Detection of Polycyclic Aromatic Hydrocarbons in Commonly Consumed Edible Oils and Their Likely Intake in the Indian Population

Manoj K. Pandey, Krishn K. Mishra, Subhash K. Khanna, and Mukul Das*

Food Toxicology Laboratory, Industrial Toxicology Research Centre, Mahatma Gandhi Marg, Lucknow-226001, India

ABSTRACT: Edible oils such as coconut, groundnut, hydrogenated vegetable, linseed, mustard, olive, palm, refined vegetable, rice bran, safflower, sesame, soybean, and sunflower were analyzed for the presence of light and heavy polycyclic aromatic hydrocarbon (PAH) residues using liquid–liquid extraction, cleanup on a silica gel column, and resolution and determination by HPLC using a fluorescence detector. Ten PAH viz. acenaphthene, anthracene, benzo(a)pyrene, benzo(e)pyrene, benz(ghi)perylene, chrysene, coronene, cyclopenta(def)phenanthrene, phenanthrene, and pyrene were monitored. Analysis of 296 oil samples showed that 88.5% (262) samples were contaminated with different PAH. Of 262 contaminated edible oil samples, 66.4% of the samples showed PAH content of more than the 25 μg/kg recommended by the German Society for Fat Science. The total PAH content was highest in virgin olive oil (624 μg/kg) and lowest in refined vegetable oils (40.2 μg/kg). The maximum content (265 μg/kg) of heavy PAH was found in olive oil and the minimum (4.6 μg/kg) in rice bran oil. Phenanthrene was present in 58.3% of the oil samples analyzed, followed by anthracene (53%). Among the heavy PAH, benzo(e)pyrene was observed in 31.2% of the samples followed by benzo(a)pyrene (25.5%). The intake of PAH was highest through olive oil (20.8 μg/day) followed by soybean oil (5.0 μg/day) and lowest through refined vegetable oil (1.3 μg/day). Based on these monitoring studies, international and national guidelines for permissible levels of PAH can be prepared so as to restrict the intake of these toxic contaminants.

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KEY WORDS: Heavy PAH, HPLC, Light PAH, and vegetable oils.

Polycyclic aromatic hydrocarbons (PAH) are highly stable contaminants that occur in air, water, soil, and food (1). PAH enter into the environment largely as releases into the air from volcanoes, forest fires, wood burning, and automobile exhaust. These compounds enter the surface water through discharges from industrial plants and wastewater treatment plants and may be released into the soil at hazardous waste sites (2,3). The primary sources of exposure to PAH are through the inhalation of tobacco smoke, wood smoke, ambient air, and by eating contaminated food materials (2,3). Some of the PAH have been shown to have carcinogenic potential (4,5). Benzo(a)pyrene [B(a)P], one of the carcinogenic PAH, has been found at variable concentrations in several foods and beverages including water, cereals, oils, and smoked meats (2,3,6). Possible sources of PAH contamination of vegetable oils are (i) atmospheric deposition onto the plant material, (ii) direct drying of oilseeds with combustion smoke, (iii) contamination through the extraction solvent, or (iv) uptake by the oilseed plants through contaminated soil (1,7).

Human exposure to PAH is high through food than through ambient air or drinking water (8). Edible oils and fats are the most important contributing sources of PAH because of their lipophilic nature. Moreover, higher levels of PAH in foods produced with added oil implicate fat origin as one of the prime sources of PAH (6,9). The presence of PAH in vegetable oils such as corn oil, coconut oil, grapeseed oil, groundnut oil, olive oil, palm oil, pumpkinseed oil, rapeseed oil, rice bran oil, soybean oil, and sunflower oil have been reported (6,10–15). In general, the levels of detected PAH in refined edible oils (corn, palm, rapeseed, soybean, and sunflower) are in the range of a few μg/kg, although higher levels have been found in virgin and unrefined oils (7,10,15–18). Although several reports exist on the presence of PAH in edible oils from the Western countries, PAH residues in commonly used edible oils from Indian markets have not been studied. Hence, we have attempted to assess the PAH residues in commonly consumed edible oils and to assess their likely intake assessment in Indian households.

MATERIALS AND METHODS

Chemicals. PAH kit comprising standard references was purchased from Aldrich Chemical Co. (Milwaukee, WI). HPLC grade acetonitrile, tetrahydrofuran, dichloromethane, dimethylsulfoxide (DMSO) were procured from E. Merck (Mumbai, India). Cyclohexane, *n*-heptane, and *n*-hexane (all HPLC grade) were purchased from Qualigens Fine Chemicals (Mumbai, India). Silica gel G was purchased from ACME Synthetic Chemicals (Mumbai, India). The RC4 filter was a product of Sartorius Biotech Pvt. Ltd. (Banglore, India).

