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RESEARCH PAPER

Polycyclic Aromatic Hydrocarbons in Different Highly Consumed Local and Imported Fruits Available in Oman

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Citation
Abdalla Shaban Lotfi Gohari Abdou, Salem Said Jaroof Al Touby and Mohammad Amzad Hossain (2024), Polycyclic Aromatic Hydrocarbons in Different Highly Consumed Local and Imported Fruits Available in Oman. *Green Energy and Environmental Technology* 3(1), 1–18.

DOI
https://doi.org/10.5772/geet.25

Received: 12 July 2023
Accepted: 19 February 2024
Published: 28 June 2024

Abstract

Vegetables and fruits are usually contaminated due to fuel combustion and environmental pollution. Therefore, before consumption, these food items have to be screened for the level of contamination. Polycyclic aromatic hydrocarbons (PAHs) exposure may increase the risk of human health conditions such as lung cancer, skin cancer, and cardiovascular disease. This work aims to extract the PAHs from the local and imported fruits peels and flesh, and quantify them using Liquid Chromatography–Mass Spectrometry (LC-MS). To achieve this aim, six locally-grown fruits and five imported fruits were collected from the Al-Hoti market, Al-Khuwair, Muscat, Oman. The dry powder samples were extracted with alcoholic sodium hydroxide for four hours. The extract was washed with methanol/distilled water (1:1), finally extracted with cyclohexane, and evaporated using a rotary evaporator at 40 °C. The results from LC-MS showed that not all major sixteen PAHs were detected in the fruits and fruit peels. One of the major PAHs, acenaphthylene was detected in all of the fruit's peel and flesh samples with concentrations ranging from 0.0483 μg/kg (lemon) to 0.1987 μg/kg (banana). 1,2-Benzanthracene was detected in all of the samples, with concentrations ranging from 0.2219 μg/kg (sweet melon peel) to 0.2257 μg/kg (grapefruit peel) and benzo[a]pyrene was detected in all fruit's peel and flesh samples, with concentrations ranging from 0.8331 μg/kg (lemon) to 0.8348 μg/kg (banana peel).

Overall, the levels of PAHs in the collected fruits' peel and flesh samples were generally low and did not exceed the permissible limits. Nevertheless, continuous monitoring of PAH levels in the fruits' peel and flesh samples is necessary to ensure that the limit is within the permissible limit for the safety of consumers.

Keywords: fruits' flesh and peel samples, polycyclic aromatic hydrocarbons, extraction, pre-concentration, quantification, LC-MS

1. Introduction

Fruits are recommended as a part of a healthy diet due to the high amounts of vitamins, minerals, especially electrolytes, dietary fibers, and phytochemicals, mainly antioxidants that they contain. Numerous studies have connected chronic illnesses, including chronic obstructive pulmonary disease, hypercholesterolemia, respiratory disorders, cardiovascular diseases, hypertension, osteoporosis, mental health concerns, and cancers to the lack of fruits and vegetables in the patients' diet [1]. Consuming fruits have been associated with a decreased risk of several non-communicable diseases. Antioxidants not only give fruits their vivid color but also function as scavengers, cleaning up free radicals before they have an adverse impact on health. This is the reason why antioxidants are topics of great interest [1].

The Sultanate of Oman is a subtropical country in the Arabian Peninsula. Low precipitation (100 mm annually) and high temperatures throughout the year characterize the climate. Geographically, the desert, which has little vegetation, makes up about 75% of the nation's total land area. Contrarily, the remaining area has a wide range of terrains and climatic conditions that enable the cultivation of a wide range of fruit crops, including tropical, subtropical, and temperate fruit crops. The Hajar Mountains in the north of the country reach an altitude of around 3000 m, where temperatures drop significantly, and several temperate-zone fruit crops and other vegetation can grow [2].

Although cultivation is mostly limited to the northern region of the nation, fruit cultivation is economically significant in Oman. The date palm is the most important fruit crop in terms of production and profitability. Banana is the second-largest fruit exported from Oman and is grown all around the country. Mango, lime and pomegranate are also significant crops in this region; the local economy also greatly benefits from the production of almonds, apricots, pears, walnuts, apples, peaches, and figs [3].

Polycyclic aromatic hydrocarbons (PAHs) are colorless, white, or pale-yellow solid organic substances. They are a ubiquitous group of several hundred chemically related molecules that are environmentally persistent and have varying toxicity. Through several different processes, they are toxic to both humans and animals.

Multiple mechanisms allow PAHs to reach the environment. They are frequently found as a mixture of two or more compounds, such as soot. Industries, vehicles, and incomplete combustion from either natural (forest and brush fires) or artificial combustion sources can produce PAHs, in addition to biological processes. Therefore, it is common to find PAHs in the air, soil, and water samples. The toxicity caused by PAHs interferes with the function of cellular membranes and the enzyme systems connected with the membrane. It has been established that PAHs are potent immune suppressants as well as mutagenic and carcinogenic agents. The effects on immune system development, humoral immunity, and host resistance have all been recorded [4].

PAHs are made of two or more benzene rings fused in linear, cluster, or angular configurations. Even though there are numerous PAHs; the majority of data reporting, assessments, and regulations give importance to 14 and 20 different PAH molecules. PAHs have two or more single or fused aromatic rings that share a pair of carbon atoms. Small PAHs have less than six fused aromatic rings, while big PAHs have seven or more. The majority of research on PAHs has been conducted on small PAHs due to their availability. Phenanthrene and anthracene, which both have three fused aromatic rings, are the simplest PAHs, according to the International Agency for Research on Cancer. Another aromatic hydrocarbon is naphthalene, which has two coplanar, six-membered rings that share an edge. Despite being referred to as a bicyclic aromatic hydrocarbon, it is not a real PAH. The PAHs 7, 12-dimethyl benzo anthracene (DMBA), and benzo(a)pyrene (BaP) have been the subject of intense research. These PAHs have been frequently examined.

