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Research Article

Molecular Characteristics, Sources, and Health Risk Assessment of Gaseous Carbonyl Compounds in Residential Indoor and Outdoor Environments in a Megacity of Northwest China

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Carbonyl compounds (CCs) in indoor air pose a significant threat to residents' health and have garnered considerable attention in recent years. However, most studies have focused on low-molecular-weight carbonyl compounds (LMW-CCs) and have underestimated the impact of high-molecular-weight ones (HMW-CCs), causing a failure to comprehensively understand their effects on health. In this study, we analyzed twenty carbonyls in the indoor and outdoor air at typical residential communities in a megacity in Northwest China by using high-performance liquid chromatography (HPLC) coupled with a photodiode array detector (DAD). The total concentration of indoor carbonyls was 1.4-3.4 times that of outdoor carbonyls. In addition, the concentration of indoor carbonyls was much higher during the heating season than that during the nonheating season. Conversely, the concentration of outdoor carbonyls was higher during the nonheating season than that during the heating season. The principal component analysis (PCA) revealed that indoor carbonyl pollution was primarily influenced by building materials, cooking fume, and wooden furniture. Formaldehyde exposure in indoor environments posed a greater health risk to children than acetaldehyde exposure. HMW-CCs were the primary contributors to indoor odor pollution, which was considered a significant cause of sick building syndrome (SBS). Our findings underscore the crucial role of HMW-CCs in indoor environments in exerting adverse impacts on health.

1. Introduction

Carbonyl compounds (CCs), including aldehydes and ketones, are commonly present in the gaseous phase of indoor environments [1]. Indoor CCs originate from various sources, such as building materials (e.g., linoleum floors, composite floors, wood-based panel materials, plywood products, and wallpaper) [2, 3], wooden furniture [4, 5], household products (e.g., glue, paint, and coating) [6], cigarette smoke [7], cooking smoke [8], air fresheners, cosmetics, and other daily consumer goods [9, 10]. Some indoor CCs are secondary products of volatile organic compounds (VOCs) reacting with ozone [11]. Additionally, certain CCs are emitted directly from automobile exhaust, biomass combustion, fuel combustion, biological sources, or generated by photochemical oxidation of VOCs in the

outdoor atmosphere in urban areas [12–14]. These compounds can enter indoor environments through air infiltration and natural ventilation [15].

Many CCs are toxic and harmful, and some are considered carcinogenic. For example, formaldehyde, one of the most common CCs [16], can induce emphysema, renal failure, and other symptoms when excessively inhaled, leading to deformity and cancer [9, 17]. Acetaldehyde can stimulate various parts of the human body, including brain nerves, facial skin, eyes, and respiratory mucosa [18, 19]. Breathing in moderate levels of acetone for short periods can cause headaches, light-headedness, confusion, and a high pulse rate, while prolonged or repeated skin contact with acetone may produce severe dermatitis [20]. Glyoxal and methylglyoxal can severely affect human health by inducing cellular damage and generating advanced glycation end products [21]. In addition, CCs can cause odor and sensory irritations, which are important aspects of sick building syndrome (SBS) and directly affect residents' perceptions of indoor air quality [22, 23]. Therefore, the presence of CCs in residential indoor environments constitutes a potential hazard to the health of residents.

Numerous studies have investigated the characteristics and sources of indoor CCs pollution and their potential effects on health [1, 24–26]. For instance, Pu et al. [27] identified the primary emission sources of formaldehyde (sofa materials, smoking, and family location), acetaldehyde (smoking and kitchen structure), acetone (kitchen structure and potted plants), and hexanal (potted plants) in residential buildings in Beijing. Huang et al. [28] found that indoor formaldehyde exposure may increase the risk of children suffering from the common cold. However, most of these studies focused on indoor low-molecular-weight (LMW, carbon number <6) CCs, such as formaldehyde, acetaldehyde, and acetone. In fact, high-molecular-weight (HMW, carbon number ≥ 6) CCs (i.e., hexanal, heptanal, octanal, nonanal, and decanal) also contribute significantly to total indoor CCs [29–31] and have significant adverse effects on human health [32, 33]. Lou et al. [22] found that some aldehydes (i.e., nonanal, decanal, and heptanal) were potential oxidation products with ozone, and they were closely associated with people's sensory irritations. Therefore, understanding both LMW-CCs and HMW-CCs in the indoor environment is necessary to mitigate their threats to health.

In this study, we measured indoor and outdoor CCs, including both LMW-CCs and HMW-CCs, in residential buildings in different areas of a megacity (Xi'an) in Northwest China. The study is aimed at (a) determining the levels of carbonyls in the indoor and outdoor environments of the residence in the city, (b) identifying the most relevant pollutant sources of carbonyls, and (c) assessing the potential cancer risks of children exposed to formaldehyde and acetaldehyde and determining olfactory pollution by HMW-CCs in the indoor environment.

2. Methods

2.1. Sample Collection. Carbonyl samples were collected from January to March 2016 (heating season) and September 2016 (nonheating season) at three residential communities in Xi'an (Figure S1): (1) a mixed commercial–residential site in Huifang community with heavy traffic (HS); (2) an urban residential site in Qujiang village (QJ), adjacent to densely populated residential buildings without any factories; and (3) a suburban site in Xiangyang community (XY), with relatively less population and commercial activities. The details of each sampling site can be referred to in Table S1. Carbonyl samples were collected by an acidified 2,4-dinitrophenylbydrazine (DNPH) impregnated cartridge (Sep-Pak DNPH-silica, Waters Corporation, Milford, MA) at a flow rate of 0.6-0.8 L/min. Indoor samples were collected for 8 h (9 : 30-17 : 30) in the living room of each household. Outdoor sampling was carried out on the open balcony of another uninhabited house. In addition, we collected 10 carbonyl samples during the cooking process (18 : 00-19 : 00) in the heating and nonheating seasons in Qujiang village. The air exchange rate (ACH) was monitored by a CO_2 analyzer (LI-820, LI-COR, USA).

