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POLYCYCLIC AROMATIC HYDROCARBONS OCCURRENCE IN CEREAL BASED-PRODUCTS - A REVIEW

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) are environmental contaminants known to have toxic properties, carcinogenic and mutagenic potential, cereals and derivative products being the most important sources of PAH exposure to humans as a result of the high intake of this kind of products. The aim of this study was to investigate the occurrence of PAH in cereals and cereal-based products and the effect of different factors on the content of these compounds. The factors that influence the PAH content of cereal-based products were the raw material used in the recipe, the category of processed product, the baking parameters (time and temperature), the type of fuel used. The maximum level for benzo(a)pyrene (BaP) and sum of BaP, benzo(a)anthracene (BaA), benzo(b)fluoranthene (BbF), and chrysene (ChR) in processed cereal-based food and baby foods for infants and young children was established by regulation no. 835/2011. This study can provide an overview of the PAH content of different cereal-based products commercialized on the market.

***Key words**: benzo(a)pyrene, bread, cereals, contamination, PAH.*

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) represent a class of organic, colourless, white or pale-yellow solid compounds (Abdel-Shafy and Mansour, 2016). These compounds are semi-volatile or non-volatile and ubiquitous. PAHs have two or more linear or angular condensed aromatic rings, the number varying from two to ten (Huang and Penning, 2014). Based on the number of fused aromatic rings, PAHs can be divided into light molecular PAHs (2-4 rings) and high molecular PAHs (> 5 rings).

PAHs can result from the incomplete combustion of organic material or heat-induced decomposition, being omnipresent pollutants in the environment (EFSA, 2008). The contamination sources can be natural or anthropogenic, PAHs being present in water, soil, air, and implicitly in cereals (Abdel-Shafy and Mansour, 2016).

PAHs have been classified by IARC as possible carcinogens (Group 2B), probable carcinogens (Group 2A), and carcinogens for humans (Group 1) (IARC, 2010). The United States Environmental Protection Agency (US EPA) have been listed 16 priority PAHs that poses carcinogenic properties as follows: acenaphthene, acenaphthylene, anthracene, benzo(a)anthracene (BaA), benzo(a)pyrene (BaP), benzo(b)fluoranthene (BbF), benzo (g,h,i)perylene, benzo(k)fluoranthene, chrysene (ChR), dibenzo(a,h)anthracene, fluoranthene, fluorene, indene(1,2,3-cd)pyrene, naphthalene, phenanthrene, and pyrene (US EPA, 2010).

In Europe, based on the risk potential EFSA (2008) selected 4 PAHs to be indicators for the carcinogenic PAHs: BaP, BaA, BbF, and ChR. Among all PAHs, BaP is considered as the most carcinogenic compound, being classified as a carcinogen (Group 1) (Joint FAO/WHO, 2005), while BbF, BaA and ChR were included in the class of possible carcinogens (Group 2B) (IARC, 2010).

Cereals are important sources for both human and animal nutrition, being widely consumed. All over the world, grains such as wheat, rice, corn and cereal derived products such as bread, biscuits, pasta and other cereal-based products are very high consumed. Cereals provide energy due to the vitamin, protein, lipid and mineral content (Kamalabadi et al., 2019; Rozentale et al., 2017).

Although processed cereals are in general characterized by low levels of PAH contamination, according to EFSA, cereals, bread and cereal-based products are major sources of human exposure to PAHs (SCF, 2002; EFSA, 2008) mainly because of their high consumption volume (Ibáñez et al., 2005; Ciecierska and Obiedziński, 2013; Abramsson-Zetterberg et al., 2014; Escarrone et al., 2014).

Additionally, PAH contamination is inevitable in bread and cereal-based products due to the high temperature of the baking process. A maximum content of PAHs is thought to form when materials are heated to temperatures ranging between 500-550°C, while the bread baking temperature is around 250°C (McGrath et al., 2007).

The Commission Regulation (EU) no. 835/2011 does not establish the maximum level for PAHs present in cereals, it only set the maximum level of 1 μg/kg for BaP, and sum of BaP, BaA, ChR, and BfF in processed cereals and baby foods for infants and young children.

Research on the PAH content of cereals, flour, bread and cereal-based products is limited. However, when examining the existing studies, some conclusions can be reached regarding the occurrence and level of PAHs in cereals and cereal-based products, and the factors that influence the content of these compounds.

In this study we realized a literature review to analyse the content of PAHs in cereals and cereal derived products, and we also evaluated the influence of different factors on the level of these contaminants.

MATERIALS AND METHODS

Data search and selection criteria

The present literature review was performed in order to evaluate the PAH contamination of cereals and cereal-based products by performing a compressive search during 2010 until 2024 of the articles indexed in Web of Science, and also from the databases of Google Academic, and Scopus. Original research articles and reviews published in English were included in the search and the following terms were used: "*polycyclic aromatic hydrocarbons*", "*PAH*", "*benzo(a) pyrene*", "*BaP*", "*cereals*", "*flours*", "*bread*", "*pasta*", "*biscuits*", "*cereal products*", and "*cereal-based products*".

Once the search was realized, the articles were analysed and the most relevant manuscripts for this study were selected. Based on the selection, this review was synthesized and divided on the category of cereal derived products: cereals and flours, bread and other cereal-based products.

RESULTS AND DISCUSSIONS

PAH content in cereals and flours

The concentration of PAHs in cereals must be monitored, the content being influenced by the degree of contamination of the region in which the grains were cultivated, the degree of industrialization of the area, the harvest year, and the way of drying the cereals. The PAH content of cereals and flours reported in literature are presented in Table 1.