PAHs generally have low vapor pressure, very low aqueous solubility, and high melting and boiling temperatures (so they are solids). Molecular weight is directly proportional to PAH resistance to oxidation and reduction while inversely proportional to the vapor pressure and the aqueous solubility. With each extra ring, PAHs become less soluble in water. Meanwhile, due to their high lipophilicity, PAHs are particularly soluble in organic solvents. PAHs exhibit several properties, including emittance, conductivity, corrosion resistance, and physiological activity. UV absorbance spectra for PAHs have a distinctive appearance. Each isomer has a varied UV absorbance spectrum because each ring structure has a distinct UV wavelength. This is incredibly useful in identifying PAHs. The majority of PAHs are also fluorescent and, upon excitation, emit specific light wavelengths [4]. The incomplete burning of organic resources, including coal, oil, and wood, is the main source of PAHs [5]. PAHs are not chemically produced for industrial use. However, several PAHs have a few commercial applications. They are mostly employed as agricultural, pharmaceutical, and chemical intermediates [4–7].

During cultivation, PAHs can be transferred from the soil and the air. They may also arise during cooking, shipping, or storage procedures before ingestion. Only small quantities of PAHs are usually found in uncooked fruits and vegetables. The US Environmental Protection Agency classifies some chemicals as priority pollutants, and the quantities range between 0.01 and 0.5 μg kg^{-1} (wet weight) (EPA). However, many studies have shown that PAH concentrations in various fruits and vegetables can surpass 0.5 μg kg^{-1} wet weight and approach 5 μg kg^{-1}. Depending on the crops' surroundings, the amounts of the aromatic hydrocarbon, or even the product itself, might vary considerably. Products cultivated near roads or in urban settings typically have higher PAH content than those grown in rural areas. In almost every fresh fruit and vegetable, trace amounts of chemicals, including phenanthrene, fluoranthene, and pyrene, have been discovered. Some of them have been discovered to have relatively significant concentrations of lighter PAHs, such as naphthalene, acenaphthene, and acenaphthylene [8].

16 PAHs are taken into account as a category since they have been recognized as being compounds of concern in terms of possible exposure and harmful effects on human health. Due to the extensive distribution of these substances and their toxicological importance, biological assessment of exposure to PAHs is of particular interest. However, there are specific differences in how each PAH affects health. In reality, several PAHs are classified by the International Agency for Research on Cancer as known, potentially, or probably carcinogenic to humans (Group 1, 2A, or 2B). Benzo[a]pyrene (Group 1), benz[a] anthracene, naphthalene, and benzo[b]fluoranthene, chrysene, and benzo[k]fluoranthene (Group 2B) are a few of these. Some PAHs are well-known carcinogens, mutagens, and teratogens, which put human health and well-being in severe danger [9]. An increased risk of lung cancer is the most significant health consequence that may be anticipated from breathing in PAHs [9].

Fruits are an essential component of every well-balanced diet since it includes a variety of essential nutrients. Numerous studies have found a link between consuming enough fruit and a decreased risk of non-communicable diseases. Furthermore, fruit fibers have been shown to delay intestinal transit speeds by producing bulk, allowing for more gradual nutrient absorption and reducing constipation [10]. Fibres can undergo colonic fermentation, which raises the concentration of short-chain fatty acids with anticancer benefits and preserves gastrointestinal health. Multiple studies have emphasized the cardiovascular disease risk-reducing ability of fruits, finding that their consumption correlated withe lower cholesterol, blood pressure, and triacylglycerol, averting early cardiovascular diseases [11–13]. According to statistics, about 3% of Omanis are now affected by skin diseases, and that number is gradually increasing and is expected to double by 2029. A diseases-related analysis showed that the number of people with cancer in Oman is now about 7% and is expected to double by 2035 [14, 15]. Among the organic toxicants, PAHs are the primary concern. These hazardous and carcinogenic substances are present in fruits and vegetables, which has resulted in many serious health diseases. Literature has shown that several illnesses, including cardiovascular diseases, cancers, hypertension, myocardial infarction (MI), atherosclerosis, and thrombosis, are related to the intake of PAHs-contaminated fruits and vegetables. Prolonged exposure to PAHs can lead to tumor growth in various organs, including skin, lungs, pancreas, esophagus, bladder, colon, and female breast.

Based on the negative impact of PAHs and to ensure the safety of the Omani nation, there has been just one research publication that demonstrated the existence of PAHs in the water of the Oman Sea [9]. According to the literature, no study had been conducted on PAHs in local and imported fruits in Oman. Therefore, this study aimed to collect fruit samples from various local markets and supermarkets in Oman and to extract PAHs from the selected fruits and peels using the standard method to define the amount of PAHs using liquid chromatography-mass spectrometry (LS-MS).

1.1. Literature review

Due to the facts above, a study was conducted in Brazil to ascertain the amount of PAHs in fruits and vegetables [16]. In order to better quantify the dietary exposure of Brazilians to PAHs, this study measured the amounts of PAHs in samples of apples, cabbages, grapes, lettuces, pears, and tomatoes. Six separate locations were used to gather samples of each fruit and vegetable, which were then examined for 16 PAHs. The analytical procedure included saponification with methanolic KOH, liquid–liquid extraction with cyclohexane, cleanup on a silica gel column, and analysis by high-performance liquid chromatography with fluorescence detection. The results showed that PAHs were present in lettuces at a mean level of 13.53 μg/kg, tomatoes at 9.50 μg/kg, cabbage at 8.86 μg/kg, apples at 4.05 g/kg, grapes at 3.77 μg/kg, and pears at 3.87 μg/kg. One of the most typical hazardous PAH member is Benzo(a)anthracene, estimated at 89% of all samples examined. There was no evidence of chrysene in any analyzed sample [17].

