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Spatiotemporal distribution of carbonyl compounds in China

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ABSTRACT

A sampling campaign was carried out at nine Chinese cities in 2010/2011. Fifteen monocarbonyls (C# = 1–9) were quantified. Temperature is the rate-determining factor of the summertime carbonyl levels. The carbonyl emissions in winter are mainly driven by the primary anthropogenic sources like automobile. A molar ratio of propionaldehyde to nonaldehyde is a barometer of the impact of atmospheric vegetation emission which suggesting that strong vegetation emissions exist in summer and high propionaldehyde abundance is caused by fossil fuel combustion in winter. Potential health risk assessment of formaldehyde and acetaldehyde was conducted and the highest cumulative risks were observed at Chengdu in summer and Wuhan in winter. Because of the strong photochemical reaction and large amount of anthropogenic emissions, high concentrations of carbonyl compounds were observed in Chengdu. The use of ethanol-blended gasoline in Wuhan is the key reason of acetaldehyde emission and action should be taken to avoid potential health risks.

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1. Introduction

Carbonyl compounds originate from both primary and secondary origin. The compounds can be emitted either directly from incomplete combustion of biomass and fossil fuel or formed indirectly from the photochemical oxidations of volatile organic compounds (VOCs) in both of natural and anthropogenic sources (Possanzini et al., 2002). Industrial emissions and vehicular exhausts were identified to be the primary sources of carbonyl compounds in urban areas (Ho et al., 2007, 2013). Increasing evidences suggest that cooking fumes (e.g. meat cooking or cooking oils) can release large amounts of high molecular weight carbonyls (Feng et al., 2005; Schauer et al., 2002). Other studies reported that aldehydes generate from secondary emission can be more than from primary emission (Altshuller, 1993). The lifetimes of carbonyls

in troposphere are different due to their various physicochemical properties and the production/removal pathways (DeMore et al., 1992). A long range transport of carbonyls and their precursors can lead to enhanced photochemical pollution in the metropolitan areas (Possanzini et al., 2002).

Several carbonyls have been recognized for their adverse human health effects due to their potential carcinogenic and mutagenic properties (CEPA, 1993; NCR, 1981; WHO, 1987), in addition of their capability of forming toxic and phytotoxic radical intermediates or stable species (Roberts, 1990). Specific carbonyls induced adverse human health effect symptoms are eyes and lung irritations (WHO, 2000). Formaldehyde is classified as Group 1 human carcinogen by the International Agency for Research on Cancer (IARC) for its carcinogenicity (IARC, 2006) and acetaldehyde is a suspected carcinogen (Baez et al., 2003; Zhang et al., 1994). Formaldehyde can cause nasopharyngeal cancer (IARC, 2004) and is suggested to be potentially associated with leukemia (Zhang et al., 2009). World-wide scientific community has been focusing on pollution level, potential sources and human exposure of carbonyls in indoor and outdoor environment ever since (Baez et al., 2003; Cavalcante et al.,

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2006; Feng et al., 2004; Gilbert et al., 2006; Marchand et al., 2006; Zhang et al., 1994).

Airborne carbonyls have been investigated at urban areas in China (Cheng et al., 2014; Ho et al., 2014; Liu et al., 2013; Louie et al., 2013; Weng et al., 2009), nonetheless only limited studies had been conducted in middle and western regions despite of rapid urbanization and industrialization in these regions. There are also lack of information about high molecular weight carbonyls (HMW carbonyls, $C\#>6$) in urban atmosphere (Chi et al., 2007; Huang et al., 2008; Wang et al., 2010). Emission from vegetation can be one of possible sources induced by ozone exposure of plants (Karl et al., 2005). Biomass, biofuel and charcoal combustion are other sources of HMW carbonyls in air (Andreae and Merlet, 2001). In the densely populated urban areas, higher HMW carbonyls emissions would be expected due to the increased consumption of fuels and large emission from industries which seriously hamper air quality in urban areas.

The aim of this study is to quantify ambient monocarbonyls and characterize their seasonal and spatial variations in urban and rural areas of China. The inhalation cancer risk of carbonyls is used to compare the potential health impact of carbonyl among different Chinese cities.

2. Experimental

A national wide survey of ambient monocarbonyl compounds were conducted simultaneously in nine sites during summer and winter season. Fifteen carbonyls in ambient air were sampled using standard 2,4-dinitrophenylhydrazine (DNPH) derivatization method followed by high-performance liquid chromatography (HPLC) analysis (USEPA, 1999) to elucidate the condition of airborne carbonyl pollution in China.

2.1. Sampling site

Nine sampling sites in different regions (covering an area of 23–39°N, 91–121°E) of China were selected to represent economically-developed and economically-developing cities. The seven urban sampling sites include Beijing [BJ], Chengdu [CD], Guangzhou [GZ], Shanghai [SH], Wuhan [WH], Xiamen [XM], and Yantai [YT] while the two rural sites were chosen to be Qinghai Lake, Qinghai [QH] and Lasa, Tibet [TB]. The details of sampling

locations are described in Table 1. Samplers were located on rooftops at ~10–20 m above ground level for two weeks during summer (August 2010) and winter (January 2011), respectively.

