*Aerosol and Air Quality Research*, 16: 1967–1980, 2016

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2015.05.0312

# Risk Assessment of Indoor Formaldehyde and Other Carbonyls in Campus Environments in Northwestern China

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## ABSTRACT

Risk assessment for indoor formaldehyde and other carbonyls was investigated at an university in Xi'an, Shaanxi, China. Eight representative locations, including six indoor workplaces and two residential units of staff apartments and a student dormitory, were chosen. The indoor pollution origins were identified according to the variability in molar composition and correlation analysis for the target species. Environmental tobacco smoke (ETS), cooking activities, and office technologies such as printers and copiers can produce different degrees of carbonyls in the workplace. A one-year demonstration study conducted in a apartment showed significance of the off-gases from lacquers and new wooden furniture. The concentrations of formaldehyde and acetaldehyde in the most sampling locations were above the recommended exposure limits, reflecting a potential health risk to workers and occupants. Chronic daily intake (CDI) and lifetime cancer hazard risk (R) were calculated to assess the carcinogenic risks of chronic exposure to the carbonyls. The R values for formaldehyde exceeded the alarm level of $1 \times 10^{-6}$ in all sampled workplaces, but lower R values were associated with acetaldehyde. The results indicate that exposure of formaldehyde is a critical occupational health and safety concern. In addition, high risks associated with formaldehyde were also measured in the staff apartment, suggesting that the refurbishing materials and wooden furniture can potentially cause health impacts to occupants. The findings are informative to be referred in establishment of indoor air quality guidelines in China.

***Keywords:*** Carbonyls; Indoor pollution; Cancer risk; University campus.

## INTRODUCTION

Carbonyls, including aldehydes and ketones, are ubiquitous among harmful pollutants in the atmosphere. This class of compounds has been receiving more regulatory, scientific, and public attention because of their active roles in atmospheric reactions and potential adverse health impacts on humans (WHO, 2010). Carbonyl compounds are the direct precursors of peroxyacylnitrates and ozone ($O_3$) from photolysis or reactions of carbonyls with a hydroxyl radical (OH) to generate peroxyradicals (Finlayson-Pitts and Pitts, 1986; Lary and Shallcross, 2000). Carbonyls act as major contributors to photochemical smog in the urban atmosphere (Atkinson, 2000). Photochemical degradation is a major source of carbonyls in nature (Moortgat, 2001). Airborne carbonyls are emitted from incomplete combustion of fossil fuels in industrial plants, incinerators, automobiles and anthropogenic biomass burning (Grimaldi *et al.*, 1995;

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Lewtas, 2007). Industrial resins used in manufacturing of polymeric products, such as paints and adhesives, are pollution sources as well (Fjällström *et al.*, 2003). Indoors, carbonyls can be directly released from wooden furniture, building materials, and household products and formed through reactions between indoor volatile organic compounds (VOCs) (e.g., alkenes) and oxidants (e.g., $O_3$) (Morrison *et al.*, 2002; Clarisse, 2003; Poppendieck *et al.*, 2007). Formaldehyde and acetaldehyde are two abundant pollutants in residential units, offices, and schools because of their existences in wooden materials used for manufacturing decoration and furniture (Yu and Brump, 1999; Yu and Kim, 2011a, b). Particular indoor sources for several carbonyls are human activities, such as cooking, and the production of environmental tobacco smoke (ETS) (Guerin *et al.*, 1992).

A few carbonyls can cause irritation to mucous membranes in eyes and in the respiratory system (WHO, 2010). Formaldehyde is classified as a human carcinogen by the International Agency for Research on Cancer (IARC) (IARC, 2006). Acetaldehyde is also a known animal carcinogen (WHO, 2000) and cause irritations to eye, mucous membrane, skin, throat and respiratory tract (Eckert *et al.*, 2009). Symptoms of exposure to acetaldehyde include nausea, vomiting, and headache. Acrolein, an unsaturated carbonyl, is an eye irritant and exacerbates asthma (Arntz *et al.*, 2012). Feng *et al.* (2006) demonstrated that the lung cancer risk was elevated from inhalation of acrolein emitted from ETS.

Indoor carbonyl concentrations have been measured worldwide in various microenvironments, such as hospitals (Yu and Crump, 2006), temples (Ho and Yu, 2002), academic institutes (Cavalcante *et al.*, 2005; Crump *et al.*, 2005; Yamashita *et al.*, 2011), subway stations and tunnels (Ho *et al.*, 2007), residential buildings (Crump *et al.*, 1997; Huang *et al.*, 2011), shopping centers (Tang *et al.*, 2009), hotels (Feng *et al.*, 2004; Chan *et al.*, 2011), cinemas (Weng *et al.*, 2009), offices (Yu and Crump, 2000; Ongwandee *et al.*, 2009), museums (Báez *et al.*, 2003) and photocopy centers (Lee *et al.*, 2006). However, such an evaluation is still not comprehensive on the Mainland of China (Wang *et al.*, 2007), which has an urgent need for specialized research because of the large changeability in meteorological conditions and human behavior observed in China.

A school campus is a micro-scale society, and its air quality can easily be disregarded. In the present investigation, the objectives were to provide an inclusive appraisal of indoor formaldehyde and other carbonyls and to raise the public awareness of the occupational and residential air quality, especially in rapid development of the economies of northwestern Chinese cities.

## EXPERIMENTAL SECTION

### *Sampling Sites*

The monitoring was conducted at an university in Xi'an, Shaanxi, China. There were approximately 5,600 of full-time professors, lecturers, and academic and supporting staffs in the university. The total undergraduate and postgraduate enrollments were ca. 3,000. Eight representative locations on the campus were chosen for the carbonyls measurement and exposure assessment. The six workplaces include (W1) a non-smoking academic office, (W2) a smoking academic office, (W3) a dining room in the student canteen, (W4) a photocopy center, (W5) an underground supermarket and (W6) a lecture room. Two residential assessments were carried out at (R1) a bedroom in a staff apartment and (R2) a student dormitory. A one-year comparison study was conducted at Site R1 by collecting airs in the room once the decoration had been finished and when the site had been occupied for one year, respectively. General descriptions of the sampling locations are listed in Table 1. The information of potential pollution sources and characteristics of each workplace and residential unit were achieved through on-site inspection and self-administered questionnaires. Two mechanical ventilations of exhaust fan or air conditioning were equipped and operated as general practices. No fresh air was supplemented with the exhaust fans. The air-conditioning units, if present, re-circulated the indoor air supplemented with a 10% outdoor fresh air supply.

