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Carbonyl compounds and BTEX in the special rooms of hospital in Guangzhou, China

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ABSTRACT

The occurrence of carbonyl compounds and benzene, toluene, ethylbenzene and xylenes (BTEXs) was assessed in the indoor and outdoor air of a hospital in Guangzhou, China. The pharmacy room, the preparing traditional Chinese medicine room, the supply room (where the medical appliances are disinfected), the laundry and the garbage room were selected as sampling sites. Acetaldehyde (ranging from 4.56 to 66.8 $\mu g\,m^{-3}$) was in all samples the most abundant among the 18 carbonyls detected, and toluene (ranging from 33.5 to 264 $\mu g\,m^{-3}$) among the BTEXs. The indoor/outdoor (I/O) concentration ratios of BTEXs in the morning were always >1.0, and close to 1.0 or slightly <1.0 in the afternoon, while the concentration ratios of carbonyls in the afternoon showed large variation. These ratios demonstrate the significance of outdoor emissions that deteriorate the indoor air quality at the various rooms of the hospital. The possible sources of BTEXs and carbonyls in these rooms are discussed with the use of specific ratios and with the use of statistical methods, like principal components analysis.

1. Introduction

As volatile organic compounds (VOCs) are classified organic compounds that have a boiling point between 50–100 °C and 240–260 °C, and the most important category of VOCs are the group of benzene, ethylbenzene, toluene, and xylenes (BTEXs) and the carbonyls, especially the two smaller molecules of this group, that is formaldehyde and acetaldehyde. The BTEXs are widely used in many household products such as paints, varnishes, waxes, solvents, detergents and can also be emitted by the use of other products, such as printers, photocopiers, etc. They are known to be toxic and genotoxic and they also actively participate in the photochemical reactions [1–5]. Carbonyl compounds are present ubiquitously in the atmosphere, and they have received scientific and regulatory attention due to their potential adverse health effects on humans and to their important role in atmospheric chemistry. Ambient carbonyls are mainly emitted from incomplete combustion of fossil fuels (e.g., motor vehicle exhaust) and/or biomass, and also formed through atmospheric photochemical reactions. Typical indoor carbonyl sources include off-gassing from building materials, furniture, and consumer products and through reactions between indoor ozone and alkenes. Human activities that include combustion processes such as tobacco smoking, cooking, and heating, are other significant indoor sources of several carbonyls [3,6–7].

On average, people spend indoors 80–90% of their time, mainly among their workplaces and their homes and that underlines the need of knowing the quality of the indoor air. That need becomes even more evident when it comes to specific places of high interest. Clearly, hospitals are places where the air quality is very important, especially for people suffering from asthma or other diseases in the respiratory system. In the past, only few studies investigated the indoor/outdoor carbonyls or BTEXs in sites of particular interest (e.g., hospitals) [1–2,8–9]. Some other studied the air quality in some parts of a hospital [10–14]. For example, the study of Dascalaki et al. [12] investigated the air quality in hospital operating rooms, whilst Wang et al. [14] reported hospital indoor respirable particles and carbonaceous composition. A previous study of our group reported the levels and distributions of carbonyls and/or BTEXs in specific places of four hospitals (injection room, ward, clinic, and emergency room) and found that all the target compounds of that study were detected in the indoor of four hospitals [2].

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The objectives of this study were to investigate further the indoor air quality of specific hospital rooms, to compare it with the outdoor air quality and to examine whether there are significant indoor sources of VOCs. The measurement data were statistically treated to further highlight the potential sources of VOCs.

2. Materials and methods

2.1. Chemicals and materials

All organic solvents used were HPLC grade. Water was re-distilled and filtered by Milli-Q before use. The 2,4-dinitrophenylhydrazine (DNPH) was purchased from Fluka (USA) and further purified by recrystallizing twice in acetonitrile (Merck, Germany). A composite stock standard solution (ChemService, USA) contained 21 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.

The cartridge was employed as the sampling medium. Then 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 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: concentrations of formaldehyde, acetaldehyde and acetone per cartridge were <0.15, <0.10 and 0.30 μg, respectively, and those of other carbonyls per cartridge were <0.10 μg [9,15–16].

2.2. Sample collection

The samples were collected from a hospital located in Guangzhou, South China. In all sampling dates, samples were collected both in the morning (AM) and in the afternoon (PM). The pharmacy room, the preparation room (where traditional Chinese medicines are produced), supply room (where disinfecting of the medical appliances takes place), the laundry room and the garbage room were selected for the collection of the samples. The indoor and outdoor samples were collected simultaneously. The indoor air samples were taken at a height of 1.2 m above the floor (outdoor samples as well) in the center and the near-wall part of each investigated room. The outdoor sampling sites were between the garbage and the preparation room and given the vicinity of the selected indoor rooms, they were considered as representative.