*To whom correspondence should be addressed at Food Toxicology Laboratory, Industrial Toxicology Research Centre, Mahatma Gandhi Marg, P.O. Box #80, Lucknow-226001, India.
E-mail: mditrc@rediffmail.com or mditrc@hotmail.com

Procurements of edible oils. A total of 296 edible oil samples consisting of hydrogenated vegetable oils (HVO), linseed oil, mustard oil, olive oil, palm oil, refined vegetable oils (RVO), rice bran oil, safflower oil, sesame oil, soybean oil, and sunflower oil were purchased from local retail markets of Lucknow. Coconut oil and groundnut oil samples were purchased from Tiruvananthapuram (Kerala) and Ahemdabad (Gujarat), respectively.

Characteristics of individual standard PAH. The excitation and emission maxima of the 10 individual standard PAH namely acenaphthene (Acen), anthracene (Anth), B(a)P, benzo(e)pyrene (B(e)P), benz(ghi)perylene (B(ghi)P), chrysene (Chry), coronene (Coro), cyclopenta(def)phenanthrene (C(d)P), phenanthrene (Phen) and pyrene (Pyr), were measured on a model LS-50 Bspectrofluorimeter (Perkin–Elmer, Uberlingen, Germany).

Resolution of standard PAH. An HPLC instrument (m/s Waters Associates, Vienna, Austria) equipped with a dual pump (model #510), Rheo dyne injector with 20-µl loop and fluorescence detector (model # 474) was used. The reversed-phase column used for the analysis was a C-18 Lichrocart (5 µm, 125-mm × 4mm; E. Merck) with a precolumn of the same type. The column was eluted at ambient temperature (25°C) with a 40-min linear gradient (curve 6) of 55 to 100% acetonitrile in water as solvent, at a flow rate of 1 mL/min. The eluate was monitored on the fluorescence detector at excitation/emission wavelengths of 250/335 nm (0–15 min) for Acen, 250/390 nm (15–18.5 min) for Phen, Anth and C(d)P; 285/390 nm (18.5–22 min) for Pyr; 242/380 nm (22–24 min) for Chry; 350/405 nm (24–27.5 min) for B(e)P; 320/427 nm (27.5–33 min) for B(a)P and B(ghi)P; and 329/443 nm (33–40 min) for Coro, respectively. Although the excitation/emission wavelengths for the individual Phen, Anth and C(d)P were 300/363, 260/398 and 308/362 nm, respectively, a common wavelength of 250/390 nm was used for these compounds as their retention times (RT) were quite close. Furthermore, B(a)P and B(ghi)P showed common excitation/emission wavelengths but different RT values. The chromatogram was recorded and processed using Waters Millenium® 32 software (Waters, Milford, MA).

Extraction and clean up. The extraction and cleanup procedure of PAH from edible oils was essentially the same as described by Menichini *et al.* (13) with slight modifications. Essentially, 10 g of edible oil sample was weighed and dissolved in 20 mL of *n*-heptane and extracted with 10 mL of DMSO in a separating funnel. Extraction with 10 mL of DMSO was repeated three times and the lower DMSO layers were separated and pooled. Cold distilled water (60 mL) was added slowly to the pooled DMSO extract and the extract was re-extracted three times with 50 mL of cyclohexane. The upper cyclohexane layers were carefully collected, pooled, and washed twice with 100 mL each of distilled water. The cyclohexane extract was concentrated to 20 mL and the entire volume was passed through a glass column (20 × 2.2 cm) containing 5 g of silica gel with a top layer of 1 g of anhydrous sodium sulfate, which was earlier pre-equilibrated with 10 mL of cyclohexane and 5 mL of dichloromethane. The elution was carried out with 10 mL of cyclohexane followed by 5 mL of dichloromethane and the eluate was collected and evaporated to dryness. The final residue was dissolved in 1 mL of acetonitrile and passed through 0.2 µm RC4 filter prior to HPLC analysis. Each solvent blank used for PAH extraction was scanned for the presence of interference peaks.

Recovery and minimum detection limits of standard PAH. Control edible oil was used, as vehicle to spike known quantities of a mixture of 10 PAH and the recovery of individual PAH was noted after extraction and clean up steps as just described. Individual standard PAH, namely, Acen, Phen, Anth, C(d)P, Pyr, Chry, B(e)P, B(a)P, B(ghi)P and Coro, were spiked with 12.5, 8.3, 4.2, 17, 8, 17, 50, 4, 8, and 8 µg/g of the respective PAH. The percentage recovery and the minimum detection limits of the standard PAH were calculated.

Confirmation of sample peaks. Sample PAH peaks in the HPLC chromatograms were confirmed by co-injecting the standard reference PAH and the enhanced peak areas were matched against those of the respective standard PAH. For additional validation of the HPLC peaks, both the sample extract and the standard PAH were separately injected and the eluted fractions were collected. UV spectra of the respective eluted peaks were matched with those of the corresponding standard PAH peak on a Bio Lamda 20 double-beam spectrophotometer (PerkinElmer). Emission spectra of the eluted fraction of the respective peaks were also matched with the eluted fractions of standard PAH on a spectrofluorimeter (LS 50 B).

