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Traditional smoking of Wallachian cheeses and sausages in Polish and Slovak parts of the Carpathian Mountains

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Abstract

Wallachian expansion brought to the Carpathian Mountains a system of shepherd economy—farming, production of Wallachian cheeses, animals adapted to life in difficult mountain conditions—mainly sheep of the Cakiel breed group. Mountain sheep's milk is used to produce traditional cheeses: bundz, bryndza podhalańska, oscypek, redykołka—on the Polish side and Slovenská bryndza, Slovenská parenica, Slovenský oštiepok, Ovčí hrudkový syr salašnícky—on the Slovak side. Also sausages are made from sheep meat. These cheeses and sausages are salted and then traditionally smoked. The source of heat and smoke is hard wood with appropriate humidity, burned in the hearth located in the shepherd's hut, over which the cheeses are placed under the roof. Among several hundred smoke components, there are also polycyclic aromatic hydrocarbons, which, on the one hand, give the smoked product its taste and aroma, and on the other hand have carcinogenic and mutagenic properties. Sausages and cheeses from Poland and Slovakia, made from milk and meat of native sheep breeds, preserved by traditional smoking, were analyzed for polycyclic aromatic hydrocarbons content. The analyzed cheeses were characterized by a trace or low content of benzo[a]pyrene and the sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b) fluoranthene and chrysene. In cheeses from outer Eastern Carpathians subprovince (Bieszczady), the high content of naphthalene, acenaphthylene, fluorene and phenanthrene is noteworthy. Polish lamb sausages were characterized by a higher content of benzo(a)pyrene and the sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene. The differences result from the method of smoking (warm or cold in the south of the Carpathians—hot in the north) and the type of wood used for smoking.

Keywords Polish and Slovak Carpathian, Wallachian culture, Traditionally smoked cheeses and meats, Polycyclic aromatic hydrocarbons

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Introduction

Wallachian nation

In the Medieval Ages, the region of the Carpathian Mountains was sparsely populated and the shepherding was the main form of economy performed by Carpathian Mountaineers. At the beginning of thirteenth century, Wallachian shepherds started to move from Balkan Mountains to the Carpathian Mountains. The oldest description of Wallachs is, known from seventh century Greek word "Vlah" (Vlachoi–plural), which was adopted by Southern Slavs in not changed form as Vlah, Vlahi, Vlasi, Vlachie, Vleh [1, 2]. Colloquially in the South Slavs (Serbia, Croatia), the word "Vlach" was describing all sheep shepherds. The wars with Byzantium and connected to them repressions against peasants caused that the nomadic population, occupied with shepherding (Vlahs) started to move with their herds of sheep and goats, to the north, along the Carpathian ridge, to find new places for pasturing and free living. The Ruthenian population named the wandering shepherds Wallachians. The Wallachians were Eastern Rite Christians, and it is rather difficult to recognize them as a national group or nation. They were rather a professional group engaged in mountain shepherding of the so-called small cattle, i.e., sheep, goats and, less often, pigs [3, 4].

Wallachian culture

The Vlah shepherds used for their herds Carpathian forests, halls, glades and where they met with the local inhabitants Ruthenians, Poles, Slavaks. The settled tribes accepted vocabulary, rituals culture and also the mountain economy shepherding system called hut or summer mountain slopes shepherding. At the beginning of thirteenth century, numerous groups of Vlahs settled onto Polish lands mainly on Red Ruthenia, Podolia, Lublin region accepting local culture, but staying involved in shepherding and animal breeding [1, 5, 6]. Then, nomadic groups of Vlachs—shepherds headed west (along the arc of the Carpathians), grazing their flocks of sheep and goats, leading a semi-nomadic lifestyle. When the grass for animals in the pastures and pastures ran out, they wandered further west [7]. In the sixteenth century, there was a new wave of Wallachian settlement and colonization under Wallachian law in the Polish part of the Carpathians, which resulted in the location of new settlements under Wallachian law located in the so-called Muszyna key in the Beskid Sądecki and to the location of villages in Skalne Podhale and the Silesian Beskids [8]. Wallachian settlement in Slovakia proceeded from two directions—eastern corridor (Maramara-Zakarpattia) and the southern corridor from the Balkans through the Hungarian Plain to the Gemeru Mountains. In the sixteenth century, another wave of settlements took place, as a result of which empty settlements were settled and older settlements were re-settled (Upper Orava) [9]. The most recently occupied by Wallachian settlements located in the territory of Czech Moravia and covering, among others, Silesian-Moravian Beskids. Wallachian colonization took place here from the mid-sixteenth and seventeenth centuries and was mainly based on migration from the nearby Cieszyn Silesia [10].

The Wallachian expansion brought to the Carpathian Mountains a system of shepherd economy—farming, production of Wallachian cheeses, animals adapted to life in difficult mountain conditions—mainly sheep of the Cakiel breed group (Vlachian sheep—Valaška, Transylvania; Podhale, Raczka, Curkana, Cigája), goats (Carpathian goat), cattle (red cattle), horses (Hutsul horse), breeding nomenclature, specific dialectal terms, nomenclature of places in the mountains, names of places located under Wallachian law, characteristic architecture and clothing, and simple cuisine [4, 11, 12]. During lactation lasting about 150 days, mountain sheep provide about 60–70 L of milk [13], while the Carpathian goat gives 350 to 498 L of milk during 230–260 days of milking. Mountain sheep's milk is used to produce traditional cheeses: bundz, bryndza podhalańska, oscypek, redykołka—on the Polish side and Slovenská bryndza, Slovenská parenica, Slovenský oštiepok, Ovčí hrudkový syr salašnícky—on the Slovak side. The word "bryndza" comes from the Wallachian language (Romanian brânză "cheese"). Other cheeses are made from bundz, which can be smoked to preserve them; sheep cheese, redykołka, Slovenská bryndza, Slovenská parenica, Slovenský oštiepok, Ovčí hrudkový syr salašnícky. These cheeses are very old products of Wallachian shepherds grazing their sheep in mountain glades. They came to Podhale together with the entire Wallachian culture, the organization of grazing, the way of running a shepherd's hut, processing milk. Although the world's oldest evidence of cheese production was found in Kujawy (according to archeological research, cheese was produced here as early as 7,500 years ago) [14], the cradle of Polish cheesemaking is the Carpathians, where cheesemaking developed in Poland as early as the eleventh and twelfth centuries. Due to the seasonality and short period of milk production, Wallachian shepherds used simple methods of cheese preservation (maturing, salting and smoking. Wallachians and then shepherd communities took special care of fire (watra, vatra) and water. The so-called slanice, salty waters, which provided the grazing animals with salt and were used to soak the oscypek cheese in salt water, the so-called "rosolenie", were also of exceptional importance. to this day—both "cold water" and "slanice" have their permanent places on the map of the "route of the Carpathian pastoral culture". The fire, continuously burning zawaternik (zawaternik,