The PAH contamination of wheat was monitored in several countries, like Poland (Ciercierska and Obiedziński, 2013; Roszko et al., 2020; Thabit et al., 2022), Russia, Germany, France, Ukraine, Latvia, Lithuania, Estonia (Thabit et al., 2022), Romania (Muntean et al., 2019a; Thabit et al., 2022), California (Kobayashi et al., 2008), Pakistan (Aamir et al., 2021) and results showed that the content varies depending on the country of origin of the grains.

The PAH content of wheat samples is influenced by the geographical area where it was cultivated and the degree of pollution. As it can be noticed in Table 1, BaP was detected only in wheat samples from Poland (Roszko et al., 2020).

Muntean et al. (2019a) studied the PAH content in winter wheat cultivated in three different regions in Romania with different pollution levels, in the period 2012-2014, and determined the presence of BaA, naphthalene and anthracene. In this study, ChR was not detected, although when Thabit et al. (2022) studied the contamination level of wheat from Romania, it was determined a content of ChR of 3.70 μg/kg, with a degree of sample contamination of 90%. As expected, pollution had an effect on the PAH contamination, the most contaminated wheat samples being those cultivated in the high traffic area, with 15 PAH content of 7.83-8.67 μg/kg, followed by those from the experimental field (4.8-5.37 μg/kg), and those in the area with historical contamination (3.06-3.80 μg/kg) (Muntean et al., 2019a). Contrary, in the study realized by Roszko et al. (2020) it was shown that the PAH content was not influenced by the region of cultivation of wheat sample from Poland.

Table 1. PAH content of cereals and flours from different regions

Cereal/ flour	**Country**	**BaP**	**BaA**	**ChR**	**BbF**	**Other PAHs in high proportion, µg/kg**	**4 PAH, µg/kg**	**Sum PAH, µg/kg**	**Reference**
		µg/kg							
Wheat	Poland	0.09	0.08	0.18	0.08	2.24 NAP	0.43	1.03 (16)	Roszko, 2020
	Poland	nd	0.05±0.00	nd	nd	1.14±0.09 PHN 0.58±0.14 FLT	*	2.35±0.15 (19)	Ciercierska, Obiedziński, 2013
	Poland	*	*	*	4.10	*	*	*	Thabit, 2022
	Russia	*	*	*	4.15	1.92 PHN	*	*	Thabit, 2022
	Germany	*	*	3.22	*	2.40 ANT	*	*	Thabit, 2022
	France	*	3.82	*	*	*	*	*	Thabit, 2022
	Ukraine	*	*	*	3.92	*	*	*	Thabit, 2022
	Latvia	*	4.20	*	*	*	*	*	Thabit, 2022
	Lithuania	*	*	*	*	5.86 FLT	*	*	Thabit, 2022
	Estonia	*	2.85	*	*	*	*	*	Thabit, 2022
	Romania	*		3.70	*	*	*	*	Thabit, 2022
	Romania	*	0.03-0.16		*	1.39÷3.60 NAP; 1.32÷2.93 FLR	*	*	Munteanu, 2019a
	Pakistan	*	*	*	*	*	*	169 (16)	Aamir et al., 2020
Maize	Pakistan	*	*	*	*	*	*	159 (16)	Aamir et al., 2020
Rice	Pakistan	*	*	*	*	*	*	53 (16)	Aamir et al., 2020
Rice	Spain	-	-	-	-	0.097±0.006 ÷ 5.5±0.3 ANT - ÷ 4.9±0.3 FLR	-	0.18÷10.54 (16)	Rascón et al., 2018
Corn	Romania	*	nd-0.07	*	*	1.23÷4.59 NAP 0.02÷0.25 ANT	*	*	Munteanu, 2019b
Barley	Poland	-	-	-	-	-	-	-	Thabit, 2022
	Russia		3.41	*	5.81	*	*	*	Thabit, 2022
	Germany	-	-	-	-	-	*	*	Thabit, 2022
	France	-	-	-	-	*	*	*	Thabit, 2022
	Ukraine		4.55	*	*	*	*	*	Thabit, 2022
	Latvia	*	*	5.61	*	*	*	*	Thabit, 2022
	Lithuania	*	*	*	*	6.21 FLR	*	*	Thabit, 2022
	Estonia	-	-	-	-	*	*	*	Thabit, 2022
	Romania	-	-	-	-	*	*	*	Thabit, 2022
Rye	Poland	nd	0.06±0.00	nd	nd	1.45±0.23 PHN 0.19±0.04 FLT	*	2.93±0.22 (19)	Ciercierska, Obiedziński, 2013
Cereals	Latvia	0.061	0.121	0.288	0.139	*	0.61	*	Rozentale et al., 2017
Wheat flour	Poland	nd	nd	nd	nd	0.64±0.09 PHN 0.19±0.01 FLT	*	1.07±0.14 (19)	Ciercierska, Obiedziński, 2013
Wheat flour	Spain	-	-	-	-	0.013±0.001 NAP 0.79±0.04 ANT	-	0.803 (16)	Rascón et al., 2018
Corn flour	Spain	-	-	-	-	0.086±0.005 NAP 2.4±0.2 ANT	-	2.486 (16)	Rascón et al., 2018
Rye flour	Poland	nd	nd	nd	nd	0.59±0.11 PHN 0.20±0.01 FLT	*	1.25±0.15 (19)	Ciercierska, Obiedziński, 2013
Wholemeal rye flour	Poland	nd	nd	nd	nd	0.51±0.11 PHN 0.21±0.01 FLT	*	1.35±0.20 (19)	Ciercierska, Obiedziński, 2013
Wheat bran	Poland	nd	0.05	nd	0.11	0.79±0.05 PHN 0.17±0.04 FLT	*	1.87±0.18 (19)	Ciercierska, Obiedziński, 2013
Rye bran	Poland	nd	0.05	nd	nd	0.76±0.09 PHN 0.59±0.01 FLT	*	2.65±0.17 (19)	Ciercierska, Obiedziński, 2013
Bran from rye grinding-wholemeal rye flour	Poland	nd	0.08	0.16	nd	1.19±0.11 PHN 0.55±0.03 FLT	*	3.65±0.23 (19)	Ciercierska, Obiedziński, 2013
White flour	Kuwait	nd	nd	nd	nd	4.33 NAP; 31.4 PHN; 5.04 FLT	*	43.1 (16)	Al-Rashdan et al., 2010
Wholewheat flour	Kuwait	nd	nd	nd	nd	12.9 NAP; 32.0 PHN; 8.14 FLT	*	56.4 (16)	Al-Rashdan et al., 2010
White rice dried by LPG, in oven and by wood	Brazil	nd	nd	nd	nd	7.0 NAP 1.4-2 PHE	*	*	Escarrone et al., 2014
Parboiled rice dried by LPG, in oven and by wood	Brazil	nd	nd	nd	nd	1.0-6.0 NAP 1.0 PHE 1.0 FLT	*	*	Escarrone et al., 2014
Brown rice, parboiled	Brazil	nd	nd÷1.5±0.7	0.9±0.1 ÷	nd	3.5±0.7 ÷ 81.4±0.0 PHE 6.2±0.8 ÷ 35.6±0.5 NAP	*	12.7÷131.6 (16)	Bertinetti et al., 2018

