

Indoor and outdoor carbonyl compounds and BTEX in the hospitals of Guangzhou, China

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Abstract

Indoor and outdoor concentration levels of 21 carbonyl compounds and five BTEX (benzene, toluene, ethylbenzene and xylenes) were measured in four hospitals of Guangzhou from 2nd January to 20th March 2004. Samples were collected in five consecutive daytimes for each hospital. Among most of the samples, acetone was the most abundant carbonyl, followed by acetaldehyde, 2-butanone or formaldehyde. Toluene was the most abundant BTEX and the others were at similar levels. The relatively higher acetone concentrations might have resulted from the high level of background in Guangzhou area due to emission of the factories and LPG-fuel vehicles, and also for the special weather conditions during sampling time. The high concentration of acetaldehyde, which was even higher than that of formaldehyde, might be resulted from the wide use of ethanol in hospital. The partial oxidation of ethanol may form acetaldehyde. The indoor concentrations of carbonyls and BTEX were found a little higher than their outdoor counterparts with only a few exceptions, which showed the anthropogenic sources for these compounds. The low correlations between most carbonyls and BTEX concentrations might be caused by their complex sources. Finally, the human exposure levels of formaldehyde and acetaldehyde in hospitals are discussed.

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1. Introduction

Carbonyl compounds are common constituents of the atmosphere (Zhang and Smith, 1999), which have received attention due to their potential adverse health

effects on humans and their important role in atmospheric chemistry (Grosjean et al., 2002; Báez et al., 2003). Vehicles emissions are believed to be the most important source of carbonyl compounds in urban areas (Granby et al., 1997) and atmospheric photochemical reaction is another important source (Grosjean et al., 1990). BTEX are also one kind of harmful volatile organic compounds (VOCs), which have bad effects on human health, such as headache, eyes irritation, chest tightness, etc. (Bukowski and Meyer, 1995). BTEX are also mainly produced from vehicular emissions (Zhao et

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al., 2004), as well as other sources, such as building and furnishing materials emissions, etc. (Lee et al., 2002).

Some studies have been reported on the indoor and outdoor concentration levels of carbonyl compounds and BTEX (Williams, 1995; Sandner et al., 2001; Báez et al., 2003; Edwards et al., 2001; Kim et al., 2001; etc.). But for hospitals, special places whose air quality is very important, especially for the sick people, only a few studies have been performed (Koda et al., 1999; Cheong and Chong, 2001; Alizadeh and Zargari, 2002; Takigawa et al., 2004), of which, only a few compounds (formaldehyde, benzene or toluene, etc.) were investigated, and the sampling was performed in some special places (such as pathology laboratory, operation room or endoscope unit, etc.). The studies of carbonyl compounds and BTEX indoors in hospitals of China have seldom been reported.

In the present study, systematic measurements were carried out both indoors and outdoors in four hospitals of Guangzhou. Clinic, ward, emergency room and injection room, where most people (doctors, nurses and patients) were present, were chosen as the sampling locations. Twenty-one carbonyl compounds and five BTEX were quantified, and their possible sources are discussed. The exposures and risks of carbonyls and BTEX in hospitals are evaluated.

2. Experimental Section

2.1. Sampling sites

Samples were collected in four hospitals in Guangzhou from 2nd January to 20th March 2004 (the locations of H1–H4 are shown in Fig. 1). In each hospital, samples were collected on 5 consecutive days in the morning. The sampling time in H1 and H4 included weekdays and weekends, and in H2 and H3, sampling was performed in weekdays.

Hospital 1 (H1) is located in a suburban residential area. All kinds of vehicles (including heavy-duty diesel trucks) are permitted to pass by and traffic jams are frequent. There are many factories around this area. The injection room, ward and the outdoors (roof of four-floor building) were selected as sampling sites. Numbers of patients and visitors were about 15–30 in injection room and 20–45 in all the wards of the same floor. The building was being decorated during the sampling time; the inpatient building was finished just a year ago.

Hospital 2 (H2) is located in a street in an urban residential area with high density of population and heavy traffic. There are hospitals, restaurants and commercial streets around it. The clinic, ward and the outdoors (roof

of ten-floor building) were selected as sampling sites. Numbers of patients and visitors were about 10–25 in clinic and 15–30 in all the wards of the same floor. Some tall buildings were being built near H2 and one building near the sampling site in H2 was being decorated during the sampling time.

Hospital 3 (H3) is a paediatric hospital in an urban commercial area. Near H3, there is a famous commercial pedestrian street. The emergency room, injection room, ward and the outdoors (roof of nine-floor building) were selected as sampling sites. Numbers of patients and visitors were about 10–15 in emergency room, 20–30 in injection room and 25–40 in all the wards of the same floor.

Hospital 4 (H4) is a tuberculosis hospital in an urban commercial area surrounded by Baiyun Mountain and Luhu Park. The emergency room, ward and outdoors (roof of three-floor building) were selected as sampling sites. Numbers of patients and visitors were about 5–10 in emergency room and 5–15 in all the wards of the same floor.

2.2. Reagents

All solvents employed were HPLC grade. Water was double distilled and filtered by Milli-Q. The 2,4-dinitrophenylhydrazine (DNPH) was purchased from Fluka (USA) and further purified by recrystallizing two times in acetonitrile (Merck, Germany). The calibration standards (ChemService, USA) contained carbonyl-DNPH derivatives, including Mix 1 (DNPH derivatives of formaldehyde, acetaldehyde, acetone, acrolein, butyraldehyde, propionaldehyde, crotonaldehyde, benzaldehyde, 2,5-dimethylbenzaldehyde, hexaldehyde, isovaleraldehyde, valeraldehyde, *o*-tolualdehyde, *m*-tolualdehyde, *p*-tolualdehyde), Mix 2 (DNPH derivatives of formaldehyde, acetaldehyde, crotonaldehyde, propionaldehyde, butyraldehyde, cyclohexanone, valeraldehyde, hexaldehyde, heptaldehyde, octylaldehyde, nonanaldehyde, decylaldehyde) and 2-butanone-DNPH derivative.