kłotek—a block of wood, most often spruce, put into the watra so that it would not go out overnight), which was traditionally supervised by a shepherd, was lit on the first day of stay in the hall. During cold smoking, which lasts for several days, the cheeses are placed on shelves under the roof of the shepherd's hut. Shepherd's hut is a characteristic wooden building standing in the pastures, where for several months a year (during grazing) there is a shepherd with juhas. It consists of two parts—a working one where the cheese is produced and a residential one. In the working part, the most important place is occupied by a watra, i.e., a bonfire. It is covered with flat stones taken from the stream. Cheese is smoked using the smoke from the watra in which alder or beech wood is burned. Above the hearth there is a pole called "jadwiga or rozwodnica" on which a cast-iron boiler (kotlik, kocieł) is hung, used to heat sheep's milk and brew entica.

During smoking, the smoke components give the characteristic golden color of oscypek or ostiepek and preserve the product. Smoke consists of several hundred ingredients, both positively affecting the quality of the smoked product, indifferent to the consumer's health, and compounds that raise doubts in terms of health. Some smoke compounds have bacteriostatic and antioxidant effects and may act as preservatives. Formaldehyde from smoke, in reaction with proteins, reduces the digestibility of products that are smoked too much. Due to their properties, phenols play an important role in smoking, as they have a specific smell and shape the sensory properties of smoked products. In addition, they have an antioxidant effect. Among several hundred smoke components, there are also polycyclic aromatic hydrocarbons, which, on the one hand, give the smoked product its taste and aroma, and on the other hand have carcinogenic and mutagenic properties. The result of many years of research on the harmfulness of PAHs was the recognition in 2002 by the Scientific Committee on Food of the European Commission of 15 compounds from the PAH group (benz[a] anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[g,h,i]perylene, benzo[a] pyrene, chrysene, cyclopenta[c,d]pyrene, dibenz[a,h] anthracene, dibenzo[a,e]pyrene, dibenzo[a,h]pyrene, dibenzo[a,i]pyrene, dibenzo[a,l]pyrene, indeno[1,2,3-cd] pyrene and 5-methylchrysene) are potentially genotoxic and carcinogenic to humans. The Scientific Committee on Food (SCF) suggested using benzo[a]pyrene as a marker of occurrence and effect of the carcinogenic PAHs in food, based on examinations of PAH profiles in food and on evaluation of a carcinogenicity studies in animals [15, 16]. The result is a number of Commission Regulations (EU) aimed at limiting the presence of PAHs in food. Today there are no regulations about smoked cheese and their maximum PAH concentrations.

Aim of this study was analysis of traditional smoked cheese in which process source of smoke and heat is burning of different type of wood on level of PAHs and safety for consumers. For sausages, we want to find if differences in smoking method between Carpathians provinces like using hot smoke in Poland and cold smoke in Slovakia fulfill Commission Regulation (EC) No. 1881/2006.

Material and methods

The analyzed cheeses were produced in the summer in small individual farms. All investigated cheeses were rennet-curd and produced from raw caprine or ovine milk. As smoked cheese and sausages are typical products for regions inhibited by Wallachians, we collected samples of most popular cheeses and sausage for analysis.

Cheeses were smoked with warm smoke from burning pieces of alder, beech, willow, spruce or pine wood.

Characteristic of analyzed cheeses and sausages

Bundz was analyzed—goat's smoked cheese, goat's smoked cheese "Vlach's type and sheeps smoked cheeses from outer Eastern Carpathians subprovince (Bieszczady), sheep cheeses smoked in a traditional way (oscypki, redykołki, bundz) from Outer Western Carpathians subprovince (Poland) and sheep cheeses smoked in a traditional way (bundz, slovensky oštiepok, slovenská parenica) from Outer Western Carpathians subprovince (Slovakia). Even both side of Carpathians were settled by Wallachians, there are differences in processing milk (smoking temperature and way of storage of cheese in sheepherd's huts) and additives used in sausage production (garlic and red pepper in north part—Poland and red pepper in south part—Slovakia). Carpathians were a line that differ smoking method of cheese—Poland direct smoking using hot smoke and Slovakia—indirect smoking using warm/cold smoke. There are also differences in smoking—in Poland cheese rested on the characteristic shelves under the roof and is smoked while in Slovakia cheese were in leather yokes while warm/cold smoke from fire placed in central part of shepherd's hut were smoking cheese.

All production stages are carried out manually with traditional highlander equipment. After milking the sheep, the milk is brought to a wooden hut with a stone-covered fireplace, where the bonfire is burning, necessary for smoking cheese.

Bundz is a sheep's not matured rennet type cheese, it is the first product made of clotted milk right after milking. Mostly made in Podhale region and western part of Podbeskidzie. Cheese curd if steamed for few minutes in 70 °C [17].