Cereal/	Country	BaP	BaA	ChR	BbF	Other PAHs in high	4 PAH,	Sum PAH,	Reference
brown rice, polished rice dried by wood				3.3±0.7					
Brown rice, parboiled brown rice, polished rice dried by rice husk	Brazil	nd	nd÷1.1±0.2	0.9±0.3 ÷ 4.9±0.7	nd	0.4±0.2 ÷ 44.4±1.9 PHE 4.7±0.1 ÷ 14.1±1.5 NAP	*	6.6÷120.8 (16)	Bertinetti et al., 2018
Brown rice, parboiled brown rice, polished rice dried by LPG	Brazil	nd	nd	nd÷ 2.4±0.2	nd	nd ÷ 22.9±1.3 PHE nd ÷ 9.9±1.8 NAP	*	nd÷63.9 (16)	Bertinetti et al., 2018
Brown rice, parboiled brown rice, polished rice dried by electric heating	Brazil	nd	nd	nd	nd	nd ÷ 4.0±0.9 PHE 2.9±0.3 ÷ 5.5±0.6 NAP	*	2.9÷9.1 (16)	Bertinetti et al., 2018

BaP- benzo(a)pyrene, BaA- benzo(a)anthracene, ChR- chrysene, BkF- benzo(k)fluoranthene, NAP- naphthalene, PHN-phenanthrene, FLT- fluoranthene, ANT- anthracene, FLR- fluorene, nd- not detectable, LPG- liquefied petroleum gas,
"-" compound not found, *- compounds were not studied or reported. PAHs are presented as range or as mean, as reported in references. The number in parentheses indicates the number of compounds included in the reported sum.

The harvest year had no influence on the PAH content of wheat samples cultivated in the same country (Munteanu et al., 2019a; Roszko et al., 2020).

Low molecular weight PAHs were mainly found in wheat samples cultivated in different European countries (Ciecierska and Obiedziński, 2013; Muntean et al., 2019a; Roszko et al., 2020; Thabit et al., 2022), phenanthrene, anthracene, fluoranthene, fluorene, naphthalene, BaA, ChR and BbF being detected, their concentration differing according to the area of origin, the degree of positivity of the samples being over 50% (Thabit et al., 2022).

Also, in a study conducted in California, the predominant PAHs in wheat cultivated in the same season and harvested from different geographical areas were the low molecular weight ones, with 2 to 4 rings, with naphthalene being the most abundant PAH (Kobayashi et al., 2008). In general, PAH profiles suggest contamination of wheat from emission sources resulting from combustion (liquid fossil fuel, coal, biomass).

When it comes to barley samples collected from different European countries, fluoranthene, BaA, ChR and BbF contaminated the samples in a proportion greater than 70% (Thabit et al., 2022).

Additionally, Muntean et al. (2019b) determined the degree of contamination of corn cultivated in regions from Romania with different pollution degrees, and the most contaminated samples were those from the area with heavy traffic, the PAH content varying between 6.34-9.28 μg/kg.

Compared to the results reported in European countries, in Pakistan, Aamir et al. (2021) determined a higher content for 16 PAHs in cereals (maize, rice and corn), probably as a consequence of pollution. A much lower content of 16 PAH was determined for the rice cultivated in Spain (Rascón et al., 2018) compared to the results reported by Aamir et al. (2021).

Besides cereals, flours were also analysed to determine the PAH contamination. The wheat, corn, rye, and wholemeal rye flours didn't present the 4 carcinogenic PAHs (Al-Rashdan et al., 2010; Ciecierska and Obiedziński, 2013; Rascón et al., 2018). BaA was detected in the bran of wheat, rye, and the mixture of rye bran and wholemeal rye flour brans, BbF was detected in wheat bran, and ChR was detected in the bran flours mixture. The PAH content of wheat and rye flours from the European countries (0.803-1.25 μg/kg) was lower than the PAH content of brans (1.35-3.65 μg/kg). The wheat, rye and wholemeal flours analysed by Ciecierska and Obiedziński (2013) were characterized by a high content of light PAH (phenanthrene, anthracene, fluoranthene and pyrene) of 93, 80 and 61% of all PAHs. The brans obtained from these cereals presented a

