

Indoor and outdoor carbonyl compounds in the hotel ballrooms in Guangzhou, China

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Abstract

Twenty-one carbonyl compounds were measured simultaneously at four hotel ballrooms in urban Guangzhou during the autumn, 2002. In each ballroom, measurements were carried out in business hours in the evening (20:30–24:00) on 7 consecutive days without any disturbance of the ballroom's normal operation. Nineteen out of the 21 target carbonyl compounds were identified in indoor and outdoor air. In the outdoor environment, formaldehyde was the most abundant carbonyl, followed by acetaldehyde, and there existed a strong correlation between formaldehyde and acetaldehyde. In the indoor air, however, acetaldehyde was the most abundant carbonyl, its concentrations seemed to be affected significantly by smoking. The indoor concentrations of carbonyls were found higher than their outdoor counterparts with only a few exceptions. Further studies concerning the indoor/outdoor ratios and mutual correlation of the carbonyls indicated that apart from direct emission from indoor materials and infiltration of outdoor air, other anthropogenic sources, e.g. tobacco smoke, also significantly contributed to carbonyl compounds. The possible sources of some high molecular weight carbonyls, e.g. nonanaldehyde, were also discussed briefly. Preliminary estimate of the exposures and risks due to carbonyls in the ballrooms was made, which indicated that long-term exposure in such places might cause increased chance of developing cancers.

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1. Introduction

Carbonyls in urban air have received scientific and regulatory attention as toxic air contaminants (Shepson et al., 1986) and important precursors to free radicals, ozone, and peroxyacyl nitrates (Carrier et al., 1986; Grosjean et al., 1993a, 1996; Carter, 1995). They can be produced directly from incomplete combustion of biomass and/or fossil fuels and also through the atmospheric photo-oxidation of hydrocarbons (Satsumabayashi et al., 1995). In indoor air, some

carbonyl compounds could be released from building materials, furniture, and consumer products and through reactions between indoor ozone and alkenes (Crump and Gardiner, 1989; Kelly et al., 1999). Cigarette smoke was another significant indoor source of several carbonyls (Löfroth et al., 1989; Morrison and Nazaroff, 2002).

Ambient air concentrations of carbonyl compounds in urban and rural areas have been widely measured (Grosjean, 1982; Shepson et al., 1991; Szilagyi et al., 1991; Possanzini et al., 1996; Slemr and Junkermann, 1996; Granby and Christensen, 1997; Montero et al., 2001; Nguyen et al., 2001; Báez et al., 2001), in the mean time their seasonal (Tanner and Meng, 1984; Christensen et al., 2000; Ho et al., 2002) and diurnal variations

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(Viskari et al., 2000; Sin et al., 2001) have also been discussed. While for indoor measurements major target compounds are formaldehyde and acetaldehyde due to their higher concentrations in indoors than those outdoors, resulting from indoor sources (Miguel et al., 1995; Brickus et al., 1998; Khoder et al., 2000). A few studies, however, have reported other carbonyl compounds presented both in indoor and outdoor air (Zhang et al., 1994a; Reiss et al., 1995; William et al., 1996; Báez et al., 2003). Zhang et al. (1994a) measured nine aldehydes in both indoor and outdoor air of six New Jersey homes, which showed the indoor to outdoor ratios (I/O) > 1 with the exception of propionaldehyde. Williams et al. (1996) and Báez et al. (2003) found that there were higher carbonyl concentrations with than without tobacco smoke, indicating the importance of tobacco smoking as a major source of carbonyls. But almost no studies about high molecular weight (HMW) carbonyls were measured in the indoor and outdoor air, especially in public entertainment places. In entertainment places with enhanced human activities in particular hours and with much more indoor decorations, like hotel ballrooms, studies of indoor carbonyls including HMW compounds are needed for human health assessment.

Guangzhou is a central city in the Pearl River Delta, one of the most developed regions in south China, where there are many popular places like ballrooms for nighttime entertainment. In the present study, four hotel ballrooms were selected for indoor and outdoor measurements of 21 carbonyls (C₁–C₁₀) during 20:30–24:00 in 7 consecutive days. The human exposure levels and their possible sources of these carbonyls were discussed.

2. Experimental methods

2.1. Sampling site

Four hotel ballrooms in urban Guangzhou were selected for sampling (Fig. 1 and Table 1). Indoor and outdoor measurements were conducted from 20 August–20 September 2002, and in each ballroom samples were collected in the evening time between 20:30 and 24:00 in 7 consecutive days. During the sampling periods, the ballrooms run normally, and guests and waiters just behaved themselves as usual without any extra restriction of their activities like drinking and smoking. Samples were simultaneously taken at an outdoor site

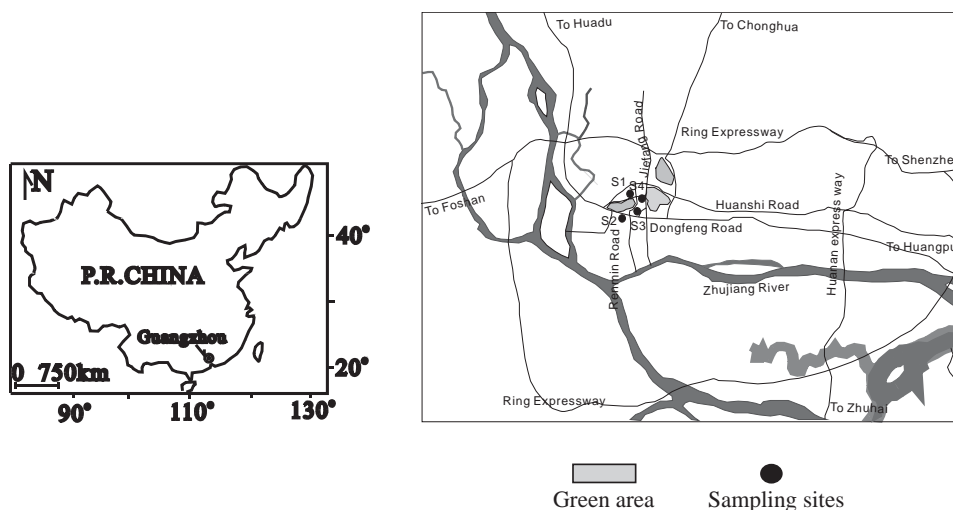


Fig. 1. Location of sampling sites.

Table 1
Description of sampling sites

| Sampling site | Area (m ³) | Floor | The last fitment | Ventilation system |
|---------------|------------------------|----------|---------------------------|----------------------------|
| S1 | 80 | Third | 4 years | Central air condition |
| S2 | 240 | Third | Carpets replaced 3 months | Individual fresh air taker |
| S3 | 220 | Third | 3 years | Individual fresh air taker |
| S4 | 120 | Basement | 3 years | Central air condition [f1] |

near the fresh air intake, and an indoor site away from people's activity in each ballroom.

