Evaluation of hazardous airborne carbonyls on a university campus in southern China

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TECHNICAL PAPER

Evaluation of hazardous airborne carbonyls on a university campus in southern China

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A comprehensive assessment of indoor carbonyl compounds for the academic staff, workers, and students was conducted on a university campus in Xiamen, China. A total of 15 representative environment categories, including 12 indoor workplaces and three residential units, were selected. The potential indoor pollution sources were identified based on the variability in the molar compositions and correlation analyses for the target carbonyls. Furnishing materials, cooking emissions, and electronic equipment, such as photocopiers, can generate various carbonyls in the workplace. Comparison studies were conducted in the clerical offices, demonstrating that off-gases from wooden furniture and lacquer coatings, environmental tobacco smoke (ETS), and the use of cleaning reagents elevated the indoor carbonyl levels. The measured concentrations of formaldehyde and acetaldehyde in most locations surpassed the exposure limit levels. The lifetime cancer hazard risk (R) associated with formaldehyde was above the concern risk level (1 × 10⁻⁶) in all of the workplaces. The results indicate that formaldehyde exposure is a valid occupational health and safety concern. Wooden furniture and refurbishing materials can pose serious health threats to occupants. The information in this study could act as a basis for future indoor air quality monitoring in Mainland China.

Implications: A university campus represents a microscale city environment consisting of all the working, living, and commercial needs of staff and students. The scope of this investigation covers 21 hazardous carbonyl species based on samples collected from 15 categories of workplaces and residential building in a university campus in southern China. Findings of the study provide a comprehensive assessment of indoor air quality with regards to workers’ health and safety. No similar study has been carried out in China.

Introduction

On average, an individual spends approximately 90% or more of their time indoors (U.S. Environment Protection Agency [EPA], 2009). Satisfactory indoor air quality is crucial to protecting the general public health, although carbonyl levels in indoor air have been reported to be much higher than those in outdoor air, indicating that local carbonyl emissions originate predominantly from indoor sources (Liu et al., 2006). In light of the growing awareness of occupational health and safety, the evaluation of indoor pollutant levels in various workplaces and residential environments has become increasingly imperative.

Chinese indoor air quality (IAQ) standards were established since the 1980s (Edwards et al., 2007). However, they were not thoroughly covered for all gaseous and particulate pollutants. Therefore, there is an urgent need for original research in Mainland China due to the wide variability in materials, climates, and human behaviors compared with other countries.

Carbonyls (aldehydes and ketones) are an important group of airborne organic compounds in atmospheric chemistry. These compounds photochemically dissociate to produce free radicals, which consequently initiate atmospheric reactions, such as ozone formation at ground level (Atkinson et al., 1992; Carlier et al., 1986; Seinfeld and Pandis, 1998). Carbonyls are also important intermediates in the formation of photochemical smog (Birks, 1998). Aldehydes and ketones can be either emitted into or formed in the atmosphere. Primary emission sources include both natural (e.g., biomass burning) and anthropogenic (e.g., automobile exhaust, cooking emission, incense burning, and environmental tobacco smoke [ETS]) sources (Clements et al., 2009; Ho et al., 2006, 2013b; Ho and Yu, 2002; Villanueva-Fierro et al., 2004; Wang et al., 2012).
The toxic health effects of many carbonyls are well documented (World Health Organization [WHO], 2000). Formaldehyde is classified as a human carcinogen by the International Agency for Research on Cancer (IARC) (IARC, 2006). The National Toxicology Program (2011), an interagency program of the U.S. Department of Health and Human Services, also named formaldehyde as a known human carcinogen in its 12th Report on Carcinogens. A National Cancer Institute (NCI) case-control study among funeral industry workers that characterized exposure to formaldehyde found an association between increasing formaldehyde exposure and mortality from myeloid leukemia (Hauptmann et al., 2009). Acetaldehyde is an irritant of the skin, eyes, mucous membranes, throat, and respiratory tract (Eckert et al., 2009). Symptoms of exposure to this compound include nausea, vomiting, and headache. Acetaldehyde is also a known animal carcinogen (WHO, 2000). In 2009, the IARC (2009) updated the classification of acetaldehyde, stating that acetaldehyde included in and generated endogenously from alcoholic beverages is a Group I human carcinogen. Acrolein causes eye irritation and odor annoyance and exacerbates asthma (Arntz et al., 2012). Feng et al. (2006) found that connections exist between acrolein gas in the ETS and the risk of lung cancer. In terms of the “noncarcinogenic health quotient” for components in ETS, acrolein dominates, contributing 40 times more than the next component, hydrogen cyanide (Hausmann, 2012). Common objects, such as furniture, carpets, fabrics, paints, and cooking fumes, as well as ETS, have been reported as indoor sources of carbonyls (Weisel et al., 2008).

In the present study, airborne carbonyl measurement was conducted in 15 types of indoor environments at a university campus in Mainland China. The scope of the investigation was to provide a comprehensive assessment of the indoor air quality with regards to the health and safety of academic staff, workers, and students. The university campus, more than just a study place, represents a microscale city consisting of all the working, living, and commercial needs of staff and students. The indoor air quality may be easily overlooked in such a peaceful and silent environment. No similar study has been conducted in Mainland China. This research can bring awareness to the Chinese public about their working and living environments, especially in the rapidly developing southern areas.