### *Collection of Samples*

Three-hour integrated air samples were collected onto Waters Sep-Pak acidified 2,4-dinitrophenylhydrazine (DNPH)-silica cartridges (Milford, MA) using an carbonyl sampler at a flow rate of 0.85 L $min^{-1}$. No any breakthrough was encountered under such sampling parameters (U.S. EPA, 1999; Herrington *et al.*, 2007; Waters, 2007). The inlet of sampler was set at 1.5 m above the ground level. Four visits were attempted to each sampling site and four samples were collected during each visit in the years of 2010 and 2011. For Site W1-W6, sampling was conducted within the normal working hours (between 08:00 and 20:00). The samples were taken during the active period of the occupants (i.e., evening) at Site R1 and R2. Three baseline samples were collected at each sampling site during the off-duty or non-activated period as well. The sampler was calibrated in the field prior to air collection and its flow rate was further checked at the end of each sampling using a Gilibrator Calibrator (W. Caldwell, NJ). A Whatman Teflon filter assembly (Clifton, NJ) and a Waters Sep-Pak ozone scrubber were installed in front of the cartridge to remove any air particle and prevent oxidization from ozone effect, respectively (Spaulding *et al.*, 1999). The collection efficiency and recovery of carbonyls would not be impacted by the ozone scrubber (Ho and Yu, 2002; Ho *et al.*, 2013a). The sampling reproducibility (> 95%) was assessed by collection of collocated field samples. One field blank was collected on each sampling trip and the results were corrected for the average of the blanks. All of the samples were sealed in Al foil protection bag and refrigerated at a temperature below 4°C once the sampling completed, properly transported, and analyzed within 14 days. Meteorological parameters (i.e., relative humidity (RH) and temperature (T)) were recorded.

### *Chemical Analysis*

Each sampled cartridge was eluted with HPLC grade acetone-free acetonitrile in a 2.0 mL volumetric flask. No any detectable leftover (i.e., DNPH and carbonyl DNP-

**Table 1.** Descriptions of workplaces and residential units on the university campus.

| | Site# | Description | Area ($m^3$) | Floor | Year of Last Decoration | Facilities | Temp[a] (°C) | RH[a] (%) | Ventilation System Equipped |
|---|---|---|---|---|---|---|---|---|---|
| Workplace | W1 | non-smoking academic office | 40 | 4 | 2006 | wooden table and bookshelves; plastic chair; desktop computer | 22.2 | 76.1 | mechanical (air conditioning) |
| | W2 | smoking academic office | 40 | 4 | 2006 | wooden table and bookshelves; plastic chair; desktop computer | 21.7 | 70.1 | mechanical (air conditioning) |
| | W3 | dining room in student canteen | 120 | 1 | 2002 | wooden tables; plastic chairs; stainless steel cupboards | 14.3 | 66.7 | Mechanical (fan) |
| | W4 | photocopy center | 56 | 1 | 2003 | wooden tables; photocopiers | 10.1 | 85.7 | Mechanical (fan) |
| | W5 | underground supermarket | 280 | −1 | 1999 | plastic shelves; refrigerators | 11.9 | 77.8 | mechanical (air conditioning) |
| | W6 | lecture room | 63 | 1 | 2002 | wooden tables and chairs | 13.5 | 77.8 | mechanical (air conditioning) |
| Residential Unit | R1 | bedroom in staff apartment | 16 | 11 | 2010 | wooden bed and wardrobe | 31.3 | 58.4 | mechanical (air conditioning) |
| | R2 | student dormitory | 32 | 4 | 2009 | wooden wardrobes, beds and tables; plastic chairs | 20.3 | 72.5 | mechanical (air conditioning) |

[a] Average values have been reported.

hydrazones) was determined after the elution (Ho *et al.*, 2007). Twenty micro-liter of the extract was injected into an Agilent 1200 high performance liquid chromatography (HPLC) system coupled with a photodiode array detector (DAD) (Santa Clara, CA). The analytes were separated with a PerkinElmer Spheri-5 ODS 5 µm C-18 reversed-phase column (4.6 × 250 mm) (Norwalk, CT) at room temperature. The mobile phase consisted of three components: Component I, 60:30:10 (v/v) of water/acetonitrile/tetrahydrofuran; Component II 40:60 (v/v) of water/acetonitrile; Component III: 100% acetonitrile. The elution program initially started with 80% I/20% II for 1 minute, followed by three linear gradients including 50% I/50% II in 8 minutes, 100% II in 10 minutes and 100% III in 6 min, and finally maintained at 100% III for 5 minutes. Throughout the elution, the flow rate was kept at 2.0 mL $min^{-1}$. Absorbance at 360 nm was used for quantification of aliphatic carbonyls while absorbance at 390 nm was applied for aromatic species. A four-point calibration over a concentration range of 0.015–3.0 mg $mL^{-1}$ for each target carbonyl from the certified standards (Supelco, Bellefonte, PA) was established, and the correlation coefficients ($r^2$) for linear regressions of the calibration curves were at least 0.999. Sixteen airborne carbonyls were quantified in this study (Table 2). No data was reported for the two unsaturated carbonyls (i.e., acrolein and crotonaldehyde). The unsaturated carbonyl DNP-hydrazones would further react with excess DNPH to form adducts, which cause multiple influences on chromatographic separations and inaccuracy in both calibrations and quantifications (Schulte-Ladbeck *et al.*, 2001; Ho *et al.*, 2011). The target carbonyls were identified and quantified according to their retention times and peak areas of the corresponding calibration standards. The limit of detection (LOD) of the quantified carbonyls were in a range of 0.0045 to 0.0098 µg $mL^{-1}$, which can be converted to 0.059–0.13 µg $m^{-3}$ with a sampling volume of 0.153 $m^3$. The method precision was found to be 0.5–3.2% with duplicate analyses.