Kumar *et al.* [18] investigated the occurrence of PAHs in fruits. The study aimed to determine the amount of PAHs in regularly consumed fruits in India and examine the potential health concerns linked to them. The scientists collected 56 fruit samples from several marketplaces in Delhi, India. Apples, bananas, guava, mango, papaya, and watermelon were among the fruits. Gas chromatography-mass spectrometry (GC-MS) was used to examine the samples for 16 key PAHs. The findings revealed that all fruit samples had detectable amounts of PAHs. The apple and banana samples had the highest quantities of PAHs, with total PAH values ranging from 0.34 to 2.46 ng/g and 0.33 to 2.39 ng/g, respectively. The watermelon samples had the lowest PAH values, with total PAH amounts ranging from 0.03 to 0.29 ng/g. The researchers also calculated the dietary intake of PAHs from fruit-eating for various age groups. The findings revealed that youngsters had a greater estimated daily intake of PAHs from fruit-eating than adults. The predicted lifetime cancer risk from PAHs in fruit intake was likewise shown to be greater in youngsters than in adults. According to the study, fruits can be a substantial source of PAH exposure, especially for youngsters. The study emphasizes the need to monitor PAH levels in fruits and take steps to prevent contamination [18].

Fernández-González *et al.* [19] analyzed the amounts and sources of PAHs in various fruits from Northwest Spain. The researchers collected 90 fruit samples from local markets and farms, including apples, pears, peaches, nectarines, plums, strawberries, raspberries, blueberries, blackberries, and cherries. GC-MS was used to examine the samples for 16 key PAHs. The findings revealed that PAHs were present in all fruit samples, with the highest quantities detected in strawberries and raspberries. PAH levels varied by fruit variety, with berries and stone fruits holding more significant amounts than pome fruits. In addition, multivariate statistical analysis was employed in the study to identify probable sources of PAH contamination. The findings indicated that PAHs in fruits mainly originated from vehicle exhaust emissions and agricultural practices such as pesticide and fertilizer usage. According to the study's findings, fruits may be a substantial source of PAH exposure, and the amounts of PAH in fruits can vary depending on the kind of fruit and the source of contamination. The study emphasizes the need to monitor fruits' PAH levels and take steps to prevent contamination [19].

2. *Materials and methods*

2.1. LC-MS instrument

The prepared pre-concentrated fruits' peel and flesh extracts were analyzed using a sensitive LC-MS with APCI mode (MS/Q-TOF, Model G6530B Accurate mass, Agilent Technologies, Singapore). Ten microliter sample from each extract was injected into the LC injector, and the oven temperature was set to 30 °C. The column temperature was set at 25 °C, and the gradient flow pump was used. The total run time of PAH samples was 45 min. The ion source and mass transfer line temperatures were set to 280 °C and 320 °C, respectively. The data obtained were full-scan mass spectra with a mass range of 30–2200 amu, and the scan rate was one per second. Agilent C_{18} reverse phase column was used with a length of 4.6 mm, diameter of 150 mm, and internal diameter of 5 μm to analyze PAHs. The mobile phase used to analyze PAHs consisted of (A) 100% H_2O + 0.1% formic acid + 10 mM AF and (B) 100% acetonitrile + 0.1% formic acid + 10 mM AF. The flow rate in mobile phase was 0.2 ml min^{-1}. UV spectroscopy was used with wavelengths ranging from 190–400 nm.

2.2. Samples collection

The Omani fruits, namely sweet melon, banana, papaya, lemon, and fig, were collected from Al-Hoti market, Al-Khuwair, Muscat, Oman, and the imported fruits, namely Turkish grapefruit, Yemeni sweet melon, Ecuadorian banana, Sri Lankan papaya, and South African lemon were sourced from Lulu hypermarket (Figure 1).One sample (Omani grapefruit) was picked by hand from a tree in the University of Nizwa. All samples were collected in February 2023 during daytime. The taxonomist identified the samples, and the voucher specimens were deposited in the lab. The collected samples were transported to the University of Nizwa for extraction.

Figure 1. Locations of sample collection.

2.3. Extraction of PAHs

The peel and flesh were initially separated for all local and imported fruits.Then, the collected peel and flesh samples were sliced separately by knife and kept on paper under the shade with a fan for a few days. While drying, the samples were checked carefully to avoid microbial contamination. After drying, the dried peels and flesh samples were crushed into a coarse powder using ball mills. The fruits' peels and coarse flesh powder were kept separately in a plastic bottle to avoid contamination. Each peel and flesh powder sample (5 gm) was taken into a 100 ml round bottom flask and 30 ml of 2% alcoholic NaOH solution was added. The mixture was refluxed using a heating mantle for 4 h [7–14]. After refluxing, the samples were filtered, and the filtrate was transferred into a separatory funnel (250 ml). Initially, the mixture was washed with methanol/distilled water (1:1, 25 ml). Then, cyclohexane (25 ml) was used to extract the PAHs for 2 min from the aqueous layer. The process was repeated twice to extract PAHs from the aqueous layer completely (Figure 2). The cyclohexane layer was washed with methanol/water solution (1:1, 12.5 ml) and finally washed with distilled water (12.5 ml). The cyclohexane layer was pre-concentrated to 2 ml using the rotary evaporator at 40 °C [20].

