



Incidence and survey of polycyclic aromatic hydrocarbons in edible vegetable oils in China



Long-Kai Shi, Dong-Dong Zhang, Yu-Lan Liu*

College of Food Science and Technology, Henan University of Technology, Zhengzhou, 450001, China

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) occurrence in eighty-five samples of edible vegetable oils from China was described for the first time. Analytes were determined based on a simplified solvent extraction and liquid chromatography-atmospheric pressure photoionization-tandem mass spectrometry method. PAHs were detected in all of the oil samples and the concentrations of BaP, PAH4 and PAH16 were 0.12–6.28, 1.48–27.84 and 9.25–154.27 $\mu\text{g}/\text{kg}$, respectively. Levels of BaP were all conformed to the upper limit 10 $\mu\text{g}/\text{kg}$ set by China. However, each had 13% of oil samples (11 samples) exceeded the maximum limit 2 and 10 $\mu\text{g}/\text{kg}$ set for BaP and PAH4 by EU. The light PAHs were primary in oil samples, and the contribution of the heavy PAHs was much less significant. A high variability of PAH levels between different oil types was observed, and peanut oils and sesame oils were two types of oils that were highly contaminated. The relationship between BaP and PAH4, BaP and PAH16 and PAH4 and PAH16 suggested that PAH4 was more suitable than BaP acted as a surrogate for the contamination level of PAHs in edible oils.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are non-polar chemical compounds that belong to a huge group of molecules containing two or more aromatic rings (Hutzler, Luch, & Filser, 2011; Petridis, Sakkas, & Albanis, 2014). Some of them are generated by incomplete combustion of organic materials (e.g., wood, coal, gas or tobacco) (Tarrant, Mills, & Williard, 2009), meanwhile, part of them are considered to be possible or probable human carcinogens (Olsson, Sadiqsis, Holmbäck, & Westerholm, 2014; Payanan, Leepitapiboon, & Varanusupakul, 2013; M. Wu, Xu, Yu, & Wang, 2011).

Due to the ubiquitous occurrence, suspected carcinogenicity and relative toxicity, sixteen PAHs have been identified as priority pollutants by the US Environmental Protection Agency (EPA) (M. Wu et al., 2011). Among them, individuals containing up to four fused rings are classified as light PAHs (LPAHs), and those containing more than four rings are called heavy PAHs (HPAHs) (Farhadian, Jinap, Abas, & Sakar, 2010). The primary source of exposure to PAHs for non-smokers and non-occupationally exposed individuals is dietary intake (Alomirah et al., 2011; Ramalhosa et al., 2012), whereas commercial edible vegetable oils are one of the most contributors due to the lipophilic property of PAHs (Amzad Hossain & Salehuddin, 2012; S. Wu & Yu, 2012). Edible oils are mainly polluted by environmental contamination of the raw materials (migration from contaminated air, water or soils) (Ciecierska & Obiedziński, 2013; Drabova et al., 2013), and also by pollution

* Corresponding author.

E-mail address: liuy17446@sina.com (Y.-L. Liu).

from direct smoke-drying, solvent extraction, package material and mineral oils (Amzad Hossain & Salehuddin, 2012; Moret & Conte, 2002).

Traditionally, the main PAHs of interest in edible oils has always been benzo[a]pyrene (BaP), which is often served as the benchmark for the presence and potential carcinogenic effects of the entire class of harmful compounds in vegetable oils. Up to now, there are many countries and organizations worldwide that set maximum limit values for PAHs in edible oils. For example, the upper residue level for BaP was set at 10 $\mu\text{g}/\text{kg}$ in edible oils according to the national standard in China (Wang & Guo, 2010). The German Society for Fat Science suggested a value of 5 $\mu\text{g}/\text{kg}$ for HPAHs and 25 $\mu\text{g}/\text{kg}$ for total PAHs (Payanan et al., 2013). Meanwhile, the European Food Safety Authority Panel on Contaminants in the Food Chain demonstrated that BaP was not a sufficient surrogate of PAHs existence in food and recommended four PAHs (benz(a)anthracene, chrysene, benzo(b)fluoranthene, BaP, defined as PAH4) as the most suitable representatives of toxic PAHs (Drabova et al., 2013). Recently, Commission Regulation of European Union No 835/2011 established the maximum legislation levels of BaP and PAH4 for edible oils, which are intended for direct consumption or use as a raw material ingredient in food, were 2 $\mu\text{g}/\text{kg}$ and 10 $\mu\text{g}/\text{kg}$, respectively (Drabova et al., 2013).

Previously, we developed an analytical technique for determining EPA 16 PAHs in edible vegetable oils by using liquid chromatography-atmospheric pressure photoionization-tandem mass spectrometry (LC-APPI-MS/MS) (Shi, Liu, Liu, & Zhang, 2015). The quantitative levels of 16 analytes in nine samples from domestic commercial oil brands were measured, and the differences in PAHs distribution profiles among three oil types were observed. So far, there is quite limited information on PAHs concentrations in edible oils commonly consumed in this one of the world's most populous country. Given the lack of information about these sort of chemical components in vegetable oils available to the public, consumers have little or no resources adopting potential safety concerns associated with these pollutants. To help further assess the dietary risks associated with oil consumption, we designed this study to obtain PAHs data on a diverse selection of oils manufactured and used in China. This is the first comprehensive effort in the last several years to report on the levels of selected PAHs measured in vegetable oils from modern oil brands in China. Meanwhile, the inclusion of multiple PAHs in various oil types provides a full-scale representation of the entire class of compounds in vegetable oils.

