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Research Paper / Araştırma Makalesi

Polycyclic Aromatic Hydrocarbons (PAHs) Contamination Levels in Some Commercial Olive Oils Sold on the Markets in Giresun, Türkiye

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ABSTRACT

Olive oil is widely consumed worldwide and well-known for its health beneficial effects. However, polycyclic aromatic hydrocarbons (PAHs) contamination in olive oils has been a serious concern and knowledge regarding PAH concentrations in olive oils is scarce. The aim of this study was the determination and evaluation of PAH contamination levels in olive oils in the market. In this study, commercial olive oils (n=14) available in the markets of Giresun (Türkiye), including extra virgin and riviera olive oil samples, were investigated. Samples were analyzed by a gas chromatography unit coupled with a mass spectrometer. Significant amounts of PAHs were determined in all olive oil samples. The PAH contents of the samples varied between 0.04 µg kg^{-1} and 4637.13 µg kg^{-1} with the mean content of 264.61 µg kg^{-1}. Regarding benzo[a]pyrene, 64.28% of the samples was incompatible with the EU requirements. None of the samples were compatible with the EU requirements in terms of PAH4 components (benzo[a]pyrene, benzo(a)anthracene, chrysene, and benzo[b]fluoranthene). Results showed that commercial olive oils studied in this research might contain PAHs categorized under potential carcinogens.

Keywords: Gas chromatography-mass spectrometer (GC-MS), Olive oil, Polycyclic aromatic hydrocarbons (PAHs), Benzo[a]pyrene, PAH4

Giresun'da Satılan Bazı Ticari Zeytinyağlarının Polisiklik Aromatik Hidrokarbon (PAH) Kontaminasyon Düzeyleri

ÖZ

Zeytinyağı dünya çapında yaygın olarak tüketilmektedir ve sağlığa yararları ile bilinmektedir. Ancak zeytinyağındaki polisiklik aromatik hidrokarbon (PAH) kontaminasyonu ciddi bir endişe kaynağıdır ve bu yağların PAH konsantrasyonlarına ilişkin bilgi azdır. Bu çalışmanın amacı, piyasada bulunan zeytinyağlarındaki PAH kontaminasyon düzeylerinin belirlenmesi ve değerlendirilmesidir. Bu çalışmada Giresun ili marketlerinden rastgele seçilen sızma zeytinyağı ve riviera zeytinyağı örnekleri (n=14) incelenmiştir. Numuneler, kütle spektrometresi ile birleştirilmiş gaz kromatografisi ünitesi ile belirlenmiştir. Yağ örneklerinin tamamında önemli miktarda PAH kalıntısı tespit edilmiştir. PAH tespit edilen örneklerde PAH miktarları 0.04 µg kg^{-1} ile 4637.13 µg kg^{-1} (ortalama 264.61 µg kg^{-1}) arasında değişmektedir. Benzo[a]piren ile ilgili olarak numunelerin %64.28'i AB gereklilikleriyle uyumlu çıkmamıştır. Zeytinyağı örneklerinin hiçbiri PAH4 bileşenleri (benzo[a]piren, benzo(a)antrasen, krizen ve benzo[b]floranten) açısından AB gerekliliklerine uygun çıkmamıştır. Sonuçlar, bu araştırmada incelenen ticari zeytinyağlarının potansiyel kanserojenler kategorisinde yer alan PAH'ları içerebileceğini göstermiştir.

Anahtar Kelimeler: Gaz kromatografisi-kütle spektrometresi (GC-MS), Zeytinyağı, Polisiklik aromatik hidrokarbonlar (PAH'lar), Benzo[a]piren, PAH4

INTRODUCTION

Olive oil is a product from the fruit of the olive tree (*Olea europaea* L.) and it is the primary source of fat in a Mediterranean diet. Olive oil is well-known for its unique composition of monounsaturated fats, antioxidants, and anti-inflammatory properties. Some of the key health benefits of olive oil include reducing the risk of heart diseases [1], containing of powerful antioxidants such as vitamin E and polyphenols [2], anti-inflammatory effects [3], cancer prevention [4], brain health and cognitive functions [5]. The use of olive oil has grown over the past 25 years not just in Mediterranean regions but also globally [6].

Polycyclic aromatic hydrocarbons (PAHs) constitute a class of organic compounds composed of multiple fused aromatic rings, generally ranging from two to six rings. The US Environmental Protection Agency (US-EPA) has listed PAHs as a priority control due to their carcinogenic, mutagenic, and teratogenic properties [7]. Names, chemical structures, molecular weights, and hazard classification of 15 priority PAHs were shown in Table 1. PAH compounds containing 2-4 rings are classified as low molecular weight PAHs (LMW PAHs) or light PAHs (LPAHs), while PAH compounds containing 5 or more rings are classified as high molecular weight PAHs (HMW PAHs) or heavy PAHs (HPAHs) [8, 9].