Calculations for intake of PAH from different edible oils. Intake of PAH was calculated based on 30 g of oil consumption per day per capita in Indian households (19). In addition, PAH intake values through edible oil and fats were back-calculated by using the formula

$$\text{intake (µg/d/adult)} = \frac{[\text{Mean of PAH content (µg)} \times 30\ \text{(g)}]}{1000\ \text{(g)}} \quad [1]$$

RESULTS

The HPLC resolution profile following fluorometric detection of the 10 standard PAH is shown in Figure 1. The lowest RT was observed for Acen (14.6 min) and the highest was for Coro (36.8 min). The minimum detectable limit of individual standard PAH ranged from 0.1 to 4.0 ng, and the recovery of individual PAH in spiked oil samples ranged from 58 to 99% (Table 1). Figure 2 shows the HPLC profile of PAH present in an unknown edible oil sample. The chromatogram was matched with that of the standard PAH chromatogram for RT. In addition, the sample extract was co-injected with the standards of suspected PAH for confirmation (Fig. 2B). At the same time, eluted peaks of oil samples having specific RT were collected and matched with the UV and emission spectra of the standard PAH to confirm the presence of PAH in the unknown samples (Table 2). This procedure was adopted for all the edible oil samples analyzed in the present study.

TABLE 1
Percentage Recovery and Minimum Detection Limits of Individual Standard Polycyclic Aromatic Hydrocarbons (PAH)

Name	% Recovery	Minimum detection limit (ng)
Acenaphthene	58	4.0
Anthracene	73	0.1
Benzo(a)pyrene	89	1.0
Benzo(e)pyrene	78	2.0
Benzo(ghi)perylene	90	0.1
Chrycene	99	0.3
Coronene	92	1.0
4H-Cyclopenta-(def)phenanthrene	72	0.2
Phenanthrene	99	0.1
Pyrene	95	2.0

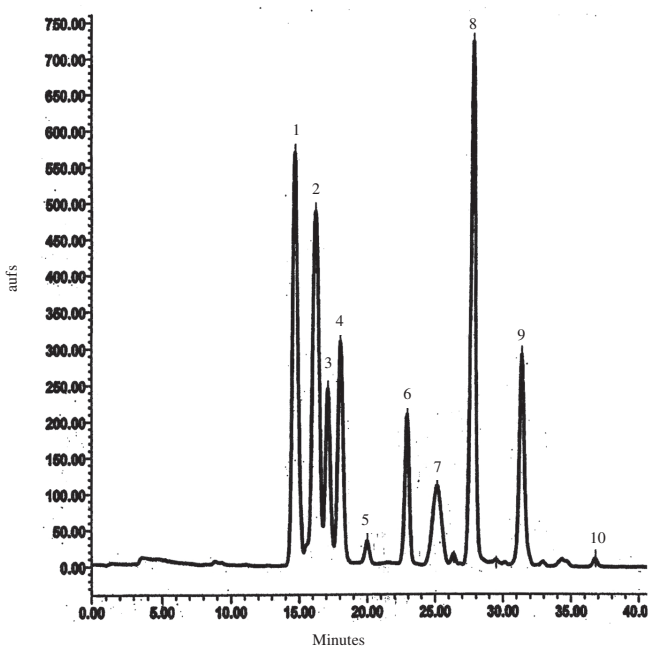

FIG. 1. HPLC separation of a mixture of standard polycyclic aromatic hydrocarbons (PAH). 1. Acenaputhene (Acen); 2. Phenanthrene (Phen) 3. Anthracene (Anth); 4. Cyclopenta(def)phenanthrene [C(d)P]; 5. Pyrene (Pyr); 6. Chrysene (Chry); 7. Benzo(e)pyrene [B(e)P]; 8. Benzo(a)pyrene [B(a)P]; 9. Benz(ghi)perylene [B(ghi)P]; 10. Coronene (Coro).

Details of the resolution on HPLC is described in the Material and Methods section. The eluate was monitored on a fluorescence detector at the excitation/emission wavelengths of 250/335 nm (0–15 min) for Acen, 250/390 nm (15–18.5 min) for Phen, Anth, and C(d)P, 285/390 nm (18.5–22 min) for Pyr; 242/380 nm (22–24 min) for Chry; 350/405 nm(24–27.5 min) for B(e)P; 320/427 nm (27.5–33 min) for B(a)P and B(ghi)P; and 329/443 nm (33–40 min) for Coro, aufs, absorbance units full scale.