2.3. Sample collection and analysis

For carbonyls, the whole method was described in detail in our previous studies (Feng et al., 2004, 2005). Owing to the interference of the laboratory air, all the laboratory operations were conducted in a vacuum glove box (ZKX2, 800×600×700 mm, Nanjing University Instrument Plant, China). The sampling medium was a Sep-Pak Silica Gel Cartridge (Waters, Millipore Corp). Each cartridge was rinsed with 10 ml of acetonitrile (ACN) slowly and then coated by passing 7 ml of the freshly made coating solution, which contained 60-ml

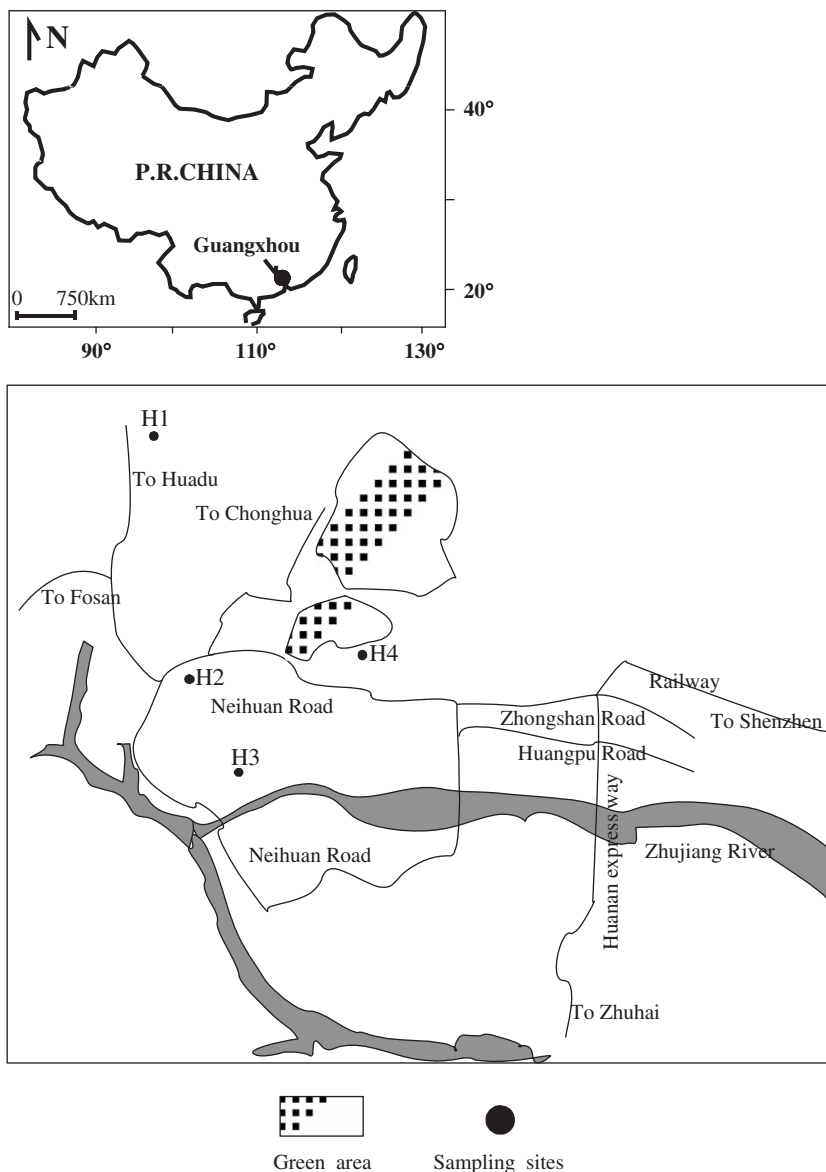


Fig. 1. Location of sampling sites.

DNPH-ACN-saturated solution and 2-ml concentrated *ortho*-phosphoric acid in 500-ml ACN, through the cartridge by gravity. When there was no more solution flowing out of the cartridge, the cartridge was dried with a gentle flow of nitrogen for 15 min. Then, the coated cartridges were wrapped in aluminum foil, sealed in hermetic Teflon bags and stored in refrigerator at 4 °C until use. Three blank cartridges from each cartridge batch were analyzed and the results were all below the EPA blank criteria.

Samples were collected by drawing the air with a sampling pump (Thomas, USA) through the cartridge.

A potassium iodide (KI) denuder was connected to the upstream of the cartridge to prevent the interference of ozone (Sirju and Shepson, 1995). The sampling duration time was about 2 h in H1, H2, H3 and about 3 h in H4, and the flow rate was 0.8–1.3 l min⁻¹, which was measured with a digital flow meter (DryCal DC Lite, Bios Corp., USA) during sampling. After sampling, each cartridge was wrapped in aluminum foil, resealed in a Teflon bag, and transported back to the laboratory and stored in the refrigerator before being analyzed. Each sampling program included one laboratory blank and one field blank. At each sampling site, two field

samples were collected with back-up cartridge to evaluate breakthrough.

