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Concentration and particle size distribution of polycyclic aromatic hydrocarbons formed by thermal cooking

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A B S T R A C T

The concentration and particle size distribution of 19 major polycyclic aromatic hydrocarbons (PAHs) emitted by thermal cooking were investigated. Corn, trout, beef, prawns, and pork were selected for grilling. The PAHs in the oil mist emitted when the food was grilled were collected according to particle size range and analysed by GC/MS. Much higher concentrations of PAHs were detected in the oil mist emitted by grilled pork, trout, and beef samples, which were rich in fat. The main components of the cooking exhaust were 3- and 4-ring PAHs, regardless of food type. The particle size distribution showed that almost all the PAHs were concentrated in particles with diameters of <0.43 μm. For pork, the toxic equivalent of benzo[*a*]pyrene accounted for 50% of the PAHs in particles with diameters of <0.43 μm. From these results, we estimated that >90% of the PAHs would reach the alveolar region of the lungs.

1. Introduction

Cooking exhaust is one of the most important components of indoor air pollution, along with cigarette smoke, volatile organic compounds (VOCs) emitted from building materials, and combustion exhausts from heaters. Chemical compounds in the emissions produced by cooking consist mainly of gases, such as NO and CO, esters, fatty acids, hydrocarbons, and mist components (Rogge, Hildemann, Mazurek, Cass, & Simoneit, 1991). These chemical compounds can be adsorbed onto kitchen walls and exhaust fans, and they negatively affect the indoor air quality and the health of the inhabitants. Cooking emissions can also contain carcinogenic polycyclic aromatic hydrocarbons (PAHs) (Chen, Wang, Hsieh, Yang, & Lee, 2012; Dyremark, Westerholm, Övervik, & Gustavsson, 1995).

PAHs constitute a large class of organic compounds that are composed of two or more fused aromatic rings. Currently, more than 100 PAHs have been found in nature (United State Department of Health, 1995). They are formed primarily through the incomplete combustion or pyrolysis of organic matter and also during various industrial processes (Lee, Novotny, & Bartle, 1981). In addition, PAHs are also produced by volcanic activity (Fuoco et al., 2012), wildfires (Zhang & Tao, 2009), fossil fuel burning (Daisey, McCaffrey, & Gallagher, 1981), and other natural processes. Thus, humans have been exposed to PAHs throughout our evolutionary history.

Studies of PAHs in food have examined the carcinogenic effects on humans. Lodovici, Dolara, Casalini, Ciappellano, and Testolin (1995) measured the PAHs in several Italian foods and found that the highest levels of PAHs were in pizza baked in wood-burning ovens and in barbecued beef and pork. Phillips (1999) reported that diet was usually the main source of human exposure to PAHs, and that the major dietary sources of PAHs were cereals and vegetables, rather than meat. Furthermore, Barranco et al. (2004) measured the PAHs in different fatty foods from a Spanish market, and found that the average concentration of the total amount of PAHs in edible vegetable oils was below 25 ng g^{-1}, whereas the total amount of heavy PAHs did not exceed 5 ng g^{-1}. Furthermore, Chung et al. (2011) reported that relatively high levels of PAHs were found in charcoal-grilled pork. The European Union's Scientific Committee on Food (Scientific Committee of Food, 2002) concluded that 15 PAHs showed clear evidence of mutagenicity and genotoxicity in somatic cells of animals *in vivo*. In addition, Canada, Korea, and China introduced standards for PAHs in foods, limiting the concentration of benzo[*a*]pyrene (BaP). The World Health Organization (1998) established guidelines for PAH concentrations in drinking water.

In addition, PAHs emissions, generated by thermal cooking, have been studied. Rogge et al. (1991) focused on cooking emissions as a source of atmospheric PAHs, and reported the levels of PAHs in emissions from grilled meat. Li, Lin, Lee, and Tsai (2003)

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reported that PAH emissions from cooking Chinese food were 29.5–130 μg m^{-3}, which was approximately 2-fold greater than that from Japanese and fast food. McDonald et al. (2003) studied the levels of PAHs in char-grilled foods and reported that PAH concentrations were 28 mg m^{-3} in chicken, 39 mg m^{-3} in steak, and 45 mg m^{-3} in hamburger. Furthermore Tanaka, Ohtake, Tsuzaki, and Miyazaki (2012) showed that heating of mackerel and pike generated over 100-fold more PAHs when the fish was cooked directly over the heat source. In addition, the amount of PAHs generated was positively correlated with the amount of fat in the food, and not with the amount of protein and carbohydrates.

Most PAHs generated by thermal cooking, particularly high molecular weight PAHs, are carried on particles or in the oil mist in the cooking exhaust. When people inhale these particles and the oil mist, the deposition site in the respiratory system depends on the particle size. In general, finer particles are capable of reaching deeper into the alveoli (United States Environmental Protection Agency, 2004), and this may pose a risk of lung cancer. Therefore, it is important to clarify the particle size distribution of PAHs in cooking emissions in order to evaluate health risks associated with exposure to cooking exhaust.