The mean values of total, light, and heavy PAH in different edible oil samples are given in Table 3. Analysis of 296 oil samples showed that almost 11.5% were devoid of any PAH, while 88.5% samples were found to be contaminated with different PAH. Imported virgin olive oil samples showed the highest levels of total PAH content (624 μg/kg) including light (359 μg/kg) and heavy PAH (265 μg/kg). Among other edible oils showing total PAH content of 85 to 150 μg/kg were, palm oil (86.9) sesame oil (93), mustard oil (92.5), rice bran oil (94), sunflower oil (114) and soybean oil (149 μg/kg). The next category of oils showing a total PAH content ranging between 60 to 70 μg/kg, were hydrogenated vegetable oils (HVO) (60.1), coconut oil (63.2), safflower oil (65.0), and groundnut oil (67.5). Linseed oil samples showed a total PAH content of 55.7 μg/kg while the minimum levels of total PAH content of 40.2 μg/kg (with 39.9 μg/kg light and 4.7 μg/kg of heavy PAH) was found in samples of refined vegetable oils (RVO).

Table 4 depicts the content of six individual light and four heavy PAH in different edible oils. Acen was found to be present in 43.5% of the oil samples with a mean value of 16.0 μg/kg. The mean value of 67.0 μg/kg for Acen content was maximally observed in sunflower oil while the lowest (1.3 μg/kg) in olive oil samples. Anth was detected in 53% samples with a mean value of 12.0 μg/kg. C(d) P was present in 44.6% oils with a mean value of 5.3 μg/kg. Chry and Pyr were present in 51.5 and 41.9% samples with a respective mean value of 79.9 and 37.2 μg/kg. Phen was present in 58.3% oils, with a mean value of 12.8 μg/kg. Among 4 heavy PAH; Coro, B(ghi)P and B(a)P were present in 22.1 to 25.5% samples while B(e)P was noted in 31.2% of edible oil samples. The mean content of B(a)P and B(ghi)P was 1.9 μg/kg and 2.1 μg/kg, respectively. The mean Coro content detected in oil samples was 4.5 μg/kg. The maximum average content among heavy PAH was that of B(e)P (27.1 μg/kg) (Table 4).

The pattern of total, light and heavy PAH in different edible oils, in terms of maximum recommended limits of German Society of Fat Sciences (GSFS) is given in Table 5. Among 262 analyzed edible oils, 66.4% samples exceeded the GSFS limit

TABLE 2
UV and Fluorescence Emission Characteristics of Specific Sample Peaks Eluted by HPLC: Comparison with Standard PAH Having Similar Retention Times

	Peak 1		Peak 3		Peak 7	
	UV λ_{max} [a] (nm)	Emission λ_{max} [b] (nm)	UV λ_{max} [a] (nm)	Emission λ_{max} [b] (nm)	UV λ_{max} [a] (nm)	Emission λ_{max} [b] (nm)
Standard	226.6	334.3	251.9	402.7	288.1	420.0
Sample	225.5	ND	251.7	402.6	288.1	437.0

[a]Fractions for peaks 1, 3, and 7 from Figure 2A were collected and the spectra were recorded.
[b]Fractions for peaks 1, 3, and 7 from Figure 2A were collected and excited at 225.5, 251.7, and 288.1 nm, respectively, followed by determination of emission spectra.
ND, not detected.

TABLE 3
PAH Content (μg/kg) in Different Edible Oils

Edible oils	Samples analyzed[a]	Samples with PAHs	Mean PAHs content (μg/kg) Total	Light	Heavy
Coconut	30	27	63.2	31.6	28.2
Groundnut	30	27	67.5	55.1	12.4
HVO	30	29	60.1	46.7	8 .5
Linseed	8	8	55.7	49.2	6.4
Mustard	30	30	92.5	60.8	35.2
Olive	6	6	624	359	265
Palm	24	24	86.9	80.0	7.0
Rice bran	5	5	94.0	89.4	4.6
RVO	30	23	40.2	39.9	4.7
Safflower	5	5	65.0	50.9	14.0
Sesame	28	28	93.0	82.4	10.5
Soybean	30	28	149	140	10.5
Sunflower	30	22	114	108	6.3
Total	296	262	123.5	91.7	31.8

[a]11.5% samples of oil showed absence of PAH residues. HVO, hydrogenated vegetable oils; RVO, refined vegetable oils; for other abbreviation see Table 1.

of total PAH (25 μg/kg) and 37% samples exceeded the recommended limit of 5 μg/kg for heavy PAH. In the individual oil categories, all the samples of olive oil and safflower exceeded total PAH limit while groundnut, rice bran, linseed, HVO, mustard, soybean, and sesame oil belonged to the category in which 60 to 75% samples exceeded the total PAH limit of GSFS. Coconut had the least number of samples (40.7%) exceeded GSFS limit of 25 ug/kg for total PAH.

