Review article
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REVIEW OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN SMOKED ANIMAL ORIGIN FOOD: A SERBIAN PERSPECTIVE

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Abstract

This review article presents various aspects of polycyclic aromatic hydrocarbons (PAH) found in smoked animal-derived foods. The article covers PAHs and smoking methods, their levels and origins, regulatory aspects, existing knowledge gaps, as well as their impact on human health and future prospects concerning smoked animal-derived foods from the Republic of Serbia. Given our tradition and the widespread appreciation for smoked meat among consumers, producers are increasingly incorporating smoking into the production process of various animal and non-animal food products to encourage consumption diversification. Over the past ten years, our research has included smoking in traditional and industrial conditions. So far, we have examined the presence of PAHs in meat products, fish products and honey. Research on PAHs in grilled and smoked cheese is underway, after which we the results of the presence of PAH compounds in all types of smoked animal origin food will be available. Our findings revealed the presence of PAHs in the majority of studies conducted in the Republic of Serbia. Therefore, it is crucial to maintain continuous monitoring of their levels. Additionally, there is a need to develop models and implement solu-

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tions aimed at reducing PAHs in animal-derived foods intended for human consumption.

Key words: smoking, food production, smoked meat, smoked fish

PREGLED SADRŽAJA POLICIKLIČNIH AROMATIČNIH UGLJOVODONIKA U DIMLJENIM PROIZVODIMA ŽIVOTINJSKOG POREKLA: PERSPEKTIVA U SRBIJI

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Kratak sadržaj

U ovom preglednom radu predstavljeni su mnogi aspekti prisustva policikličnih aromatičnih ugljovodonika u dimljenoj hrani životinjskog porekla. Obrađeni su najznačajniji PAH-ovi, vrste dimljenja, njihov sadržaj i izvori, regulativa, nedostaci u sadašnjim saznanjima, uticaj PAH-ova na zdravlje ljudi i buduća stanovišta vezana za dimljenu hranu životinjskog porekla iz Republike Srbije. Poznavajući našu tradiciju i prihvaćenost dimljenog mesa među potrošačima, naši proizvođači sve više koriste dimljenje u cilju povećanja potrošnje ostalih vrsta hrane životinjskog i ne životinjskog porekla. Tokom prethodnih deset godina, naša istraživanja su obuhvatala dimljenje u tradicionalnim i industrijskim uslovima. Do sada smo ispitivali prisustvo PAH-ova u mesnim proizvodima, ribljim proizvodima i medu. U toku su istraživanja PAH-ova u pečenom i dimljenom siru, nakon čega bismo imali rezultate prisustva PAH jedinjenja u svim vrstama dimljene hrane životinjskog porekla. Rezultati istraživanja su pokazali da su PAH-ovi prisutni u većini studija sprovedenih u Republici Srbiji, zbog čega ih treba kontinuirano pratiti i raditi na modelima i rešenjima koji dovode do

njihove redukcije u namirnicama životinjskog porekla koje su namenjene za ishranu ljudi.

Ključne reči: dimljenje, proizvodnja hrane, dimljeno meso, dimljena riba

INTRODUCTION

Smoked products, and smoked meat in particular, hold a prominent place in the diets of people in the Republic of Serbia and the wider Balkan region due to their popularity. Smoked food, such as smoked meat and smoked fish products are very popular and consumed quite often in our region (Babić et al., 2017; Novakov et al., 2022).

With a deep-rooted tradition and high consumer acceptance of smoked meat, producers are leveraging smoking techniques more frequently in the food production process. Smoked meat and meat products, smoked fish and fish products, as well as smoked cheese are available on the Serbian market.

Although fish consumption in the Republic of Serbia is comparatively lower than in EU countries (FAO, 2022; FAO, 2024), fish and fish products are significant in the Serbian diet, particularly during the Christmas and Easter fasting periods. The taste of smoked carp, as one the mostly common fish species in Serbian market, resembles the taste of other traditional smoked products and therefore, affects the increase of the fish consumption (Babić et al., 2018a). Consumption of smoked fish has significantly increased over the past few decades in many European countries including the Republic of Serbia (Tešić et al., 2013; FAO, 2022; FAO, 2024), due to their receptive sensory properties, high nutritional value which includes polyunsaturated fatty acids, minerals, vitamins and proteins favorable content (Bansal and Kim, 2015; Bongiorno et al., 2015). Lately, the growing consumer dissatisfaction with industrially produced food and the growing interest in nonconventional products urges producers to look for other alternatives - traditional and authentic food, such as smoked food.