2.2. Materials and reagents

All solvents employed were HPLC grade. Water was double distilled and filtered by Milli-Q. The acetonitrile (ACN) and the 2,4-dinitrophenylhydrazine (DNPH) were purchased from Merck (Germany) and Fluka (USA), respectively, and the calibration standard (Carbonyl-DNPH Mix 2) containing 15 components (DNPH derivatives of acetaldehyde, acetone, acrolein, benzaldehyde, butyraldehyde, crotonaldehyde, 2,5-dimethylbenzaldehyde, formaldehyde, hexaldehyde, isovaleraldehyde, propionaldehyde, *o*-tolualdehyde, *m*-tolualdehyde, *p*-tolualdehyde, valeraldehyde) and other six individual components (2-butanone, cyclohexanone, heptaldehyde, octylaldehyde, nonanaldehyde, decylaldehyde) were purchased from ChemService (West Chester, USA).

2.3. Carbonyl sampling and analysis

The whole method was based on EPA method TO-11A (US EPA, 1999). The sampling medium was a Sep-Pak Silica Gel Cartridge (Waters, Millipore Corp.). DNPH was purified by recrystallization in HPLC grade ACN three times. Each cartridge was rinsed by attaching a syringe filled with 10 ml of ACN and coated slowly by passing 7 ml of the freshly made coating solution, which contained 60 ml DNPH-ACN-saturated solution and 4 ml concentrated ortho-phosphoric acid in 500 ml ACN, through the cartridge by gravity. When there was no more solution flowing out of the cartridge, the cartridge was dried with a gentle flow of nitrogen for 15 min, and then wrapped in aluminum foils, sealed in Teflon bags, and stored in refrigerator at 4°C until use. Three blank cartridges from each cartridge batch were analyzed and the results were below the EPA blank criteria except acetone (formaldehyde < 0.14 µg/cartridge; acetaldehyde < 0.09 µg/cartridge; acetone < 2.53 µg/cartridge and other aldehydes or ketones concentration < 0.07 µg/cartridge). The cartridge was reported to be good for at least 6 months when stored under 4°C (Levin et al., 1985; Druzik et al., 1990). The storage time of our cartridges was less than 30 days.

Samples were collected by drawing the air with a sampling pump (Thomas, USA) through the cartridge. A potassium iodide (KI) denuder was connected to the upstream of the cartridge to prevent the interference of ozone (Sirju and Shepson, 1995). The samples were collected in the evening between 20:30 and 24:00 each day at a flow rate of 0.8–1.21 min⁻¹. The flow rate was measured with a digital flow meter (DryCal DC Lite, Bios Corp., USA) during sampling. After sampling, each cartridge was wrapped in aluminum foil, resealed with a

Teflon bag, and transported back to the laboratory and stored in the refrigerator before being analyzed. Each sampling program included one laboratory blank and one field blank and in each sampling sites two field samples collocated with back-up cartridge to evaluate breakthrough.

The sampled cartridges were eluted slowly with 5 ml of ACN into a 5-ml volumetric flask, store in refrigerated conditions until the samples were analyzed. The eluted samples were stable at 4°C for up to 1 month (US EPA, 1999). A 20-µl aliquot was injected into the HPLC system through an autosampler. The HPLC system was HP1100. The analytical conditions were as follows: Agilent SB-C18 reverse column (250 × 4.6 mm, 5 µm); gradient mobile phase: 60–70% ACN of water solution for 20 min, 70–100% ACN for 3 min, 100% ACN for 4 min, 100–60% ACN for 1 min, and then 60% ACN for 5 min; mobile-phase flow rate: 1 ml min⁻¹; detector: UV at 360 nm.

2.4. Quality assurance and quality control

Identification and quantification of carbonyl compounds were based on their retention times and peak areas, respectively. The instrument was calibrated using five standard concentrations (from 0.5 to 10 µg ml⁻¹) covering the concentration of interest for ambient work. There were very good linear relationships ($R^2 > 0.999$) between the concentrations and responses for all carbonyls identified. Cartridge collection efficiency was determined with two cartridges in series, and over 99% of carbonyl compounds were found in the first cartridge. Second elution tests indicated complete recovery of all of the carbonyls. Relative standard deviations (RSDs) for replicate analyses were less than 5%. RSDs of collocated samples were often below 13%. Method detection limits (MDLs) was determined by using seven replicate analyses of the working standards at the lowest concentration, for which an approach was described in detail in 40 CFR Part 136B (Code of Federal Regulation, 2001). The MDLs of present study were found to range 0.05–0.15 µg m⁻³ for various carbonyls of 120 l sampling volume.

3. Results and discussion

3.1. Indoor and outdoor levels

Twenty-one carbonyl compounds were measured in the outdoor air and indoor air, of which 19 compounds were identified as: formaldehyde, acetaldehyde, propionaldehyde, crotonaldehyde, butyraldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, hexaldehyde, *p*-tolualdehyde, *m/o*-tolualdehyde, 2-butanone, cyclohexanone, heptaldehyde, octylaldehyde, nonanaldehyde,

decylaldehyde and 2,5-dimethylbenzaldehyde. The concentrations of these carbonyls in the outdoor air and indoor air were summarized in Table 2. Acrolein, a common component in environmental tobacco smoke and vehicle emissions, could not be detected in this

study, just as Zhang et al. (1994a) and Reiss et al. (1995) had reported. 2,5-dimethylbenzaldehyde was only found in three outside samples, and *p*-tolualdehyde only in five indoor samples and *m/o*-tolualdehyde in 80% of the samples collected. Because of the high background of

Table 2
Indoor and outdoor concentrations ($\mu\text{g m}^{-3}$) of carbonyl compounds