The figures for likely daily intake of PAH from different edible oils are given in Table 6. Based on a daily edible oil consumption figure of 30 g/d per capita in the Indian population, olive oil constituted the maximum risk oil category with a mean intake value of 20.8 μg/d. Soybean oil was the next downward category of edible oil, having a mean intake value of 5.0 μg/d. The mean intake of 1.3 μg/d for PAH from the RVO category was the lowest.

DISCUSSION

The Prevention of Food Adulteration Act of India has no clause on the permissible levels for PAH in edible oils and other foods and this seems to be the case in many other countries. It is baffling to note that in spite of several reports of high levels of PAH in edible oils, their toxicity and even carcinogenic potential of some of the PAH (4,5,8), the regulatory directive fixing legal limits for these contaminants has not been forthcoming from even most developed countries of the world. GSFS has taken the lead in setting their own recommended limit of 25 ppb for total PAH and 5ppb for heavy PAH (15,17, 20,21). Recently, Spain has set a limit of 2 ppb for B(a)P in olive residue oils (16).

B(a)P is often taken as a prototype compound for PAH contamination. The total dietary intake of B(a)P in UK was assessed to have come from two food groups, namely cereals and oils/fats (6,22). Several investigators believe that fats and oils represent one of the major sources of PAH in the diet because of the lipophilic nature (12,22–23). Among a large number of vegetable oils analyzed by Balenoic *et al.* (24), almost all showed total PAH content of above 25 ppb. Our results also reveal that all the analyzed edible oil samples do show the presence of PAH, though 33.6% samples may fall within GSFS limit of 25 ppb and 66.4% exceed it. The present study is the first report on the presence of PAH in mustard oil, the second most popular oil in India, as well as in other typical oil used in Indian markets such as linseed, safflower and sesame oil.

The major dietary intake of some PAH in the UK was estimated to be from oils and fats, with 28% from butter, 20%

TABLE 4
Pattern of Individual Light and Heavy PAH[a] in Different Edible Oils

Oils	Acen		Phen		Anth		C(d) P		Pyr		Chry		B(e)P		B(a)P		B(ghi)P		Coro	
(Sample no)	No.	Content[c]	No.	Content	No.	Content	No.	Content	No.	Content	No.	Content	No.	Content	No.	Content	No.	Content	No	Content
Coconut (27)	11	15.0	16	5.0	8	2.0	13	1.0	12	3.3	9	2.0	8	5.0	6	0.8	1	0.07	3	22.2
Groundnut (27)	4	13.0	13	12.0	20	66.0	13	2.2	13	9.0	14	778	11	10.0	4	0.2	6	3.0	5	3.0
HVO (29)	18	5.2	17	6.2	17	6.2	12	3.0	15	3.0	21	5.0	12	2.0	9	2.0	12	2.0	10	1.0
Linseed (8)	4	7.6	4	3.0	5	6.8	6	9.4	3	1.7	6	20.7	4	0.8	3	1.5	5	2.1	3	2.1
Mustard (30)	8	12.0	16	13.0	16	10.0	17	5.0	6	2.0	14	22.0	7	14.0	7	0.3	4	0.5	5	20.0
Olive (6)	2	1.3	3	39.1	3	2.5	2	10.5	4	265	2	32.5	4	250	2	3.3	2	1.8	1	4.4
Palm oil (24)	11	11.4	13	5.0	5	1.0	6	5.0	8	41.0	6	3.0	1	0.02	3	2.5	7	3.0	2	0.2
Rice bran (5)	1	10.3	2	1.3	1	0.6	2	4.1	3	73.1	—	—	—	—	1	4.5	—	—	1	0.2
RVO (23)	15	7.0	15	7.0	15	9.0	13	6.0	10	27.0	19	10.0	11	5.0	9	4.0	12	3.0	9	2.0
Safflower (5)	4	11.6	1	3.1	3	5.6	3	5.6	1	0.8	5	26.6	2	1.4	2	2.6	4	9.3	2	0.8
Sesame (28)	7	2.0	9	12.0	9	4.0	6	7.0	13	74.0	9	23.0	4	52.0	11	2.1	4	1.1	6	2.0
Soybean (28)	17	13.0	28	31.0	24	19.0	13	3.0	16	12.0	20	23.0	13	11.0	6	0.40	5	0.40	6	0.2
Sunflower (22)	12	67.0	16	17.0	13	10.0	11	3.0	6	0.95	10	1.0	5	1.0	4	0.13	2	0.8	5	0.2
Total (262)	114	16.0	153	12.8	139	12.0	117	5.3	110	37.2	135	79.9	82	27.1	67	1.9	64	2.1	58	4.5
	(43)[d]		(58)[c]		(53)[c]		(44)		(41)		(51)		(31)		(25)		(24)		(22)	

[a]Acen, acenaphthene; Phen, phenanthrene; Anth, anthracene; C(d)P, cyclopenta(def)phenanthrene; Pyr, pyrene; Chry, chrysene; B(e)P, benzo(e)pyrene; B(a)P, benzo(a)pyrene; benz(ghi)perylene; Coro, coronene; for other abbreviations see Tables 1 and 3.
[b]No., number of samples showing the presence of individual PAH.
[c]Content of individual PAHs are represented as mean value (μg/kg).
[d]Data in parentheses represent the percent of total oil samples containing individual PAH.