2. Materials and methods

2.1. Materials and chemicals

Certified EPA 16 PAHs standard mixture that was dissolved in acetonitrile with the concentration of 200 $\mu\text{g}/\text{mL}$ was provided by O2si (Charleston, South Carolina, USA), detailed information of analytes are shown in Table 1. The labeled internal standards, Nap-d8, Acy-d8, Ace-d10, Fl-d10, Phe-d10, Ant-d10, Pyr-d10, Chr-d12, BaA-d12, DBaA-d14 were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany), Flu-d10, BbFlu-d12, BkFlu-d12, BaP-d12, BghiP-d12, IP-d12 were purchased from C/D/N Isotopes (Pointe-Claire, Quebec, Canada). Purities of PAH standards and internal standards were guaranteed between 97.9% and 99.9%. Working mixed standard solutions containing all the analytes and analogues were prepared by dilution of the stock standard solutions with acetonitrile and stored at $-20\text{ }^{\circ}\text{C}$ in darkness to avoid volatilisation and photodegradation.

Acetonitrile and toluene (HPLC grade) were obtained from Sigma (St. Louis, MO, USA). Ultrapure water (18 M Ω cm resistiv-

ity) was prepared by using a Milli-Q system purchased from Millipore (Billerica, MA, USA). Nine types (eighty-five brands) of commercial edible oils were randomly purchased from the retail markets in Zhengzhou in the year of 2015. All the samples were stored in their original packages, assigned with unique labels and stored at $-20\text{ }^{\circ}\text{C}$ until needed.

2.2. Oil sample preparation and analytical condition

Oil sample preparation scheme and analytical method for PAHs determination was described in detail in a previous published paper (Shi et al., 2015). Sample preparation scheme was concisely described as below: 1 g (± 0.01 g) of oils were weighted at a 15 mL Teflon-type centrifuge tube. 60 μL of the internal standard solution (1 $\mu\text{g}/\text{mL}$ of each) and 8 mL of acetonitrile were added, the mixtures were shaken vigorously at 2000 rpm for 9 min. Then centrifuged at 6000 rpm for 3 min ($-20\text{ }^{\circ}\text{C}$), the top layer was carefully transferred into a glass test tube and the extraction process was repeated once again with 8 mL of acetonitrile. All the extracts were collected together and filtrated through a 0.22 μm nylon syringe filter, then dried under a gentle flow of nitrogen to about 200 μL . 5 μL of this solution was injected into the LC-APPI-MS/MS for analysis. Table 2 shows the LC conditions and all of the required instrumental parameters. Precursor ions and daughter ions of 16 PAH standards and their isotope internal standards are given in Table S1, data of the method validation (linear correlation, recoveries, limit of detection, limit of quantification, accuracy and precision) are provided in Table S2 and Table S3.

2.3. Data analysis

Chromatographic peaks were carefully reviewed using the Analyst software version 1.5.1 (Applied Biosystems, Foster City, CA), and any errors and integration corrections were made manually, if necessary. All of the standards and sample concentrations were measured using standard peak areas versus internal standard peak areas. Data analysis was performed using Origin 8.0 and Microsoft Office Excel 2013.

3. Results and discussion

3.1. PAHs in commercial edible vegetable oil

For this study, eighty-five samples of nine types of edible vegetable oils collected from China market were analyzed, including rapeseed oils ($n = 10$), soybean oils ($n = 15$), olive oils ($n = 8$), peanut oils ($n = 10$), sunflower oils ($n = 10$), corn oils ($n = 12$), sesame oils ($n = 10$), cottonseed oils ($n = 5$) and rice bran oils ($n = 5$). Table 3 shows the minimum, maximum and mean levels of contamination according to the type of edible oils studied, concentrations of individual PAHs and as well as LPAHs, HPAHs, PAH4 and total PAHs (defined as PAH16) are all presented.

16 analytes were found in all of the oil samples. PAH16 concentrations in 85 oil specimens were ranged from 9.25 to 154.27 $\mu\text{g}/\text{kg}$. The average of the total contents of 16 PAHs was 41.10 $\mu\text{g}/\text{kg}$. The concentrations of BaP were ranged from 0.12 to 6.28 $\mu\text{g}/\text{kg}$, and 1.29 $\mu\text{g}/\text{kg}$ was the mean. Meanwhile, PAH4 amounts were ranging from 1.48 to 27.84 $\mu\text{g}/\text{kg}$, and 7.70 $\mu\text{g}/\text{kg}$ was the mean. Individual levels of PAHs ranged from 0.01 $\mu\text{g}/\text{kg}$ (BaA level of an olive oil sample) to 40.54 $\mu\text{g}/\text{kg}$ (Phe level of a sesame oil), showing a broad range of PAHs concentrations among the different types of edible oils. Phe, Fluo and Pyr were the three kinds of compounds with the highest concentrations, with the average values of 85 samples were 8.60, 5.44 and 4.23 $\mu\text{g}/\text{kg}$

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