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Content of Polycyclic Aromatic Hydrocarbons in Traditionally Smoked Meat Products from North Serbia (Vojvodina)

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Abstract: This study examined the safety of meat products from north Serbia (Vojvodina), smoked in traditional conditions, from a PAH point of view, and assessed the possibility of their reduction in these types of products. Samples of dry cured meat products, bacons and dry fermented sausages smoked in six different chambers on the territory of Vojvodina were examined. The contents of 16 polycyclic aromatic hydrocarbons, from the United States Environmental Protection Agency list (16 US-EPA PAHs), and sensory quality of meat products were determined. The total content of 16 US-EPA PAHs in dry cured meat products was in the range from 99.73 µg/kg to 412.76 µg/kg; in bacons it was in the range from 36.43 µg/kg to 188.86 µg/kg; and in dry fermented sausages in the range from 47.23 µg/kg to 270.60 µg/kg. The lowest contents of 16 US-EPA PAHs compounds were determined in meat products smoked in traditional conditions during 3–5 days (3–4 h per day) at a distance of 2.5 m between the fire and products. Generally, it can be concluded that shortening of smoking process is justified, because products of good sensory quality and with decreased content of PAHs compounds were obtained. Benzo[a]pyrene, whose maximum allowed content in smoked meat products is 2 µg/kg, was below the limit of detection in all examined traditional meat products from Vojvodina. Also, contents of PAH4, sum of benz[a]anthracene, chrysene, benzo[a]pyrene and benzo[b]fluoranthene, were in the range from ND to 2.22 µg/kg, still greatly lower than the set maximum value. These results indicated the safety of dry cured meat products, bacons and dry fermented sausages from the territory of north Serbia (Vojvodina), as defined by EU Regulation 2023/915 criteria for PAHs contents.

Keywords: traditional meat products; safety; PAHs; sensory quality

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1. Introduction

Smoked meat products are very popular in human diet and have a significant economic value for the meat industry. About 40–60% of the total meat products are smoked. In the smoking process, volatile components, produced by thermal degradation of wood, penetrate into meat and meat products, with the aim of prolonging shelf life and contributing to the formation of the specific aroma, taste and colour of final products [1–3].

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds that may have harmful effects on human health, which can cause DNA damage or mutation and cancerous processes in the human body. They are formed by incomplete combustion of organic matter and are transferred to meat products during smoking. Their content can vary, depending on the method and conditions of smoking [2,4–6].

The European Commission has set maximum limits for benzo[a]pyrene—BaP (<2 μg/kg) and PAH4 (benz[a]anthracene, chrysene, benz[b]fluoranthene and benzo[a]pyrene) in smoked meat products (<12 μg/kg), which Serbian regulations are aligned with. However, since 2020, some EU member states allow higher limits for both BaP (<5 μg/kg) and PAH4 (<30 μg/kg) [7] in traditional smoked meat products. Reports that warn about the possibility of higher PAH content in meat products (especially in traditional ones) stimulated the interest of the meat industry and scientific institutions, encouraging them to examine the PAHs in smoked meat products and the conditions during production processes that can reduce their contents [8–13]. Determination of the PAH content in meat products is very important, especially when the smoking process is more intense, the smoking conditions are minimally controlled, or the produced smoke does not undergo a purification process. Due to the potential harmful impact of PAHs on health, there have been constant attempts to reduce their content in smoked products. Different factors (such as: type of wood, method, temperature and duration of smoking, usage of liquid smoke, position of the product in the smoking chamber, type of products, fat content and diameter of product, type of casing, drying and storage duration, etc.) directly affect the PAH content, and by controlling them for different types of products, the reduction of could be achieved [2,5,12,14–26]. The presence of polycyclic aromatic hydrocarbons has only been analyzed in a few traditional meat products from Serbia, and especially from Vojvodina [5,17].

Therefore, the aim of this research was to determine the content of 16 PAHs compounds, taken from the United States Environmental Protection Agency list, in dry cured meat products, bacons and dry fermented sausages, which were collected from traditional producers in the territory of Vojvodina (Šid, Temerin, Sremska Mitrovica, Ada and Subotica). Also, the connection between sensory characteristics and the content of the PAHs compound was examined.