| Compound | | S1 | | S2 | | S3 | | S4 | | GM ^a ±GSD ^b | N | Max. | Min. |
|--------------------------|---------|-----------------|---|-----------------|---|-----------------|---|-----------------|---|-----------------------------------|----|--------|-------|
| | | GM ^c | N | GM ^c | N | GM ^c | N | GM ^c | N | | | | |
| Formaldehyde | Outdoor | 9.59 | 6 | 15.38 | 7 | 13.19 | 5 | 11.81 | 7 | 12.37±7.33 | 25 | 29.00 | 6.43 |
| | Indoor | 58.04 | 7 | 23.66 | 7 | 18.32 | 7 | 31.12 | 7 | 29.74±21.88 | 28 | 63.00 | 26.26 |
| Acetaldehyde | Outdoor | 4.57 | 6 | 9.24 | 7 | 8.04 | 5 | 6.58 | 7 | 6.90±4.76 | 25 | 17.31 | 3.12 |
| | Indoor | 42.17 | 7 | 221.07 | 7 | 33.15 | 7 | 76.97 | 7 | 69.84±121.05 | 28 | 416.41 | 22.90 |
| Acetone | Outdoor | Dq | | Dq | | Dq | | Dq | | | | | |
| | Indoor | Dq | | Dq | | Dq | | Dq | | | | | |
| Acrolein | Outdoor | Nd | | Nd | | Nd | | Nd | | | | | |
| | Indoor | Nd | | Nd | | Nd | | Nd | | | | | |
| Propionaldehyde | Outdoor | 0.74 | 6 | 1.23 | 7 | 1.63 | 5 | 1.35 | 7 | 1.18±0.95 | 25 | 3.13 | 0.35 |
| | Indoor | 2.18 | 7 | 2.87 | 7 | 1.80 | 7 | 3.09 | 7 | 2.43±1.20 | 28 | 4.15 | 1.31 |
| Crotonaldehyde | Outdoor | 0.38 | 6 | 0.43 | 7 | 0.32 | 5 | 0.45 | 7 | 0.40±0.26 | 25 | 0.87 | 0.19 |
| | Indoor | 0.49 | 7 | 0.51 | 7 | 0.45 | 7 | 0.96 | 7 | 0.57±0.39 | 28 | 1.34 | 0.28 |
| 2-butanone | Outdoor | 3.03 | 6 | 7.75 | 7 | 6.48 | 5 | 5.05 | 7 | 5.29±6.12 | 25 | 20.50 | 0.90 |
| | Indoor | 4.62 | 7 | 8.52 | 7 | 4.76 | 7 | 7.31 | 7 | 60.8±4.89 | 28 | 20.52 | 2.15 |
| Butyraldehyde | Outdoor | 0.50 | 6 | 1.10 | 7 | 0.96 | 5 | 1.08 | 7 | 1.01±2.93 | 25 | 12.91 | 0.08 |
| | Indoor | 2.61 | 7 | 3.70 | 7 | 1.95 | 7 | 3.26 | 7 | 2.80±1.53 | 28 | 5.31 | 1.54 |
| Benzaldehyde | Outdoor | 0.76 | 6 | 1.14 | 7 | 1.11 | 5 | 1.03 | 7 | 1.00±0.62 | 25 | 2.22 | 0.52 |
| | Indoor | 1.20 | 7 | 0.79 | 7 | 0.82 | 7 | 0.99 | 7 | 0.93±0.40 | 28 | 1.96 | 0.49 |
| Isovaleraldehyde | Outdoor | 0.23 | 6 | 0.35 | 7 | 0.47 | 5 | 0.49 | 7 | 0.37±0.25 | 25 | 0.86 | 0.18 |
| | Indoor | 0.80 | 7 | 2.82 | 7 | 0.73 | 7 | 1.62 | 7 | 1.28±1.40 | 28 | 4.02 | 0.27 |
| Cyclohexanone | Outdoor | 0.68 | 6 | 1.99 | 7 | 2.43 | 5 | 2.33 | 7 | 1.67±2.54 | 25 | 6.00 | 0.06 |
| | Indoor | 5.74 | 7 | 1.62 | 7 | 1.67 | 7 | 3.37 | 7 | 2.69±3.03 | 28 | 7.00 | 0.48 |
| Valeraldehyde | Outdoor | 0.21 | | 0.46 | 7 | 0.44 | 5 | 0.35 | 7 | 0.35±0.29 | 25 | 1.06 | 0.07 |
| | Indoor | 1.02 | 7 | 0.78 | 7 | 0.44 | 7 | 0.56 | 7 | 0.67±0.39 | 28 | 1.36 | 0.30 |
| <i>p</i> -Tolualdehyde | Outdoor | Nd | | Nd | | Nd | | Nd | | | | | |
| | Indoor | | | | | | | | | 0.06±0.08 ^d | 5 | | |
| <i>m/o</i> -Tolualdehyde | Outdoor | 0.20 | 4 | 0.44 | 7 | 0.58 | 5 | 0.18 | 7 | 0.30±0.57 | 23 | 0.70 | Nd |
| | Indoor | 0.13 | 7 | 0.32 | 7 | 0.30 | 7 | 0.43 | 7 | 0.55±0.69 | 28 | 1.19 | 0.10 |
| Hexaldehyde | Outdoor | 0.48 | 6 | 0.77 | 7 | 0.89 | 5 | 0.74 | 7 | 0.70±0.45 | 25 | 1.71 | 0.22 |
| | Indoor | 2.19 | 7 | 1.42 | 7 | 1.00 | 7 | 1.11 | 7 | 1.36±0.77 | 28 | 3.05 | 0.80 |
| 2,5-dimethylbenzaldehyde | Outdoor | | | | | | | | | 0.64±0.06 ^d | 3 | | |
| | Indoor | Nd | | Nd | | Nd | | Nd | | | | | |
| Heptaldehyde | Outdoor | 0.32 | 6 | 0.47 | 7 | 0.51 | 5 | 0.59 | 7 | 0.46±0.21 | 25 | 0.85 | 0.18 |
| | Indoor | 0.56 | 7 | 0.51 | 7 | 0.43 | 7 | 0.49 | 7 | 0.49±0.16 | 28 | 0.82 | 0.35 |
| Octaldehyde | Outdoor | 0.06 | 6 | 0.19 | 7 | 0.35 | 5 | 0.27 | 7 | 0.18±0.28 | 25 | 1.08 | 0.01 |
| | Indoor | 0.82 | 7 | 0.42 | 7 | 0.36 | 7 | 0.36 | 7 | 0.46±0.35 | 28 | 1.55 | 0.26 |
| Nonanaldehyde | Outdoor | 0.88 | 6 | 1.47 | 7 | 2.44 | 5 | 2.36 | 7 | 1.63±1.31 | 25 | 3.77 | 0.29 |
| | Indoor | 1.88 | 7 | 1.46 | 7 | 2.01 | 7 | 1.54 | 7 | 1.71±0.65 | 28 | 2.66 | 0.96 |
| Decylaldehyde | Outdoor | 0.15 | 6 | 0.32 | 7 | 0.49 | 5 | 0.40 | 7 | 0.31±0.26 | 25 | 0.83 | 0.06 |
| | Indoor | 0.87 | 7 | 0.23 | 7 | 0.52 | 7 | 0.38 | 7 | 0.45±0.42 | 28 | 1.64 | 0.18 |
| Total | Outdoor | 23.92 | 6 | 45.60 | 7 | 41.06 | 5 | 35.75 | 7 | 36.72±21.95 | | 87.97 | 14.92 |
| | Indoor | 128.15 | 7 | 276.19 | 7 | 70.65 | 7 | 136.37 | 7 | 135.90±126.70 | | 457.43 | 58.86 |

N: number of samples; Dq: Did not quantify; Nd: Not detected.

^aGM: Geomean in all sampling sites.

^bGSD: Geometrical standard deviation.

^cGM: Geomean in one sampling site.

^dGSD calculated using only those samples in which carbonyls were detected.

acetone in our laboratory, acetone in the samples could not be quantified accurately and so was not reported here.

