



Levels and Health Risk of Carbonyl Compounds in Air of the Library in Guangzhou, South China

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ABSTRACT

The concentrations of 18 carbonyl compounds and their health risks for people were investigated in indoor and outdoor air of three different rooms of a library in Guangzhou, South China. Indoor air samples were collected during normal activities of staff and students, and carbonyl compounds were analyzed by high-performance liquid chromatography. The results show that the total concentrations of 18 carbonyls ranged from 8.8 to 73.9 $\mu\text{g m}^{-3}$ with a mean value of 24.7 $\mu\text{g m}^{-3}$. Acetone was the most abundant species with a mean value of 10.8 $\mu\text{g m}^{-3}$ in indoor air, followed by formaldehyde and acetaldehyde. The levels of carbonyls in this study were relatively lower than most of other microenvironments in the literature. The average total concentration of carbonyls in the electronic reading room was higher than those in stack room and compact stack room. Diurnal variation in carbonyl concentrations was not distinct in the three rooms. The indoor/outdoor ratios and Spearman correlation coefficients demonstrated that endogenous emission sources contributed significantly to most of indoor carbonyls, while outdoor infiltration might be one of the main sources for acetaldehyde. The personal exposure, lifetime cancer risk, and non-cancer chronic health impacts through inhalation associated with formaldehyde and acetaldehyde were estimated and discussed.

Keywords: Carbonyl compounds; Library; Indoor air; Health risk.

INTRODUCTION

Indoor air quality is of great concern because a lot of hazardous chemicals have been frequently detected in the indoor air. Carbonyl compounds are among the most important categories of chemicals, and are of an increasing concern due to their adverse effects on public health. Some carbonyls, such as formaldehyde, acetaldehyde, and propionaldehyde, are listed as air toxics in the Clean Air Act Amendments of 1990 (US EPA, 1991; Yao *et al.*, 2015). Formaldehyde, one of the most abundant carbonyls in air, has been classified as a human carcinogen by the International Agency for Research on Cancer (IARC) due to its carcinogenicity (IARC, 2006). Formaldehyde is used extensively in construction, wood processing, furniture, textiles, carpeting, and in the chemical industry (Bernabe *et al.*, 2015; Leuchner *et al.*, 2016). By 2007, the production

of formaldehyde in China had reached a staggering 12,000 kt, accounting for about 34% of the total global output (Tang *et al.*, 2009). Acetaldehyde, another abundant airborne carbonyl, has been classified as a suspected human carcinogen by United States Environmental Protection Agency (US EPA, 2003).

Indoor air quality is very vital because people spend about 80%–90% of their time in indoor (Cavalcante *et al.*, 2006). Recently, the levels, distribution, and risks of carbonyl compounds in indoor air of various microenvironments have been widely investigated, including dwellings, shopping centers, furniture stores, office rooms, schools, hospitals, stations, cinemas, restaurants, etc. (Cavalcante *et al.*, 2006; Lü *et al.*, 2006; Lovreglio *et al.*, 2009; Feng *et al.*, 2010; Sarigiannis *et al.*, 2011; Jiang *et al.*, 2013; Cheng *et al.*, 2014; Du *et al.*, 2014; Villanueva *et al.*, 2015; Huang *et al.*, 2016). Many studies have reported that the carbonyl levels of indoor air were higher than those of outdoor along with seasonal and diurnal variation (Weng *et al.*, 2009; Lü *et al.*, 2010; Feng *et al.*, 2010; Zhang *et al.*, 2012; Jiang *et al.*, 2013; Du *et al.*, 2014; Villanueva *et al.*, 2015). The indoor sources significantly contributed to carbonyls, such as indoor emission from materials (e.g., wood products, furniture),

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indoor chemical formation, and anthropogenic activities (Weng *et al.*, 2009; Feng *et al.*, 2010).

Due to the relatively low boiling points and high vapor pressures of carbonyls, human can be exposed to carbonyls through inhalation and dermal contact (Sarigiannis *et al.*, 2011; Chen *et al.*, 2012; Ramirez *et al.*, 2012; Du *et al.*, 2014; Villanueva *et al.*, 2015). People exposed to chemicals in indoor environment have an increasing probability of getting degenerative diseases (Cavalcante *et al.*, 2006). Importantly, the modeled personal inhalation exposure to some carbonyls (e.g., formaldehyde and acetaldehyde) of indoor air in urban of China is higher than the cancer benchmark concentrations by one or two orders of magnitude (Du *et al.*, 2014). However, most of the previous studies have only focused on the carbonyl levels and their risk in residences, offices and the public places for entertainment and leisure (e.g., ballrooms, shopping centers, stations) (Weng *et al.*, 2009; Sarigiannis *et al.*, 2011; Du *et al.*, 2014). Thus, it is very important to investigate the levels of carbonyls in more microenvironments and to assess their risk of human exposure.

The library is a favorable place for study. Many students and readers would like to stay here for reading, reviewing and writing. Cavalcante *et al.* (2006) reported that staff or employees spend 40 h per week, and students spend 10–12 h per week in the library. The indoor air quality of library may be easily overlooked in such a peaceful and silent environment (Ho *et al.*, 2014). Nevertheless, a previous study showed that formaldehyde was highly concentrated in library, and the air quality with respect to carbonyl levels in the library was similar to or worse than that in some laboratories where chemistry reagents were daily handled (Cavalcante *et al.*, 2006). The cancer risk for professionals was 5–30-fold higher than that for students, and that in some libraries was even higher when compared to particular laboratories (Cavalcante *et al.*, 2006). Therefore, more attention should be paid to the air quality in the library and health risk of human exposure to contaminants.