content of light PAH of 70, 63 and 74%, respectively.
The 16 PAH content of white and wholewheat flours from Kuwait was much higher (43.1-56.4 μg/kg) than the content of 19 HAP of wheat flours from Europe (Al-Rashdan et al., 2010). This contamination might be the result of pollution of the region in which the grains were cultivated (Kobayashi et al., 2008; Abdel-Shafy and Mansour, 2016; Muntean et al., 2019a).
A common practice for maintaining the quality of grain is to dry it to a specific moisture content and maintain it. Reducing the moisture content is achieved in most countries by burning gasses and wood, hence exposure to smoke. It was observed that by drying white rice by burning wood and by using the oven, phenanthrene was accumulated at a concentration of 1.4 and 2 μg/kg, respectively (Escarrone et al., 2014). By drying white rice with liquefied petroleum gas (LPG) the formation of naphthalene (7 μg/kg) resulted, which was reduced by 14% when parboiled rice was used. In parboiled rice dried in oven, a 4 μg/kg naphthalene content was determined, while when this rice was dried by wood, besides naphthalene (5 μg/kg), phenanthrene (1 μg/kg), and fluoranthene (1 μg/kg) were found (Escarrone et al., 2014).
Contrary, when Bertinetti et al. (2018) studied the influence of drying method on the 16 PAH content of rice, the highest concentration of 16 PAHs (131.6 μg/kg) resulted when wood was used for drying which was 2.9-fold lower for rice husk, 8.3-fold lower for LPG, and 17.1-fold lower when electric heating was used. The concentration of PAHs was found to increase by 120.6-663.3% in the case of rice that was parboiled and dried with different fuels. Increasing the drying temperature from 40°C to 60°C increased PAH contamination by 64.39% (Bertinetti et al., 2018). As in the study realized by Escarrone et al. (2014), the predominant PAHs were naphthalene and phenanthrene.

PAH content in bread products

The occurrence and the PAH formation in the bakery chain, from raw materials to finished products, have been studied by several authors (Ciecierska and Obiedziński, 2013; Rozentale et al., 2017).
Contamination of bread with PAHs can depend both on the contamination of the raw bakery materials (Ciecierska and Obiedziński, 2013), mainly flour, and on the baking process (Chawda et al., 2017; Karşi et al., 2021). An important issue is also the temperature of the heat treatment, taking into account its impact on the level of contamination of the bread.
The PAH content of bread made with wheat flour (Rozentale et al., 2017; Al-Rashdan et al., 2010; Kacmaz, 2016b), rye flour (Ciecierska and Obiedzinski, 2013; Rozentale et al., 2017), and wholemeal rye flour (Ciecierska and Obiedzinski, 2013) commercialized in different countries is synthesized in Table 2. Results showed that the contamination with PAHs of breads was influenced by the flour used, but also by the country of production and assortment.
Rascón et al. (2018) studied the contamination level with the EPA's 16 PAH content of breads (breadcrumbs, breadsticks, wheat cakes and flat bread) marketed in Spain and determined a content ranging between 1.55 and 8.77 μg/kg. Samples were characterized by a high content of anthracene of 0.07-4.8 μg/kg. From the 4 priority PAH only ChR was found in bread.
Additionally, the highest percentage of 4 HAPs for wheat and rye breads sold on the Latvian market, was represented by ChR, with a content ranging between 0.108-0.611 μg/kg (Rozentale et al., 2017). The mean concentration of BaP in bread made with wheat flour was 0.064 μg/kg, while the level of 4 PAHs was 0.49 μg/kg. Among the analysed samples marketed in Latvia, rye bread was the most contaminated with PAHs, with an average content of 0.084 μg/kg for BaP, and 0.71 μg/kg for 4 PAHs, respectively. A total of 14% of the analysed bread samples from Latvia (35) exceeded the 4 PAH level of 1 μg/kg settled by the EU regulation (Rozentale et al., 2017).
The contamination with PAH was also investigated for bread marketed in Kuwait (Al-Rashdan et al., 2010). The content of 16 PAHs in bread sold in this country varied between 1.06 and 287.5 μg/kg, higher than the ones reported in Spain (Rascón et al., 2018), Latvia (Rozentale et al., 2017), and Iran (Kamalabdai et al., 2019) for the wheat bread. Among the 16 analysed bread samples from Kuwait, 5 samples exceeded the BaP level (5.57-16.5 μg/kg) imposed by the European legislation;

for the rest of the samples BaP was not detected. Ciecierska and Obiedzinski (2013) determined the amount of 19 PAH in raw materials, doughs and breads baked at different conditions (temperature 230-260°C, time 40 min).

