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## The variability of acrylamide content in potato French fries depending on the oil used and deep-frying conditions

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### ABSTRACT

The research aimed to investigate the variability of the acrylamide content in French potato fries depending on the type of oil and the length and conditions of deep-frying. Deep-frozen pre-fried potato French fries primarily intended for catering establishments were deep-fried parallel in two oils (multi-component oil and rapeseed oil) at the same conditions (175 °C/4 min and 200 °C/3 min) until the limit for total polar compounds (TPCs) content (24%) was reached. The samples were analysed immediately after removal from the package, after the first frying and when the TPCs was exceeded. High-performance liquid chromatography/electrospray ionization tandem mass spectrometry (HPLC/ESI-MS/MS) was used to determine acrylamide. Mathematical and statistical evaluation of the results was according to the indicators of descriptive characteristics, i.e., arithmetic mean, standard deviation (SD), and coefficient of variation (%). Analysis of variance (ANOVA) was used to compare groups, i.e., the assumption of agreement of variance was verified by the F test (F). All pairwise differences in means were tested using Tukey's HSD test (Honest Significantly Different) and Scheffe's test. The critical value of  $\alpha$ , compared to the standardized difference between the means, was established using our chosen risk of 5%. The highest acrylamide values were measured in samples deep-fried in rapeseed oil at 200  $^{\circ}$ C/3 min in sample 2b (451.13 µg/kg when deep-fried immediately) and in sample 2d (383.24 µg/kg after exceeding TPCs). The lowest values of acrylamide were found in samples deep-fried in multi-component oil at a temperature of 200 °C/3 min in sample 1d (183.35 µg/kg after exceeding TPCs) and at a temperature of 175 °C/4 min in sample 1c (240.75 µg/kg after exceeding TPCs). The decreased tendency of acrylamide in both types of oils and variants of temperature after exceeding TPCs compared to the state immediately after frying is confirmed for all samples. Potato-based products are a significant source of acrylamide production and subsequent consumption. Monitoring its presence in food is, therefore, an important legislative requirement.

Keywords: French fries, Oil, Acrylamide, Deep-frying, Liquid chromatography

### INTRODUCTION

Potatoes (*Solanum tuberosum* L.) rank as the fifth highest-produced commodity for human consumption. In 2019, 371 million metric tons of potatoes were produced worldwide, with China, India and Ukraine accounting for 45% of global production [1]. Potato tubers need to be processed before consumption. Due to their culinary versatility, different cooking practices are applied, with the most widespread being boiling, microwaving, toasting, roasting and frying [2]. Frying is widely used in industrial and domestic sectors because it can create unique sensory properties in fried food [3]. Several chemical reactions occur in the oil during frying and deep-frying, such as oxidation, hydrolysis, oxidative and thermal polymerization, which may cause the formation of various process contaminants [4]. The benefits of the frying are its speed and operational simplicity. Even though deep-frying is an old and popular process, it is still poorly understood [5]. French fries are a semi-finished product and a finished product that is sold frozen in a wide variety of assortments, usually in the form of strips of various thicknesses, but also in the form of slices, crescents, and quarters, so with or without the skin, or even as whole

small tubers. Traditional fries are the most popular variant, defined as fried potato pieces in the shape of strips with a cross-section of  $10 \times 10$  mm and a length exceeding 6-7 cm. When pre-fried, they contain about 4% fat, while ready-to-eat French fries can contain about 7% fat [6]. Repeated frying and deep-frying of food in the same vegetable oil leads to the formation of new compounds resulting from hydrolytic, thermal, and oxidative reactions [7], [8]. Interactions of oils with food matrices lead to further changes affecting food quality and digestibility. These are changes in fatty acid composition, decreased nutritional value considering vitamins and antioxidant compounds, and increased accumulation of dimer and polymerized molecules and other chemical compounds with toxic potential for human health [9], [10].

