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## Original Research Article

## Spatial and temporal analysis of ambient carbonyls in a densely populated basin area of central Taiwan

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## ABSTRACT

The spatial and temporal distributions of ambient carbonyls during the southwest and northeast monsoon seasons in a densely populated basin area were analyzed. The results show that the major carbonyls in ambient were ketones during the both monsoon seasons. The concentrations of carbonyls at daytime and nighttime were in the order: urban > rural > suburban and urban > suburban > rural areas, respectively. The higher carbonyl concentrations occurred in urban area all day, but maximum variation of carbonyl concentrations occurred in rural area. The orders of carbonyl concentrations at daytime and nighttime were irregular during the northeast monsoon season; the variations of carbonyl concentrations in the area were all low. The effects of urban heat island phenomena on the area were negligible, especially during the northeast monsoon season. The ranges of average ratio (northeast to southwest monsoon seasons) of total carbonyl concentrations in urban, suburban, rural areas were 1.0–1.6, 1.5–3.5, and 1.8–2.4, respectively. Because the clear results were obtained during the both monsoons that ambient carbonyls increased with increasing volatile organic compound (VOC) concentrations, the simulation results of VOCs by CMB8.2 model were used to understand the contribution of carbonyls from the primary sources. The order of VOCs and carbonyls source contributions were vehicle exhaust > hi-tech industry > traditional industry > ship and gasoline stations during the both monsoon seasons. Therefore, in order to effectively reduce the carbonyls in ambient air, pollution from vehicle and hi-tech industry must be managed and controlled.

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## 1. Introduction

Carbonyl compounds in ambient air are well known for their adverse effects on human health, especially on eye and lung irritations [1,2]. A large amount of data has implicated reactive carbonyls as neurotoxic mediators of oxidative damage in the progression of Alzheimer's disease and other neurodegenerative diseases [3]. Exposure of red blood cells to carbonyl compounds leading to Maillard reaction. It caused a marked decrease in red blood cell deformability [4]. Several studies have indicated that carbonyl emissions from cook stoves when the combustion is incomplete [5,6]. Kabir et al. [7] quantified the emission concentrations of a number of carbonyl compounds during the combustion of commonly used barbecue charcoal. Kim et al. [8] quantified

the emission concentrations of 13 carbonyl compounds released directly from various anthropogenic source processes in a large industrial complex. Researchers have also identified motor vehicles as the leading sources of carbonyls emission in non-industrial area [9–11].

The gas-phase reaction of ozone with alkenes is of critical importance in atmospheric chemistry. It leads several products including carbonyls [12]. Most secondary production of formaldehyde is expected to occur during the atmospheric oxidation of alkenes. Additionally, it is formed more slowly from both the oxidation of alkanes and aromatic compounds [13,14].

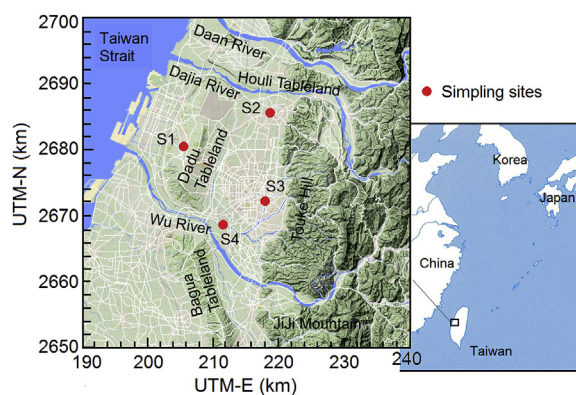
The topography of Taichung area (about 2500 km<sup>2</sup>) is complex, which includes coast, tablelands, basin, hills, and mountains (Fig. 1). At the end of 2012, approximately 2.3 million and 0.9 million peoples lived in the basin and coastal areas, respectively. A densely populated area is located within the basin center, with surrounded by rural area. Additionally, topographical and meteorological factors, such as basin and monsoons, can drastically affect

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**Fig. 1.** Location of sampling sites in Taichung Basin. The Taichung Basin faces south toward the Wu River, north toward the Dajia River, East toward the Touke Hill, West toward the Dadu Tableland, Southwest toward the Bagua Tableland, and Southeast toward the Jiji Mountain.

levels of carbonyls and volatile organic compounds (VOCs) in ambient air of the area. The weather conditions in the area are different. For example, urban heat island effect occurs in the basin area; the clear monsoon effect takes place in the coastal area. Also, a city with a million people would cause a 4.4 °C warming [15]; urban heat island effect in this basin with intensive and frequent occurrence resulting in more pollutant accumulation in the basin. Therefore, characterizing the variation of ambient air pollutants, especially carbonyls and VOCs, with sources, monsoons, and terrain in the target area is crucial.