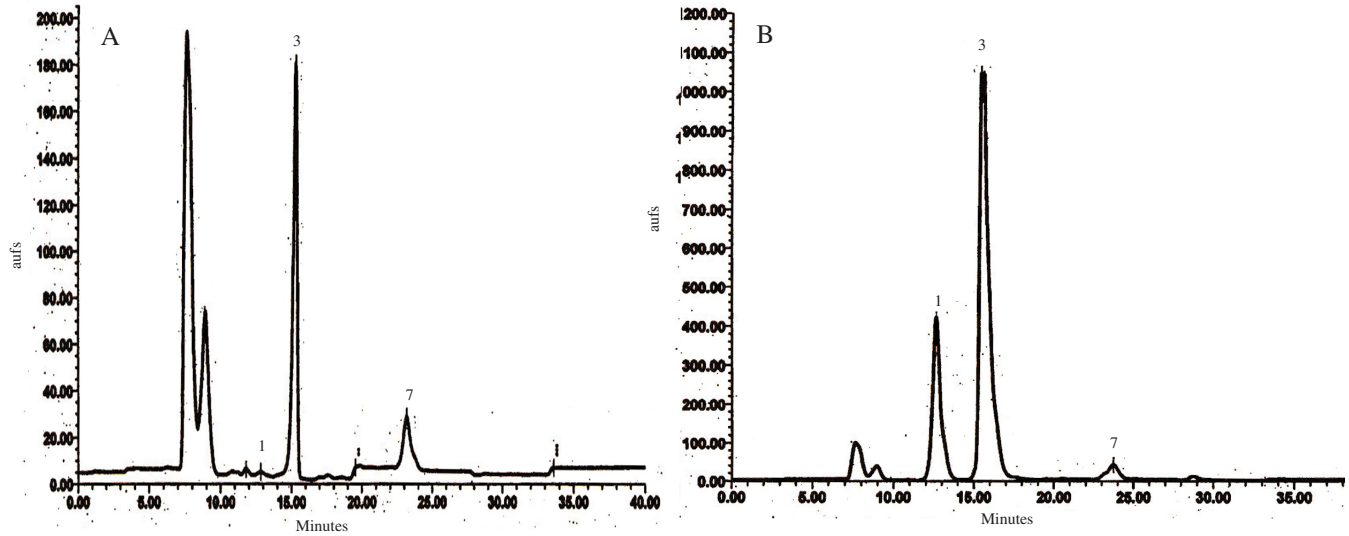

FIG. 2. HPLC profile of PAH in an unknown edible oil sample. (A) Peaks 1, 3, and 7 were identified as Acen, Anth, and B(e)P, respectively. (B) Above 1, 3, and 7 were confirmed with coinjection of Acen, Anth, and B(e)P, respectively. Details of peaks confirmation is described in the Material and Methods section. The eluate was monitored on a fluorescence detector at the excitation/emission wavelengths of 250/335 nm (0–15 min) for Acen; 250/390 nm (15–18.5 min) for Phen, Anth, and C(d)P; 285/390 nm (18.5–22 min) for Pyr; 242/380 nm (22–24 min) for Chry, 350/405 nm (24-27.5 min) for B(e)P; 320/427 nm (27.5–33 min) for B(a)P and B(ghi)P; and 329/443 nm (33–40 min) for Coro. For abbreviations see Figure 1.

from cheese, and 17% from margarine, in respective dietary survey groups (22). In Sweden, the annual intake per person of the sum of 11 PAH was about 1 mg (2). In Germany, the total dietary intake of B(a)P was determined to be 0.14–1.0 μg/wk (25), while in Italy; it was 0.2 μg/wk (26). In the U.S., the total dietary intake of PAH has been fixed at 0.16–1.6 μg/d/person for B(a)P (23). In our study, maximum dietary intake of total PAH is through imported olive oil (20.8 μg/d) with maximum dietary intake of heavy PAH (8.8 μg/d) and light PAH (11.9 μg/d). In case of soybean and sunflower oil the intake of total PAH was 5.0 and 3.8 μg/d respectively. Keeping in view the daily consumption of edible oil by individuals and the potential toxicity and carcinogenecity of the PAH, the PAH levels in different edible oils should be kept as low as possible.