Cheeses from Bieszczady are maturated rennet type cheese similar to balkan type cheese.

The Slovensky oštiepok (PGI-SK-0549 26/09/2008 COMMISSION REGULATION (EC) No. 943/2008 of 25 September 2008) is a semi-hard and semi-fat cheese, which can be submitted to a smoking or steaming procedure. It is produced from sheep's milk (of the races Valaska, Zoslachtená Valaska and Cigájao), from a mixture of sheep's milk with cows' milk, or even only with cows' milk [18], weight between 300 and 800 g. Smoking process: cheeses were hanged in leather yokes close to roof of sheepherd's hut and smoked using cold smoke from beech wood for about 6 days.

The Slovenská parenica (PGI-SK-0485 11/07/2008 COMMISSION REGULATION (EC) No. 656/2008 of 10 July 2008) is a traditional Slovak Cheese, a soft, steamed, lightly smoked cheese wound into two rolls 6–8 cm in diameter and 5–8 cm high, connected in an 'S'-shape. The rolls are bound with cheese string or chain with weight about 450–500 g [19]. Parenica is a semi-firm, non-ripening, semi-fat, steamed and usually smoked cheese, although a non-smoked version is also produced. It is made of unprocessed sheep's milk from grazing sheep of the Wallachian, improved Wallachian, Cigaya and East Friesian breeds or a mixture of fresh raw, unprocessed sheep's milk and fresh raw, unprocessed cow's milk, containing at least 50% sheep's milk. Smoking process: cheese's ribbon is tied by another cheese "thread" or "chain" and placed on wooden desk. After drying parenica is smoked about 2 h using burning pieces of beech wood.

Oscypek (PDO-PL-0451 COMMISSION REGULATION (EC) No. 510/2006 of 2 September 2006) is a smoked sheep cheese made in Podhale. It has shape of a double cone or spindle. It is between 17 and 23 cm in length, between 6 and 10 cm in width at its widest point and weighs between 600 and 800 g. When cut it has a light cream color, darker at the rind, but a shade closer to white is also permissible. It is made by hand from the sheep's milk of Polish Mountain sheep, sometimes with a small admixture of milk from the Polish Red cow and produced from May to September. Smoking is carried out using cold smoke and lasts between 3 and 7 days. [20]. Smoking process: brining and smoking from 4 to 7 days, cold smoking in temperature about 30 °C from burning pieces of spruce or pine.

Redykołka (PDO-PL-0588 COMMISSION REGULATION (EC) No. 1176/2009 of 30 November 2009) is a small cheese (30–60 g) made of unpasteurized milk of the Polish Mountain sheep. The addition of cow's milk from Polish Red cow breed is allowed, however, not exceeding 40 percent of the total milk weight. Redykołka is produced in the period from May to September. The manufacturing process resembles the production of oscypek (redykołka is also produced in the same area), except that the cheese is formed into other shapes and smoked for 3–7 days [21]. Smoking process: cold smoking in temperature about 30 °C from burning pieces of spruce or pine. Because of size of redykołka (about 30 g) they are placed on special wooden racks in upper part of shepherd's hut or on the wall next to fire place.

In addition, sheep meat sausages produced by a traditional method and traditionally smoked in Slovakia and Poland were analyzed. The sausage is a medium-minced meat product made of lamb meat and fat, stuffed into natural small intestine casings. The raw material was ground in a meat grinder using 10-mm screens. The natural casings (20–30 mm in diameter) were filled with a stuffer and left for about 2 h to dry the casing surface and deposit the ingredients. It also contains salt and usually pepper and garlic (Poland) or pepper, and red pepper (Slovakia). After stuffing, the sausage is hung at 20–30 °C and then undergoes smoking. The parameters of this process vary slightly among different producers; however, typically the surface is dried at 40–60 °C in low density smoke as the first phase, followed by smoking at 45–60 °C, with the smoke density ranging from medium to very high (Slovak). In Poland the final phase, which is conducted at 75–85 °C in low density smoke, is intended to denature the proteins in the stuffing. At the end of the smoking process, sausages are cooled in cold air for several minutes to lower their internal temperature to about 40 °C. After manufacturing, sausages were chilled to 6 ± 1 °C.

All products mentioned above are hand-made from centuries (and non-commercial) in shepherd's huts using traditional technology with hand-made processing of clotted milk.

Sample collections

Cheeses were bought from producers in sheepherd's huts in duration between May and June.. Cheeses were bought in weekly intervals just after production process finished. From each analyzed cheese were collected 5 repetitions (each 1 kg weight). Sausages were bought in weekly intervals directly from producers just after production process finished. Each sample of sausage weight 1 kg in total was collected from different parts of smokehouse. Four repetitions of each sausage were collected.

The samples collected were packed in aluminum foil and handled in accordance with the Commission regulation (EU) 836/2011. All samples were transported in refrigerated conditions temperature 4 °C. [22]. Every repetition of cheese and sausage (number of cuts in aggregative sample were between 3 and 20) was sent to analysis next day after collection.

The collected cheese and sausage samples were crushed (number of cuts for each cheese and sausage were between 3 and 20). Crushed samples were stored in 4 °C overnight and next day send to analysis every week.

CIE-L*a*b* color parameters

Color measurements were taken using a Minolta CM-600d spectrophotometer (Konica Minolta, Osaka, Japan) in the CIE-LAB space Commission Internationale del'Eclairage, 1976 [23] with illuminant D65, 10° standard observer and an aperture diameter of 4 mm. Lightness (L*), redness (a*), and yellowness (b*) were recorded. CIE-L*a*b* color parameters of sheep cheeses were measured on surface and in middle (on the cross-section of the cheese). CIE-L*a*b* color parameters of sausage were measured on a cross-section of the sausage.