Short-chain fatty acids, aldehydes, ketones, alcohol, and nonvolatile products generated by lipid oxidation have higher polarity than triglycerides of fresh edible oil. These are considered total polar materials /compounds (TPM/TPCs), and fried food with TPM has been found to exhibit various ill-health effects [11]. Total polar compounds (TPCs) the content in deep-fried oils is a reliable indicator of oxidative degradation of frying oils. For public health concerns, the content of TPCs in frying oil is regulated at not more than 25%, in Taiwan [12]. Several authors [13-16] have determined TPM in a wide range of food. Temperature and frying time affect the kinetics of a wide spectrum of oxidation products and the formation of acrylamide [17]. Acrylamide (CH2=CH-CONH, 2propenamide) is a white, odourless, crystalline solid with a relative molecular weight of 71.08 g x mol<sup>-1</sup> [18]. Acrylamide is produced during the heat treatment of food. Due to its simple structure, it can be formed by various mechanisms, which include the reactions of carbohydrates, proteins, amino acids, lipids, and probably other minor food components. However, it primarily reacts with the amino acid asparagine and glucose [19]. Asparagine content is relatively higher in potato-based products than the reduced sugar content. Reducing sugar level influences the acrylamide formation of the products [20], [21]. Acrylamide is formed in food at a temperature of 120-210 °C, but most of it is formed in the temperature range of 120-170 °C. However, its formation is also possible at a temperature of 100 °C, when an N-glycoside is formed, cleaved between the bonds of C-N atoms, and an intermediate product is formed from which acrylamide can be formed [22]. The primary formation mechanism of acrylamide in food is the reaction of the free amino acid, asparagine, and reducing sugars at temperatures above 120 °C through the Maillard reaction [23]. Acrylamide is produced in foods rich in carbohydrates, which are processed at high temperatures by frying, baking, and roasting. These are most often potato crisps, French fries, and baked potatoes. A slightly lower content is found in bread, breakfast cereals (roasted), coffee, coffee substitutes, gingerbread, crackers, wafers, and biscuits [24].

Acrylamide is a chemical processing contaminant that is generated during the thermal treatment of heatprocessed foodstuffs such as fried potato, cereal-based products, and roasted coffee. This compound presents a public health concern and a priority for the European Food Safety Authority (EFSA). In 2017, the European Commission published a regulation to control acrylamide levels within the primary sources of exposure [25]. In 2019, a new recommendation for monitoring the presence of acrylamide in other foods was also introduced [26]. The US Food and Drug Administration (FDA) has also provided guidance to the industry regarding monitoring and controlling acrylamide in food products, although it has not established maximum recommended levels [27]. French fries are one of the main contributors to the dietary intake of acrylamide [28], [29], although estimated levels of acrylamide intake often fail to take into account consumer behaviour during the preparation of this food [30]. Fried potatoes (French fries, crisps, and hash browns) are prone to acrylamide formation due to the high content of precursors in fresh tubers and the intensity of the thermal treatment applied during frying [31], [32]. Acrylamide in food potentially increases the risk of developing cancer for consumers in all age groups. Since acrylamide is present in a wide range of everyday foods, this concern applies to all consumers, but children are the most exposed age group on a body weight basis [33]. Acrylamide exhibits several toxic effects at once, increasing its overall toxicity. In addition to carcinogenic, neurotoxic, mutagenic, teratogenic, and genotoxic properties, its influence on reproduction has also been proven [22]. The potato industry is continuously perfecting acrylamide mitigation strategies, including selecting suitable potato varieties, controlling transport and storage conditions, monitoring and adjusting process conditions, and using alternative technologies. An essential strategy is to provide end users with information about suitable food preparation methods [34, 35, 36, 37, 38, 39].

The research aimed to investigate the variability of the acrylamide content ( $\mu g/kg$ ) in French potato fries depending on the type of oil and the length and conditions of deep-frying.

### **Scientific Hypothesis**

H1: We expect that acrylamide in French fries will rise at higher frying temperatures.

H2: After the first frying of French fries, the acrylamide content will be lower than after reaching the TPCs limit value, which means wear/burn-through oil.

H3: We expect differences in acrylamide content in French fries fried in rapeseed and multi-component oil.

