Assessing impacts of factors on carbonyl compounds emissions produced from several typical Chinese cooking

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ABSTRACT

Carbonyl compounds are important constituents of photochemical smog and important precursors of photochemical smog formation. In order to investigate concentrations and types of carbonyls generated during cooking process, the influence of cooking styles, food materials, oil types and purification facilities were investigated. Silica cartridge impregnated with 2,4-dinitrophenylhydrazine (2,4-DNPH) and high performance liquid chromatography (HPLC) were adopted for collecting and analyzing samples. Results showed that the emission factors (EFs) of carbonyls (C1-C8) produced from cooking emissions varied within the range of 0.669—1.596 µg/kg. In relation to cooking styles, Barbecue (1.596 µg/kg) > Frying (1.530 µg/kg) > Teppanyaki (1.229 µg/kg) > Stir-frying (0.699 µg/kg), while C1-C3 carbonyls accounted for over 85% for the carbonyls concentrations. Regarding the food materials, generally, carbonyl emissions from meat dishes were greater than for non-meat dishes. Among different oil types used, cooking with sunflower oil resulted in the highest carbonyl emissions. In addition, carbonyls produced from frying carried the highest health risk. The high-voltage electrostatic methodology applied to remove carbonyls did not show as efficient for this purpose. Formaldehyde was found to be the largest contributor to ozone formation potential (OFP).

1. Introduction

Carbonyl compounds are a group of chemically active volatile organic compounds (VOCs) involved in many atmospheric, photochemical reactions, such as ozone and peroxyacetyl nitrate (PANs) formation [1]. Carbonyls are of critical importance as products of photo-oxidation from gas-phase hydrocarbons, which are a major source of free radicals and the precursors to organic-aerosol formation in urban air. In addition, carbonyls have received increasing attention for their adverse health effects of irritants and carcinogens [2—5].

Cooking fume is one of the major sources of carbonyl compounds [6]. On one hand, cooking fuels, including charcoal and wood, could produce carbonyls during combustion [3,7]. On the other hand, pyrolysis and hydrolysis of edible oils and food materials at high temperatures can also produce carbonyls [8]. However, GB/T18883-2002 of China standard for indoor air only regulates the emitted concentrations of formaldehyde (0.10 mg/m^3.h), and not for other compounds present.

There are two main analytical methodologies used for sampling and subsequent analysis of carbonyls: One includes collection on dinitrophenylhydrazine (DNPH) silica cartridges, followed by a high performance liquid chromatography (HPLC) used for analysis. The other one is using pentafluorobenzyl hydroxylamine (PFBHA) as a derivative of the adsorption column for sampling, and then gas chromatography-mass spectrometer (GC-MS) [9,10]. Current studies were mainly conducted in real restaurants [2,11—13]. It was found that fast food restaurants contribute the most to the total carbonyl emissions, whereas deep-fry cooking was the main contributor. Cooking with rapeseed salad oil produced the largest

amount of carbonyls in comparison to other oil types. Few studies evaluated factors influencing levels of carbonyl emission in the laboratory, where the amount of food and oil, as well as ventilation conditions were controlled during cooking process.

This study conducted lab measurements to investigate carbonyl concentrations emitted from several typical Chinese cooking dishes using the most common food material and oil types being impacted by factors including cooking styles, oil types, food materials, and purification facilities. In addition, we investigated the role carbonyls have on the environment. Its ozone formation potential (OFP) was investigated, which was further used to assess the impact of VOCs on ozone formation for indoor and outdoor air [14]. Health impact of exposure to carbonyls produced during cooking was also assessed.

2. Methodology

2.1. Sample collections and analysis

The amount of food material (5 kg), oil consumption (200 ml), oil temperature (260 °C) and cooking time (1 h) were controlled strictly. Surface water content was maintained constant for each test by spraying the same amount of water after drying of food materials. The measurement setup is shown as Fig. 1.

When the cooking starts, cooking fumes pass through the purification system and the sampling is done on a silica cartridges impregnated with 2,4 DNPH (WATERS Sep-Pak DNPH-Silica). TO-11 method was adopted to collect and analyze carbonyls from cooking fume (US EPA) [15]. Teflon filter was set in front to remove particulate matter. The gas flowrate was 0.7 L/min, and the sampling time was for about one hour. An ozone scrubber was connected to the inlet of the 2,4-DNPH silica cartridge to prevent interference from ozone. All sampled cartridges were stored in laboratory refrigerators before analyzing 2,4-DNPH silica cartridge with high performance liquid chromatography (HPLC, Agilent 1260; Column: Zorbax RRHD Eclipse Plus C18 (2.1 × 100 mm, 1.8 μm)). Each 2,4-DNPH silica cartridge was eluted with 5.0 mL acetonitrile (HPLC grade) solution and transferred to a 2 mL volumetric flask before analyzing. The injection volume \was 18 μl. Mobile phase consisted of acetonitrile and water in the ratio 58%: 42%. The flow rate was 0.9 mL/min. The wavelength used was 365 nm and 430 nm.