The sampled cartridges were eluted slowly with 2 ml of ACN into a 2-ml volumetric flask and stored in refrigerated conditions until analysis. The HPLC system (HP1100, Agilent, USA) was used for the measurement of carbonyl compounds. A 10- μ l aliquot was injected into the HPLC system through an autosampler. The analytical conditions were as follows: Agilent SB-C18 reverse column (250 mm \times 4.6 mm \times 5 μ m); gradient mobile phase: 60–70% ACN of water solution for 20 min, 70–100% ACN for 3 min, 100% ACN for 4 min, 100–60% ACN for 1 min and then 60% ACN for 5 min; mobile-phase flow rate: 1 ml min⁻¹; detector: UV at 360 nm.

For BTEX, the samples were collected by the commercial stainless steel canister (Polar Ware Company, USA). All canisters were cleaned using ultra-pure N₂ (>99.999%) and then evacuated before sampling. Sampling and analysis were according to the US EPA Compendium Method To-14 A (US EPA, 1997). Sampling inlets were placed at about 1.2 m above the ground, through which air was drawn by opening the valve.

For analysis, BTEX samples in the canisters were concentrated in the Model 7100 Preconcentrator, and injected into an HP 6890 gas chromatography coupled to an HP 5973 mass-selective detector (GC/MSD). A RESTEK RTX-1 capillary column, 60 m \times 320 μ m \times 1.0 μ m, was used in this system. The initial temperatures was held at 40 °C for 2 min, and then increased at a rate of 6 °C min⁻¹ to 230 °C and held for 5 min. For BTEX, the detailed method was in our previous studies (Wang et al., 2002; Zhao et al., 2004).

2.4. Quality assurance and quality control

For carbonyls, identification and quantification of carbonyl compounds were based on their retention times and peak areas, respectively. The instrument was calibrated using five standard concentrations (from 0.5 to 10 μ g ml⁻¹) covering the concentration of interest. The linear relationships ($R^2 > 0.999$) between the concentrations and responses for all carbonyls identified were very good. Calibration standard was run daily to ensure the instruments are stabilized. Cartridge collection efficiency was determined with two cartridges in series and over 99% of carbonyl compounds were found in the first cartridge, which indicated complete recovery of all of the carbonyls. Relative percent differences (RPDs) for duplicate analysis were less than 5%. Method detection limits (MDLs) were determined by using seven replicate analyses of the working standards at the lowest con-

centration. The MDLs of present study were found to range from 0.05 to 0.15 μ g m⁻³ for various carbonyls of 120-l sampling volume.

For BTEX, compounds were identified by their retention times and their mass spectra. Standard gas mixtures (1.0 ppm, Supeclo To-14 Calibration Mix) were first dynamically diluted with zero air, then sampled and analyzed using identical conditions to those for the field samples, and seven-point calibration (0.0, 1.0, 5.0, 10.0, 20.0, 40.0, 50.0 ppbv) was performed for quantifying the BTEX in the air samples. The detection limits of our method for all compounds were <0.2 ppb.

3. Results and discussion

3.1. Indoor and outdoor concentration levels

Twenty-one carbonyl compounds were measured in the ambient air. The sum of *m*-tolualdehyde and *o*-tolualdehyde was reported because they could not be separated by the analytical method. Arithmetic mean concentrations of carbonyl compounds were listed in Table 1. *p*-Tolualdehyde was not detected in any of the ambient air samples, acrolein and crotonaldehyde were present in only a few samples.

Of the 21 carbonyls listed in Table 1, acetone was the most abundant carbonyl in all samples, followed by acetaldehyde, formaldehyde. The mean concentration of acetaldehyde in the injection room was the highest in all sampling sites in H1 and H3, most carbonyls concentrations were higher in H1 and H2 than in H3 and H4 (Table 1).

BTEX were measured in the hospital air, including benzene, toluene, ethylbenzene, *o*-xylene and *m,p*-xylene (these two compounds could not be separated entirely in GC and were quantified as the sum). Of the BTEX listed in Fig. 2, toluene was the most abundant BTEX in most samples and the other four BTEX did not show obvious difference in concentrations (Fig. 2).

Similar to the carbonyls, most BTEX concentrations were higher in H1 and H2 than in H3 and H4, which might be related to the higher emissions of those compounds from the environments around H1 and H2. Although the quality of the indoor air was poorer than that of the outdoor air, outdoor sources strongly influence indoor air (Baek et al., 1997). Many factories were near H1 and its inpatient building had just been built for about one year. Furthermore, the first floor and the second floor of the building in H1 were being decorated during the sampling time. Also some tall buildings were being built near H2 and one building in H2 was being decorated during the sampling time. Interior decoration

Table 1
Concentrations of carbonyl compounds in the sampling sites ($\mu\text{g m}^{-3}$) (Arith.mean \pm S.D.)