## MATERIAL AND METHODOLOGY Samples

Deep-frozen pre-fried potato French fries primarily intended for catering establishments were deep-fried parallel in two oils (multi-component oil and single-component oil) (Figure 1). Composition of pre-fried and deep-frozen potato French fries: potatoes, sunflower oil. Nutritional information in 100 g of deep-frozen product: fats 4.5 g, saturated fatty acids 0.5 g, carbohydrates 24 g, sugars 0.5 g, proteins 3 g, salt 0.1 g.

For deep-frying French fries, edible multi-component and rapeseed oils have a wide spectrum of long-term and short-term thermal food preparation (cooking, steaming, frying, baking) and a cold kitchen (salads, marinades, sauces etc.).



Figure 1 Deep-frozen pre-fried potato French fries preparation.

Multi-component oil (1): sunflower oil, sunflower oil HO, rapeseed oil, anti-foam agent E 900. 100 g of the product contains 9.2 g of saturated fatty acids, 56.4 g of monounsaturated fatty acids, 34.4 polyunsaturated fatty acids and 0 g of carbohydrates.

Single-component (rapeseed) oil (2): 100% rapeseed oil suitable for frying. 100 g of the product contains 91.5 g of fat, of which 8.6 g are saturated fatty acids, 0 g are carbohydrates.

## Chemicals

Acetic acid HPLC grade (Fischer Scientific, Loughborough, UK), d3-acrylamide (2,3,3-D<sub>3</sub>-AA) (98%) (Cambridge Isotope Laboratories, Androver, USA), ethyl acetate 99%, (Centralchem, Bratislava, Slovak Republic), K<sub>4</sub>[Fe(CN)<sub>6</sub>.3H<sub>2</sub>O p.a. (Centralchem, Bratislava, Slovak Republic), ZnSO<sub>4</sub>.7 H<sub>2</sub>O p.a. (Centralchem, Bratislava, Slovak Republic), ZnSO<sub>4</sub>.7 H<sub>2</sub>O p.a. (Centralchem, Bratislava, Slovak Republic), Mathematical Slovak Republic), methanol gradient grade  $\geq$ 99.9% (Sigma-Aldrich Laborchemikalien, Germany).

Deep-fat fryers Bartscher SNACK III A162810E with smart thermostat (Bartscher, Germany). Oil tester Testo 270 (Testo SE, Germany).

Vacuum rotatory evaporator Heidolph WB 2000 (Heidolph Instruments, Schwabach, Germany)

HPLC system 1200 series (Agilent Technologies, Santa Clara, California, USA) coupled to an Agilent 6460 Triple Quad detector equipped with an ESI interface.

Centrifuge Sigma 2-16KC (Sigma, Osterode am Harz, Germany).

Nylon syringe filter (Q-Max RR Syringe Filters, Frisenette ApS, Knebel, Denmark).

## Laboratory Methods

The method for determining acrylamide was according to Ciesarová et al. [40]. Samples were extracted to acetic acid (0.2 mM) water solution and pre-extracted to ethyl acetate to avoid the negative impact of salts in the chromatography system. The sample preparation was as follows: 2.0000 g of a homogenized sample was weighed into a 10 mL centrifuge tube with a cap, then 100  $\mu$ L of the internal standard solution (0.0020 g of d3-acrylamide in 100 mL of water) and 18 mL of acetic acid (0.2 mM) were added. After shaking by a vortex mixer for 30 s, the mixture was sonicated for 5 min. Then, 1 mL of Carrez solution I (15 g of (K<sub>4</sub>[Fe(CN)<sub>6</sub>].3H<sub>2</sub>O in 100 mL of water) and 1 mL of Carrez solution II (30 g of (ZnSO<sub>4</sub>.7 H<sub>2</sub>O) in 100 mL of water) were added and mixed for