The detailed method of sampling has been presented elsewhere [2,9]. Briefly, samples for carbonyls analysis were collected by drawing the air with a sampling pump (Thomas, USA) through the cartridge. The sampling duration was 2 h, and the flow rate was 0.75–1.17 L min^{-1}. After sampling, each cartridge was wrapped in aluminum foil, resealed in a Teflon bag, transported back to the laboratory and stored in the refrigerator before being analyzed. Each sampling program included one laboratory and one field blank. At each sampling site, two field samples were collected with back-up cartridge to evaluate breakthrough.

For BTEXs, the samples were collected by the commercial stainless steel canister (Polar Ware Company, USA). All canisters were cleaned using ultra-pure N_2 (>99.999%) and then evacuated before sampling. Sampling and analysis were according to the US EPA Compendium Method To-14 A [2].

2.3. Analysis procedure, quantification and QA/QC

For the analysis procedure, the cartridges were eluted slowly with 2 mL of acetonitrile (ACN) into a 2-mL volumetric flask. A 10-μL aliquot was injected into the HPLC system (HP1100, Agilent, USA) through an auto-sampler. The analytical conditions were as follows: an Agilent SB-C18 reverse column (250 mm × 4.6 mm × 5 μm); 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.

Identification of carbonyl compounds was based on the comparison of retention time between samples and the standard solution containing 21 carbonyls. Quantification was performed by integration of peak areas. The instrument was calibrated using five standard concentrations (from 0.5 to 10 μg mL^{-1}) covering the concentrations of interest. The strong linear relationships ($R^2 > 0.999$) were obtained between the concentrations and responses for all carbonyls identified. Calibration standard was run daily to ensure the instruments being stabilized. Cartridge collection efficiency was determined with two cartridges in a series, and over 99% of carbonyl compounds were found in the first cartridge, indicating almost no breakthrough of the first cartridge. Relative percent differences 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 concentration. The MDLs ranged from 0.05 to 0.15 μg m^{-3} for various carbonyls of 120-L sampling volume. It should be noted that this study presents the sum concentrations of *m*-xylenes and *p*-xylenes, and those of *m*-tolualdehyde and *o*-tolualdehyde, because *m*-xylenes and *p*-xylenes could not be separated by the analytical method, and the same for *m*-tolualdehyde and *o*-tolualdehyde.

Samples in the canisters for BTEXs analysis 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 × 320 μm × 1.0 μm, 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 [2]. The compounds of interest were identified by their retention times and their mass spectra. Standard gas mixtures (1.0 ppm, Supelco 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. Concentrations of BTEX and carbonyls

All BTEXs were detected in the hospital indoor air. The sum concentrations of the five compounds (ΣBTEXs) varied from 65.8 to 819 μg m^{-3} (Table 1). Toluene (ranging between 33.5 and 264 μg m^{-3}) was the most abundant among BTEXs, followed by *m*,*p*-xylenes. Benzene, exhibited significant lower concentration ranging from 4.23 to 27.2 μg m^{-3}. During AM samplings, the highest concentrations of both individual compounds and ΣBTEXs were

Table 1
Concentrations of BTEX in special sites of hospitals ($\mu g\,m^{-3}$).

	Pharmacy		Preparation		Supply		Laundry		Garbage		Outdoor		Min	Max	Mean
	AM[a]	PM[b]	AM	PM	AM	PM	AM	PM	AM	PM	AM	PM			
Benzene	27.2	9.56	9.25	5.02	10.73	7.7	12.5	4.23	19.4	4.45	9.96	9.05	4.23	27.2	10.8
Toluene	264	111	87.1	40.9	119	89.9	80.5	33.5	150	36.1	76.2	88.4	33.5	264	98.0
Ethylbenzene	184	56.9	55.0	14.2	81.9	57.2	43.6	8.41	126	16.4	45.7	44.6	8.41	184	61.1
m,p-Xylenes	239	76.7	75.4	20.9	145	101	66.0	14.7	171	22.9	66.5	66.2	14.7	239	88.8
o-Xylenes	105	32.4	33.6	7.86	62.0	41.5	27.2	5.06	79.8	11.8	28.2	30.3	5.06	105	38.7
ΣBTEX[c]	819	286	260	88.8	418	297	230	65.8	546	91.7	227	239	65.8	819	297

[a] AM, sample collected in the morning.
[b] PM: sample collected in the afternoon.
[c] ΣBTEX: The sum concentration of five BTEX.

observed in the pharmacy room, followed by the garbage room. BTEXs concentrations of these two rooms were considerably higher than those in the laundry and the traditional Chinese medicine preparation rooms. Moreover, the total BTEXs levels in AM samples for all rooms were higher than the outdoor levels. During PM samplings, the concentrations of individual compounds and ΣBTEXs in each room were far lower than those in the corresponding AM. Currently, there is a guideline in China regulating the indoor concentration of BTEXs (110 $\mu g\,m^{-3}$ for benzene, 200 $\mu g\,m^{-3}$ for toluene, 200 $\mu g\,m^{-3}$ for Xylenes) [17]; and as it can be seen, the concentrations found in the hospital of Guangzhou are far lower than the aforementioned guidelines with two exceptions, that are toluene and xylenes in the preparing traditional Chinese medicine room).