Smoking is considered as one of the oldest procedures for preserving food (Djinovic et al., 2008), but nowadays, smoking primarily serves to impart the characteristic color, aroma, flavor, and appearance to smoked food products. (Fasano et al., 2016). During the pyrolysis of organic compounds, many chemical contaminants including polycyclic aromatic hydrocarbons (PAHs) are formed (Sun et al., 2019). PAHs are a large group of ubiquitous, persistent or-

ganic pollutants consisting of two or more fused aromatic rings in their chemical structural (Babić et al., 2018b). PAHs originate from two main sources: natural (biogenic and geochemical) and anthropogenic (Yebra-Pimentel et al., 2015). Naturally, PAHs occur in fossil fuels, but are also formed during the incomplete combustion of organic materials such as sawdust, charcoal, diesel, wood and vegetation. Most of the PAHs have been proved to have teratogenicity, carcinogenicity and mutagenicity, posing a great threat to human health. Benzo(a)pyrene, as the most common compound in smoke, is metabolized in the human body into benzopyrene diol epoxide, which is then covalently bound to cellular macromolecules, including DNA. In addition, PAHs interferes with normal cell function, disrupting the cell membrane and enzyme system (ATSDR, 2012; Bogdanović et al., 2019). Many studies have shown that the major source of exposure to PAHs is through food consumption (Bansal and Kim 2015; Domingo and Nadal 2015; Yebra-Pimentel et al., 2015). EFSA (2008) estimated the dietary exposure for the sum of PAH8 (benz(a)anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene, dibenz(a,h)anthracene (DhA) and benzo(ghi)perylene (BgP)) as 1.73 mg/day per person.

The main goal of this study is to highlight PAHs as very important chemical contaminants in smoked products of animal origin, to present the national and European legislation related to the present issue and give an overview of the level of contamination of smoked meat and fish products found on the Serbian market.

PAHs IN SMOKED PRODUCT OF ANIMAL ORIGIN ON THE SERBIAN MARKET

The content of PAHs in smoked meat and meat products on the Serbian market has been examined in several studies. Djinovic et al. (2008) investigated the levels of PAHs in beef ham, pork ham, bacon without skin, bacon with skin, "*čajna*" sausage and "*Sremska*" sausage originating from Serbia, produced by meat industry Zlatiborac. The tested products did not exceed the maximum permitted limits. Škrbić et al. (2014) investigated PAH levels in smoked dry fermented sausage "*Petrovská klobása*" from Serbia. The highest total concentration of 16 PAHs was found in the samples smoked in traditional smokehouse, being almost 15 times higher than the total 16 PAHs content in sausages smoked in industrial smokehouse. The most abundant PAH was phenanthrene. PAH4 was below the maximum allowed legislation levels. In a study conducted by Kartalović et al. (2015) the content of PAH4 in household-

produced smoked meat were analyzed. Ham produced in traditional drying facilities and smoking cabinets in Serbia contained 11.51 μg/kg of total PAHs. Kartalović et al. (2022) analyzed the content of PAHs in traditionally smoked and dry-cured bacon "*Slavonska slanina*" and determined that it was safe for consumption. PAH4 were determined as follows: 14.84 μg/kg in the middle, 10.60 μg/kg on the surface and 17.37 μg/kg in the skin in traditional smoked meat samples, while the PAH4 content in the samples smoked in industrial conditions were below the level of quantification. Škaljac et al. (2022) found benzo[a]pyrene and PAH4 were below the limits of detection in all analyzed samples of "*Petrovská klobása*" smoked in industrial conditions. Vranešević et al. (2022) examined the content of PAHs and the possibilities of their reduction in traditional (homemade) smoked dry-cured pork loin produced in Serbia. Đinović-Stojanović et al. (2023) investigated benzo(a) pyrene and sum of PAH4 compounds levels in our types of dry fermented sausages collected from market in Belgrade, Serbia. They found that all analyzed samples PAH's levels were under limits regulated by Serbian and EU legislation. Škaljac et al. (2023) analyzed the total content of 16 PAHs in traditional dry fermented beef sausage from Serbia and found the content of BaP to be lower than the limit of detection, while contents of PAH4 were from ND to 7.42 μg/kg. Škaljac et al. (2024) examined the content of PAHs in dry cured meat products, bacon and dry fermented sausages smoked in six different chambers from the territory of Vojvodina. Benzo[a]pyrene and PAH4 were lower than the set maximum legislation values.