1 min. After that, the mixture was centrifuged in Sigma 2-16KC at 8720 x g for 10 min at 5 °C. A volume of 5 mL of the clear supernatant was transferred to a separator funnel; 5 mL of ethyl acetate was added and mixed well. The ethyl acetate layer was removed, and the extraction step was repeated twice with 5 mL of ethyl acetate. 3 x 5 mL of ethyl acetate layers were collected and evaporated in a vacuum rotatory evaporator Heidolph WB 2000 at 35 °C to dryness. The residue was dissolved in 1 mL of acetic acid solution (0.2 mM) and filtered through a 0.45 µm pore size nylon syringe filter. LC-MS/MS analysis was performed with an HPLC system 1200 series coupled to an Agilent 6460 Triple Quad detector equipped with an ESI interface. The analytical separation was performed on Atlantis dC18 (100 mm, 3 µm) column (Waters, Milford, MA, USA) using an isocratic mixture of 5 mL of methanol, 1 mL of acetic acid and 500 mL of deionized water at a flow rate of 0.4 mL/min at 25 °C. The ESI mass spectrometry detection was performed in a positive ESI+ mode with drying gas (N<sub>2</sub>) flow 8 L/min and 350 °C temperature, nebulizer pressure 50 psi, capillary voltage 2.5 kV. Data acquisition was performed using multiple reaction monitoring (MRM) with the transition for acrylamide: 72  $\rightarrow$  55 and d3-acrylamide: 75  $\rightarrow$  58. The quantification of acrylamide was calculated with a calibration curve of the standard compound in the range from 10 to 4000 ng/10 mL. The analysis time was 11 min; the retention time of acrylamide and d3-acrylamide was 2.0 min. The LOD of the applied procedure was 10 µg/L; LOQ was 15 µg/L.

### **Description of the Experiment**

Number of samples analyzed: 9.

Number of repeated analyses: 3.

Number of experiment replication: All experiments were done at least in triplicates.

**Design of the experiment:** Deep-frozen pre-fried potato French fries were deep-fried parallel in two oils (multi-component oil and single-component oil) at the same conditions (175 °C/4 min and 200 °C/3 min) until the limit for TPCs content (24%) was reached. The samples were analysed immediately after removal from the package, after the first deep-frying and when the TPCs were exceeded. Four commercial deep-fat fryers of capacity 8 L were used for frying French fries samples. Fresh oils were loaded into the fryer separately and heated to 175 and 200 °C. The same batch of French fries (100 g – one frying cycle) was deep-fried, and the same frying conditions (175 °C/4 min and 200 °C/3 min) were applied. Afterwards, the French fries were placed in a plate, and extra oil was sucked using tissue paper. The frying procedure was held constantly for several days (6 h per day according to reached 24% TPCs content). At the end of each frying day, the deep fryer was shut off, and the oil was cooled down. French fries sampling intervals for the determination of acrylamide content are listed above.

Labelling of samples:

- Control pre-fried and deep-frozen potato French fries.
- 1a French fries deep-fried in multi-component oil, 175 °C/4 min, sampling immediately after the first deep-frying.
- 1b French fries deep-fried in multi-component oil, 200 °C/3 min, sampling immediately after the first deep-frying.
- 1c French fries deep-fried in multi-component oil, 175 °C/4 min, sampling after exceeding TPCs.
- 1d French fries deep-fried in multi-component oil, 200 °C/3 min, sampling after exceeding TPCs.
- 2a French fries deep-fried in single-component oil, 175 °C/4 min, sample immediately after the first deep-frying.
- 2b French fries deep-fried in single-component oil, 200 °C/3 min, sampling immediately after the first deep-frying.
- 2c French fries deep-fried in single-component oil, 175 °C/4 min, sampling after exceeding TPCs.
- 2d French fries deep-fried in single-component oil, 200 °C/3 min, sampling after exceeding TPCs.



**Figure 2** Fried potato chips fried in parallel in two oils (multi-component oil and single-component oil) under the same conditions (175 °C/4 min and 200 °C/3 min) up to the limit for TPC content.

Measurement of TPCs content was performed by oil tester Testo 270. The TPCs content enables a statement on the deterioration of deep-frying oils because of heat. TPCs estimation was based on constant dielectric changes directly measured on hot oil with deep frying oil tester Testo 270.

Table 1 Evaluation of the oil quality based on measurement of TPCs content by Testo 270.

Display indication	Classification
>1% and <14% polar compounds	Fresh oil
>14% and <18% polar compounds	Slightly used
>18% and <22% polar compounds	Used, but still OK
>22% and <24% polar compounds	Much used, it is recommended to exchange
>= 24 % polar compounds	Oil deterioration

LC-MS/MS was used to determine acrylamide. The results were evaluated using the software MassHunter Workstation version B.04.00 and Agilent MassHunter Workstation version B. 04.01.