2. Materials and Methods

2.1. Samples of Meat Products

Samples of meat products (dry cured meat products (P), bacons (S) and dry fermented sausages (K)) from six traditional producers (1–6) in the territory of Vojvodina (Šid, Temerin, Sremska Mitrovica, Ada and Subotica, Serbia) were examined. Samples P1, P2, P5 and PL6 were obtained by smoking and drying pork loin, and sample PN6 was obtained by smoking and drying pork neck meat. The sausages of the K1 and KCH4 (55 mm in diameter) groups were stuffed in collagen casing, and sausages K2, KNL3, KNL4, K5 (26–30 mm) and KNH3 (80–90 mm) were stuffed in natural casing. Smoking conditions in traditional chambers are presented in Table 1 (time of smoking type of wood and distance between fire and products).

After smoking, the processes of fermentation, drying and ripening followed, which were under the influence of outdoor conditions (temperature and relative humidity). The products of dry cured meat products were ready for consumption after 40 days, bacons after 60 days and sausages were ready after 20, 70 and 120 days for diameters of 26–30 mm, 55 mm and 80–90 mm, respectively.

Samples for the analyses were taken at the end of the drying process, when the meat products were prepared for consumption. Contents of 16 polycyclic aromatic hydrocarbons (US-EPA) were determined in three samples from each meat product. Also, the sensory quality and nutritional quality (moisture, fat and mineral content) of these products were examined.

Table 1. Processing conditions (time of smoking; type of wood; distance between fire and products) in traditional chambers.

Traditional Chambers/Samples	Time of Smoking	Type of Wood/Distance between Fire and Products
1 (P1, S1, K1)	4 days/10 h per day	beech/2.5 m
2 (P2, S2, K2)	4 days/10 h per day	beech/2 m
3 (S3, KNL3, KNH3)	7 days with pauses every second day/10–12 h per day	beech/2–2.5 m
4 (S4)	5 days/3-4 h per day	beech/2.5 m
4 (KNL4, KCH4)	3 days/4 h per day	beech+ sweet cherry/2.5 m
5 (P5)	4 days/3 h per day	beech/2 m
5 (S5)	7 days/4 h per day	beech/2 m
5 (P5)	6 days/3 h per day	beech/2 m
6 (PL6, PN6)	4 days/3 h per day	beech/2.5 m

2.2. PAH Determination

Monitoring of 16 PAHs compound content, recommended by the United States Environmental Protection Agency, was the subject of this study: naphthalene (Nap); acenaphthylene (Acy); acenaphthene (Ace); fluorene (Fln); anthracene (Ant); phenanthrene (Phe); fluoranthene (Flt); pyrene (Pyr); benz[a]anthracene (BaA); chrysene (Chr); benzo[k]flouranthene (BkF); benzo[b]fluoranthene (BbF); benzo[a]pyrene (BaP); benzo[ghi]perylene (BgP); dibenz[a,h]anthracene (DhA) and indeno[1,2,3-cd]pyrene (IcP) were determined. GCMS 7890B/5977A (gas chromatograph–mass selective detector (GC–MSD) (Agilent, Santa Clara, CA, USA) was used for PAH analysis. The method for sample preparation was described in detail by Mastanjević et al. [11].

In brief, acetonitrile and water were added to 3 g of sample and vortexed for 1 min. Then, the sample was supplemented with anhydrous $MgSO_4$ and anhydrous CH_3COONa and centrifuged for 5 min at 3000 rpm. The supernatant (1 mL) was transferred to a 5 mL tube with anhydrous $MgSO_4$ (150 mg), PSA (100 mg) and C18 (50 mg), and again centrifuged for 5 min at 3000 rpm. Obtained supernatant (0.5 mL) was transferred to a glass vial, evaporated under nitrogen gas, reconstituted in hexane and analyzed on an Agilent DB-5MS column (Agilent, Santa Clara, CA, USA). As described by Petrović et al. [27] and Mastanjević et al. [11] the sample was injected on 280 °C using spit-less mode and the following temperature program was applied: hold at 50 °C for 0.4 min; 50–195 °C (25 °C/min) and hold for 1.5 min; 195–265 °C (8 °C/min), and maintain at 315 °C for 1.25 min after increasing at a rate of 20 °C/min. The MSD temperature was 280 °C. A carrier gas flow of 1.2 mL/min was used. The identification of PAHs was verified by comparing target ions and retention times and the list of characteristic ions for all PAHs were described by Petrović et al. [27] and are given in Supplementary Table S1. The average values for precision, reproducibility, accuracy, linearity, LOQ, and LQD for method validation can be found in Supplementary Table S2, which shows that they conformed to the requirements of the SANTE/11312/2021 document for residue validation [28]. To ensure the results, blank matrix was used; smoked meat (from PT activity, blank material) was spiked with a known concentration of PAHs and calibration through matrix was carried out. The PAHs components had recoveries ranging from 85.9 to 100%, which showed a good performance of the PAH measurement method [29].