Guangzhou, the capital of Guangdong Province located in subtropics, is one of the largest industrial center and the fastest expanding cities in China. The rapid industrialization and urbanization has severely deteriorated the environmental quality during the past decades. Our previous studies indicated that carbonyls were frequently detected in ambient and indoor air (i.e., hospital) in Guangzhou, in some cases the levels of carbonyls being higher than those in other regions or countries (Lü *et al.*, 2006, 2010a, b). However, the levels and health risk of carbonyls of the library in China even in southeast Asia were scarcely reported. Thus, the aims of the present work are to investigate the levels and distribution of 18 carbonyls in indoor and outdoor air of a library in Guangzhou, South China, and to analyze the main sources of carbonyls. Based on these results, the health risks of occupational exposure to carbonyls for staff were discussed.

MATERIALS AND METHODS

Chemicals and Materials

All organic solvents used were in high-performance liquid

chromatography (HPLC) grade. Water was re-distilled and filtered by Milli-Q prior to use. A composite stock standard solution was purchased from Chem Service (USA), containing 18 carbonyl–DNPH (2,4-dinitrophenylhydrazine) derivatives, including formaldehyde, acetaldehyde, acetone, propionaldehyde, crotonaldehyde, 2-butanone, butyraldehyde, benzaldehyde, isovaleraldehyde, cyclohexanone, valeraldehyde, *m*-toulaldehyde, *o*-toulaldehyde, hexaldehyde, heptaldehyde, octylaldehyde, nonanaldehyde, decylaldehyde. The sampling cartridges were Sep-Pak DNPH–Silica cartridges manufactured by Waters Corporation (Milford, MA).

Air Sampling

Air samples were collected in a library of a university located in Guangzhou from 11th to 13th January, 2011. This library covers three buildings with an area of 32,000 m² and a collection of 7.86 million volumes, and provides more than 2,200 seats in general reading room and 260 seats in the electronic reading room. The stack room (L1), electronic reading room (L2), and compact stack room (L3) located in first floor of each building were selected for sampling (Fig. 1). The details of sampling sites and meteorological conditions are listed in Table 1. Smoking is strictly forbidden in the library. In each site, air samples were collected during three periods: morning (8:00–11:00; local time), afternoon (15:00–18:00), night (20:00–8:00) for three consecutive days.

The sampling procedure followed the method described in our previous study (Lü *et al.*, 2006). Briefly, samples were collected by drawing the air with a sampling pump (Thomas, USA) through the DNPH-coated cartridge. A potassium iodide (KI) denuder was connected to the upstream of the cartridge to prevent the interference of ozone. The carbonyl sampler was installed at about 1.2 m above the ground. The sampling site of outdoor air was set-up in the point which has almost the same distance (about 8 m) to the three building. The sampling duration time during daytime (morning and afternoon) was about 3 h at a flow rate of 0.95–1.1 L min⁻¹, while that at night was about 12 h at a flow rate of 0.2 L min⁻¹ to prevent a breakthrough. After sampling, each cartridge was wrapped immediately in aluminum foil and then stored in the refrigerator in a Teflon bag for analysis. During sampling, one laboratory blank and one field blank were conducted each day, respectively. At each sampling site, two field samples were taken with a back-up cartridge to evaluate breakthrough.

Analytical Procedure, Quality Assurance and Quality Control

The cartridges above were eluted slowly with acetonitrile (ACN) into a 2-mL rinsed volumetric flask (Lü *et al.*, 2006). The extract was stored in a refrigerator for subsequent analysis. The reverse phase HPLC system (HP1100, Agilent, USA) coupled with a ultra-violet (UV) detector was used for chemical analysis. A SB-C18 reverse column (250 mm × 4.6 mm × 5 μm, Agilent, USA) was used for separation. A 10-μL aliquot was injected into the HPLC system through an auto-sampler. The gradient program of mobile phase was 60%–70% ACN of water solution for 20 min, 70%–100%



Fig. 1. Sampling sites in a library of Guangzhou. L1, stack room (with an area of 520 m²); L2, electronic reading room (with an area of 320 m²); L3, compact stack room (with an area of 476 m²).

Table 1. Description of locations sampled in the library.

Sampling sites	Usage (ventilation system)	Decoration conditions	Temp (°C)	Relative humidity (%)	Numbers of readers
L1	Stack room (natural ventilation)	Rubber flooring, 50 new wood shelves, wood tables and chairs, new books	11–14	48–68	13–40
L2	Electronic reading room (natural ventilation)	Tile floor, fabric curtains, plastic tables and chairs, computes, photocopier	8–14.5	52–65	4–83
L3	Compact stack room (exhaust fan)	Tile floor, 30 old wood shelves, old books	8.5–15	47–60	2–3
Outdoor			4–13	48–68	

ACN for 3 min, 100% ACN for 4 min, 100%–60% ACN for 1 min and then 60% ACN for 5 min. The flow rate of mobile-phase was 1 mL min⁻¹. The analytes were detected by UV at 360 nm.