Compounds	H1			H2			H3			H4			
	I (N=5)	W (N=4)	O (N=5)	C (N=5)	W (N=5)	O (N=5)	E (N=5)	I (N=5)	W (N=4)	O (N=5)	E (N=5)	W (N=5)	O (N=5)
Formaldehyde	11.4 \pm 2.9	8.1 \pm 2.6	13.4 \pm 3.7	6.0 \pm 3.1	8.7 \pm 3.7	12.9 \pm 4.4	10.8 \pm 5.6	9.0 \pm 3.8	9.6 \pm 4.1	9.1 \pm 2.0	5.3 \pm 1.5	6.0 \pm 1.8	6.7 \pm 2.5
Acetaldehyde	14.3 \pm 7.0	13.0 \pm 3.7	11.7 \pm 5.3	21.4 \pm 9.5	13.9 \pm 2.7	17.8 \pm 6.1	7.9 \pm 2.0	17.3 \pm 5.5	8.9 \pm 2.6	8.0 \pm 2.4	9.4 \pm 3.0	10.4 \pm 2.9	9.3 \pm 3.0
Acrolein	Nd	Nd	Nd	3.7 \pm 5.7 ^a	Nd	Nd	Nd	Nd	0.33 ^b	Nd	Nd	Nd	Nd
Acetone	49 \pm 25	41 \pm 14	29 \pm 8.8	31 \pm 16	28 \pm 6.7	19.3 \pm 7.1	19 \pm 4.7	24 \pm 6.4	15.1 \pm 3.7	14.5 \pm 2.6	17.9 \pm 5.9	19.3 \pm 6.2	16.9 \pm 6.0
Propionaldehyde	3.4 \pm 1.7	3.4 \pm 1.1	2.8 \pm 1.5	6.1 \pm 5.9	3.5 \pm 0.6	2.9 \pm 0.5	2.0 \pm 0.3	2.4 \pm 0.4	1.8 \pm 0.5	2.1 \pm 0.5	2.4 \pm 0.8	2.4 \pm 0.7	2.4 \pm 0.9
Crotonaldehyde	Nd	0.7 \pm 0.1 ^c	Nd	1.9 \pm 2.3 ^d	Nd	0.7 ^b	Nd	0.8 ^b	Nd	Nd	Nd	Nd	Nd
2-Butanone	18 \pm 14	19 \pm 12	9.2 \pm 8.1	14.4 \pm 11.3	14.6 \pm 10.7	8.5 \pm 3.6	4.1 \pm 1.9	4.9 \pm 1.2	3.0 \pm 0.7	5.4 \pm 1.4	7.3 \pm 3.0	7.7 \pm 2.8	8.5 \pm 2.8
Butyraldehyde	2.6 \pm 1.1	2.5 \pm 0.6	2.1 \pm 0.9	3.8 \pm 1.6	2.5 \pm 0.5	2.5 \pm 0.4	1.7 \pm 0.1	2.1 \pm 0.3	2.0 \pm 0.5	2.1 \pm 1.1	1.8 \pm 0.5	1.9 \pm 0.5	1.9 \pm 0.5
Benzaldehyde	2.7 \pm 1.2	2.7 \pm 1.0	2.1 \pm 1.0	2.5 \pm 0.5	2.6 \pm 0.4	2.3 \pm 0.3	1.4 \pm 0.3	2.2 \pm 1.2	1.3 \pm 0.3	1.8 \pm 0.2	2.3 \pm 0.5	2.4 \pm 0.7	2.4 \pm 0.6
Isovaleraldehyde	1.0 \pm 0.4	1.0 ^b	0.9 \pm 0.2 ^a	1.1 \pm 0.1 ^d	1.1 \pm 0.2 ^a	1.2 \pm 0.2	1.0 \pm 0.1	1.1 \pm 0.1	0.9 \pm 0.1	1.0 \pm 0.2	0.8 \pm 0.1	1.0 \pm 0.2	0.9 \pm 0.2
Cyclohexanone	11.2 \pm 6.2	3.1 \pm 4.4	2.2 \pm 1.3	7.3 \pm 1.3	2.9 \pm 2.1	2.8 \pm 1.0	2.6 \pm 0.9	3.4 \pm 1.0	4.0 \pm 1.8	2.4 \pm 0.5	3.7 \pm 1.3	4.0 \pm 1.1	3.7 \pm 1.0
Valeraldehyde	1.4 \pm 0.5	7.0 \pm 4.1	2.5 \pm 3.1	1.9 \pm 0.3	3.2 \pm 3.8	1.4 \pm 0.1	1.2 \pm 0.1	1.4 \pm 0.1	1.1 \pm 0.2	1.2 \pm 0.2 ^d	1.0 \pm 0.2	1.2 \pm 0.2	1.2 \pm 0.3
<i>p</i> -Tolualdehyde	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd
<i>m/o</i> -Tolualdehyde	1.6 ^b	1.6 \pm 0.1 ^a	1.8 \pm 0.4 ^d	1.4 ^b	1.8 \pm 0.1 ^a	1.6 \pm 0.2	1.5 \pm 0.2 ^d	1.7 \pm 0.1	1.1 ^b	1.6 \pm 0.4	1.1 \pm 0.1 ^d	1.2 \pm 0.2 ^c	1.0 ^b
Hexaldehyde	2.5 \pm 0. 6	2.0 \pm 0.3	2.0 \pm 0.2	3.5 \pm 1.6	2.2 \pm 0.6 ^d	2.0 \pm 0.3	1.9 \pm 0.4	2.8 \pm 0.3	2.0 \pm 0.5	1.8 \pm 0.3	1.3 \pm 0.3	1.6 \pm 0.3	1.4 \pm 0.3
2,5-Dimethylbenzaldehyde	1.0 \pm 0.1	1.4 \pm 0.3	1.0 \pm 0.2 ^a	1.2 \pm 0.8 ^d	1.1 \pm 0.1 ^a	0.9 \pm 0.1 ^c	Nd	Nd	1.70 ^b	Nd	Nd	Nd	Nd
Heptaldehyde	1.2 \pm 0.6	0.8 \pm 0.2	0.7 \pm 0.1 ^d	1.2 \pm 0.4	1.0 \pm 0.6	1.1 \pm 0.3	0.6 \pm 0.2	0.6 \pm 0.1	0.6 \pm 0.1	0.5 \pm 0.1	0.6 \pm 0.3 ^d	0.5 \pm 0.1	0.5 \pm 0.1 ^d
Octylaldehyde	1.0 \pm 0.3	1.4 \pm 1.3	0.7 \pm 0.1	0.9 \pm 0.2	1.0 \pm 0.4	0.6 \pm 0.1	0.6 \pm 0.2	0.7 \pm 0.1	0.6 \pm 0.2	0.6 \pm 0.1	0.5 \pm 0.2	0.6 \pm 0.2	0.5 \pm 0.2
Nonanaldehyde	4.0 \pm 1.5	2.6 \pm 1.0	2.3 \pm 0.5	2.9 \pm 1.4	3.1 \pm 0.9	2.8 \pm 0.7	1.9 \pm 1.2	2.4 \pm 0.2	2.2 \pm 1.4	2.1 \pm 0.8	1.8 \pm 0.7	2.3 \pm 1.3	2.0 \pm 0.9
Decylaldehyde	0.9 \pm 0.3	0.7 \pm 0.2	0.7 \pm 0.1	0.8 \pm 0.3	0.8 \pm 0.3	0.6 \pm 0.1	0.6 \pm 0.2	0.8 \pm 0.1	0.8 \pm 0.3	0.7 \pm 0.2	0.5 \pm 0.1	0.7 \pm 0.5	0.5 \pm 0.1
Total	127 \pm 57	111 \pm 29	84 \pm 30	109 \pm 52	90 \pm 19	81 \pm 8.5	59 \pm 12	77 \pm 13	55 \pm 5.7	55 \pm 6.4	57 \pm 14	62 \pm 13	59 \pm 15