**Experimental**

**Sampling locations**

The study was conducted at a university in Xiamen, Fujian, China. The university has a faculty and staff of ca. 3000, of which 1200 are full-time teachers, including >600 professors and associate professors. The current enrollment of full-time students is approximately 1700. Fifteen representative workplaces and residential units in the campus were selected for the measurement and assessment of the occupational and residential exposures. A campus map shows their locations on the campus (Figure 1). The 12 categories included (1) departmental general offices, (2) faculty offices, (3) supporting staff offices, (4) lecture theaters, (5) small lecture rooms, (6) a chemistry laboratory, (7) a library, (8) a stationary store, (9) fast food courts, (10) staff restaurants, (11) a commercial bank, and (12) grocery stores. Three residential assessments were performed at (13) faculty apartments, (14) student dormitories, and (15) safe guard hostels. Table 1 lists the detailed descriptions of the workplaces and the residential units. The number of workers refers to the workers who were working in the sampling collection rooms and does not include any employees in other separated areas, offices, or pantries. The characteristics and potential pollution sources of the sampling locations were obtained through on-site investigation. More than one location was selected for each category of workplaces and residential unit, except for the chemistry laboratory (category 6), stationary store (category 7), library (category 8), and commercial bank (category 11). No mechanical ventilation systems were in operation in either the workplaces or residential units during the sampling periods. From category 1 to category 13, typical mechanical ventilations such as air-conditioning and exhaust fans were employed as usual practices. The air-conditioning units, if present, recirculated the indoor air supplemented with 10% outdoor fresh air supply. However, no fresh air is supplied with the exhaust fans. Natural ventilation (e.g., windows and doors) was found in the student dormitories (category 14) and safe guard hostels (category 15).

**Sample collection**

Air samples were collected in silica cartridges impregnated with acidified 2,4-dinitrophenylhydrazine (DNPH) (Sep-Pak DNPH-silica, 55–105 μm particle size, 125 Å pore size; Waters Corporation, Milford, MA, USA) at a flow rate of 0.7 L min⁻¹ for 240 min (EPA, 1999). The selected sampling time ensured that the collected carbonyls would not consume >30% of the derivatizing agent coated on the cartridge. Sampling was conducted during normal working hours between 08:00 a.m. and 06:00 p.m. For sites 13–15, the samples were collected within the occupant’s active period (mostly between 05:00 p.m. and 12:00 a.m.). In total, five visits were made for each sampling location from July 2012 to June 2013, and four samples were collected during each visit. Both samples were taken at a height of 1.5 m above the ground. Baseline sampling was conducted in each location during off-duty periods. No breakthrough was acknowledged at such sampling flow rates and sampling times (EPA, 1999; Herrington et al., 2007; Waters Corporation, 2007). The flow rates were checked in the field at the beginning and end of each sampling period using a calibrated flow meter (Glibrator Calibrator; Gilian Instruments, W. Caldwell, NJ, USA). A Teflon filter assembly (Whatman, Clifton, NJ, USA) and an ozone scrubber (Sep-Pak; Waters Corporation) were installed in front of the DNPH-silica cartridge to remove any particulate matter and prevent possible ozone interference, respectively (Spaulding et al., 1999). The recovery of the carbonyls collected in the process was not affected by the ozone trap (Ho and Yu, 2002; Ho et al., 2013a). Collocated samples were collected to determine sample collection reproducibility, which was found to be >95%. A cartridge was designated as a field blank on each sampling trip and was handled the same way as the sample cartridges. The amount of carbonyls detected in the cartridges was corrected for the field blank when the air concentrations of carbonyls were computed. Three baseline samples were
Figure 1. A map showing the sampling locations on the campus (the representations of numerical location are shown in Table 1).
<table>
<thead>
<tr>
<th>Sample Area No.</th>
<th>Description</th>
<th>Number of Locations</th>
<th>Area (m²)</th>
<th>Estimated Number of Related Workers/Occupants</th>
<th>Year of Last Decoration</th>
<th>Main Facilities</th>
<th>Environmental Conditions</th>
<th>Ventilation System Equipped</th>
</tr>
</thead>
<tbody>
<tr>
<td>Workplace</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>Departmental offices</td>
<td>1</td>
<td>45–64</td>
<td>600</td>
<td>2008</td>
<td>Mechanical</td>
<td>Temp (°C): 13–22</td>
<td>RH (%): 77–89</td>
</tr>
<tr>
<td>2</td>
<td>Faculty offices</td>
<td>3</td>
<td>13–22</td>
<td>1200</td>
<td>2008</td>
<td>Mechanical</td>
<td>Temp (°C): 14–22</td>
<td>RH (%): 67–91</td>
</tr>
<tr>
<td>3</td>
<td>Supporting staff offices</td>
<td>3</td>
<td>45–62</td>
<td>500</td>
<td>2008</td>
<td>Mechanical</td>
<td>Temp (°C): 17–22</td>
<td>RH (%): 71–89</td>
</tr>
<tr>
<td>4</td>
<td>Lecture rooms</td>
<td>3</td>
<td>120–176</td>
<td>1200</td>
<td>2010</td>
<td>Mechanical</td>
<td>Temp (°C): 13–22</td>
<td>RH (%): 73–87</td>
</tr>
<tr>
<td>5</td>
<td>Chemistry laboratory</td>
<td>1</td>
<td>90</td>
<td>200</td>
<td>2003</td>
<td>Mechanical</td>
<td>Temp (°C): 15–22</td>
<td>RH (%): 68–89</td>
</tr>
<tr>
<td>6</td>
<td>Library</td>
<td>1</td>
<td>120</td>
<td>10</td>
<td>2011</td>
<td>Mechanical</td>
<td>Temp (°C): 15–23</td>
<td>RH (%): 70–89</td>
</tr>
<tr>
<td>7</td>
<td>Stationary store</td>
<td>3</td>
<td>250–300</td>
<td>80</td>
<td>2002</td>
<td>Mechanical</td>
<td>Temp (°C): 12–28</td>
<td>RH (%): 65–82</td>
</tr>
<tr>
<td>10</td>
<td>Student apartments</td>
<td>3</td>
<td>90–110</td>
<td>2400</td>
<td>2011</td>
<td>Mechanical</td>
<td>Temp (°C): 13–24</td>
<td>RH (%): 75–84</td>
</tr>
<tr>
<td>Residential Unit</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>Faculty dormitories</td>
<td>3</td>
<td>35–42</td>
<td>1700</td>
<td>2010</td>
<td>Mechanical</td>
<td>Temp (°C): 12–16</td>
<td>RH (%): 65–93</td>
</tr>
<tr>
<td>14</td>
<td>Student apartments</td>
<td>3</td>
<td>30</td>
<td>13–31</td>
<td>2006</td>
<td>Mechanical</td>
<td>Temp (°C): 12–16</td>
<td>RH (%): 63–95</td>
</tr>
<tr>
<td>15</td>
<td>Safety guard</td>
<td>3</td>
<td>12–16</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
collected during nonoperation hours on each site. The DNPH-coated cartridges were stored in a refrigerator at <4 °C after sampling and before analysis, and they were analyzed in our laboratory within 2 weeks. The temperature and relative humidity (RH) were recorded during the sampling period.