Table 2. PAH content of bread commercialized in different regions

Bread	Country	BaP	BaA	ChR	BbF	Other PAHs in high proportion, µg/kg	4 PAH, µg/kg	Sum PAH, µg/kg	Reference
		µg/kg							
Bread (breadcrumb, breadsticks, wheat cakes and flat bread)	Spain	-	-	- ÷ 0.67±0.04	-	0.07±0.004 ÷ 4.8±0.3 ANT - ÷ 3.2±0.2 FLU	- ÷ 0.67	1.55÷8.77 (16)	Rascón et al., 2018
Commercial bread	Spain	-	-	- ÷ 0.48±0.03	-	0.041±0.002 ÷ 0.59±0.03 NAP 0.42±0.02 ÷ 2.4±0.1 ANT	- ÷ 0.48	1.29÷4.8 (16)	Rascón et al., 2018
Commercial bread home-toasted	Spain	- ÷ 0.98±0.05	-	0.24±0.01 ÷ 1.8±0.1	-	1.5±0.1 ÷ 3.8±0.2 NAP 0.97±0.05 ÷ 7.7±0.4 ANT	0.24 ÷ 3.45	10.31÷17.82 (16)	Rascón et al., 2018
White bread	Kuwait	nd÷16.5	nd÷2.31	nd÷1.53	nd÷7.27	*	*	1.06÷287.5 (16)	Al-Rashdan et al., 2010
Sandwich white bread	Kuwait	nd÷9.04	nd÷6.22	nd÷4.19	nd÷5.58	*	*	28.3÷149.3 (16)	Al-Rashdan et al., 2010
Bread	Belgium	<0.03÷0.20	*	*	*	*	0.11÷0.22	*	Kacmaz et al., 2016a
Bread	Turkey	0.17±0.05	0.03±0.02	0.05±0.03	0.05±0.02	*	0.28±0.09	*	Kacmaz, 2016b
Wheat bread	Latvia	0.064	0.094	0.202	0.133	*	0.49	*	Rozentale et al., 2017
Rye bread	Latvia	0.084	0.178	0.279	0.166	*	0.71	*	Rozentale et al., 2017
Wheat-rye bread baked at different temp	Poland	nd	0.05÷0.09	nd÷0.23	nd÷0.15	0.65÷2.28 PHN 0.49÷1.96 FLT	*	1.59÷7.37 (19)	Ciercierska, Obiedziński, 2013
Rye bread baked at different temp	Poland	nd	0.07÷0.18	0.15÷0.27	nd	2.57÷5.99 PHN 1.58÷3.21 FLT	*	5.62÷13.04 (19)	Ciercierska, Obiedziński, 2013
Wholemeal rye bread baked at different temp	Poland	nd÷0.24	0.06÷0.29	0.15÷0.53	nd÷0.23	1.67÷5.65 PHN 0.49÷3.62 FLT	*	2.67÷13.55 (19)	Ciercierska, Obiedziński, 2013
Brown bread	Kuwait	nd÷11.1	nd÷1.79	nd÷0.7	nd÷3.32	*	*	29.2÷56.4 (16)	Al-Rashdan et al., 2010
Senan bread-industrial	Iran	nd	-	-	-	*	*	*	Eslamizad et al., 2016
Sangak bread-traditional	Iran	1.97	-	-	-	*	*	*	Eslamizad et al., 2016
Sangak bread	Iran	-	-	-	-	1.7± - PYR 0.59±0.31 ANT 0.40±0.37 FLT	*	2.25±2.05 (8)	Khaniki et al., 2021
Sangak bread	Iran	-	-	-	-	5.57±5.21 PHN 14.59±5.52 ANT	*	10.08±6.38 (10)	Peiravian et al., 2021
Lavash bread-traditional	Iran	nd	nd÷2.12 (0.14)	nd÷2.58 (0.17)	nd÷2.58 (0.33)	50.10 NAP; 4.50 ACE; 0.68 PHN	*	64.87 (16)	Kamalabdai et al., 2019
Taftoon bread-traditional	Iran	nd	nd÷9.20	nd÷11.22	nd	60.62 NAP; 1.76 PHN; 1.73 ANT	*	68.97 (16)	Kamalabdai et al., 2019
Baguette - industrial	Iran	nd	nd÷5.15	nd÷7.16	nd	61.51 NAP 4.48 PYR 3.35 ACE	*	118.66 (16)	Kamalabdai et al., 2019
Taftoon bread	Iran	-	-	-	-	0.88±1.1 FLT 0.59±0.34 ANT 0.54±0.17 PYR	*	1.15±1.03 (8)	Khaniki et al., 2021
Barbari bread	Iran	-	-	-	-	1.06±1.15 FLT 0.99±0.75 PYR 0.62±0.25 ANT	*	2.64±2.15 (8)	Khaniki et al., 2021
Lavash bread	Iran	-	-	-	-	0.9±- PYR 0.73±0.3 ANT 0.48±0.24 ACY	*	2.25±1.67 (8)	Khaniki et al., 2021
Industrial bread	Iran	-	-	-	-	1.23± - PYR 0.43±0.17 ANT 0.35±0.18 NAP	*	1.66±1.49 (8)	Khaniki et al., 2021
Tandoor bread	India	1.18÷17.41	4.12÷14.51	0.24÷ 12.08	0.52÷8.92	3.14÷51.88 PHN 5.22÷31.8 ANT	*	113.36÷211.19 (16)	Chawda et al., 2017

Bread	Country	BaP	BaA	ChR	BbF	Other PAHs in high proportion, µg/kg	4 PAH, µg/kg	Sum PAH, µg/kg	Reference
		µg/kg							
						15.52÷49.83 FLT			
Tawa bread	India	0.19÷2.62	0.44÷3.47	15.37÷ 20.09	0.18÷2.67	7.31÷11.15 PHN 0.35÷5.34 ACE 3.41÷10.17 FLT	*	59.64÷77.12 (16)	Chawda et al., 2017
Bread baked at household electricity	Turkey	nd**	0.02±0.004 **	0.013± 0.003**	nd**	7.63±1.85 ACE** 3.12±0.35 NAP**	*	2.68±3.33* *(16)	Karşi et al., 2023
Bread countryside firewood	Turkey	0.013± 0.014**	0.035±0.021 **	0.32±0.30 **	nd**	3.57±2.74 ACE* 2.58±1.24 NAP*	*	2.71±3.23* * (16)	Karşi et al., 2023
Bread-commercial firewood	Turkey	0.059± 0.052**	0.092± 0.040**	0.125± 0.034**	nd**	1.08±0.387 NAP* 1.68±0.358 FLR* 1.64±0.552 PHE*	*	4.40±1.52* *(16)	Karşi et al., 2023
Bread – commercial natural gas	Turkey	0.023±0. 020**	0.078± 0.045**	0.059± 0.021**	nd**	1.19±0.702 NAP* 1.31±0.255 FLR*	*	3.58±1.74** (16)	Karşi et al., 2023

BaP- benzo(a)pyrene, BaA- benzo(a)anthracene, ChR- chrysene, BkF- benzo(k)fluoranthene, NAP- naphthalene, PHN-phenanthrene, FLT- fluoranthene, ANT- anthracene, FLR- fluorene, PYR- pyrene, ACE- acenaphthene, ACY- acenaphthylene, nd- not detectable, "-" compound not found, *- compounds were not studied or reported, **- expressed in µg/kg d.w. PAH are presented as range or as mean, as reported in references. The number in parentheses indicates the number of compounds included in the reported sum.