2.3. Sensory Quality

The sensory analyses were performed by the 8-member panel employees of the Faculty of Technology and Institute for Food Technology, who had previous experience of testing different traditional meat products. The samples of traditional meat products were cut into

slices, approximately 3 mm thick, and served at room temperature on white plastic plates. The panelists were asked to score samples by using a 1 (changed and atypical property) to 5 (typical, optimal and exceptional property) scale for sensory characteristics: appearance of surface, appearance and composition of cut, colour, favour/taste and texture/juiciness [30]. Also, average score of total sensory acceptability was calculated for each sample. The sensory analysis was carried out in strict adherence to the ethical principles outlined in the Declaration of Helsinki. The evaluation team, responsible for conducting the analysis, possessed the requisite qualifications and expertise in the relevant field.

2.4. Nutritional Quality

Content of moisture, fat and total ash (mineral content) in traditional meat products were determined according to methods recommended by the International Organization for Standardization [31–33].

2.5. Statistical Analysis

The software package (STATISTICA 12.0; TIBCO Software Inc., Palo Alto, CA, USA) was used for statistical analysis. The mean value ± standard deviation (SD) was used to present all results. The differences were tested using the variance analysis (ANOVA) and Duncan's multiple range test, where a $p < 0.05$ criterion indicated 95% statistical significance.

3. Results and Discussion

The presence of polycyclic aromatic hydrocarbons has been analyzed in a small number of traditional meat products from Serbia, and especially from the territory of Vojvodina. This study gives more detailed information about the qualitative and quantitative content of PAHs compounds in these types of products. The results of the content of 16 US EPA PAHs in meat products (dry cured meat products, bacons and dry fermented sausages) from Vojvodina smoked in traditional conditions are presented in Table 2. The PAHs contents determined in traditional meat products from Vojvodina were: Nap (1.95–26.88 μg/kg), Fln (6.94–70.86 μg/kg), Ant (13.82–222.29 μg/kg), Phe (0.83–57.14 μg/kg), Flt (ND–30.38 μg/kg), Pyr (ND–25.43 μg/kg), BaA (ND–1.21 μg/kg), Chr (ND–1.8 μg/kg), while contents for other investigated PAHs were below the limit of detection (ND). Predominantly, over 90% of PAHs compounds obtained in the analyzed traditional meat products were light PAHs (with two or three rings), and only Pyr, BaA and Chr of higher molecular PAHs compounds were detected in the analyzed samples. This was in accordance with the results for traditional meat products previously reported by other authors [11,13,34].

Table 2. Content of polycyclic aromatic hydrocarbons (μg/kg) in traditional meat products from north Serbia (Vojvodina).