2.2. Analytical Method. A total of 20 carbonyls were analyzed by a high-pressure liquid-chromatography system (HPLC, Series 1200; Agilent Technology, Santa Clara, CA) coupled with a photodiode array detector (DAD). The carbonyls included formaldehyde (C1), acetaldehyde (C2), acetone (A3K), propionaldehyde (*n*-C3), 2-butanone (MEK), butyraldehyde (*i,n*-C4), benzaldehyde (Benz), isovaleraldehyde (*i*-C5), *n*-valeraldehyde (*n*-C5), *o*-tolualdehyde (*o*-tol), *m*-tolualdehyde (*m*-tol), *p*-tolualdehyde (*p*-tol), *2,5*-dimethylbenzaldehyde (*2,5*-DB), hexaldehyde (C6), heptanaldehyde (C7), octanaldehyde (C8), nonanaldehyde (C9), decanaldehyde (C10), glyoxal (Gly), and methylglyoxal (mGly), respectively. A more detailed description of the analytical method may be found in research by Dai et al. [8]. Identification and quantification of CCs were based on retention time and peak areas of the corresponding calibration standards, respectively. A strong linear relationship ($R^2 > 0.999$) was found between the concentrations and responses for each target carbonyl. The relative standard deviation for duplicate analysis was lower than 5%. Further information of quality control and quality assurance (QA/QC) can be referred to in research by Lui et al. [12] and Dai et al. [34].

2.3. Data Analysis. Statistical analyses were conducted by using SPSS 26 and Excel 2021. The normality of the data distribution was checked with the Kolmogorov–Smirnov test. Spearman correlation coefficients were calculated to examine the relationships between two kinds of CCs. Statistical significance was determined at $p < 0.05$ for all analyses. Principal component analysis (PCA) with varimax rotation and multiple linear regression analysis were applied to the carbonyl concentration data to identify the sources of CCs. For PCA, components with an eigenvalue greater than one were extracted. Each factor from the PCA was associated with source characterization by its most representative CCs. Prior to statistical analysis, values below the limit of detection were substituted by LOD/2 to estimate means and standard deviations [35]. Multiple linear regression analysis was performed using a stepwise procedure with a

significance level of p set at <0.05 for inclusion variables and >0.10 for exclusion them.

2.4. Children's Inhalation Exposure Estimation and Health Risk Evaluation. Compared with adults, children are more susceptible to the adverse effects of environmental pollutants due to their higher respiratory rate, more vulnerability, and longer exposure time [36, 37]. In this study, we employed the methodology proposed by the U.S. Environmental Protection Agency (EPA) to calculate personal inhalation exposure and assess the health risks of children aged 0-6 years [38]. The formula was as follows:

$$\begin{aligned} \mathrm{E_{ij}} &= C_{ij} \times \mathrm{IR}_j \times t_j, \\ \mathrm{RQ} &= \frac{E_{ij}}{\mathrm{NSRL_{child}}}, \end{aligned} \tag{1}$$

where E_{ij} was the daily inhalation dose of pollutant (i) in the microenvironment (j), μg/day; C_{ij} was the concentration of the pollutant (i) in the microenvironment (j), namely, the measured concentration of carbonyls in the living room, $\mu g/m^3$; IR_j was the inhalation rate in the microenvironment (j), values from the Chinese exposure factors handbook (children) [39], as shown in Table S2 in the Supplementary Data, m^3/h; t_j was the exposure time in the microenvironment (j), namely, children's daily time spent in different rooms, as shown in Table S2 in the Supplementary Data, h/day; RQ was risk quotients. An RQ value of 1.0 or less indicates an insignificant impact on human health, while an RQ value of larger than 1.0 represents that children's inhalation dose exceeds the threshold of 10^{-5} lifetime cancer risk, or in other words, the child is confronted with the potential cancer risk. $\mathrm{NSRL_{child}}$ is child-specific no significant risk levels, and the procedure for calculating child-specific $\mathrm{NSRL_{child}}$ is clarified in the Supplementary Data (S1).

2.5. Odor Analysis. The odor intensity resulting from the presence of a CC component was quantified using odor activity value (OAV), which represents the ratio of the mass concentration of a single chemical compound to its corresponding odor threshold (OT) [23, 40]. The OAV was calculated using the following formula:

$$\mathrm{OAV}_i = \frac{C_i}{\mathrm{OT}}, \tag{2}$$

where OAV_i was the odor activity value of pollutant (i) (dimensionless), C_i was the mass concentration of pollutant (i) ($\mu g/m^3$), and OT was the odor threshold values used for the calculation was obtained from Jiang et al. [23] and Yoshio and Nagata [41].

3. Results and Discussion

3.1. Concentration and Molecular Distribution of Carbonyl Compounds

3.1.1. Outdoor Carbonyl Compounds. The concentrations of total measured CCs, $PM_{2.5}$, and meteorological parameters at various outdoor sites in the heating season were summarized in Table S3 (indoor: HS-1, HS-2, HS-3, HS-4, QJ-1, QJ-2, QJ-3, QJ-4, QJ-5, XY-1, XY-2, XY-3, XY-4, and XY-5, outdoor: HS-O, QJ-O, and XY-O). The outdoor temperature in XY and HS was higher than that in QJ due to the different sampling periods, while the outdoor humidity remained relatively constant across all sampling sites. The total concentrations of outdoor CCs at the three sites were comparable. Figure 1 and Table S4 present the molecular distributions of outdoor CCs at HS, QJ, and XY. Consistent with previous studies [42–48], acetone (A3K), acetaldehyde (C2), and formaldehyde (C1) were the most abundant compounds in CCs at all three outdoor sites. The concentrations of outdoor A3K, C2, and C1 were 9.6-21.2 $\mu g/m^3$, 5.7-12.2 $\mu g/m^3$, 6.0-9.7 $\mu g/m^3$, respectively, and the sum of LMW C1, A3K, and C2 accounted for 37.3-66.3% of the total CCs. Additionally, methylglyoxal (mGly), glyoxal (Gly), and nonanaldehyde (C9) were found to have high loading at outdoor sites. Especially in HS, the concentrations of mGly, Gly, and C9 were 9.7 $\mu g/m^3$, 8.7 $\mu g/m^3$, and 5.4 $\mu g/m^3$, respectively, and their sum of accounted for 40.8% of the total CCs. These results confirmed that HMW-CCs and Di-CCs (Gly and mGly) also contributed substantially to outdoor environments.