For all types of bread obtained, the PAH content was higher after baking than in the dough, increasing as the baking temperature elevated, confirming the fact that PAHs are formed during heat treatment. Among all 19 PAHs determined, 4 low molecular weight PAHs (phenanthrene, fluorene, pyrene and anthracene), account for about 90% of the total PAHs. The 4 EU marker PAHs were also reported to be detected at low levels. BaP was detected only in the crust of wholemeal rye bread (0.24 µg/kg) baked at the highest temperature (260°C). Regardless of bread type and baking temperatures, the concentration of 19 PAHs in bread portions followed the trend: crumb < loaf of bread < crust. For the bread obtained from a mixture of wheat and rye flour, the content of 19 PAH varied between 1.59-7.37 µg/kg, for that obtained from rye between 5.62-13.04 µg/kg, and for that from whole rye flour it varied between 2.67-13.55 µg/kg (Ciecierska and Obiedzinski, 2013), showing that the contamination is dependent on the type of flour used.

Kacmaz et al. (2016a) studied the level of the 4 EU priority PAHs in bread samples traded on the Belgium market and determined an average content ranging between 0.11-0.22 µg/kg. BaP content ranges from below the LOQ (0.03 µg/kg) to 0.20 µg/kg. In another study realized by the same author, it was found that the total content of 4 PAHs in bread commercialized in Turkey varied between 0.16-0.46 µg/kg with an average of 0.28 µg/kg. The largest share in the sum of 4 PAHs is represented by BaP with an average content of 0.17 µg/kg (Kacmaz, 2016b).

The level of BaP in traditional Sangak bread traded in the Iranian market was determined by several authors (Eslamizad et al., 2016; Khaniki et al., 2021; Peiravian et al., 2021). The BaP content determined ranged between undetectable and 1.97 µg/kg. In addition to BaP, Peiravian et al. (2021) detected the presence of phenanthrene and anthracene in the analysed breads, while Khaniki et al. (2021) determined the presence of pyrene, anthracene and fluoranthene.

In the study realized by Eslamizad et al. (2016) it was also analysed the bread obtained under industrial conditions (Senan), and BaP was not detected in these samples. Also, Kamalabadi et al. (2019) determined the content of 16 PAHs in 117 assortments of traditional bread (62 Lavash and 55 Taftoon) and 52 assortments of industrial bread (baguette) sold in the Iranian market. Lavash bread is 2-3 mm thickness and baked at 332°C, 80 s, while Taftoon bread is 3-5 mm thickness and baked at 315°C, 2.5 min. The baguette, on the other hand, is 5-6 cm thickness and is baked at a lower temperature (245°C), for a longer time (14 min). The most abundant PAH detected was naphthalene (average 57.41 µg/kg). BaP content was below the LOD value (0.2 µg/kg). The bread obtained under industrial conditions presented the highest average content of 16 PAH (118.66 µg/kg) as a result of the greater thickness of the bread and the longer baking time compared to the other analysed breads. Taftoon bread had higher PAH content compared to Lavash bread due to longer baking time and baking temperature (Kamalabadi et al., 2019).

In another study carried out in Iran, four traditional bread assortments (Taftoon, Barbari, Lavash, and Sangak) and one industrial assortment sold on the local market were analysed from the point of view of the content of 8 PAHs (Khaniki et al., 2021). With the exception of Taftoon bread which had a PAH content of 1.15 µg/kg, the other assortments of traditional bread had a content of 2.25-2.64 µg/kg, higher than the one obtained for industrial bread (1.66 µg/kg). Contrary to the study of Kamalabadi et al. (2019), in general the level of PAHs was higher in traditional breads, compared to those obtained industrially. From the analysed bread, Barbari bread, which is a barley bread, had the highest PAH content.

In the assortments of breads commercialized in Iran, naphthalene, phenanthrene, fluoranthene, anthracene, acenaphthylene, and pyrene were also determined (Khaniki et al., 2021).

In addition to the grain harvesting region, the type of flour used, and the bread assortments specific to each country, the PAH content of these products is also influenced by the baking method, the type of oven and fuel used for burning.

The effect of baking method on PAH concentration in bread was reported by Chawda et al. (2017) who studied the influence of two ovens, with coal and with gas on the Tandoori and tawa breads, respectively, sold in the Indian market. The authors obtained a concentration of 16 PAHs ranging between 113.36-211.19 µg/kg in the first mode of baking for Tandoori bread, and for the second mode (tawa bread), the level was between 59.64-77.12 µg/kg. The level of 16 PAHs in bread baked with gas oven was about 2.5-3.5-fold lower than for bread baked with coal ovens. In both assortments of breads, 3- and 4-ring PAHs were predominant.

By using the gas as a heating source for the oven, bread presented a higher content of ChR than when the coal was used as a thermal agent. Additionally, Karşi et al. (2021) investigated the effect of different fuel ovens on the 16 EPA priority PAHs in bread samples baked in Turkey. The bread was made in 6 commercial bakeries using different oven fuels (2 using firewood and 4 of them using natural gas), 5 traditional countryside bakeries (firewood) and 2 ovens of household (electricity). PAHs with 2 to 4 rings were predominant in the bread samples. By using electric ovens, the lowest content of 16 HAP was obtained (2.68 ± 3.33 µg/kg). When wood-fired ovens were used, the content of 16 PAHs was higher for breads made in commercial bakeries (4.40 ± 1.52 µg/kg) compared to those made in traditional bakeries (2.71 ± 3.23 µg/kg). For the samples baked in natural gas ovens, a PAH content of 3.58 ± 1.74 µg/kg was obtained. In conclusion, for samples baked in wood-fired ovens (rural and commercial ovens) the level of 4 PAHs found was 2 to 10-fold higher than when ovens with natural gas or with electricity were used (Karşi et al., 2021).