Table 1. Names, chemical structures, molecular weights and hazard classification of 15 priority PAHs

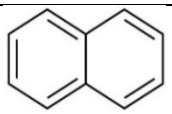
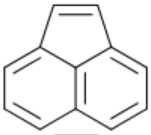
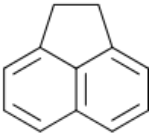
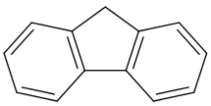
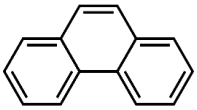
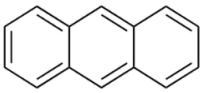
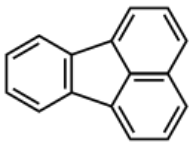
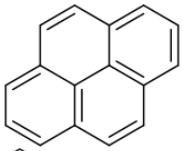
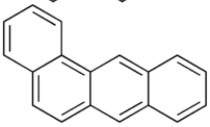
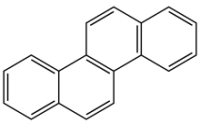
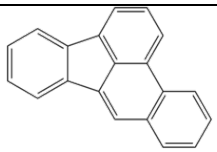
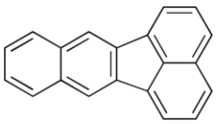
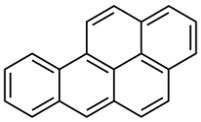
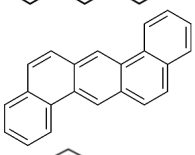
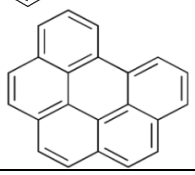
No	PAH and Abbreviation	Chemical Structure	Ring No	MW (g/mol)	Hazard Classification	Reference
1	Naphthalene (Nap)		2	128.2	Possibly carcinogenic to humans according to IARC (Group 2B)	[10, 11, 34]
2	Acenaphthylene (Acy)		3	154.2	Acute and chronic hazardous air pollutant according to EPA	[10, 11, 12]
3	Acenaphthene (Ac)		3	166.2	Unclassifiable as to carcinogenicity in humans according to IARC (Group 3), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
4	Fluorene (Fle)		3	178.2	Unclassifiable as to carcinogenicity in humans according to IARC (Group 3), acute and chronic hazardous air pollutant according to EPA	[11, 12, 13]
5	Phenanthrene (Phe)		3	178.2	Unclassifiable as to carcinogenicity in humans according to IARC (Group 3), chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
6	Anthracene (Ant)		3	202.3	Unclassifiable as to carcinogenicity in humans according to IARC (Group 3), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
7	Fluoranthene (Flu)		4	202.3	Unclassifiable as to carcinogenicity in humans according to IARC (Group 3), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
8	Pyrene (Pyr)		4	228.3	Unclassifiable as to carcinogenicity in humans according to IARC (Group 3), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
9	Benzo[a]anthracene (BaA)		4	228.3	Possibly carcinogenic to humans according to IARC (Group 2B), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
10	Chrysene (Chr)		4	252.3	Possibly carcinogenic to humans according to IARC (Group 2B), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]

Table 1. Names, chemical structures, molecular weights and hazard classification of 15 priority PAHs (continuing)

No	PAH and Abbreviation	Chemical Structure	Ring No	MW (g/mol)	Hazard Classification	Reference
11	Benzo[b]fluoranthene (BbF)		5	252.3	Possibly carcinogenic to humans according to IARC (Group 2B), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
12	Benzo[k]fluoranthene (BkF)		5	252.3	Possibly carcinogenic to humans according to IARC (Group 2B), chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
13	Benzo[a]pyrene (BaP)		5	278.4	Carcinogenic to humans (Group 1) according to IARC, acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
14	Dibenzo[a,h]anthracene (DBahA)		5	276.3	Probably carcinogenic to humans (Group 2A) according to IARC, acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]
15	Benzo[g,h,i]perylene (BghiPe)		6	276.3	Unclassifiable as to carcinogenicity in humans according to IARC (Group 3), acute and chronic hazardous air pollutant according to EPA	[10, 11, 12, 13]

PAHs are ubiquitous environmental contaminants that arise from both natural and anthropogenic sources, including incomplete combustion of organic matter, industrial processes, vehicular emissions, and natural events such as forest fires and volcanic eruptions [14, 15].

Due to hydrophobicity and electrochemical stability of PAHs, their persistence in nature is high [16]. The significance of PAHs lies in their widespread distribution, persistence in the environment, and potential adverse effects on human health and ecosystems. Due to their lipophilic nature, PAHs can accumulate in various environmental compartments, including soil, sediment, water, and air, leading to long-term exposure risks for organisms across levels [17]. Hydrophobicity, stability, and toxicity properties all rise in direct proportion to their molecular weight of PAHs [16].

Moreover, recent research [18] has shed light on the role of PAHs as persistent organic pollutants with long-range atmospheric transport capabilities, leading to global distribution patterns and bioaccumulation in remote regions, including polar environments.

PAHs are of particular concern due to their known or suspected adverse health effects. Several high molecular weight PAHs, such as benzo[a]pyrene (BaP), have been classified as priority pollutants by regulatory agencies due to their toxicological significance. Exposure to PAHs has been associated with respiratory diseases, cardiovascular disorders, and various types of cancer, particularly in populations living in urban areas or in close proximity to industrial facilities [19].