The concentration of outdoor acetone increased gradually from the urban center (HS: 9.6 $\mu g/m^3$) to the suburban area (XY: 21.2 $\mu g/m^3$). However, formaldehyde (C1) and acetaldehyde (C2) exhibited higher concentrations at QJ than at HS and XY, as QJ is located in the residential area of Xi'an where traditional heating methods involving coal and/or biomass materials are still prevalent, leading to significant emissions of C1, C2, and other pollutants [49]. This was supported by the significantly higher $PM_{2.5}$ concentration in the atmospheric environment of QJ (225.4 $\mu g/m^3$) compared with HS (111.9 $\mu g/m^3$) and XY (110.8 $\mu g/m^3$) (Table S3). In contrast, XY is situated in the suburbs of Xi'an, with fewer residents and no apparent sources of pollution, suggesting that the high concentration of A3K in XY may be largely influenced by regional transport. The concentrations of Gly (8.7 $\mu g/m^3$) and mGly (9.7 $\mu g/m^3$) in the outdoor environment of HS were higher than those in the outdoor environments of QJ (0.8 $\mu g/m^3$ and 1.0 $\mu g/m^3$) and XY (1.6 $\mu g/m^3$ and 8.0 $\mu g/m^3$). Since HS is the main tourist area in Xi'an with dense human flow and busy traffic, the increase in Gly and mGly in this area may be mainly attributed to vehicle exhaust and/or combustion activities [13, 50].

3.1.2. Indoor Carbonyl Compounds. The average concentrations and relative molecular contributions of indoor CCs at HS, QJ, and XY are shown in Figure 2. The concentrations of CCs in indoor environments at these sites were 1.4-3.4 times those in the corresponding outdoor environments. The indoor CCs concentrations could be ranked in descending order as follows: QJ (186.6 $\mu g/m^3$)>XY (96.2 $\mu g/m^3$)>HS (84.1 $\mu g/m^3$). Formaldehyde (C1), acetaldehyde (C2), acetone (A3K), hexanal (C6), and nonanal (C9) exhibited higher concentrations in QJ than in the other two sites. The houses in QJ were constructed within the past five years

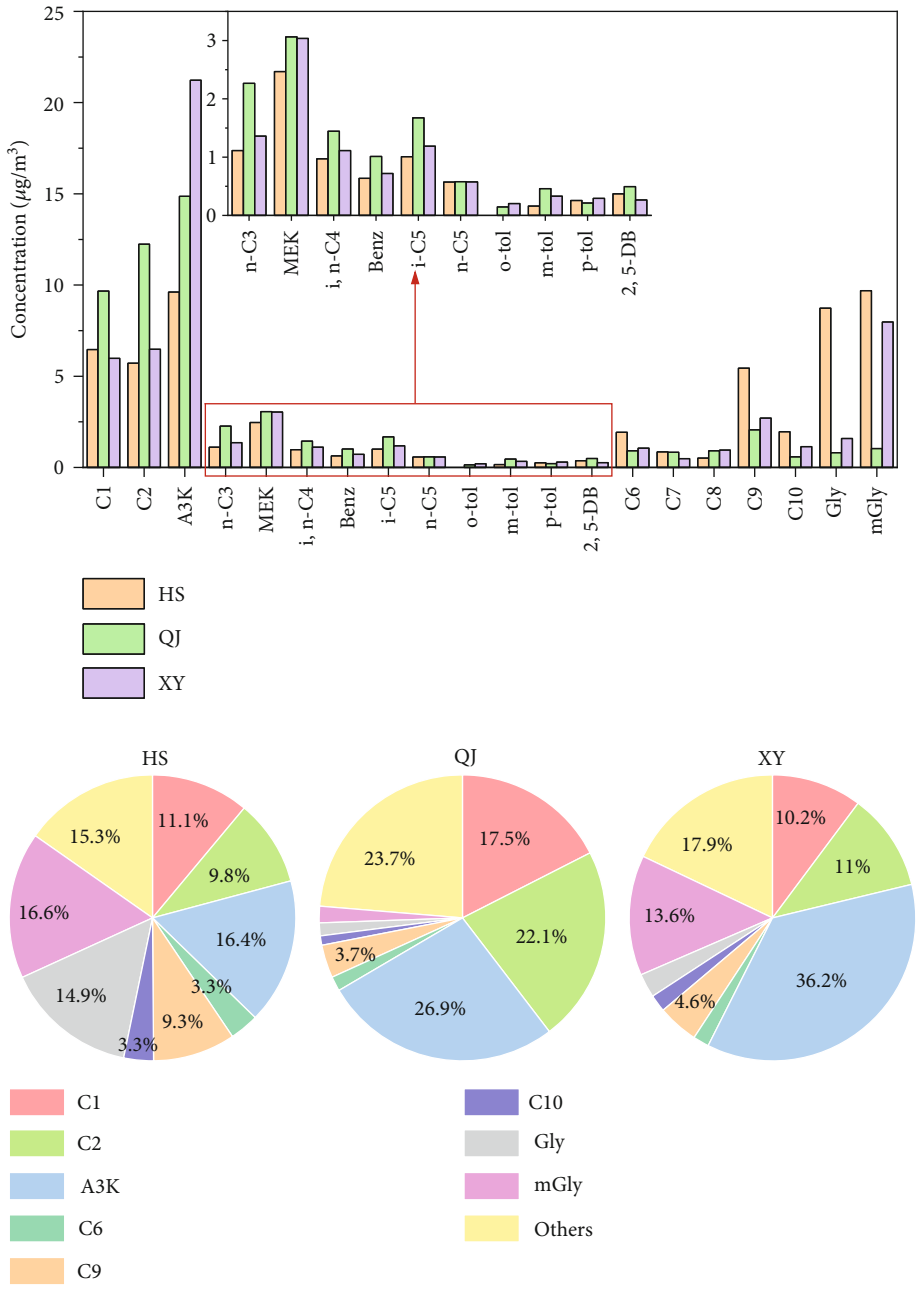

Figure 1: The concentrations and relative molar contributions of outdoor carbonyls at HS, QJ, and XY.