Arith.mean: arithmetic mean, S.D.: standard deviation, Nd: not detected.

N: number of samples, I: injection room, W: ward, O: outdoor, C: clinic, E: emergency room.

^a Three samples average.

^b Only one sample.

^c Two samples average.

^d Four samples average.

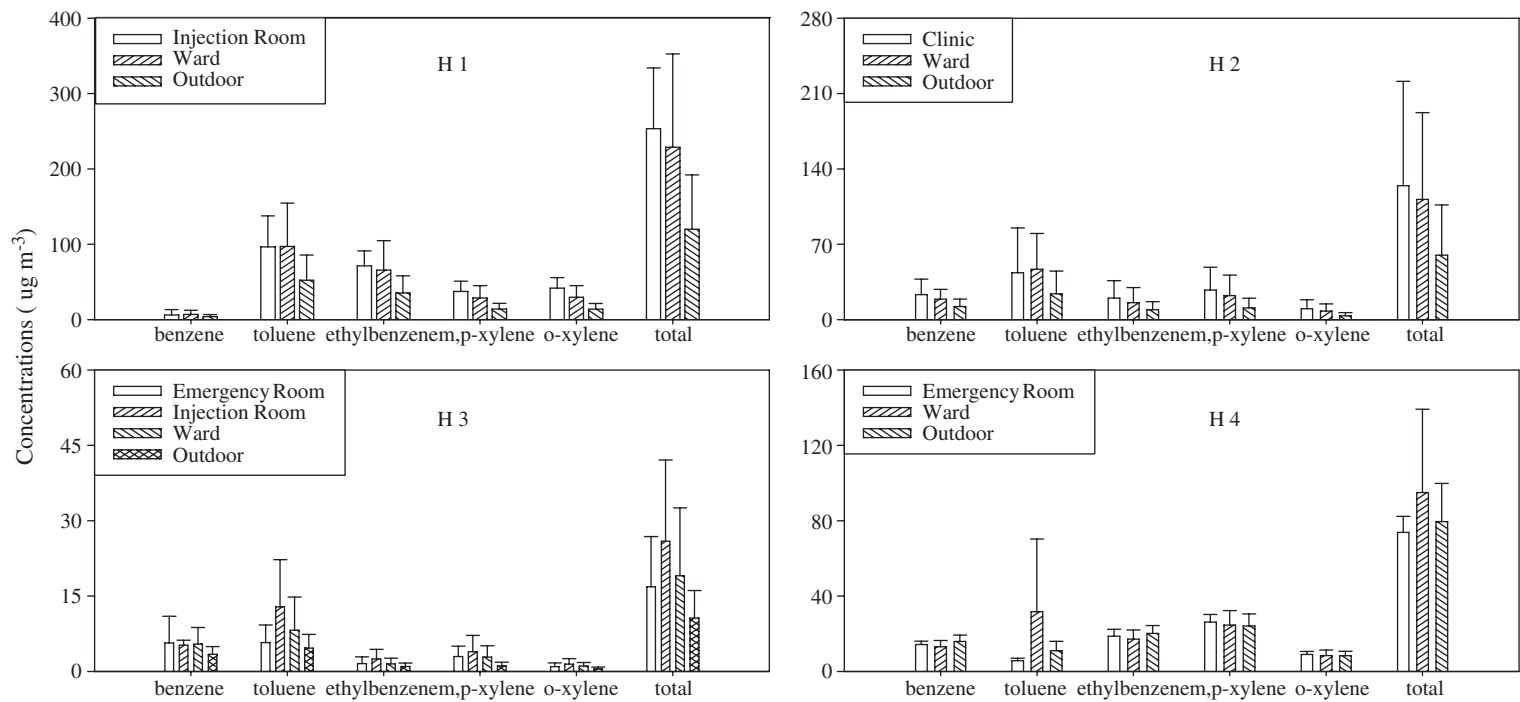


Fig. 2. Concentrations of BTEX in the sampling sites ($\mu\text{g m}^{-3}$) (Arith.mean \pm S.D.). Arith.mean: arithmetic mean, S.D.: standard deviation.

