Environmental Pollution 227 (2017) 24–30

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Chemical characteristic of $PM_{2.5}$ emission and inhalational carcinogenic risk of domestic Chinese cooking☆

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ARTICLE INFO

Article history:
Received 26 December 2016
Received in revised form
11 April 2017
Accepted 13 April 2017

Keywords:
Indoor pollution
$PM_{2.5}$
Cooking
Source profile
Carcinogenic risk

ABSTRACT

To illustrate chemical characteristic of $PM_{2.5}$ emission and assess inhalational carcinogenic risk of domestic Chinese cooking, 5 sets of duplicate cooking samples were collected, using the most used 5 types of oil. The mass abundance of 14 elements, 5 water-soluble ions, organic carbon (OC), elemental carbon (EC) and 11 polycyclic aromatic hydrocarbons (PAHs) were calculated; the signature and diagnostic ratio of cooking in the domestic kitchen were analyzed; and carcinogenic risks of heavy metals and PAHs via inhalation were assessed in two scenarios. The analysis showed that OC was the primary composition in the chemical profile; Na was the most abundant element that might be due to the usage of salt; Cr and Pb, NO_3^- and SO_4^{2-}, Phe, FL and Pyr were the main heavy metals/water-soluble ions/PAHs, respectively. Phe and FL could be used to separate cooking and stationary sources, while diagnostic ratios of BaA/(BaA + CHR), BaA/CHR, BaP/BghiP and BaP/BeP should be applied with caution, as they were influenced by various cooking conditions. Carcinogenic risks of heavy metals and PAHs were evaluated in two scenarios, simulating the condition of cooking with no ventilation and with the range hood on, respectively. The integrated risk of heavy metals and PAHs was 2.7×10^{-3} and 5.8×10^{-6}, respectively, during cooking with no ventilation. While with the usage of range hood, only Cr(VI), As and Ni might induce potential carcinogenic risk. The difference in the chemical abundance in cooking sources found between this and other studies underlined the necessity of constructing locally representative source profiles under real conditions. The comparison of carcinogenic risk suggested that the potentially adverse health effects induced by inorganic compositions from cooking sources should not be ignored. Meanwhile, intervention methods, such as the operation of range hood, should be applied during cooking for health protection.

1. Introduction

Source identification of personal particulate matter (PM) exposure has been conducted worldwide for pollution control strategies and health effects research. Sources of indoor PM should be of great concern (Chen et al., 2012; Saito et al., 2014; Taner et al., 2013). On one hand, people spend the majority of their time indoors (Abdullahi et al., 2013; See and Balasubramanian, 2006; Slezakova et al., 2009) and could be exposed to indoor sources from a short distance; On the other hand, heavy metals and organic compounds emitted from these sources were concentrated on $PM_{2.5}$ (particulate matter with aerodynamic diameter less than or equal to 2.5 μm), which may deposit deeper into the alveoli and induce adverse health impacts (Che et al., 2014; Huang et al., 2011; Hung et al., 2007; Liao et al., 2006; Saito et al., 2014; Slezakova et al., 2009; Taner et al., 2013). However, the available information related to the chemical characterization of indoor PM sources was very limited, which hindered the estimation of their contribution to carcinogenic risk.

Cooking is one of the most significant indoor PM sources (Abdullahi et al., 2013; He et al., 2004; Lai and Ho, 2008; Massey

☆ This paper has been recommended for acceptance by Dr. Chen Da.

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http://dx.doi.org/10.1016/j.envpol.2017.04.033
0269-7491/

et al., 2012; Saito et al., 2014; See and Balasubramanian, 2006; Wan et al., 2011). Abdullahi et al. (2013) stated that previous research had mainly focused on mass concentration, size distribution and chemical composition of PM emitted from cooking. Some studies monitored in commercial restaurants (Chen et al., 2012; He et al., 2004; Lee et al., 2001; See et al., 2006; Taner et al., 2013; Zhao et al., 2007a; Zhu and Wang, 2003), and some simulated cooking with a single raw material or activity like oil-heating, meat charbroiling or tofu frying (Schauer et al., 1999, 2002; See and Balasubramanian, 2008; Zhang et al., 2009; Zhu and Wang, 2003). However, these results were not representative of chemical characterization of exposure to real domestic cooking. Moreover, relevant research primarily focused on organic compounds such as carbonyls and polycyclic aromatic hydrocarbons (PAHs) (Chen et al., 2012; He et al., 2004; Li et al., 2003; Saito et al., 2014; Zhu and Wang, 2003). The features of inorganic compositions (heavy metals, water-soluble ions, elemental carbon (EC), etc.) were usually overlooked. Some of the heavy metals were also considered as possible carcinogens according to International Agency for Research on Cancer (IARC) (IARC, 2006, 2012), like PAHs. It had been reported that heavy metals were likely to affect human respiratory and nervous systems (Fang et al., 2010; Li et al., 2013). As people usually cook at home using various raw materials and cooking methods, comprehensive information on chemical profiles of $PM_{2.5}$ emitted from cooking in domestic houses was urgently needed, for both carcinogenic risk assessment and personal source apportionment.