Of all the carbonyls identified in the outdoor environment, formaldehyde ($12.37 \pm 7.33 \mu\text{g m}^{-3}$) and acetaldehyde ($6.90 \pm 4.76 \mu\text{g m}^{-3}$) were the most abundant carbonyls, followed by 2-butanone, propionaldehyde, butyraldehyde and benzaldehyde. The least was octylaldehyde ($0.18 \pm 0.28 \mu\text{g m}^{-3}$). The HMW carbonyls showed the concentrations less than $0.5 \mu\text{g m}^{-3}$ except nonanaldehyde. The total concentration of all carbonyls had a geomean outdoor level of $36.72 \pm 21.95 \mu\text{g m}^{-3}$, in which formaldehyde accounted for $33.68 \pm 6.79\%$ and acetaldehyde for $18.79 \pm 3.91\%$. In the indoor environment, the carbonyls had a geomean level of $136.26 \pm 126.74 \mu\text{g m}^{-3}$ with acetaldehyde to be the most abundant carbonyl compound ($69.84 \pm 121.05 \mu\text{g m}^{-3}$). Acetaldehyde and formaldehyde accounted for $51.25 \pm 32.60\%$ and $21.83 \pm 8.33\%$ of the total indoor carbonyls, respectively. A wide range of the carbonyl concentrations were observed both outdoors and indoors over the 7-day sampling period.

Formaldehyde and acetaldehyde were the two most abundant carbonyls in the urban air of Guangzhou, which was comparable to that in other urban areas in the world (Table 3). The concentrations of formaldehyde and acetaldehyde were similar to Rome, Italy (Possanzini et al., 1996). Brazil (Miguel et al., 1995) was unique due to the extensive use of ethanol fuel throughout that country. Except for Mexico (Báez et al., 1995), the carbonyl concentrations in Guangzhou were much higher than that of many other cities. For example, although Hong Kong was very close to Guangzhou, the carbonyl concentrations in Guangzhou were three times higher than that in Hong Kong.

3.2. Mutual correlations, I/O ratios and source implications

The correlations between formaldehyde, acetaldehyde and other carbonyls were shown in Table 4. The

significant correlation between formaldehyde and acetaldehyde ($R = 0.95$) implied that the two carbonyls had almost the same sources. With the exception of some HMW carbonyls, all the other carbonyls were reasonably well correlated with formaldehyde and acetaldehyde. Based on the study of Possanzini et al. (1996) and Ho et al. (2002), vehicular exhaust was the primary source in winter while both vehicular exhaust and photochemical reactions were the major sources in summer. Our sampling period was between 20 August and 20 September 2002, it was just autumn of Guangzhou, and the sampling time for all the samples collected was after 8 at night, indicating that vehicular exhaust was the major source of carbonyls. In the mean time photochemical reactions may also make some contribution, because part of the carbonyls produced during the daytime might survive until night.

The formaldehyde/acetaldehyde (C_1/C_2) ratios and the acetaldehyde/propionaldehyde (C_2/C_3) ratios were

Table 4
Correlation coefficients for some carbonyls outdoors

| Compounds | R (formaldehyde) ^a | R (acetaldehyde) |
|--------------------------|---------------------------------|--------------------|
| Formaldehyde | 1.00 | 0.95 |
| Acetaldehyde | 0.95 | 1.00 |
| Propionaldehyde | 0.84 | 0.88 |
| Crotonaldehyde | 0.50 | 0.60 |
| 2-Butanone | 0.73 | 0.78 |
| Butyraldehyde | 0.55 | 0.56 |
| Benzaldehyde | 0.93 | 0.94 |
| Isovaleraldehyde | 0.65 | 0.70 |
| Cyclohexanone | 0.65 | 0.58 |
| Valeraldehyde | 0.85 | 0.90 |
| <i>m/o</i> -Tolualdehyde | 0.67 | 0.75 |
| Hexaldehyde | 0.85 | 0.76 |
| Heptaldehyde | 0.40 | 0.45 |
| Octylaldehyde | 0.26 | 0.30 |
| Nonanaldehyde | -0.04 | 0.11 |
| Decylaldehyde | 0.01 | 0.10 |
| Total | 0.96 | 0.97 |

^a Pearson coefficient.

Table 3
Comparison of mean concentrations ($\mu\text{g m}^{-3}$) of the most abundant carbonyls with other urban cities

| Location | Sampling time | HCHO | CH ₃ CHO | References |
|------------------------|-----------------------------|-----------|---------------------|-------------------------------|
| Atlanta, USA | July–August 1992 | 3.3–3.7 | 4.7–5.7 | Grosjean et al. (1993b) |
| California state, USA | 1990–1998 | 3.3 | 2.0 | CARB (1999) |
| Copenhagen, Denmark | February–June 1994 | 3.2 | 1.8 | Granby and Christensen (1997) |
| Hong Kong, China | April 1999–April 2000 | 4.13–5.27 | 1.73–2.53 | Ho et al. (2002) |
| Kuopio, Finland | May 1997–January 1998 | 1.3–2.8 | 1.1–3.2 | Viskari et al. (2000) |
| Mexico City, Mexico | March–May 1995 | 43.5 | 28.6 | Báez et al. (1995) |
| Rio de Janeiro, Brazil | 3 May–1 November 2000 | 10.84 | 10.43 | Grosjean et al. (2002) |
| Rome, Italy | January–March 1995 | 13.7 | 8.2 | Possanzini et al. (1996) |
| Takasaki, Japan | Summer 1983–1986 | 3.1–14 | 2.3–12 | Satsumabayashi et al. (1995) |
| Guangzhou, China | 20 August–20 September 2002 | 13.29 | 7.6 | Present study |

calculated (Table 5). It was reported that C_1/C_2 ratios usually varied from 1–2 (urban) to about 10 (rural or forested areas); therefore, they could be used as an indicator of biogenic sources of formaldehyde (Shepson et al., 1991). The average C_1/C_2 ratio value in this study was 1.81 that was very close to the value reported by Grosjean (1982) (ratio = 1.80). C_2/C_3 ratios were often used as effective indicators of anthropogenic origin for ambient carbonyls, since propionaldehyde was believed to be associated only with anthropogenic emissions, in contrast, other carbonyls had both anthropogenic and natural sources. Then, C_2/C_3 ratios would be high in rural atmospheres and low in polluted urban air. The average value was 6.2 in the present study, which was quite similar to the value reported by Grosjean (1988) (ratio = 6.0), indicating the presence of anthropogenic sources in the urban areas of Guangzhou. Comparing with the ratios of other urban areas (range 1.7–6.0) (Grosjean, 1988; Zhang et al., 1994a; Possanzini et al., 1996), the ratio of Guangzhou was a little higher. While

in Hong Kong C_2/C_3 ratio was 8.38 (Ho et al., 2002), which was higher than that in Guangzhou.