Traditional Meat Product	Nap	Acy	Ace	Fln	Ant	Phe	Flt	Pyr	BaA	Chr	BkF	BbF	BaP	BgP	DhA	IcP	Σ 16 US EPA PAHs	PAH4
P1	7.00 ^{c} ± 0.65	<LOQ	<LOQ	43.60 ^{h} ± 1.23	121.27 ^{h} ± 3.91	24.66 ^{f} ± 0.83	6.88 ^{e} ± 0.05	6.50 ^{gh} ± 0.38	0.74 ^{b} ± 0.08	0.71 ^{c} ± 0.11	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	211.36 ^{h} ± 7.24	**1.45 ^{c} ± 0.19**
P2	4.90 ^{b} ± 1.01	<LOQ	<LOQ	70.86 ^{l} ± 3.12	222.29 ^{j} ± 2.98	57.14 ^{h} ± 4.22	30.38 ^{i} ± 2.05	25.43 ^{m} ± 0.98	0.81 ^{c} ± 0.12	0.94 ^{d} ± 0.09	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	412.76 ^{j} ± 14.57	**1.75 ^{d} ± 0.21**
P5	2.86 ^{a} ± 0.17	<LOQ,	<LOQ,	52.62 ^{j} ± 0.97	129.48 ^{i} ± 1.54	36.52 ^{g} ± 1.17	18.93 ^{h} ± 1.63	19.17 ^{l} ± 0.74	1.21 ^{d} ± 0.05	1.01 ^{d} ± 0.07	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	261.81 ^{i} ± 6.34	**2.22 ^{e} ± 0.12**
PL6	8.73 ^{de} ± 1.11	<LOQ	<LOQ	21.77 ^{f} ± 1.21	52.63 ^{e} ± 0.88	8.79 ^{c} ± 1.06	4.53 ^{d} ± 0.10	2.71 ^{cd} ± 0.03	<LOQ,	0.57 ^{b} ± 0.06	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	99.73 ^{de} ± 4.45	**0.57 ^{a} ± 0.06**
PN6	3.11 ^{a} ± 0.07	<LOQ,	<LOQ,	23.29 ^{f} ± 1.80	111.36 ^{g} ± 1.87	18.59 ^{d} ± 0.33	13.37 ^{g} ± 1.05	7.73 ^{h} ± 0.33	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	177.45 ^{g} ± 5.45	**<LOQ,**
S1	21.38 ^{h} ± 0.95	<LOQ	<LOQ	20.62 ^{ef} ± 1.45	46.40 ^{d} ± 2.23	8.49 ^{c} ± 0.08	2.45 ^{c} ± 0.18	1.55 ^{bc} ± 0.07	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	100.91 ^{e} ± 3.06	**<LOQ,**
S2	1.95 ^{a} ± 0.27	<LOQ	<LOQ	22.59 ^{f} ± 0.81	109.65 ^{g} ± 5.27	21.76 ^{e} ± 2.01	19.59 ^{h} ± 0.84	13.32 ^{j} ± 1.31	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	188.86 ^{g} ± 5.95	**<LOQ,**
S3	5.87 ^{bc} ± 0.17	<LOQ,	<LOQ,	6.94 ^{a} ± 0.77	22.04 ^{b} ± 1.33	2.51 ^{ab} ± 0.16	4.39 ^{d} ± 0.09	0.69 ^{ab} ± 0.03	<LOQ,	0.48 ^{a} ± 0.04	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	42.91 ^{ab} ± 2.59	**0.48 ^{a} ± 0.04**
S4	8.95 ^{de} ± 1.25	<LOQ	<LOQ	8.29 ^{a} ± 0.14	13.82 ^{a} ± 2.01	2.16 ^{ab} ± 0.08	1.98 ^{bc} ± 0.17	0.79 ^{ab} ± 0.06	<LOQ,	0.44 ^{a} ± 0.02	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	36.43 ^{a} ± 3.73	**0.44 ^{a} ± 0.02**
S5	10.08 ^{ef} ± 0.94	<LOQ,	<LOQ,	15.43 ^{cd} ± 1.85	39.55 ^{c} ± 3.11	10.67 ^{c} ± 1.26	6.43 ^{e} ± 0.44	5.19 ^{fg} ± 0.06	0.43 ^{a} ± 0.03	0.62 ^{b} ± 0.05	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	88.41 ^{d} ± 7.74	**1.05 ^{b} ± 0.08**
K1	11.48 ^{f} ± 0.77	<LOQ	<LOQ	13.03 ^{bc} ± 1.10	18.58 ^{ab} ± 0.87	0.83 ^{a} ± 0.03	0.55 ^{ab} ± 0.07	4.77 ^{ef} ± 0.81	<LOQ,	<LOQ,	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	49.24 ^{b} ± 3.65	**<LOQ,**
K2	15.76 ^{g} ± 0.30	<LOQ	<LOQ	67.39 ^{k} ± 2.16	117.37 ^{h} ± 4.93	36.04 ^{g} ± 1.85	18.21 ^{h} ± 2.03	15.84 ^{k} ± 0.88	<LOQ,	<LOQ,,	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	270.60 ^{i} ± 8.09	**<LOQ,**
KNL3	10.04 ^{ef} ± 1.11	<LOQ,	<LOQ,	17.92 ^{de} ± 1.42	55.46 ^{e} ± 8.33	9.59 ^{c} ± 1.06	<LOQ,	<LOQ,	<LQD	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	93.01 ^{de} ± 11.92	**<LOQ,**
KNH3	3.27 ^{a} ± 0.22	<LOQ	<LOQ	12.23 ^{bc} ± 0.69	38.65 ^{c} ± 1.33	4.49 ^{b} ± 0.16	6.33 ^{e} ± 0.64	5.03 ^{f} ± 0.37	<LOQ,	<LOQ,	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	70.00 ^{c} ± 3.41	**<LOQ,**
KNL4	7.38 ^{cd} ± 0.73	<LOQ,	<LOQ,	28.44 ^{g} ± 4.07	57.25 ^{e} ± 3.91	10.38 ^{c} ± 0.80	6.26 ^{e} ± 0.34	11.44 ^{i} ± 1.14	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	<LOQ,	121.16 ^{f} ± 3.17	**<LOQ,**
KCH4	3.10 ^{a} ± 0.34	<LOQ	<LOQ	9.72 ^{ab} ± 0.81	23.10 ^{b} ± 3.64	3.54 ^{b} ± 0.67	4.17 ^{d} ± 0.84	3.60 ^{de} ± 0.91	<LOQ,	<LOQ,	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	47.23 ^{ab} ± 5.19	**<LOQ,**
K5	26.88 ^{i} ± 2.27	<LOQ	<LOQ	48.89 ^{i} ± 4.96	85.28 ^{f} ± 3.91	18.51 ^{d} ± 0.89	11.43 ^{f} ± 1.07	10.64 ^{i} ± 2.04	<LOQ,	1.80 ^{e} ± 0.05	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	203.44 ^{h} ± 7.37	**1.80 ^{d} ± 0.05**