#### **Statistical Analysis**

The SAS Enterprise Guide Version, 1.5 system program, performed the mathematical and statistical evaluation of the results. The obtained data were statistically evaluated according to the indicators of descriptive characteristics, i.e.  $\bar{X}$  – arithmetic mean, SD – standard deviation and coefficient of variation (%). Analysis of variance (Kruskal-Wallis test, ANOVA) was used to compare groups, i.e., the assumption of agreement of variance was verified by the F test (F). All pairwise differences in means were tested using Tukey's HSD test (Honest Significantly Different) and Scheffe's test. The critical value of  $\alpha$ , compared to the standardized difference between the means, was established using our chosen risk of 5%.

#### **RESULTS AND DISCUSSION**

Several companies operating in the Slovak market today offer a wide range of potato products. French fries are delivered to catering establishments as food for ordinary consumers as 1st and 2nd level convenience, i.e., peeled sliced potatoes in the shape of fries or pre-fried frozen potato French fries intended for deep-frying (semi-finished product). As there is experience from gastronomic practice, their quality is different, not to mention the wrong choice of oil for deep-frying or unsuitable conditions for deep-frying. French fries can be prepared at home or by professional food services from fresh or deep-frozen par-fried potato, with the most extended heat preparing methods being baking and frying.

Setting goals and implementing the experiment was based on Slovak legislation requirements [41]. The legislation states that only fats designed for heat preparation and the given purpose can be used for continuous frying and deep-frying of prepared dishes. These fats and oils can be used for a maximum of 24 hours, while their operating temperature must not exceed 180 °C unless otherwise specified by the manufacturer. Food and catering establishments must also have regularly calibrated temperature measuring devices. However, practice shows that operators of fast-food restaurants often violate these requirements and use deteriorated oils for frying. Concerning the above study, this paper will report on the effect of various oil types and deep-frying conditions on the acrylamide content in French fries. The first prerequisite for evaluating the results was the creation of a calibration curve. Calibration was performed by diluting the acrylamide stock solution (5 mg in 100 mL of water) in the range of 10-4000 ng/10 mL with 50  $\mu$ L of the internal standard (d3-acrylamide).



Figure 3 Calibration curve for acrylamide content ( $\mu$ g/kg) determination.

A supervised learning algorithm's inference of a linear relationship allows prediction a variable based on another variable using simple linear regression. The regression plot of standardized residuals versus standardized predicted values indicates that the points are randomly and evenly dispersed around the line of mean throughout the plot, as shown in Figure 4. No significant trend was observed from the scatter plot. The residual plot suggests no violation of Linearity.

Since frying oil degradation is greatly accelerated by foods, the frying condition should be controlled carefully. The decisive parameter for determining the content of acrylamide in French fries was the monitoring of the TPCs value, which is related to the quality and deterioration of the oil. As already mentioned, deep-frozen pre-fried potato French fries primarily intended for catering establishments were deep-fried parallel in two oils (multi-component oil and single-component oil) at the same conditions (175 °C/4 min and 200 °C/3 min) until the limit for TPCs content (24%) was reached. The samples were analysed immediately after removal from the package, after the first frying and when the TPCs were exceeded.

A lower TPCs content (3.5) was found in fresh rapeseed oil (samples 2) compared to fresh multi-component oil (4.5). Exceeding the TPCs values and thus the oil deterioration occurred after several hours of continuous deep-frying of French fries. The results are shown in Table 2.



Figure 4 Residual plot (standardize residuals against predicted y).

