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Distribution of Polycyclic Aromatic Hydrocarbons in Traditional Dry Cured Smoked Ham Slavonska Šunka

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Abstract: Slavonska šunka is a traditional dry-cured smoked ham. Smoking with open fire commonly results in the accumulation of polycyclic aromatic hydrocarbons (PAH) molecules in tissues. The objective of this research was to assess the types and concentrations of 16 PAHs in 30 samples of traditional dry-cured smoked ham Slavonska šunka. In general, all samples had high values of anthracene while higher levels of acenaphthylene were present in some samples. In sample SS1, the maximal value for anthracene reached 228.03 µg/kg, while sample SS28 had the lowest value of this PAH—33.38 µg/kg. SS1 had increased values for several other PAHs: benzo[a]anthracene (54.03 µg/kg), acenaphtene (19.90 µg/kg) and phenanthrene (27.11 µg/kg). Cancerogenic benzo[a]pyrene content was below the limit of quantification for all samples. PAH4 (benzo[a]anthracene, chrysene, benzo[b]fluoranthene and benzo[a]pyrene) concentrations were above legislative recommendation (>30 µg/kg) in seven samples. All the samples of Slavonska šunka had significant PAH16 concentrations, with the lowest value of 61.30 µg/kg (SS28) and the highest 462.83 µg/kg (SS1). The highest concentrations of PAHs were determined in the Vukovar–Srijem county samples. Another conclusion from this research is that samples smoked with elm tree showed high PAH concentrations in comparison to the ones smoked with beech and hornbeam.

Keywords: dry-cured smoked ham; Slavonska šunka; PAH content; traditional smoking

1. Introduction

Traditional meat products have always been highly valued for their taste. The production of dry-cured smoked ham in a traditional way using open fire ensures the unique experience of smoky aroma. The popularization of original and organic products resulted in returning to traditional production methods, mainly by small family businesses. Traditional meat products are extremely wanted, the demand and supply are always high. The production of high-quality Slavonska šunka takes a certain amount of time. The formation of characteristic aroma, taste and smell of Slavonska šunka is a timely process, but consumers appreciate the final outcome. Slavonska šunka is a traditional smoked dry-cured product that has a great potential for receiving a protected geographical indication (PGI) according to the European Commission of European Union (EU) legislative recommendation. However, due to the smoking procedure using open fire, there is a high risk that it may contain certain amounts of different polycyclic aromatic hydrocarbons (PAHs). PAHs are general contaminants in smoked foods such as meat and fish [1,2]. Many traditional meat products from European counties (Portugal, Spain, Greece, etc.) receive a lot of attention because they might contain PAHs [1–8]. Gomes

et al. conducted a research on Portuguese [1], and Škaljac et al. [7,8] conducted research on traditional sausages originating from Serbia. They concluded that the traditional sausages smoked in industrial conditions showed lower PAH contamination. On the other hand, Roseiro et al. investigated PAHs in Portuguese [3] and Lorenzo et al. [5,6] in Spanish traditional sausages exposed to traditional smoking procedure (open fire) and reported relatively high PAH16 content. Incomplete wood combustion during the smoking procedure can result in significantly higher polycyclic aromatic hydrocarbon amounts than those set by legislation. They pose a threat for human health because, according to the International Agency for Research on Cancer (IARC) [9], some of them are carcinogenic and mutagenic [10,11]. All food processing that includes grilling, cooking, smoking or roasting has a tendency to increase PAH contamination of processed food [12–14].