(as shown in Table S1), and thus more indoor pollutants were emitted from building materials and furniture [51, 52].

Regarding Di-CCs, the relatively higher concentration of Gly and mGly in HS could be ascribed to the fact that the houses in HS were self-built buildings with 2-3 stories, which had poor airtightness and were more susceptible to outdoor CCs. In addition, the pollutants from the fuel combustion for heating could not be completely discharged from the house, thereby resulting in indoor CCs pollution [53].

HMW-CCs, especially C6, C9, and C10 in the three residential areas, accounted for 18.3%, 19.5%, and 25.0% of the total CCs, respectively. Within indoor environments, the ozonolysis of fatty acids resulted in the formation of C10, C9, and other aldehydes [54], which were linked to ozone-initiated chemistry on the human skin [54, 55] and interior surface (clothing fabric, painted wall, or carpet) [56]. HMW-CCs might be associated with cooking-related activities [57]. Here, the concentration of HMW-CCs in the indoor environments was significantly higher than that in the outdoor environments, suggesting that these CCs may be emitted by the human body and cooking-related activities. Additionally, the concentration of Di-CCs in the indoor and outdoor environments changed similarly, with HS>XY>QJ, indicating that they may be influenced by outdoor infiltration.

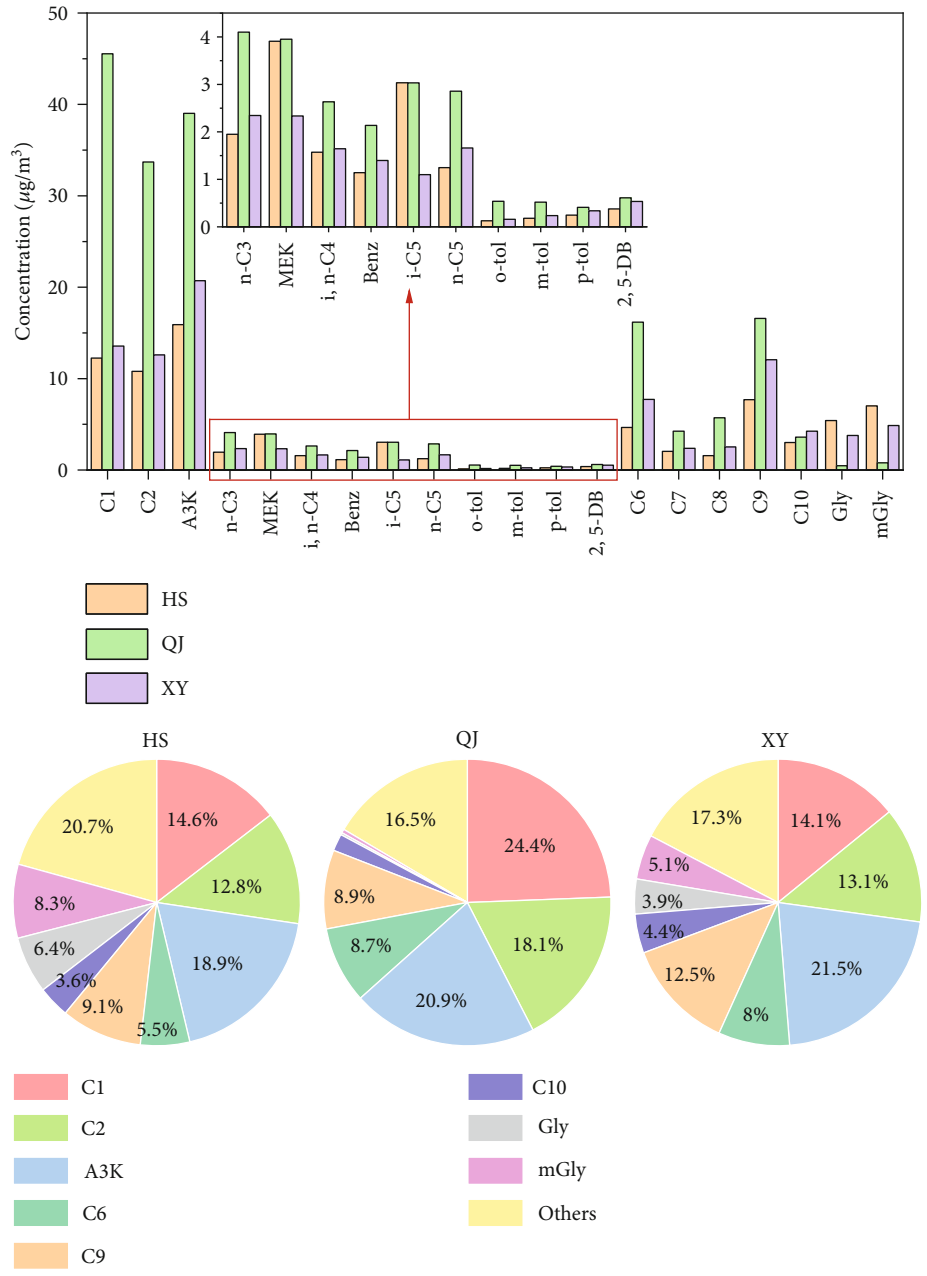

Figure 2: The concentrations and relative molar contributions of indoor carbonyls at HS, QJ, and XY.