Carbonyl compounds were identified and quantified by their retention time and peak areas of the corresponding calibration standards (Lü *et al.*, 2006, 2010a). The calibration curve was established by five standard concentrations (from 0.5 to 10 µg mL⁻¹) covering the levels of interest. Strong linear relationships ($R^2 > 0.99$) were found between the concentrations and responses for all carbonyls identified. A calibration standard was run each day to ensure the

stabilization of the instruments. The collection efficiency of cartridge was determined with two cartridges in series. The results show that over 99% of carbonyl compounds were recovered from the first cartridge, demonstrating the complete recovery of all carbonyls. The relative standard deviation of duplicate samples was < 5%. The detection limits for target carbonyls were determined using seven replicate analyses of the working standards at the lowest concentration. The detection limits of this study ranged from 0.05 to 0.15 µg m⁻³ for various carbonyls (in 120-L sampling volume). The analytical procedures met the requirements of US EPA Method TO-11a (US EPA, 1999).

The sum concentrations of isovaleraldehyde and cyclohexanone, *m*-tolualdehyde and *o*-tolualdehyde are presented simultaneously because they were not separated by the analytical method.

RESULTS AND DISCUSSION

Concentrations of Carbonyls in the Library

In this study, eighteen carbonyl compounds were detected in the air of a library in a university, Guangzhou. The average concentrations of carbonyl compounds measured at each site of the library are presented in Table 2. The total concentrations of 18 carbonyls ($\sum_{18\text{carb}}$) ranged from 8.76 to 73.9 $\mu\text{g m}^{-3}$ with a mean value of 24.7 $\mu\text{g m}^{-3}$. Some studies have reported the carbonyl levels of indoor air. For example, the total concentrations of 19 carbonyls in hospitals of Guangzhou, China, were between 18 and 106 $\mu\text{g m}^{-3}$ (mean: 40.6 $\mu\text{g m}^{-3}$) (Lü *et al.*, 2010b), and those in subway stations of Shanghai varied from 28.8 to 124 $\mu\text{g m}^{-3}$ (Feng *et al.*, 2010). The total concentrations of 12 carbonyls in an academic building of Beijing ranged from 20.7 to 166 $\mu\text{g m}^{-3}$ (Jiang and Zhang, 2012). Compared with the levels above, the $\sum_{18\text{carb}}$ in this study were considerably lower.

Among 18 carbonyls measured, acetone was the most abundant species, followed by formaldehyde and acetaldehyde. The concentrations of acetone varied from 1.36 to 25.4 $\mu\text{g m}^{-3}$ with an average value of 10.8 $\mu\text{g m}^{-3}$ (Table 2), accounting for ~49.3%, 53.8%, 54.4%, and 53.6% of the $\sum_{18\text{carb}}$ in the L1, L2, L3 (Fig. 2), and outdoor air, respectively. Formaldehyde and acetaldehyde showed mean values of 4.66 $\mu\text{g m}^{-3}$ and 4.15 $\mu\text{g m}^{-3}$, and contributed ~39.7% and ~33.5% to the $\sum_{18\text{carb}}$, respectively. In the high molecular weight carbonyls ($\text{C}_6\text{--}\text{C}_{12}$), nonanaldehyde was the most abundant species with a mean value of 0.95 $\mu\text{g m}^{-3}$. Similarly, Dai *et al.* (2012) reported that nonanaldehyde was the most abundant compound among high molecular weight mono-carbonyls ($\text{C}\# > 5$) and di-carbonyls in urban atmosphere of Xi'an, China. The average concentrations of the other compounds were less than 1.0 $\mu\text{g m}^{-3}$ (Table 2), accounting for < 10% of the $\sum_{18\text{carb}}$. This distribution trend is similar to the previous studies in Hong Kong (Cheng *et al.*, 2014) and Xiamen, China (Ho *et al.*, 2014). In the literature, some scientists have reported the levels of carbonyls in the library (Cavalcante *et al.*, 2006; Ho *et al.*, 2014). For example, Ho *et al.* (2014) and Cavalcante *et al.* (2006) reported that formaldehyde, acetaldehyde, and acetone of indoor air in the library were the most abundant compounds with average concentrations < 10.0 $\mu\text{g m}^{-3}$ (except with formaldehyde in a library of Xiamen), in accord with our results. Nevertheless, the average levels of formaldehyde, acetaldehyde and acetone in this study were considerably lower than those in subway stations and subway trains of Shanghai, China (Feng *et al.*, 2010), at home of Beijing, China (Tang *et al.*, 2009; Du *et al.*, 2014) and Puertollano (Iberian Peninsula; Villanueva *et al.*, 2015), residential indoor of United states (Liu *et al.*, 2006) and South Italy (Lovreglio *et al.*, 2009), and in office of China (Du *et al.*, 2014). In all rooms tested in this study, formaldehyde concentrations were below the air quality guideline value (100 $\mu\text{g m}^{-3}$) set by the World

Health Organization. On the whole, the concentrations of most carbonyls in this study did not show distinct differences between indoor and outdoor air.