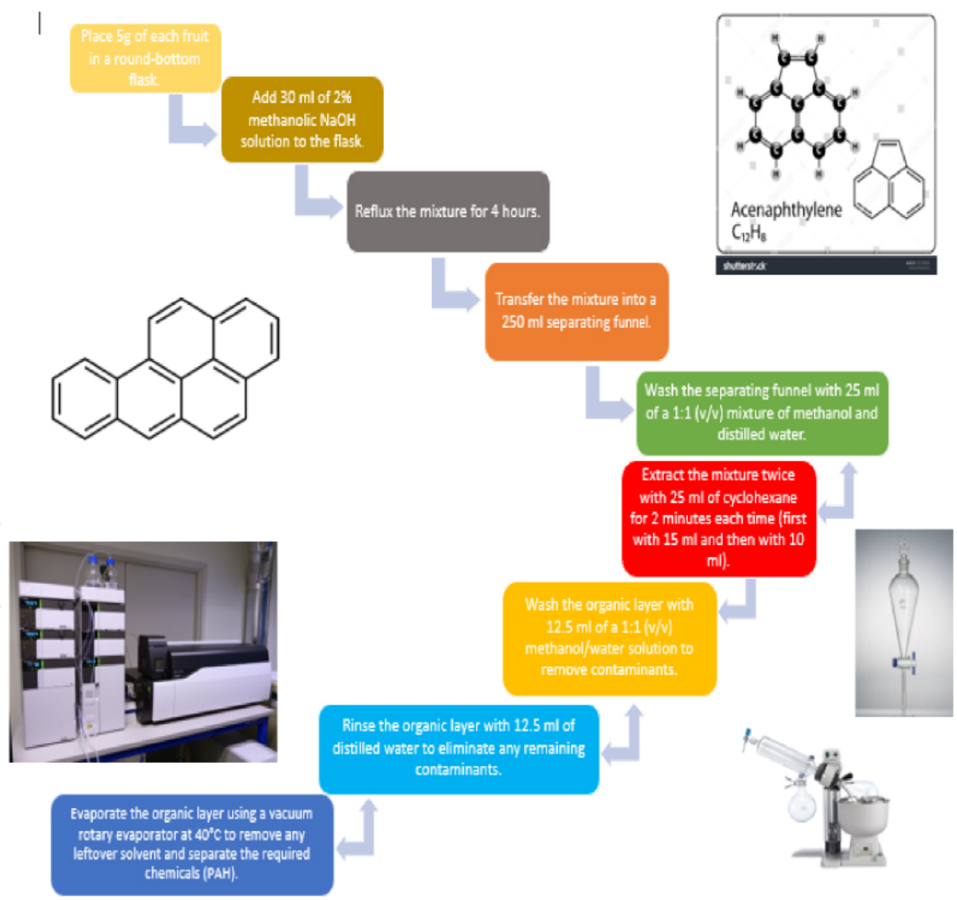

Figure 2. Flow chart of PAHs' extraction procedure.

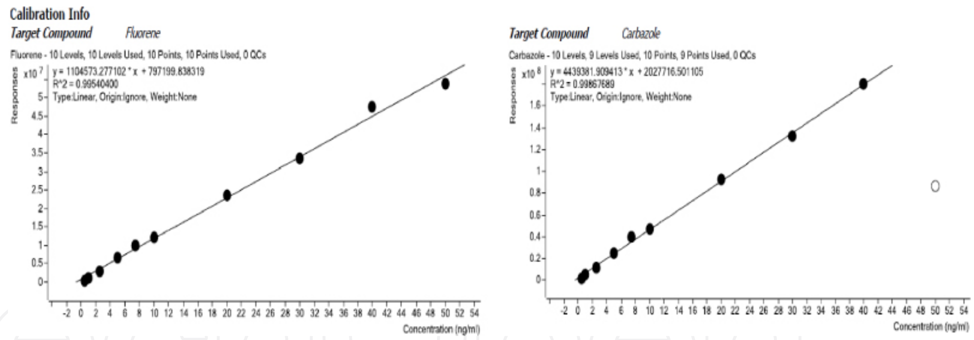

Figure 3. Calibration curves of the PAHs standards.

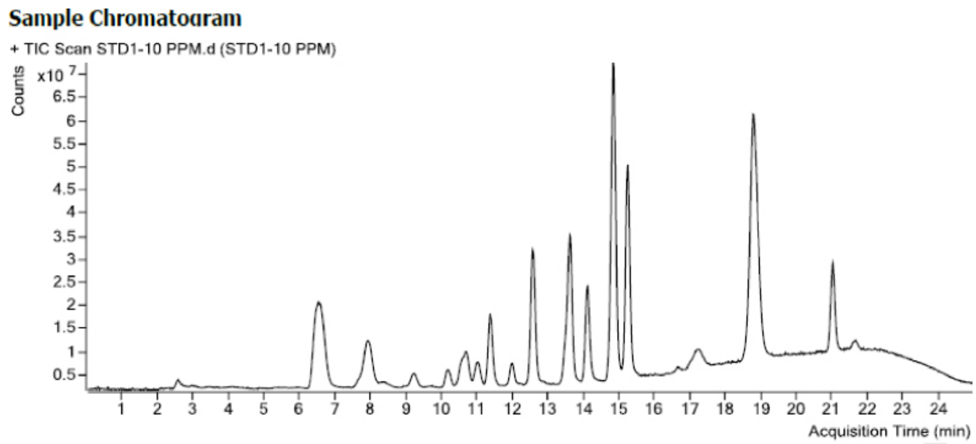

Figure 4. Chromatogram of the 10 ppm standard solution.