The most common source of PAHs in food is smoking (a type of wood and its moisture content, casing), but environmental contamination (exhaustion fumes, wildfire, and other combustion-prone processes) also significantly contribute to the contamination of food [15,16]. This means that PAH content in meat products can be related to the contamination of feed used in pig breeding. For that reason, the US Environmental Protection Agency (US EPA) defined 16 PAHs (naphthalene (Nap), acenaphthylene (Anl), acenaphthene (Ane), fluorene (Flu), phenanthrene (Phen), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (InP), dibenz[a,h]anthracene (DahA) and benzo[g,h,i]perylene (BghiP)) (PAH16) as priority environmental pollutants [17]. The European Food Safety Authority (EFSA), on the other hand, defined that the concentrations of BaP and the sum of the concentrations of four PAHs: BaP, BaA, BbF and Chr (PAH4) [18], should be a reference when determining PAHs in food. According to the European Commission (EU) Regulation no. 1881/2006, 835/2011 and XX/2019 [19–21], the maximum permissible concentration of BaP in meat products is 2 μg/kg and the sum PAH4 concentrations should not exceed 12 μg/kg. However, since traditional meat products are prone to higher levels of PAHs, a special regulation regarding PAH concentrations in such products has been issued by the European Commission (EU) [21] in which the maximum BaP is set to 5 μg/kg, and the sum of the PAH4 is limited to 30 μg/kg.

The aim of this research was to determine and quantify the types and concentrations of 16 PAHs in 30 samples of Slavonska šunka, a traditional dry-cured smoked ham, bought at the city market and labeled as traditional food.

2. Materials and Methods

2.1. Sample Production and Sampling

In order to carry out this study, 30 samples of Slavonska šunka, manufactured by representative homemade producers, were chosen (supplementary material). Hams were manufactured in a non-industrial environment, using traditional techniques. The influence of climate and region of origin was the main factor in processing the hams. Upon collection, the samples were transported to the laboratory in a very short time, and stored in a refrigerator at a temperature below 4 °C. All samples were prepared according to the traditional manufacturing procedure without the addition of nitrites or ascorbic acid. Hams were primarily processed, i.e., shaped in such a way that sacrum and pelvic bones (ilium, ischium and pubis) were taken out from the hams. After taking out the pelvic bones, ham's muscles were rounded in a semicircle so that the bottom is about 6 cm from the femur head. The raw hams were traditionally dry salted with an indeterminate amount of salt. This means that the hams were rubbed and then put in salt and left to rest. The rest stage lasted around 30 days and it varied based on the raw ham weight. The temperature at this stage should be around 8 °C and relative humidity about 85%. The hams were turned every seven days. The temperature and relative humidity of the air at this stage should be from 18 to 20 °C and 70 to 90%. After the smoking, Slavonska šunka has to undergo the ripening phase. This phase takes up the longest amount of time during the production and it can last more than 10 months, depending on ham's initial weight. During

this period, hams are kept in a dark room with the temperature ranging from 14 to 17 °C with relative humidity reaching from 70 to 80%. After this stage, Slavonska šunka is ready for consumption. Before the PAH determinations, samples were packed in glass jars and stored in the dark at −30 °C. The description of conditions during the processing of Slavonska šunka is shown in Table 1.

Table 1. Description of conditions during the processing of Slavonska šunka.

Abbreviation	Place of Production	Smoking Time (Days)	Processing Time (Days)	Wood Used for Smoke Production	Processing Conditions during Smoking
SS1–SS12	Vukovar-Srijem county	Aprox. 30 every second day for 2–3 h	cca. 400	Mostly elm	Uncontrolled (natural climatic conditions)
SS13–SS20	Osijek-Baranja county	Aprox. 30 every second day for 2–3 h	cca. 400	Mostly beech and hornbeam	Uncontrolled (natural climatic conditions)
SS21–SS24	Brod-Posavina County	Aprox. 30 every second day for 2–3 h	cca. 400	Mostly beech and hornbeam	Uncontrolled (natural climatic conditions)
SS25–SS30	Požega-Slavonia County	Aprox. 30 every second day for 2–3 h	cca. 400	Mostly beech and hornbeam	Uncontrolled (natural climatic conditions)

2.2. GC-MS Analysis

Were subjected to chromatographic separation according to Mastanjević et al. [10]. All analyses were done in triplicate.