Results are expressed as means ± standard deviations (n = 3); In the same column different letters mean that values are significantly different (p <0.05).

The total content of 16 US-EPA PAHs in dry cured meat products was in the range from 99.73 µg/kg (PL6 group) to 412.76 µg/kg (P2 group), with significant difference ($p < 0.05$) between all investigated groups. Samples of P2 group were smoked around 40 h (4 days/10 h per day), with 2 m of distance between fire and products; samples of PL6 and PN6 groups were smoked only 12 h (4 days/3 h per day), with 2.5 m of distance between fire and products (Table 1). The longer smoking time and lower distance between fire and product resulted in higher content of 16 US-EPA PAHs. Further, samples of the PL6 group had significantly lower ($p < 0.05$) total content of 16 US-EPA PAHs, compared to the samples of the PN6 group (99.73 µg/kg; 177.45 µg/kg respectively). This result indicated that meat products with lower total fat content (PL6-19.73%; PN6-38.26%—Table 3) smoked in the same conditions had lower content of 16 US-EPA PAHs. The impact of higher total fat content on the higher PAH content were also reported by other authors [2,13,35]. Pöhlmann et al. [22] found that PAH content clearly increases with fat content in smoked Frankfurter-type sausages, which can be explained by the liposoluble character of these compounds.

Table 3. The content of moisture, total fat and total ash (%) in traditional meat products from north Serbia (Vojvodina).

Traditional Meat Product		Moisture Content	Total Fat Content	Total Ach Content
dry cured meat products	P1	40.37 ^{m} ± 0.16	23.07 ^{e} ± 0.06	6.56 ^{k} ± 0.06
	P2	44.66 ^{n} ± 0.21	10.69 ^{b} ± 0.16	5.50 ^{h} ± 0.07
	P5	53.18 ^{o} ± 0.34	8.19 ^{a} ± 0.15	5.17 ^{g} ± 0.02
	PL6	33.21 ^{j} ± 0.13	19.73 ^{d} ± 0.19	7.97 ^{m} ± 0.03
	PN6	23.71 ^{f} ± 0.22	38.26 ^{i} ± 0.05	7.03 ^{l} ± 0.03
bacons	S1	20.08 ^{d} ± 0.05	62.84 ^{p} ± 0.07	4.99 ^{f} ± 0.05
	S2	23.29 ^{e} ± 0.25	49.76 ^{m} ± 0.42	6.39 ^{j} ± 0.01
	S3	10.30 ^{a} ± 0.19	70.87 ^{r} ± 0.02	2.72 ^{a} ± 0.02
	S4	15.02 ^{b} ± 0.05	51.76 ^{o} ± 0.19	4.91 ^{e} ± 0.01
	S5	33.14 ^{j} ± 0.07	39.65 ^{j} ± 0.13	7.93 ^{m} ± 0.04
dry fermented sausages	K1	34.99 ^{l} ± 0.01	33.89 ^{g} ± 0.05	4.59 ^{b} ± 0.01
	K2	19.96 ^{d} ± 0.21	41.53 ^{k} ± 0.05	4.85 ^{d} ± 0.02
	KNL3	18.42 ^{c} ± 0.26	44.11 ^{l} ± 0.08	4.55 ^{b} ± 0.01
	KNH3	33.78 ^{k} ± 0.03	18.92 ^{c} ± 0.04	6.16 ^{i} ± 0.02
	KNL4	26.65 ^{g} ± 0.22	36.35 ^{h} ± 0.27	5.03 ^{f} ± 0.03
	KCH4	28.85 ^{h} ± 0.20	26.13 ^{f} ± 0.12	4.71 ^{c} ± 0.02
	K5	31.25 ^{i} ± 0.12	41.49 ^{n} ± 0.01	4.68 ^{c} ± 0.06

Results are expressed as means ± standard deviations ($n = 3$); In the same column different letters mean that values are significantly different ($p < 0.05$).