PAH contamination in foods has recently become a serious issue. In 2001, a processing problem caused a general contamination of European olive pomace oils. In 2005, the European Commission (EC) maximum levels for BaP were adopted for oils and fats. Vegetable oils, naturally free of PAHs, are contaminated by some processes. However, the PAHs' content in vegetable oils can be radically reduced during refining [20].

Various comprehensive studies from different aspects have been conducted on olive oil by researchers such as optimization of olive oil extraction [21, 22], determination of antioxidative capacity of olive oils [23], the effect of some additives on olive oil yield and quality parameters [24]. In the extent of contaminants determination; phthalate residues in olive oils [25], metal contamination on olive oils [26], mycotoxin residues in olive oils [27] and pesticide residues in olive oils [28] were examined in the literature. Studies on the detection of PAH residues in Turkish olive oil are limited. To the best of our knowledge, there are 3 studies [16, 29, 30] carried out in Türkiye for the detection of PAH residues in olive oils which are about BaP levels in olive oil samples.

Understanding the sources, fate, transport, and toxicological effects of PAHs is essential for developing effective mitigation strategies, regulatory policies, and environmental monitoring programs to safeguard human health and ecological integrity.

Advancements in analytical techniques, such as gas chromatography-mass spectrometry (GC-MS) [31, 9] and high-performance liquid chromatography (HPLC) [32, 33], have enabled the accurate detection and

quantification of PAHs in environmental matrices at trace levels. Additionally, molecular and biomarker approaches have provided insights into the sources and transformation pathways of PAHs in various ecosystems [23].

Bertoz et al. [11] stated that information regarding PAH concentrations in olive oils that are widely eaten worldwide is somewhat scarce. It is an important issue to reveal informative findings about PAH contamination of olive oils. The aim of this study was determination and evaluation of PAH contamination levels in olive oils in the market. In this article, a survey of 14 olive oils sampled from the Turkish market included extra virgin olive oil (EVOO) and riviera olive oil (ROO) samples, was done. 15 of 16 USA-EPA priority pollutants were selected for the analyzes. Considerable amounts of PAHs were determined in all the oil samples.

MATERIALS and METHODS

Sample Collection

Olive oils (n=14) including EVOO and ROO were analyzed for its PAH content. Refined oils are marketed as blends with unrefined oils because the refining methods negatively impact the flavor, aroma, and color of olive oil [10]. The ROO samples in the current study were blends of refined and extra virgin olive oils. The olive oils were purchased from different grocery stores in Şebinkarahisar district of Giresun province. All of samples were originated from Türkiye. Samplings of the olive oils were performed in triplicate on unopened containers which had been stored at room temperature before the analysis.

Solvents and Chemicals

Chromatography grade hexane, dichloromethane, acetonitrile, and purified water (Millipore, Bedford, MA, USA) were used. The PAH standard mixture was obtained from Sigma–Aldrich (Missouri, USA) and consisted of 15 compounds (100 ng/mL; 96–99.9% purity). These compounds were as naphthalene (Na) (1000 lg/mL), acenaphthylene (Ap) (2000 lg/mL), acenaphthene (Ac) (1000 lg/mL), fluorene (F) (200 lg/mL), phenanthrene (Pa) (100 lg/mL), anthracene (A) (100 lg/mL), fluoranthene (Fl) (200 lg/mL), pyrene (P) (100 lg/mL), benz-[a]anthracene (BaA) (100 lg/mL), chrysene (Ch) (100 lg/mL), benzo[b]fluoranthene (BbF) (200 lg/mL), benzo[k]fluoranthene (BkF) (100 lg/mL), benzo[a]pyrene (BaP) (100 lg/mL), dibenz[a,h]anthracene (DBahA) (200 lg/mL), benzo[g,h,i]perylene (BghiP) (200 lg/mL).

Sample Preparation

PAH extraction processes from olive oils were carried out in accordance with a previously validated method [34]. Since various matrix interferences along with PAHs were extracted from the sample, clean-up step of the samples was required for analytical determination. Solid phase extraction (SPE) cartridges containing octadecyl phase (Chromabond, 5 mL, Macherey-Nagel, Duren, Germany) were used for the cleaning-up step.

Olive oils (2.5±0.001 g) accurately weighed into a 10 mL volumetric flask, and the oil was diluted to 10 mL using n-hexane. Next, 1.0 mL of the sample solution was placed onto a 5-g silica SPE cartridge that had already been cleaned with 20 mL of dichloromethane, vacuum-dried, and conditioned with 20 mL of n-hexane. PAHs were eluted using a 70:30 (v/v) mixture of dichloromethane and n-hexane. Following the discharge of the initial 8 mL of eluate, an 8 mL fraction containing the PAH component was collected in a conical vial. Approximately one drop per second was used to control the flow rate.

Minimizing volatile PAH losses, the collected fraction was concentrated under a nitrogen stream to around 20-30 µL of volume, allowing the leftover solvent to evaporate spontaneously at ambient temperature. After being dissolved in 100 µL of acetonitrile, the residue was introduced into the GC device.