Smoked carp, trout and salmon are the most commonly consumed types of smoked fish in Serbia. Smoked mackerel, herring and sprats, usually canned, can also be found. Also, non-smoked fish, especially shellfish, originating from polluted areas or areas where oil has been spilled, should be checked for the PAHs presence. There are several studies regarding PAHs levels in fish produced or marketed in Serbia. Babić et al. (2018a, 2018b) analyzed PAH levels in smoked common carp meat in industrial and traditional conditions with and without filters and found that the meat was safe for consumers in accordance with European and national legislation. Novakov et al. (2017) determined the total PAHs levels of 17.67 μg/kg in canned tuna, 15.12 μg/kg in canned sardines and 57.19 μg/kg in smoked sprouts. In canned smoked sprouts benzo(a) pyrene concentration reached 8.29 μg/kg, which was above the legislation limit. Novakov et al. (2021) analyzed mussels from the Serbian market and found PAHs levels to be under the legal limits.

In addition to meat and fish, PAHs can also be found in other products of animal origin. Thus, Petrović et al. (2021) analyzed pesticides, antimicrobial

drugs and PAH concentrations in honey from Serbia and found that 6.6% of polyfloral honey samples contained high PAH8 (58.9 -656.2 ng/g) concentrations.

INFLUENCE OF DIFFERENT FACTORS AFFECTING PAH LEVELS IN FOOD

Numerous factors influence the PAH content in food, making it a focal point of a majority of research studies in this field. Some investigations are focused on the differences in PAH content in traditional and industrial smoking, some on the distance from the fire to the product, some on the filters that can absorb harmful substances from the smoke (Babić et al., 2018b). There are also differences in PAHs content when different types of casings are used (Mastanjević et al., 2020). There are also studies that have shown that plastic packaging can absorb a certain amount of PAHs during product storage (Semanova et al., 2016).

Traditional and especially homemade traditional manufacturing is not standardized and is uncontrolled. PAH4 level in samples of "*Hercegovačka pečenica*" subjected to traditional smoking highly exceeded maximum limits set by the EU Regulation (12 μg/kg) by up to 10 times (Puljić et al., 2019). Pancetta, dry pork neck ("*buđola*"), pork tenderloin and "*Slavonska kobasica*" were smoked in traditional smokehouse and in an industrial chamber to see variations and influence of different smoking type on chemical contaminants content (Kartalović et al., 2020; Mastanjević et al., 2020). "*Slavonska kobasica*" contained BaP above 5 μg/kg in 4 samples and PAH4 level above 30 μg/kg in 3 samples, meaning they were not in accordance with EU regulation (Mastanjević et al., 2020). There are ways to apply traditional smoking but with the use of certain filters. In the investigations conducted by Babić et al. (2018a, 2018b) samples of common carp meat were smoked in traditional conditions using different filters. The zeolite filter appeared to be the best one solution through traditional production of smoked common carp meat. The casing type, especially for sausages is also an important way of the PAHs reduction. Mastanjević et al. (2020) found that higher total content of the 16 PAHs in sausages stuffed in natural casings than those in collagen casings. Semanova et al. (2016) proved the migration of PAHs from smoked sausages to LDPE, where the total PAH4 content decreased from 30.1 to 5.7 μg/kg, during 180 min of interaction with packaging material, proving that packaging type and length of storage also may have an important impact on PAH reduction. Direct smoking when a kiln is placed directly below food during the drying process is