Sample analysis

A total of 21 carbonyls were quantified (Table 2). Unsataturated carbonyls, including acrolein and crotonaldehyde, were detected, but their abundances were not reported in the study. The unsaturated carbonyl DNPH-hydrazone could react with excess reagent to form adducts, which could not be quantified accurately due to chromatographic and response factor issues (Ho et al., 2011; Schulte-Ladbeck et al., 2001).

Each DNPH-coated cartridge was eluted with 2.0 mL of acetone-free acetonitrile (high-performance liquid chromatography [HPLC] grade; Mallinckrodt Laboratory Chemicals, Phillipsburg, NJ, USA) into a volumetric flask. Testing was performed to demonstrate that any DNPH or its derivatives remaining in the cartridge were undetectable after the 2.0 mL elution (Ho et al., 2007). Certified calibration standards of monocarbonyl DNPH-hydrzones were purchased from Supelco (Bellefonte, PA, USA). The dicarbonyl calibration standards were prepared by mixing glyoxal and methylglyoxal (Sigma, St. Louis, MO, USA) in acetonitrile with DNPH in an acidic aqueous solution. The mixtures were allowed to stand at room temperature for at least 6 hr for complete derivatization. The final volume of each calibration mixture was brought to 2.0 mL with 8.2 (v/v) acetonitrile/pyridine (HPLC grade; Sigma). The concentrations of the dicarbonyl DNPH-hydrzones in the calibration standards ranged from 0.01 to 4.0 µg mL⁻¹. Linearity was indicated by a correlation coefficient (r²) value greater than 0.999. The cartridge extracts and calibration standards were analyzed by injecting 20 µL of the samples into a HPLC system (series 1200; Agilent Technology, Santa Clara, CA, USA) equipped with a photodiode array detector (DAD). The column for separation was a 4.6 × 250 mm Spheri-5 ODS 5 µm C-18 reversed-phase column (PerkinElmer, Norwalk, CT, USA) operated at room temperature. The mobile phase consisted of three solvent mixtures: (A) 6:3:1 (v/v/v) water/acetonitrile/tetrahydrofuran, (B) 4:6 (v/v) water/acetonitrile and (C) acetonitrile. The gradient program was 80% A/20% B for 1 min, followed by a linear gradient to 50% A/50% B in 8 min, 100% B for 10 min, 100% C for 6 min, and, finally, 100% C for 5 min. The flow rate was 2.0 mL min⁻¹ throughout the run. The absorbances at 360, 390, and 420 nm were used for the identification of the aliphatic monocarboxyls, aromatic monocarboxyls (benzaldehyde, tolualdehydes, and 2,5-dimethylbenzaldehyde), and dicarboxyls (glyoxal and methylglyoxal), respectively. The identification and quantification of carbonyl compounds were based on the retention times and the peak areas of the corresponding calibration standards, respectively. The minimum detection limit (MDL) was obtained by analyzing a minimum of seven replicates of a standard solution containing the analytes at a concentration of 0.015 µg mL⁻¹. The MDLs of the target carbonyls ranged from 0.0042 to 0.0092 µg mL⁻¹, which is equal to 0.07–0.27 ppb, with a sampling volume of 0.164 m³ (refer to Table 2).