Regarding the different rooms in the library, the average $\sum_{18\text{carb}}$ in L2 (28.2 $\mu\text{g m}^{-3}$) was higher than in L1 (25.9 $\mu\text{g m}^{-3}$) and L3 (20.0 $\mu\text{g m}^{-3}$), and the average concentrations of formaldehyde and acetone in L2 were also higher than those in the other rooms, while the concentrations of acetaldehyde and acetone in L2 were comparable to those in L1 and L3 (Table 2). This might be related to the emission sources of indoor materials and human activities. In the indoor air of workplace, some carbonyl compounds may be released from indoor sources, including building materials, furniture, decorating materials, consumer products, electronic equipments (such as photocopiers), and through reactions between indoor ozone and alkenes (Santarsiero and Fuselli, 2008). In L2, there were many electronic equipments including 96 computers and 2 photocopiers, which could generate indoor ozone from oxygen gas by electrical discharge and by the action of high-energy electromagnetic radiation, in addition to the formation of various volatile organic compounds (VOCs) (Lee *et al.*, 2006; Ho *et al.*, 2014). A previous study reported that the concentrations of carbonyls (13 compounds, including formaldehyde, acetaldehyde, and acetone) and total VOCs in the copy rooms were considerably higher than those in office rooms and meeting rooms of the same academic building (Jiang and Zhang, 2012). In L1 (stack room), there are wood shelves, wood tables and chairs, books, etc. Generally, formaldehyde is used as an adhesive for producing wood-based materials. Formaldehyde emission from solid wood, plywood, blockboard used for furnishing materials was high in the first week after being manufactured, while then decreased noticeably (Bohm *et al.*, 2012). In this study, the wood shelves, wood tables and chairs of L1 were installed one year ago, and this room was always ventilated during work time. Therefore, formaldehyde emission of L1 was relatively low. In L3 (compact stack room), there were old wood shelves, old books and tile floor, and thus the average total concentrations and individual carbonyl levels were the lowest among the three rooms. Similarly, Molloy *et al.* (2012) reported that elevated concentrations of formaldehyde were observed in dwellings constructed in recent decades compared to older buildings.

Diurnal Variations of Carbonyl Concentrations

Diurnal variation in carbonyl concentrations was not distinct in the three rooms of the library, though the average concentrations of individual carbonyls and the average $\sum_{18\text{carb}}$ decreased in the order of night > afternoon > morning (Table 2). Similarly, in one office room of an academic building, carbonyl compounds and total VOC concentrations tended to be higher at night than in the daytime (Jiang and Zhang, 2012). Change in ventilation is one of the most important factors affecting the diurnal variations of carbonyl compounds (Jiang and Zhang, 2012). Carbonyls in the indoor air are thought to be derived mainly from the indoor emissions, indoor chemical formation and outdoor infiltration (Zhang *et al.*, 1994).

Table 2. Concentrations of carbonyl compounds in the sampling sites ($\mu\text{g m}^{-3}$) (Mean \pm S.D.).