Deep-fat fryers	Oil type	Frying conditions	Oil deterioration (h)	Number of deep- frying cycles
1	multi-component oil	175 °C/4 min	77	154
2	multi-component oil	200 °C/3 min	74	148
3	rapeseed oil	175 °C/4 min	26	52
4	rapeseed oil	200 °C/3 min	25	50

Table 2	TPCs	and dee	n-frving	process
I abit 2	11 03	and ucc	p-n ying	process

Zeleňáková et al. **[5]** examined the thermo-degradative changes of rapeseed and sunflower oils during deepfrying French fries (170 °C, 4 min). One of the investigated parameters was the TPCs value. Rapeseed oil found 3.3% of TPCs and the limit (24%) was achieved on the fourth day. The total time for the deterioration of deepfrying rapeseed oil was  $23\frac{1}{2}$  hours. On the contrary, in fresh sunflower oil on the first day was TPCs content 5.5% and the limit of 24% was reached on the third day. The total time for the deterioration of deep-frying sunflower oil was  $17\frac{1}{2}$  hours. The oil with higher oleic acid content achieved the 24% TPCs after 22 hours (at room temperature) and  $26\frac{1}{2}$  hours (in the refrigerator) of deep-frying. The low erucic acid rapeseed oil was less stable – 19 hours and  $22\frac{1}{2}$  hours, respectively **[42]**.

Samula -					
Sample	Ā	X <sub>min</sub>	X <sub>max</sub>	SD	v [%]
Control	40.33 <sup>g</sup>	38.00	42.00	2.08	5.16
1a	362.91 <sup>bc</sup>	345.50	373.45	15.19	4.19
1b	334.40 <sup>cd</sup>	323.73	346.73	11.59	3.47
1c	240.75 <sup>e</sup>	227.09	249.16	11.93	4.96
1d	183.35 <sup>f</sup>	176.56	190.77	7.13	3.89
2a	322.19 <sup>d</sup>	316.51	333.05	9.41	2.92
2b	451.13 <sup>a</sup>	443.12	456.94	7.17	1.59
2c	265.08 <sup>e</sup>	246.57	283.73	18.58	7.01
2d	383.24 <sup>b</sup>	374.66	388.15	7.46	1.95

**Table 3** Descriptive characteristics of acrylamide content (ug/kg) in French fries deep-fried in two types of oil and under different time-temperature conditions.

Note:  $\bar{X}$  – mean;  $X_{min}$  – minimum;  $X_{max}$  – maximum; SD – standard deviation; v – coefficient of variation. The different letters <sup>a,b,c,d,e,f,g</sup> listed with the mean values in the columns represent statistically significant differences between the observed varieties (p < 0.005).

The highest acrylamide values in French fries fried in multi-component oil were measured in sample 1a at a temperature of 175 °C/4 min (362.91 µg/kg in immediate frying). The lowest value was found in sample 1d using a temperature of 200 °C/3 min (183.35 µg/kg after exceeding the TPCs). A statistically significant difference (p < 0.005) was found between all samples from each other, except for when a significant difference was not found between the samples that were fried immediately 1a at a temperature of 175 °C/4 min and 1b at a temperature of 200 °C/3 min (p = 0.097). In the case of using rapeseed oil, the highest values of acrylamide were found in sample 2b deep-fried at a temperature of 200 °C/3 min ( $451.13 \mu$ g/kg in immediate frying) and the lowest value was found in the case of sample 2c deep-fried at a temperature of 175 °C/4 min (265.08 µg/kg after exceeding the TPCs). Statistically significant differences in frying French fries in rapeseed oil were found between all samples from each other (p < 0.005). The decreased tendency of acrylamide in both types of oils and variants of temperature after exceeding TPCs compared to the state immediately after frying is confirmed for all samples. A complete visual presentation of the results is given in Figure 5.

In principle, it can be concluded that deep-frying influences the acrylamide content in potato French fries. The results also showed that the multi-component oil showed better thermal degradation properties expressed by the TPCs indicator and is more suitable for continuous frying.

Its deterioration (24% TPCs) occurred after 74 and 77 hours of deep-frying (148 and 154 cycles), depending on the combination of temperatures and time. Rapeseed oil reached the TPM limit after only 25 and 26 hours of deep-frying (50 and 52 cycles). The final content of acrylamide in French fries deep-fried in multicomponent oil at its deterioration was 183.35 and 240.75  $\mu$ g/kg, respectively. In French fries deep-fried in rapeseed oil, these values were higher and detected in a shorter time.

