

POLYCYCLIC AROMATIC HYDROCARBONS IN CHEESES AS A RESULT OF THE SMOKING PROCESS: A REVIEW

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Abstract

The presence of smoke when smoking cheeses can have negative effects on the health of consumers. Polycyclic Aromatic Hydrocarbons (PAH) result from pyrolysis processes as well as incomplete combustion without organic sources. The factors that influence the concentration of PAH in cheeses are: temperature, moisture content, type of wood used in the smoking process, origin of the raw material (cow's milk, sheep's milk). The main PAH compound that specifies their presence in food products is benzo[a]pyrene. The interest directed at them is due to the carcinogenic and mutagenic effects they have on human well-being.

Key words: cheese, contamination, pollution, polycyclic aromatic, toxicology.

INTRODUCTION

Accelerated urbanization and industrialization result in the production of various pollutants such as polycyclic aromatic hydrocarbons (Mojiri et al., 2019). In some European countries it is forbidden to smoke cheeses through smoke, due to the fact that smoking food can contain contaminants that have negative effects on human health. This type of contaminants are polycyclic aromatic hydrocarbons (Guillen et al., 1997). As a result of industrial activity, human actions, the incomplete combustion of organic materials or their pyrolysis process results in the formation of more than 200 chemical substances (Aygun et al., 2005; Ishizaki et al., 2010).

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds containing two or more aromatic rings, made up of carbon and hydrogen atoms, which are pale yellow, white or colorless compounds (Patel et al., 2020). Aromatic rings can be molecularly arranged linearly, angularly or in groups (Abdel-Shafy & Mansour, 2016). According to their molecular mass, PAH can be divided into two categories: low molecular mass (composed of two/three aromatic rings) and high molecular mass (composed of four/more aromatic rings). Contaminations in soil, water and air play an

important role because they have carcinogenic and mutagenic effects (Titato & Lancas, 2006). Depending on their molecular mass, the emission of PAHs is of two types: in the form of particles or gas (Lee & Vu, 2010). Regarding the structure of PAHs, they are divided as follows: alternating PAHs (consisting of 6 carbon rings) and non-alternating PAHs (consisting of 6 carbon rings and an additional ring composed of less than 6 carbon atoms), for example fluorine (Gupte et al., 2016). Following the studies carried out, it has been proven that it has a carcinogenic potential for human health, thus it constitutes a global problem.

MATERIALS AND METHODS

To present a database for this study we conducted a systematic search of scientific publications in the Google Scholar, ScienceDirect PubMed and Scopus databases. In the tables below are highlighted concentrations of PAH from different milk products (raw milk, commercial milk, pasteurised milk, smoked cheese), the minimum and maximum values of benzo[a]pyrene (BaP) in cheeses under the influence of different smoking conditions are also presented.

RESULTS AND DISCUSSIONS

Factors influencing PAH

PAH have the following characteristics: low solubility in water, high melting and boiling points, low vapor pressure. All these elements depend on their structure (Lee & Vu, 2010). They are the result of incomplete combustion of organic materials and are produced in various forms, such as in commercial processes or in car workshops (Suchanová et al., 2008).

Thermal methods have a strong influence on the development of polycyclic aromatic hydrocarbons. Thus, Howard & Fazio (1983) suggest that the main factors that influence the hydrocarbons in food are spread over several levels:

1. Heat treatment temperature;
2. The nature of the energy source and
3. Direct or indirect contact with the power source.

Aydinol & Ozcan (2013) mentioned that the smoking process at temperatures higher than 425°C lead to the formation of toxic PAHs, so they recommend that the furnace temperature should not be higher than 425°C.

Another category of factors that influence PAH production are: temperature, oxygen concentration, humidity, type of wood used or type of smoke generation (Guillen & Sopelana 2005; Howard & Fazio, 1980).

THE PRESENCE OF PAH IN CHEESE

Cheese is a food that contains a wide range of varieties, it can be characterized according to the nature of the raw material used (buffalo, cow, sheep, goat), the manufacturing process (the type of curd used, the addition or absence of molds, the ripening time). The smoking process is an owner's option. Smoked cheese exists at a European level in every country, with a variety of cheeses valued for their organoleptic characteristics (McIlveen & Vallely, 1996).

Hydrocarbon concentrations were analyzed in 7 categories of smoked cheeses (Guillén & Sopelana, 2004). The highest concentrations found are correlated with the hydrocarbons that have the lowest weights (naphthalene, acenaphthylene, fluorene, phenanthrene), as compounds increase, hydrocarbon levels decrease or tend to cease to exist.

