



# Correlations between polycyclic aromatic hydrocarbons and polar components in edible oils during deep frying of peanuts

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## ARTICLE INFO

### Article history:

### Keywords:

Deep-frying oils  
Total polar components  
Polar component compositions  
Polycyclic aromatic hydrocarbons  
Correlations

## ABSTRACT

Peanuts were deep-fried for 192 h in soybean oil and two blends (super palm olein oil/soybean oil/high oleic sunflower oil, at volume ratios of 2:2:1 and 3:2:1). For each type of oil, changes in the total polar components (TPCs), the polar component composition (polymerized products, oxidized products and hydrolysis products), and polycyclic aromatic hydrocarbons (PAHs) and the correlations between them were investigated. These parameters were found to increase as the deep-frying time increased; the increase was fastest for soybean oil. In this study, the TPCs content showed higher correlations with polymerized products and oxidized products than with hydrolysis products. In addition, the Pearson correlation coefficients between PAHs and polymerized products and between PAHs and oxidized products were higher (ranging, 0.766 to 0.917), which proved that the changes in PAHs concentrations are related to oxidative stability of oils. These results may be helpful in studying the mechanism of PAHs accumulation and in developing ways to reduce PAHs contents during frying processes.

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## 1. Introduction

Deep-frying is a traditional, popular food preparation method around the world. Fried foods possess good flavor, a crisp texture, and an attractive appearance, which makes them appealing to consumers (Li, Li, Wang, Cao, & Liu, 2017; Sebastian, Ghazani, & Marangoni, 2014). Because deep-frying is normally carried out between 150 °C and 190 °C and involves exposure to oxygen and moisture (Alizadeh, Nayebzadeh, & Mohammadi, 2016), it generated short chain fatty acids, aldehydes, ketones, epoxides and hydrocarbons, free fatty acids (FFA), mono- and diacylglycerols, glycerol, and organic contaminants such as polycyclic aromatic hydrocarbons (PAHs) (Hua, Zhao, Wu, & Li, 2016; Weisshaar, 2014). PAHs are a large group of toxic contaminants that contain two or more fused aromatic rings (Shi, Liu, Liu, & Zhang, 2015). It is well known that some have carcinogenic, mutagenic, and teratogenic properties (Li, Wu, Wang, & Akoh, 2016). Generally, the toxicity of PAH increases with the number of aromatic rings in the PAH molecule (Li, Wu, Zeng, Wang, & Yu, 2016; Nisbet & Lagoy, 1992). Benzo[a]pyrene was long considered a marker of the carcinogenic

PAHs in food, but the European Food Safety Authority (EFSA) Panel on Contaminants in the Food Chain (CONTAM Panel) (EFSA, 2008) concluded that PAH4 (i.e., the sum of benzo[a]anthracene [BaA], benzo[a]pyrene [BaP], benzo[b]fluoranthene [BbFlu], and chrysene [Chr]) and PAH8 (i.e., the sum of BaA, Chr, BbFlu, benzo[k]fluoranthene [BkFlu], BaP, dibenzo[a,h]anthracene [DBahA], benzo [g,h,i]perylene [BghiP], and indeno[1,2,3-c,d]pyrene [IP]) are more accurate indicators of PAHs levels than BaP.

Ingestion in food is a major route by which humans are exposed to PAHs (Purcaro, Moret, & Conte, 2013). The safety of fried food is closely related to the quality of the deep-frying oil (Weisshaar, 2014). Because intensive mass exchange occurs between the frying oil and the fried food, the majority of harmful substances will be transferred to the final fried food, which may have detrimental health effects (Aladedunye & Przybylski, 2009a; Bansal, Zhou, Barlow, Lo, & Neo, 2010; Casal, Malheiro, Sendas, Oliveira, & Pereira, 2010). Thus, the quality of frying oils should be controlled.