2.3. Statistical Analysis

Results of PAHs grouped according to the production site (county) and wood type were subjected to the analysis of variance (ANOVA) and Fisher's least significant difference (LSD), with significance defined at $p < 0.05$. Statistica 12.7 (StatSoft Inc., Tulsa, OK, USA, 2015) was used for statistical analysis. The Chi-square (χ^2) test was used to examine the potential differences in PAH4 content between the different counties.

3. Results and Discussion

In order to get an insight into the amounts and types of PAHs an average consumer is exposed to when consuming a traditional Slavonska šunka, this research was conducted on samples obtained from the local market. The research was performed on samples produced at small family farms and smoked using a traditional method. As the smoking is done with open fire, PAH contamination cannot be avoided and moreover, can reach disturbingly high concentrations. Several authors associate the wood type with the composition and concentration of PAHs in smoked meat products. Moisture content, concentration of oxygen in smoking chambers, temperature of wood combustion, etc. are also important factors for the emergence of PAHs in smoked meat products [3,4,22–26]. According to different sources [1,3,8,11,27–32], casing (natural, collagen, cellulose) acts as an obstacle and may disrupt the diffusion and deposition of smoke components into smoked foods.

The results of detected and quantified PAHs are shown in Table 2. Most of the heavy PAHs, the ones with four or more benzene rings were not quantified in this research since their levels stayed below the level of quantification (LOQ). Chry was detected and quantified in only one sample, SS1 with 45.23 μg/kg. BbF, BkF, BaP, DahA, BghiP and InP were below the LOQ

Table 2. Polycyclic aromatic hydrocarbons (PAH) contents (μg/kg) in Slavonska šunka.

	Nap	Anl	Ane	Flu	Ant	Phen	Flt	Pyr	BaA	Chry	PAH 4	PAH 16
SS1	6.55	20.62	19.90	35.06	228.03	27.11	26.30	<LOQ	54.03	45.23	99.26	462.83
SS2	0.66	39.09	<LOQ	44.26	168.63	24.27	11.30	<LOQ	28.56	<LOQ	28.56	316.78
SS3	<LOQ	36.46	4.73	32.32	115.56	14.93	8.93	<LOQ	44.26	<LOQ	44.26	257.19
SS4	<LOQ	16.63	4.27	25.68	100.15	13.53	9.05	<LOQ	40.54	<LOQ	40.54	209.86
SS5	5.05	23.24	16.13	17.6	97.96	18.62	17.49	<LOQ	27.90	<LOQ	27.90	224.04
SS6	1.53	4.84	1.40	16.10	153.92	24.86	23.46	<LOQ	22.41	<LOQ	22.41	248.49
SS7	<LOQ	14.68	<LOQ	36.47	151.71	23.04	14.46	<LOQ	22.77	<LOQ	22.77	263.14
SS8	<LOQ	<LOQ	14.66	41.15	154.01	20.17	13.11	<LOQ	21.52	<LOQ	21.52	264.63
SS9	3.53	6.34	2.42	16.57	148.93	24.36	22.46	<LOQ	20.91	<LOQ	20.91	245.51
SS10	<LOQ	15.63	1.27	24.18	95.15	13.25	8.82	<LOQ	28.54	<LOQ	28.54	186.85
SS11	<LOQ	1.37	4.25	24.86	86.86	5.84	7.37	<LOQ	47.49	<LOQ	47.49	178.06
SS12	<LOQ	13.24	12.13	27.64	77.96	8.62	7.49	<LOQ	27.90f	<LOQ	27.90	174.99
SS13	3.11	9.13	<LOQ	26.36	102.66	19.21	8.35	6.81	<LOQ	<LOQ	<LOQ	175.64
SS14	2.52	2.25	4.68	15.29	59.81	3.62	4.76	<LOQ	33.77	<LOQ	33.77	126.71
SS15	<LOQ	6.22	2.29	20.96	72.71	6.79	5.50	5.41	10.12	<LOQ	10.12	130.03
SS16	<LOQ	1.27	4.15	23.86	84.36	4.84	6.87	<LOQ	27.49	<LOQ	27.49	152.86
SS17	2.42	2.15	4.53	13.79	59.31	3.32	4.26	<LOQ	31.76	<LOQ	31.77	121.56
SS18	<LOQ	3.74	2.13	17.63	57.96	4.62	3.99	<LOQ	32.90	<LOQ	32.90	122.99
SS19	<LOQ	2.55	3.26	18.99	64.24	4.91	4.52	<LOQ	7.16	<LOQ	7.16	105.65
SS20	<LOQ	<LOQ	19.77	14.83	61.16	2.32	3.91	4.90	14.23	<LOQ	14.23	121.12
SS21	19.39	2.15	<LOQ	14.10	63.61	5.01	5.83	<LOQ	8.71	<LOQ	8.71	118.81
SS22	<LOQ	<LOQ	4.26d	11.22	54.95	3.61	4.09	4.24	25.58	<LOQ	25.58	107.96
SS23	<LOQ	<LOQ	4.13	11.72	55.45	3.61	4.08	4.20	25.08	<LOQ	25.08	108.27
SS24	<LOQ	<LOQ	0.98	13.65	58.97	6.85	4.31	3.93	11.74	<LOQ	11.74	100.44
SS25	<LOQ	1.47	0.61	17.16	66.52	4.05	4.83	<LOQ	5.04	<LOQ	5.04	99.69
SS26	<LOQ	0.47	0.41	17.66	67.52	4.55	4.68	<LOQ	4.54	<LOQ	4.54	99.84
SS27	<LOQ	<LOQ	<LOQ	15.23	45.55	3.74	4.76	3.41	14.15	<LOQ	14.15	86.84
SS28	8.38	3.14	<LOQ	7.33	33.38	1.65	7.43	<LOQ	<LOQ	<LOQ	<LOQ	61.30
SS29	9.88	1.13	<LOQ	9.33	39.88	2.65	5.93	<LOQ	<LOQ	<LOQ	<LOQ	68.80
SS30	<LOQ	<LOQ	<LOQ	11.73	42.05	2.24	3.76	4.91	11.15	<LOQ	11.15	75.84