All examined groups of dry cured meat products had a total sensory acceptability score over 4.3 (Table 4) and there were no significant differences ($p > 0.05$) between investigated groups. Dry cured meat products with reduced content of PAHs compounds could be produced without impairing the sensory quality (i.e. longer time of smoking is not necessary or justified). The smoking duration of 4 days (3 h per day) can be considered optimal in the traditional production of dry cured meat products in the territory of Vojvodina, from both a sensory and PAH viewpoint.

Table 4. Sensory characteristics of traditional meat products from north Serbia (Vojvodina).

Traditional Meat Product		Appearance of Surface	Composition of Cut	Colour	Favour/Taste	Texture/ Juiciness	Total Sensory Acceptability
dry cured meat products	P1	$5.00^{d} \pm 0.00$	$4.55^{cd} \pm 0.37$	$4.40^{cde} \pm 0.22$	$4.45^{def} \pm 0.45$	$4.25^{bcde} \pm 0.35$	$4.53^{cd} \pm 0.23$
	P2	$4.80^{cd} \pm 0.45$	$4.85^{d} \pm 0.22$	$5.00^{e} \pm 0.00$	$4.25^{cdef} \pm 0.25$	$4.35^{cde} \pm 0.34$	$4.65^{cd} \pm 0.14$
	P5	$4.50^{bcd} \pm 0.87$	$4.50^{cd} \pm 0.61$	$3.95^{abcd} \pm 0.93$	$4.40^{def} \pm 0.89$	$4.45^{cde} \pm 0.67$	$4.36^{bcd} \pm 0.77$
	PL6	$4.75^{cd} \pm 0.43$	$4.75^{cd} \pm 0.25$	$4.60^{cde} \pm 0.22$	$4.45^{def} \pm 0.33$	$4.70^{de} \pm 0.33$	$4.65^{cd} \pm 0.19$
	PN6	$4.40^{bcd} \pm 0.22$	$4.65^{cd} \pm 0.42$	$4.80^{de} \pm 0.27$	$4.60^{ef} \pm 0.45$	$4.25^{bcde} \pm 0.25$	$4.54^{cd} \pm 0.30$
bacons	S1	$4.75^{cd} \pm 0.43$	$4.80^{cd} \pm 0.27$	$4.80^{de} \pm 0.27$	$4.60^{ef} \pm 0.42$	$4.30^{bcde} \pm 0.45$	$4.65^{cd} \pm 0.23$
	S2	$4.25^{bcd} \pm 0.75$	$4.20^{bcd} \pm 0.76$	$3.90^{abcd} \pm 0.72$	$3.35^{b} \pm 0.60$	$3.80^{abcd} \pm 0.57$	$3.90^{abc} \pm 0.60$
	S3	$4.10^{abc} \pm 0.76$	$4.30^{bcd} \pm 0.57$	$4.20^{bcde} \pm 0.57$	$3.25^{b} \pm 0.61$	$3.90^{abcd} \pm 0.55$	$3.95^{abc} \pm 0.55$
	S4	$4.60^{bcd} \pm 0.65$	$4.50^{cd} \pm 0.40$	$4.45^{cde} \pm 0.27$	$4.50^{ef} \pm 0.35$	$4.20^{bcde} \pm 0.27$	$4.45^{cd} \pm 0.32$
	S5	$4.40^{bcd} \pm 0.45$	$3.65^{ab} \pm 0.96$	$3.00^{a} \pm 0.61$	$3.45^{bc} \pm 0.27$	$3.65^{abc} \pm 0.49$	$3.63^{ab} \pm 0.50$
dry fermented sausages	K1	$4.65^{bcd} \pm 0.22$	$3.95^{abc} \pm 0.57$	$3.65^{abc} \pm 0.70$	$3.85^{bcde} \pm 0.52$	$4.20^{bcde} \pm 0.33$	$4.06^{abc} \pm 0.37$
	K2	$4.40^{bcd} \pm 0.42$	$4.40^{bcd} \pm 0.38$	$4.25^{cde} \pm 0.35$	$3.85^{bcde} \pm 0.49$	$4.20^{bcde} \pm 0.27$	$4.22^{bcd} \pm 0.27$
	KNL3	$3.45^{a} \pm 0.57$	$3.40^{a} \pm 0.82$	$3.25^{ab} \pm 0.71$	$3.60^{bcd} \pm 0.42$	$3.40^{ab} \pm 0.65$	$3.42^{a} \pm 0.59$
	KNH3	$3.85^{ab} \pm 1.14$	$4.55^{cd} \pm 0.11$	$4.45^{cde} \pm 0.41$	$4.10^{bcdef} \pm 0.22$	$4.45^{cde} \pm 0.37$	$4.28^{bcd} \pm 0.34$
	KNL4	$4.45^{bcd} \pm 0.45$	$3.60^{ab} \pm 1.08$	$3.25^{ab} \pm 1.94$	$2.40^{am} \pm 1.64$	$3.30^{a} \pm 1.99$	$3.40^{a} \pm 1.28$
	KCH4	$5.00^{d} \pm 0.00$	$4.75^{cd} \pm 0.25$	$4.90^{de} \pm 0.22$	$4.80^{f} \pm 0.27$	$4.95^{d} \pm 0.11$	$4.88^{d} \pm 0.12$
	K5	$4.25^{bcd} \pm 0.56$	$4.35^{bcd} \pm 0.34$	$4.35^{cde} \pm 0.34$	$4.30^{cdef} \pm 0.45$	$4.40^{cde} \pm 0.22$	$4.33^{bcd} \pm 0.30$