Table 2
The ratio of indoor/outdoor (I/O) carbonyl concentrations

Compound	AM	S.D.	N	Max	Min
Formaldehyde	0.88	0.46	43	2.13	0.11
Acetaldehyde	1.29	0.84	43	5.25	0.54
Acetone	1.41	0.55	43	3.62	0.73
Propionaldehyde	1.20	0.71	43	5.32	0.64
2-Butanone	1.29	0.87	43	4.36	0.37
Butyraldehyde	1.12	0.40	43	2.70	0.42
Benzaldehyde	1.07	0.41	43	2.77	0.54
Isovaleraldehyde	1.00	0.16	35	1.33	0.74
Cyclohexanone	2.26	4.15	43	27.14	0.20
Valeraldehyde	1.58	1.74	40	8.38	0.19
<i>m/o</i> -Tolualdehyde	0.90	0.26	19	1.38	0.50
Hexaldehyde	1.24	0.46	42	3.47	0.76
2,5-Dimethylbenzaldehyde	1.58	0.64	8	2.77	0.82
Heptaldehyde	1.20	0.52	38	3.24	0.32
Octylaldehyde	1.34	0.57	43	4.10	0.77
Nonanaldehyde	1.16	0.50	43	2.35	0.40
Decylaldehyde	1.19	0.38	43	2.15	0.68
Total	1.20	0.40		2.80	0.84

AM: the arithmetic mean of every pair I/O for all the sampling sites.
S.D.: standard deviation, N: number of samples.

works and the use of industrial solvents could significantly increase the indoor levels of VOCs (Lee et al., 2002).

Previous studies of carbonyl and BTEX levels in hospitals were showed as follows: the indoor formaldehyde concentrations were 369–2829 $\mu\text{g m}^{-3}$ (in pathology division, Koda et al., 1999), 123–369 $\mu\text{g m}^{-3}$ (in the administration offices of a hospital building, Cheong and Chong, 2001) and 20 $\mu\text{g m}^{-3}$ (in the rooms of newly constructed hospital, Takigawa et al., 2004), respectively. According to the study performed in pathologic laboratory and operation room of governmental and private hospitals (Alizadeh and Zargari, 2002), the formaldehyde concentrations were 750 $\mu\text{g m}^{-3}$ and 492 $\mu\text{g m}^{-3}$, 541 $\mu\text{g m}^{-3}$ and 357 $\mu\text{g m}^{-3}$, respectively. The concentrations of BTEX were 463 $\mu\text{g m}^{-3}$ for toluene, 252 $\mu\text{g m}^{-3}$ for ethylbenzene, 476 $\mu\text{g m}^{-3}$ for *m,p*-xylene and 250 $\mu\text{g m}^{-3}$ for *o*-xylene (in the rooms of newly constructed hospital, Takigawa et al., 2004). Comparing to the data listed in Table 1 and Fig. 2, it could be seen that in our study, the concentration levels were several to hundreds of times lower. This might result from the special use of reagent in the particular sampling sites of the previous studies and also may be due to the good ventilation of the sampling sites in present study.

3.2. Indoor/outdoor ratios

By comparison of the indoor and outdoor carbonyl and BTEX concentrations, it was found that, except

formaldehyde, *m/o*-tolualdehyde and a few BTEX samples, the ratios of indoor/outdoor (I/O) were all >1 (Fig. 2, Table 2), resulting from the indoor sources for most compounds (e.g., emission of construction and decoration materials, some medical reagents, etc.). It was reported that indoor ozone chemistry could play a role in generating indoor aldehydes, which were generated by the reaction of ozone with VOCs (Weschler et al., 1992). As also shown in Fig. 2 and Table 2, most I/O ratios were only a little higher than 1, which showed the important role of ventilation. During the sampling time, the windows and doors were always open, outdoor infiltration might be one of the main source of indoor carbonyls. And it should be pointed out that in H1 and H2, the concentrations of acetone and 2-butanone indoors were obviously higher than outdoors (Tables 1 and 2), which might be due to the decorations in H1 and H2 near the sampled room. These compounds could have been emitted from some paints and diffused into the sampling room (Crump and Gardiner, 1989).

3.3. The concentration ratios of carbonyl compounds

Formaldehyde/acetaldehyde (C_1/C_2) and acetaldehyde/propionaldehyde (C_2/C_3) ratios were calculated (Table 3). The C_1/C_2 ratios usually varied from 1–2 (urban area) to about 10 (rural or forest area); therefore, it could be used as a measure of the biogenic source of formaldehyde (Shepson et al., 1991). However, the C_1/C_2 ratios was not constant and often showed large variations, so care must be taken if using such ratios to identify the source of carbonyl compounds. The average C_1/C_2 ratio value in the present study was 0.82, which might be from the result of the widespread use of ethanol. In Rio de Janeiro (Brazil), acetaldehyde/formaldehyde ratios measured in the 1980s were obviously >1 (units of ppb/ppb), with average values of up to 3.0 (Grosjean et al., 2002), due to the extensive use of ethanol as fuel throughout the country. C_2/C_3 ratios were often used as an indicator of anthropogenic origin for ambient carbonyls, since propionaldehyde was believed to be associated only with anthropogenic emissions. C_2/C_3 would be high in rural atmospheres and low in polluted urban air. The average

Table 3
The concentration ratios of formaldehyde/acetaldehyde (C_1/C_2) and acetaldehyde/propionaldehyde (C_2/C_3)

Ratio	N	AM	S.D.	Max	Min
C_1/C_2	63	0.82	0.45	2.21	0.10
C_2/C_3	63	5.00	1.75	13.95	2.28

N: number of the samples.

AM: arithmetic mean, S.D.: standard deviation.

C_2/C_3 ratios value in this study was 5.0, which was quite similar to that of some urban cities, e.g., Rome: 5.2 (Possanzini et al., 1996). It implied that anthropogenic emission was an important source of carbonyls in urban areas of Guangzhou.