2.4. Standard preparation

One milligram of each PAH standard namely acenaphthene (ACE), acenaphthylene (ANP), azulene (AZL), 1,2-benzanthracene (BAA), 2,3-benzanthracene (BZA), benzo[a]pyrene (BAP) benzo[e]pyrene (BEP), chrysene (CHR), anthracene (ANC), coronene (COR), 4H-cyclopenta-phenanthrene (CPDP), fluoranthene (FLA), fluorene (FLE), carbazole (CRB), indene (IND), naphthalene (NAP), and perylene (PER) (99% purity, Sigma-Aldrich Chemical Company, Germany) was dissolved in a 10 ml volumetric flask with acetone solvent.The prepared stock solution was 100 ppm. From the stock solution, ten concentrations, such as 0.5, 1, 2.5, 5, 7.5, 10, 20, 30, 40, and 50 ppm, were prepared by serial dilution with addition of acetone solvent. The standard calibration curves of the PAHs were prepared in VersaCount, and the mixture standard is presented in Figures 3 and 4.

3. Results

PAHs are organic compounds with several fused aromatic rings. They are produced by partial combustion of organic materials such as coal, oil, gas, and wood and are found in various environmental media, including air, water, soil, and sediments. This can lead to leaching into fruits and, hence, into the human body. PAHs have been related to several health problems. PAHs are carcinogenic organic compounds, and are directly related to various cancers, including lung, skin, bladder, liver, and gastrointestinal cancers. In addition, long-term exposure to these carcinogenic compounds causes respiratory difficulties, including respiratory irritation, asthma, lung damage, and development and reproduction abnormalities [15, 19, 20]. Due to these human health problems, we investigated the carcinogenic PAHs in the fruits available in Oman. In this study, Omani and imported fruits, both peel and flesh, were used to extract PAHs using the solvent/solvent extraction method described in Literature. Each extract was pre-concentrated by a rotary evaporator until 2 ml at 40 °C. The pre-concentrated sample and standard at various concentrations (0.5–50 ppm) were analyzed using LC-MS, and the sample analysis results of the collected local and imported fruit peels and flesh samples for seventeen PAHs. The results are presented in Tables 1 and 2.

In the Omani fruit samples, only three PAHs were detected: acenaphthylene (ANP), 1,2-benzanthracene (BAA), and benzo[a]pyrene (BAP) as shown in Tables 1 and 2. The concentrations of the three PAHs, ranked from highest to lowest, are BAP > BAA > ANP.

According to the findings in Tables 1 and 2, only three PAHs, acenaphthylene (ANP), 1,2-benzanthracene (BAA), and benzo[a]pyrene (BAP), were detected in the imported fruits. Remarkably, all fruits' peels and flesh samples showed the presence of these three PAHs. In terms of concentration, benzo[a]pyrene had the highest concentration, followed by BAA > ANP (Tables 1 and 2).

4. Discussion

Fruits are part of a balanced diet that is essential for human well-being. Pollution has expanded dramatically in soil and water, reaching magnitudes that threaten to taint fruits, posing a toxicity risk to humans and animals who consume them. Emissions of PAHs from vehicular traffic and industrial operations significantly influence environmental pollution. PAHs are among the toxic and carcinogenic pollutants, posing severe risks to human health and marine organisms. As a result, this study examined the level of PAH contamination in fruits cultivated in various parts of Oman and in imported fruits and ascertain whether they conform to international standards and guidelines to ensure the safety of fruits and to develop

Table 1. Concentrations of PAHs in Omani fruits and their peels.

PAHs	Lemon (A1)	Lemon peel (A3)	Papaya (A5)	Papaya peel (A7)	Banana (A9)	Banana peel (A11)	Grapefruit peel (A15)	Grapefruit (A13)	Sweet melon (A17)	Sweet melon peel (A19)	Fig (A21)
Fluorene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Carbazole	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Azulene	ND	ND	ND	ND	ND	0.413	ND	ND	ND	ND	ND
Naphthalene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Indene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Acenaphthylene	0.048 ± 0.12	0.052 ± 0.09	0.048 ± 0.68	0.049 ± 0.09	0.199 ± 0.10	0.059 ± 0.77	0.054 ± 0.54	0.052 ± 0.21	0.050 ± 0.16	0.051 ± 0.74	0.048 ± 0.24
Acenaphthene	ND	ND	ND	ND	ND	ND	ND		ND	ND	ND
Anthracene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
4h-cyclopenta def phenanthrene		ND		ND	ND		ND	ND		ND	ND
Fluoranthene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
2,3-Benzanthracene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Chrysene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1,2-Benzanthracene	0.22 ± 0.25	0.224 ± 0.65	0.222 ± 0.71	0.222 ± 0.15	0.223 ± 0.17	0.222 ± 0.44	0.223 ± 0.14	0.226 ± 0.39	0.222 ± 0.53	0.222 ± 0.08	0.223 ± 0.23
Perylene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Benzo[e]pyrene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Benzo[a]pyrene	0.833 ± 0.10	0.834 ± 0.23	0.833 ± 0.81	0.834 ± 0.45	0.834 ± 0.22	0.833 ± 0.89	0.834 ± 0.13	0.833 ± 0.17	0.833 ± 0.63	0.835 ± 0.13	0.834 ± 0.23
Coronene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

ND: Not detected.

Table 2. PAH concentrations in imported fruits and their peels.