### *Carcinogenic Risks Calculation*

In this study, inhalation is the main exposure route of interest. Carcinogenic risks were calculated for chronic exposure to carbonyls in the workplaces and residential units. Such estimation with a cancer endpoint is expressed in terms of the probability of rising cancer from continuous exposure to the carbonyl compound in a lifetime. Exposure duration and frequency, body weight, and lifetime of the receptor are critical parameters for computation of chronic daily intake (CDI) for a carcinogen (U.S. EPA, 1989), which is calculated by the equation of:

$$CDI = \frac{Ca \times IR \times ET \times EF \times ED}{BW \times AT \times 365} \quad (1)$$

where Ca is the concentration of carcinogenic substance (mg $m^{-3}$), IR is the rate of inhalation ($m^3$ $hour^{-1}$), ET is the time of exposure (hour $day^{-1}$), EF is the frequency of exposure (day $year^{-1}$), ED is the duration of exposure (y), BW is the average body weight of receptor's (kg), and AT

**Table 2.** Statistical data on exposure frequency and duration and sampling location and collection periods.

| | Class of Indoor Places | Site# | People Involved | Number of People | Number of Hour (h) Accessed per Day | Number of Day(d) Accessed per Week | Sampling Point | Sample Collection Period |
|---|---|---|---|---|---|---|---|---|
| Workplace | non-smoking academic office | W1 | academic staffs | ~2,500 | 8 | 5 | center of office | office hours (09:00–17:00) |
| | smoking academic office | W2 | academic staffs | ~500 | 8 | 5 | center of office | office hours (09:00–17:00) |
| | student canteen | W3 | workers | ~60 | 8 | 5 | center of student canteen | lunch hours (10:30¯13:30), dinner hours (17:00¯20:00) |
| | | | students | ~3,000 | 2 | 5 | | |
| | photocopy center | W4 | workers | ~10 | 8 | 5 | center of photocopy center | working hours (09:00–17:00) |
| | | | customers | ~3,000 | < 0.5 | 1 | | |
| | underground supermarket | W5 | workers | ~20 | 8 | 5 | beside cashier counter | operation hours (08:00–17:00) |
| | | | customers | ~500 | < 0.5 | 2 | | |
| | lecture room | W6 | teachers | ~2,700 | 8 | 5 | back of lecture theater | lecture period (08:00–18:00) |
| | | | students | ~3,000 | 8 | 5 | | |
| Residential Unit | staff apartment | R1 | academic staffs and relatives | ~3,800 | 12 | 7 | center of living room | rest period (19:00¯23:00) |
| | student dormitory | R2 | students | ~3,000 | 12 | 7 | center of bedroom | rest period (19:00¯23:00) |

is the average lifetime (years). The standard BWs and ATs for adults and children have been defined by the United States Environmental Protection Agency (U.S. EPA) (U.S. EPA, 1997a, b). In further, based on the CDI values, the lifetime cancer hazard risk (R) is thus estimated as (U.S. EPA, 1989, 1997a):

$$R = CDI \times PF \quad (2)$$

where PF is the cancer potency factor for a particular carcinogen (kg day$^{-1}$ mg$^{-1}$), which are available from the open access in Integrated Risk Information System (IRIS) (U.S. EPA, 2012).

## RESULTS AND DISCUSSION

### *Exposure Assessment Strategy*

Six workplace and two residential sampling sites were selected based on their representativeness to the campus. Our self-administered questionnaires show that the staffs and students totally spend an average of more than 90% of their times indoor daily. Statistical data on exposure frequency and duration of different groups of people involved are summarized in Table 2. All samples were collected at a fixed point if available and mostly in the center of the rooms, except for the lecture room (W6) where the sampler was installed at the back of the theater. Therefore, potential variations and uncertainties to the airborne levels may include different sample location of the sampling site. In order to ensure the representativeness of the data set, the sampling events were conducted during normal working hours or human-active periods.

### *Carbonyls in Workplace*

Ventilation systems, sizes of sampling locations, and potential indoor pollution sources can greatly vary the carbonyl levels in the workplaces. Table 3 summarizes the carbonyls concentration measured in the sampled workplaces. Extra activity that could produce additional pollutants was not allowed in each site during the sampling period. No variation was found in either absolute values or molar composition based on our preliminary diurnal and seasonal sample sets. The carbonyls concentrations were statistically equivalent demonstrated by Student's *t*-test, with a 95% confidence level for the airs taken at the same sampling site in the different periods. The mean was thus calculated to show individual carbonyls that occurred in the workplaces. Molar composition profiles for the targeted carbonyls are graphed in Fig. 1. Few carbonyls (i.e., *o*- and *p*-and tolualdehyde and 2,5-dimethylbenzaldehyde) were below LOD in > 98% of the valid samples.

Because of the uniqueness of each sampling site, the absolute concentrations should not directly be compared among the indoor workplaces. Some carbonyls could impose high risk in health even at a trace level, so the levels of carbonyls do not necessarily reflect the potential of health hazard. The CDI and R are thus more indispensable, and these risks are discussed in the following sections. Nonetheless, the molar composition profiles allow us to illustrate and apportion the potential indoor pollution origins at selected sites.