Nineteen out of the 21 carbonyls were detected in the air samples and their concentrations are presented in Table 2. Acrolein and isovaleraldehyde were not detected in any samples. Furthermore, the acetone concentrations in the samples were not quantified because of the high acetone concentration on blank cartridges. The sum concentrations of the 18 carbonyl compounds detected ranged from 18.0 to 106 $\mu g\,m^{-3}$, being in similar levels to those observed in other studies [2]. Acetaldehyde (varying between 4.56 and 66.8 $\mu g\,m^{-3}$) was the most abundant carbonyls in all samples (Table 2), accounting for 25.3 to 62.8% of the total concentrations of the 18 carbonyls (Table 3). Butyraldehyde ranged between 1.42 and 9.40 $\mu g\,m^{-3}$ and accounted for 4.5–23.7% of the total concentrations of 18 carbonyls. Formaldehyde was detected in very low levels (1.03–7.55 $\mu g\,m^{-3}$) and its higher values were in the garbage room (6.62–7.55 $\mu g\,m^{-3}$). Similarly to Benzene, there exists a guideline in China for the levels of formaldehyde in the indoor air (100 $\mu g\,m^{-3}$) [17], which was in no case exceeded by the hospital rooms in this study.

Similarly to the BTEXs, during AM the concentrations of carbonyls in all rooms were greater than those in the corresponding PM (except for formaldehyde, 2-butanone, heptaldehyde in the pharmacy room) (Table 2). This phenomenon could be partly explained by the operation or usage of these rooms. The pharmacy room was filled with various medicines and there was heavy odor of medicine; its door was closed for safety in AM and opened in PM. In AM the garbage room was full of hospital rubbish which was also giving the sense of bad odor which was fully eliminated during PM samplings. The laundry room was operative in AM and in PM this room was used to deal with the clean clothes. Concerning the supply room, there were a lot of medical appliances which are disinfected by ozone or other solutions. The relatively higher concentrations of formaldehyde and acetaldehyde at that room could be the result of secondary formation of carbonyls, through the reaction of the free ozone with non-saturated organic compounds.

3.2. Indoor/outdoor (I/O) concentration ratios

As shown in Table 1, the indoor concentrations of BTEXs in AM were higher than those in outdoor air, but the respectives in PM, as well as carbonyls in some cases, were lower than those in the out-

Table 2
Concentrations of carbonyl compounds in special sites of hospitals ($\mu g\,m^{-3}$).

	Pharmacy		Preparation		Supply		Laundry		Garbage		Outdoor	Min	Max	Mean
	AM[a]	PM[b]	AM	PM	AM	PM	AM	PM	AM	PM	PM			
Formaldehyde	2.89	5.22	1.19	1.03	1.32	2.33	1.28	1.85	7.55	6.62	1.1	1.03	7.55	2.94
Acetaldehyde	14.3	12.2	17.9	10.2	20.9	13.7	10.9	4.56	66.8	21.4	9.55	4.56	66.8	18.4
Acrolein	ND[c]	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	–	0.00	0.00
Propionaldehyde	3.3	3.05	3.14	1.08	2.80	1.24	2.20	0.89	5.34	2.56	1.83	0.89	5.34	2.49
Crotonaldehyde	0.1	0.14	0.12	0.07	0.08	0.1	0.09	ND	0.37	0.21	0.30	0.07	0.37	0.16
2-Butanone	0.23	3.65	0.18	0.1	0.18	0.07	0.18	0.09	0.38	0.17	1.15	0.07	3.65	0.58
Butyraldehyde	7.79	1.83	9.4	4.47	6.8	4.07	6.65	4.15	9.38	6.03	1.42	1.42	9.40	5.64
Benzaldehyde	2.33	1.94	1.22	0.82	0.9	0.75	1.12	0.81	2.96	1.51	1.74	0.75	2.96	1.46
Isovaleraldehyde	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	–	0.00	0.00
Cyclohexanone	0.66	0.57	0.71	0.31	0.38	0.38	0.6	0.35	0.96	0.53	0.28	0.28	0.96	0.52
Valeraldehyde	2.70	2.38	2.30	1.37	6.75	3.93	1.69	1.33	4.97	2.76	0.60	0.60	6.75	2.80
p-Tolualdehyde	0.08	0.12	ND	ND	0.12	0.14	0.10	ND	ND	ND	0.18	–	0.18	0.07
m/*o*-Tolualdehyde	0.24	0.16	0.37	0.12	0.46	0.24	0.27	0.44	0.56	0.13	0.66	0.12	0.66	0.33
Hexaldehyde	1.03	0.79	ND	ND	0.16	ND	ND	ND	0.32	0.14	0.08	–	1.03	0.23
2,5-Dimethyl-benzaldehyde	8.92	6.79	1.17	0.65	1.96	1.19	1.33	1.17	2.85	1.78	0.15	0.15	8.92	2.54
Heptaldehyde	0.17	0.37	0.25	0.16	0.34	0.22	0.08	0.21	0.54	0.3	0.25	0.08	0.54	0.26
Octylaldehyde	0.64	0.47	0.53	0.33	0.58	0.42	0.36	0.34	0.78	0.50	0.39	0.33	0.78	0.49
Nonanaldehyde	1.11	0.88	1.09	0.91	2.08	1.61	1.07	1.36	2.30	1.64	1.63	0.88	2.30	1.43
Decylaldehyde	0.2	0.11	0.16	0.17	0.43	0.48	0.28	0.47	0.38	0.34	0.32	0.11	0.48	0.30
Total	46.7	40.7	39.7	21.8	46.2	30.8	28.2	18.0	106	46.6	21.6	18.0	106	40.6