In industrial cheese, the PAH concentration was lower (almost 10 times) compared to smoked cheese (Esposito et al., 2015). Suchanova et al. (2008) obtained similar results with different cheese smoking methods, in cow's cheese the most significant level of PAH was obtained. During the smoking of cheeses, accumulations of solid particles can form that have PAHs in their composition and are distributed on the surface of the cheese, a situation encountered in this case was with cow cheese, which had a high concentration of PAH, with values between 75 and 113 µg/kg (Suchanová et al., 2008) in compared to commercial smoked cheese which has a lower content, from 2.3 to 57 µg/kg represented the total values of 12 PAH (Pagliuca et al., 2003). Following the study by Gul et al. (2015) who evaluated the PAH content of smoked and non-smoked industrial cheeses, it was concluded that traditional smoked cheese contains higher amounts of PAH than industrial cheese. This statement is also supported by Pluta et al. (2020), who obtained low levels of PAH in industrial cheeses. Similar PAH results were found in smoked cheeses: naphthalene, acenaphthylene and phenanthrene (Polak et al., 2022), as well as in other studies, the structure with a low molecular mass was found (fluorene, acenaphthene and naphthalene) and high molecular mass compounds (benzo(gly)perylene, benzo(a)pyrene and benz(a)anthracene) (Guillén & Sopelana, 2004). At the same time, the most abundant PAHs are those containing 2, 3 or 4 aromatic rings (Guillén & Sopelana, 2004; Guillén et al., 2007). The PAH analysis of the cheeses from the point of view of the number of aromatic rings was grouped into several categories as follows: 2 or 3 aromatic rings (naphthalene, anthracene, acenaphthene, phenanthrene fluorene), 4 aromatic rings (chrysene, benz(a)anthracene, fluoranthene), 5 or 6 aromatic rings (dibenz(a,h)anthracene, benzo(b)fluoranthene, benzo[a]pyrene, benzo(k)fluoranthene, benzo(g,h,i)perylene, indeno[1,2,3-cd]pyrene (I[1,2,3-cd]P) (Suchanová et al., 2008).

Sheep milk whey contains the lowest number of hydrocarbons although it contains a higher amount of hydrocarbons compared to other samples. The highest concentration of hydrocarbons was found inside the cheese

(1037.23 µg/kg), in the middle and inside the contents were similar (153.70 and 88.96 µg/kg) (Guillén & Sopolana, 2004). Compounds with a mass greater than 228 (greater than benz(a)anthracene, chrysene or triphenylene) were found in cow, sheep and goat milk samples (Guillén & Sopolana, 2004).

The most contaminated part of the cheese with PAH is found on the surface layers of the cheese, where the amounts are 3 to 6 times more abundant than those of the entire sample. This observation is in agreement with that obtained by Guillen & Sopolana (2004). Removal of 1 to 2 mm of the surface layer reduces PAH concentration by 50-100% (Suchanová et al., 2008).

Table 1. Average PAH level in different dairy products

Product category	The amount of PAH	Reference
Raw milk	5.427 ng/g	Naccari et al. (2011)
Pasteurised milk	6.518 ng/g	Naccari et al. (2011)
UHT milk	5.941 ng/g	Naccari et al. (2011)
	(Semiskimmed)	
	7.752 ng/g	Naccari et al. (2011)
	(Whole milk)	
Commercial milk	0.99 lg/kg	Kishikawa et al. (2003)
Raw milk	1.010 lg/kg (from farm)	Abou-Arab et al. (2014)
	0.36 lg/kg (Commercial)	Abou-Arab et al. (2014)
Smoked cheese	175.43–1037.22 lg/kg	Guillen and Sopolana (2004)
Circassian cheese	19.57 lg/kg (Traditionally smoked)	Gul et al. (2015)
	0.77 lg/kg (Traditional nonsmoked)	Gul et al. (2015)
	6.36 lg/kg (Industrial smoked)	Gul et al. (2015)
	0.49 lg/kg (Industrial nonsmoked)	Gul et al. (2015)
Industrial smoked cheese	10.10 lg/kg	Suchanova et al. (2008)
Smoked cheese	242 lg/kg (smoked with almond shells)	Guillen et al. (2007)
	85 lg/kg (smoked with Dry prickly pear)	Guillen et al. (2007)
Cheese I	10.75 lg/kg	Martorell et al. (2010)
Cheese II	11.75 lg/kg	Martorell et al. (2010)
Cheese III	12.80 lg/kg	Martorell et al. (2010)

High concentrations of polycyclic aromatic hydrocarbons do not always contain a large number of polycyclic aromatic hydrocarbons. For example, the cow's milk cheese sample with a total concentration of 367.53 µg/kg had a content of 28 polycyclic aromatic hydrocarbons, compared to goat's milk cheese which had a total concentration of 194.65 µg/kg and 41 polycyclic aromatic hydrocarbons (Guillén & Sopolana, 2004). The minimum PAH content was detected in non-smoked cheese (1.83 µg/g) (Rawash et al., 2018).