Multi-component oil with a special composition is rich in essential fatty acids with an excellent balance of stable monounsaturated and polyunsaturated fatty acids. It does not produce toxins or contains allergens, nuts, soy, palm oil or GMO raw materials. Heat-resistant and long-lasting compared to classic rapeseed oil. High smoke point 220 °C and more, higher oxidation resistance [43], [44].



Figure 5 Visual presentation of acrylamide content ( $\mu g/kg$ ) in individual samples with error bars. Note: Oil 1 – multi-component oil; Oil 2 – rapeseed oil.

The results from acrylamide were compared by ANOVA (Tukey test) whereby we performed pairwise comparisons of samples. To compare all possible simple and complex pairs of means, the Scheffe's test was performed, which has a narrower confidence interval (Table 4), confirming the ANOVA results.

F test		366.02+++						
Sample	1a	1b	1c	1d	2a	2b	2c	2d
Control	+	+	+	+	+	+	+	+
1a		-	+	+	+	+	+	-
1b			+	+	-	+	+	+
1c				+	+	+	-	+
1d					+	+	+	+
2a						+	+	+
2b							+	+
2c								+

**Table 4** Statistical evaluation of differences in acrylamide content ( $\mu g/kg$ ) in French fries depending on the type of oil, the deep-frying conditions, and the total frying time by Scheffe's test P<sub>0.05</sub>.

Note: - Statistically insignificant difference according to the Scheffe's test (p > 0.05); + Statistically significant difference according to the Scheffe's test (p < 0.05).

Figure 6 shows the percentage increase in acrylamide in individual French fries samples. Compared to the control, the most significant increase in acrylamide occurred in sample 2b, where the value of acrylamide increased 11 times. In samples 2a, 1b, 1a and 2d, the value of acrylamide was 7.9-9.4 times higher. The lowest increase of 4.5 times more compared to the control was in the 1d sample.



#### Acrylamide increase in %

Figure 6 Percentage expression of acrylamide increase in French fries.

Regarding the frying conditions, it is known that both the temperature and time of frying determine the kinetics of a wide spectrum of oxidation products and acrylamide formation. This formation starts at temperatures above 120 °C and the maximum rate takes place at temperatures higher than 170-180 °C [45], [46]. In our research, the same batch of French fries (100 g) was deep-fried, and the same frying conditions (175 °C/4 min and 200 °C/3 min) were applied.

The highest acrylamide content in rapeseed oil was formed at 200 °C and linearly occurred with an increment of temperature. The heat sensitivity of the Maillard reaction occurring between reducing sugars and asparagine may cause an increase of the acrylamide content [47, 48, 49]. Likewise, Yang et al., Lu et al. and Mariotti-Celis et al. [50, 51, 52] have reported that acrylamide formation was affected by an increment of frying temperature which agrees with our results.

In the case of multi-component oil, it was recorded for each pair of samples either immediately frying or deepfried in oil after exceeding the TPCs, when a higher acrylamide content was detected at a lower frying temperature.

Ahrné et al. **[53]** measured lower acrylamide values in bread samples when baked at a higher temperature. They stated that the water content plays an important role in the degradation process of acrylamide. They reported that acrylamide concentration tended to decrease as crust temperature increased and water content decreased.

For acrylamide analysis, oil type and lipid oxidation profile plays an important role in the acrylamide concentration in the fried products [54].

French fries are susceptible to acrylamide formation due to the high content of precursors in the tuber and the intensity of the heat treatment used. The chemical composition of potatoes varies, among others, according to cultivar, place of growth, agricultural practices, and maturity at harvest or storage conditions [55].

Reducing the content of acrylamide in potato products can be achieved by choosing varieties with low sugar content, adequate storage and transport, suppression of germination, blanching, the addition of sodium diphosphate, treatment with asparaginase, coarser cutting and frying at a maximum temperature of 175 °C [34], [55], [39].

Recent findings by Lee Kuek et al. **[56]** also confirmed that oil types significantly influenced the acrylamide formation in French fries during intermittent frying.

For instance, based on a real-food investigation, Granda and Moreira [46] have measured the concentration profiles of acrylamide in potato chips and have reported that a counteracting effect exists, in which the acrylamide formation rate rapidly increased at the early stage of frying and then gradually decreased after some time.