Compounds	L1			L2			L3			Outdoor		
	Morning	Afternoon	Night	Morning	Afternoon	Night	Morning	Afternoon	Night	Morning	Afternoon	Night
Formaldehyde	3.12 \pm 1.00	3.84 \pm 1.11	4.36 \pm 1.37	6.97 \pm 6.12	5.82 \pm 3.78	7.59 \pm 3.08	2.61 \pm 0.58	2.73 \pm 0.46	3.83 \pm 0.95	2.25 \pm 1.93	3.64 \pm 1.56	8.56 \pm 7.98
Acetaldehyde	2.62 \pm 0.56	3.31 \pm 0.98	3.59 \pm 1.50	4.56 \pm 2.98	2.76 \pm 0.30	3.25 \pm 0.66	2.42 \pm 0.57	3.54 \pm 0.96	4.20 \pm 0.04	5.52 \pm 5.54	5.64 \pm 5.24	5.99 \pm 4.59
Acetone	9.12 \pm 4.14	12.6 \pm 5.04	12.8 \pm 5.22	9.80 \pm 4.32	9.88 \pm 1.82	15.5 \pm 3.45	8.25 \pm 5.11	7.20 \pm 1.36	9.73 \pm 2.74	6.98 \pm 4.43	6.15 \pm 3.90	11.2 \pm 12.4
Propionaldehyde	0.40 \pm 0.01	0.61 \pm 0.18	0.59 \pm 0.21	0.44 \pm 0.11	0.39 \pm 0.03	0.47 \pm 0.17	0.38 \pm 0.09	0.40 \pm 0.07	0.58 \pm 0.14	0.36 \pm 0.06	0.44 \pm 0.18	0.44 \pm 0.24
Crotonaldehyde	Nd	Nd	0.14a	Nd	Nd	0.10 \pm 0.04b	Nd	Nd	0.1a	Nd	0.16a	0.11 a
2-Butanone	0.48 \pm 0.11	0.65 \pm 0.29	0.84 \pm 0.58	0.58 \pm 0.23	0.46 \pm 0.13	1.11 \pm 1.02	0.48 \pm 0.16	0.60 \pm 0.25	1.29 \pm 0.83	0.51 \pm 0.19	0.65 \pm 0.51	0.81 \pm 0.65
Butyraldehyde	0.28 \pm 0.06	0.30 \pm 0.01	0.31 \pm 0.13	0.44 \pm 0.38	0.17 \pm 0.03	0.26 \pm 0.06	0.20 \pm 0.05	0.19 \pm 0.04	0.31 \pm 0	0.58 \pm 0.62	0.44 \pm 0.38	0.40 \pm 0.27
Benzaldehyde	0.37 \pm 0.14	0.59 \pm 0.24	0.57 \pm 0.19	0.39 \pm 0.16	0.36 \pm 0.05	0.59 \pm 0.20	0.32 \pm 0.01	0.45 \pm 0.10	0.63 \pm 0.18	0.35 \pm 0.08	0.52 \pm 0.37	0.61 \pm 0.40
Iso + Cyclo	0.48 \pm 0.12	0.62 \pm 0.24	0.70 \pm 0.31	0.57 \pm 0.13	0.51 \pm 0.09	0.92 \pm 0.39	0.50 \pm 0.15	0.50 \pm 0.06	0.77 \pm 0.34	0.45 \pm 0.09	0.53 \pm 0.24	0.71 \pm 0.39
Valeraldehyde	0.20 \pm 0.02	0.34 \pm 0.21	0.26 \pm 0.12	0.14 \pm 0.03	0.11 \pm 0.01	0.15 \pm 0.09	0.14 \pm 0.04	0.13 \pm 0.02	0.2 \pm 0.04	0.07 \pm 0.03	0.10 \pm 0.02	0.16 \pm 0.14
m/o-Tolualdehyde	0.76 \pm 0.01	1.19 \pm 0.30	1.05 \pm 0.11	0.65 \pm 0.34	0.84 \pm 0.22	1.17 \pm 0.32	0.48 \pm 0.05	0.70 \pm 0.03	0.70 \pm 0.19	0.33 \pm 0.34	0.57 \pm 0.32	2.16a
Hexaldehyde	0.64 \pm 0.05	0.81 \pm 0.07	0.87 \pm 0.18	0.39 \pm 0.12	0.36 \pm 0.20	0.42 \pm 0.10	0.28 \pm 0.02	0.34 \pm 0.05	0.40 \pm 0.01	0.28 \pm 0.10	0.24 \pm 0.04	0.43 \pm 0.37
Heptaldehyde	0.20 \pm 0.01	0.27 \pm 0.02	0.26 \pm 0.02	0.21 \pm 0.02	0.20 \pm 0.05	0.23 \pm 0.04	0.17 \pm 0	0.21 \pm 0.06	0.22 \pm 0.05	0.16 \pm 0.03	0.15 \pm 0.03	0.20 \pm 0.09
Octylaldehyde	0.19 \pm 0.06	0.33 \pm 0.06	0.30 \pm 0.07	0.23 \pm 0.08	0.20 \pm 0.14	0.32 \pm 0.19	0.12 \pm 0.01	0.18 \pm 0.12	0.21 \pm 0.10	0.09 \pm 0.03	0.07 \pm 0.03	0.23 \pm 0.32
Nonanaldehyde	0.85 \pm 0.10	1.38 \pm 0.29	1.29 \pm 0.18	1.03 \pm 0.20	0.95 \pm 0.43	1.22 \pm 0.56	0.79 \pm 0.35	1.27 \pm 0.90	1.34 \pm 0.92	0.58 \pm 0.09	0.55 \pm 0.06	0.92 \pm 0.83
Decylaldehyde	0.28 \pm 0.06	0.54 \pm 0.19	0.47 \pm 0.16	0.46 \pm 0.23	0.45 \pm 0.43	0.88 \pm 0.69	0.18 \pm 0.01	0.23 \pm 0.08	0.22 \pm 0.02	0.13 \pm 0.10	0.14 \pm 0.03	0.67 \pm 0.9
Total	20.0 \pm 6.22	27.4 \pm 8.64	28.3 \pm 9.99	26.7 \pm 9.09	23.5 \pm 5.71	68.5 \pm 5.58	17.3 \pm 6.34	18.7 \pm 1.20	24.7 \pm 4.46	17.9 \pm 7.96	18.7 \pm 6.96	29.2 \pm 24.2

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

L1: stack room, L2: electronic reading room, L3: compact stack room.

^a Only one sample.^b Two samples average, iso+cyclo: isovaleraldehyde and cyclohexanone.

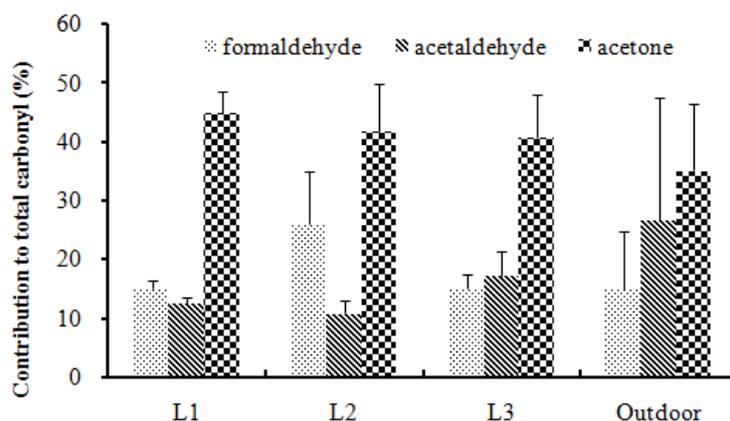


Fig. 2. Contribution of formaldehyde, acetaldehyde, and acetone to the total concentrations of 18 carbonyls.