Instrumental Analysis

The chromatographic separation of the analytes was performed using GC-MS (7890A GC with 5975C Inert MSD, Agilent Technologies) on a HP-5ms capillary column (30 m x 250 µm i.d. x 0.25 µm film thickness, Agilent Technologies, USA). The column was a C18 reversed phase, 250 x 3 mm ID, 5 µm particle size thermostatted at 38℃ with a column heater. Head-space equipment (Agilent, 7694E) was used before gas chromatography to capture the PAH compounds.

Purified helium was used as a carrier gas at a constant flow rate of 1.5 mL/min and the pressure was 18.633 psi. The injection port's initial temperature was set to 30℃. The temperature program of the oven was as follows: 55°C held for 1 min, followed by a 25°C/min ramp to 320°C for 5 min.

In a pulsed splitless mode, one microliter of the sample was processed. Data was acquired using selected ion monitoring (SIM) mode. The MS Source and MS Quad temperatures were set to 230°C and 150°C, respectively.

RESULTS and DISCUSSION

There are limited studies conducted in Türkiye on the investigation of PAH contamination of olive oils [16, 29, 30], and they are only upon the determination of BaP levels in olive oils. In our study, 15 priority PAH active substances were examined unlike other studies. In addition to BaP levels; PAH4 levels also were presented in this study and some evaluations were made by comparing them with regulatory limits and with the results of other researchers.

A collection of 14 samples from commercially available olive oil brands were analyzed using GC-MS. Significant results have been obtained from the samples. Regarding BaP, except for 3 samples of EVOO and 2

samples of ROO, other samples (64.28%) were not compatible with the EU requirements. In terms of the total amount of BaP, BaA, Chr and BbF substances (called PAH4), none of the olive oil samples were not compatible with the EU requirements. The PAH amounts of the samples occurrence detected, varied between 0.04 µg kg^{-1} and 4637.13 µg kg^{-1}. The mean of the occurrence was 264.61 µg kg^{-1} in PAH detected samples.

Heavy PAHs (HPAHs) are typically more stable and hazardous than light PAHs (LPAHs) [11]. In this study, the amounts of LPAHs such as 2-ring and 3-ring in all olive oil samples (especially in EVOO) were higher compared to HMW PAHs. PAHs are compounds with low water solubility, and water solubility tends to decrease with each additional ring in the structure [11]. LMW PAHs with less lipophilic properties and more hydrophilic properties were detected at a higher rate in this study. The results were compatible with the survey of Scientific Committee on Food (SCF) indicated that the low molecular weight PAHs were the most prevalent type of PAHs found in foods [35].

Determination and quantification of the PAH compounds was made in light of their retention times (RT). RTs identified by using standard chromatogram obtained from mix samples of 15 PAH component and evaluation were made by chromatographic comparison with the standard chromatogram. GC-MS standard chromatogram at the concentration of 100 µg kg^{-1} is presented in Figure 1.

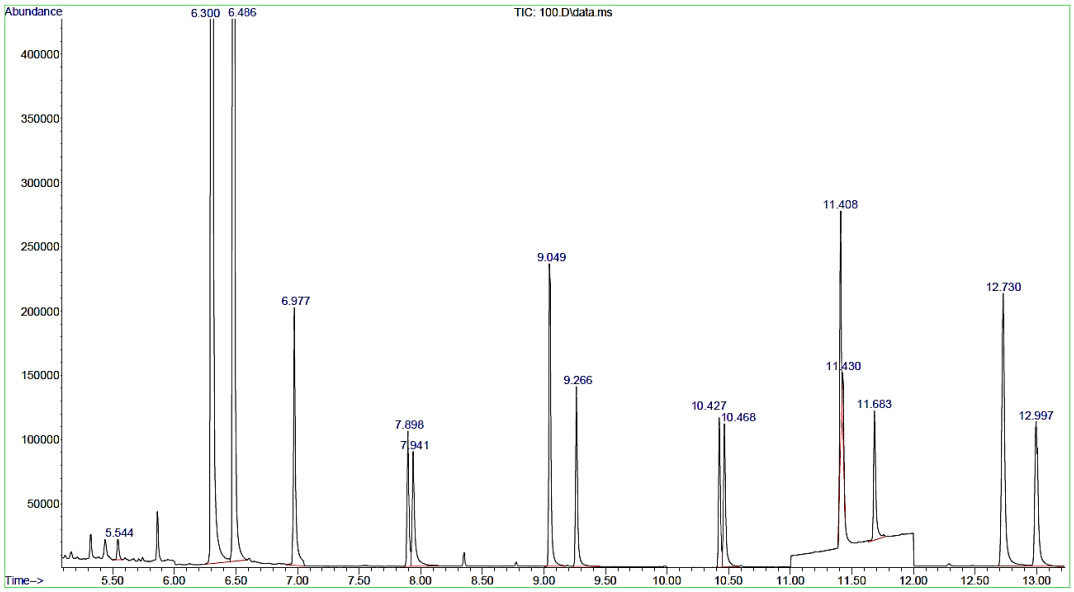


Figure 1. GC-MS spectrum of standard chromatogram (100 µg kg^{-1})

The results of PAH concentrations in EVOO samples were presented in Table 2 and the results of PAH concentrations in ROO samples were presented in Table 3.

