the four sampling stations. It is formed in the atmosphere from the incomplete fuel combustion and photochemical oxidation of toluene and other aromatic hydrocarbons. The high toluene concentration at the CT where many gasoline-fueled vehicles passed through possibly caused the especially high concentration of benzaldehyde at this location.

ST was located outside a semi-closed bus terminal, with over 99% of vehicles passing this station being buses (diesel vehicles). This site has the lowest traffic volume count among the four sampling station (with only around 130 vehicles per hour). However, it had similar concentration levels of most carbonyl compounds as CT. The average concentrations of FA, AA, and benzaldehyde were 8.57, 9.01, and 2.05 $\mu\text{g}/\text{m}^3$, respectively. The semi-confined bus terminal prevents a considerable

dispersion of pollutants, which resulted in high pollutant levels. Also the diesel-fueled buses (high emission rate of FA and AA) may have contributed to the high carbonyl concentrations at this location.

CU was located near to the bus and taxi terminal, which is an open area with good dispersion. The traffic volume was low, therefore most carbonyl concentrations were the lowest among the four sampling locations. The average concentrations of FA, AA, and benzaldehyde at this station were 7.43, 8.19, and 3.03 $\mu\text{g}/\text{m}^3$, respectively.

3.4. Comparison with other studies

Table 3 illustrates the comparison of the concentrations of FA and AA in the roadside microenvironments in Hong Kong with other urban cities, and the average concentrations in Hong Kong are well within the normal ranges reported in these cities. In general, the absolute amount of carbonyls would be affected by the sampling location, sampling time, environmental conditions, meteorological factors, vehicular fleets and compositions of fuels used. The mean concentrations of FA in Hong Kong were comparable to those measured in North London and Eastern Finland. But the concentration levels were lower than in West London, Brazil and Egypt. Also the FA/AA ratios in Hong Kong roadside were lower than in other cities. The ratios were around 1 or below in Hong Kong, while the ratios were between 1 and 2 in North London and Eastern Finland, and around 10 in West London. Similar concentration levels of FA and AA were obtained in Hong Kong roadside environments. However, the Mexico City in Mexico and the Cairo in Egypt had much higher concentration levels of these two pollutants than in Hong Kong roadsides due to the incomplete combustion of different fuel compositions.

4. Conclusion

The concentrations of carbonyl compounds were determined at roadside HKPU station from January 2002 to February 2002 to determine their diurnal variations. Also spatial variations of carbonyl compounds were determined by collecting samples at four other roadside monitoring locations. Nine carbonyl species were identified and quantified. The most abundant carbonyl was FA at the HKPU roadside microenvironment. The main sources of carbonyls at HKPU station came from vehicle exhaust, the FA/AA ratio varied from 1.27 to 1.35, which were slightly lower than other cities in previous studies. Temporal variations of carbonyl concentrations were observed. Both FA and AA showed peaks in the time period 09:00–12:00. The peaks were caused by the morning and evening traffic peak hours, as well as lower mixing height and degradation reaction. Strong correlations between FA and AA were observed, especially in the time period 18:00–21:00, indicating that they were coming from the same sources. Direct vehicular emissions were their principal source at nighttime, while both vehicular emissions and photochemical reactions were major sources in the daytime. When comparing the four roadside sampling stations, KC was found to have the highest average FA and AA concentrations. It is due to the high diesel traffic volume and atmospheric photooxidation, especially in summertime. The highest benzaldehyde concentration at the CT was probably due to the high toluene concentration, and photochemical oxidation of toluene resulted in high concentration of benzaldehyde. Also, the limited dispersion of air pollutants resulted in high carbonyl concentrations at the ST.

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