Many other scientific teams have been engaged in analysing the acrylamide content in food [18], [33], [40], [57-59].

In the context of the work's objectives, it is important not to forget the legislative requirements regarding the amount of acrylamide in food.

Potato-based products are one of the main contributors to acrylamide exposure [28] due to their acrylamide content and frequency of consumption. The benchmark level of the amount of acrylamide in French potato fries ready for direct consumption is  $500 \ \mu g/kg$ , according to the current legislation. However, although acrylamide content does not exceed the current benchmark level, like in our case, the Commission Regulation 2017/2158 [25] requires food processors and food business operators in Europe to take measures that lead to the reduction of the presence of acrylamide in products according to the ALARA principle, while these measures are proportionate to the size and nature of the operations. Moreover, a mandatory maximum level of acrylamide in a wide range of foods is expected to be set soon.

Food establishments must implement these requirements to produce semi-finished potato products in catering facilities and households. There are still shortcomings in the last two mentioned.

#### CONCLUSION

French fries are fried or deep-fried small potato wedges that are widely consumed around the world. According to surveys, they are among the most popular side dishes, even though they are associated with an unhealthy eating style. Fried potatoes (French fries, chips) are susceptible to acrylamide formation due to the high content of precursors in fresh tubers and the intensity of heat treatment applied during frying. Only the highest quality potatoes can be used to make top-quality French fries. Constant innovations that improve and speed up processes, greener processes, revolutionary ideas and more inventive thinking, manufacturers can guarantee and provide gastronomic establishments with top-quality potato French fries with a low acrylamide content. The correct choice of raw material is very important because it affects the preparation of safe and high-quality potato French fries. The decreased tendency of acrylamide in both types of oils and variants of temperature after exceeding TPCs compared to the state immediately after frying is confirmed for all samples. The highest acrylamide values were measured in samples deep-fried in rapeseed oil at 200 °C/3 min in sample 2b (451.13 µg/kg when deep-fried immediately) and in sample 2d (383.24 µg/kg after exceeding TPCs). The lowest values of acrylamide were found in samples deep-fried in multi-component oil at a temperature of 200 °C/3 min in sample 1d (183.35 µg/kg after exceeding TPCs). Evaluation of established scientific hypotheses:

H1: We expect that acrylamide in French fries will rise at higher frying temperatures. This hypothesis was confirmed only partially. An increasing trend was confirmed only for rapeseed oil (oil 2). On the contrary, when frying French fries in multi-component oil, there was a slight decrease in the acrylamide content at a combination of 200 °C/3min compared to a temperature of 175 °C.

H2: After the first frying of French fries, the acrylamide content will be lower than after reaching the TPCs limit value, which means wear/burn-through oil. This hypothesis was rejected because the opposite trend was noted in all analyses.

H3: We expect differences in acrylamide content in French fries fried in rapeseed and multi-component oil. This hypothesis was confirmed.

In principle, it can be concluded that deep-frying influences the acrylamide content in potato French fries. The results also showed that the multi-component oil showed better thermal degradation properties expressed by the TPCs indicator and is more suitable for continuous frying. From a legislative point of view, it can be stated that

none of the analysed samples of French potato fries (semi-finished product, first deep-frying, limit value for TPCs) exceeded the set content of acrylamide (500  $\mu$ g/kg). Determining the content of acrylamide in French fries and other food products is especially important concerning human health. The results presented in this article represent a valuable source of information for further research in the subject area. Variability of acrylamide content depends on many factors such as temperature (>120 °C), high carbohydrate content, low protein content, free asparagine, reducing sugars, pH, water content, ammonium bicarbonate, high concentration of competing amino acids. The acrylamide content can be effectively reduced by carefully selecting and storing raw materials and by changing procedures during the heat preparation of dishes. In conclusion, it should be emphasized that acrylamide is not the only quality indicator of potato French fries. No less important are sensory and textural characteristics, nutritional value (especially fatty acids profile), and thermo-degradative properties, which are accompanied by chemical reactions like oxidation, polymerization of triglycerides (TAGs), and hydrolysis.

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The authors declare no conflict of interest.

#### **Ethical Statement:**

This article does not contain any studies that would require an ethical statement.

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