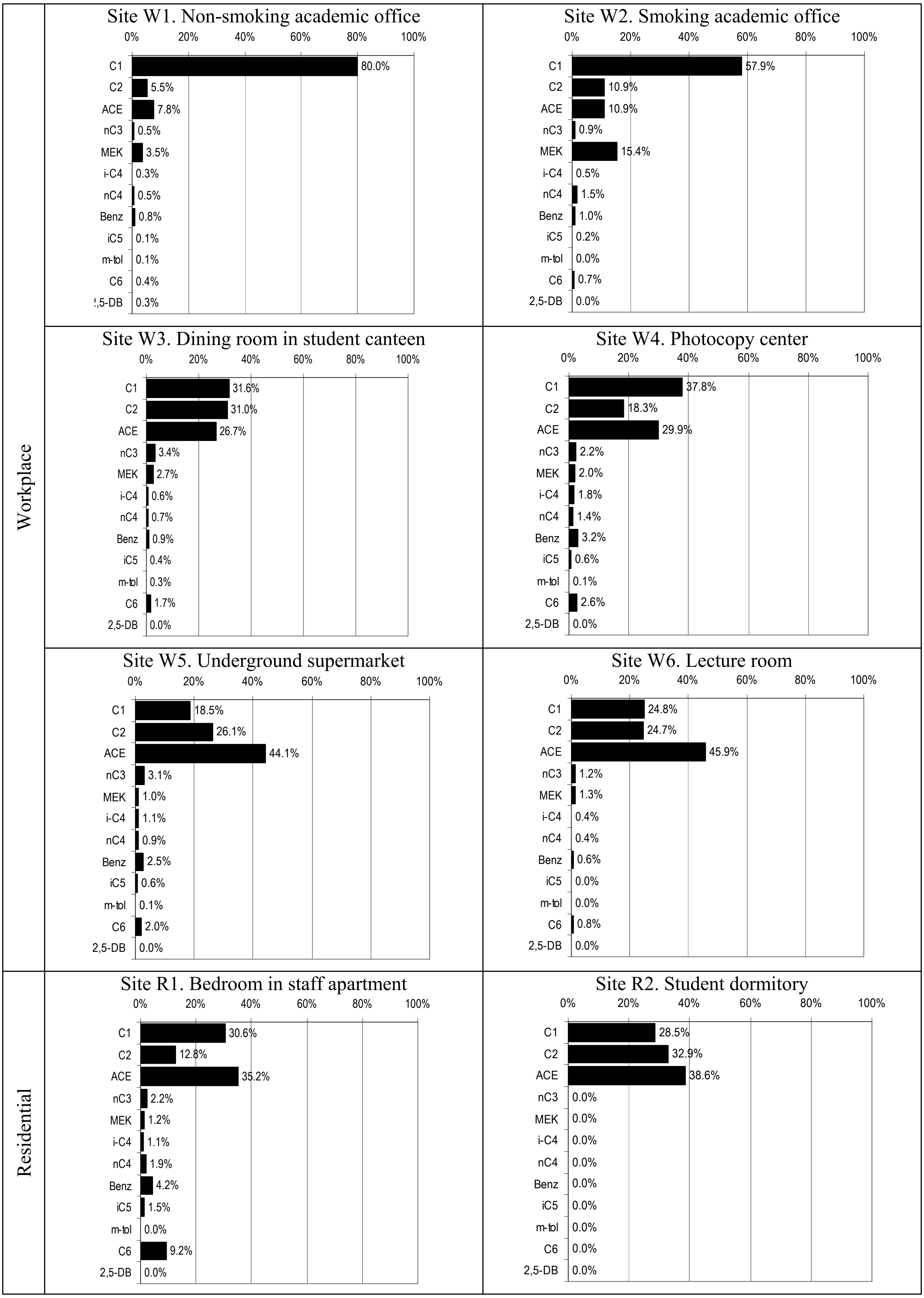
In Fig. 1, the composition profiles of carbonyls demonstrate the difference of ETS contribution in site W1 and W2 with very similar settings. Both W1 and W2 were similar in size and had been completely refurnished six years ago. Both sites were filled with the same furniture, equipment, and ventilation (i.e., windows amount and air-conditioning), except tobacco smoking was permitted at Site W2. The office windows and door were kept closed during the sampling period. No any neither mobile nor stationary indoor pollution sources were identified in these two offices. The contributions of acetaldehyde and methyl ethyl ketone (MEK) at Site W2 exceeded the values at Site W1 by an average factor of 1.9 and 4.4, respectively. These values are in agreement with our previous environmental chamber study, where these two carbonyls are the organic markers for ETS (Wang *et al.*, 2012). Furthermore, compared with other non-smoking workplaces, the highest contribution of MEK was observed in the Site W2 (15.4%). A strong association is linked between the indoor carbonyls and ETS (Loefroth *et al.*, 1989; Katsoyiannis, 2006).

In this study, different molar composition profiles were found for the academic offices (Site W1 and W2) and the public workplaces (Site W3–W6). Formaldehyde was dominant carbonyl at Site W1 and Site W2, which accounted for 80.0% and 57.9%, respectively, of the total quantified carbonyls. However, at all the public sites, the contributions of acetone and acetaldehyde were very similar or higher than the contribution of formaldehyde. The discrepancies of carbonyl distributions could be caused by the ventilation, air circulation, and indoor pollution sources. In the dining room of the student canteen (Site W3), formaldehyde and acetaldehyde are the two largest molar contributors. Cooking emissions from the semi-open kitchen may influence the air quality in the dining area. Our profiles were in agreement with the research results that large amounts of carbonyls (such as formaldehyde, acrolein, and acetaldehyde) can be emitted from combustion of fuels and commercial cooking (Zhang and Smith, 1999; Schauer *et al.*, 2001; Ho *et al.*, 2006). No unique carbonyl profile was observed in the photocopy center (Site W4). We found that the formaldehyde and acetaldehyde levels in normal working hours were 46–58% and 49–71%, respectively, higher than those in the off-duty or non-operational periods. Office technologies including electronic copiers and printers can produce ozone by the reactions of high energy electromagnetic radiation and electrical discharge from oxygen, consequently leading to the volatile organic compounds (VOCs) formations (Yu and Crump, 1998; Dales *et al.*, 2008). Besides, no any unique pattern was observed on the carbonyl composition profiles for the underground supermarket (Site W5) and lecture room (Site W6). Other than normal daily usage of cleaning products such as floor cleaners and furniture detergents, no specific pollution sources were found in these workplaces. Customers, academic staff, workers and students frequently and continuously use the public areas on the campus. Their entrance, stopover, and exiting can impose variability on the carbonyl levels and profiles but their influence would be minimal.