PAHs	Lemon (A2)	Lemon peel (A4)	Papaya (A6)	Papaya peel (A8)	Banana (A10)	Banana peel (A12)	Grapefruit (A14)	Grapefruit peel (A16)	Sweet melon (A18)	Sweet melon peel (A20)
Fluorene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Carbazole	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Azulene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Naphthalene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Indene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Acenaphthylene	0.051 ± 0.14	0.048 ± 0.40	0.049 ± 0.77	0.068 ± 0.11	0.054 ± 0.29	0.646 ± 0.19	0.048 ± 0.27	0.064 ± 0.81	0.049 ± 0.13	0.051 ± 0.32
Acenaphthene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Anthracene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
4h-cyclopenta def phenanthrene	ND	ND		ND	ND	ND	ND	ND	ND	ND
Fluoranthene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
2,3-Benzanthracene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Chrysene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1,2-Benzanthracene	ND	0.223 ± 0.56	0.223 ± 0.92	0.223 ± 0.26	0.223 ± 0.33	0.223 ± 0.28	0.223 ± 0.45	0.222 ± 0.23	0.222 ± 0.17	0.222 ± 0.40
Perylene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Benzo[e]pyrene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Benzo[a]pyrene	0.833 ± 0.88	0.838 ± 0.15	0.833 ± 0.10	0.833 ± 0.76	0.833 ± 0.19	0.833 ± 0.11	0.833 ± 0.73	0.834 ± 0.49	0.833 ± 0.74	0.835 ± 0.09
Coronene	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

ND: Not detected.

methods to prevent pollution. The data provided by this study may benefit both the Omani national and global research communities. In addition, the data from the study will be helpful for research scholars to carry out future work.

Diet is one of the major sources of PAHs in humans [21–23]. Long-term exposure in the human body can affect the major organs and lead to different cancers. Therefore, one of the main aim of scientists/researchers and responsible authorities is to detect and quantify the content of PAHs that are accumulated in the fruits and vegetables grown in Oman. Several fruits and vegetables are cultivated to fulfill local needs, but to cover their shortage, vegetables and fruits are imported from neighboring countries. Oman's agricultural production sector is weak due to hot weather and the lack of fertile land and water resources. Oman is an oil-producing country in the Persian Gulf; therefore, the Omani people are busy with oil-related activities throughout the year. The widespread use of fossil fuels is a vital reason for producing PAHs in the environment. However, there is no definite indicator in Oman to evaluate the levels of PAHs in vegetables and fruits in the agricultural sector. This study aimed to quantify the level of PAHs in samples of commonly consumed fruit from local markets and imported fruits from supermarkets. The results obtained were used to calculate a preliminary estimation of the contribution of these fruits as a source of PAH exposure in the Omani population.

In light of the facts mentioned above, a study was carried out in Brazil to ascertain the amount of PAHs present in fruits and vegetables [16]. In order to better quantify the dietary exposure of Brazilians to PAHs, this study measured the amounts of PAHs in samples of apples, cabbages, grapes, lettuces, pears, and tomatoes. Six separate locations were used to gather fruit and vegetable samples, then examined for 16 PAHs. The analytical procedure included saponification with methanolic KOH, liquid–liquid extraction with cyclohexane, cleanup on a silica gel column, and analysis by high-performance liquid chromatography with fluorescence detection. The results showed that PAHs were present in lettuces at a mean level of 13.53 μg/kg, tomatoes at 9.50 μg/kg, cabbage at 8.86 μg/kg, apples at 4.05 g/kg, grapes at 3.77 μg/kg, and pears at 3.87 μg/kg. One of the most typical hazardous PAH members is Benzo(a)anthracene, estimated at 89% of all samples examined. There was no evidence of chrysene in any analyzed sample [24].

Kumar *et al.* [18] determined the occurrence of PAHs in fruits. The study ascertained the presence of PAHs in regularly consumed fruits in India and examined the potential health concerns linked to them. The scientists collected 56 fruit samples from several marketplaces in Delhi, India. Apples, bananas, guava, mango, papaya, and watermelon were among the commonly consumed fruits. GC-MS was used to examine the samples for 16 key PAHs. The findings revealed that all fruit samples had detectable amounts of PAHs. Apple and banana samples had the highest quantities of PAHs, with total PAH values ranging from 0.34 to 2.46 ng/g and 0.33 to 2.39 ng/g, respectively. Watermelon samples had the lowest PAH values, with total PAH amounts ranging from 0.03 to 0.29 ng/g. The researchers also calculated the dietary intake of PAHs from fruit-eating for various age groups. The findings revealed that youngsters had a greater estimated daily intake of PAHs from fruit-eating than adults. The predicted lifetime cancer risk from PAHs in fruit intake was likewise shown to be greater in youngsters than in adults. According to the study, fruits can be a substantial source of PAH exposure, especially for youngsters. The study emphasized the need to monitor fruits' PAH levels and take steps to prevent contamination [18].

Fernández-González *et al.* [19] analyzed the amounts and sources of PAHs in various fruits from Northwest Spain. The researchers collected 90 fruit samples from local markets and farms, including apples, pears, peaches, nectarines, plums, strawberries, raspberries, blueberries, blackberries, and cherries. GC-MS was used to examine the samples for 16 key PAHs. The findings revealed that PAHs were present in all fruit samples, with the highest quantities detected in strawberries and raspberries. PAH levels varied by fruit variety, with berries and stone fruits holding more significant amounts than pome fruits. In addition, multivariate statistical analysis was employed in the study to identify probable sources of PAH contamination. The findings indicated that PAHs in fruits mainly originated from vehicle exhaust emissions and agricultural practices such as pesticide and fertilizer usage. According to the study's findings, fruits may be a substantial source of PAH exposure, and the amounts of PAH in fruits can vary depending on the kind of fruit and the source of contamination. The study emphasizes the need to monitor PAH levels in fruits and take steps to prevent contamination [19].