Polycyclic aromatic hydrocarbons (PAHs)

The determination of PAHs included 8 from 15 compounds recognized in 2002 by The Scientific Committee on Food as potentially genotoxic and carcinogenic to humans (benz(a)anthracene, chrysene, benzo(b) fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, dibenz(a,h)anthracene, benzo(g,h,i)perylene), as well as naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene and pyrene. The analysis was carried out by ALS Czech Republic, s.r.o. (Certificate of accreditation no. 128/2017) on request of ALS FOOD & PHARMACEUTICAL POLSKA Sp. z o. o. (Method CZ_SOP_D06_06_180 except chap. 10.3.3.1–10.3.3.8 (US EPA 429, STN EN 16619). PAHs in cheese and sausage samples were determined using the isotope dilution method with the use of HR-GC-MS. The procedure of determination of PAHs fulfilled the conditions and the requirements of the US EPA Method 429 (1997) and also met the requirements and the criteria of ISO 11338–1:2003, ISO 11338–2:2003 and the Commission regulation (EU) 836 /2011.

Analysis included spiking the sample with $^{2}H_{x}$-labeled extraction standards, specific extraction method by matrix, cleanup of the raw extract and HR-GC-MS measurement after addition of $^{2}H_{x}$-labeled injection standard.

Extraction

10–25 g of weighed sample was spiked with extraction standard containing $^{2}H_{x}$-labeled PAH congeners (d8-naphthalene, d8-acenaphthylene, d10-phenanthrene, d10-anthracene, d10-fluoranthene, d12-benz[a] anthracene, d12-chrysene, d12-benzo[b]fluoranthene, d12-benzo[a]pyrene, d12-indeno[1,2,3-c,d]pyrene, d14-dibenz[a,h]anthracene, d12-benzo[g,h,i]perylene; Wellington Laboratories Inc.). The spiked sample was mixed with hydromatrix (Agilent Technologies) and transferred into extraction thimble. The extraction with toluene in Soxhlet extractor took 20 h. The toluene extract was concentrated in the rotary vacuum evaporator and then re-dissolved in hexane. The raw extract was pre-cleaned by shaking with 70% sulfuric acid at laboratory temperature.

Cleanup

Pre-cleaned hexane extract was transferred onto the top of a multilayer silica-gel column. The column was prepared by ALS Czech Republic, s.r.o. and packed from bottom to top: silica, silver nitrate silica, basic silica, silica and anhydrous sodium sulfate. The extract was eluted first by hexane and then by DCM/hexane (2:1, v/v). The hexane fraction was discarded, the second fraction was concentrated using the modified Kuderna–Danish concentrator (made by ALS Czech Republic, s.r.o.) up to 0.5–1 cm^3.

Preparation before injection

The final extract containing PAHs was spiked with $^{2}H_{x}$-labeled injection standard (d10-acenaphthene, d10-pyrene, d12-benzo[e]pyrene; Wellington Laboratories Inc.). Finally, 2–4 µL was injected to HR-GC–MS (Finnigan MAT 95XP/Agilent Technologies GC 6890N).

HR-GC–MS analysis

The chromatographic conditions of the HR-GC: column type: Rxi -17MS (30 m × 0.25 mm, film 0.25 µm; Restek), injector temperature: 270 °C, transfer line: 280 °C, splitless time: 2 min, carrier gas (helium) flow rate: 1.0 mL/min, oven temperature program: 85 °C (1 min), rate 1: 4.3 °C/min to 320 °C (maintained at this level for 5.5 min), rate 2: 10 °C/min to 340 °C (maintained at this level for 9 min). The working conditions of the HRMS: Resolution: ≥ 8000; EI ionization; ion source temperature 280 °C, MID (Multiple Ion Detection) mode.

Limits of quantification LOQ and limits of detection (LOD) for this method are:

Polycyclic aromatic Hydrocarbons	Limit of quantification LOQ (µg/kg)	Limit of detection LOD (µg/kg)
Naphthalene	20.0	0.0033
Acenaphthylene	2.0	0.0022
Acenaphthene	1.0	0.0044

Polycyclic aromatic Hydrocarbons	Limit of quantification LOQ (μg/kg)	Limit of detection LOD (μg/kg)
Fluorene	2.5	0.008
Phenanthrene	5.0	0.0027
Anthracene	0.3	0.0033
Fluoranthene	2.5	0.0027
Pyrene	1.5	0.0027
Benzo(a)anthracene	0.9	0.0051
Chrysene	0.9	0.0069
Benzo(b)fluoranthene	0.9	0.012
Benzo(k)fluoranthene	1.0	0.015
Benzo(a)pyrene	0.9	0.0099
Indeno(1,2,3-cd)pyrene	1.0	0.0076
Dibenzo(a,h)anthracene	1.0	0.01
Benzo(g,h,i)perylene	1.0	0.0092

Results and discussion

Tables 1, 2, 3 show PAH levels (μg/kg) and CIE-L*a*b* color parameters traditionally smoked cheeses from Polish and Slovak Carpathians, while Table 4 shows PAH levels (μg/kg) and CIE-L* a*b* color parameters sheep traditionally smoked sausages from Polish and Slovak Carpathians.