The highest amount of PAH compounds was Nap in all samples. Nap was detected mostly in B1 brand in both EVOO and ROO samples. The interesting part of the result was that the B1 was a major brand that has a dominating power in the market. The Nap concentrations in samples ranged between 1054.66 µg kg^{-1} and 2732.59 µg kg^{-1} (the mean: 2698.00 µg kg^{-1}). The highest amounts of PAH components in all samples after Nap were Ac, Acy and BbF. The mean levels of these PAHs were 153.62 µg kg^{-1}, 137.45 µg kg^{-1} and 87.68 µg kg^{-1}, respectively.

Phe was the compound detected in the least number of samples. Phe was detected in only one sample as 14.42 µg kg^{-1} in which was EVOO sample of B4 brand. Despite there is no regulatory limit for Phe set by law, 14.42 µg kg^{-1} concentration was a significant amount compared to threshold limit of PAH4 designated by law as 10 µg kg^{-1}. Flu followed Phe in being present in the fewest number of samples. Flu was detected in both EVOO and ROO samples of 2 brands coded as B1 and B2. Flu was detected as 20.09 µg kg^{-1} in the EVOO sample of B1 brand and 21.11 µg kg^{-1} in the ROO sample of B1 brand. For B2 brand, Flu was detected as 6.67 µg kg^{-1} in the EVOO sample and 3.15 µg kg^{-1} in the ROO sample.

In terms of detected PAH compound at the lowest amount in all samples, BaA was the lowest. BaA was detected at a minimum level of 0.04 µg kg^{-1} in EVOO samples and detected at a minimum level of 0.69 µg kg^{-1} in ROO samples.

About PAH limit values, the EU Regulation (Regulation No. 835/2011) and the Turkish Food Codex Regulation (Contaminants Regulation No. 28157/2011) are compatible with each other. According to the regulations, the maximum limit was designated 2 µg kg^{-1} for BaP and 10 µg kg^{-1} for the PAH4 compounds consist of BaP, BaA, Chr and BbF.

Table 2. PAHs contents of randomly selected some samples comprising of extra virgin olive oils (μg kg^{-1})

	Extra virgin olive oil (EVOO)*						
PAHs	B1-E	B2-E	B3-E	B4-E	B5-E	B6-E	B7-E
Nap	3250.92±0.06	2984.85±0.35	2558.86±0.03	2531.97±0.36	2855.37±0.12	2426.7±0.08	2519.47±0.23
Acy	511.73±0.42	136.62±0.12	65.35±0.14	58.89±0.07	68.25±0.06	58.78±0.19	62.54±0.34
Ac	898.99±0.21	56.22±0.09	27.62±0.08	19.66±0.18	33.78±0.38	28.11±0.22	10.95±0.14
Fle	N.D.**	21.89±0.41	8.98±0.24	7.08±0.02	12.00±0.15	9.62±0.10	N.D.
Phe	N.D.	N.D.	N.D.	14.42±0.09	N.D.	N.D.	N.D.
Ant	15.35±0.12	16.33±0.17	21.52±0.19	16.33±0.23	17.90±0.08	22.29±0.34	12.20±0.03
Flu	20.09±0.24	6.67±0.15	N.D.	N.D.	N.D.	N.D.	N.D.
Pyr	21.02±0.06	18.88±0.09	14.36±0.05	11.61±0.23	12.89±0.16	11.81±0.02	10.35±0.11
BaA	8.16±0.14	0.50±0.03	6.07±0.21	5.61±0.05	0.04±0.01	N.D.	N.D.
Chr	10.71±0.01	11.10±0.14	N.D.	N.D.	N.D.	3.59±0.09	3.56±0.12
BbF	53.43±0.18	46.53±0.07	133.89±0.02	197.98±0.14	36.88±0.03	17.74±0.05	40.32±0.33
BkF	26.54±0.22	24.03±0.16	66.54±0.29	99.69±0.03	19.38±0.17	8.81±0.09	20.59±0.29
BaP	32.50±0.10	14.38±0.29	14.99±0.11	14.47±0.09	N.D.	N.D.	N.D.
DBahA	42.41±0.06	N.D.	52.16±0.06	48.81±0.06	48.95±0.06	42.54±0.06	47.45±0.06
BghiPe	29.12±0.06	11.04±0.06	127.70±0.06	11.78±0.06	6.08±0.06	3.57±0.06	7.18±0.06

*Number of replicates = 3, B: Brand, E: Extra virgin olive oil, **N.D., Not Detectable

Table 3. PAHs contents of randomly selected some samples comprising of riviera olive oils (μg kg^{-1})