[a] AM: sample collected in the morning
[b] PM: sample collected in the afternoon.
[c] ND: not detectable.

Table 3
Percentage distribution (%) of various carbonyls to the total concentrations.

	Pharmacy		Preparation		Supply		Laundry		Garbage		Outdoor
	AM	PM	AM	PM	AM	PM	AM	PM	AM	PM	PM
Formaldehyde	6.2	12.8	3.0	4.7	2.9	7.6	4.6	10.3	7.1	14.2	5.1
Acetaldehyde	30.6	30.0	45.0	46.9	45.2	44.3	38.6	25.3	62.8	45.8	44.2
Propionaldehyde	7.1	7.5	7.9	5.0	6.1	4.0	7.8	4.9	5.0	5.5	8.5
Crotonaldehyde	0.2	0.3	0.3	0.3	0.2	0.2	0.3		0.4	0.5	1.4
2-Butanone	0.5	9.0	0.5	0.5	0.4	0.2	0.6	0.5	0.4	0.4	5.3
Butyraldehyde	16.7	4.5	23.7	20.5	14.7	13.2	23.6	23.0	8.8	13.0	6.6
Benzaldehyde	50	4.8	3.1	3.8	2.0	2.4	4.0	4.5	2.8	3.2	8.0
Cyclohexanone	1.4	1.4	1.8	1.4	0.8	1.2	2.1	1.9	0.9	1.1	1.3
Valeraldehyde	5.8	5.8	5.8	6.3	14.6	12.8	6.0	7.4	4.7	5.9	2.8
p-Tolualdehyde	0.2	0.3	-	-	0.3	0.5	0.4	-	-	-	0.8
m/*o*-Tolualdehyde	0.5	0.4	0.9	0.6	1.0	0.8	1.0	2.4	0.5	0.3	3.1
Hexaldehyde	2.2	1.9	-	-	0.4	-	-	-	0.3	0.3	0.4
2,5-Dimethyl-benzaldehyde	19.1	16.7	3.0	3.0	4.2	3.9	4.7	6.5	2.7	3.8	0.7
Heptaldehyde	0.4	0.9	0.6	0.7	0.7	0.7	0.3	1.2	0.5	0.6	1.2
Octylaldehyde	1.4	1.2	1.3	1.5	1.3	1.4	1.3	1.9	0.7	1.1	1.8
Nonanaldehyde	2.4	2.2	2.7	4.2	4.5	5.2	3.8	7.6	2.2	3.5	7.5
Decylaldehyde	0.4	0.3	0.4	0.8	0.9	1.6	1.0	2.6	0.4	0.7	1.5

Table 4
Indoor/outdoor (*I*/*O*) concentration ratios of BTEX in hospitals.

	Pharmacy		Preparation		Supply		Laundry		Garbage	
	AM	PM	AM	PM	AM	PM	AM	PM	AM[a]	PM[a]
Benzene	2.73	1.06	0.93	0.55	1.08	0.85	1.26	0.47	1.95	0.49
Toluene	3.47	1.25	1.14	0.46	1.56	1.02	1.06	0.38	1.97	0.41
Ethylbenzene	4.03	1.28	1.20	0.32	1.79	1.28	0.95	0.19	2.75	0.37
m,*p*-Xylenes	3.60	1.16	1.13	0.32	2.18	1.53	0.99	0.22	2.57	0.35
o-Xylenes	3.71	1.07	1.19	0.26	2.20	1.37	0.96	0.17	2.83	0.39
ΣBTEX	3.62	1.20	1.15	0.37	1.85	1.25	1.01	0.28	2.41	0.38

door air (Tables 1 and 2). In AM samples, the *I*/*O* ratios of individual BTEX compounds and ΣBTEX in pharmacy, supply and garbage rooms were >1.0, and the highest ones were observed in pharmacy room (ranging from 2.73 to 4.03) (Table 4), proving the existence of indoor sources for BTEXs in these rooms; whereas those in preparation room and laundry room were close to 1.0. This does not exclude the existence of indoor sources, but in any case these are less significant than in the pharmacy, the supply and the garbage rooms.