Table 2. Minimum detection limits (MDL) for the target carbonyls

<table>
<thead>
<tr>
<th>Anonym</th>
<th>Carbonyl</th>
<th>CAS#</th>
<th>MW</th>
<th>Class</th>
<th>MDL (ppb, a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>Formaldehyde</td>
<td>50-00-0</td>
<td>30</td>
<td>Aliphatic monocarboxyl</td>
<td>0.12</td>
</tr>
<tr>
<td>C2</td>
<td>Acetaldehyde</td>
<td>75-07-0</td>
<td>44</td>
<td>Aliphatic monocarboxyl</td>
<td>0.13</td>
</tr>
<tr>
<td>ACE</td>
<td>Acetone</td>
<td>67-64-1</td>
<td>58</td>
<td>Aliphatic monocarboxyl</td>
<td>0.27</td>
</tr>
<tr>
<td>nC3</td>
<td>Propionaldehyde</td>
<td>123-38-6</td>
<td>58</td>
<td>Aliphatic monocarboxyl</td>
<td>0.08</td>
</tr>
<tr>
<td>MEK</td>
<td>Methyl ethyl ketone</td>
<td>78-93-3</td>
<td>72</td>
<td>Aliphatic monocarboxyl</td>
<td>0.07</td>
</tr>
<tr>
<td>iC4</td>
<td>iso-Butyraldehyde</td>
<td>78-84-2</td>
<td>72</td>
<td>Aliphatic monocarboxyl</td>
<td>0.17</td>
</tr>
<tr>
<td>nC4</td>
<td>n-Butyraldehyde</td>
<td>123-72-8</td>
<td>72</td>
<td>Aliphatic monocarboxyl</td>
<td>0.17</td>
</tr>
<tr>
<td>Benz</td>
<td>Benzaldehyde</td>
<td>100-52-7</td>
<td>106</td>
<td>Aromatic monocarboxyl</td>
<td>0.11</td>
</tr>
<tr>
<td>iC5</td>
<td>iso-Valeraldehyde</td>
<td>590-86-3</td>
<td>86</td>
<td>Aliphatic monocarboxyl</td>
<td>0.13</td>
</tr>
<tr>
<td>nC5</td>
<td>n-Valeraldehyde</td>
<td>110-62-3</td>
<td>86</td>
<td>Aliphatic monocarboxyl</td>
<td>0.16</td>
</tr>
<tr>
<td>o-tol</td>
<td>o-Tolualdehyde</td>
<td>529-20-4</td>
<td>120</td>
<td>Aromatic monocarboxyl</td>
<td>0.18</td>
</tr>
<tr>
<td>m-tol</td>
<td>m-Tolualdehyde</td>
<td>529-20-4</td>
<td>120</td>
<td>Aromatic monocarboxyl</td>
<td>0.19</td>
</tr>
<tr>
<td>p-tol</td>
<td>p-Tolualdehyde</td>
<td>104-87-0</td>
<td>120</td>
<td>Aromatic monocarboxyl</td>
<td>0.21</td>
</tr>
<tr>
<td>C6</td>
<td>Hexaldehyde</td>
<td>66-25-1</td>
<td>100</td>
<td>Aliphatic monocarboxyl</td>
<td>0.17</td>
</tr>
<tr>
<td>C7</td>
<td>Heptaldehyde</td>
<td>111-71-7</td>
<td>114</td>
<td>Aliphatic monocarboxyl</td>
<td>0.21</td>
</tr>
<tr>
<td>C8</td>
<td>Octaldehyde</td>
<td>124-13-0</td>
<td>128</td>
<td>Aliphatic monocarboxyl</td>
<td>0.22</td>
</tr>
<tr>
<td>C9</td>
<td>Nonaldehyde</td>
<td>124-19-6</td>
<td>142</td>
<td>Aliphatic monocarboxyl</td>
<td>0.20</td>
</tr>
<tr>
<td>C10</td>
<td>Decaldehyde</td>
<td>112-31-2</td>
<td>156</td>
<td>Aliphatic monocarboxyl</td>
<td>0.23</td>
</tr>
<tr>
<td>2,5-DB</td>
<td>2,5-Dimethylbenzaldehyde</td>
<td>5779-94-2</td>
<td>134</td>
<td>Aromatic monocarboxyl</td>
<td>0.24</td>
</tr>
<tr>
<td>gly</td>
<td>Glyoxal</td>
<td>107-22-2</td>
<td>58</td>
<td>Dicarboxyl</td>
<td>0.23</td>
</tr>
<tr>
<td>mgly</td>
<td>Methylglyoxal</td>
<td>78-98-8</td>
<td>72</td>
<td>Dicarboxyl</td>
<td>0.19</td>
</tr>
</tbody>
</table>

Notes: a The MDL is the minimum detection limit of a carbonyl on a cartridge. The MDL expressed as ppb, is calculated using a sampled air volume of 0.168 m³ (at a flow rate of 0.7 L/min for 240 min). b iso-Butyraldehyde and n-butyraldehyde were co-eluted in the HPLC analysis.
The measurement precision ranges from 0.7% to 3.3% for the target carbonyls.

Health risk calculation

The carcinogenic risks of chronic exposure to carbonyls were assessed in this study. The risk estimation with a cancer endpoint is expressed in terms of the probability of developing cancer from a lifetime of continuous exposure to carbonyls. In the cancer risk assessment, the academic and supporting staffs were all full-time workers in the selected sites, and the main exposure route of interest was inhalation. The chronic daily intake (CDI) of a carcinogenic contaminant is controlled by various factors, such as exposure frequency, exposure duration, and the body weight of the receptor. The equation used to calculate CDI in mg kg\(^{-1}\) day\(^{-1}\) is

\[
CDI = \frac{Ca \times ET \times ED}{BW \times AT \times 365}\tag{1}
\]

where \(Ca\) is the contaminant concentration (mg m\(^{-3}\)), \(IR\) is the inhalation rate (m\(^3\) hr\(^{-1}\)), \(ET\) is the exposure time (hr day\(^{-1}\)), \(EF\) is the exposure frequency (day yr\(^{-1}\)), \(ED\) is the exposure duration (year), \(BW\) is the body weight (kg), and \(AT\) is the average lifetime (years). The EPA has established standard values for average body weights and lifetimes for adults and children (EPA, 1994).