A total of six fruits, such as grapefruit, sweet melon, banana, papaya, lemon, and fig, which were grown in Oman, were collected from the Al-Hoti market, Al-Khuwair, Muscat. Soon after the collection of fruits, peels were separated from the flesh, and all of them were sliced and dried for powder formation. The carcinogenic PAHs were extracted from the powder of the peels and flesh using the well-established solvent-solvent extraction method. The PAH extract from each sample was pre-concentrated and injected into the LC-MS to determine PAHs. The results for PAHs in the fruits grown in Oman are presented in Tables 1 and 2. Tables 1 and 2 showed that among the seventeen PAHs, only three were detected: ANP, BAA, and BAP. The concentrations of the three PAHs, ranked from highest to lowest, are BAP > BAA > ANP. The highest amount of ANP was found in the papaya peels and the lowest in the fig among the six local fruits. Grapefruit contained the highest levels of 1,2-benzanthracene (BAA) and the lowest levels were in sweet melon. Similarly, almost the same amount of benzo[a]pyrene (BAP) was obtained in all the fruits grown in Oman. The other PAHs were not detected in the Omani fruits by LC-MS. None of the seventeen (PAHs) are used as pesticides. They are typically associated with environmental contaminants that lead to serious health risks if exposure occurs at high levels. The present experimental data is unable to be compared with the reference data due to the absence of an official list of the Maximum Residue Limit (MRL) published by the Omani government. Overall, these results suggest that the levels of PAHs in the selected fruits' flesh and peels are generally low, within the permissible limit.

On the other hand, five imported fruits, Turkish grapefruit, Yemeni sweet melon, Ecuadorian banana, Sri Lankan papaya, and South African lemon, were examined in this present study for the extraction and quantification of PAHs were sourced from Lulu Hypermarket, Muscat. Among the analyzed PAHs, only three PAHs, acenaphthylene (ANP), 1,2-benzanthracene (BAA), and benzo[a]pyrene (BAP) were detected in the fruits' samples by LC-MS. The results are presented in Tables 3 and 4. Remarkably, all analyzed samples contained these three PAHs in varied concentrations. In terms of concentration, BAP had the highest concentration, followed by BAA > ANP. The highest amount of ANP was in the papaya peels and the lowest in the fig among the five imported fruits. For 1,2-benzanthracene (BAA), the highest amount was grapefruit, and the lowest was sweet melon. Similarly, almost the same amount of benzo[a]pyrene (BAP) was found in all the fruits imported from other countries. The remaining PAHs could not be found in the local and imported fruits. No significant difference was seen between the analyzed PAHs in the local Omani and imported selected fruits. Interestingly, the local and the imported fruit samples contained the same three PAHs: BAP, BAA, and ANP. Also, a similar trend was found in the samples, either in the peels or the flesh samples. The other members of analyzed PAHs were absent in both local and imported fruit samples. The maximum permissible limit of benzo[a]pyrene (BAP) in fruit samples, either processed or raw fruits, was set to 10 μg/kg by the European Union [21]. On the other hand, the permissible limit for other members of PAHs in the selected fruits was not available. The Environmental Protection Agency [22] in the United States has established a maximum concentration limit (MCL) for BAP in drinking water of 0.2 μg/kg. However, no permissible limit for PAHs in fruits is established [22]. Additionally, the World Health Organization (WHO) did not set specific maximum limits for BAP, BAA, and ANP in fruits and food products. However, the WHO has established a provisional tolerable monthly intake (PTMI) for benzo[a]pyrene, acenaphthylene, and 1,2-Benzanthracene to be 10 μg/kg, 60 μg/kg, and 30 μg/kg of body weight [23], respectively.

5. Conclusion

Sensitive LC-MS and gradient flow with a UV detector were used to analyze each extracted sample and quantify different PAHs in the fruit peels and flesh. This study extracted the PAHs using the QuEChERS method from different fruit samples grown in Oman or imported from different countries. Long-term intake of PAHs through our diet causes serious health problems such as various cancers, liver, and cardiovascular diseases. Therefore, we must check the level of PAHs in the fruits before the consumption of fruits. The experimental results showed that only three PAHs out of 17, namely BAP, BAA, and ANP, were present in the analyzed samples. The other members were absent from all the analyzed samples. The amount of BAP, BAA, and ANP in the analyzed fruit samples was within the permissible limit. According to the standards and regulations of the European Union (EU) and the provisional tolerable monthly intake (PTMI) list set by the World Health Organization (WHO), the concentrations of these PAHs are lower than the standard levels. Nevertheless, the country must take action and set regulations to control the PAH levels in fruits and keep them within the standard levels. It is critically important to continue monitoring the levels of PAHs in fruits and their peels to ensure their safety for human consumption. Regular testing should be conducted using validated analytical methods to effectively monitor the levels of PAHs in fruits and their peels. These tests should focus on detecting specific PAH compounds known to be harmful to human health. Additionally, measures can be taken to reduce the levels of PAHs in fruits, such as implementing good agricultural practices to prevent contamination during cultivation, processing, and storage. Furthermore, peeling or washing fruits before consumption can help to reduce PAH levels in some cases.

Data availability

The source data for Figures and Tables are available on request from the corresponding author Mohammad Amzad Hossain.

Funding

Not Applicable

Conflict of interest

The authors declare that there is no conflict of interest.

Acknowledgements

The authors are grateful to the University of Nizwa for providing all lab and instrumental facilities. The authors are also grateful to Mr. Partha Sarathy Elumalai for his excellent contribution towards analyzing the samples. The authors appreciate the efforts of the Writing Center at the University of Nizwa throughout the revising and editing process.