**Table 3.** Carbonyl concentrations (μg $m^{-3}$) in the indoor campus environments.

| | Workplace | | | | | | Residential Unit | | |
|---|---|---|---|---|---|---|---|---|---|
| | Private | | Public and Open | | | | | | |
| | Site W1 | Site W2 | Site W3 | Site W4 | Site W5 | Site W6 | Site R1 | | Site R2 |
| | | | | | | | Bedroom in Staff Apartment | | |
| | Non-smoking Academic Office | Smoking Academic Office | Dining Room in Student Canteen | Photocopy Center | Under-ground Super-market | Lecture Room | New Decorated | Occupied for a Year | Student Dormitory |
| Number of Samples | 16 | 16 | 16 | 16 | 16 | 16 | 20 | 20 | 16 |
| Carbonyls | | | | | | | | | |
| formaldehyde | 72.2 ± 31.0 | 55.8 ± 24.1 | 8.01 ± 0.87 | 5.18 ± 1.51 | 1.75 ± 0.67 | 3.02 ± 1.69 | 86.6 ± 8.71 | 38.3 ± 12.2 | 0.92 ± 0.45 |
| acetaldehyde | 7.38 ± 3.7 | 14.5 ± 4.51 | 11.5 ± 0.27 | 4.08 ± 0.88 | 3.94 ± 1.66 | 4.41 ± 1.78 | 52.9 ± 4.22 | 32.0 ± 7.62 | 1.58 ± 0.76 |
| acetone | 13.7 ± 1.33 | 20.1 ± 3.65 | 13.1 ± 2.43 | 8.36 ± 1.24 | 7.93 ± 2.33 | 10.9 ± 1.22 | 191.9 ± 13.2 | 94.0 ± 8.67 | 2.43 ± 0.78 |
| propanal | 0.95 ± 0.51 | 1.65 ± 0.66 | 1.65 ± 0.11 | 0.62 ± 0.25 | 0.57 ± 0.21 | 0.28 ± 0.16 | 12.1 ± 1.33 | 9.50 ± 1.81 | bd |
| methyl ethyl ketone | 9.76 ± 7.14 | 38.6 ± 27.6 | 1.63 ± 0.13 | 0.69 ± 0.27 | 0.28 ± 0.31 | 0.38 ± 0.21 | 8.1 ± 0.61 | 6.40 ± 0.64 | bd |
| iso-+n-butyraldehyde | 0.63 ± 0.28 | 0.98 ± 0.18 | 0.38 ± 0.15 | 0.58 ± 0.19 | 0.25 ± 0.11 | 0.18 ± 0.11 | 7.4 ± 1.12 | 7.10 ± 1.11 | bd |
| benzaldehyde | 1.80 ± 1.01 | 5.82 ± 4.27 | 0.64 ± 0.14 | 0.77 ± 0.55 | 0.30 ± 0.12 | 0.28 ± 0.17 | 19.6 ± 5.23 | 9.30 ± 2.31 | bd |
| iso-valeraldehyde | 2.13 ± 0.87 | 2.59 ± 0.78 | 0.63 ± 0.22 | 1.35 ± 0.87 | 0.65 ± 0.14 | 0.30 ± 0.21 | 34.3 ± 3.87 | 12.2 ± 2.23 | bd |
| n-valeraldehyde | 0.37 ± 0.17 | 0.43 ± 0.16 | 0.31 ± 0.21 | 0.24 ± 0.05 | 0.16 ± 0.12 | bd[a] | 11.9 ± 1.67 | 8.90 ± 2.68 | bd |
| m-tolualdehyde | 0.29 ± 0.08 | 0.48 ± 0.09 | 0.50 ± 0.71 | 0.16 ± 0.04 | 0.24 ± 0.08 | bd | 1.4 ± 1.08 | 0.10 ± 0.08 | bd |
| hexanal | 1.65 ± 0.67 | 2.34 ± 1.46 | 1.44 ± 0.81 | 1.13 ± 0.98 | 0.60 ± 0.13 | 0.48 ± 0.14 | 87.0 ± 14.3 | 41.8 ± 10.4 | bd |
| total carbonyls | 110.8 ± 46.5 | 143.4 ± 60.2 | 39.8 ± 3.44 | 23.2 ± 3.65 | 16.7 ± 2.66 | 20.2 ± 2.51 | 513.2 ± 39.5 | 259.6 ± 43.5 | 4.93 ± 1.34 |

[a] bd represents below the method detection limit.




* Compound names and symbols are shown in Table 2.

**Fig. 1.** Carbonyl molar composition profiles in indoor workplaces and residential units.

***Carbonyls in Residential Unit***

The carbonyls were quantified in the air samples collected in the same bedroom in the staff apartment (Site R1) after the interior decoration had just been completed and after the apartment had been occupied by two residents for one year (see Table 3). Higher carbonyl levels collected in the newly decorated room were invariably shown with acetone (191.9 ± 13.2 µg $m^{-3}$) representing the most dominant carbonyl compound, followed by hexanal (87.0 ± 14.3 µg $m^{-3}$) and formaldehyde (86.6 ± 8.7 µg $m^{-3}$). The three compounds accounted for 35.2%, 9.2% and 30.6%, respectively, of the total quantified carbonyls expressed in molar ratio. The wardrobe, bed, and floorboards inside the bedroom were primarily made of solid wood. Indoor carbonyls could be released from lacquer coatings and decorating-, refurbishing- and pressed-wood materials (Brown, 1999; Kelly *et al.*, 1999; Brown, 2002). Acetone is widely utilized as lacquer for furniture finishes, potentially contributing to the high level of acetone in the newly decorated room; on the other hand, hexanal is not a commonly and frequently dominant airborne organic compound in the micro-environments and ambient airs. The high hexanal level can be attributed to dry wood emissions from the degradation process (Svedberg *et al.*, 2004). The presence of hexanal can be used an indicator for rancidity. Therefore, the emission of hexanal from the freshly made wooden furniture is undoubtedly particular and causes indoor pollution intensely.

In contrast, lower carbonyl concentrations were found after the apartment had been occupied for one year. The concentrations of acetone, hexanal and formaldehyde decreased at a percentage of 51.0%, 52.0%, and 55.8%, respectively. Such declines were also observed for other carbonyl compounds. No any electronic appliance (e.g., radiator and heaters) or pollutant-generating human activities (e.g., smoking) in the bedroom. The off-gas rates from the apartment layout and related furniture materials plausibly were reduced during that time. There were many factors that can cause large variations in the abundances of carbonyls such as people who brought in or dispersed the pollutants, occasional airing through opening windows, and air circulation with the living room. These factors can potentially lead an increase or a decrease in the carbonyl concentrations but did not affect the molar compositions, as evidenced by the stable profiles (± 3.2%) (Fig. 2).