Concerning carbonyls, *I*/*O* ratios showed large variation (Table 5). For example, *I*/*O* ratios for the total concentrations of carbonyls were >1.0 in the pharmacy, supply and garbage rooms, but was <1.0 in the laundry room. As for the individual carbonyl compounds, *I*/*O* ratios in all rooms were >1.0 only for buturaldehyde, cyclohexanone, valeraldehyde and 2,5-dimethylbenzaldehyde, suggesting potential indoor sources. In contrast, the *I*/*O* ratios in all rooms were <1.0 for crotonaldehyde, *p*-tolualdehyde and nonanaldehyde. As for the rest of the rooms, the *I*/*O* ratios for 12 out of 18 carbonyls detected were >1.0 in pharmacy and garbage rooms, whereas *I*/*O* ratios >1.0 were found only for five carbonyls (i.e., acetaldehyde, butyraldehyde, cyclohexanone, valeraldehyde, and 2,5-dimethylbenzaldehyde) in the preparation room, eight carbonyls (i.e., formaldehyde, acetaldehyde, butyraldehyde, cyclohexanone, valeraldehyde, 2,5-dimethylbenzaldehyde, octylaldehyde, and decylaldehyde) in the supply room and six carbonyls (i.e., formaldehyde, butyraldehyde, cyclohexanone, valeraldehyde, 2,5-dimethylbenzaldehyde, and decylaldehyde) in the laundry room, suggesting the different fate and sources of the carbonyls in the various rooms.

Table 5
Indoor/outdoor (*I*/*O*) concentration ratios of carbonyls in hospitals (PM).

	Pharmacy	Preparation	Supply	Laundry	Garbage
Formaldehyde	4.75	0.94	2.12	1.68	6.02
Acetaldehyde	1.28	1.07	1.43	0.48	2.24
Acrolein	0.00	0.00	0.00	0.00	0.00
Propionaldehyde	1.67	0.59	0.68	0.49	1.40
Crotonaldehyde	0.47	0.23	0.33	0.00	0.70
2-Butanone	3.17	0.09	0.06	0.08	0.15
Butyraldehyde	1.29	3.15	2.87	2.92	4.25
Benzaldehyde	1.11	0.47	0.43	0.47	0.87
Isovaleraldehyde	0.00	0.00	0.00	0.00	0.00
Cyclohexanone	2.04	1.11	1.36	1.25	1.89
Valeraldehyde	3.97	2.28	6.55	2.22	4.60
p-Tolualdehyde	0.67	0.00	0.78	0.00	0.00
m/*o*-Tolualdehyde	0.24	0.18	0.36	0.67	0.20
Hexaldehyde	9.88	0.00	0.00	0.00	1.75
2,5-Dimethylbenzaldehyde	45.27	4.33	7.93	7.80	11.87
Heptaldehyde	1.48	0.64	0.88	0.84	1.20
Octylaldehyde	1.21	0.85	1.08	0.87	1.28
Nonanaldehyde	0.54	0.56	0.99	0.83	1.01
Decylaldehyde	0.34	0.53	1.50	1.47	1.06
Total	1.88	1.01	1.43	0.83	2.15

The higher concentrations of VOCs during the morning were expected because the hospital is thoroughly cleaned early in the morning, and the use of cleaning products that contain also compounds like terpenes, can lead to direct or secondary emissions of VOCs. The use of electronical products (like printers or copiers) could also contribute to secondary emissions of VOCs. Unexpectedly, the *I*/*O* ratios were >1 in many PM samples. This was not expected because for many of them the outdoor air is the main source. The case of human activities during PM, the individual ventilation of each room (the use of air-conditioning systems that filter air, or of simple fans) together with the different fate of individual VOCs could be the explanation for this.

3.3. The concentration ratios of carbonyl compounds

The ratios of formaldehyde/acetaldehyde concentrations (C_1/C_2) could be used as an indicator of the source of carbonyls. C_1/C_2 normally vary from 1 to 2 in a polluted environment (e.g.,

Table 6
Concentration ratios of formaldehyde/acetaldehyde (C_1/C_2) and acetaldehyde/propionaldehyde (C_2/C_3).