Values are means of three measurements; LOQ—limit of quantification.

Light PAHs, containing up to four benzene rings—Nap, Anl, Ane, BaA and Pyr were detected in some of the samples. Nap was quantified in 11 samples of Slavonska šunka. A majority of samples had unquantifiable levels of this PAH. The highest level of this PAH was detected in SS21, 19.39 µg/kg. Anl was below LOQ in seven samples. The highest concentration of Anl was detected in sample SS2, amounting 39.09 µg/kg. Ane was also below the LOQ in eight samples and the highest concentration was detected in SS1 with 19.09 µg/kg. BaA was quantified in the majority of samples, with the lowest quantified value of 4.54 µg/kg in sample SS26. The highest value was quantified in sample SS1 and was 54.03 µg/kg. This PAH stayed below the LOQ in only three samples. Pyr was quantified in eight samples, while in 22 samples it stayed below the LOQ. The highest level was detected in sample SS13 and it amounted to 6.81 µg/kg. Most of the samples ranged between 3 and 5 µg/kg.

Flu, Ant, Phen, and Flt appeared to be the most abundant light PAHs as they were quantified in all of the samples. Flu showed the highest concentration in sample SS2 with 44.26 µg/kg, and its minimum value was detected SS28 with 7.33 µg/kg. Ant showed very high levels (>100 µg/kg) in nine samples, SS1–4, SS6–9 and SS13. The highest concentration of Ant was quantified in sample SS1 with 228.03 µg/kg. Other samples were below 100 µg/kg and the lowest concentration quantified for this PAH was 33.38 µg/kg in sample SS28. Phen concentrations stayed below 30 µg/kg in all samples, and the highest level was found in SS1, 27.11 µg/kg.