In the present study, in winter there is only natural ventilation via windows and doors for L1 and L2, and mechanical ventilation via exhaust fans for L3 (Table 1). From midnight (around 22:00) till early morning (around 8:00), the doors and windows of library rooms were closed, and the air exchange rates were very low. Thus, carbonyl compounds released from furnishing materials remained in the rooms, thus resulting in the relatively higher average concentrations of carbonyls at night. Nevertheless, no significant difference was observed in carbonyl concentrations among night, afternoon, morning. This pattern is similar to the diurnal variations of carbonyls in urban ambient air of Guangzhou (Lü *et al.*, 2010), implying that carbonyl concentrations in the library might be also affected by outdoor air.

Indoor/Outdoor Ratios of Carbonyls and Sources

Outdoor infiltration is one of important sources of indoor carbonyls. The indoor/outdoor ratio is usually applied to infer penetration to indoor environments and indoor sources. In this study, the average ratios of indoor/outdoor were almost all > 1 (except acetaldehyde, butyraldehyde and benzaldehyde) (Table 3), indicating that endogenous emission sources were responsible for indoor carbonyls. Especially for some carbonyl compounds (including crotonaldehyde, valeraldehyde, *m/o*-tolualdehyde, hexaldehyde, heptaldehyde, octylaldehyde, nonanaldehyde, isovaleraldehyde and cyclohexanone), their indoor/outdoor ratios are close to or higher than 1.0, implying that they are resulted from indoor emission. In the rooms tested, there are many books, wood furniture, computers, photocopiers, curtain, decoration materials, etc, which are important emission sources of carbonyls in the library. Nevertheless, it should be noted that indoor/outdoor ratios of formaldehyde, acetone and decylaldehyde varied greatly from 0.33 to 3.88 (Table 3), suggesting that both indoor and outdoor sources influenced indoor levels of the three compounds to the some degree. Regarding acetaldehyde and butyraldehyde, their indoor/outdoor ratios were less than 1.0, indicating their outdoor sources (including outdoor infiltration). This might be related with ventilation of library rooms. During the sampling time, the windows and doors were open, and

Table 3. Indoor/outdoor (*I/O*) concentration ratios of carbonyls in the library.

	Mean	S.D.	Min	Max
Formaldehyde	1.21	0.80	0.45	3.09
Acetaldehyde	0.59	0.13	0.44	0.82
Acetone	1.35	0.34	0.87	2.05
Propionaldehyde	1.15	0.18	0.88	1.38
Crotonaldehyde	1.45	0.24	1.27	1.73
2-Butanone	1.07	0.26	0.71	1.59
Butyraldehyde	0.59	0.18	0.35	0.78
Benzaldehyde	0.97	0.14	0.69	1.13
Iso + Cyclo	1.10	0.12	0.95	1.29
Valeraldehyde	1.79	0.81	0.94	3.29
<i>m/o</i> -Tolualdehyde	1.63	0.44	0.96	2.34
Hexaldehyde	1.67	0.81	0.94	3.44
Heptaldehyde	1.30	0.22	1.06	1.82
Octylaldehyde	2.17	1.08	0.91	4.45
Nonanaldehyde	1.70	0.43	1.32	2.51
Decylaldehyde	2.03	1.27	0.33	3.88
Total	1.27	0.46	0.84	2.34

AM: the arithmetic mean of every pair *I/O* for all the sampling sites.

S.D.: standard deviation, N: number of samples.

Iso + cyclo: isovaleraldehyde and cyclohexanone.

outdoor infiltration might be one of the main sources of indoor acetaldehyde and butyraldehyde.

Pearson correlations based on concentrations of indoor carbonyls were used to investigate the inter-correlations among carbonyl species and to clarify the potential emission sources of carbonyls in the library. The Pearson correlation matrix of 15 carbonyls is demonstrated in Table 4. Significant correlations were found between acetaldehyde and propionaldehyde, between acetaldehyde and butyraldehyde as well as benzaldehyde, etc, indicating that these compounds came from almost the same sources. As mentioned above, these carbonyls mainly came from indoor sources. Some compounds, such as acetone, *m/o*-tolualdehyde, octylaldehyde, decylaldehyde were significantly correlated with each other, implying that they were from almost the same sources. As stated above, some carbonyls (*i.e.*,

Table 4. Pearson correlation coefficients for carbonyls in indoor air of the library.

Compounds	1	2	3	4	6	7	8	9	10	11	12	13	14	15	16
Formaldehyde 1	1														
Acetaldehyde 2	-.14	1													
Acetone 3	.499*	.235	1												
Propionaldehyde 4	.075	.537**	.558**	1											
2-butanone 5	-.039	.414*	.333	.709**	1										
Butyraldehyde 6	-.274	.848**	.188	.414*	.340	1									
Benzaldehyde 7	.172	.478*	.630**	.846**	.724**	.336	1								
Iso + cyclo 8	.196	.423*	.604**	.753**	.914**	.309	.815**	1							
Valeraldehyde 9	.048	.333	.579**	.821**	.332	.252	.746**	.488*	1						
m/o-tolualdehyde 10	.497*	-.144	.670**	.329	.028	-.198	.411*	.247	.458*	1					
Hexaldehyde 11	.080	.113	.439*	.510**	.039	.186	.447*	.130	.673**	.554**	1				
Heptaldehyde 12	.159	.317	.402*	.365	-.032	.237	.474*	.131	.573**	.561**	.688**	1			
Octylaldehyde 13	.525**	.173	.629**	.235	-.159	.086	.390	.099	.440*	.720**	.614**	.870**	1		
Nonanaldehyde 14	.077	.197	.056	-.024	-.226	.065	.180	-.151	.143	.258	.341	.822**	.709**	1	
Decylaldehyde 15	.748**	.006	.652**	.077	-.216	-.079	.222	.076	.219	.732**	.327	.526**	.844**	.383	1

*, **: statistically significant at the probability level 0.05 and 0.01, respectively.