Several deteriorative reactions, including oxidation, hydrolysis, polymerization, pyrolysis, isomerization, and cyclisation, occur during frying (Song, Kim, Kim, & Lee, 2017). In general, total polar components (TPCs) and their different compositions are used to monitor the degree of degradation in frying oils (Mba, Dumont, & Ngadi, 2016). TPCs levels are considered one of the most reliable parameters because they reflect all three types of fatty acid

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degradation products: oxidized, polymerized and hydrolysis products (Bansal et al., 2010; Santos, Cruz, Cunha, & Casal, 2013). The mechanism for the accumulation of PAHs during the frying process has not yet been conclusively determined (Li, Wu, Zeng, et al., 2016). In recent years, a few group (An, Liu, & Liu, 2017; Farhadian, Jinap, Faridah, & Zaidul, 2012; Wongmaneeprapitip & Vangnai, 2017; Zhao, Wu, Gong, Li, & Zhuang, 2017) have studied the correlation between PAHs contents and antioxidant capacity of oils. An et al. (2017) showed that the PAHs level of oil used for frying can be predicted based on its correlation with the TPCs value. However, according to AOCS Official Method Cd 20–91, TPCs are defined as “those components in oils and fats which are determined by column chromatography.” The TPCs level of frying oil is thus a mixture of oxidized, polymerized, and hydrolysis products, as described in ISO 16931:2009 (2009). In some cases, even in oils with the same TPCs value, the specific TPCs compositions may be very different (Bansal et al., 2010). Therefore, in this study, the polar component composition, including PTG, DTG, OTG, DAG, MAG, FFA and unsaponifiables, was further analyzed by the HPSEC method according to ISO 16931:2009 (2009) and correlated to PAHs. These components are related to the reactions of polymerization, oxidation, and hydrolysis. By taking all of these indices into consideration, the correlation between PAHs and polar component composition may be evaluated more effectively and systematically.

The peanut is a kind of ideal food around the world. Raw peanuts are commonly thermal treated, such as frying (Farhadian et al., 2012). Fried peanuts are a popular fried food. In China, nearly all commercial restaurants and supermarkets have fried peanuts for sale. Peanuts and their derived products are vulnerable to oxidation processes because they contain about 50%–55% lipids. PAHs have been found in both raw and fried peanuts (Zhao et al., 2017). However, no studies to date have paid attention to the retention of PAHs in the oil used for frying peanuts.

In this study, the main objective is to determine the values of PAHs and polar components (TPCs and polar component composition) in soybean oil and two blends of soybean oil with super palm olein oil and high oleic sunflower oil under commercial frying conditions. In addition, the correlation between the values of TPCs, the polar component composition, and PAHs will be analyzed.

## 2. Materials and methods

### 2.1. Materials and reagents

Oils for frying were purchased from Cargill Corp (Shanghai, China): high-grade soybean oil (SO), two blended oils consisting of high-grade soybean oil, super palm olein oil (melting point 8 °C) (SPOO) and high oleic sunflower oil (HOSO). The SPOO: SO: HOSO (%v/v/v) blends were prepared 4.8 L at a time, as follows: 2:2:1 (%v/v/v) (Blend 1) and 3:2:1 (%v/v/v) (Blend 2); a new batch was readied for the next use once the blend was nearly exhausted. The composition of the three fresh oils in terms of major fatty acids is listed in Table 1. Raw peanuts (Fenghua 1) were obtained from the farmers' market in Yimengshan (Shandong, China). All peanuts were peeled with an initial moisture content and oil content of 14.37% and 54.15%, respectively.

Acetonitrile, n-Hexane, dichloromethane, and tetrahydrofuran (HPLC grade) were purchased from J&K Scientific Ltd. (Beijing, China). Other solvents and reagents (analytical grade) were purchased from Sinopharm Chemical Reagent (Shanghai, China). Ultra-pure water was purified by a Milli-Q Advantage System (Billerica, MA).