In some previous studies, the measurement of ambient carbonyls based on 2,4-dinitrophenylhydrazine (DNPH) or O-(2,3,4,5,6-pentafluorobenzyl)hydroxylamine (PFBHA) derivatisations is utilized. It is followed by separation and detection of the hydrazones with high-performance liquid chromatography/UV–Vis absorption (HPLC/UV–V) or gas chromatography/mass spectrometry (GC/MS) [16–19]. The procedures of these methods are complicated and these methods are unable to measure ambient VOCs at the same time. In present study, US EPA TO-15 Method was employed to measure ambient carbonyls and VOCs. This method determines VOCs in air collected in specially-prepared canisters, analyzed by GC/MS, which applies to ambient concentrations of VOCs above 0.5 ppbv and typically requires VOC enrichment by concentrating up to one liter of a sample volume. The sample air was collected and stored in canisters with subsequent GC/MS analysis. It is due to the fact that  $\alpha,\beta$ -unsaturated carbonyls are unstable when collected in electropolished canisters [20]. Therefore in this study, some specially prepared (without humidification) silcosteel canisters with air sampling kites were selected for collecting ambient air samples to avoid this problem.

In previous studies, several approaches have been applied for understanding the source apportionment of carbonyls. The multi-linear regression (MLR) method [21,22] and positive matrix factorization (PMF) model [23] are two commonly used techniques. The MLR method separates primary and secondary carbonyls based on their correlations with markers for primary emission (e.g., CO, ethyne) and secondary production (e.g., O<sub>3</sub>, glyoxal). The PMF technique is used to derive a set of source composition profiles. Each identifies a mix of compounds associated with a particular category of emissions (e.g., on-road emissions, solvent manufacturing) [24]. However, the MLR method heavily depends on the selection of markers, and improper markers may bias source apportionment results [23]. The uncertainty from rotational ambiguity in the PMF solutions could affect the species loadings in the source profile matrix [25]. Thus, this study uses bimestrial field

measurements and the chemical mass balance model (CMB8.2) to analyze VOC emissions and further understanding of carbonyl emissions from various possible sources in the area. For the simulation of CMB8.2 model, 10 source profiles of VOC emissions were established in this study.

## 2. Materials and methods

### 2.1. Field measurements

Fig. 1 shows the map of Taichung Basin with sampling sites. Taichung Basin faces south toward the Wu River; north toward the Dajia River; east toward the Touke Hill; west toward the Dadu Tableland; southwest toward the Bagua Tableland; and southeast toward the Jiji Mountain. Four sampling sites were selected to measure the ambient concentration of carbonyls. Sampling site S1 was chosen for its location outside the basin. In the basin, the sampling sites S2 and S4 were selected to follow the pollutant transport in which there were two passes channeling air masses in the basin. Sampling site S3 was selected because of its proximity to a prevailing strong heat island effect at the center of basin [26,27].

Ho et al. [28] pointed out that no photochemical reactions occur between 06:00 and 09:00 (early morning); the period of less photochemical reactions between 09:00 and 12:00 (morning); the ground-level atmosphere mixes rapidly increasing photochemical reactions, causing VOC fraction variations between 12:00 and 15:00 (noon); the later period of photochemical reactions between 15:00 and 18:00 (afternoon); and the period after photochemical reaction between 18:00 and 21:00 (evening). In this study, bimestrial samples from May 2011 to February 2012 were collected simultaneously at sites S1–S4 for 3 d (72 h). The samples were taken four times per day for the periods of 4 h: 08:00–12:00 (morning), 12:00–16:00 (afternoon), 16:00–20:00 (evening), and 20:00–24:00 (night).

The ambient air samples at four sampling sites (S1–S4) were simultaneously collected following the US EPA Compendium Method TO-15. A specially prepared (without humidification) in vacuum silcosteel canisters (RESTEK SilcoCan cat. no. 24114) was used with air sampling kites (RESTEK Veriflo SC423XL flow controllers). After sampling, the ambient carbonyls and VOC samples were analyzed by GC/MS (Shimadzu QP-5050A) with thermal-adsorption equipment (ENTECH 7100A Preconcentrator) according to US EPA Method TO-15. A 0.25 mm Chrompack DB-1 60 m-length capillary column was employed in the GC/MS. 102 standard curves spectrum gases were calibrated using the standard gases in U.S. EPA Method TO-14A calibration mix (RESTEK cat. no. 34400), Method TO-15 ozone precursor mixtures (RESTEK cat. no. 34420), and prepared standard gases. The detection limits of VOC concentrations were in the  $\mu\text{g m}^{-3}$  range.

The equipment blanks were analyzed in a laboratory with high purity nitrogen. The results were all lower than 2 times of the method detection limit. Because these standard curves did not contain all compounds in ambient air, a method of relative response factor (RRF) was used to quantitatively determine compounds for each non-standard curve in the air sample. In addition, the experiments were conducted on 102 standard gases. Also, a number of pure statistical RRF equations were proposed to estimate the concentrations of other alkanes, alkenes, aromatics, carbonyls, alcohols, and chlorohydrocarbons. These equations depend on a number of the VOC properties. The most important of which are the molecular weight, number of double bond, boiling point, number of branched chain, number of carbon, number of chlorine, stereo structure, and positions of function group. These RRF equations are as follows:

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