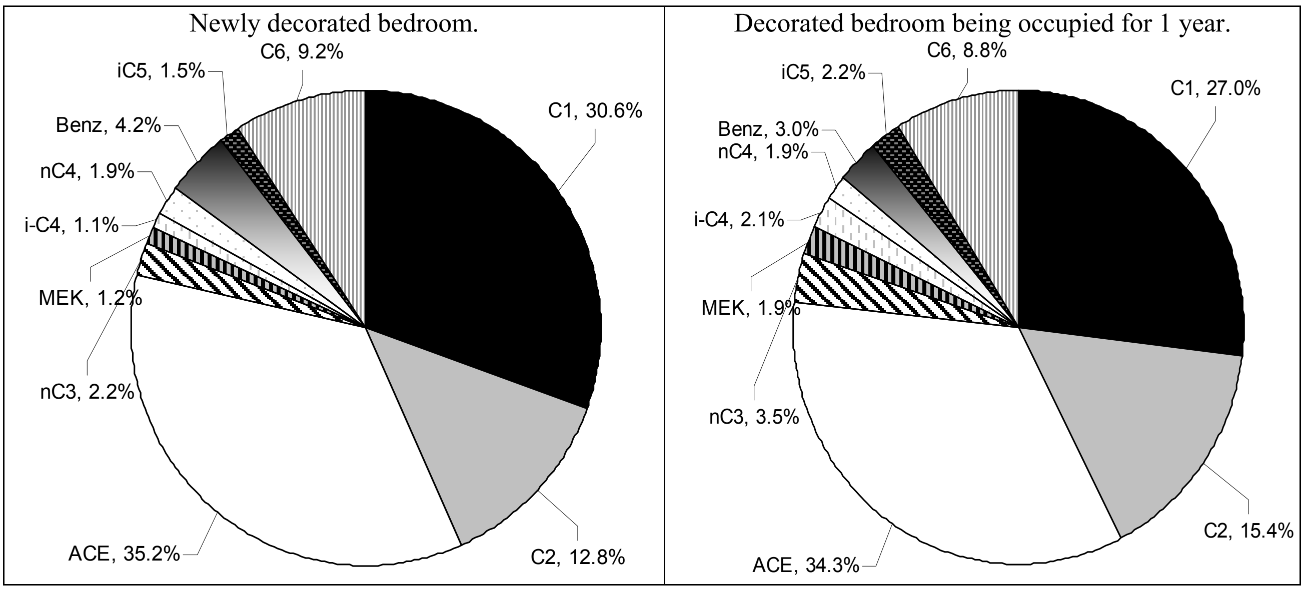
In the student dormitory (Site R2), only three carbonyl compounds (formaldehyde, acetaldehyde, and acetone) were detectable. Similar to other indoor sites, smoking was prohibited, and no any particular pollution source was identified indoor. The dormitory had been furnished over ten years prior to the sampling effort, and the emissions from the aged furniture were expectedly limited.

***RH Influences***

The off-gassing of airborne carbonyls from any materials can be greatly controlled by RH. Even though the air-conditioning was being operated during the sampling period, the RH is > 70% on average in the sampling locations (except Site R1). Few studies demonstrate that the emission rates of gases (e.g., VOCs) can be promoted at humid environments (Kuang *et al.*, 2009; Nnadili *et al.*, 2011). Therefore, the off-gassing of carbonyls is likely to be facilitated under high RH.

***Comparison with Occupational Guideline***

Levels of the three most abundant carbonyls, including formaldehyde, acetone, and acetaldehyde, in the workplaces were compared with international occupational guidelines. Among the target carbonyls, formaldehyde had the highest concentrations in all sampling sites. Its average concentrations in the academic offices (Site W1 and W2) exceeded the recommended exposure limit (REL) of 20 µg $m^{-3}$ (16.3 ppbv) for an 8- or 10-hour time-weighted average (TWA) exposure and/or ceiling, which is defined by the National Institute of




* Compound names and symbols are shown in Table 2.

**Fig. 2.** Comparison of carbonyl molar compositions in the decorated bedroom in the staff apartment (Site R1).

Occupational Safety and Health (NIOSH). These academic offices consisted of wooden furniture and possible carbonyl emission sources including ETS. Their contributions to formaldehyde should not be thus underestimated.

Acetone was the second abundant carbonyls in the sampled workplaces. Even though acetone is not categorized as a mutagen, carcinogen, or chronic neurotoxic to human, it is still recognized as an organic having low acute and chronic toxicity if inhaled and/or ingested that evidenced from extensive medical researches. The acetone concentrations in the workplaces did not exceed any of the threshold limit value (TLV) ceiling of 475 mg $m^{-3}$ (200 ppmv) and the short-term exposure limit (STEL) of 1,188 mg $m^{-3}$ (500 ppmv) for an 8-hour TWA exposure, recommended by the American Conference of Governmental Industrial Hygienists (ACGIH). In addition, the levels of acetone in this study were at least two orders of magnitudes below the general industry's permissible exposure limit (PEL) of 2,400 mg $m^{-3}$ (1000 ppbv) for an 8-hour TWA exposure and 1,800 mg $m^{-3}$ (750 ppmv) for the STEL-15 minutes, guided by the Occupational Safety and Health Administration (OSHA). Both of the consumptions of household detergents and cleaning products, and off-gassing from lacquer layer on the surface of furniture can raise certain degrees of acetone levels in the indoor environments.

Acetaldehyde is another well-known carcinogen which was found to be the next most abundant carbonyl in the workplaces. The concentrations of acetaldehyde were far below the ACGIH's TLV of 45 mg $m^{-3}$ (25 ppmv) and OSHA's general industry PEL for a 8-hour TWA of 360 mg $m^{-3}$ (2000 ppmv). There is a wide range of indoor and outdoor sources for acetaldehyde such as automobile exhausts, incense burning, and cooking emissions. Other carbonyls such as MEK and hexanal had significant contributions to the carbonyl levels in many sampled workplaces. Even though no occupational exposure guideline can be compared, these organics at a high concentration level possibly cause other symptoms to human health such as irritations to respiratory system and eyes.