3.1.3. Seasonal Variation. The concentrations of the ten most abundant CCs in both indoor and outdoor environments of QJ in heating and nonheating seasons are shown in Figure 3 (indoor: QJ-1, QJ-2, QJ-3, QJ-4, and QJ-5; outdoor: QJ-O). Interestingly, the seasonal variation of indoor and outdoor CCs was the opposite. The concentrations of indoor CCs (147.2-235.1 $\mu g/m^3$) in the heating season were higher than those in the nonheating season (70.9-137.7 $\mu g/m^3$), whereas outdoor CCs showed higher concentrations in the nonheating season ($68.9 \pm 28.2\ \mu g/m^3$) than those in the heating season ($55.3 \pm 9.4\ \mu g/m^3$). The seasonal variation of outdoor CCs was attributed to the relatively higher temperature (21.6 ± 4.3°C in the nonheating season vs. 0.8 ± 2.9°C in the heating season) and stronger solar radiation in the nonheating season, which favored the photochemical reaction of atmospheric organic matter [58]. The impact of plant emissions was also an important factor [59]. For indoor CCs, the concentrations of LMW-CCs, C1, C2, and A3K in the heating season were obviously higher than those in the nonheating season. Especially, the concentration of C2 in the heating season (24.1-48.5 $\mu g/m^3$) was more than twice that in the nonheating season (7.5-12.9 $\mu g/m^3$). Similar seasonal variations of CCs in urban residences were observed in Baotou, another city in northwest China [9]. This was reasonable because the indoor temperature in the heating season and nonheating season in most cities in northwest China was comparable (20-25°C), but the window was less open in the heating season. Thus,

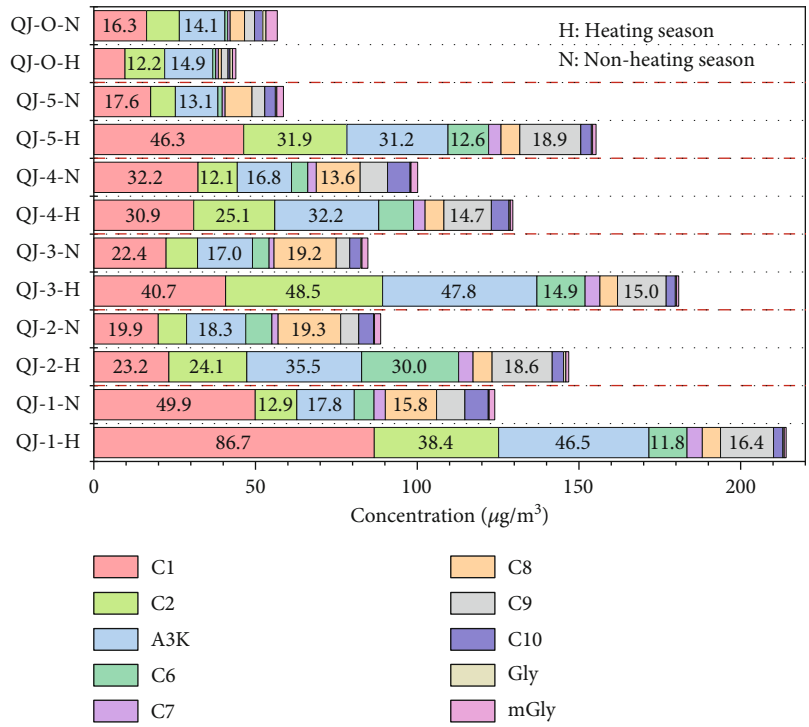

Figure 3: The concentrations of indoor and outdoor carbonyls at QJ in two seasons. The numbers in the figure represent the concentration of the carbonyls.

the excellent airtightness of modern buildings could cause a continuous accumulation of gaseous pollutants in indoor environments.

Previous studies have demonstrated that indoor pollutant concentrations decrease with strong ventilation [9, 25, 60]. To investigate the impact of ventilation on indoor CCs pollution, we employed the CO_2 tracer gas decay method to determine the air exchange rate (ACH) [60]. The detailed procedure for calculating ACH was described in Supplement S2. Our results indicated that the ACH in the nonheating season (2.6-48.6 h^{-1}) was 1-2 orders of magnitude higher than that in the heating season (0.1-0.8 h^{-1}), further highlighting that the low ACH served as a significant contributor to severe indoor CCs pollution in the heating season (Table S5).

3.2. Source Identification

3.2.1. Correlation and PCA. Spearman correlations were calculated for almost all CCs quantified in the indoor environments to determine common sources (Table S6). Except for 2,5-DB, C10, Gly, and mGly, most CCs showed strong correlations with each other, with Spearman coefficients (ρ) between 0.5 and 0.9, indicating that they coexisted in the indoor environments and were emitted from the same sources. C1 presented a strong correlation with C2 ($\rho = 0.89$), A3K ($\rho = 0.83$), C3 ($\rho = 0.86$), and C4 ($\rho = 0.85$), suggesting similar sources for these four CCs. *N*-C5 presented a strong correlation with C6 ($\rho = 0.90$) and C7 ($\rho = 0.91$), suggesting that the three CCs shared similar sources.

Factor analysis was used to quantitatively analyze indoor CCs sources in this study. Factor 1 was characterized by C1, C2, A3K, C3, and C4, which were mainly emitted from building materials and cooking fumes [61–65]. In the indoor environment, pressed wood products using adhesives that contain urea-formaldehyde (UF) resins were likely to be the most significant sources of C1 (available at http://www.epa.gov/iaq/formalde.html). The Spearman coefficients between these CCs ranged from 0.77 ($p < 0.01$) to 0.92 ($p < 0.01$), indicating that they were common indoor sources. Factor 2 presented high loadings with *n*-C5 and C6, indicating the source of wooden furniture [66, 67]. Factor 3 was dominated by Gly and mGly, which were emitted directly to the atmosphere through biomass combustion and then again formed by oxidation of VOCs [14]. Factor 4 was characterized by high levels of Benz, which was used as a special top-flavor and found in some cosmetics and personal care products (available at https://www.vedantu.com/chemistry/benzaldehyde), such as cosmetics, air fresheners, and cleaning agents [68]. A3K and C2 also showed high loadings, indicating that Factor 4 represented emissions from personal care products. Factor 5 was identified as cigarette smoke due to the high contributions of MEK and *i*-C5. MEK showed higher levels in smoking houses and apartments [29]. The twiddle factor loadings and variances of the extracted principal components are shown in Table 1. Building materials and cooking fumes were the most important sources of indoor CCs, contributing to 54.9% of the indoor CCs. The identified release source of wooden furniture also made a substantial contribution (13.6%) to CCs. The rest of the CCs were caused by outdoor infiltration,

Table 1: Component matrix from PCA of indoor CCs.