Detailed information on the measurement procedure is contained in Table 1. Four factors were considered to be important for carbonyls emissions: cooking style (including barbecue, teppanyaki, frying and stir-frying); food materials (vegetables, meat and aquatic products) oil types (soybean oil, corn oil and sunflower oil) and the purification equipment (on/off). Control test was conducted by sampling carbonyls from the ambient air. In addition, the fuel we used was from the same batch with same quality, which were mixed evenly, and fully burned after 2 min, then the measurement started. For each cooking, we weighted all the ingredient and materials before the measurement. All the ingredient and

materials were taken from the same batch, respectively. And also the speed and frequency of the stir-frying process were controlled by a timer. Furthermore, we repeated three times in the same condition.

A high-voltage electrostatic hood (BF-JD-4, Shenzhen ENT Corporation, China Shenzhen, air volume = 20 m³/min) were used for removing cooking emissions and cleaning room. It makes oil mist charged by the high-voltage electrode, and collected on the plate. Its purification efficiency is beyond 90% as claimed by the producer, and practical and no secondary pollution.

2.2. Emission factor calculation

Emission factors were calculated by multiplying the average concentration (C) by the cooking time (t), the flow from the metal container (F), the dilution ratio (DR), and dividing by the amount of food used (M_{Food}) [16].

$$EF = \frac{C \times DR \times t \times F}{M_{Food}} \quad (1)$$

where, C (μg/m³) is the average emission, DR is the dilution ratio, t (h) is the cooking time, F (m³/h) is flow rate, M_{Food} (kg) is the amount of food.

2.3. Health risk assessment and OFP

In this study, cancer risks were assessed by assuming inhalation of carbonyls produced by cooking emissions and calculated using US EPA (2011) guidelines as follows:

$$CR = slope\ factor \times LADD \quad (2)$$

$$LADD = \frac{C \times IR \times AF \times ED}{BW \times AT} \quad (3)$$

where $LADD$ (mg^{-1} kg^{-1} day^{-1}) is the lifetime average daily dose, C(mg/m³) is the pollutant concentration, IR(m³/day) is the intake rate, AF (%) is the absorption fraction, ED (day) is the exposure duration, BW(kg) is the body weight, and AT (day) is the average time days. The values of input parameters were IR = 18 m³/day, AF = 100%, ED = 1.7 h × 24 h^{-1} × 5 day/week × 52.14 week/year × 5 year = 92.34 day, BW = 64.56 kg, and AT = 27,959 days (=76.6 year) [17]. Total cooking time for three meals per day was 1.7 h in this study [17]. The slope factor in Eq. (2) was determined from reference dose (RFD, mg^{-1} kg^{-1} day^{-1}) and according to the Integrated Risk Information System [18]. Note that only formaldehyde and acetaldehyde are carcinogenic among these substances. The CR value below 10^{-6} has been set as to be acceptable or tolerable for regulating purposes [19].

The OFP of an individual carbonyl compounds can be calculated as multiplying the emission by its corresponding maximum increment reactivity (MIR) factor [20]. In this study, the total OFP is the summation of the OFPs corresponding to all carbonyl compounds.

3. Results and discussion

3.1. Carbonyls categories classified by carbon numbers

The emission factors of each carbonyl compound monitored in this study are given in Table 2.

The concentrations of C1-C3, in case of all four cooking styles, accounted for over 80% of the total carbonyls detected, while the content of C4-C8 was relatively low. Concentrations of chain-like
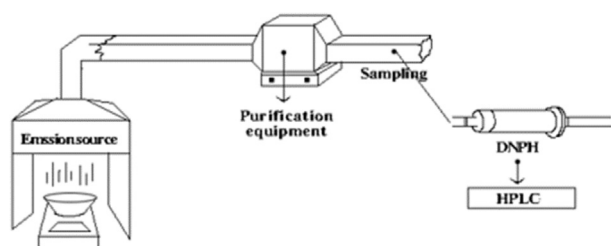

Fig. 1. Diagram of sampling methodology.