***Comparison with Residential Guideline***

Residential guidelines were also used to compare with the carbonyls levels measured in the residential units. Our measured formaldehyde concentrations all exceeded the inhalation reference exposure level of 3.0 µg $m^{-3}$ (2.5 ppbv) proposed by the Office of Environmental Health Hazard Assessment (OEHHA), that indicates a substantial risk for the occupants to chronic exposure to this toxic. Fortunately, the concentrations were well below the recommendation indoor level of 100 µg $m^{-3}$ (81.8 ppbv) for a 30-minute average exposure, which is established by the World Health Organization (WHO) (WHO, 2010). Other health concerns of exposure to formaldehyde such as inflammatory, hyperplastic and degenerative changes of the nasal mucosa and irritations to eyes and upper and lower airway should not be underestimated. The acetaldehyde concentrations at both of the residential units surpassed the daily inhalational exposure guideline level of 9 µg $m^{-3}$ (5 ppbv) defined by the U.S. EPA (U.S. EPA, 2012), but were below the WHO's tolerable concentration of 2000 µg $m^{-3}$ (1.11 ppmv) for a 24-hour average exposure, demonstrating that there is still a potential risk of deleterious non-cancer influences on human health.

***Cancer Hazard Risks***

Inhalation exposure is highly related to the receptor's living pattern, type of activity and exposure duration and frequency. These parameters are critical in the estimation of CDI and R. Due to the carcinogenicity classified by U.S.EPA (U.S.EPA, 2012), cancer hazard risk potentials associated with the high abundant formaldehyde and acetaldehyde were estimated to the exposure to the workplaces and residential units in this study. It is worth noting that there are many assumptions recommended by U.S.EPA regarding to the carcinogenic assessment. The inhalation volume (in unit of $m^3$ $hour^{-1}$) of a male and female light duty worker is 0.8 and 0.5, respectively, and of a male and female moderate duty worker is 2.5 and 1.6, respectively. According to our inspection and questionnaires collected, the works for academic and supporting staffs in indoor workplaces are classified as light duty job, while workers in the student canteen, photocopy center and underground supermarket are categorized as moderate duty. In general, all employers are assumed to work eight hours per day and five days per week in a 40-year working period. For the risk assessment in residential units, the inhalation volume (in unit of $m^3$ $hour^{-1}$) of a male and female occupant is 0.7 and 0.3, respectively, at a rest mode. A 24-hour exposure and seven days per week are assumed for residential living. The absorption factor for both workers and occupants is estimated at a level of 90% (U.S. EPA, 1985). An average BW (kg) for a male and female is 70 and 60, respectively, and an average lifetime (years) for a male and female is 69 and 72, respectively (U.S. EPA, 1994). According to the record in IRIS (U.S.EPA, 2012), PF (in unit of kg $day^{-1}$ $mg^{-1}$) for formaldehyde and acetaldehyde are 0.045 and 0.0077, respectively Table 4 lists the estimated CDI and R associated with formaldehyde and acetaldehyde in the workplaces and residential units. For formaldehyde, the R values were in a range of $4.49 \times 10^{-6}$ and $4.22 \times 10^{-4}$, and $2.15 \times 10^{-6}$ to $2.02 \times 10^{-4}$, for males and females, respectively. The risks for acetaldehyde ranged from $1.30 \times 10^{-6}$ to $4.36 \times 10^{-5}$ and $6.23 \times 10^{-7}$ to $2.09 \times 10^{-5}$ for males and females, respectively. In typical, a R value of $< 1 \times 10^{-6}$ represents below the "concern level" while a R value of $> 1 \times 10^{-4}$, as an indication for the "alarm level", implies an urgent necessity to take proper action in protection of human health (Lee *et al.*, 2006). Obviously, higher cancer risks to the workers were seen for formaldehyde than those of acetaldehyde. In the workplaces, the R values associated with formaldehyde were all above the "concern level" and even exceeded the "alarm level" in the academic office. For acetaldehyde, the R values were well below "concern level" in all of the sampling locations. Our results prove that exposure to formaldehyde in the workplace is a realistic safety and occupational health concern. For residential units, high R values were also determined at the bedroom in the staff apartment either newly decorated or having been occupied for one year, indicating that the refurbishing materials and wooden furniture could