The establishment of source profiles should also consider the local emission characteristics, as the results of source apportionment based on source profiles from other countries might substantially cause the difference (Kong et al., 2011, 2013; Patil et al., 2013). The composition of PM emitted from cooking was strongly associated with cooking style (See et al., 2006), cooking method (He et al., 2004; McDonald et al., 2003; See and Balasubramanian, 2008; Zhu and Wang, 2003), type of oil (Buonanno et al., 2009; Zhang et al., 2009; Zhu and Wang, 2003), raw materials (Buonanno et al., 2009; McDonald et al., 2003), and even additives (Saito et al., 2014; Torkmahalleh et al., 2013; Yassin et al., 2012). It had been reported that PM emissions of vegetable-cooking was much lower than that of fatty-food cooking, while cooking with oil (such as stir-frying, sautéing, deep-frying, etc.) might produce higher amounts of PM than cooking with water (i.e. boiling and steaming) (Abdullahi et al., 2013; He et al., 2004; See and Balasubramanian, 2008). Chinese food is famously healthy with an abundance of vegetables cooked via mixed methods (but mainly with oil), and differences in $PM_{2.5}$ emissions have already been found between the Chinese cooking style and others (See et al., 2006). Therefore, the published cooking source profiles, which primarily described the emissions of cooking with a single raw material using the same cooking methods, were not representative of the complexity of Chinese cooking. As cooking exposure usually takes place at home, source profiles of real dishes in a domestic kitchen should be established to fill a gap of knowledge in China.

In this study, 5 sets of $PM_{2.5}$ cooking samples were collected in a domestic kitchen using 5 types of commonly used oils. Detected chemical components included 14 elements, 5 water-soluble ions, organic carbon (OC), EC and 11 PAHs. The primary purpose of this study was to: (1) form $PM_{2.5}$ source profiles of cooking in actual household environments; (2) obtain source markers and diagnostic ratios of cooking sources; and (3) assess carcinogenic risks of $PM_{2.5}$ bounded heavy metals/PAHs emitted during cooking. The results would provide basic information for personal PM source apportionment, as well as health impact assessment of inhabitants.

2. Material and methods

2.1. Sampling

2.1.1. Sampling sites and instruments

The samplings were conducted in an empty apartment with a kitchen and two spare rooms. The interior surfaces of the rooms consisted of the most common architectural materials in China, such as latex and flat wall paint. The fuel for cooking was natural gas, from which the emission has negligible contribution to particulate matter formation. No range hood was equipped in the kitchen and the experiments were carried out with closed glass windows and wooden doors, simulating no major ventilation and minimum turbulence to disperse the particles. As a result, we assumed that the $PM_{2.5}$ mass collected during the sampling was mainly due to the cooking with relatively small contribution from the existing $PM_{2.5}$ background in the apartment.

Personal exposure monitors (PEM-$PM_{2.5}$; BGI Inc., Waltham, MA, USA) and pumps (Buck Inc, Orlando, FI, USA) were used for the sampling. Each monitoring system consisted of two pumps connected with two samplers: one was loaded with 37 mm quartz-fiber filter (Pall-Gelman, Ann Arbor, MI, USA) and the other with 37 mm polypropylene-fiber filter (Pall-Gelman, Ann Arbor, MI, USA). Quartz-fiber filters were used to analyze water-soluble ions, OC, EC and PAHs; polypropylene-fiber filters were used to analyze elements. During experiments, sampling monitors were placed at a vertical distance of 1.2 m aboveground and at a horizontal distance of 0.3 m away from the wok; pumps operated at a flow of 4 ± 0.2 L min^{-1} and were calibrated using a soap-film flow meter (Buck Inc, Waltham, MA, USA) before and after sampling; two monitoring systems were occupied for replication.

2.1.2. Sample collection

The experiments were designed to simulate $PM_{2.5}$ formation occurring during domestic Chinese cooking. The selected 5 types of cooking oil, cooking methods and main raw materials of this study were the most commonly used in domestic Chinese cooking, as shown in Table 1. On each sampling day, 7 dishes were cooked successively using only one type of oil, and the accumulated $PM_{2.5}$ emissions were collected on the filter; the cooking lasted for 1 h duration, while the sample collection started as soon as the cooking began and lasted 6 h until there was no obvious lampblack smoke. Between each dish, the wok was thoroughly washed with detergent and tap water; between each sampling day, the apartment was put under forced ventilation for a duration of 24 h to clear the air from any residue inherited from the carried out experiments. As 2 monitoring systems were placed in the kitchen, 5 sets of duplicate filters for cooking emissions were collected in total. The duplicate concentrations of mass and chemical compositions were averaged for data analysis.

2.2. Mass and chemical analysis

2.2.1. Mass analysis

Before sampling, polypropylene-fiber and quartz-fiber filters were pre-heated at 60 °C and 800 °C for 2 h each. Then, they were equilibrated in a controlled laboratory with constant relative humidity (35% ± 1%) and temperature (22 °C ± 1 °C) for at least 48 h. All of the filters were first exposed to a low-level radioactive source to remove the static charge, and then weighed using a microbalance (Mettler MX5, Mettler-Toledo, Greifensee, Switzerland) with a sensitivity of ±1 μg. After three measurements, the average value of each filter was used.