The lifetime cancer hazard risk \((R)\) is calculated using the equation:

\[
R = CDI \times PF\tag{2}
\]

where \(PF\) is the cancer potency factor in units of kg day\(^{-1}\) mg\(^{-1}\) of a specific carcinogenic substance. The values of \(PF\) were obtained from the Integrated Risk Information System (IRIS) developed by the EPA (EPA, 2012).

Results and Discussion

Indoor workplace carbonyl levels

The concentrations of carbonyls in the sampled workplaces varied widely depending on the sizes, potential indoor pollution sources, and ventilation systems of the workplaces (refer to Table 3). During the sampling, any activity potentially generating additional pollutants was prohibited in all workplaces. No diurnal or seasonal variations in either the absolute values or molar compositions were found for our sample sets. The concentrations of the carbonyls were statistically the same according to Student’s \(t\) test, with a 95% confidence level for the samples collected at the same location at different times. The average value was thus taken to express the individual carbonyls that existed in different workplaces.

The absolute concentrations should not be compared among the indoor workplaces. A compound can be found at a trace level but can still cause very serious health problems. In contrast, another compound can be found at relatively high concentrations but may not be very toxic to humans and animals. The cancer or hazard potentials are more relevant and are discussed in other sections. However, the molar composition profiles have enormous value to illustrate and interpret the potential indoor pollution sources at particular sites.

The carbonyl molar composition profiles are plotted in Figure 2 for comparison of the relative abundance of the different carbonyls in the various categories of nonsmoking workplaces. The mean values are plotted because more than one location of the same category was sometimes available (except for categories 6–8, 11). The molar carbonyl profiles of the clerical offices and classrooms (categories 1–5) were dissimilar from those of the public workplaces (categories 6–12) on the university campus. Formaldehyde was the largest carbonyl contributor in the clerical locations, where it accounted for 57.4–66.3% of the total quantified carbonyls. These workplaces mainly consisted of wooden facilities, such as desks, benches, ledges, seats, and floorboards. Wooden materials and lacquer coatings used for furnishing and decoration can be a source of indoor carbonyls (e.g., formaldehyde and acetone) (Brown, 1999, 2002; Kelly, 1999). Hexanal is not a universally abundant carbonyl in most indoor and outdoor environments. Its relatively large concentration (9.1–13.32%) can be ascribed to emissions from the dried wood. Hexanal, acting as a measure of rancidity, is produced during the degradation process of wood (Svedberg et al., 2004). Hence, its emission from the wooden facilities is identifiable and significantly affects the indoor air quality.