By comparison of the indoor and outdoor carbonyl concentrations, it was found that “indoor > outdoor” (I/O ratios > 1) was very common for almost every pair of samples measured (Table 6). The I/O ratios in S1 were higher than that in other three places except for one or two carbonyls (e.g. acetaldehyde), and the I/O ratios for almost all components in S3 were the lowest. The I/O ratio of acetaldehyde was especially high in S2. The difference between the I/O ratios might result from the different ventilation conditions and indoor source strengths. As described in Table 1, the area of S1 was the smallest and without independent ventilation system, thus the carbonyls would be accumulated in the room. Conditions were similar in S4, which was also not so big in size and located in the basement without good ventilation. As a consequence, both sites S1 and S4 resulted in higher I/O ratios. S3 was much bigger in size with good ventilation, and the I/O ratios were obviously much smaller. For S2, there were some special reasons for its high I/O ratio of acetaldehyde (discussed later).

Carbonyl compounds present in the indoor air were thought to be resulting from the indoor emissions, indoor chemical formation and outdoor infiltration. There was always possibility for the penetration of a pollutant presented in the outdoor air into the houses. In our present study, there was a continuous exchange of air with the outside by ventilation system. Therefore, all contaminants of outdoor air were likely to be presented

Table 5

The concentration ratios of formaldehyde/acetaldehyde (C_1/C_2) and acetaldehyde/propionaldehyde (C_2/C_3)

| Ratio | N | Mean | SD | Max. | Min. |
|-----------|----|------|------|-------|------|
| C_1/C_2 | 25 | 1.81 | 0.28 | 2.42 | 1.30 |
| C_2/C_3 | 25 | 6.16 | 2.35 | 13.87 | 3.25 |

Mean: arithmetic mean; SD: standard deviation.

Table 6

The ratios of the indoor/outdoor (I/O) carbonyl concentrations

| Compound | GM ^a | | | | GM ^b | GSD | N | Max. | Min. |
|--------------------------|-----------------|-------|------|-------|-----------------|-------|----|-------|------|
| | S1 | S2 | S3 | S4 | | | | | |
| Formaldehyde | 6.03 | 1.54 | 1.41 | 2.63 | 2.44 | 2.90 | 25 | 8.56 | 0.98 |
| Acetaldehyde | 9.12 | 23.94 | 4.03 | 11.70 | 10.88 | 16.90 | 25 | 55.94 | 2.52 |
| Propionaldehyde | 3.04 | 2.34 | 1.13 | 2.28 | 2.14 | 2.11 | 25 | 6.20 | 1.00 |
| Crotonaldehyde | 1.30 | 1.20 | 1.35 | 2.13 | 1.47 | 1.49 | 25 | 4.73 | 0.44 |
| 2-Butone | 1.55 | 1.10 | 0.86 | 1.45 | 1.23 | 0.87 | 25 | 3.00 | 0.58 |
| Butyraldehyde | 2.90 | 3.36 | 1.53 | 3.01 | 2.69 | 4.91 | 25 | 20.87 | 0.20 |
| Benzaldehyde | 1.65 | 0.70 | 0.79 | 0.96 | 0.96 | 0.87 | 25 | 3.42 | 0.50 |
| Isovaleraldehyde | 3.44 | 7.95 | 1.40 | 3.29 | 3.59 | 4.41 | 25 | 13.53 | 0.60 |
| Cyclohexaone | 8.26 | 0.81 | 0.86 | 1.45 | 1.68 | 12.24 | 25 | 49.57 | 0.23 |
| Valeraldehyde | 4.93 | 1.67 | 1.12 | 1.60 | 1.98 | 3.08 | 25 | 12.66 | 0.86 |
| <i>m/o</i> -Tolualdehyde | 0.56 | 1.48 | 0.95 | 4.87 | 1.63 | 5.43 | 23 | 15.94 | 0.17 |
| Hexaldehyde | 4.74 | 1.84 | 1.15 | 1.51 | 1.99 | 2.84 | 25 | 10.39 | 0.93 |
| Heptaldehyde | 1.84 | 1.09 | 0.90 | 0.84 | 1.10 | 0.89 | 25 | 3.57 | 0.58 |
| Octylaldehyde | 13.54 | 2.28 | 1.03 | 1.36 | 2.58 | 12.00 | 25 | 39.55 | 0.35 |
| Nonanaldehyde | 2.23 | 0.99 | 0.82 | 0.67 | 1.04 | 1.52 | 25 | 6.19 | 0.29 |
| Decylaldehyde | 5.95 | 0.72 | 1.03 | 0.96 | 1.39 | 3.29 | 25 | 9.27 | 0.40 |
| Total | 4.92 | 6.06 | 1.71 | 3.81 | 3.93 | 3.90 | | 13.56 | 1.33 |

GSD: geometrical standard deviation; N: number of samples.

^aGM: for one sampling site, the geomean of every pair I/O.

^bGM: for all the sampling sites, the geomean of every pair I/O.

in indoors, which originated, to a large extent, from automobile or plants emissions. For some carbonyls, the concentrations of outdoor sources were higher than that of indoors, resulting in I/O ratios < 1 of these carbonyls (e.g. some HMW carbonyls). Another important source was indoor chemical formation. Previous studies had found that carbonyls could be produced by the complex chemical reaction by ozone or other oxidants with some hydrocarbon emissions from the carpets (Weschler et al., 1992) or tobacco smoke (Shaughnessy et al., 2001). Nonetheless, direct emissions of these carbonyls, either from construction materials and furnishings or from human activity and presence, might be the dominant sources.

Indoor materials (such as carpets, textiles, furniture) and household products (such as perfumes, air freshener spray) could introduce carbonyls into the indoor air (Crump and Gardiner, 1989; Zhang et al., 1994b). In the present study, even there were no people in S1 on the first 4 days (Fig. 2 and Table 6), the indoor carbonyl concentrations were still higher than of outdoors (the mean I/O = 6.03), indicating the major sources from indoor materials.