The analyzed cheeses were characterized by a varied level of polycyclic aromatic hydrocarbons. These cheeses were smoked with warm smoke resulting from burning pieces of alder, beech, willow, spruce or pine wood. The highest amount of benzo(a)pyrene was found in redykołki, small sheep smoked cheese—4.50 ± 1.35 μg/kg (Table 2). In this cheese, the highest content of the sum of 4 PAHs (benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene) was also found—24.90 ± 2.58 μg/kg. Redykołki are small cheeses—they

Table 1 The PAH levels (μg/kg) and CIE-L*a*b* color parameters cheeses smoked in a traditional way from Outer Eastern Carpathians subprovince

Polycyclic aromatic Hydrocarbons	Outer Eastern Carpathians subprovince (Bieszczady)			
	Bundz—goat's smoked cheese, not matured rennet type cheese	Goat's smoked cheese Vlach's type cheese	Sheep smoked cheese	Sheep smoked cheese
Naphthalene	210 ± 63.0	970 ± 291	940 ± 282	580 ± 174
Acenaphthylene	200 ± 60.0	840 ± 252	450 ± 135	350 ± 105
Acenaphthene	9.30 ± 2.79	48.0 ± 14.4	28.0 ± 8.40	130 ± 39.0
Fluorene	100 ± 30.0	140 ± 42.0	120 ± 36.0	110 ± 33.0
Phenanthrene	120 ± 36.0	330 ± 99.0	140 ± 42.0	85 ± 25.5
Anthracene	32.0 ± 9.60	87.0 ± 26.1	37.0 ± 11.1	46.0 ± 13.8
Fluoranthene	7.30 ± 2.19	20.0 ± 6.00	17.0 ± 5.10	12.0 ± 3.60
Pyrene	6.00 ± 1.80	16.0 ± 4.80	12.0 ± 3.60	10.0 ± 3.00
Benzo(a)anthracene	< 0.80	1.70 ± 0.51	1.60 ± 0.48	1.46 ± 0.44
Chrysene	< 0.80	1.60 ± 0.48	1.70 ± 0.51	1.50 ± 0.45
Benzo(b)fluoranthene	< 0.80	0.97 ± 0.291	< 0.75	< 0.66
Benzo(k)fluoranthene	< 0.74	< 0.59	< 0.63	< 0.56
Benzo(a)pyrene	< 0.80	0.85 ± 0.255	< 0.88	< 0.75
Indeno(1,2,3-cd)pyrene	< 0.80	0.78 ± 0.234	< 0.65	< 0.46
Dibenzo(a,h)anthracene	< 0.14	< 0.41	< 0.25	< 0.24
Benzo(g,h,i)perylene	< 0.80	0.81 ± 0.243	< 0.58	< 0.56
Sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene	< 0.80	5.12 ± 1.54	3.30 ± 0.99	2.96 ± 0.88
CIE-L*a*b* color parameters (surface)				
Lightness L*	62.44 ± 4.11	68.25 ± 3.98	69.42 ± 2.63	72.28 ± 1.72
Redness a*	21.64 ± 3.04	16.83 ± 2.61	16.83 ± 2.71	13.85 ± 2.05
Yellowness b*	46.41 ± 3.73	44.25 ± 3.35	41.58 ± 5.14	34.11 ± 2.41
CIE-L*a*b* color parameters (middle)				
Lightness L*	84.97 ± 1.64	89.25 ± 1.41	90.11 ± 0.44	86.45 ± 0.68
Redness a*	2,54 ± 0.31	3.34 ± 0.24	4.82 ± 0.20	5.74 ± 0.12
Yellowness b*	16.42 ± 0.96	22.11 ± 0.69	25.28 ± 0.97	28.17 ± 0.65

Table 2 The PAH levels (μg/kg) and CIE-L*a*b* color parameters sheep cheeses smoked in a traditional way from Outer Western Carpathians subprovince (Poland)

Polycyclic aromatic hydrocarbons	Outer Western Carpathians subprovince (Poland)					
	Oscypek Sheep smoked cheese Koniaków	Oscypek Sheep smoked cheese Leśnica	Oscypek Sheep smoked cheese Nowy Targ	Sheep smoked cheese Słopnice	Redykołki Small sheep smoked cheese	Bundz smoked cheese, not matured rennet type cheese Koniaków
Naphthalene	54.0 ± 16.2	70.0 ± 21.0	81.0 ± 24.3	1300 ± 390	1200 ± 360	< 18
Acenaphthylene	130 ± 39.0	96.0 ± 28.8	80.0 ± 24.0	220 ± 66.0	1200 ± 360	< 2
Acenaphthene	3.70 ± 1.11	4.80 ± 1.44	7.0 ± 2.10	23.0 ± 6.90	38.0 ± 11.4	1.8 ± 1.0
Fluorene	50.0 ± 15.0	31.0 ± 9.30	84.0 ± 25.2	61.0 ± 18.3	400 ± 120	11.0 ± 2.50
Phenanthrene	170 ± 51.0	60.0 ± 18.0	110 ± 33.0	760 ± 228	790 ± 237	13.00 ± 4.00
Anthracene	48.0 ± 14.4	14.0 ± 4.20	35.0 ± 10.5	100 ± 30.0	190 ± 57.0	2.00 ± 0.50
Fluoranthene	19.0 ± 5.70	6.50 ± 1.95	22.0 ± 6.6	45.0 ± 13.5	94.0 ± 28.2	< 2.5
Pyrene	14.0 ± 4.20	4.40 ± 1.32	21.0 ± 6.3	1.80 ± 0.54	67.0 ± 20.1	2.30 ± 1.50
Benzo(a)anthracene	< 0.85	< 0.61	1.1 ± 0.33	1.70 ± 0.51	8.60 ± 2.58	< 0.50
Chrysene	< 0.82	< 0.61	1.2 ± 0.36	2.60 ± 0.78	7.30 ± 2.19	< 0.75
Benzo(b)fluoranthene	< 0.82	< 0.61	< 0.85	1.10 ± 0.33	4.50 ± 1.35	< 0.50
Benzo(k)fluoranthene	< 0.82	< 0.54	< 0.85	< 0.73	2.10 ± 0.63	< 0.50
Benzo(a)pyrene	< 0.82	< 0.61	< 0.85	0.74 ± 0.22	4.50 ± 1.35	< 0.50
Indeno(1,2,3-cd) pyrene	< 0.82	< 0.61	< 0.50	< 0.48	2.30 ± 0.69	< 0.50
Dibenzo(a,h)anthracene	< 0.39	< 0.067	< 0.50	< 0.48	< 0.61	< 0.50
Benzo(g,h,i)perylene	< 0.82	< 0.61	< 0.50	0.74 ± 0.22	1.90 ± 0.57	< 0.50
Sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b) fluoranthene and chrysene	< 0.85	< 0.61	2.30 ± 0.69	6.14 ± 0.78	24.90 ± 2.58	< 0.75
CIE-L*a*b* color parameters (surface)						
Lightness L*	78.32 ± 1.32	80.12 ± 1.63	76.64 ± 0.98	82.11 ± 1.12	69.45 ± 7.41	70.43 ± 6.12
Redness a*	11.46 ± 1.02	12.05 ± 0.86	14.16 ± 1.06	10.84 ± 2,15	17.52 ± 4.18	20.42 ± 3.24
Yellowness b*	34.86 ± 0.96	36.23 ± 1.24	32.82 ± 1.11	31.80 ± 0.88	41.32 ± 5.04	42.56 ± 4.11
CIE-L*a*b* color parameters (middle)						
Lightness L*	72.27 ± 1.10	77.32 ± 1.35	71.78 ± 0.98	88.97 ± 1.12	77.43 ± 2.85	90.2 ± 1.38
Redness a*	4.65 ± 0.29	3.88 ± 0.18	4.17 ± 0.24	2.10 ± 0.13	4.17 ± 0.39	2,92 ± 0.16
Yellowness b*	31.97 ± 0.75	23.80 ± 0.82	30.83 ± 0.68	23.07 ± 0.41	23.98 ± 1.23	22.5 ± 0.997