Results are expressed as means ± standard deviations (n = 8); In the same column different letters mean that values are significantly different ($p < 0.05$).

The total content of 16 US-EPA PAHs in bacons was in the range from 36.43 µg/kg (S4 group) to 188.86 µg/kg (S2 group), with significant differences ($p < 0.05$) between all investigated groups, except between S3 and S4. The content of PAHs was significantly lower in bacon samples than in dry cured meat products smoked in the same conditions, although bacon samples had significantly higher fat content (from S5—39.65% to S3—70.87%), which can be explained by the fact that the skin on the bacon is a significant barrier to PAHs compounds penetration. Đinović et. al [17] determined a significantly higher total sum of the 16 EU priority PAHs in bacon without skin (22.7 µg/kg), compared to bacon with skin (12.2 µg/kg) smoked in traditional conditions for 15 days. Samples of S2 group had significantly ($p < 0.05$) lower fat content (49.76%) compared to the S3 group of samples (70.87%) and significantly ($p < 0.05$) higher content of 16 US-EPA PAHs (S2—188.86 µg/kg; S3—42.91 µg/kg—Table 2), showing that the smoking conditions (smoking time and distance between fire and products) had a much greater impact on PAH content than the fat content. On the basis of the score for sensory quality, it can be concluded that the optimal smoking time for bacons was 5 days (3–4 h per day) at a distance of 2.5 m between fire and products, as this resulted in samples with the lowest PAH content (S4—36.43 µg/kg) and the highest sensory score (4.45).

Further, the total content of 16 US-EPA PAHs in dry fermented sausages was in the range from 47.23 µg/kg (KCH4 group) to 270.60 µg/kg (K2 group), with significant differences ($p < 0.05$) between all investigated groups, except between K1 and KCH4 groups. It is important to note that sausage samples in collagen casing (K1 and KCH4 groups) had lower content of 16 US-EPA PAHs, compared with sausages in natural casing (K2, KNL3, KNH3, KNL4 and K5 groups). This confirms the findings of other researchers who reported a significant difference in PAH levels depending on the casing type, indicating that synthetic casings reduced the content of PAHs in smoked sausages [5,35]. Natural casings have high porosity with large average pore size, which enables the fat to pass through the casing and coat its outer layer, giving it a sticky texture. These features of the casing, along with the product's crinkled surface, ensure that the smoke particles can easily attach to and stick to

the sausage surface [2,35]. The significantly ($p < 0.05$) higher PAHs content of KNL3 group, smoked under the same conditions and in the same type of casing (natural) as the sausages of the KNH3 group (93.01 μg/kg; 70.00 μg/kg respectively), can be explained by referring to the smaller diameter of KNL3 sausages and the greater penetration of PAHs compounds into the center of the product. Lorenza et al. [9] determined higher total content of 16 EPA PAHs in "*Androlla*" sausages than in "*Botillo*" sausages, underlining the importance of surface/mass ratio factor for PAH contamination. "*Androlla*" being of lower diameter than "*Botillo*" showed a larger surface per unit of volume, which favored the adsorption of PAH. Producers using the traditional smoking method should pay attention to the size of diameter and type of the casing, because collagen casings with a higher diameter can contribute to the reduction of PAH content in sausages.