Human activities such as tobacco smoke were considered to be the main source of some carbonyls (Crump and Gardiner, 1989; Shaughnessy et al., 2001). There were hundreds of different volatile organic compounds including carbonyls presented in environment tobacco smoke (Shaughnessy et al., 2001), and the major carbonyl was acetaldehyde, of which the concentration could be 10 times more than that of the other carbonyls including formaldehyde (Crump and Gardiner, 1989). The contribution of smoking to the carbonyls in the ballrooms could be clearly illustrated by our investigation in S1 (Fig. 2). In S1, while there were no

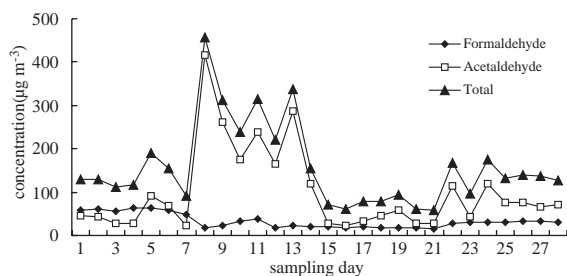


Fig. 2. The variation of indoor formaldehyde, acetaldehyde and the total carbonyl concentrations in indoor of four ballrooms. S1—sampling days: 1–7, the number of people for each day, respectively: 0, 0, 0, 26, 12, 0; S2—sampling days: 8–14, the number of people: 25–30, 16, 8, 10–20, 0, 15–20, 4–14; S3—sampling days: 15–21, the number of people: 15–35, 25–30, 15–31, 50, 56, 12–24, 13; S4—sampling days: 22–28, the number of people: 62, 29, 45, 10–30, 20, 10, 20; Most of the people smoked; for S2, there were still many people uncounted in the independent rooms around.

people on the first 4 days, the acetaldehyde concentrations were lower than formaldehyde. Then on the following 2 days, there were at least 10 smokers there, and the acetaldehyde concentrations increased significantly and were higher than formaldehyde. On the last day, while again there were no smokers, the acetaldehyde concentration dropped again to the same level as the first 4 days. As shown in Fig. 2, for all the four sampling sites, the total carbonyl concentrations had the same trends as acetaldehyde, and the acetaldehyde concentrations increased with the number of smokers. As mentioned above, S2 had the highest I/O ratio (55.94) and highest concentration of acetaldehyde. An explanation of this phenomenon might be due to heavier smoking and/or other unknown sources for the situation in S2.

The HMW carbonyls (heptaldehyde, octylaldehyde, nonanaldehyde, decylaldehyde) were less correlated with formaldehyde and acetaldehyde, indicating different source contributions originated most possibly from biogenetic emissions apart from vehicular exhaust and photochemical reaction. HMW carbonyls such as nonanaldehyde were found to be released by some kind of plants (Ciccioli et al., 1993). There were large areas of trees and grass around the four hotels, especially around S1 and S4 (Fig. 1), which meant that the emission of plants might be the potential sources of these HMW carbonyls detected. So on one hand, these HMW carbonyls might have stronger sources outdoor than indoor; On the other hand, during the night (our sampling time), the elimination of these HMW carbonyls mainly depended on their reaction with OH radical (Atkinson, 2000); the concentration of the OH radical in such a “dirty” indoor air might be higher than that of outdoor air (Sarwara et al., 2002); So the elimination rate of such compounds might be higher in the indoor than that in the outdoor environment. Considering these two factors, it was not surprising that the I/O ratios of these HMW carbonyls were occasionally less than 1.

3.3. Exposure and risk

The exposure (E) for an individual (i) due to intake processes (inhalation and ingestion) can be calculated from the equation of the US EPA (US EPA, 1992):

$$E_i = C_j IR_i t_{ij},$$

where C is the concentration of the pollutant ($\mu\text{g m}^{-3}$), IR is the inhalation rate ($\text{m}^3 \text{h}^{-1}$), t is the exposure time (h day^{-1}), and j the microenvironment.

For gaseous compounds in air, inhalation was considered to be a major exposure route, therefore, IR corresponds to the inhalation rate (Zhang et al., 1994a, b). Indoor inhalation rates were estimated for an average person based upon the exposure factor (US

EPA, 1990). In this study, ballrooms were selected to calculate the exposure (E), of which the exposure time (t) was based upon residence time. For these special places, a mean of 6 h was determined according to the opening time of these ballrooms (from 20.00 p.m. to 2.00 a.m.). The mean and the 95th percentile exposure for indoor formaldehyde and acetaldehyde of the ballrooms (Table 7) were estimated in comparison with some other places. The exposure for formaldehyde and acetaldehyde in the ballrooms in Guangzhou was higher than in the residential and office in Mexico City, known as one of the most polluted cities in the world (Báez et al., 2003).

Formaldehyde has been classified by the EPA Group as B1, probable human carcinogen of medium carcinogenic hazard with an inhalation unit risk estimate of $1.3 \times 10^{-5} (\mu\text{g m}^{-3})^{-1}$ (US EPA, 1991a). Acetaldehyde has been classified as B2, probable human carcinogen of low carcinogenic hazard, with an inhalation unit risk estimate of $2.2 \times 10^{-5} (\mu\text{g m}^{-3})^{-1}$ (US EPA, 1991b). Cancer risks for formaldehyde and acetaldehyde were computed by using the inhalation unit risk, mean concentrations and the highest concentrations (95th percentile) (Table 7). Comparing to another study by Báez et al. (2003), the risk in this study was obviously higher than that in Mexico, even though there were no air-conditioning system in the house and office investigated in the Mexico City. The risk might represent the high-end estimates (increased chance of developing cancer) for an individual who worked in the ballrooms or often went to such places, where the person

continuously breathed air heavily polluted by formaldehyde and acetaldehyde.

Cancer risks for formaldehyde and acetaldehyde should be viewed as preliminary because parameters such as the ventilation rate, time spent outside and inside houses and offices, transportation media, the duration and type of physical activity, e.g. work, rest, and light-to-moderate activity, were not determined, and because of insufficient data. But for our research on ballrooms, the site for dancing and singing, it could be anticipated that people there would achieve a more intensive activity than usual, resulting in higher inhalation rates and higher risks comparing to just calm stay at residential houses or offices.

4. Conclusions

Formaldehyde, acetaldehyde, propionaldehyde, crotonaldehyde, butyraldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, *p*-tolualdehyde, *m/o*-tolualdehyde hexaldehyde, 2,5-dimethylbenzaldehyde, 2-butanone, cyclohexanone, heptaldehyde, octylaldehyde, nonanaldehyde and decylaldehyde were identified and quantified in four hotel ballrooms in Guangzhou during the autumn, 2002. The total concentration of the 18 carbonyls investigated in the outdoor air was $36.72 \pm 21.95 \mu\text{g m}^{-3}$. Formaldehyde was the most abundant compound, accounting for $33.68 \pm 6.79\%$ of the total carbonyls, followed by acetaldehyde, accounting for $18.79 \pm 3.91\%$. But for indoor carbonyls, the

Table 7
The carbonyl concentration (C), exposure (E) and risks of indoor air

| Parameter | Formaldehyde | | | | Acetaldehyde | | | |
|--------------------------------|--------------|-----------------|----------------------|----------------------|--------------|-----------------|-----------------------|----------------------|
| | Arith. mean | 95th percentile | Risk | | Arith. mean | 95th percentile | Risk | |
| | | | Arith. mean | 95th risk | | | Arith. mean | 95th risk |
| <i>Residential^a</i> | | | | | | | | |
| $C (\mu\text{g m}^{-3})$ | 37 | 49 | 4.9×10^{-4} | 6.4×10^{-4} | | | 4.3×10^{-5} | 6.1×10^{-5} |
| $E (\mu\text{g day}^{-1})$ | 236 | 310 | — | — | 122 | 176 | — | — |
| <i>Office^b</i> | | | | | | | | |
| $C (\mu\text{g m}^{-3})$ | 26.2 | 34.4 | 3.4×10^{-4} | 4.5×10^{-4} | 19.3 | 32.7 | 4.2×10^{-5} | 7.2×10^{-5} |
| $E (\mu\text{g day}^{-1})$ | 132 | 173 | — | — | 97 | 165 | — | — |
| <i>Ballroom^c</i> | | | | | | | | |
| $C (\mu\text{g m}^{-3})$ | 33.1 | 55 | 4.4×10^{-4} | 7.2×10^{-4} | 100.1 | 225.7 | 22.6×10^{-5} | 51×10^{-5} |
| $E (\mu\text{g day}^{-1})$ | 124 | 209 | — | — | 378 | 853 | — | — |

The data of ^a, ^b were cited from the study of Báez et al. (2003); The data of ^c was according to this study.