weigh from 30 to 60 g; therefore, the large surface area to mass means that more compounds are deposited in these cheeses during smoking. Cheeses smoked slowly over several days with warm smoke in shepherd's huts (oscypek, slovensky oštiepok and slovenská parenica) were characterized by low levels of benzo(a)pyran and sum 4 PAHs. Michalski and Germuska [24] found in Slovak cheeses from < 0.1 to 3.8 μg/kg of benzo[a]pyran depending on the time (from 30 to 180 min) and smoking temperature (from 13 up to 30 °C) higher levels of PAHs were found in home-smoked cheeses. In addition, they showed that the surface of smoked cheeses (rind) is three to six times more contaminated with PAH compared to the entire sample. Removal of the skin reduced the total PAH content by approximately 50–100%. The content of PAHs in smoked cheeses depends on the type of wood used for smoking [25], straw, wood shavings [26]. In "mozzarella di bufala campana" cheeses smoked with burnt straw, Anastasio et al. [26] found from 0.38 to 2.12 μg/kg of benzo[a]pyran, while in "mozzarella di bufala campana" cheeses smoked with wood chips and liquid smoke preparation from 0.19 to 0.80 μg/kg and from 0.18 to 0.50 μg/

Table 3 The PAH levels (μg/kg) and CIE-L*a*b* color parameters sheep cheeses smoked in a traditional way from Outer Western Carpathians subprovince (Slovakia)

Polycyclic aromatic Hydrocarbons	Outer Western Carpathians subprovince (Slovakia)				
	Slovensky oštiepok Sheep smoked cheese Zázrivá	Sheep smoked cheese Revišné	Sheep smoked cheese Zázrivá	Slovenská Parenica semi-firm, non-ripening, semi-fat, steamed and smoked cheese	Bundz smoked cheese, not matured rennet type cheese Oravská Polhora
Naphthalene	78.0 ± 23.4	200 ± 60.0	72.0 ± 21.60	37.0 ± 11.10	140 ± 42.0
Acenaphthylene	9.80 ± 2.94	61.0 ± 18.30	5.00 ± 1.50	9.60 ± 2.88	94.0 ± 28.2
Acenaphthene	3.00 ± 0.90	37.0 ± 11.10	4.20 ± 1.26	3.30 ± 0.99	26.0 ± 7.80
Fluorene	35.0 ± 10.50	100 ± 30.00	59.0 ± 17.7	21.0 ± 6.30	360 ± 108.0
Phenanthrene	41.0 ± 12.30	56.0 ± 16.80	31.0 ± 9.30	23.0 ± 6.90	300 ± 90.0
Anthracene	7.20 ± 2.16	16.0 ± 4.80	7.10 ± 2.13	3.50 ± 1.05	65.0 ± 19.20
Fluoranthene	5.90 ± 1.79	6.30 ± 1.89	3.90 ± 1.17	4.10 ± 1.23	38.0 ± 11.40
Pyrene	5.10 ± 1.53	14.0 ± 4.20	3.40 ± 1.02	4.10 ± 1.23	30.0 ± 9.0
Benzo(a)anthracene	< 0.50	1.50 ± 0.45	< 0.50	< 0.85	2.30 ± 0.69
Chrysene	< 0.90	1.60 ± 0.48	< 0.90	< 0.85	1.80 ± 0.54
Benzo(b)fluoranthene	< 0.50	< 0.90	< 0.50	< 0.47	1.50 ± 0.45
Benzo(k)fluoranthene	< 0.50	< 0.90	< 0.50	< 0.47	1.20 ± 0.36
Benzo(a)pyrene	< 0.50	< 0.90	< 0.50	< 0.47	1.10 ± 0.33
Indeno(1,2,3-cd)pyrene	< 0.50	< 0.50	< 0.50	< 0.47	< 0.48
Dibenzo(a,h)anthracene	< 0.50	< 0.50	< 0.50	< 0.47	< 0.48
Benzo(g,h,i)perylene	< 0.50	< 0.90	< 0.90	< 0.47	< 0.48
Sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene	< 0.90	3.10 ± 0.93	< 0.90	< 0.85	6.70 ± 2.01
CIE-L*a*b* color parameters (surface)					
Lightness L*	72.54 ± 0.88	78.25 ± 10.51	76.61 ± 5.20	80.20 ± 8.24	54.59 ± 3.68
Redness a*	14.30 ± 1.06	12.58 ± 5.44	13.11 ± 2.23	10.86 ± 3.12	21.14 ± 1.38
Yellowness b*	37.06 ± 1.23	34.62 ± 7.64	35.47 ± 4.33	25.46 ± 5.23	47.69 ± 2.75
CIE-L*a*b* color parameters (middle)					
Lightness L*	79.75 ± 5.02	91.31 ± 0.09	88.22 ± 1.02	59.83 ± 6.23	84.92 ± 1.89
Redness a*	1.74 ± 0.16	2.63 ± 0.03	2.33 ± 0.12	20.59 ± 2.35	2.72 ± 0.43
Yellowness b*	14.67 ± 0.46	16.98 ± 0.36	15.53 ± 0.78	47.59 ± 1.97	21.19 ± 1.37