	Pharmacy		Preparation		Supply		Laundry		Garbage		Outdoor	Min	Max	Mean
	AM	PM	AM	PM	AM	PM	AM	PM	AM	PM	PM			
C_1/C_2	0.20	0.43	0.07	0.10	0.06	0.17	0.12	0.41	0.11	0.31	0.12	0.06	0.43	0.19
C_2/C_3	4.34	3.99	5.69	9.47	7.46	11.0	4.94	5.12	12.5	8.34	5.22	3.99	12.5	7.10

urban areas) to about 10 in a less polluted background environment (e.g., rural areas) [18]. In this study, C_1/C_2 was calculated and listed in Table 6. C_1/C_2 ratios ranged from 0.06 to 0.43 (with a mean of 0.19). These ratios were lower than those reported in the hospitals (including ward, emergency room and injection room) of Guangzhou (mean ratio = 0.82) [2] and in the hotel ballrooms of Guangzhou, China (ranging from 1.3 to 2.42 with a mean of 1.81) [9]. The results of this study were in accordance with the finding in Rio de Janeiro (Brazil) that C_1/C_2 ratios were <1 with average values of 0.33 [19].

The concentration ratio of acetaldehyde/propionaldehyde (C_2/C_3) has been widely used as an effective indicator of anthropogenic source for carbonyls in the ambient air [9,18,20–21], because propionaldehyde was believed to be associated mainly with anthropogenic emissions, while other carbonyls had both natural and anthropogenic sources [20]. Thus, C_2/C_3 ratio would be high in rural atmospheres and low in contaminated urban air. In the present study, C_2/C_3 ratios were between 3.99 and 12.51 (with a mean of 7.10) (Table 6), being similar to the value recorded in the hotel ballrooms (average ratio = 6.2) [9] and hospitals of Guangzhou, China (average ratio = 5.0) [2], Hong Kong (average ratio = 8.4) [20] and Rome (average ratio = 5.2) [18]. The results suggested that there are anthropogenic sources in various rooms of hospital. Nevertheless, the use of the C_2/C_3 ratios should be made cautiously in diagnosing the effects of anthropogenic source processes for the carbonyl pollution in air, since some studies reported that the ratios often show large variations [20].

3.4. Potential sources of BTEXs and carbonyls

To elucidate the distribution pattern and possible emission sources, correlation analysis of the concentrations of carbonyls and BTEXs were performed. Their Pearson's correlation matrix is shown in Table 7. Significant correlations were found between all BTEXs ($R^2 > 0.92$), suggesting their common sources. Lü et al. [2] reported that there were good correlations between the concentrations of BTEXs (except benzene) in the hospitals (including ward, emergency room and injection room) and suggested that vehicle emission could be an important source. Ward, emergency room and injection room were kept under good ventilation, so the outdoor air could penetrate to the indoor environment. Nevertheless, in the present study, vehicle emission might not be the most important source for BTEXs in the garbage, pharmacy and supply room, because these rooms are poorly ventilated which might lead to the accumulation of BTEXs. The high concentrations of BTEXs in these rooms are due to indoor sources. A previous study by Statheropoulos et al. [22] indicated that benzene, toluene and *m*-xylene were among the most prominent volatile organic compounds (VOCs) in urban waste disposal bins. These results could explain the higher concentrations of BTEXs in the garbage rooms of the present study (Table 1).

As for carbonyls, significant correlations were found between some of the carbonyls (e.g., between formaldehyde and acetaldehyde, between acetaldehyde and propionaldehyde), while low correlations were observed among many carbonyls (Table 7), possibly implying the complex sources of carbonyls.

Principal component analysis (PCA) is a multivariate statistical method that is applied widely in atmospheric characterization studies to aid in the analysis and subsequent interpretation of a set of correlated variables. For example, Santarsiero and Fuselli [23] employed PCA to elucidate the indoor and outdoor air carbonyl compound correlations. In this study, PCA was used to investigate the distribution of different carbonyls and possible potential sources. The majority of the variance (83.9%) was explained by five eigenvectors principal components (PCs), i.e., 42.89% for PC1, 21.47% for PC2, 13.22% for PC3, 8.27% for PC4 and 6.22% for PC5, respectively. Fig. 1 is showing the loading plot for carbonyls and scores plot for locations of PC1 *vs.* PC2. As seen in the loading plot for carbonyls (Fig. 1a), the 17 carbonyl compounds could be clustered in three groups. Group A clustered eight compounds, that is formaldehyde, acetaldehyde, propionaldehyde, crotonaldehyde, benzaldehyde, cyclohexanone, heptaldehyde and octylaldehyde. Among Group A, positive significant correlations ($P < 0.05$) were found between formaldehyde and the other compounds, between acetaldehyde and the other com-