Guillén & Sopolana (2004) used 4 different types of plant material to smoke the cheese: almond skins (*Prunus dulcis*), dry fiber (*Opuntia ficus indica*) and canary pine wood or needles (*Pinus canariensis*). PAH concentrations smoked with almond shells were higher (42.46-739.55 µg/kg) compared to those smoked with dry fiber (29.21-193.11 µg/kg). The presence of very light (naphthalene or acenaphthylene) or heavy (benz(a)anthracene, chrysene, benzopyrene or benzo(gy)perylene) PAH was noted.

Cancerogenic effects

Depending on the PAH compounds, the degree of cancer can be characterized as follows: from moderate to strong (benzo[a]pyrene, dibenz[a,h]anthracene, dibenz[a,j]anthracene, 3-methylcholanthrene and 5-methylchrysene), relatively weak or inactive (benzo[e]pyrene, benzo[c]phenanthrene, dibenz[a,c]anthracene, fluoranthene and chrysene) (Adeyeye, 2020). The most toxic PAH is Benzo(a)pyrene and in traditionally and industrially ripened cheeses the level of B[a]P was 0.69 and 0.25 µg/kg (Bukowska et al., 2022). The primary element that indicates the occurrence of PAH in food is benzo[a]pyrene, according to animal studies (EFSA, 2008).

PAH are based on 3 types of effect categories: mutagenic, teratogenic and carcinogenic (Jiang et al., 2014). The degree of toxicity increases as the ring structure increases (Hankin et al., 1996). Between the years 1980-2000, they began to study the adverse effects they have on organisms, such as the rat, daphnia (Sun et al., 2021). Since 2000, research on the immunotoxicity, neurotoxicity, genotoxic impact, reproductive and endocrine disrupting effects of PAH has started to be a new topic

(Barron, 2004). PAH exposure is of many types and their effects are correlated with exposure duration, health status, chemical or toxicity (Kim et al., 2013). Among the factors mentioned the most important factor is the duration of exposure (WHO, 1998). On the other hand, the inhalation of PAH and the effects on the cardiovascular system, gastrointestinal system, hematological and musculoskeletal effects are less common. Occupational exposure in an environment with high PAH concentrations in the short term has as symptoms nausea, vomiting, confusion and skin and eye irritation (Goudarzi et al., 2017b). People who have asthma can be affected by short-term exposure to PAH, and people with coronary heart disease are more likely to get thrombosis (Kim et al., 2013). More severe health conditions such as skin, lung or digestive tract cancer occur as a result of repeated exposure to PAH over a longer period of time (Bach et al., 2003; Olsson et al., 2010). Exposure to low doses of PAH causes eye irritation that can result in cataracts (Mumtaz & George, 1995).

Influence of smoke from PAH

In general, smoke conditions can influence PAH levels obtained from smoking and smoked foods (Maga, 1988). One element that reflects the existence of PAH compounds is benzo[a]pyrene contamination demonstrating the existence of a link to smoke generation conditions (Toth & Potthast, 1984). Parameters that influence the production of PAHs during the smoking period are: the type of wood used, the type of smoke generation, the percentage of humidity, the temperature and the oxygen concentration (Howard & Fazio, 1980).

The most used method of preventing the spoilage process in the range of food products is smoking, but nowadays, food smoking is mainly done for the sensory qualities and not for the preservation effect (Bratzler et al., 1969). Smoke performs bacteriostatic and antioxidant roles, imparts flavor and color to high protein foods (Burt, 1988). Cheese can be smoked using two methods: smoking with natural smoke or smoking with liquid smoke. For meat and fish, liquid smoke is used as a preservative and flavoring agent (Hatulla & Luoma, 2001). For example Wendorff et al.,

(1993) state that phenolic compounds in smoke can prevent mold growth in smoked Cheddar cheese. One way to diversify the flavors is to smoke the cheese. Riha and Wendorff (1993) studied what is the perception of consumers on the choice of the type of cheese and later it was concluded that the most important characteristic of the cheese is the color. In a consumer survey, Hendrick et al. (1960) found that hardwood smoke flavored cheeses were preferred over liquid smoke cheeses.

During the baking of Cheddar cheese under the influence of smoke, the moisture percentage was located between 35.50-37.06. Usually after 3 months of ripening the moisture level increases in smoked cheeses (Rehman et al., 2003).