kg, respectively. Pagliuca et al. [27] in the "Diavoletto" cheese produced in the Sorrento peninsula and smoked with various plant-derived materials; they found from 0.12 to 6.21 μg of PAHs/kg. Cheeses from Outer Eastern Carpathians subprovince (Bieszczady) were characterized by a darker surface (crust) and higher values of redness and yellowness (Table 1). Bundz—smoked cheese from Oravská Polhora—was characterized by similar features. Cheeses from the Outer Western Carpathians subprovince from both Poland and Slovakia were characterized by a lighter (straw) color of the surface and lower values of redness and yellowness. This is due to a different way of smoking in shepherd's huts. In the halls, the cheeses are aged on shelves at the roof of the shepherd's hut, over a slowly smoldering watra (bonfire) in which spruce or pine wood is burned, and they are smoked (smoked) with warm smoke for several days. Fast (several hours) smoking in a special smokehouse gives a product with a darker surface (skin). Because smoked cheeses are particularly appreciated by consumers due to their unique taste and smell [28] and are popular among consumers, research is conducted on their safety, especially on the content of polycyclic aromatic hydrocarbons. However, taking into account the high solubility of PAHs in fats and the high fat content in cheese, these impurities may be retained in the product during the smoking process [29].

Table 4 The PAH levels (ug/kg) and CIE-L*a*b* color parameters sheep sausages smoked in a traditional way

Polycyclic aromatic hydrocarbons	Poland		Slovakia	
	Sheep sausage	Sheep sausage Koniaków	Sheep sausage	homemade sheep sausage
Naphthalene	120 ± 36.0	84.0 ± 25.20	530 ± 159.0	110 ± 33.0
Acenaphthylene	160 ± 48.0	48.0 ± 14.40	480 ± 144.0	20.0 ± 6.00
Acenaphthene	4.20 ± 1.26	4.10 ± 1.23	56.0 ± 16.8	6.70 ± 2.01
Fluorene	35.0 ± 10.50	57.0 ± 17.10	730 ± 219.0	91.0 ± 27.30
Phenanthrene	160 ± 48.0	120 ± 36.00	650 ± 195.0	41.0 ± 12.30
Anthracene	34.0 ± 10.20	24.0 ± 7.20	130 ± 39.0	9.50 ± 2.85
Fluoranthene	78.0 ± 23.40	76.0 ± 22.80	78.0 ± 23.40	4.60 ± 1.38
Pyrene	71.0 ± 21.30	68.0 ± 20.40	77.0 ± 23.10	4.70 ± 1.41
Benzo(a)anthracene	11.0 ± 3.30	7.90 ± 2.37	3.60 ± 1.08	< 0.47
Chrysene	11.0 ± 3.30	8.30 ± 2.49	3.30 ± 0.99	< 0.85
Benzo(b)fluoranthene	7.00 ± 2.10	6.20 ± 1.86	2.50 ± 0.75	< 0.47
Benzo(k)fluoranthene	4.40 ± 1.32	4.30 ± 1.29	2.60 ± 0.78	< 0.47
Benzo(a)pyrene	5.10 ± 1.53	4.50 ± 1.35	1.90 ± 0.57	< 0.47
Indeno(1,2,3-cd)pyrene	3.10 ± 0.93	1.70 ± 0.51	< 0.97	< 0.47
Dibenzo(a,h)anthracene	< 0.72	< 0.50	< 0.49	< 0.40
Benzo(g,h,i)perylene	3.20 ± 0.96	2.20 ± 0.66	1.20 ± 0.36	< 0.47
Sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene	34.10 ± 3.30	26.90 ± 8.07	11.30 ± 3.39	< 0.85
CIE-L*a*b* color parameters				
Lightness L*	57.03 ± 3.15	46.86 ± 3.54	43.04 ± 5.27	38.66 ± 7.50
Redness a*	14.36 ± 0.83	17.81 ± 0.94	26.32 ± 3.03	25.04 ± 3.68
Yellowness b*	13.73 ± 0.38	15.24 ± 0.42	44.46 ± 2.95	27.15 ± 6.65

Most of the previous studies on the presence of PAHs in smoked cheeses concerned benzo(a)pyran [28]. Guillen and Sopelana [29] found over 320 compounds in smoked cheese of various origins. Among these compounds were various acids, phenolic derivatives and a high number of PAHs of differing molecular weights as main flavor components. The level of benzo(a)pyrene ranged from non-detected to 0.91 μg/kg. Guillen and Sopelana [29] have found both compounds of low molecular weight (naphthalene, acenaphthene, fluorene) and of high molecular weight (benz(a)pyrene, benzo(a)pyrene, benzo(g,h,i) perylene). In the analyzed goat and sheep, cheeses from the outer Eastern Carpathians subprovince (Bieszczady) the high content of naphthalene, acenaphthylene, fluorene and phenanthrene is noteworthy (Table 1). To sum up, it should be stated that the analyzed cheeses were characterized by a trace or low content of benzo(a)pyrene and the sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene. This is the result of gentle smoking of cheeses with warm smoke, because the properties of the smoke produced and its composition depend, among others, on the temperature of the pyrolysis process. During smoking, the cheese begins to "sweat", i.e., fat begins to appear on its surface, which, combined with the smoke, preserves the product.