**Table 4.** Calculations of chronic daily intake (CDI) and lifetime cancer hazard risk (R).

| Description | Duty Mode | IR ($m^3$ $h^{-1}$) | | ET (h $week^{-1}$) | EF (week $y^{-1}$) | ED (yr) | formaldehyde | | | | | acetaldehyde | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | Conc. (μg $m^{-3}$) | male | | female | | Conc. (μg $m^{-3}$) | male | | female | |
| | | male | female | | | | | CDI[a] (× $10^{-4}$) | R[b] (× $10^{-4}$) | CDI (× $10^{-4}$) | R (× $10^{-4}$) | | CDI (× $10^{-4}$) | R (× $10^{-4}$) | CDI (× $10^{-4}$) | R (× $10^{-4}$) |
| *Workplace* | | | | | | | | | | | | | | | | |
| Non-smoking academic office | light | 0.8 | 0.5 | 40 | 52 | 40 | 72.2 | 24.5 | 1.12 | 17.1 | 0.78 | 7.38 | 2.51 | 0.0193 | 1.75 | 0.0135 |
| Smoking academic office | light | 0.8 | 0.5 | 40 | 52 | 40 | 55.8 | 19 | 0.862 | 13.2 | 0.603 | 14.5 | 4.94 | 0.038 | 3.45 | 0.0266 |
| Dining room in student canteen | moderate | 2.5 | 1.6 | 40 | 52 | 40 | 8.01 | 8.5 | 0.387 | 6.09 | 0.277 | 11.5 | 12.2 | 0.0941 | 8.75 | 0.0673 |
| Photocopy center | moderate | 2.5 | 1.6 | 40 | 52 | 40 | 5.18 | 5.5 | 0.25 | 3.93 | 0.179 | 4.08 | 4.33 | 0.0334 | 3.1 | 0.0239 |
| Underground supermarket | moderate | 2.5 | 1.6 | 40 | 52 | 40 | 1.75 | 1.85 | 0.0844 | 1.33 | 0.0604 | 3.94 | 4.18 | 0.0322 | 2.99 | 0.023 |
| Lecture room | light | 0.8 | 0.5 | 40 | 52 | 40 | 3.02 | 1.03 | 0.0466 | 0.716 | 0.0326 | 4.41 | 1.5 | 0.0115 | 1.05 | 0.0081 |
| *Residential Unit* | | | | | | | | | | | | | | | | |
| Bedroom in staff apartment being newly decorated | resting | 0.7 | 0.3 | 144 | 52 | 40 | 86.6 | 92.7 | 4.22 | 44.4 | 2.02 | 52.9 | 56.6 | 0.436 | 27.1 | 0.209 |
| Decorated bedroom in staff apartment being occupied after 1 year | resting | 0.7 | 0.3 | 144 | 52 | 40 | 38.3 | 41 | 1.87 | 19.6 | 0.894 | 32 | 34.3 | 0.264 | 16.4 | 0.126 |
| Student dormitory | resting | 0.7 | 0.3 | 144 | 52 | 40 | 0.92 | 0.986 | 0.0449 | 0.473 | 0.0215 | 1.58 | 1.69 | 0.0163 | 0.809 | 0.0062 |

[a] CDI in unit of mg $kg^{-1}$ $day^{-1}$.

[b] R according to the assumptions of: (i) average body weights for male and female are of 70 kg and 60 kg, respectively; (ii) a lifetime for male and female are 69 years and 72 years, respectively.

**Table 5.** Comparison of lifetime cancer hazard risk (R) with other micro-environments in China.

| Indoor Environments | Sex | Formaldehyde | Acetaldehyde |
|---|---|---|---|
| dwellings affected by cooking activities (Huang *et al.*, 2011) | - | $1.05 \times 10^{-5}$–$1.57 \times 10^{-5}$ | $3.05 \times 10^{-6}$ |
| industrial workplaces | male | $2.01 \times 10^{-5}$–$2.37 \times 10^{-4}$ | $2.41 \times 10^{-6}$–$5.13 \times 10^{-5}$ |
| (Ho *et al.*, 2013b) | female | $2.68 \times 10^{-5}$–$3.16 \times 10^{-4}$ | $3.12 \times 10^{-6}$–$6.84 \times 10^{-5}$ |
| campus in Southern China | male | $0.16 \times 10^{-4}$–$6.37 \times 10^{-4}$ | $0.84 \times 10^{-7}$–$0.83 \times 10^{-4}$ |
| (Ho *et al.*, 2014) | female | $0.11 \times 10^{-4}$–$4.56 \times 10^{-4}$ | $0.59 \times 10^{-7}$–$0.59 \times 10^{-4}$ |
| campus in Northern China | male | $4.49 \times 10^{-6}$–$4.22 \times 10^{-4}$ | $2.15 \times 10^{-6}$–$2.02 \times 10^{-4}$ |
| (this study) | female | $1.30 \times 10^{-6}$–$4.36 \times 10^{-5}$ | $6.23 \times 10^{-7}$–$2.09 \times 10^{-5}$ |

create a significant health impact on occupants. It is critical to point out that the cancer risks for the customers or students who use the indoor micro-environments (e.g., W3–W6) on campus could not be accurately assessed due to their low exposure frequency and short exposure duration. Further short-term personal exposure tests will be conducted in our future studies.

Table 5 compares the R values for formaldehyde and acetaldehyde in various workplaces or residential units in China (Huang *et al.*, 2011, Ho *et al.*, 2013b, 2014). Those values were estimated with the same assumptions suggested by U.S.EPA. The comparison demonstrates that the cancers risks in several indoor setting (e.g., academic offices) were even higher than those of the industrial workplaces and dwellings influenced by cooking activities. This observation further suggests that the airborne and inhalation levels of formaldehyde and acetaldehyde on the campus should not be underestimated.

Owing to a lack of guidelines and risk indices, the chemical hazard potentials or cancer risks for other less abundant carbonyls (e.g., benzaldehyde and tolualdehyde isomers) are unable to be evaluated systematically. Nevertheless, the determination of these trace compounds is still valuable for source apportionment and preservation of data to legislate indoor air quality regulation in Mainland of China.

### *Uncertainties and Limitations*

Even though the cancer risks could reflect the hazard potentials to the carbonyls in the sampled indoor places, both environmental (e.g., climates, ventilations, number and frequencies of visitors) and occupational (e.g., work practices and changes over time) factors would greatly increase their uncertainties in health assessments. Few additional assumptions must be remarked. The risks were estimated under the particular working conditions and climate shown in this study. The numbers and frequencies of visitors and the practices of workers were all consistent. During the sampling events, the ventilation systems were working properly and no sudden or unpredictable pollution source was influenced to the sampled places.

## CONCLUSIONS

The air measurements on the university campus provide a more and clear understanding on the indoor pollution sources and better estimations on the potential health risk to different carbonyls. The results prove that there is a substantial risk for the academic staffs, workers and occupants to chronic exposure to formaldehyde. The carbonyls levels can be raised by the emission from refurbishing materials and wooden furniture, additionally with anthropogenic indoor pollution activities such as cigarette smoking. These pieces of information suggest the significance for conducting more regular and on-site indoor air measurements at different public micro-environments in Mainland of China.

## ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (NSFC-21107084), Xi'an Jiaotong University (No. 0814100), the State Key Laboratory of Loess & Quaternary Geology (SKLLQG0804), the Hong Kong Polytechnic University (G-YX3L and G-YF23), and the Research Grants Council of Hong Kong (RGC 5197/05E, PolyU 5175/09E and BQ01T).

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*Received for review, May 25, 2015*
*Revised, September 25, 2015*
*Accepted, January 11, 2016*