The results of the sample from a type of processed (industrial) cheese in which a liquid smoke flavor was used, the PAH content found had high values ($181 \mu\text{g kg}^{-1}$), respectively ($9.8 \mu\text{g kg}^{-1}$) of the carcinogenic ones (Suchanová et al., 2008). Gilbert & Knowles (1975) state that natural smoking provides a higher degree of preservation than liquid smoking. Benzo[a]pyrene develops during smoking, so as an alternative the method of liquid smoking began to be used to reduce the carcinogenic effect of the compounds. To reduce the risk of PAH formation, it is recommended that the combustion temperature be lower than 300°C (Tulay et al., 2011).

The influence of the type of wood on the PAH content

The elements that generate smoke production are: air supply, combustion temperature, wood moisture (Guillén et al., 2000). PAH content is lower in wet wood compared to dry wood because the temperature at which smoke is produced from wet wood is low (Maga, 1988). Wood moisture has effects on smoke production, for example a low moisture level results in incomplete combustion of the wood, as opposed to a high moisture level that decreases the combustion performance and increases the appearance of smoke, so to reduce the released particles it is recommended a moisture concentration between 15 and 30% (Simoneit et al., 2000).

That is why a low air supply during pyrolysis reduces PAH production (Guillén et al., 2000).

The production of PAHs is influenced by the type of wood used, for example specific resinous (conifers: firs or pines) have positive effects on the development of resin, which is why they can contribute to the decomposition of crisis (Ch), benzo[a]-pyrene (BaP), benzo[a]anthracene (BaA)(Simoneit, 2002) benzo[k]fluoranthene (BkF), ndeno(1,2,3-cd)pyrene (IP), or benzo[k]fluoranthene (BkF) (Simoneit et al., 2000), compared to hardwood smoke which contains reduced amounts of Ch, BaA and BaP (Oros & Simoneit, 2001).

Maga, (1988) proposed the use of hardwoods to reduce the PAH content of smoked products in exchange for the use of resinous lemmas.

The results of the study by Guillen et al., (2000) illustrate that benzo[a]pyrene, naphthalene, pyrene and phenanthrene were identified. Low molecular weight compounds contain the highest concentrations of PAH. There are differences in the content of PAH and the origin of the raw material, for example beech, oak and cherry shoots contain low amounts of PAHs, in contrast to vine shoots where high concentrations of PAHs are found (due to the high content of methyl derivatives and naphthalene). Benzo[a]pyrene content was high in cheeses smoked with spruce, pine, willow, alder or beech wood (Migdał et al., 2020).

Cherry, beech and oak contain lower amounts of PAHs than vine shoots. Poplar, although a hardwood or softwood, produces higher amounts of PAHs compared to the 4 types of wood previously mentioned (Pallu, 1971). To obtain smoke flavors with low contents of carcinogenic PAH. At temperatures between 270°C and 380°C significant amounts of smoke are produced, and above 500°C the smoke production increases (Migdał et al., 2020).

Among the smoke flavors analyzed, the wood that recorded the highest naphthalene and methyl derivative contents was vine shoots (Guillen et al., 2000).

The analyzed studies demonstrated that one of the most important elements that have an impact on the PAH content is the raw material, namely the wood used in the smoking process. The highest PAH values were detected in cheeses smoked with softwoods such as conifers compared to hardwoods. At the same time, traditional smoked cheeses contain higher

amounts of PAHs than industrially smoked cheeses.

Table 2. B[a]P concentration in cheeses

B[a]P content	The method used	Reference
0.011–0.87	Commercially and home-made smoked cheeses	Suchanová et al. (2008)
ND–0.54	Rind of commercial smoked cheeses	Guillén & Sopolana (2004)
<0.11–3.7	Home-made smoked cheeses	Michalski & GERMUSKA (2003)
<0.22–1.65	Effect of different smoking conditions on B[a]P	Michalski & GERMUSKA (2003)
0.04–0.38	Effect of different smoking conditions incl. smoke-flavoured and liquid flavoured cheeses on PAHs	Pagliuca et al. (2003)
ND–0.90	Commercially smoked cheeses	Garcia et al. (1996)
0.15–0.74	Investigation of different smoking conditions on B[a]P	Anastasio et al. (2004)
<0.15–4.1	Investigation of different smoking conditions on PAH	Bosset et al. (1998)

ND*= not detected

CONCLUSIONS

PAH in cheeses present a mutagenic and carcinogenic factor on the health of consumers, which is why their control is necessary to reduce the negative effects they can have. To ensure a high level of preservation of the cheeses, natural smoking is recommended. It is advisable to use hardwoods instead of softwoods to reduce the PAH content in the smoke. One way to reduce the carcinogenic effects of PAH compounds is the liquid fumigation method.

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