According to the results obtained in this study, traditional meat products from Vojvodina are safe for consumption, from a PAH point of view, because BaP content was lower than the limit of detection for all analyzed samples, while contents of PAH4 were greatly below the maximal allowed value (from ND to 2.22 μg/kg). Also, obtained results indicate that it is possible to produce traditional meat products with typical sensory characteristics, with a lower content of PAHs compounds (BaP < 2 μg/kg; PAH4 < 12 μg/kg), and that it is not necessary for traditional products from Vojvodina to apply the new regulatory criteria EU Regulation (BaP < 5 μg/kg; PAH4 < 30 μg/kg) [7].

Higher levels of 16 EPA PAHs (609–679 μg/kg) and PAH4 (from ND to 9.90 μg/kg) were determined in Croatian dry fermented sausages by Mastanjević et al. [11], in Portuguese traditional meat products (16 US-EPA PAHs = 877.37–2609.81 μg/kg; PAH4 = 3.47–6.94 μg/kg) by Santos et al. [13], in Cypriot traditional meat products by Kafouris et al. [8] (PAH4 = 5.9–15.2 μg/kg), as well as in dry cured meat products from Bosnia and Hercegovina by Puljić et al. [12] (16 US-EPA PAHs = 145–2474 μg/kg; PAH4 = 12.7–32.5 μg/kg). In contrast, traditional meat products from Spain [9,10] and Italy [36] showed lower amounts of 16 US EPA PAHs (<100 μg/kg), while dry fermented sausages from Bosnia and Hercegovina [37] had similar amounts of 16 US EPA PAHs (<300 μg/kg), compared to the results obtained in this study. The variations in the PAHs levels could be attributed to the traditional smoking process, which is specific for each geographical area and the type of meat product. Several factors during the traditional smoking process, such as temperature, humidity, type of wood, smoke intensity, number of smoking days, type of meat products, etc., influence the type and the level of PAHs compounds in traditional meat products [2,5,9,10,17,21,38,39]. In order to draw an accurate conclusion about the quality and quantity of PAHs compounds in traditional meat products, it is essential to conduct more studies of PAHs content in different products from across numerous geographical areas.

Generally, in this study, the highest contents of 16 US-EPA PAHs compounds were determined in samples smoked in chamber 2, where the smoking process was more intense, beech was used as a type of wood and there was a distance of 2 m between the fire and the products. The lowest contents of 16 US-EPA PAHs compounds were determined by sample producer in chamber 4, where the smoking process was adapted to each type of product, and a mixture of beech and Spanish cherry wood was used. The smoking time was significantly shorter than for chamber 2, and it was expected that the samples would have a lower content of PAHs compounds. Also, for samples of dry cured meat products, the lowest content of 16 US-EPA PAHs was determined in samples from chamber number 6, where the smoking process was only performed for 4 days for 3 h/day. On the basis of the scores for total sensory acceptability (S4-4.45; KCH4-4.88; PL6-4.65 and PN6-4.54), it can be concluded that shortening the smoking process in chamber 4 and 6 was justified, because products of good sensory quality with reduced content of PAHs were obtained. The obtained results indicate that, in order to decrease the levels of PAH in traditional smoked meat products from the territory of Vojvodina, manufacturers must consider shortening the smoking process and establishing a higher distance between fire and products.

4. Conclusions

The total content of 16 US-EPA PAHs in dry cured meat products was in the range from 99.73 μg/kg to 412.76 μg/kg; in bacons it was in the range from 36.43 μg/kg to 188.86 μg/kg; and in dry fermented sausages in the range from 47.23 μg/kg to 270.60 μg/kg. The obtained results indicate that manufacturers must, in seeking to decrease the level of PAH in the traditional production of smoked meat products from the territory of Vojvodina, consider a shorter smoking process and establishing a larger distance between fire and products. Also, in the sausages, the production usage of collagen casings with a higher diameter can contribute to the reduction of PAH content in the final product. All examined samples of traditional meat products from Vojvodina had BaP content lower than the limit of detection and contents of PAH4 greatly lower than the set maximum value, thus confirming the safety of these products. This study showed that it is possible to produce traditional meat products (dry cured meat products, bacons and dry fermented sausages) in the territory of Vojvodina that combine typical sensory characteristics with lower content of PAHs compounds (BaP < 2 μg/kg; PAH4 < 12 μg/kg), and that this does not necessarily require applying the new regulatory criteria—EU Regulation 2023/915 (BaP < 5 μg/kg; PAH4 < 30 μg/kg).

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/fermentation10020104/s1, Retention time (RT), molecular mass (MW), primary (target) ion (T) and secondary (qualifier) ion (Q) can be found in Supplementary Table S1. The average values for precision, reproducibility, accuracy, linearity, LOQ, and LQD for method validation can be found in Supplementary Table S2.

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