	Riviera olive oil (ROO)*						
PAHs	B1-R	B2-R	B3-R	B4-R	B5-R	B6-R	B7-R
Nap	4637.13±0.21	2408.31±0.07	2444.27±0.19	2503.84±0.02	2872.97±0.18	2722.73±0.41	1054.66±0.11
Acy	100.49±0.14	121.86±0.04	61.51±0.17	58.68±0.09	61.24±0.29	56.80±0.27	61.27±0.34
Ac	77.25±0.01	68.81±0.16	23.07±0.02	18.80±0.26	29.79±0.05	N.D.**	3.77±0.38
Fle	N.D.	22.52±0.36	6.83±0.14	6.32±0.25	11.84±0.43	28.58±0.03	7.89±0.03
Phe	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Ant	52.91±0.19	18.32±0.43	16.81±0.23	11.33±0.23	24.66±0.15	30.04±0.14	22.62±0.14
Flu	21.11±0.44	3.15±0.06	N.D.	N.D.	N.D.	N.D.	N.D.
Pyr	20.59±0.19	17.71±0.04	12.26±0.09	11.55±0.06	13.18±0.46	11.30±0.36	12.01±0.23
BaA	15.99±0.15	14.75±0.12	0.69±0.02	14.31±0.20	6.52±0.24	N.D.	N.D.
Chr	18.38±0.35	N.D.	N.D.	19.90±0.08	12.49±0.21	4.31±0.14	2.93±0.08
BbF	30.07±0.32	42.61±0.07	199.32±0.08	202.97±0.28	31.62±0.22	68.51±0.10	38.64±0.17
BkF	14.95±0.29	21.77±0.19	99.97±0.34	102.06±0.24	15.72±0.27	34.81±0.23	20.00±0.12
BaP	3.31±0.05	15.16±0.13	15.95±0.16	14.72±0.10	N.D.	N.D.	51.17±0.08
DBahA	30.16±0.14	N.D.	51.00±0.04	43.77±0.29	47.24±0.16	42.16±0.07	52.44±0.19
BghiPe	39.49±0.22	7.17±0.06	13.30±0.06	3.09±0.05	37.34±0.19	26.76±0.02	5.99±0.04

*Number of replicates = 3, B: Brand, R: Riviera olive oil, N.D., Not Detectable; **N.D. = Not Detected

Based on the evaluation of the PAH profiles in food, SCF recommended using BaP as a marker of presence and impact of the carcinogenic PAHs in food [36]. Despite the fact that maximum BaP level should have been 2 μg kg^{-1} according to the law, commercial brands except for samples B5, B6 and B7 of EVOO samples, did not meet the regulatory threshold levels for BaP. Detected BaP levels above the limit in EVOO samples ranged from 14.38 μg kg^{-1} to 32.50 μg kg^{-1} with the mean 19.09 μg kg^{-1}. Baloğlu and Bayrak [29], found BaP concentrations between 0.330 μg kg^{-1} and 0.870 μg kg^{-1} for 3 samples of 20 extra virgin olive oils in Ankara province. Şekeroğlu et al. [30], found BaP concentrations between 2.7 μg kg^{-1} and 38.8 μg kg^{-1} of 5 olive oil samples in Gaziantep province. Çıtak [16] found BaP concentrations between 2.6 μg kg^{-1} and 19.2 μg kg^{-1} for 7 samples of 9 extra virgin olive oils in İzmir province and found between 3.0 μg kg^{-1} and 36.1 μg kg^{-1} for 14 samples of 15 extra virgin olive oils in Aydın province. In ROO samples in our study, commercial brands except for samples B5 and B6, did not meet the regulatory threshold levels for BaP. Detected BaP levels above the limit in ROO samples ranged from 3.31 μg kg^{1} to 51.17 μg kg^{-1} with the mean 20.06 μg kg^{-1}. Baloğlu and Bayrak [29], found the BaP levels up to 2.465 μg kg^{1} in 12 samples of 20 riviera olive oils. Şekeroğlu et al. [30], found the BaP concentrations between 16.2 μg kg^{-1} and 74.9 μg kg^{-1} of 5 riviera olive oil samples. Çıtak [16] found between 3.0 μg kg^{-1}and 13.7 μg kg^{-1} BaP levels in 2 samples of riviera olive oils. BaP residues of extra virgin olive oils and riviera olive oils in our study were quite high compared to Baloğlu and Bayrak [29]. BaP residues detected in our study were compatible with the results of Şekeroğlu et al. [30] and Çıtak [16].

Elaridi et al. [31] indicated that two different samples of one brand had 9.45 μg kg^{-1} and 11.9 μg kg^{-1} BaP levels from 25 bottled olive oil brands marketed in Lebanon. Similar conditions such as such as SPE and GC-MS were used in the study. The results of Lebanon olive oil samples were above the regulatory limits. Our results were higher than Lebanon olive oil samples in Elaridi et al. [31]'s study.

Fromberg et al. [20] reported that BaP concentrations in 46 EVOO samples were from <0.2 μg kg^{-1} to 0.4 μg kg^{-1}, and BaP concentrations in 6 olive oil (OO) samples were from <0.2 μg kg^{-1} to 0.2 μg kg^{-1}. In the study of Fromberg et al. [20], SPE method and GC-MS adopted as in our study and the BaP occurrence in the samples was below the permissible limits.

In the report of EC [37], BaP levels in virgin olive oils (VOO) from 671 samples were varied between 0.015 µg kg^{-1} and 32 µg kg^{-1}; BaP levels in olive oils (OO) from 280 samples were ranged between 0.03 µg kg^{-1} and 89 µg kg^{-1}. EC report indicated that some olive oil samples in the market had lower amounts than the legislation but some of them had higher than the upper BaP limit. The BaP levels of VOO samples were in line with our study.