Few studies have examined the particle size distribution of PAHs emitted from cooking exhaust. Koyano et al. (2001) collected particulate PAHs emitted from grilled fish, and divided them into three particle size ranges: >10 μm, 10–2.5 μm, <2.5 μm. Most of the PAHs were found in the particles with diameters of <2.5 μm. Kleeman et al. (2008) reported the concentration of PAHs in ultrafine particles ($PM_{0.1}$) produced by char-grilled foods. See and Balasubramanian (2008) reported the chemical characteristics of fine particles ($PM_{2.5}$) emitted by steaming, boiling, stir-frying, pan-frying, and deep-frying in a domestic kitchen. Deep-frying produced the largest amount of $PM_{2.5}$ and most of the chemical components, followed by pan-frying, stir-frying, boiling, and steaming. Furthermore Tanaka, Akai, Tsuzaki, and Miyazaki (2012) analysed PAHs in cooking emissions from grilled fish. They reported that most PAHs were concentrated in particles 1.1–2.1 μm and >11 μm in size. However, there have been few reports describing the influence of food types on the particle size distribution of PAHs emitted by cooking.

The objectives of this study were to evaluate the effects of the protein, carbohydrate, and fat content of food on the formation of PAHs and the particle size distribution, and to analyse the health risk of the respiratory tract deposition of the inhaled particles that carry PAHs.

2. Materials and methods

2.1. Target PAHs

The following 19 common PAHs, shown in Table 1, were selected for analysis; naphthalene (Nap), acenaphthylene (Anl), acenaphthene (Ane), fluorene (Flu), phenanthrene (Phen), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz[*a*]anthracene (BaA), chrysene (Cry), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*e*]pyrene (BeP), BaP, perylene (Prl), indeno[1,2,3-*cd*]pyrene (InP), dibenz[*a*,*h*]anthracene (DahA), benzo[*g*,*h*,*i*]perylene (BghiP), and coronene (Cor). The toxic equivalency factor (TEF), where the toxicity of other PAHs is normalised to that of BaP (Nisbet & Lagoy, 1997), and the assessment of the carcinogenicity of the target PAHs by IARC (2010), are also shown in Table 1.

2.2. Reagents

The 19 PAH standards were purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan) and AccuStandard Inc. (New Haven, CT). Dioxin analysis grade hexane, dichloromethane, and methanol (Wako Pure Chemical Industries) were used for sample preparation and analysis. The stable isotope reagents, naphthalene-d10, phenanthrene-d10, 1,4-dichlorobenzene-d4, fluoranthene-d10, acenaphthrene-d10, perylene-d12, and chrysene-d12 (AccuStandard.) were used as PAH internal standards.

Table 1
Target compounds measured in this study.

Compound	Abbreviation	Number of rings	IARC[a]	TEF[b]
Naphthalene	Nap	2	2B	0.001
Acenaphthylene	Anl	3	–	0.001
Acenaphthene	Ane	3	–	0.001
Fluorene	Flu	3	3	0.001
Phenanthrene	Phen	3	3	0.001
Anthracene	Ant	3	3	0.01
Fluoranthene	Flt	4	3	0.001
Pyrene	Pyr	4	3	0.001
Benz[a]anthracene	BaA	4	2A	0.1
Chrysene	Cry	4	3	0.01
Benzo[*b*]fluoranthene	BbF	5	2B	0.1
Benzo[*k*]fluoranthene	BkF	5	2B	0.1
Benzo[*e*]pyrene	BeP	5	3	–
Benzo[*a*]pyrene	BaP	5	2A	1
Perylene	Prl	5	3	–
Indeno[*1,2,3-cd*]pyrene	InP	6	2B	0.1
Dibenz[*a,h*]anthracene	DahA	5	2A	5
Benzo[*g,h,i*]perylene	BghiP	6	3	0.01
Coronene	Cor	7	3	–

[a] IARC (2010).
[b] Nisbet and Lagoy (1997).

2.3. Selected foods

The following foods were selected for thermal cooking: prawns (Australia), corn (Japan), trout (Chile), beef (round cuts, Australia), and pork (rib, Japan). According to the Standard Tables of Food Composition in Japan (Ministry of Education, Sports, & Technology, 2005), prawns contain the most protein (21.6 g 100 g^{-1}-edible portion) very little carbohydrate (trace amount), and fat (0.6 g 100 g^{-1}-edible portion), whereas corn contains the most carbohydrates (16.7 g 100 g^{-1}-edible portion) and a small amount of protein (3.6 g 100 g^{-1}-edible portion) and fat (1.7 g 100 g^{-1}-edible portion). Furthermore, the fat and protein contents of trout (14.7 g 100 g^{-1}-edible portion, 20.8 g 100 g^{-1}-edible portion), beef (9.6 g 100 g^{-1}-edible portion, 21.2 g 100 g^{-1}-edible portion), and pork (40.1 g 100 g^{-1}-edible portion, 13.4 g 100 g^{-1}-edible portion) were high, whereas trout contained small amounts of carbohydrate (trace amount, 0.5 g 100 g^{-1}-edible portion, and trace amount, respectively). Prawns were classified as protein-rich, corn was classified as carbohydrate-rich, and trout, beef, and pork were classified as fat- and protein-rich.

2.4. Thermal cooking

A home cooker, fuelled by methane, was used to thermally cook the foods. Each food was grilled directly over medium heat, allowing the gas flame to come into contact with the food. The cooking conditions were determined by preliminary cooking experiments and by cooking methods generally used in Japan. Cooking conditions were as follows: prawns, cooking time of 6 min for one 150 g portion, total amount of 2100 g; corn, cooking time of 8 min for one 600 g portion, total amount of 9000 g; trout, cooking time of 5 min for one 140 g portion, total amount of 3500 g; beef, cooking time of 3 min for one 3 × 4 cm 150 g piece, total amount of 1500 g; pork, cooking time of 17 min for one 150 g portion, total amount of 1500 g. No additives, such as oil, water, seasonings, and