3.4. Mutual correlations and source implications

The correlations between the 14 carbonyl compounds and the 5 BTEX compounds detected in the ambient air are shown in Table 4. Low correlations were found between most carbonyls and BTEX, possibly implying complex sources.

Usually, direct emissions from motor vehicles and other combustion sources are believed to be the most important sources of carbonyls. But other sources, e.g., photochemical reactions, might only contribute a minor part of such compounds in the present study. The sampling time of the present study was from January to March 2004, the late winter and early spring in Guangzhou. The weather was usually cloudy and the temperature was low, so photochemical reactions might be much weaker than in summer. For the formaldehyde concentration, this might be the reason for the relatively lower level measured in the present study than that of the previous work (Feng et al., 2005). Both studies were carried out in Guangzhou, but the samples were taken in winter and summer, respectively. The difference in formaldehyde concentrations might also reflect the importance of photochemical reactions as a source of atmospheric formaldehyde.

The high ambient acetone concentrations detected in the present study were partly from the high outdoor level of acetone in Guangzhou. Acetone is widely used as industrial solvent in many paint manufacturing and other industries (there are large numbers of such industries distributed in Guangzhou). And it might also come from the long lifetime of acetone for removal by photolysis and OH reaction, which are approximately 40 and 20 days in the atmosphere, respectively. But for formaldehyde and acetaldehyde, the lifetimes are only about 1.5 h to 1 day (Shepson et al., 1991; Atkinson, 2000). As described above, the weather status during the sampling was favorable for the long duration of emitted acetone. Another reason might be the emission of buses and taxis using LPG (liquefied petroleum gas) as fuel. The numbers of these types of vehicle have increased rapidly during recent years in Guangzhou. In a similar study in Mexico City (Báez et al., 2003), high concentration levels of acetone were also mainly derived from the emission from LPG fuel. In Mexico City, LPG is widely used fuel in hotels, houses and many small industries.

The concentration of 2-butanone was relatively higher than in previous studies conducted in other cities (Possanzini et al., 1996; Grosjean et al., 2002) and in Guangzhou (Feng et al., 2004; Feng et al., 2005). The correlation between 2-butanone and acetone was >0.8 , which might show the similar sources for these two carbonyl compounds. Similar to acetone, 2-butanone is an important component of industrial solvents and disinfectants (Müller et al., 2002), and also has a relatively long life time in the atmosphere (Atkinson, 2000). The result above showed that both acetone and 2-butanone might mainly come from the emission of reagents used in industry, and they both accumulated in the air under the weather conditions while sampling.

The relatively high level of acetaldehyde concentrations both indoors and outdoors, which was higher than most data of other studies (Christensen et al., 2000; Ho et al., 2002; Feng et al., 2005), might have resulted from the wide employment of ethanol as disinfectant in hospital. Ethanol could evaporate to air and react with OH to form acetaldehyde. As reported in the previous study, ethanol was also used to clean surfaces (notably glass windows) in Brazil, and the acetaldehyde concentration was much higher indoors than that outdoors, implying a the source of acetaldehyde indoors (Brickus et al., 1998). And for indoors, it might also come from the emissions from people (Zhang et al., 1994). Acetaldehyde is a product of human metabolism, and many patients and doctors were near the sampling sites when the samples were collected. There were so many patients in the injection rooms that the direct emissions of metabolites from humans in the injection rooms might be another reason for the highest levels of acetaldehyde concentration detected in all sampling sites in H1 and H3.

The correlations between propionaldehyde and butyraldehyde, nonanaldehyde and decylaldehyde were all >0.8 , implying common sources. Recent studies found that some plants could emit many kinds of carbonyls, most of which were high molecular weight carbonyls (Villanueva et al., 2004). There are many flowers, grass and green plants in Guangzhou throughout the year due to its climate, and there were large quantities of trees and grass around the sampling sites, so the emission of plants might be another source of carbonyls, especially for some high molecular weight carbonyls (Feng et al., 2004).

Except benzene, the correlations between the concentrations of other BTEX compounds (toluene, ethylbenzene, *m,p*-xylene and *o*-xylene) were good in general, suggesting that they might have common sources, and most important sources may be vehicle emissions. In Guangzhou, poor correlations between

Table 4
Concentration correlations of carbonyl compounds and BTEX in the ambient air in the four hospitals

Compounds	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Formaldehyde 1																		
Acetaldehyde 2	0.28*																	
Acetone 3	0.22	0.54**																
Propionaldehyde 4	0.15	0.75**	0.57**															
2-Butanone 5	0.07	0.50**	0.84**	0.62**														
Butyraldehyde 6	0.17	0.78**	0.59**	0.82**	0.58**													
Benzaldehyde 7	0.11	0.46**	0.62**	0.48**	0.72**	0.52**												
Cyclohexanone 8	0.10	0.34**	0.66**	0.33**	0.44**	0.42**	0.34**											
Valeraldehyde 9	-0.004	0.24	0.43**	0.24	0.57**	0.29*	0.43**	-0.20										
Hexataldehyde 10	0.12	0.46**	0.26*	0.29*	0.03	0.53**	0.08	0.35**	-0.02									
Heptaldehyde 11	0.16	0.60**	0.51**	0.55**	0.54**	0.60**	0.40**	0.36**	0.31*	0.37**								
Octylaldehyde 12	0.04	0.18	0.29*	0.19	0.23	0.26*	0.15	0.09	0.46**	0.27*	0.39**							
Nonanaldehyde 13	0.19	0.28*	0.17	0.15	0.07	0.28*	0.12	0.16	0.17	0.58**	0.54**	0.52**						
Decylaldehyde 14	0.13	0.23	0.18	0.13	0.02	0.28*	0.06	0.19	0.14	0.59**	0.42**	0.48**	0.85**					
Benzene 15	-0.028*	0.21	0.10	0.16	0.25	0.24	0.30*	-0.001	0.21	-0.03	0.32*	0.07	0.23	0.20				
Toluene 16	0.16	0.11	0.48**	0.14	0.32*	0.17	0.22	0.25	0.36**	0.13	0.2	0.33*	0.35**	0.25	0.17			
Ethylbenzene 17	0.12	0.07	0.52**	0.11	0.33*	0.12	0.26	0.30*	0.38**	0.05	0.18	0.37**	0.38**	0.26	0.07	0.91**		
<i>m,p</i> -Xylene 18	-0.04	0.11	0.39**	0.11	0.31*	0.12	0.34**	0.26*	0.26	-0.02	0.19	0.19	0.34**	0.24	0.56**	0.79**	0.80**	
<i>o</i> -Xylene 19	0.15	0.07	0.57**	0.10	0.34**	0.11	0.24	0.46**	0.24	0.06	0.20	0.27*	0.38**	0.24	0.10	0.85**	0.95**	0.80**