2.2. Sample collection

A 24-h interval integrated samples (from 10:00 a.m. onwards) were collected daily in this study. Air samples were collected in silica cartridges impregnated with acidified 2,4-dinitrophenylhydrazine (DNPH) (Sep-Pak DNPH-silica, 55–105 μm particle size, 125 Å pore size; Waters Corporation, Milford, MA) at flow rate of 0.7 L min^{-1} . Total 127 and 130 samples were collected in summer and winter, respectively. Collection efficiencies were measured under different field conditions by sampling carbonyls into two identical cartridges connected in series. They were calculated as 100% $(1 - A_b/A_f)$, where A_f and A_b were the amounts of carbonyl collected on the front and the back sampling tubes, respectively. No detectable breakthrough was found at these sampling flow rates and sampling times. The flow rates were checked at the field from start and end of each sampling period using a calibrated flow meter (Gilibrator Calibrator; Gilian Instruments, W. Caldwell, NJ). A Teflon filter assembly (Whatman, Clifton, NJ) with an ozone scrubber (Sep-Pak; Waters Corporation) was installed in front of the DNPH-silica cartridge in order to remove any particulate matters and prevent possible disturbance from ozone (Spaulding et al., 1999). Collocated samples were collected to examine the collection reproducibility (>95%) in the field for all of the samplers. One cartridge was reserved as a field blank on each sampling trip and was handled in same manner as the other sample cartridges. The amounts of carbonyls detected in the cartridges were corrected for field blank under air concentrations of the carbonyls analysis. The DNPH-coated cartridges were stored in a refrigerator at <4 °C after sampling and before analysis. Different meteorological parameters such as temperature, air pressure, relative humidity (RH), rainfall, and wind speed and direction were recorded during the sampling period.

2.3. Analytical methods

A total of 15 carbonyls were quantified, including formaldehyde (C1), acetaldehyde (C2), propionaldehyde (C3), *iso* + *n*-butyraldehyde (*iso* + *n*C4), benzaldehyde (*benz*), *iso*-valeraldehyde (*iso*-C5), *n*-valeraldehyde (*n*C5), *o*-tolualdehyde (*o*-tol), *m*-tolualdehyde (*m*-tol), *p*-tolualdehyde (*p*-tol), hexaldehyde (C6), 2,5-dimethylbenzaldehyde (2,5-DB), heptaldehyde (C7), octaldehyde (C8) and nonaldehyde (C9). Unsaturated carbonyls including acrolein and crotonaldehyde were detected but not reported in the study. The unsaturated carbonyl DNP-hydrazones could react with excess reagent to form adducts, leading to ambiguity in accurate quantification due to chromatographic interferences (e.g. double peaks) and response factor issues (Ho et al., 2011; Schulte-Ladbeck et al., 2001). In-laboratory experiments demonstrated that the collection efficiencies were >93 ± 5% for all target carbonyls under the same flow rate, temperature and RH.

Each DNPH-coated cartridge was eluted with 2.0 mL acetone-free acetonitrile (HPLC/GCMS grade, J&K Scientific Ltd., Ontario, Canada) solution and transferred to a volumetric flask. Previous studies demonstrated that neither DNPH nor DNPH derivatives remained in the cartridge after 2.0 mL acetone-free acetonitrile solution elution (Ho et al., 2007). Certified calibration standards of the monocarbonyl DNP-hydrazones were purchased from Supelco (Bellefonte, PA) and diluted in concentration ranges of 15–3000 $\mu\text{g mL}^{-1}$. The calibration solutions were allowed to stand at room temperature for 6 h for complete derivatization. The final volume of each calibration solution was filled up to 2.0 mL with 8:2

Table 1
Sampling site description in nine cities.

City	Population (million)	Description	Latitude	Longitude	Height ^a (m)
Beijing	19.6	Capital of China	39°58' N	116°23' E	14
Chengdu	14.0	Continental & industrial	30°39' N	104°1' E	15
Guangzhou	12.7	Industrial & commercial	23°7' N	113°21' E	10
Shanghai	23.0	Industrial & commercial	31°18' N	121°30' E	20
Wuhan	9.8	Industrial & commercial	30°31' N	114°21' E	10
Xiamen	3.5	Coastal & commercial	24°28' N	118°5' E	8
Yantai	7.0	Coastal & commercial	37°28' N	121°26' E	10
Lasa, Tibet	0.6	Continental & plateau	29°38' N	91°2' E	15
Qinghai Lake	0.1	Rural area & plateau	36°58' N	99°53' E	12

^a Height of air inlet of the sampler above the ground.

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