Van der Wielen et al. [38] revealed the amount of BaP from 170 samples was up to 85 µg kg^{-1} in OO samples. Pupin and Toledo [39] detected BaP concentrations of OO samples from 32 samples ranged between 0.5 µg kg^{-1} and 164 µg kg^{-1}. These results suggested that the highest BaP concentrations from these two studies were considerably higher than in our study.

BaP concentrations in 6 EVOO samples in the study of Krajian and Odeh [40], ranged from 0.08 µg kg^{-1} to 5.79 µg kg^{-1} with mean value 1.52 µg kg^{-1}. The BaP levels in EVOO samples were lower than the levels of our study. However, the levels of Krajian and Odeh [40] were higher than the permissible limits of EU. BaP concentrations in Rascón et al. [41] were all lower than the amount of this study. Rascón et al. [41] determined BaP concentrations from 0.018 µg kg^{-1} to 0.075 µg kg^{-1} in 8 EVOO samples. These range of BaP levels were quite low.

Ju et al. [42] found that BaP occurrence in the samples was below the limit levels. BaP concentration in EVOO sample was 0.481 µg kg^{-1}. In OO sample, BaP level was below Limit of Quantification (LOQ) value in the study of Ju et al. [42]. Shi et al. [43] declared that BaP concentrations obtained from 8 samples were in the range of 0.27 µg kg^{-1} and 0.89 µg kg^{-1}. These amounts were compatible with the requirements of legislation and quite lower than the levels of our study.

The data obtained in our study about the compounds that constitute of PAH4 was as follows: BaA detected samples from EVOO and ROO samples, had the occurence level between 0.04 µg kg^{-1} and 15.99 µg kg^{-1}. BaA was not detectable in B6 and B7 brands both of EVOO and ROO samples. Chr detected samples among of EVOO and ROO varied between 2.93 µg kg^{-1} and 19.90 µg kg^{-1}. Chr was not detectable in B3, B4 and B5 brands of EVOO samples. In ROO samples, Chr was not detectable in B2 and B3 brands. There was no sample that BbF could not be detected in the study. BbF detected samples among of EVOO and ROO varied between 17.74 µg kg^{-1} and 202.97 µg kg^{-1}.

According to the legislation, PAH4 should not be above 10 µg kg^{-1} in fats and oils for human consumption or used as food ingredients. However, in this study; PAH4 in EVOO samples ranged from 21.33 µg kg^{-1} to 218.06 µg kg^{-1}. PAH4 in ROO samples ranged from 50.63 µg kg^{-1} to 251.9 µg kg^{-1}. Even the lowest result of all samples was found to be above the EU Regulation and Turkish Regulation limit value. As a matter of fact, all olive oil brands had PAH4 contamination in their products.

PAH4 amounts in Elaridi et al. [31] were 19.3 µg kg^{-1} and 26.7 µg kg^{-1}. These values were lower than of our study. However, the values were higher than the permissible limits of combination of 4 PAH components.

Gharbi et al. [8] declared the PAH4 level was between 0.2 µg kg^{-1} and 0.7 µg kg^{-1}. The PAH4 amounts of EVOO and OO samples in Ju et al. [32], were 2.508 µg kg^{-1} in EVOO sample and 0.540 µg kg^{-1} in OO sample. These levels were quite lower than the amounts of our study, and below the upper limit of legislation. PAH4 concentrations in 6 EVOO samples in the study of Krajian and Odeh [40], ranged from 0.34 µg kg^{-1} and 20.2 µg kg^{-1} with mean value 7.66 µg kg^{-1}. The PAH4 levels were lower than the levels of our study but some of the samples were higher than the permissible limits of EU.

Rascón et al. [41], PAH4 concentrations were determined between 0.035 µg kg^{-1} and 0.667 µg kg^{-1} in 8 EVOO samples. PAH4 occurrence of 8 samples in Shi et al. [43] was declared as 1.48 µg kg^{-1} and 4.67 µg kg^{-1}. These amounts were compatible with the requirements of legislation.

The sum of PAH content of 45 compounds in Ekner et al. [9] was from 9.17 µg kg^{-1} to 94.7 µg kg^{-1}. Krajian and Odeh [40] revealed that the total amount of 16 PAH compounds in 6 EVOO samples ranged from 33.4 µg kg^{-1} to 82.4 µg kg^{-1}. Rascón et al. [41], the sum of PAH content varied between 0.074 µg kg^{-1} and 4.321 µg kg^{-1} in 8 EVOO samples. In 8 olive oil samples of the study Shi et al. [33], the sum of PAH content ranged from 17.90 µg kg^{-1} to 57.55 µg kg^{-1}.

The highest level for the sum of PAH amount from the literature was determined as 94.7 µg kg^{-1} [9]. In our study, PAH detected samples were ranged from 0.04 µg kg^{-1} to 4637.13 µg kg^{-1}. 27 results of our study were higher than 94.7 µg kg^{-1} concentration level. This is the rate 16.17% of all the samples and mostly consisting of Nap component.