* $p < 0.05$.

** $p < 0.01$.

Table 5
Comparison of exposure risks of formaldehyde and acetaldehyde in the indoor air

Parameter	Formaldehyde				Acetaldehyde			
	Arith. mean	95th percentile	Risk		Arith. mean	95th percentile	Risk	
			Mean	95th percentile			Mean	95th percentile
<i>Office</i> ^a								
<i>C</i> ($\mu\text{g m}^{-3}$)	26.2	34.4	3.4×10^{-4}	4.5×10^{-4}	19.3	32.7	4.3×10^{-5}	6.1×10^{-5}
<i>E</i> ($\mu\text{g day}^{-1}$)	132	173	–	–	97	165	–	–
<i>Ballroom</i> ^b								
<i>C</i> ($\mu\text{g m}^{-3}$)	33.1	55	4.4×10^{-4}	7.2×10^{-4}	100.1	225.7	22.6×10^{-5}	51×10^{-5}
<i>E</i> ($\mu\text{g day}^{-1}$)	124	209	–	–	378	853	–	–
<i>Hospital</i> ^c								
<i>C</i> ($\mu\text{g m}^{-3}$)	8.3	10.9	1.1×10^{-4}	1.4×10^{-4}	12.9	20.3	2.8×10^{-5}	4.5×10^{-5}
<i>E</i> ($\mu\text{g day}^{-1}$)	41.8	54.9	–	–	65	102	–	–

For exposure calculus, the inhalation rate of air was estimated for an average person ($\text{IR} = 0.63 \text{ m}^3 \text{ h}^{-1}$) according to EPA exposure factors (US EPA, 1990). The inhalation unit risk estimates of formaldehyde and acetaldehyde were 1.3×10^{-5} and $2.2 \times 10^{-6} (\mu\text{g m}^{-3})^{-1}$, respectively. A mean residence time of 8 h (official working time) was considered as exposure time (t) for a and c, and 6 h in the ballroom was used for b.

^a Báez et al. (2003).

^b Feng et al. (2004).

^c This study.

benzene and other BTEX species might imply that benzene had important sources other than vehicle emission (Wang et al., 2002).

3.5. Exposure and risk

The exposure (E) for an individual (i) due to intake process (inhalation and ingestion) can be calculated from the equation of the US EPA (US EPA, 1992):

$$E_i = C_j \text{IR}_i t_{ij}$$

Where C is the concentration of the pollutant ($\mu\text{g m}^{-3}$), IR is the inhalation rate ($\text{m}^3 \text{ h}^{-1}$), t is the exposure time (h day^{-1}) and j is the microenvironment.

In this study, hospitals were selected to calculate the exposure (E), of which the exposure time (t) was based upon residence time, a mean residence time of 8 h was considered. The mean and the 95th percentile exposure for indoors of hospitals (Table 5) were estimated in comparison with some other places. The exposure for formaldehyde and acetaldehyde in the hospitals in Guangzhou was lower than in the ballrooms in Guangzhou (Feng et al., 2004) and office in Mexico City (Báez et al., 2003).

For BTEX exposure risk, only benzene is simply evaluated for the lack of such information. The results showed that all the air samples in this study did not exceed the benzene limit of $30 \mu\text{g m}^{-3}$ (the 30-min ambient air quality standard defined by USA; Zhao et al., 2004).

The risk might represent the relatively good air conditions in hospitals of Guangzhou. After the SARS event, people here were more concerned about the air ventilation of the hospital and the results of present study showed its good effect on air quality.

4. Conclusion

In four hospitals of Guangzhou, air samples were collected to evaluate the carbonyl compounds and BTEX levels both inside and outside. 21 kinds of carbonyls were detected, among which acetone was the most abundant, followed by acetaldehyde and formaldehyde. Toluene was the most abundant compound of BTEX. This level was relatively lower for BTEX and higher for carbonyl compounds compared to previous studies in Guangzhou. The low correlations between carbonyls and BTEX showed the complex sources of vehicular exhaust, industrial, plants, building, decoration materials, etc. Emissions from factories, the weather conditions while sampling and the increase in numbers of LPG-fuel vehicles might cause the relatively high level of acetone, and the wide use of ethanol in hospital might result in the high level of acetaldehyde concentrations and low concentration ratio of C_1/C_2 carbonyls. The relatively high correlation between acetone and 2-butanone might indicate a similar source, e.g., from industrial use as a reagent. Risks caused by formaldehyde and acetaldehyde in hospitals were low compared to some previous work.

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