Crude coconut oils have been shown to contain a very high range of total PAH, from 2600 to 3700 ppb, compared with the corresponding refined products (7). Among 11 categories of edible oils sampled, most of the unrefined oils contained more than 25 ppb of total PAH (20). Similarly, virgin olive oil, which is generally unrefined, was shown to contain high levels of total

TABLE 5
Pattern of Total, Light, and Heavy PAH in Different Edible Oils in Terms of Maximum Recommended Limits of German Society for Fat Science (GSFS)[a]

Edible oils	Analyzed samples with PAH	No. of samples exceeding the GSFS limits[b]		
		Total	Light	Heavy
Coconut	27	11 (40.7)	7 (25.9)	9 (33.3)
Groundnut	27	19 (70.3)	19 (70.3)	7 (25.9)
HVO	29	21 (72.4)	22 (75.8)	12 (41.4)
Linseed	8	5 (62.5)	5 (62.5)	4 (50.0)
Mustard	30	21 (70.0)	21 (70.0)	7 (23.3)
Olive	6	6 (100.0)	5 (83.3)	6 (100.0)
Palm	24	18 (75.0)	18 (75.0)	12 (50.0)
Rice bran	5	3 (60.0)	3 (60.0)	1 (20.0)
RVO	23	12 (52.0)	12 (52.0)	5 (21.7)
Safflower	5	5 (100.0)	4 (80.0)	3 (60.0)
Sesame	28	21 (75.0)	21 (75.0)	13 (46.4)
Soybean	28	20 (71.4)	17 (60.7)	11 (39.2)
Sunflower	22	12 (54.5)	11 (50.0)	7 (31.8)
Total	262	174 (66.4)	165 (62.9)	97 (37.0)

[a]The GSFS prescribed limit for light PAHs is 25 μg/kg and for heavy PAH is 5 μg/kg. For other abbreviations see Tables 1 and 3.
[b]Values in parentheses indicate the percent of samples.

TABLE 6
Daily Intake of Total, Light, and Heavy PAH from Different Edible Oils[a]

Edible oils	Analyzed samples with PAH	Mean intake value (μg/day)[a]		
		Total	Light	Heavy
Coconut	27	2.1	1.1	0.94
Groundnut	27	2.3	1.8	0.47
HVO	29	2.0	1.5	0.28
Linseed	8	1.9	1.6	0.21
Mustard	30	3.1	2.0	1.2
Olive	6	20.8	11.9	8.8
Palm	24	2.9	2.7	0.23
Rice bran	5	3.1	3.0	0.15
RVO	23	1.3	1.3	0.15
Safflower	5	2.1	1.7	0.46
Sesame	28	3.1	2.7	0.35
Soybean	28	5.0	4.6	0.35
Sunflower	22	3.8	3.6	0.21

[a]The intake values are based on 30 g of oil consumption per day per adult. For abbreviations see Tables 1 and 3.

PAH (15). Our data confirms that high levels of PAH in virgin olive and coconut oil samples as observed earlier (13,17) and least levels in the samples of RVO. This may be due to the fact that crude oil preparation requires special treatment like drying in a kiln by direct heating with air, that contains combustion gas, which may generate PAH (17). Furthermore, PAH in edible oils could arise from atmospheric deposition to plants or through contamination of extraction solvents use (1,7). It has been shown that specific refining steps like deodorization may drastically reduce the content of these contaminants and should be an integral part of the edible oil refining process so that the risk of PAH contamination can be minimized (17). Thus, good manufacturing practices should be adopted right from the initial phase of the collection of seeds and continue on through the processing of oilseeds so as to bring PAH contamination in edible oils down to the minimum achievable levels. International and national guidelines for permissible levels of PAH in edible oils may have to be prepared and adhered to in order to restrict intake of these toxic substances.