still the most commonly used technique. Homemade smokehouses are mostly handmade according to the needs and capabilities of the producer. Traditional smoked meat products are not under monitoring and there is no strict regulation of product variables (type of raw materials, additives, smoking conditions, relative humidity etc.). The production processes (smoking, drying and ripening) of the homemade traditional smoked products takes from one to three months. One of the most important problems in traditional smoking in households is the deposition of char particles and more volatile compounds on the surface of meat products, which results in penetration from the surface area toward inner part of smoked meat products during the storage period (Kartalović et al., 2022). The amount of PAH4 in smoked samples subjected to traditional smoking highly exceeded maximum limits set by the EU by up to 10 times (Puljić et al., 2019). The consumption of this kind of smoked products can be potentially harmful to consumers' health and that is the reason why the "as low as reasonably achievable" (ALARA) principle is in force in all EU countries. On the other hand, the amounts of the PAH4 in all smoked samples subjected to industrial smoking processes were below the limit of quantification (Puljić et al., 2019). PAH reduction in smoked products can also be affected by the distance between the fire and the product. Dry fermented beef sausages smoked in traditional condition at distance of 2 m from the smoke sours (kiln) had significantly higher contents of 16 PAHs and PAH4 than the sausages smoked at distance of 4–5 m (Škaljac et al., 2023). When errors in the smoking process occur, the rinsing method should be considered in homemade manufacturing, in order to obtain safe products (Vranešević et al., 2022).

REGULATIONS

The monitoring of PAHs has been strongly recommended by the European Union, as well as regulations by the Republic of Serbia. BaP and PAH4 (BaP, Chr, BaA and BbF) were chosen as a marker for the occurrence and effects of carcinogenic PAHs in food. Maximum level for BaP is 2 μg/kg and for the sum of PAH4 is 12 μg/kg (Commission Regulation (EU) No 915/2023; Official Gazette of the Republic of Serbia, 81/2019, 126/2020, 90/2021, 118/2021 and 127/2022).

Despite the application of GHP and GMP and HACCP to the possible extent, the current maximum levels for PAHs are not achievable in several EU Member States in certain traditionally smoked meats and meat products and traditionally smoked fish and fish products, where smoking practices cannot be altered without changing significantly the organoleptic characteristics of

the food. In order not to close many small and medium size enterprises in Croatia, Ireland, Spain, Poland, Latvia, Cyprus, Sweden, Finland, Portugal and Slovakia, a derogation for local production and consumption is maintained without a time limit for certain traditionally smoked products (meat, meat products, fish and fish products) only in those Member States, namely 5 μg/kg for BaP and 30 μg/kg for PAH4 (Commission Regulation (EU) No 915/2023).

Allowed levels of BaP and PAH4 for muscle meat of most smoked fish and smoked fish products including smoked crustaceans, crabs and crab-like crustaceans are 2 and 12.0 μg/kg, respectively. Similar to European Union legislation for meat products, the EU (Commission Regulation (EU) No1255/2020) allowed Latvia, Finland and Sweden to place some traditional fish and fishery products, smoked and market in their territory, with the BaP content level lower than 5.0 μg/kg and PAH4 level lower than 30 μg/kg.

CONCLUSION

There are many techniques for quantification of PAHs. Gas chromatography-mass spectrometry (GC-MS), which has limited level of LOQ, is most commonly used, including studies that we have conducted. Our activities today are directed towards the improvement of knowledge of the sensitive detection methods for PAHs in each particular smoked food type which would considerably help consumers to avoid the risk associated with PAHs consumption. In order to decrease the level of PAHs and reduce the risk of PAHs occurrence in smoked meat products, traditional producers should be educated how to use the improved/novel smoking techniques and adjust the smoking parameters. This should result in safer smoked meat products. The results of conducted studies call for a need to monitor PAHs in traditional smoked products. In addition, likely revision of the existing rulebook for traditional smoked products might also be necessary.

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Author's contribution

J.V., N.N. and S.V.K. made contributions to the idea of the publication, organisation of work and writing the manuscript. J.V., N.N., N.P. and S.V.K. were

involved in the writing of the manuscript. K.M. reviewed the manuscript. S.Š. and B.K. gave the final approval of the manuscript to be published.

Competing interest

The authors declare that they have no competing interests.

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