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Effect of frying and aluminium on the levels and migration of parent and oxygenated PAHs in a popular Chinese fried bread youtiao

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ABSTRACT

This study investigated the effects of frying conditions on the change, migration and correlation of polycyclic aromatic hydrocarbons (PAHs) and oxygenated PAHs (oxy-PAHs) in youtiao, a typical Chinese fried bread. PAH migration was evaluated using an isotope tracer method. High concentrations of oxy-PAHs, especially 9-fluorenone (1.75 μg/kg) and anthracene-9,10-dione (6.41 μg/kg), were detected. The results revealed that 1) prolonged continuous frying decreased parent PAH and oxy-PAH concentrations, 2) frying temperature and aluminium content did not have significant effects on parent PAH concentrations, 3) PAH migration rates (2.02–18.48%) varied significantly based on the frying oil type, and 4) the sum of five oxy-PAH concentrations was significantly correlated with the sum of 16 parent PAH concentrations in soybean oil-fried youtiao. Three oxy-PAHs (9-fluorenone, anthracene-9,10-dione, and 9,10-dihydrobenzo[*a*]pyren-7(8h)-one) were significantly correlated with their respective parent PAHs. The results might be helpful for further study and evaluation on oxy-PAHs and PAHs in fried foods.

1. Introduction

PAHs (Polycyclic aromatic hydrocarbons) represent a family of toxic compounds that contain two or more fused aromatic rings. Several PAHs, including the 16 US-EPA (U.S. Environmental Protection Agency) priority PAHs, have carcinogenic, mutagenic and immunomodulatory effects (Boström et al., 2002; Poster, Schantz, Sander, & Wise, 2006; Rengarajan et al., 2015). Based on the number of condensed aromatic rings, they can be divided into light (2–4 rings) and heavy (5 or more rings) PAHs. The heavy PAHs are more stable and toxic than the light ones. According to recent studies, BaP (benzo[*a*]pyrene) is not an adequate marker of PAHs in foods. On the other hand, scientists recommend the use of PAH8 (benzo[*a*]anthracene, chrysene, benzo[*k*]fluoranthene, benzo[*b*]fluoranthene, benzo[*a*]pyrene, indeno[1,2,3-*c*,*d*]pyrene, dibenzo[*a*,*h*]anthracene and benzo[*g*,*h*,*i*]perylene) and PAH4 (benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, and benzo[*a*]pyrene) as PAH markers (EFSA, 2008; Purcaro, Moret, & Conte, 2013). Due to their lipophilic nature, fried and oily foods may contain high concentrations of PAHs (Moret & Conte, 2000). Additionally, PAHs can be formed, or modified, at high temperature cooking, such as frying. Many researchers have investigated the concentrations of PAHs in fried foods and frying oils (Perelló, Marti-Cid, Castell, Llobet, & Domingo, 2009; Purcaro, Navas, Guardiola, Conte, & Moret, 2006; Rose et al., 2015; Wu & Yu, 2012).

Oxy-PAHs (oxygenated PAHs) are defined as PAHs with one or more carbonylic oxygen(s) attached to the aromatic ring structure, including ketones and quinones (Lundstedt et al., 2007). They are not only emitted from the same sources as PAHs, incomplete combustion, but also formed from oxidation of PAHs (Lundstedt et al., 2007). Even though their mechanism of toxicity is not thoroughly understood, oxy-PAHs are considered to be more toxic than their parent PAHs (Walgraeve, Demeestere, Dewulf, Zimmermann, & Van Langenhove, 2010). High concentrations of oxy-PAHs have been reported in food, such as fish and smoked meat (Bandowe et al., 2014; Chen et al., 2014).

Youtiao or oil stick, which is a long golden-brown deep-fried twisted stick of bread, is commonly consumed for breakfast and produced by cafeterias, food stalls, KFC and food factories in China. It is a very popular, typical and traditional Chinese fried flour food, for all ages, for thousand years. Youtiao is also consumed in Indonesia, Laos, Malaysia, Myanmar and Singapore. In a previous study, 16 US-EPA PAHs were detected in commercial youtiao samples in China (Li, Wu, Wang, & Akoh, 2016). These results revealed

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Table 1
Effect of frying temperature on PAH concentrations in youtiao.