The sum of 4 PAHs (PAH4) is shown in Table 2 which gives an overview of samples that can be declared as potentially harmful to human health. The EU regulation no. 1881/2006, 835/2011 and XX/2019 [19–21], prescribes that the sum of PAH4 in traditional meat products should be lower than 30 µg/kg. According to this regulation, four samples analyzed in this survey (SS1, SS3, SS4, SS17, and SS18) exceed the prescribed values. Sample SS1 showed concurringly high concentration of PAH4, 99.26 µg/kg.

Twenty-two samples had acceptable values for PAH4 below 30 µg/kg and three samples had below—LOQ values for PAH4 SS13, SS28, and SS29. These samples can be considered safe for human consumption.

In Table 2 the sums of PAH16 are also displayed. They appear to be high. SS1 had the highest value for PAH16 462.83 µg/kg followed by SS2 with 316.78 µg/kg. However, no legal regulation for PAH16 in food commodities has been set so it is not possible to estimate if those concentrations are harmful to human health. Bogdanović et al. [33] reported lower levels for PAH4 in Croatian dry-cured hams. This is probably due to the fact that these samples were produced in industrial conditions. The samples that showed high PAH4 and BaP concentrations also have high PAH16 concentrations probably due to smoke intensity (duration and temperature). Prolonged smoking or smoking with an unsuitable wood type (oak, pine or other types of softwood) may contribute to elevated PAHs levels in meat products.

Distribution of PAHs in Slavonsla šunka samples according to the place of production is shown in Table 3. The highest concentration of PAH4 and PAH16 were detected in Vukovar–Srijem county who exhibited significantly higher PAH values in comparison to other counties. Individual PAHs concentrations were also detected in Vukovar-Srijem county, with the exception of Pyr. This is probably due to the fact that certain producers pay more attention to combustion conditions, such as distance from the fire, wood type or fire height.

Table 4 represents the PAH4 occurrence in different counties according to Chi-square (χ^2) test. The results of this test are in accordance with the results shown in Table 3. The results indicate that 50.0% of samples originating from Vukovar–Srijem county contains low concentrations of PAHs, which is, in comparison to other counties, the highest incidence. Moderate concentrations of PAHs are mostly found in Osijek–Baranja county, with the 46% incidence. This is a high incidence for moderate concentrations since other counties showed 23% (Brod–Posavina) and 31% (Vukovar-Srijem). The lowest incidence of moderately contaminated samples is found in Požega–Slavonija county, with 0%. Highly contaminated samples were evenly distributed in all counties with 29%. The exception was found in Brod–Posavina county that resulted in 14% of high contamination in samples.

Table 3. PAH contents (μg/kg) in different counties.

PAH	Brod-Posavina	Osijek-Baranja	Požega-Slavonia	Vukovar-Srijem
Nap	4.85 ± 8.40 [a]	0.89 ± 1.28 [a]	3.04 ± 4.32 [a]	1.57 ± 2.26 [a]
Anl	0.54 ± 0.93 [b]	4.51 ± 4.03 [b]	1.04 ± 1.09 [b]	16.26 ± 12.49 [a]
Ane	2.34 ± 1.88 [a,b]	5.88 ± 5.84 [a,b]	0.17 ± 0.25 [b]	6.28 ± 6.78 [a]
Flu	12.67 ± 1.23 [b]	19.93 ± 4.81 [b]	13.07 ± 3.90 [b]	28.57 ± 9.46 [a]
Ant	58.25 ± 3.46 [b]	71.13 ± 14.17 [b]	49.15 ± 13.15 [b]	136.45 ± 40.26 [a]
Phen	4.77 ± 1.33 [b]	6.47 ± 4.84 [b]	3.15 ± 1.04 [b]	19.09 ± 6.23 [a]
Flt	4.58 ± 0.73 [b]	5.52 ± 1.56 [b]	5.23 ± 1.17 [b]	14.80 ± 6.37 [a]
Pyr	3.09 ± 1.79 [a]	1.90 ± 2.73 [a]	1.39 ± 2.01 [a,b]	0.00 ± 0.00 [b]
BaA	17.78 ± 7.63 [b,c]	20.59 ± 12.04 [b]	5.82 ± 5.29 [c]	32.63 ± 11.26 [a]
Chry	0.00 ± 0.00 [a]	0.00 ± 0.00 [a]	0.00 ± 0.00 [a]	4.11 ± 13.00 [a]
PAH4	17.78 ± 7.63 [b]	20.59 ± 12.04 [b]	5.82 ± 5.29 [b]	36.74 ± 21.72 [a]
PAH16	108.87 ± 6.54 [b]	136.84 ± 23.60 [b]	82.05 ± 14.69 [b]	259.76 ± 74.24 [a]