Bread can be subjected to toasting, a common method by which slices of bread are heated before consumption, and like this being contaminated with PAHs. Rey-Salgueiro et al. (2008) studied the effect of sandwich bread toasting treatment on PAH levels using several conditions: direct toasting (with flame, coal-grill or gas oven) or indirect toasting (with electric oven). By using the electric oven and the toaster with gas the formation of PAHs in toasted bread was not reported, while when using the grill and flame the PAHs were detected. These differences may occur as a result of the deposition of PAHs from the smoke resulting from roasting. During electric oven toasting, the contamination with PAH can be the result of macronutrients pyrolysis (Rey-Salgueiro et al., 2008).

Also, Rascón et al. (2018) analysed the effect of toasting in an electric home toaster on the level of 16 PAHs in wholegrain, multiseed, black bread, sliced wheat bread, and white bread. BaP was determined only in samples subjected to the toasting treatment. For the non-toasted samples, the highest content of 16 PAH was determined for the black bread (4.8 µg/kg) and for whole grains breads (4.49 µg/kg). By subjecting the samples to heat treatment, the level of 16 PAH increased considerably being 3.7-, 5.61-, 2.71-, 7.99-, and 5.07-fold higher for wholegrain, multiseed, black bread, sliced bread and white bread, respectively, compared to untoasted bread. All samples presented naphthalene and anthracene in high

concentrations. By toasting, the level of 4 HAP exceeded the legal limit allowed.
Thus, it can be concluded that by subjecting bread samples to thermal treatments, the level of PAH contamination increases considerably, being influenced by temperature, time and type of fuel used.

PAH content in other cereal-based products
In addition to bread, other cereal products such as cookies, breakfast cereals, biscuits and pasta sold in different markets have been analysed for the PAH content and the reported results are presented in Table 3.
The PAH contamination of these samples is influenced by the raw materials used (Rascón et al., 2018), the ingredients added (Kacmaz, 2016b), and also by the technological process and the fuel used for the thermal treatment.
The content of 4 PAHs in breakfast cereal samples traded on the Turkish market varied between 0.07-0.87 µg/kg, with an average of 0.37 µg/kg (Kacmaz, 2016b). The breakfast cereals analysed in this study included major ingredients, either alone or in a mixture: rice, oats, bran and fruits, wheat, maize, and chocolate powder. The largest share in the sum of 4 PAHs was represented by BaP with an average concentration of 0.22 µg/kg. The breakfast cereals (granola, chocolate granola and milk-filled cereal) commercialized on the Spanish market, presented a higher content of BaP, ranging between 3.1-4.2 µg/kg (Rascón et al., 2018). This can be the result of the high temperatures used in their production, the fat presence and also the ingredients drying process. The content of 4 PAHs for these samples exceeded the limit of 1 µg/kg imposed by the Commission Regulation (EU) no. 835/2011.

Table 3. PAH content of processed cereals from different regions

Processed cereals	Country	BaP	BaA	ChR	BbF	Other PAHs in high proportion, µg/kg	4 PAH, µg/kg	Sum PAH, µg/kg	Reference
		µg/kg							
Breakfast cereals	Turkey	0.22±0.07	0.09±0.06	0.11±0.06	0.08±0.04	*	0.37± 0.26	*	Kacmaz, 2016b
Breakfast cereals	Spain	3.1±0.2 ÷ 4.2±0.2	-	- ÷1.9±0.1	-÷3.5±0.2	- ÷ 3.9±0.2 NAP 0.08±0.004 ÷ 3.3±0.2 ANT	- ÷ 7.7	3.5÷22.3 (16)	Rascón et al., 2018
Cookies	Spain	-	-	- ÷ 0.35±0.02	-	0.26±0.01÷0.72±0.04 NAP 1.4±0.1÷2.9±0.2 ANT	-	1.66÷3.97 (16)	Rascón et al., 2018
Short cake	Nigeria	6.8±10.6	8.3±16.7	6.8±13.3	36.7±86.6	36.35±88.9 NAP 74.8±187.0 ACY 53.4±95.7 ACE	-	303.9±237.7 (16)	Iwegbue et al., 2015
Digestive biscuits	Nigeria	1.1 ± 2.3	2.4±3.4	10.9±16.9	9.8±11.6	43.0±44.6 ACY 39.2±65.0 PHE 78.3±113.3 ANT	-	271.4±192.4 (16)	Iwegbue et al., 2015
Cookies	Nigeria	nd	1.8±1.6	14.2±27.1	70.1±124.9	47.5±33.6 ACY 54.5±88.1 ANT	-	219.6±212.7 (16)	Iwegbue et al., 2015
Shortbread	Nigeria	0.1±0.1	28.7±55.6	22.0±30.1	53.1±41.9	69.3±45.3 ANT 37.9±27.4 PYR	-	340.9±227.9 (16)	Iwegbue et al., 2015
Wafers	Nigeria	10.3±22.2	7.7±12.7	16.8±11.4	39.2±68.2	96.2±146.7 NAP 16.8±21.2 PYR	-	236.8±121.3 (16)	Iwegbue et al., 2015
Cream crackers	Nigeria	nd	0.4±0.5	2.9±3.7	3.3±4.4	49.9±57.4 ANT 105.5±116.2 ACY	-	276.9±275.1 (16)	Iwegbue et al., 2015
Pringles	Nigeria	14.2±18.8	2.0±1.9	17.5±24.1	3.9±3.2	61.2±92.3 NAP 82.1±53.6 ACY	-	224.7±90.1 (16)	Iwegbue et al., 2015
Pasta	Spain	-	-	- ÷ 0.92±0.06	-	0.018±0.001 ÷ 0.24±0.01 NAP 0.15±0.01 ÷ 0.99±0.05 ANT	- ÷ 0.9	0.168÷1.98 (16)	Rascón et al., 2018
Pasta/ noodles	Nigeria	357±580 ÷ 834±1050	- ÷ 1270±590	- ÷ 871±140	-÷ 1490±240**	- ÷ 1008±950 PHN	564±610 ÷ 5615±930 **	564±610 ÷ 7889±730	Charles et al., 2017
Pasta locally produced	Nigeria	0.2±0.0 ÷ 3.0±2.0	0.3±0.3 ÷ 7.0±1.0	0.1±0.0 ÷ 2.0±1.0	0.1±0.0 ÷ 2.0±3.0	nd – 300.0±20.0 ANT nd – 300.0±30.0 PHE	1.0±0.0 ÷ 10.0±0.0	9.0±2.0 ÷ 800.0±30.0	Ihedioha et al., 2019
Pasta imported	Nigeria	0.2±0.1 ÷ 0.7±0.3	nd ÷ 1.0±0.0	nd ÷ 0.3±0.0	0.2±0.1÷ 0.7±0.2	nd – 4.0±0.7 NAP	0.4±0.1 ÷ 2.0±0.0	2.0±3.0 ÷ 7.0±2.0	Ihedioha et al., 2019