In the public workplaces, the contribution of formaldehyde varied greatly, from 20.4% to 44.8%, whereas acetone, acetaldehyde, and nonaldehyde were close to or even greater than the concentration of formaldehyde. This result can be attributed to the discrepancies of the indoor pollution sources, air circulation, and ventilation. The contribution of acetone was the highest (46.8%) in the chemistry laboratory (category 6). Acetone serves as a common solvent or intermediate in various organic syntheses and chemical reactions (WHO, 1998; Huang et al., 2007). The distribution of carbonyls was even in the stationary store (category 7), which is equipped with eight photocopiers. Electronic equipment, such as photocopiers, can 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) (Dales et al., 2008; Yu and Crump, 1998; Lee et al., 2006). Similar carbonyl profiles were also seen in the library (category 8). The carbonyl formation from the application of photocopiers can affect air quality. The off-gases from a large amount of wooden bookshelves also contributed to the indoor air pollution in the library. Formaldehyde and nonaldehyde were the two largest molar contributors in the cooking-related workplaces. The open cooking activities at each kiosk in the fast food courts (category 9) and the semiopen kitchen design in the staff restaurants (category 10) allow emissions to be exhausted directly into the dining areas. Our molar compositions were consistent with the studies that found that fuel combustion and cooking activities produce high levels of carbonyls, particularly formaldehyde, acetaldehyde, and nonaldehyde (Ho et al., 2006; Schauer et al., 2001; Zhang and Smith, 1999). The carbonyl composition profiles did not show any specific pattern for the commercial bank (category 11) and grocery store (category 12). Most of these facilities are made of galvanized steel rather than solid wood. Apart from the consumption of cleaning detergents and floor
<table>
<thead>
<tr>
<th>Category</th>
<th>Number of Samples</th>
<th>C1</th>
<th>C2</th>
<th>ACE</th>
<th>nC3</th>
<th>MEK</th>
<th>i+4C6</th>
<th>benz</th>
<th>iC5</th>
<th>nC5</th>
<th>o- tol</th>
<th>m- tol</th>
<th>p- tol</th>
<th>C6</th>
<th>C7</th>
<th>C8</th>
<th>C9</th>
<th>C10</th>
<th>2,5-DB</th>
<th>gly</th>
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<td>57.6±14.4</td>
<td>4.12±2.45</td>
<td>9.32±3.19</td>
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<td>0.64±0.134</td>
<td>0.96±0.44</td>
<td>0.60±0.22</td>
<td>0.22±0.19</td>
<td>0.84±0.36</td>
<td>9.56±2.43</td>
<td>1.24±0.43</td>
<td>0.64±0.27</td>
<td>0.48±0.16</td>
<td>0.84±0.35</td>
<td>0.88±0.54</td>
<td>0.53±0.24</td>
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<td>3.34±1.20</td>
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<td>0.58±0.22</td>
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<td>0.36±0.17</td>
<td>0.30±0.12</td>
<td>0.40±0.16</td>
<td>4.38±1.33</td>
<td>0.54±0.20</td>
<td>6.64±0.18</td>
<td>0.76±0.22</td>
<td>42.0±12.3</td>
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<tr>
<td>Lecture theaters</td>
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<td>8.60±3.32</td>
<td>1.03±0.66</td>
<td>1.33±0.76</td>
<td>0.26±0.18</td>
<td>0.17±0.06</td>
<td>0.18±0.08</td>
<td>0.09±0.06</td>
<td>0.31±0.15</td>
<td>0.33±0.21</td>
<td>1.99±0.86</td>
<td>0.18±0.13</td>
<td>2.4±0.08</td>
<td>0.27±0.15</td>
<td>150.0±5.66</td>
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</tr>
<tr>
<td>Small lecture rooms</td>
<td>80</td>
<td>13.4±5.26</td>
<td>1.37±0.76</td>
<td>1.45±0.71</td>
<td>0.37±0.23</td>
<td>0.22±0.11</td>
<td>0.27±0.14</td>
<td>0.13±0.07</td>
<td>0.28±0.24</td>
<td>0.45±0.25</td>
<td>1.87±1.54</td>
<td>0.12±0.08</td>
<td>0.22±0.06</td>
<td>0.36±0.16</td>
<td>205.0±8.77</td>
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<td>67.2±24.9</td>
<td>45.6±17.2</td>
<td>154.4±21.2</td>
<td>16.7±4.32</td>
<td>12.4±3.65</td>
<td>15.8±4.16</td>
<td>3.98±1.86</td>
<td>1.76±0.82</td>
<td>4.34±1.33</td>
<td>0.78±0.45</td>
<td>0.56±0.23</td>
<td>1.13±0.52</td>
<td>0.87±0.32</td>
<td>0.62±0.27</td>
<td>0.76±0.41</td>
<td>1.33±0.44</td>
<td>1.68±0.54</td>
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<td>Staff apartment</td>
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<td>26.5±7.23</td>
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<td>10.4±4.35</td>
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<td>Student dormitory</td>
<td>80</td>
<td>18.1±5.12</td>
<td>2.35±0.98</td>
<td>3.24±1.35</td>
<td>1.07±0.43</td>
<td>0.43±0.28</td>
<td>0.86±0.54</td>
<td>0.26±0.19</td>
<td>0.57±0.31</td>
<td>0.66±0.41</td>
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<td>0.23±0.13</td>
<td>1.39±0.16</td>
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<td>30.8±7.68</td>
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<td>2.34±0.76</td>
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<td>1.34±0.76</td>
<td>1.96±1.13</td>
<td>73.1±20.3</td>
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</tbody>
</table>

Notes: * Anonyms as defined in Table 2. bd = below minimum detection limit.
cleaners, there were no obvious indoor pollution sources in these two categories of workplace. The academic staff, workers, students, and other customers frequently access these public locations. Their visits can pose variability in the carbonyl concentrations and compositions, but their influences cannot be taken into account.

Figure 3 compares the concentration levels of carbonyls in each category of workplace during the on- and off-duty periods. The results indicate that a substantial increase in carbonyl concentrations was directly related to the work duty in certain workplaces, such as those of the library, the stationary store, the fast food courts, and the staff restaurants. For instance, in the fast food courts, the concentrations of formaldehyde, acetaldehyde and hexaldehyde during normal working hours were 70.2%, 70.0%, and 68.7%, respectively, higher than during the nonoperation periods. However, the baseline levels of formaldehyde and acetone in these workplaces were high, even when no activity was taking place. Poor ventilation systems cannot efficiently remove or dilute polluted indoor air. In addition, secondhand emissions from factory walls or other absorbable surfaces can retain high baseline levels of these carbonyls (Katsumata et al., 2008).

Effect of cleaning reagents

Organic cleaning agents are not usually used in the academic offices. A comparison study was conducted in two identical supporting staff offices to investigate its effect on indoor carbonyls. Half a year before sampling, daily cleaning with floor cleaners and detergents was performed in one office, and another office was cleaned with pure water only. Figure 4a shows the carbonyl composition profiles for the two offices. The contributions of formaldehyde, acetaldehyde, acetone, and n-/iso-butyraldehyde in the organic solvent-cleaned office were much higher than in the water-cleaned office. Such variations were consistent with previous findings that these compounds can be released from cleaning reagents and floor cleaners (Huang et al., 2011).