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REFERENCES

1. Welling, P., and B. Kaandorp, Determination of polycyclic aromatic hydrocarbon (PAH) in edible vegetable oils by liquid chromatography and programmed fluorescence detection, *Z. Lebensmi. Unters. Forsch. 183*:111–115 (1986).
2. DeVos, R.H., W. Van Dokkum, W. Schouten, and P. DeJong-Berkhout, Polycyclic aromatic hydrocarbon in Dutch total diet samples (1984–86), *Food Chem. Toxicol. 28*:263–268 (1990).
3. Gomma, E., A. Gray, J.I. Rabie, S.L.B. Clemente, and A.M. Booren, Polycyclic aromatic hydrocarbon in smoked food products and commercial liquid smoke flavorings, *Food Addit. Contam. 10*:503–521(1993).
4. International Agency for Research on Cancer, *Monographs on the Evaluation of the Carcinogenic risk of Chemicals to Humans. Polynuclear Aromatic Compounds. Part 1: Chemicals, Environment and Experimental Data*, IARC, Lyons, 1983, Vol. 32.
5. International Agency for Research on Cancer, *Monographs on the Evaluation of the Carcinogenic risk of Chemicals to Humans. Overall Evaluation of Carcinogenecity: An Updating of IARC Monographs*, Vol. 1–42, IARC, Lyons, 1987.
6. Dennis, M.J., R.C. Massey, G. Cripp, I. Venn, N. Horwanth, and G. Lee, Factor affecting the PAHs content of cereal, fats and other food products, Food Additives and Contaminants, 8: 517–530 (1991).
7. Larsson, B.K., A.T. Eriksson, and M. Cervenka, Polycyclic aromatic hydrocarbon in crude and deodorized vegetable oils, *J. Am. Oil Chem. Soc. 64*:365–370 (1987).
8. International Programme on Chemical Safety, Environmental Health Criteria 202, Selected Non- Heterocyclic Polycyclic Aromatic Hydrocarbons, IPCS, Geneva, Switzerland, 1998.
9. Lawrence, J.F., and D.F. Weber, Determination of polycyclic aromatic hydrocarbons in some Canadian commercial fish, shellfish, and meat products by liquid chromatography with confirmation by capillary gas chromatography–mass spectrometry, *J. Agric. Food Chem. 32*:789–794 (1984).
10. Cejpek, K., J. Hajslova, V. Kocourek, M. Tomaniova, and J. Cmolik, Changes in PAH during production of rapeseed oil, *Food Addit. Contamin. 15*:563–574 (1998).
11. Grimmer, G., and A. Hildenbrandt, Content of polycyclic aromatic hydrocarbon in crude vegetable oils, *Chem. Ind. 25*:2000–2002 (1967).
12. Hopia, A., H. Pyysalo, and K. Wickstorm, Margarines, Butter and vegetable oils as sources of polycyclic aromatic hydrocarbons, *J. Am. Oil Chem. Soc. 63*:889–893 (1986).
13. Menichini, E., A. Bocca, F. Merli, D. Ianni, and F. Monfredini, Polycyclic aromatic hydrocarbons in olive oils on the Italian market, *Food Addit. Contam. 8*:363–367 (1991).
14. Pupin, A. M., M.C. Toledo, Benzo(a)pyrene in Brazilian vegetable oils, *Ibid. 13*: 639–646 (1996).
15. Speer, K., E. Steeg, P. Hortzmann, Th. Kühn, A. Montag, Determination and distribution of polycyclic aromatic hydrocarbons in native vegetable oils, smoked fish products, mussels and oysters, and bream from the river Elbe. *J. High Res. Chromatogr. 13*:104–111 (1990).
16. Barranco, A., R.M. Alonso-Salces, A. Bakkali, L. A. Berrueta, B. Gallo, F.Vicente, and M. Sarobe, Solid phase clean -up in the liquid chromatographic determination of polycyclic aromatic hydrocarbons in edible oils, *J. Chromatogr. A. 988*:33–40 (2003).
17. Swetman, T., S. Head, and D. Evans, Contamination of Coconut oil by PAH, *INFORM 10*:706–712 (1999).
18. Thomas, A., Uber die entfemung von schadestoffen bei der Dampfung von Speiseolen und-fetten, *Fette Seifen Anstrichm. 84*:133–136 (1982).
19. NNMB (National Nutrition Monitoring Bureau); Nutritional status of rural population. National Institution of Nutrition/Indian Council of Medical Research, Hyderabad, India (1996).
20. Gertz, Ch., and H. Kogelheide, Undersunchung und Beurtilung von PAH in Speisefetten und Ölen, *Fat Sci. and Technol. 96*:175–180 (1994).
21. Moret, S., and L.S. Conte, Polycyclic aromatic hydrocarbons in edible fats and oils: occurrence and analytical methods, *J. Chromatogr. A. 882*:245–253 (2000).
22. Dennis, M. J., R. C. Massey, D. J. McWeeny, M.E. Knowles, and D. Watson, Analysis of polycyclic aromatic hydrocarbon in UK total diets, *Food Chem. Toxicol. 21*:569–574. (1983).
23. Santodonato, J., D. Basu, and P. Howard, Multimedia human exposures and carcinogenic risk assessment for environmental PAH, in *Polynuclear Aromatic Hydrocarbons: Chemistry and Biological Effects*, edited by A. Bjorseth and A.J. Dennis, Battle Press, Columbus, OH, 1980, pp. 435–454.
24. Balenovic, J., I. Petrovic, and M. Perkovac, *Proceedings of Euro Food Chem VIII*, Vienna, Austria, 18–20 September, 2: 275 (1995).
25. State Committee for Air Pollution Control (Cancer Risk from Air Pollution). Development of evaluation criteria for carcinogenic pollutants. Minister for Environmental and Agriculture for Northrhine-West Falia, Dusseldorf, 1992 (in German), pp. 71.
26. Menichine, E., Urban air pollution by polycyclic aromatic hydrocarbons: levels and sources of variability, *Sci. Tot. Environ. 116*:109–135 (1991).

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