Values are means ± SD. Values in the same row with different superscripts ([a–c]) are significantly different ($p < 0.05$).

Table 4. PAH4 (μg/kg) occurrence in different counties.

County	Low		Moderate		High			
	(*n*)	(%)	(*n*)	(%)	(*n*)	(%)	χ^2	*p*
Brod-Posavina	0	0	3	23.1	1	14.3	10.323	0.1117
Osijek-Baranja	1	10	6	46.2	2	28.6		
Požega-Slavonia	4	40	0	0	2	28.6		
Vukovar-Srijem	5	50	4	30.8	2	28.6		
	10	100	13	100	7	100		

PAH4 content was recorded into variables with three levels (Low = <12 μg/kg; Moderate = 12–30 μg/kg; High = >30 μg/kg).

The effect of different wood types on PAH content is shown in Table 5. It is visible that the samples smoked using elm tree for combustion exhibited higher values for PAHs than the samples smoked using beech and hornbeam.

Table 5. Effect of different types of wood on PAH content (μg/kg).

PAH	Beech and Hornbeam	Elm
Nap	2.41 ± 4.90 [a]	1.57 ± 2.26 [a]
Anl	2.58 ± 3.41 [b]	16.26 ± 12.49 [a]
Ane	3.33 ± 4.83 [a]	6.28 ± 6.78 [a]
Flu	16.24 ± 5.33 [b]	28.57 ± 9.46 [a]
Ant	61.48 ± 15.70 [b]	136.45 ± 40.26 [a]
Phen	5.06 ± 3.73 [b]	19.09 ± 6.23 [a]
Flt	5.23 ± 1.35 [b]	14.80 ± 6.37 [a]
Pyr	1.99 ± 2.42 [a]	0.00 ± 0.00 [b]
BaA	15.33 ± 11.52 [b]	32.63 ± 11.26 [a]
Chry	0.00 ± 0.00 [a]	4.11 ± 13.00 [a]
PAH4	15.33 ± 11.52 [b]	36.74 ± 21.72 [a]
PAH16	113.65 ± 30.26 [b]	259.76 ± 74.24 [a]

Values are means ± SD. Values in the same row with different superscripts ([a,b]) are significantly different ($p < 0.05$).

4. Conclusions

Light PAHs Nap, Anl, and Ane were detected in some of the samples and Nap were quantified in 11 samples of Slavonska šunka. Regarding heavy PAHs, only BaA was detected in 27 samples and Chry was quantified in only one sample. PAH4 concentrations were elevated (>30 µg/kg) in four samples (13.3%), meaning they are not in accordance with the EU regulation (EC No 835/2011 and EC No XX/2019). Even though the sum of PAH16 is not legally regulated, the occurrence of PAH16 seems to be closely related to PAH4 and BaP concentrations. As far as the site of production goes, the applied statistical analysis revealed that the highest concentrations of PAHs are found in samples from Vukovar-Srijem county. It is obvious that wood type has a big impact on PAHs occurrence and the samples smoked with elm showed significantly higher PAH concentrations.

PAHs concentration in traditional meat products needs to be reduced in the future and for that reason, the ALARA (as low as reasonably achievable) [21] principle was set in force in the EU. However, the legislative recommendations regarding the type of wood used for smoking, minimal heights for meat products hung in smokehouses during the exposure to the (open fire) smoking, ventilation regulations and casings used for filling that would help standardize smoking procedure should be issued in order to reduce the PAH content in traditional meat products.

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