PAHs	PAH concentrations (μg/kg wet mass)				
	160 °C	170 °C	180 °C	190 °C	200 °C
Light PAHs					
NA	1.95 ± 0.85[a]	0.99 ± 0.03[a]	1.30 ± 0.52[a]	0.93 ± 0.02[a]	1.10 ± 0.47[a]
Ap	0.12 ± 0.07[a]	0.06 ± 0.00[a]	0.10 ± 0.05[a]	0.08 ± 0.00[a]	0.07 ± 0.06[a]
Ac	0.29 ± 0.14[a]	0.15 ± 0.01[a]	0.23 ± 0.11[a]	0.21 ± 0.04[a]	0.34 ± 0.18[a]
F	1.10 ± 0.57[a]	0.60 ± 0.03[a]	1.00 ± 0.47[a]	1.28 ± 0.19[a]	1.21 ± 0.70[a]
Phe	3.24 ± 1.64[a]	1.90 ± 0.11[a]	2.96 ± 1.40[a]	3.29 ± 0.22[a]	2.81 ± 1.73[a]
Ant	0.61 ± 0.35[a]	0.30 ± 0.01[a]	0.47 ± 0.24[a]	0.46 ± 0.01[a]	0.30 ± 0.20[a]
Fl	1.64 ± 0.63[a]	0.99 ± 0.07[a]	2.43 ± 1.26[a]	0.84 ± 0.03[a]	0.72 ± 0.28[a]
Pyr	1.25 ± 0.47[a]	1.12 ± 0.01[a]	1.62 ± 0.91[a]	0.64 ± 0.00[a]	0.59 ± 0.28[a]
BaA	0.85 ± 0.22[a]	1.08 ± 0.24[a]	1.85 ± 1.00[a]	0.76 ± 0.02[a]	1.92 ± 0.32[a]
Chr	1.32 ± 0.18[a]	1.43 ± 0.07[a]	1.47 ± 0.42[a]	0.80 ± 0.00[a]	2.03 ± 0.65[a]
Total Light PAHs	12.40 ± 5.12[a]	8.63 ± 0.58[a]	13.44 ± 6.38[a]	9.29 ± 0.53[a]	11.10 ± 4.87[a]
Percentage (%)	89.73	58.35	74.66	55.33	71.92
Heavy PAHs					
BbF	0.63 ± 0.29[a]	2.05 ± 0.24[a]	1.70 ± 0.66[a]	1.34 ± 0.56[a]	2.02 ± 0.60[a]
BkF	0.14 ± 0.07[a]	0.57 ± 0.06[a]	0.44 ± 0.19[a]	0.62 ± 0.43[a]	0.53 ± 0.14[a]
BaP	0.40 ± 0.20[a]	1.38 ± 0.20[a]	1.00 ± 0.43[a]	1.22 ± 0.66[a]	0.89 ± 0.21[a]
IP	0.13 ± 0.02[a]	1.03 ± 0.23[a]	0.66 ± 0.30[a]	1.48 ± 0.90[a]	0.47 ± 0.02[a]
DBahA	0.08 ± 0.01[a]	0.33 ± 0.08[a]	0.16 ± 0.08[a]	1.50 ± 1.33[a]	0.15 ± 0.12[a]
BghiP	0.05 ± 0.01[a]	0.78 ± 0.14[a]	0.60 ± 0.26[a]	1.33 ± 0.79[a]	0.27 ± 0.06[a]
Total Heavy PAHs	1.42 ± 0.60[a]	6.16 ± 0.95[a]	4.56 ± 1.92[a]	7.50 ± 4.67[a]	4.34 ± 1.15[a]
Percentage (%)	10.27	41.65	25.34	44.67	28.08
Σ (BaP + DBahA)	0.48 ± .021[a]	1.71 ± 0.28[a]	1.16 ± 0.51[a]	2.72 ± 1.99[a]	1.04 ± 0.33[a]
PAH4	3.20 ± 0.45[a]	5.94 ± 0.40[a]	6.02 ± 1.34[a]	4.12 ± 0.87[a]	6.86 ± 0.96[a]
PAH8	3.60 ± 0.46[a]	8.65 ± 0.49[b]	7.88 ± 1.41[b]	9.05 ± 2.03[b]	8.28 ± 0.98[b]
Σ 16 PAHs	13.81 ± 5.72[a]	14.78 ± 1.53[a]	18.00 ± 8.30[a]	16.78 ± 5.20[a]	15.44 ± 6.02[a]

Different lowercase letters within a row represent significant differences ($p < 0.05$).
PAH4 includes the sum of BaA, Chr, BbF, and BaP.
PAH8 includes the sum of BaA, Chr, BkF, BbF, BaP, Ip, DBahA, and BghiP.

that the sum of PAH4 concentrations in these samples exceeded the permissible limit (1 μg/kg) established by the European Commission for processed cereal-based foods (European Commission Regulation 835/2011), while BaP concentrations were lower than the Chinese permissible limit (5 μg/kg) in cereal-based foods (National Standards of China GB 2762-20127).