References

1 Cannoosamy K, Pem D, Bhagwant S, Jeewon R. Is a nutrition education intervention associated with a higher intake of fruit and vegetables and improved nutritional knowledge among housewives in Mauritius. *Nutrients*. 2016;**8**(12):723.

2 Oman Ministry of Finance. Geography [Internet]. 2022. Available from: https://fm.gov.om/about-oman/state/geography/.

3 Al-Yahyai R, Al-Said F, Opara L. Deciduous fruit crops of Oman. *Acta Horticul*. 2014;**2014**(1059):185–187.

4 Abdel-Shafy H, Mansour M. A review on polycyclic aromatic hydrocarbons: source, environmental impact, effect on human health and remediation. *Egypt J Petrol*. 2016;**25**(1):107–123.

5 Kulshrestha A, Massey D, Masih J, Taneja A. Source characterization of trace elements in indoor environments at urban, rural and roadside sites in a semi-arid region of India. *Aerosol Air Qual Res*. 2014;**14**(6):1738–1751.

6 Masih J, Masih A, Kulshrestha A, Singhvi R, Taneja A. Characteristics of polycyclic aromatic hydrocarbons in indoor and outdoor atmosphere in the North central part of India. *J Hazard Mater*. 2010;**177**(1–3):190–198.

7 Akyüz M, Çabuk H. Gas–particle partitioning and seasonal variation of polycyclic aromatic hydrocarbons in the atmosphere of Zonguldak, Turkey. *Sci Total Environ*. 2010;**408**(22):5550–5558.

8 Paris A, Ledauphin J, Poinot P, Gaillard J. Polycyclic aromatic hydrocarbons in fruits and vegetables: origin, analysis, and occurrence. *Environ Pollut*. 2018;**234**: 96–106.

9 Kim K, Jahan S, Kabir E, Brown R. A review of airborne polycyclic aromatic hydrocarbons (PAHs) and their human health effects. *Environ Int*. 2013;**60**: 71–80.

10 Ahmed M, Mostafa G, Rasbi S, Askar A. Residues of polycyclic aromatic hydrocarbons in fish and water from Sultanate of Oman. *Int J Environ Health Res*. 1998;**8**(1):77–84.

11 Moh.gov.om. Health Releases Cancer Statistics in Sultanate of Oman 2019 - Media Center Display Page - Ministry of Health [Internet]. 2022. Available from: https://www.moh.gov.om/en/-/-2-10.

12 Ministry of Health, Sultanate of Oman. النظرة المستقبلية 2050 [Future Vision 2050] [Internet]. 2022. Available from: https://moh.gov.om/documents/2317925/0/%D8%A7%D9%84%D9%86%D8%B8%D8%B1%D8%A9+%D8%A7%D9%84%D9%85%D8%B3%D8%AA%D9%82%D8%A8%D9%84%D9%8A%D8%A9+2050/c7d80af9-9aab-de57-6545-370ad7e68275.

13 Mallah M, Changxing L, Mallah M, Noreen S, Liu Y, Saeed M. Polycyclic aromatic hydrocarbon and its effects on human health: an overeview. *Chemosphere*. 2022;**296**: 133948.

14 Atler K, Muñoz D, Knowles C, Noe A, Fruhauf C, McGirr K, et al. Improving diabetes outcomes with a multimodal intervention for less advantaged adults with type 2 diabetes. *OBM Integr Compl Med*. 2021;**6**(4):043.

15 Lattimer J, Haub M. Effects of dietary fiber and its components on metabolic health. *Nutrients*. 2010;**2**(12):1266–1289.

16 Adebawo O, Salau B, Ezima E. Fruits and vegetables moderate lipid cardiovascular risk factors in hypertensive patients. *Lipids Health Dis*. 2006;**5**: 14.

17 Rojo Camargo M, Toledo M. Polycyclic aromatic hydrocarbons in Brazilian vegetables and fruits. *Food Control*. 2003;**14**(1):49–53.

18 Kumar V, Balomajumder C, Roy P. Polycyclic aromatic hydrocarbons (PAHs) in fruits: a study from the northwestern Indo-Gangetic Plain, India. *Food Addit Contam: B*. 2015;**8**(1):51–58.

19 Fernández-González R, Iglesias-Otero MA, Martínez-Carballo E, Simal-Gándara J. Polycyclic aromatic

hydrocarbons in fruits from northwest Spain: occurrence and potential sources. *Food Chem Toxicol*. 2017;**108**(Pt B):579–585.

20 Hossain MA, Salahuddin SM, Kabir MJ. Comparison study of polycyclic aromatic hydrocarbons (PAHs) in Bangladeshi compressed natural gas (CNG) and diesel engine vehicles by GC-MS. *Asian J Energy Environ*. 2009;**10**(01):11–18.

21 Hossain MA, Salahuddin SM, Abu Hanif M, Kundu PK. Carcinogenic polycyclic aromatic hydrocarbon (PAH), anthracene in cabbage samples from Bangladesh. *Asian J Food Agro Ind*. 2009;**2**(02):315–320.

22 US Environmental Protection Agency (EPA). Polycyclic Aromatic Hydrocarbons (PAHs) - Basic Information [Internet]. 2019. Available from: https://www.epa.gov/air-research/polycyclic-aromatic-hydrocarbons-pahs-basic-information.

23 European Union. Commission Regulation (EU) No 836/2011 of 19 August 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs. *Offi J Eur Union*. 2011;**L215**: 4–8.

24 FAO/WHO. Maximum levels for certain contaminants in food [Internet]. 2019. Retrieved from http://www.fao.org/3/ca5429en/ca5429en.pdf.