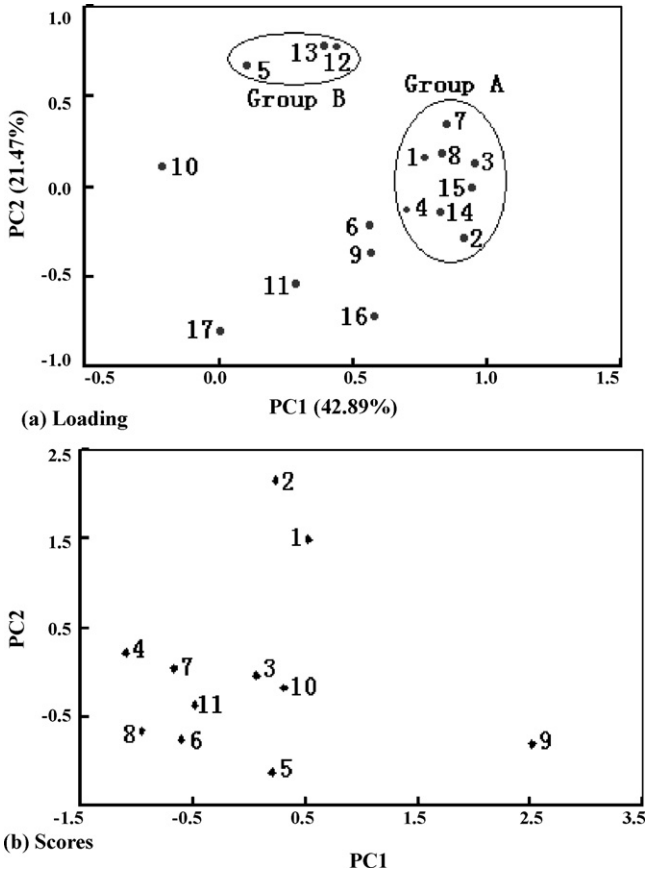

Fig. 1. Loading plot for carbonyls (a) and scores plot for locations (b) of PC1 *vs.* PC2. The corresponding compounds in (a): 1, formaldehyde; 2, acetaldehyde; 3, propionaldehyde; 4, crotonaldehyde; 5, 2-butanone; 6, butyraldehyde; 7, benzaldehyde; 8, cyclohexanone; 9, valeraldehyde; 10, *p*-tolualdehyde; 11, *m*/*o*-tolualdehyde; 12, hexaldehyde; 13, 2,5-dimethylbenzaldehyde; 14, heptaldehyde; 15, octylaldehyde; 16, nonanaldehyde; 17, decylaldehyde. The corresponding sites in (b): 1, pharmacy room (AM); 2, pharmacy room (PM); 3, preparation room (AM); 4, preparation room (PM); 5, supply room (AM); 6, supply room (PM); 7, laundry room (AM); 8, laundry room (PM); 9, garbage room (AM); 10, garbage room (PM); 11, outdoor (PM).

Table 7
Pearson correlation coefficients for carbonyls in indoor and outdoor air of hospitals.

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
Formaldehyde 1	1																					
Acetaldehyde 2	**0.70**	1																				
Propionaldehyde 3	**0.65**	**0.84**	1																			
Crotonaldehyde 4	**0.60**	**0.71**	**0.64**	1																		
2-Butanone 5		*0.58*	**0.62**		1																	
Butyraldehyde 6			**0.68**	**0.84**	**0.74**	1																
Benzaldehyde 7	**0.62**	**0.75**	**0.89**	*0.46*			**0.73**	1														
Valeraldehyde 9		*0.57*	*0.50*			*0.46*			1													
p-Tolualdehyde 10										1												
m/*o*-Tolualdehyde 11				*0.47*							1											
Hexaldehyde 12			*0.48*		*0.51*		**0.66**					1										
2,5-Dimethyl-benzaldehyde 13			*0.45*		*0.45*		*0.58*	*0.41*				**0.98**	1									
Heptaldehyde 14	**0.73**	**0.80**	**0.72**	**0.66**			**0.61**	*0.49*	*0.58*					1								
Octylaldehyde 15	**0.60**	**0.82**	**0.92**	*0.53*		**0.67**	**0.76**	**0.78**	**0.67**				*0.48*	**0.71**	1							
Nonanaldehyde 16		**0.69**	*0.42*	*0.56*					**0.70**		**0.62**			**0.66**	*0.56*	1						
Decylaldehyde 17									*0.41*		*0.43*					**0.73**	1					
Benzene 18			**0.66**			*0.48*		**0.62**				**0.71**	**0.70**		**0.71**			1				
Toluene 19			**0.60**			*0.40*	**0.64**	*0.51*				**0.81**	**0.81**		**0.70**			**0.96**	1			
Ethylbenzene 20		*0.45*	**0.68**			*0.49*	**0.69**	*0.58*	*0.44*			**0.73**	**0.72**		**0.79**			**0.96**	**0.98**	1		
m,p-Xylenes 21		*0.45*	**0.65**			*0.48*	**0.60**	*0.52*	*0.56*			**0.66**	**0.66**		**0.79**			**0.92**	**0.96**	**0.98**	1	
o-Xylenes 22		*0.50*	**0.69**			*0.51*	**0.64**	*0.56*	*0.56*			**0.66**	**0.65**		**0.82**			**0.93**	**0.96**	**0.99**	**1.0**	1