Compounds	F1 Building materials and cooking fumes	F2 Wooden furniture	F3 Outdoor infiltration	F4 Personal care products	F5 Cigarette smoke
C1	**0.897**	0.048	-0.271	0.167	0.138
C2	**0.695**	0.320	-0.255	**0.443**	0.151
A3K	**0.591**	0.317	-0.218	**0.582**	0.136
C3	**0.738**	0.406	-0.198	0.342	0.165
MEK	0.113	0.030	-0.041	0.225	**0.951**
C4	**0.620**	0.484	-0.174	0.455	0.246
Benz	0.345	0.214	-0.147	**0.860**	0.110
iC5	0.174	0.089	-0.104	-0.035	**0.960**
nC5	0.179	**0.859**	-0.256	0.250	0.169
Tol	0.362	0.477	-0.237	0.553	0.060
DB	0.326	0.727	0.401	0.083	-0.208
C6	0.126	**0.890**	-0.285	0.212	0.109
Gly	-0.261	-0.151	**0.898**	-0.184	-0.113
mGly	-0.272	-0.145	**0.906**	-0.147	-0.041
% variance	54.9	13.6	10.6	7.6	3.8
Accumulative (%)	54.9	68.5	79.1	86.7	90.5

Boldface denotes high factor loadings considered to be marker species.

personal care products, and cigarette smoke, contributing to 10.6%, 7.6%, and 3.8% of the CCs, respectively.

Indoor CCs sources identified in this study were compared with other limited source apportionment studies available worldwide. In the present study, different indoor sources within Chinese households contributed to more than 90% of measured indoor CCs. Guo [69] found that over 76% of indoor VOCs concentrations in households in Hong Kong, China, were attributed to the off-gassing of building materials. The highest source contribution of off-gassing of building materials could be explained by the fact that most furniture in households in Hong Kong was made of pressed wood products with adhesives containing urea-formaldehyde resins [70]. The source apportionment in Edmonton, Canada, which employed PCA, suggested that the emission of household products had the largest contribution (43.2%) to indoor VOCs [71]. In a recent study, Liu et al. [72] reported that more frequent high-emitting activities (such as cooking), and high emissions from building materials and furnishings were the primary source of indoor VOCs in the United States. Overall, the observed concentrations of CCs within residences were influenced by common indoor sources, which had been reported in comparatively similar ways in different studies.

3.2.2. Cooking Contribution. In a study conducted by Liang et al. [73], only two aldehydes (hexanal and nonanal) were detected at different stages of interior decoration in an apartment in Beijing. Another study by Liang et al. [74] revealed that aldehyde levels increased gradually after one year of occupancy, with the type of aldehydes increasing from 2 to 10. This indicated that aldehydes were closely associated with human activities in Chinese households with cooking activities being a significant contributor to indoor air pollution. Ho et al. [75] illustrated that long-chain saturated carbonyls, e.g., heptanal, octanal, and nonanal, accounted for at least 40% of carbonyls in kitchens that frequently used heated cooking oils. Cheng et al. [76] measured the concentrations of CCs in the restaurant, kitchen, and fume exhaust and observed that most CCs (>50%) were removed by the kitchen ventilator. However, these pollutants could still be transported to the dining room and other areas and thus affected indoor air quality. Pei et al. [24] investigated the concentrations of VOCs in unventilated kitchens before and after cooking and found that the concentrations of indoor HMW-CCs (hexanal, nonanal, octanal, pentanal, and heptanal) increased 2-5 times during the cooking period.

The concentration variations of CCs in the living room before and during cooking in QJ are shown in Figure 4. The concentrations of indoor LMW-CCs C1, C2, and A3K increased significantly during the cooking process. The increase of the three LMW-CCs was even more pronounced in the nonheating season, with increments of 73% (QJ-3), 47% (QJ-4), and 143% (QJ-3), respectively. A3K is a moderately toxic reagent that can cause damage to the central nervous system and liver in humans [77]. Therefore, the potential risk of A3K exposure during cooking cannot be overlooked. The contribution of HMW-CCs to indoor CCs during the cooking process increased more significantly in the heating season. For instance, C9 and C10 increased by 42% and 71%, respectively. Liu et al. [72] reported that C9 and C10 could be emitted from ozone reactions on various indoor surfaces, such as surface oil films originating from cooking [78]. These results confirmed that cooking activity was a significant source of CCs.