BaP- benzo(a)pyrene, BaA- benzo(a)anthracene, ChR- chrysene, BkF- benzo(k)fluoranthene, NAP- naphthalene, PHN-phenanthrene, ANT- anthracene, ACE- acenaphthene, ACY- acenaphthylene, PYR- pyrene, **- sum of benzo(b,j,k) fluoranthene, sum of benzo(b,j,k)fluoranthene is used in calculation, nd- not detectable, "-" compound not found, *- compounds were not studied or reported. PAH are presented as range or as mean, as reported in references. The number in parentheses indicates the number of compounds included in the reported sum.

Cookies commercialized on the Spanish market presented a 16 PAH content ranging between 1.66 and 3.97 μg/kg (Rascón et al., 2018). These cookies presented naphthalene and anthracene in a high proportion (0.26-2.9 μg/kg).
The content of 16 PAHs in different assortments of biscuits and cookies, commercially available in the Nigerian market was studied by Iwegbue et al. (2015), determining a PAH content ranging between 18.4-880.3 μg/kg, with an average of 219.6 μg/kg. The authors stated that variations in PAH content in different assortments of biscuits can be the result of the raw materials, baking method, type of fuel used in baking and oven thermal conditions. Biscuit samples from China presented higher concentrations of 16 PAHs than those commercialized on the European and the United States markets, showing that the area of production influences the PAH content. In general, the biscuits presented PAHs in relation to the number of aromatic rings in the following order: 3-rings > 4-rings > 5-rings > 2-rings > 6-rings. Phenanthrene, fluoranthene, and pyrene, which are low molecular PAHs, tend to form during the baking process at temperatures above 220°C (Houessou et al., 2007). By increasing the baking temperature (260°C), a significant level of pyrene, ChR and BaA was formed.
Another cereal-based product, quite popular worldwide, where PAHs can form is pasta: noodles, spaghetti, macaroni, etc. Ihedioha et al. (2016) assessed the PAHs content in Nigerian and imported brands of pasta and found 16 PAHs ranging between 9-800 μg/kg and for BaP between 0.2-3 μg/kg in Nigerian brands, and a content much lower in imported brands, of 2-7 μg/kg, and 0.2-0.7 μg/kg, respectively. PAH content varied between pasta brands, but also within the same brand due to heat treatment applied (temperature, time), distance to the energy source, oxygen availability, fat content and type of fuel used.
When Charles et al. (2018) determined the concentrations of 16 PAHs in different brands of noodles available on the same market, very high concentration of 16 PAHs were found, ranging between 564-7.889 μg/kg, while the sum of carcinogenic PAHs was found to be 564-5.615 μg/kg. BaP was detected at a concentration of 357-834 μg/kg, demonstrating serious health problems, especially since this type of product is widely consumed by children.
In Europe, the contamination with 16 PAHs in different cereal based-products purchased from various supermarkets was studied by Rascón et al. (2018), and for pasta it was determined a much lower content (0.168-1.98 μg/kg) than the one reported in Nigeria (Ihedioha et al., 2016; Charles et al., 2018) but nevertheless, the content was higher than the limit imposed by European regulations. BaP was not detected in the analysed pasta.
For most cereal-derived products, the predominant PAHs were naphthalene, anthracene and phenanthrene.

CONCLUSIONS

This study can provide an overview of the degree of PAH contamination of cereals and cereal derived products, and the factors that influence the formation of these contaminants.
Results of the study showed that cereals tend to be contaminated with low molecular PAH, with 2 to 4 rings.
The level of contamination with PAHs of cereals is influenced by the degree of pollution of the area where it is cultivated, the harvest year and also the way of drying the grains.
When it comes to cereal derived products, the PAH content is dependent of the flour type used in the manufacturing recipe, the region of production and assortment specific to each country, the baking process which implies baking time and temperature, but also the type of fuel used as a thermal agent.
The PAH content is higher in bread than in the dough from which it was obtained, showing that during baking PAH contamination can occur. When using natural gas or electric ovens, the PAH content in bread samples was lower than when using wood or grill for baking it.
In cereal-based products such as cookies, biscuits, pasta, the content of PAH is affected by the raw materials and ingredients used, the fat content of samples, and the heat treatment applied.
The presence of these pollutant environment contaminants in cereals and cereal-based products should be monitored and measures should be taken when the maximum level of 1

μg/kg for BaP, and sum of BaP, BaA, ChR, and BfF in processed cereals and baby foods for infants and young children is exceeded.

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