Several chemical reactions take place during frying, e.g. oxidation, polymerization, hydrolysis, pyrolysis, isomerization and cyclisation. Frying condition (processing temperature and time) could have a strong effect on levels of oil oxidation in products (Koh & Surh, 2015). However, the effect of frying condition on PAH formation in fried foods is unknown. Little information is available concerning the contamination level of oxy-PAHs in fried foods.

Aluminium is an indispensable composition in the traditional formula of youtiao for more than thousand years. Alum, e. g. aluminium sulfate or aluminium potassium sulfate, is commonly used as a raising agent for youtiao. The addition of alum is to achieve a crispy and fluffy texture for products. However, many studies have shown that chronic exposure to aluminium can have a negative impact on human health, so the FAO/WHO recommended a provisional tolerable weekly intake of 2 mg/kg, and the National Standard of China was clear that fried food should contain no more than 100 mg/kg. In our previous survey, youtiao samples with high levels of aluminium content also suffered high PAHs contaminations (Li et al., 2016). Therefore, it is worth understanding whether aluminium could affect the levels of PAHs during frying.

Moreover, oil degradation products act as wetting agents and increase oil viscosity (Bouchon, 2009), which may have a pronounced effect on PAH migration from the frying oil into the food product. Therefore, PAH and oxy-PAH concentrations in fried foods should be investigated.

The main objective of this research was to evaluate the effects of different frying conditions (temperature, time, oil types and aluminium content) on the change, migration and correlation of parent and oxy-PAHs during the deep-frying of youtiao, a popular and typical Chinese fried bread youtiao.

2. Materials and methods

2.1. Materials and reagents

Low-gluten wheat flour, soybean oil and shortening oil were supplied by Yihai Kerry Co. Ltd. (Shanghai, China). Salt and sodium bicarbonate were purchased from a local supermarket in Shanghai, China. Aluminium potassium sulfate (purity ⩾ 99.5%) was supplied by Jianheng Industrial Co. Ltd. (Hunan, China). Fast youtiao raising agent (Angel Yeast Co. Ltd., Hubei, China) was used for the preparation of aluminium-free youtiao samples. The ingredients of the fast youtiao raising agent included salt, sodium bicarbonate (23%), corn starch, calcium carbonate (8%), tartaric acid (6%), sodium dihydrogen phosphate (6%), citric acid (4%), and xylanase (0.5%). *n*-Hexane, acetonitrile, acetone, methanol, toluene, and dichloromethane, which were of HPLC grade, were provided by CNW (Darmstadt, Germany). Water was purified using a Milli-Q water purification system (Millipore Co., Milford, USA). Supelclean ENVI-18 solid phase extraction (SPE) cartridges (2 g, 12 ml) and Florisil SPE cartridges (1 g, 6 ml) were acquired from Supelco Inc. (Bellefonte, PA). A standard PAH mixture (0.2 mg/ml in dichloromethane) from AccuStandard (New Haven, USA) consisted of the following 16 PAHs, naphthalene (NA), acenaphthylene (Ap), acenaphthene (Ac), fluorene (F), anthracene (Ant), phenanthrene (Phe), fluoranthene (Fl), pyrene (Pyr), benzo[*a*]anthracene (BaA), chrysene (Chr), benzo[*k*]fluoranthene (BkF), benzo[*b*]fluoranthene (BbF), benzo[*a*]pyrene (BaP), indeno[1,2,3-*c*,*d*]pyrene (Ip), dibenzo [*a*,*h*]anthracene (DBahA), and benzo[*g*,*h*,*i*]perylene (BghiP). 9-Fluorenone (9FO, 99.5%) and anthracene-9,10-dione (ATQ, 99.0%) were supplied by Dr. Ehrenstorfer (Augsburg, Germany).