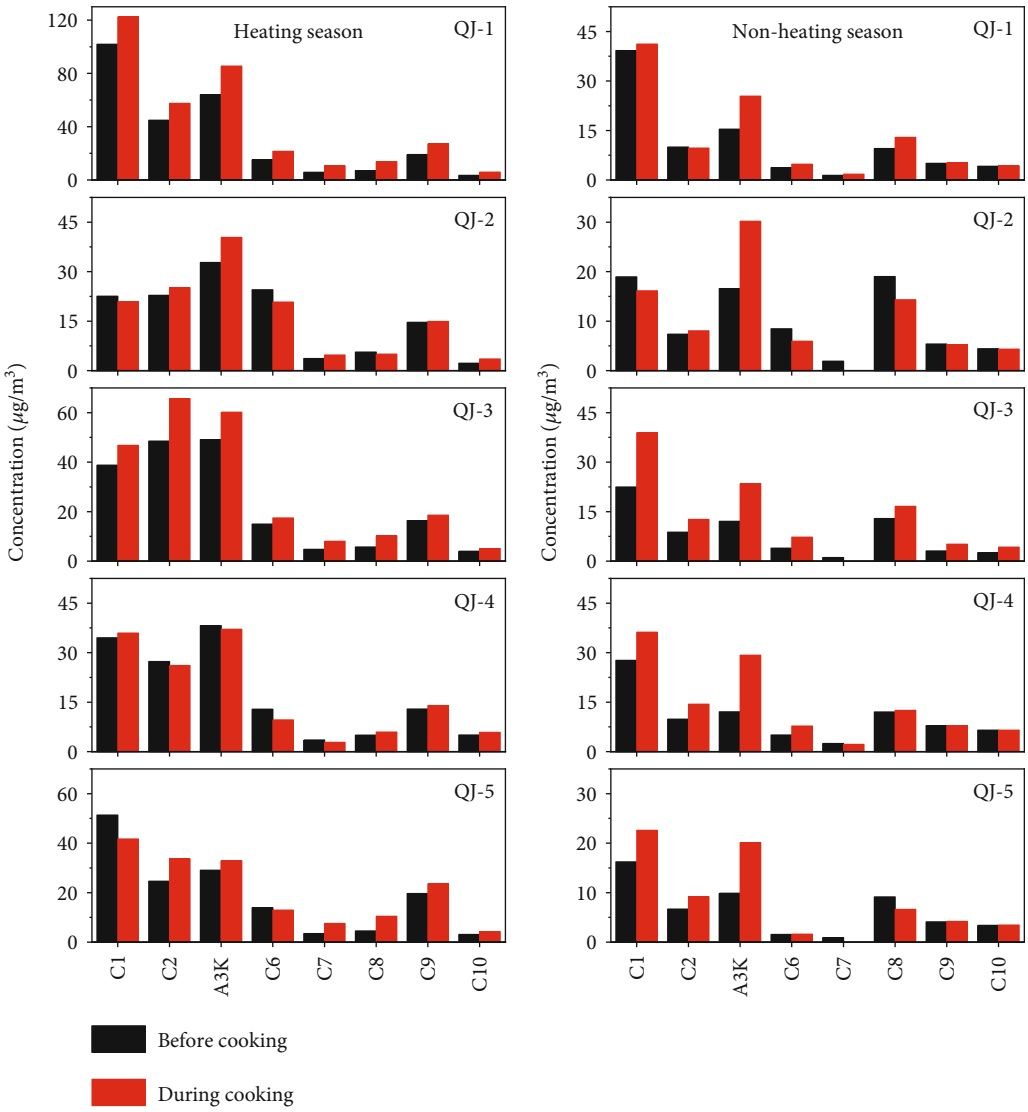

Figure 4: Changes of CC concentrations during cooking and before cooking.

3.3. Inhalation Exposure Estimation and Health Risk Assessment of Children. Table 2 summarizes the daily inhalation doses of formaldehyde and acetaldehyde (E_{ij}) for children in each household during the heating season. The highest daily inhalation doses of formaldehyde and acetaldehyde for children aged <2 years were 398.5 μg/day and 223.0 μg/day, while for children aged 2-6 years, they were 580.8 μg/day and 325.0 μg/day, respectively. The daily average inhalation doses of formaldehyde and acetaldehyde for children aged <2 years (209.5 and 154.5 μg/day) and 2-6 years (305.3 and 225.1 μg/day) in QJ were approximately 3-4 times higher than those in HS (aged <2 years: 50.8 and 44.3 μg/day; aged 2-6 years: 74.1 and 64.6 μg/day) and XY (aged <2 years: 63.1 and 57.3 μg/day; aged 2-6 years: 92.0 and 83.5 μg/day). Fan et al. [1] reported that the inhalation doses of formaldehyde and acetaldehyde for children in the child's bedroom were nearly ten times that in the living room due to prolonged exposure time. Therefore, indoor CCs pollution in the bedrooms of children warrants greater attention.

The child-specific NSRLs were utilized to assess the health risks of children exposed to formaldehyde and acetaldehyde in their homes. Table 2 summarizes the risk quotient (RQ) of each child's exposure to these chemicals during the heating season. The inhalation dose exceeded the safety limit (RQ > 1.0) of the exposure dose recommended by the Office of Environmental Health Hazard Assessment (OEHHA). The results showed that children in all three communities were at risk of developing cancer (cancer risk > 10^{-5}). During the heating season, the RQ of formaldehyde exposure for children aged <2 years in households was 45.7-773, and for children aged 2-6 years, it was 10.9-185. These findings demonstrated that children's exposure to formaldehyde was tens to hundreds of times the "Safe Harbor" level. The values of RQ for acetaldehyde exposure in all households ranged from 15.4 to 192.3 for children aged <2 years and

Table 2: Daily inhalation doses and risk quotients of formaldehyde and acetaldehyde by children in each household.

Site	Formaldehyde				Acetaldehyde			
	Birth to <2 years old		2 to <6 years old		Birth to <2 years old		2 to <6 years old	
	E_{ij} (μg/day)	RQ	E_{ij} (μg/day)	RQ	E_{ij} (μg/day)	RQ	E_{ij} (μg/day)	RQ
HS-1	62.9	122.1	91.7	29.1	59.4	51.2	86.6	12.2
HS-2	46.3	89.9	67.5	21.5	51.2	44.2	74.6	10.5
HS-3	23.5	45.7	34.3	10.9	17.9	15.4	26.0	3.7
HS-4	70.5	136.7	102.7	32.6	48.7	42.0	70.9	10.0
QJ-1	398.5	773.2	580.8	184.5	176.6	152.3	257.4	36.4
QJ-2	106.7	207.0	155.5	49.4	110.7	95.4	161.3	22.8
QJ-3	187.3	363.3	272.9	86.7	223.0	192.3	325.0	45.9
QJ-4	141.9	275.3	206.8	65.7	115.2	99.3	167.9	23.7
QJ-5	212.9	413.0	310.3	98.6	146.8	126.5	213.9	30.2
XY-1	73.9	143.3	107.6	34.2	49.1	42.3	71.6	10.1
XY-2	49.4	95.9	72.0	22.9	45.6	39.3	66.4	9.38
XY-3	88.9	172.4	129.5	41.2	119.2	102.8	173.8	24.5
XY-4	48.3	93.7	70.4	22.4	35.7	30.8	52.0	7.35
XY-5	55.1	106.9	80.3	25.5	36.7	31.7	53.5	7.56

Table 3: Odor activity values of indoor pollutants at HS, QJ, and XY.