Iso + cyclo: isovaleraldehyde and cyclohexanone.

formaldehyde, acetone, propionaldehyde) were derived from both indoor and outdoor sources. The major sources of indoor carbonyls are building materials and furniture. In the library, varnished wood furniture (shelves, tables and chairs), curtain, and decoration materials were potential sources of carbonyls. Computers were used in every library room, particularly in L2 (also including copying machines). Additionally, this university has many chemical-physical laboratories that employ various reagents and solvents (e.g., acetone). Some researchers also found high acetone concentrations of indoor and outdoor air in Guangzhou (Wang *et al.*, 2007; Lü *et al.*, 2006, 2010a). This might be attributed to the usage of acetone as industrial solvent in many paint manufacturing and other industries located around Guangzhou. The lifetime of acetone for removal by photolysis and OH reaction (20–40 day in the atmosphere) was longer than other carbonyl compounds (1.5 h to 1 day for formaldehyde and acetaldehyde) (Atkinson, 2000). However, low correlations were observed between nonanaldehyde and most of carbonyl species, possibly implying complex sources and effect factors.

In fact, carbonyl concentrations in indoor air are influenced by many factors such as direct sources of indoor, infiltration from outdoor, human activities, diffusion in building materials, meteorological conditions, their sinks and lifetime, etc. For example, acetaldehyde is a product of the human metabolism and present in human expired air (Zhang *et al.*, 1994). There were more students in L1 and L2 than in L3 when the samples were collected (Table 1), which might affected the distribution of carbonyls in the three rooms. Building materials and wood shelves could facilitate the diffusion of chemicals through their pores (Katsoyiannis *et al.*, 2012), and infiltration of carbonyls might occur between ambient air and indoor air. Additionally, this study was conducted in winter, the temperature was relatively low (4–15°C), and the relative humidity ranged from 47% to 68% (Table 1). Our previous studies demonstrated that carbonyl concentrations in summer were higher than those in winter in Guangzhou (Lü *et al.*, 2010a). Here, it is typically subtropical monsoon climate, cool and dry in winter (Table 1), hot and humid in summer. Higher ambient temperature and higher relative humidity in summer might enhance photochemical reactions and generate more carbonyls (Lü *et al.*, 2010a). The meteorological conditions in this study are not favorable for the photochemistry reaction and release of carbonyls from building materials, books and furniture (Missia *et al.*, 2010).

Exposure and Risk

The library is one of most important study places for students, and it is also the work place for staff. People in the library can be exposed to carbonyls mainly via inhalation.

In this study, the personal exposure (E_i) for individual compound (i) of carbonyls can be calculated from the equation supplied by the US EPA:

$$E_i = C_i \times IR \times t \quad (1)$$

where C_i is the concentration of carbonyl ($\mu\text{g m}^{-3}$), IR is the inhalation rate ($\text{m}^3 \text{h}^{-1}$), t is the exposure time (h d^{-1}).

Indoor inhalation rates were estimated for an average person ($IR = 0.63 \text{ m}^3 \text{ h}^{-1}$) according to US EPA exposure factors (US EPA, 1990). The exposure time for staff in the library was 8 h d^{-1} , based on the official working time. The students stay in the library generally for less than 8 h d^{-1} (Cavalcante *et al.*, 2006), thus the exposure for students to carbonyls was not estimated here.

Formaldehyde and acetaldehyde are carcinogenic compounds. The estimated lifetime cancer risk (R) posed by the carcinogenic compounds can be calculated by the methodology proposed by the US EPA (1986):

$$R = C \times IUR \quad (2)$$

where C is the exposure concentration ($\mu\text{g m}^{-3}$), IUR is the inhalation unit risk, which is the upper-bound excess lifetime cancer risk estimated to result from continuous exposure to a compound at a concentration of $1 \mu\text{g m}^{-3}$ in air. IUR is the cancer potency factor for inhaled compounds (OEHHA, 2009). Formaldehyde was listed as Group B1 by the US EPA, probable human carcinogen of medium carcinogenic hazard with an IUR value of $1.3 \times 10^{-5} (\mu\text{g m}^{-3})^{-1}$ (US EPA, 1991a), while acetaldehyde was classified as Group B2, probable human carcinogen of low carcinogenic hazard, with an IUR estimate of $2.2 \times 10^{-6} (\mu\text{g m}^{-3})^{-1}$ (US EPA, 1991b).