A mixed standard of PAHs (200 µg/mL in acetonitrile), containing BaA, Chr, BbFlu, BkFlu, BaP, DBaA, BghiP, and IP, was purchased from O2si (Charleston, South Carolina). The components

**Table 1**  
Fatty acids composition (area %) of fresh oils.

| Fatty acids | SO           | Blend 1      | Blend 2      |
|-------------|--------------|--------------|--------------|
| C12:0       | –            | 0.09 ± 0.01  | 0.12 ± 0.00  |
| C14:0       | 0.08 ± 0.01  | 0.46 ± 0.00  | 0.55 ± 0.02  |
| C16:0       | 10.92 ± 0.16 | 18.37 ± 0.11 | 20.82 ± 0.28 |
| C16:1       | 0.08 ± 0.00  | 0.14 ± 0.02  | 0.15 ± 0.01  |
| C18:0       | 4.17 ± 0.08  | 3.66 ± 0.03  | 3.65 ± 0.06  |
| C18:1       | 21.33 ± 0.34 | 43.56 ± 0.51 | 44.12 ± 0.40 |
| C18:2       | 52.05 ± 1.48 | 28.07 ± 0.24 | 25.62 ± 0.19 |
| C18:3       | 7.16 ± 0.05  | 3.21 ± 0.07  | 2.78 ± 0.02  |
| C20:1       | 0.26 ± 0.00  | 0.25 ± 0.01  | 0.24 ± 0.02  |
| ΣSFA        | 15.17 ± 0.25 | 22.58 ± 0.07 | 25.13 ± 0.24 |
| ΣMUFA       | 21.67 ± 0.34 | 43.95 ± 0.54 | 44.51 ± 0.43 |
| ΣPUFA       | 59.21 ± 1.53 | 31.28 ± 0.17 | 28.39 ± 0.17 |

Note: Mean ± SD; SO, soybean oil; Blend 1, super palm olein oil (melting point 8 °C), soybean oil, and high oleic sunflower oil at a ratio of 2:2:1 (%v/v/v); Blend 2, super palm olein oil (melting point 8 °C), soybean oil, and high oleic sunflower oil at a ratio of 3:2:1 (%v/v/v); ΣSFA, the sum of saturated fatty acids; ΣMUFA, the sum of monounsaturated fatty acids; ΣPUFA, the sum of polyunsaturated fatty acids.

used as internal standards were perdeuterated PAHs (2000 µg/mL in dichloromethane): chrysene-d12 (Chr-d12) and perylene-d12 (Per-d12) were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). The purities of the PAH standards and internal standards were between 95.0% and 99.5%. All standards were stored at –20 °C in the dark.

### 2.2. Preparation of oils for deep-frying

The frying was performed in a commercial fryer (EF - 81, Wailaan Kitchen Equipment, Guangzhou, China) with a maximum oil capacity of 5 L and a nominal power, Q, of 2500 W. Three experiments were performed at 150 °C. About 4.5 L of SO, Blend 1, and Blend 2 were placed in turn in the deep fryer with temperature control. Raw peanuts (450 ± 1.0 g) were deep-fried for 5 min every 10 min (each frying batch time was 10 min, including 5 min frying time and 5 min waiting time). The frying oil samples (30 mL) were collected and placed in sealed bottles after every 6 h of frying, and fresh oil was added to restore the volume to 4.5 L. At the end of each day of frying, the deep fryer was shut off and the oil was cooled down. The oil was then filtered to remove solid residues through a sieve (80 mesh). The overall time that the oil was heated was approximately 192 h (12 h per day). The oil was sampled for analysis at 0, 6, 12, 18, 24, 30, 42, 54, 66, 78, 96, 120, 144, 168, and 192 h. All samples were stored at –20 °C.

### 2.3. Determination of fatty acid composition

The fatty acid composition was determined via fatty acid methyl esters and analyzed by an Agilent 7820A gas chromatograph (Agilent Technologies, California, USA) equipped with a Trace TR-FAME capillary column (PEG-20 M, 60 m × 2.5 mm × 2.5 µm, Thermal Fisher, MA, USA) and a flame ionization detector (FID). The details were in accordance with AOCS Official Method Ce 1–62.

### 2.4. Determination of PAHs

The analytical method for determining PAHs levels was the procedure described by Yu, Wang, Jin, Dong, and Wang (2014), with some modifications. Approximately 0.5 g of the oil sample was measured into a 15-mL glass test tube. A total of 50 µL of the internal standard solution (200 µg/mL) and 3 mL of n-Hexane were added. The mixtures were shaken for 1 min with a vortex mixer and transferred onto a molecularly imprinted polymer (MIP)-PAH SPE cartridge (ANPEL, Shanghai, China) activated with 5 mL of

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