^a Satellite house without air-conditioning system.

^b Office at research center also without air-conditioning system.

^c For exposure calculus, the inhalation rate of air was estimated for an average person ($IR = 0.63 \text{ m}^3 \text{ h}^{-1}$) according to EPA exposure factors (US EPA, 1990). A mean residence time of 8 h (official working time) was considered as exposure time (t) (for 1, 2) and 6 h in our study (the time of opening was 20.00 p.m. to 2.00 a.m.) was used.

most abundant compound was acetaldehyde, accounting for $51.25 \pm 32.60\%$ of the total carbonyls identified ($136.26 \pm 126.74 \mu\text{g m}^{-3}$), followed by formaldehyde ($21.83 \pm 18.33\%$). A wide range of the carbonyl concentrations was observed in both outdoor and indoor air over the 7-day sampling period.

The average C_1/C_2 and C_2/C_3 ratios were about 1.81 and 6.16, respectively. Indoor carbonyl compounds concentrations were generally found to be higher than that of outdoors. The indoor acetaldehyde concentrations had a strong relationship with smoking. For example, the much higher concentration of indoor acetaldehyde might mainly derive from smoking. The outdoor carbonyls (with the exception of HMW carbonyls) had good correlations with formaldehyde and acetaldehyde, indicating a principal source of vehicular emissions. The sources of some HMW carbonyls were different from that of low molecular ones, resulting in the unusual I/O ratios. It was found that, especially for carbonyls, the contamination in the ballrooms was more serious than that reported in the common residential houses and offices.

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References

- Atkinson, R., 2000. Atmospheric chemistry of VOCs and NO_x . *Atmospheric Environment* 34, 2063–2101.
- Báez, A.P., Belmont, R., Padilla, H., 1995. Measurement of formaldehyde and acetaldehyde in the atmosphere of Mexico City. *Environmental Pollution* 86, 166–187.
- Báez, A.P., Padilla, H., Cervantes, J., Pereyra, D., Torres, M.C., Garcia, R., Belmont, R., 2001. Preliminary study of the determination of ambient carbonyls in Xalapa City, Veracruz, Mexico. *Atmospheric Environment* 35, 1813–1819.
- Báez, A.P., Padilla, H., Garcia, R., Torres, M.C., Rosas, I., Belmont, R., 2003. Carbonyl levels in indoor and outdoor air in Mexico City and Xalapa, Mexico. *The Science of the Total Environment* 302, 211–226.
- Brickus, L.S.R., Cardoso, J.N., De aquino neto, F.R., 1998. Distributions of indoor and outdoor air pollutants in Rio de Janeiro, Brazil: implications to indoor air quality in bayside offices. *Environmental Science and Technology* 22, 3485–3490.
- California Air Resources Board (CARB), 1999. California ambient air quality data 1990–1998. Planning and Technical Support Division, Los Angeles, USA.
- Carlier, P., Hannachi, H., Mouvier, G., 1986. The chemistry of carbonyl compounds in the atmosphere—A review. *Atmospheric Environment* 20, 2079–2099.
- Carter, W.P.L., 1995. Computer modeling of environmental chamber studies of maximum incremental reactivities of volatile organic compounds. *Atmospheric Environment* 29, 2513–2527.
- Christensen, C.S., Skov, H., Nielsen, T., Lohse, C., 2000. Temporal variation of carbonyl compound concentrations at a semi-rural site in Denmark. *Atmospheric Environment* 34, 287–296.
- Ciccioli, P., Brancaleoni, E., Frattoni, M., Cecinato, A., Brachetti, A., 1993. Ubiquitous occurrence of semi-volatile carbonyl compounds in tropospheric samples and their possible source. *Atmospheric Environment* 27A, 1891–1901.
- Crump, D.R., Gardiner, D., 1989. Sources and concentrations of aldehydes and ketones in indoor environments in the UK. *Environmental International* 15, 455–462.
- Druzik, C.M., Grosjean, D., Van Neste, A., Parmar, S.S., 1990. Sampling of ambient carbonyls with DNPH-coated cartridges and liquid chromatography analysis with diode array detection. *International Journal of Environmental Analytical Chemistry* 38, 495–512.
- Granby, K., Christensen, C.S., 1997. Urban and semi-rural observations of carboxylic acids and carbonyls. *Atmospheric Environment* 31, 1403–1415.
- Grosjean, D., 1982. Formaldehyde and other carbonyls in Los Angeles ambient air. *Environmental Science and Technology* 16, 254–262.
- Grosjean, D., 1988. Aldehydes, carboxylic acids and inorganic nitrate during NSMCS. *Atmospheric Environment* 22, 1637–1648.
- Grosjean, D., Williams, E.L., Grosjean, E., 1993a. Peroxyacyl nitrates at southern California Mountain forest locations. *Environmental Science and Technology* 27, 110–121.
- Grosjean, E., Williams II, E.L., Grosjean, D., 1993b. Ambient levels of formaldehyde and acetaldehyde in Atlanta, Georgia. *Journal of Air and Waste Management Association* 43, 469–474.
- Grosjean, E., Grosjean, D., Fraser, M.P., Cass, G.R., 1996. Air quality model evaluation data for organics. 3. peroxyacetyl nitrate and peroxypropionyl nitrate in Los Angeles air. *Environmental Science and Technology* 30, 2704–2714.
- Grosjean, D., Grosjean, E., Moreira, L.F.R., 2002. Speciated ambient carbonyls in Rio de Janeiro, Brazil. *Environmental Science and Technology* 36, 1389–1395.
- Ho, K.F., Lee, S.C., Louie, P.K.K., Zou, S.C., 2002. Seasonal variation of carbonyls compound concentrations in urban area of Hong Kong. *Atmospheric Environment* 36, 1259–1265.
- Kelly, T.J., Smith, D.L., Satola, J., 1999. Emission rates of formaldehyde form materials and consumer products found in California homes. *Environmental Science and Technology* 33, 81–88.
- Khoder, M.I., Shakour, S.A., Farag, S.A., Hameed, A.A.A., 2000. Indoor and outdoor formaldehyde concentrations in homes in residential areas in Greater Cairo. *Journal of Environmental Monitor* 2, 123–126.
- Levin, J.O., Anderson, K., Lindahl, R., Nilsson, C.A., 1985. Determination of sub-part-per-million levels of formaldehyde in air using active or passive sampling on