The results showed that commercially available olive oils contained probably and possibly carcinogenic PAHs. For instance, among 14 olive oil samples, one commercial brand of samples (6.67%) contained residue of PAHs carcinogenic to humans (Group 1), one commercial brand (6.67%) contained residue of PAHs probably carcinogenic to humans (Group 2A), four commercial brands (26.67%) contained residue of PAHs possibly carcinogenic to humans (Group 2B), seven commercial brands (46.67%) contained residue of PAHs unclassifiable as to carcinogenicity in humans (Group 3B). Compared to Elaridi et al. [31], the percentiles were lower in our study concerning to hazard classification.

Considering the results; all olive oil samples analyzed were contaminated with PAH residues little or much. This is an issue that should be taken into consideration seriously. Bertoz et al. [11] stated that PAHs can be found in high levels in edible oils because of their lipophilic nature. Consuming the foods with these occurrence levels will have negative health effects on

people both in the short and in the long term. For instance, PAHs can be metabolized to reactive compounds in the human body that show the ability to damage structural proteins and DNA. This can be resulted in mutations, developmental malformations, tumors, and cancer [44, 45].

It is essential to take precautions to minimize PAH occurrence in olive oils. For this reason, it is useful to be aware of the sources of contamination. According to Rascón et al. [41] environmental pollution, production methods, or the composition of the oil were the main causes of PAHs in olive oils. For instance, particles arising from air pollution and smoke contain PAH components caused olive oil contamination. The PAH components adhere to olive peel through polluted air transferred to the oil when the oil is extracted [41, 46].

Fuel emission from mechanical harvesters was the main source of PAH contamination for olive oils followed by traffic emission [9, 11]. In olive oil production, olive harvesting is carried out in two ways including collecting olives manually from tree or harvesting olives by harvester machines [47]. Incomplete combustion of the fuel from harvester machines releases various PAH compounds. These PAHs accumulate on the surface of olives and cause the contamination of olives.

Using high temperature during the grinding of olives and the extraction of olive pulp may cause PAH formation at the batch. For instance, higher levels of PAHs in the samples may have resulted from the hot-pressing procedure used when processing the olives [31].

Elaridi et al. [31] indicated that the firms close to industrial regions might accounted for the PAH pollution levels in olive oils. In our study, it is difficult to allege that all the production plants of olive oil samples with high PAH contents were generally close to industrial areas. From this angle, it is understood that the main source of contamination in the olive oil samples produced in Akhisar (Manisa) and Ayvalık (Balıkesir), was the automation systems used during harvest or manufacturing conditions. Remarkably, although olive oils factories located in İzmir were more vulnerable to the negative effects of industrialization, the lower levels of PAH occurrence were determined from the samples produced by İzmir plants.

During the manufacturing processes lubricants, extraction solvents, and detergents may also contaminate the olive oil with PAHs. According to Bertoz et al. [11], lubricating oil used during conveyor belt installation, debris exposed during plant maintenance, detergents for cleaning the lines in the factory and so on are the other sources of contamination. Ščimko et al. [47] declared that recycled polyethylenes such as plastic bottles contain PAH residues can contaminate the olive oils by means of diffusion from the bottle. In addition to this, Moret and Conte [48] expressed that the migration of PAHs to olives can be caused by jute olive bags whose jute fibers have been treated with mineral oils.

It is possible to reduce PAH contamination in olive oils through the filtration process during the olive oil production [9, 20, 40]. Before bottling, natural olive oils are physically filtered [49]. For this purpose, the tanks are heated to 20-28℃ in the cold months and the oils are aged for ten days to a month. Next; olive oils are filtered through the systems consisting of various materials such as hydrophilic cotton, cellulose and soil (diatomaceous earth, kieselguhr). However; the flavor, aroma, and color of the olive oils are all negatively impacted by the refining operations. Therefore, blending refined olive oils with unrefined olive oils can be a solution to avoid the loss [9].

PAH content in vegetable oils can be decreased by bleaching and deodorization processes [11, 42, 50]. The reasons for the decrease are removing some of PAHs by the adsorption property of bleaching clay in the bleaching step and removal some of PAHs while removing undesirable volatile compounds in the deodorization step. However, these steps are not included in the olive oil production processes. Therefore, outside the olive oil industry, it is possible to reduce the occurrence of PAHs in vegetable oils through bleaching and deodorization processes [11]. While deodorization is an effective method for removing LMW PAHs from crude vegetable oils, neutralization using activated coal or activated earth is a more efficient way to remove HMW PAHs [9, 51, 52, 53].

CONCLUSION

In this study, commercial olive oil samples (n=14) were analyzed for the presence of various PAHs. Considering the results, all olive oil samples analyzed were contaminated with PAH residues. The PAH amounts of the samples occurrence detected, varied between 0.04 μg kg^{-1} and 4637.13 μg kg^{-1} (the mean 264.61 μg kg^{-1}) . Regarding BaP, 64.28% of the samples were not compatible with the EU requirements. None of the olive oil samples were not compatible with the EU requirements in terms of PAH4 compounds. This is an issue that should be taken into consideration seriously. It is clear that consuming the foods with these residue levels will have negative health effects on people both in the short and in the long term. Therefore, it is essential to take necessary precautions to minimize PAH residues in olive oils.

Conflict of Interests

The authors declare that they have no conflict of interests.

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