Effect of tobacco smoking

Figure 4b evaluates the composition profiles for two smoking and nonsmoking size-equivalent faculty offices. The two offices
were wholly refurnished 4 yr prior to this study and were furnished with all the same working units, equipment, and ventilation (i.e., air-conditioning and number of windows). The office doors and windows were constantly kept closed. There was a lack of any stationary or mobile indoor pollution sources in the offices. The average contributions of methyl ethyl ketone (MEK), n-/iso-butyraldehyde, benzaldehyde, and n-valeraldehyde in the smoking office exceeded the average contributions in the nonsmoking office by a factor of 2.5–4.3. This result is consistent with our findings in an environmental chamber study in which these carbonyls were the organic carbonyl tracers for ETS (Wang et al., 2012). In addition, compared with other nonsmoking workspaces, the highest contribution of MEK and benzaldehyde was seen (4.2% and 3.7%, respectively). A durable association between the indoor carbonyls and ETS was thus shown (Hodgson et al., 1996; Marchand et al., 2006; Bari et al., 2011; Panagopoulos et al., 2011).

Effect of furnished materials

Carbonyl concentrations were measured at two freshly refurnished faculty offices that were equipped with similar-sized desks, bookcase combinations, cabinets, and drawers made of wooden and galvanized steel. The swivel chairs, windows, lamps, curtains, and floorboards were equivalent. As shown in Figure 4c, formaldehyde was the highest carbonyl contributor in the wooden-furnished offices, followed by hexanal and acetone. The three most abundant species accounted for 61.4%, 11.3%, and 10.2% of the total quantified carbonyls on the basis of molar ratios. A different carbonyl profile was seen for the samples collected in the galvanized steel-furnished office. Acetone had the highest carbonyl contribution (34.3%). Formaldehyde and acetaldehyde were the next two largest contributors, accounting for 28.7% and 22.4%, respectively. The results further prove that hexanal is an essential indoor source marker for freshly made wooden facilities. Acetone is widely utilized in lacquers for either wooden- or galvanized steel-furniture finishes, pointing to its significance in the newly refurnished rooms (WHO, 1998).

Campus residential carbonyl levels

Carbonyl concentrations were measured in the campus residential units (see Table 3). There was no equipment (e.g., household heaters) that could potentially generate indoor pollutants. However, a few factors may influence the variations of carbonyl levels. For example, visitors may bring in polluting substances, or the windows may be opened occasionally permitting air exchange in the rooms. We assumed that these uncertainties may dilute or elevate the carbonyl levels but did not vary the carbonyl composition, based on their steady profiles (relative standard deviation [RSD] = ±4.3%). In the living room of the faculty apartments (category 13), formaldehyde was the most dominant carbonyl, followed by acetone, nonaldehyde, acetaldehyde, and hexanal. The dining tables and chairs, television benches, cabinets, decorations, and floorboards were made of solid wood. In addition to the wooden-furnishing and
decorating materials, daily household cooking activities can contribute to the indoor pollution, a theory supported by the high contribution of nonaldehyde (8.7%). Cooking emissions can be released from the kitchens by occasionally opening doors during or post cooking. Human activities, such as the use of cleaning reagents and sprays, may also generate different degrees of carbonyls (Singer et al., 2006; Guo et al., 2000). In the student dormitories (category 14), formaldehyde, acetone, and hexanal were the top three carbonyls. Simple wooden furniture, including beds, wardrobes, desks, chairs, and floorboards, was placed in the dormitories. Both smoking and cooking were prohibited, and no explicit indoor pollution source was present. The application of household chemical products was also limited. The composition profile was similar to the clerical workplaces (categories 1–5), suggesting that the off-gases from wooden furniture are the most dominant source. The carbonyl contributions were mixed in the safe guard hostels (category 15). Only a wooden bed, wardrobe, and small cabinet were present in these small semiclosed rooms built next to the gates of the university. However, smoking is allowed; hence, the compositions of the ETS source markers MEK (5.9%), 1-/iso-butyraldehyde (3.8%), benzaldehyde (3.2%), and n-valeraldehyde (3.3%) were higher than those in the other two residential units.

**RH influences**

It should be noted that RH is a potential factor in controlling off-gassing of formaldehyde and other carbonyls from the materials. Despite the use of air-conditioning, the average RH at the sampling sites was above 70%. The off-gassing rates of VOCs

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**Figure 4.** Variations in carbonyl composition profiles in the comparison studies.
and other gases can be enhanced under high RH environments (Sidheswaran et al., 2013; Kuang et al., 2009; Nnadili et al., 2011). It is thus believable that the off-gassing of carbonyls is more critical in humid southern China.

**Occupational guideline comparison**

Formaldehyde was one of the most abundant carbonyls found in the workplaces. The National Institute of Occupational Safety and Health (NIOSH)-recommended exposure limit (REL) for formaldehyde for an 8- or 10-hr time-weighted average (TWA) exposure and/or ceiling is 16.3 ppb, (20 μg m⁻³). The average concentrations of formaldehyde in many of the indoor workplaces exceeded this exposure limit. These workplaces were mainly equipped with a large proportion of wooden facilities and other potential indoor pollution sources, such as chemicals and cooking emissions.

Acetaldehyde was often the next most abundant carbonyl species in the total carbonyl mixing ratio. The American Conference of Governmental Industrial Hygienists (ACGIH) threshold limit value (TLV) ceiling was set at 25 parts per million by volume (ppm), which is equivalent to 45 mg m⁻³; the Occupational Safety and Health Administration (OSHA) set its permissible exposure limit (PEL) for general industry at 200 ppm, (360 mg m⁻³) for every 8-hr TWA. The mixing ratios for acetaldehyde measured in this study were at least 2 orders of magnitude below the established exposure limits.