Compounds			Odor threshold/(μg/m^3)	Odor activity value		
				HS	QJ	XY
LMW-CCs	Formaldehyde	C1	670.3	0.02	0.07	0.02
	Acetaldehyde	C2	2.95	3.66	11.4	4.27
	Acetone	A3K	108900	<0.01	<0.01	<0.01
	Propionaldehyde	*n*-C3	2.59	0.75	1.58	0.91
	2-Butanone	MEK	1416.4	<0.01	<0.01	<0.01
	i,n-Butyraldehyde	*i,n*-C4	1.64	0.96	1.61	1.00
	i-Valeraldehyde	*i*-C5	0.38	7.99	7.98	2.89
	Valeraldehyde	*n*-C5	1.58	0.79	1.81	1.05
HMW-CCs	Hexaldehyde	C6	1.25	3.72	12.9	6.18
	Heptanaldehyde	C7	0.8	2.56	5.32	2.98
	Octanaldehyde	C8	0.06	26.2	95.3	42.0
	Nonanaldehyde	C9	2.16	3.56	7.68	5.59
	Decanaldehyde	C10	2.79	1.08	1.29	1.52

from 3.7 to 45.9 for children aged 2-6 years. Compared with children's exposure to acetaldehyde, formaldehyde exposure had a greater impact on children's health, particularly for children aged <2 years. This was consistent with the results of a study by Bradman et al. [18], which reported higher RQ values for formaldehyde exposure (12.0-51.7) than those for acetaldehyde exposure (2.3-9.8).

3.4. Odor Pollution of HMW-CCs Emitted from Indoor Environments. Chinese urban residents spend almost 90% of their lifetime indoors, with 72% of that time spent in residential indoor environments [25]. Consequently, the emission and concentration of indoor CCs are strongly associated with human health. Odor activity value (OAV) was used for estimating odor intensity and evaluating the contribution of CCs to perceived odor. When the OAV was below 1, the CC theoretically cannot be perceived by the general population, whereas an OAV above 1 indicated that the CC was expected to contribute to the perceived odor [23]. Table 3 shows the OAV of the CCs in the indoor environment of HS, QJ, and XY. Generally, the odor thresholds of LMW-CCs, namely, C1 and A3K, in the indoor environment of residences were relatively high, resulting in a lower OAV for these compounds. In contrast, C2 showed a significant odor activity with activity values greater than 3. The OAV of HMW-CCs, namely, C6, C7, C8, C9, and C10 was greater than 1. Notably, the OAV of C8 was greater than 10, indicating that HMW-CCs should be the focus in exploring the odor pollution of CCs in the indoor environment.

Indoor chemical factors are considered important causes of SBS. Some decorative materials contain chemical components that may induce SBS. C6 may induce general

symptoms of SBS, while C8 and C10 can significantly cause skin symptoms of SBS [32]. Therefore, it is necessary to consider setting limits for HMW-CCs in the indoor environments. Several studies have shown that older buildings increase the risk of SBS. However, Fu et al. [79] found that housing built in the last ten to fifteen years also contributed to the occurrence of SBS, possibly due to chemical residues in newer buildings, such as new decorations and new furnishings. The present study observed that new buildings in QJ had stronger odor pollution and may pose greater SBS risks for health.

4. Conclusions

The prevalent and abundant CCs in the indoor and outdoor environment of residential buildings were LMW-CCs, such as formaldehyde, acetaldehyde, and acetone. The concentrations of HMW-CCs (hexanal, nonanal, and decanal) were significantly higher indoors than outdoors. Indoor CCs concentrations were higher during the heating season, while outdoor CCs concentrations were higher during the nonheating season. This seasonal contrast was due to the influence of temperature on outdoor CCs pollution and heating and ventilation on indoor CCs pollution. PCA of indoor measurements identified five components that accounted for 90.5% of the total variance, including building materials and cooking fumes, wooden furniture, outdoor infiltration, personal care products, and cigarette smoke. During the cooking period, LMW-CCs contributed more to indoor CCs in the nonheating season, while HMW-CCs contributed more in the heating season. Formaldehyde exposure posed a greater health risk to children than acetaldehyde exposure in the indoor environment. The main source of odor pollution in residential indoor environments was HMW-CCs with low odor thresholds, including hexanal, heptanal, octanal, nonanal, and decanal. Currently, China only regulates indoor formaldehyde levels, with no relevant regulations for other CCs that have adverse effects on human health. Our findings can provide valuable insights into the profile of indoor CCs pollution in urban regions in China and highlight the need for relevant standards on indoor air quality.

Data Availability

The data used to support the findings of this study are available from the corresponding authors upon request.

Conflicts of Interest

The authors declare no competing financial interest.

Authors' Contributions

Lu Li did the conceptualization, methodology, validation, data curation, formal analysis, investigation, and writing, which of the original draft. Wenting Dai performed the conceptualization, resources, writing, which includes the review and editing, supervision, and funding acquisition. Minxia Shen was assigned to data curation. Xinyi Niu focused on data curation and investigation. Tafeng Hu was assigned to resources. Jing Duan worked on data curation and investigation. Junji Cao was tasked with resources and supervision. Zhenxing Shen did the resources. Kin Fai Ho did the writing, which includes review and editing. Jianjun Li worked on the conceptualization, writing, which includes review and editing, and supervision.

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Supplementary Materials

Supporting Materials S1: adjustment of child-specific NSRLs and cancer risk evaluation. S2: test method for ventilation frequency. Figure S1: location of the sampling areas. Table S1: descriptions of sampling locations. Table S2: the values of parameters for inhalation exposure calculation and health risk assessment for children. Table S3: the variations of T, RH, CC, and $PM_{2.5}$ concentrations at different sampling sites in the heating season. Table S4: characteristics of CCs in indoor and outdoor environments ($\mu g/m^3$). Table S5: the number of air changes of households in the heating season and the nonheating season. Table S6: Spearman correlation calculated taking into account all CCs quantified in indoor environments. *(Supplementary Materials)*

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