- 2,4-dinitrophenylhydrazine-coated glass fiber filters and high-performance liquid chromatography. *Analytical Chemistry* 57, 1032–1035.
- Löfroth, G., Burton, R.M., Forehand, L., Hammond, S.K., Seila, R.L., Zvejinger, R.B., Lewtas, J., 1989. Characterization of environmental tobacco smoke. *Environmental Science and Technology* 23, 610–614.
- Miguel, A.H., De aquino neto, F.R., Cardoso, J.N., Vasconcellos, P.D.C., Pereira, A.S., Marquez, K.S.G., 1995. Characterization of indoor air quality in the cities of São Paulo and Rio de Janeiro, Brazil. *Environmental Science and Technology* 29, 338–345.
- Montero, L., Vasconcellos, P.C., Souza, S.R., Pires, M.A.F., Sanchez-Ccoylo, O.R., Andrade, M.F., Carvalho, L.R.F., 2001. Measurements of atmospheric carboxylic acids and carbonyl compounds in São Paulo City, Brazil. *Environmental Science and Technology* 35, 3071–3081.
- Morrison, G.C., Nazaroff, W.W., 2002. Ozone interactions with carpet: secondary emissions of aldehydes. *Environmental Science and Technology* 36, 2185–2192.
- Nguyen, H.T., Takenaka, N., Bandow, H., Maeda, Y., Oliva, S.T.D., Botelho, M.M.F., Tavares, T.M., 2001. Atmospheric alcohols and aldehydes concentrations measured in Osaka, Japan and in Sao Paulo, Brazil. *Atmospheric Environment* 35, 3038–3057.
- Possanzini, M., Di Palo, V., Petricca, R., Fratarcangeli, R., Brocco, D., 1996. Measurements of lower carbonyls in Rome ambient air. *Atmospheric Environment* 30, 3757–3764.
- Reiss, R., Ryan, P.B., Tibbatts, S.J., Koutrakis, P., 1995. Measurement of organic acids, aldehydes, and ketones in residential environments and their relation to ozone. *Journal of Air and Waste Management Association* 45, 811–822.
- Sarwara, G., Corsia, R., Kimuraa, Y., Allena, D., Weschler, C.J., 2002. Hydroxyl radicals in indoor environments. *Atmospheric Environment* 36, 3973–3988.
- Satsumabayashi, H., Kurita, H., Chang, Y.S., Carmichael, G.R., Ueda, H., 1995. Photochemical formations of lower aldehydes and lower fatty acids under long-range transport in central Japan. *Atmospheric Environment* 29, 255–266.
- Shaughnessy, R.J., Mcdaniels, T.J., Weschler, C.J., 2001. Indoor chemistry: ozone and volatile organic compounds found in tobacco smoke. *Environmental Science and Technology* 35, 2758–2764.
- Shepson, P.B., Kleindeinst, T.E., Edney, E.O., Nero, C.M., Cupitt, L.T., Claxton, L.D., 1986. Acetaldehyde: the mutagenic activity of its photooxidation products. *Environmental Science and Technology* 20, 1008–1012.
- Shepson, P.B., Hastie, D.R., Schiff, H.I., Polizzi, M., Bottenheim, J.W., Anlauf, K., Mackay, G.I., Karecki, D.R., 1991. Atmospheric concentrations and temporal variations of C1–C3 carbonyl compounds at two rural sites in central Ontario. *Atmospheric Environment* 25A, 2001.
- Sin, D.W.M., Wong, Y., Louie, P.K.K., 2001. Trends of ambient carbonyl compounds in the urban environment of Hong Kong. *Atmospheric Environment* 35, 5961–5969.
- Sirju, A., Shepson, P.B., 1995. Laboratory and field investigation of the DNPH cartridge technique for the measurement of atmospheric carbonyl compounds. *Environmental Science and Technology* 29, 384–392.
- Slemr, J., Junkermann, W., 1996. Temporal variations in formaldehyde, acetaldehyde and acetone and budget of formaldehyde at a rural site in southern Germany. *Atmospheric Environment* 30, 3667–3676.
- Szilagi, I., Demeter, A., Turanyi, T., Berces, T., 1991. Non-methane hydrocarbon and aldehyde measurements in Budapest, Hungary. *Atmospheric Environment* 25A, 2103–2110.
- Tanner, R.L., Meng, Z., 1984. Seasonal variations in ambient atmospheric levels of formaldehyde and acetaldehyde. *Environmental Science and Technology* 18, 723–726.
- US Environmental Protection Agency (US EPA), 1990. *Exposure Factors Handbook*. Office of Health and Environmental Assessment, Washington, DC, EPA 600y8-89y043.
- US Environmental Protection Agency (US EPA), 1991a. Integrated risk information system (IRIS) on acetaldehyde. On-line IRIS, CASRN 75-07-0.
- US Environmental Protection Agency (US EPA), 1991b. Integrated risk information system (IRIS) on formaldehyde. On-line IRIS, CASRN 50-00-0.
- US Environmental Protection Agency (US EPA), 1992. *Guidelines for Exposure Assessment*. Office of Research Development, Office of Health and Environmental Assessment, Washington, DC, EPA 600Z-92y001.
- US Environmental Protection Agency (US EPA), 1999. *Compendium method to-11A. Determination of formaldehyde in ambient air using adsorbent cartridge followed by high performance liquid chromatography (HPLC)*, [Active Sampling Methodology] [S].
- Viskari, E., Vartiainen, M., Pasanen, P., 2000. Seasonal and diurnal variation in formaldehyde and acetaldehyde concentrations along a highway in Eastern Finland. *Atmospheric Environment* 34, 917–923.
- Weschler, C.J., Hodgson, A.T., Wooley, J.D., 1992. Indoor chemistry: ozone, volatile organic compounds, and carpets. *Environmental Science and Technology* 26, 2371–2377.
- Williams, I.D., Revitt, D.M., Hamilton, R.S., 1996. A comparison of carbonyl compound concentrations at urban roadside and indoor sites. *The Science of the Total Environment* 189/190, 475–483.
- Zhang, J., He, Q., Liou Paul, J., 1994a. Characteristics of aldehydes: concentrations, sources, and exposures for indoor and outdoor residential microenvironments. *Environmental Science and Technology* 28, 146–152.
- Zhang, J., Wilson, W.E., Liou, P.J., 1994b. Indoor air chemistry: formation of organic acids and aldehydes. *Environmental Science and Technology* 28, 1975–1982.