Off-gases from lacquers on the outer layer of furniture and the daily consumption of organic solvents (e.g., detergents) elevate acetone levels in workplaces. The health effects of acetone have been studied extensively, and the compound is generally classified by the EPA (EPA, 2012). Inhalation exposure is frequently associated with exposure frequency, duration, and activity patterns; these elements are essential factors in the calculation of chronic daily intake and lifetime cancer hazard risk. Several assumptions have been made based on suggestions by the EPA in regards to the relative carcinogenic assessment. The volume of air inspired by a typical light duty worker is assumed to be 0.8 and 0.5 m³ hr⁻¹ for men and women, respectively, and by a moderate duty worker is assumed to be 2.5 and 1.6 m³ hr⁻¹ for men and women, respectively, and the absorption factor of carbonyl is estimated at 90% (EPA, 1985). Based on our on-site investigation, we classified workers in the fast food courts, faculty restaurants, and groceries as moderate duty, compared with the light job duty for academic, research, and clerical staff in other indoor workplaces. It has also been assumed that a worker generally spends 8 hr per day and 5 days per week during his/her 40 working years in the workplace. For the residential units, the volume of air inspired by a resting occupant is assumed to be 0.7 and 0.3 m³ hr⁻¹ for men and women, respectively, with an absorption factor of 90%. Residential living was assumed to be a 24-hr exposure, 7 days per week. An average body weight of 70 kg (male) and 60 kg (female) and an average lifetime of 69 (male) and 72 (female) yr are assumed (EPA, 1994). The PF values for formaldehyde and acetaldehyde are 0.045 and 0.0077 kg day⁻¹ mg⁻¹, respectively (EPA 2012). The estimated chronic daily intake and lifetime cancer hazard risk for formaldehyde and acetaldehyde are summarized in Table 4. The lifetime cancer hazard risks associated with formaldehyde ranged from 0.16 × 10⁻⁴ to 6.37 × 10⁻⁴ and from 0.11 × 10⁻⁴ to 4.56 × 10⁻⁴ for males and females, respectively. For acetaldehyde, the risks were 0.84 × 10⁻⁷ to 0.83 × 10⁻⁴ and 0.59 × 10⁻⁷ to 0.594 × 10⁻⁴ for males and females, respectively. Risk values below one in a million (<1 × 10⁻⁶) are typically considered below the level of concern, whereas a risk value above 100 in a million (>1 × 10⁻⁴) signifies an immediate need to initiate interventions to protect human health (Lee et al., 2006). Formaldehyde poses a higher cancer risk to the workers compared with acetaldehyde. The lifetime cancer hazard risks associated with acetaldehyde in all of the workplaces were well below 1 × 10⁻⁴. However, for formaldehyde, the values were all above the concern risk level (1 × 10⁻⁶). The risks for the fast food courts and staff restaurants...
Table 4. Chronic daily intake (CDI) and lifetime cancer hazard risk ($R$) for formaldehyde and acetaldehyde in the workplaces and residential units

<table>
<thead>
<tr>
<th>Description</th>
<th>Duty Mode</th>
<th>Male ET (hr/day)</th>
<th>Male EF (day/yr)</th>
<th>Male ED (yr)</th>
<th>Conc. (µg/m³)</th>
<th>CDI ($\times 10^{-4}$)</th>
<th>$R$ ($\times 10^{-4}$)</th>
<th>Female ET (hr/day)</th>
<th>Female EF (day/yr)</th>
<th>Female ED (yr)</th>
<th>Conc. (µg/m³)</th>
<th>CDI ($\times 10^{-4}$)</th>
<th>$R$ ($\times 10^{-4}$)</th>
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<tr>
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<td>0.5</td>
<td>8 5 × 52</td>
<td>40</td>
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<td>166</td>
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<td>0.5</td>
<td>8 5 × 52</td>
<td>40</td>
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<td>8 5 × 52</td>
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<td>1.28</td>
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**Notes:**
- Chronic daily intake in mg kg⁻¹ day⁻¹.
- Lifetime cancer hazard risk based on the following assumptions: average body weights of 70 kg for male and 60 kg for female, and a lifetime of 69 yr for male and 72 yr for female.
were 3.7–4.5 times higher than the alarm level (1 × 10⁻⁴). The results demonstrate that formaldehyde exposure in the workplace is a valid occupational health and safety concern, and proper actions should be taken promptly to protect the workers. High risks were also measured in the residential units in the campus. This result reflects the fact that the wooden furniture and refurbishing materials can pose serious health impacts to the occupants.

The cancer or hazard potentials of the other minor carbonyl compounds cannot be assessed because of a lack of guidelines and health factors. However, their measurements are still advantageous for the identification of potential indoor sources and source apportionment. The data can also be preserved to enact indoor air quality regulations that have not yet been developed in Mainland China.

Conclusion

We investigated the carbonyl levels and their potential indoor sources in the workplaces and residential units on a university campus. The high risks of formaldehyde were demonstrated to be a concern to the academic staff, workers, and students. The results show that off-gases from wooden furniture and refurbishing materials can elevate certain carbonyl levels. Other anthropogenic pollution sources, such as ETS and uses of cleaning reagents, can be greatly influenced indoor air quality by direct emission or generation of other VOCs. In order to dilute formaldehyde and other carbonyls levels, advancements and specific design of mechanical or natural ventilations are suggested. The information supports the importance of establishing routine indoor air quality monitoring and health relevance assessment for the workplaces and residential units of Mainland China.

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References


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