The situation is different in the case of lamb sausages in Table 4.

Polish lamb sausages were characterized by a higher content of benzo(a)pyrene and the sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene (exceeding the permissible limit contained in Commission Regulation (EC) No. 1881/2006 [30] compared to Slovak sausages. The content of 4.5–5.1 μg/kg benzo(a)pyrene and 26.9–34.1 μg/kg sum of benzo(a) pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene disqualifies these products. These sausages may be sold and consumption is allowed by derogation from Commission Regulation (EU) 2020/1255 [31]. Sausage of sum of 4 PAHs 34.10 ± 3.30 is exceeding limits and cannot be sold for consumption. Producer of this sausage was aware of this results and started changing in smoking process by decreasing smoke temperature The end result of traditional smoking, i.e., the level of benzo(a)pyrene and sum of 4 polycyclic aromatic hydrocarbons (benzo(a) pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene) in meat and meat products, dairy products

and smoked fish, may depend, among others, on the raw material. Studies indicate that the raw material (milk, meat) contains trace amounts of PAHs [32–34]. It is the result of contamination of soil, air, water, accumulation of PAHs in plants used as animal feed and the deposition of these compounds in muscles and presence in milk. The type of cheese (its size) and the type of sausage (its thickness and composition) may determine the contents of both BaP and total PAHs4. The most difficult sausages in this respect are thin sausages, e.g., kabanos, frankfurters. Also, small cheeses, e.g., redykołki, are characterized by a higher content of PAH [35]. Waszkiewicz-Robak et al. [36] found that the total amount of PAHs produced during the smoking of the product increases when the raw material contains more fat [37]. Only gentle smoking and then baking of such products can reduce the content of these compounds.

The quality of spices, the method of their drying and the quality of functional additives (the quality of added soy protein or vegetable fiber) may affect the level of PAHs. According to Kubiak [38], the increasing use of smoking additives to improve the quality and organoleptic properties of the product has become a significant source of PAHs in cured meats. On the southern side of Carpathians (Slovakia), dried red pepper is used as a spice for sausages, on the northern side (Poland)—additive of fresh garlic (bulb vegetable) and pepper is used. Those additives may affect a* value which showed higher numbers for Slovakian sausages (Table 4). It also must be stated that using natural casings (pork, lamb or beef intestines) during traditional smoking may contribute to the penetration of PAHs into the meat and thus to their higher content in the final product [39].

A very important factor affecting the quality and safety of smoked meats, cheeses, fish and fruit is the wood used for smoking, its type, hardness, and humidity. The best smoking effect is obtained from the wood of fruit trees from the felling of old trees or from the annual pruning of fruit trees, especially plums, cherries, sweet cherries, apricots and apple trees, because the wood of these trees is rich in hemicellulose. The type of wood is also the color of the smoked product. Smoke from beech, maple, ash and linden wood gives smoked products a golden brown and golden yellow color, while pear and apple wood smoke gives red to dark brown color. Oak wood imparts a dark yellow to brown color to the products [39]. In Poland, alder is the most widely used for smoking; it gives products a color ranging from lemon to brown, depending on the concentration of smoke. In addition, if the humidity parameters are incorrect, the product will become heavily tarred. Alder is a cheap and efficient wood; moreover, it contains little tannins (3–5%), thanks to which the product smoked in this way does not show bitterness. Recommendation to use hardwood is justified, because during the pyrolysis process of hardwood 1.5–4.5 times less benzopyrene is produced than in the case of softwood (fir, pine) [39]. Hardwood with a compact structure burns slower with a "shorter" flame and the volatile compounds formed are not as exposed to oxidation to carbon dioxide as in the case of soft wood [40, 41]. As our results presented in Tables 2 and 3 showed cheeses from Poland where soft wood (pine) was used were characterized by higher L* value which indicate they were more light/pale. Similar relationship were reported for sausages where those from Poland were lighter compared to Slovak (Table 4). Lowest L* value was found in cheeses from Bieszczady (Table 1). On the southern side of the Carpathians, ripening, fermented sausages are produced, while on the northern side they are intensively smoked and there are also differences in the method of smoking (warm or cold in the south of the Carpathians—hot in the north).

Conclusions

The analyzed cheeses were characterized by a trace or low content of benzo[a]pyrene and the sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene. This is the result of gentle smoking of cheeses with cold or warm smoke, because the properties of the smoke produced and its composition depend, among others, on the temperature of the pyrolysis process. In cheeses from outer Eastern Carpathians subprovince (Bieszczady), the high content of naphthalene, acenaphthylene, fluorene and phenanthrene is noteworthy. Polish lamb sausages were characterized by a higher content of benzo(a)pyrene and the sum of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene and chrysene compared to Slovak sausages. Salting, maturing, and smoking cheeses and meats are the main preservation methods. This method still dominates in the production of sheep cheeses or mixed cheeses in Polish and Slovak Carpathians. There are differences in the production and smoking of sausages, which can be observed in PAHs and color results.

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Author contributions
Conceptualization was done by WM; methodology was done by WM, MW, SM and ŁM; experiment and data collection were done by WM, SM, MW, JD, AP-K, MF-F, AM, ŁM; data analysis was done by WM, MW, ŁM and AM; original draft preparation was done by WM; writing and review and editing were done by WM, MW and ŁM.

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Availability of data and materials
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Declarations

Ethics approval and consent to participate
Not applicable.

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All authors declare consent to the publication of this article.

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