Correlation coefficients significant at the 99% level are boldfaced; Correlation coefficients significant at the 95% level are italicized.

pounds (Table 7), implying that these compounds of Group A might have similar sources to those of formaldehyde and acetaldehyde. Group B contained three compounds (e.g., 2-butanone, hexaldehyde and 2,5-dimethylbenzaldehyde) which showed significant correlations ($P < 0.05$) (Table 7). The remaining six compounds (i.e., butyraldehyde, valeraldehyde, *p*-tolualdehyde, *m*/*o*-tolualdehyde, nonanaldehyde and decylaldehyde) displayed quite scatter and generally showed low correlations among them.

The score plot of PC1 *vs.* PC2 (Fig. 1b) showed the distribution of the various sampling locations. Obviously, the distribution was different in the six sampling sites as well as in the different time. For example, the samples of the pharmacy room both in AM and PM, and that of the garbage room in AM were considerably different from the remaining sampling sites. The former two samples were associated with PC2 axis, while the latter one was closely associated with PC1 axis. Combining the information obtained from the score and loading plots, might reveal that the different distribution of carbonyl compounds at the various rooms and sampling times was attributed to their different sources.

Generally, the outdoor emission sources of carbonyls include motor vehicles, gasoline evaporation, various industrial emission, photochemical processes and biogenic emission; indoor emission sources included the use of solvents, tobacco smoke, varnishes, decoration materials, etc. According to the results of the *I*/*O* ratios in this study, the primary emission sources of certain carbonyls could be indoor primary or secondary emissions. Formaldehyde and acetaldehyde are often emitted form building materials, like from the adhesive used in the manufacture of resins, plastics, coatings, and fabrics [24–25]. The furniture can emit significant formaldehyde levels as well. The room age, the presence of carpets and numerous consumer products have been shown to be the factors affecting the indoor carbonyl concentrations, especially that of formaldehyde and acetaldehyde [26]. These factors might have impact on carbonyl concentrations of indoor air in different rooms of hospital.

As exhibited in Table 2, acetaldehyde was the most abundant compound in indoor and outdoor air, which might be attributed to the emission of acetaldehyde from hospital waste or the wide use of ethanol (alcohol) as disinfectant in hospital [27]. High concentrations of acetaldehyde were recorded in urban waste disposal bins [22]. Moreover, ethanol was one of the most prominent VOCs in urban waste disposal bins [22]. Ethanol could evaporate into air and be transformed into acetaldehyde, something that obviously increases the concentration of acetaldehyde in the hospital.

On the other hand, ozone is often used for sterilization in hospitals, especially in the supply room. Indoor chemistry of ozone plays an important role in the secondary formation of carbonyls, especially in the presence of VOCs emitted by cleaning products, like terpenes [28]. The formation of carbonyls through these reactions has been very well documented in the paper of Morisson and Nazaroff [7].

Furthermore, direct emissions from motor vehicles and other combustion sources are an important source of carbonyls in the ambient air. In the city of Guangzhou as well as in the overall Guangdong Province, the number of registered motor vehicles has increased by about 125% between 1998 and 2004 [29]. The hospital of the present study is adjacent to a road with heavy traffic, which can be the reason for increased background levels of carbonyls.

4. Conclusion

This study investigated 21 carbonyls and five BTEXs in the pharmacy, preparation, supply, laundry and garbage rooms of a hospital in Guangzhou, China. During AM, the carbonyls and BTEXs

concentrations in all rooms were higher than those counterparts determined during PM. In AM, the I/O ratios of individual BTEXs and ΣBTEXs for all rooms were >1.0, implying the indoor sources for BTEXs in these rooms, whereas those in PM were close or lower than 1.0. The I/O ratios of carbonyls in the afternoon were varied. C_1/C_2 and C_2/C_3 ratios showed large variation and suggested the anthropogenic sources of carbonyls. The results of the correlation and the principal component analysis implied indoor sources for formaldehyde, acetaldehyde, etc. The usage of special reagents or the accumulation might contribute partly to the carbonyls and BTEX concentrations, especially in AM samples. A further study employing compound-specific carbon isotopic compositions is considered necessary to identify the sources of carbonyls in the hospital.

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