For non-cancer hazard assessment, inhalation chronic reference exposure limits ($RELs$) are applied as an indicator of potential non-cancer health impacts. REL is referred to the concentration at or below which no adverse non-cancer health effects are anticipated following long-term exposure (OEHHA, 2003; Du *et al.*, 2014). Non-cancer chronic inhalation health impacts were calculated through the personal exposure (E) divided by a specific REL for compound as following equation:

$$HQ = E/REL \quad (3)$$

where HQ is the hazard quotient for compound; E is the personal exposure to compound. Inhalation chronic $RELs$ for formaldehyde and acetaldehyde are 9 and $140 \mu\text{g m}^{-3}$, respectively (OEHHA, 2003; Du *et al.*, 2014). According to the ratio, an $HQ \leq 1$ suggests that adverse health effects are not expected to result from exposure to this chemical; while the $HQ > 1$ indicates that the probability of human health adverse effects increases with long term exposure (OEHHA, 2003; Du *et al.*, 2014).

The calculated data of personal exposure (E_i), lifetime cancer risk (R), and HQ are presented in Table 5. The personal exposure levels ranged from 2.17 to $55.8 \mu\text{g d}^{-1}$ for formaldehyde and from 10.2 to $39.7 \mu\text{g d}^{-1}$ for acetaldehyde, being comparable with or lower than those reported in

shopping centers, stations, ballrooms, and office of China and Mexico (Báez *et al.*, 2003; Weng *et al.*, 2009; Du *et al.*, 2014). The estimated lifetime cancer risk varied from 5.6×10^{-6} to 1.63×10^{-4} for formaldehyde and from 4.44×10^{-6} to 1.73×10^{-5} for acetaldehyde (Table 5), being comparable with those in Puertollano (Iberian Peninsula; Villanueva *et al.*, 2015) and European Union countries (Sarigiannis *et al.*, 2011), but lower than those reported in Hangzhou, China (Weng *et al.*, 2009) and Mexico (Báez *et al.*, 2003). The highest cancer risk for both formaldehyde and acetaldehyde was found in L2 (Table 5). Till now, no regulation for acceptable/tolerable lifetime cancer risks has been proposed by any country or international organization (Sarigiannis *et al.*, 2011). As an administrative practice, an acceptable/ tolerable lifetime cancer risk was set as 10^{-6} , i.e., at one additional cancer case per million exposed persons (Sarigiannis *et al.*, 2011). In this study, the estimated lifetime cancer risks of both formaldehyde and acetaldehyde were in the range of 10^{-6} or higher, suggesting that special attention should be paid to the human health exposure to formaldehyde and acetaldehyde in the library.

In terms of non-carcinogenic effects, the HQ of formaldehyde and acetaldehyde ranged from 0.05 to 1.40 and from 0.01 to 0.06, respectively, and the average HQs in each room for both compounds are less than 1.0 (Table 5), indicating that health adverse effects are not expected to derive from exposure to the two carbonyls. Nevertheless, in room L2, the HQ for formaldehyde was greater than 1.0 in three samples (HQ ranging from 1.13–1.40), implying that long-term exposure may result in health adverse effects.

However, it should be pointed out that although formaldehyde and acetaldehyde in this library did not pose high risks as other public places (e.g., shopping centers, furniture stores, stations) (Weng *et al.*, 2009; Du *et al.*, 2014), health impacts of carbonyls in the library are not negligible. The risk assessment in this study was based on the toxicity data of individual compounds. Potential interactions of multiple chemicals may occur in complex mixtures (Ragas *et al.*, 2011). Interactive effects of exposure to more than one carbonyl compounds should be considered, and thus the risk levels might not be over- or underestimated. Moreover, dermal exposure route is important for some compounds (Chen *et al.*, 2012), and its risk also needs considering. In addition, the risk of exposure to carbonyls was assessed only for staff in this study. In fact, there are a lot of students in every room of the library, especially in L2 where some students stayed more than 8 h d^{-1} (in some cases $> 12 \text{ h d}^{-1}$ or $> 50 \text{ h}$ per week) to gather information and write papers. Their risk of exposure to carbonyls might be higher than the staff.

Table 5. Estimated exposure values (E) and health risks of formaldehyde and acetaldehyde in the library.

Room	Formaldehyde			Acetaldehyde		
	$E (\mu\text{g d}^{-1})$	Risk ($\times 10^{-5}$)	HQ	$E (\mu\text{g d}^{-1})$	Risk ($\times 10^{-6}$)	HQ
L1	19.4 ± 5.7	4.62 ± 1.32	0.38 ± 0.19	16.4 ± 5.4	6.68 ± 1.85	0.02 ± 0.01
L2	34.3 ± 20.1	8.32 ± 5.98	0.76 ± 0.44	17.8 ± 8.7	8.05 ± 4.70	0.03 ± 0.01
L3	11.8 ± 7.5	3.49 ± 0.58	0.26 ± 0.17	13.4 ± 8.6	6.80 ± 2.10	0.02 ± 0.01

CONCLUSION

Eighteen carbonyls were detected in the indoor and outdoor air of a library in Guangzhou. The average concentrations of carbonyl in electronic reading room (L2) were relatively higher than the other rooms. The diurnal variations of carbonyl compounds were not clear-cut. Formaldehyde, acetaldehyde and acetone were the most abundant compounds that accounted for more than 65% to the $\sum_{18\text{carb}}$. The results of correlation analysis indicate that indoor emission and outdoor infiltration were the predominant source of indoor carbonyls in the library. The occupational exposure levels and risk of both formaldehyde and acetaldehyde in the library were comparable to or lower than other studies. The results of this study provide a new insight not only into the levels of carbonyls in the library of Guangzhou, China, but also into the health risks of human exposure to these compounds.

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