Apple processing wastes as potential source of new edible oil

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Summary
Large quantities of apple pomace remain as waste material after apple juice production. The idea of the study assumed that, due to the large share of seeds, apple pomace could serve as a source of edible oil. The aim of the study was to assess the oil content and fatty acids composition of oil extracted from seeds separated from dried apple pomace, and to compare its composition to the canola and grape seed oils. The oil yield from apple pomace seeds was approximately 159 $g \cdot kg^{-1}$. The oil contained a high level of unsaturated fatty acids, predominantly linoleic acid. Polyunsaturated fatty acids were more abundant in apple seed oil than in canola oil, but less abundant than in grape seed oils. The level of monounsaturated acids was lower than in canola oil but higher than in grape seed oil. The obtained results suggest that apple pomace might serve as a material for new edible oil production of high nutritional value. Hereby, the large quantities of wastes remaining after industrial apple processing could be utilized in an innovative and profitable way.

Keywords
apple pomace; apple seed oil; fatty acid composition

Currently, new assortments of edible oils are successfully introduced to the food market, e.g. avocado, hemp, pumpkin seed or hazelnut oils. Some of them show health benefits or special sensory properties. These new oils are, in general, more expensive then usual cooking oils, although some of them are produced from food industry wastes, like grape seed oil. [1]. Grape pomace is a by-product of grape juice and vine production. Grape seeds contain large amounts of oil (up to 200.0 $g \cdot kg^{-1}$) and can be used for edible oil production [2, 3]. Grape seed oil has achieved market success and nowadays is commonly used in frying, baking, and other types of cooking [4]. Similarly to grape seed oil, which is obtained from grape pomace, apple seed oil can be produced from apple pomace.

Apple juice is one of the most popular fruit juices in human diet. According to FAOSTAT database (Food and the Agriculture Organization of the United Nations, Rome, Italy), over 90 countries around the world grow apples commercially and together produce approximately 80 million tons each year. The biggest apple producer is China, representing over 40 % of the world's total [5], USA is the second largest producer with over 5 million tons a year. Poland is the largest apple producer in Europe, having produced over 4 million tons in 2017.

Most of produced apples are used for apple juice, apple juice concentrate and cider production. Currently the biggest producers of apple juice concentrate are China, USA and Poland, representing over 50 % of World's total production [5]. During the processing of apples for juice or cider, huge quantities of solid residues (peel, core, seed, calyx, stem and rest of tissue) are generated, which are generally waste material [6]. These residues, commonly known as apple pomace, account for approximately 250.0 $g \cdot kg^{-1}$ of the weight of the original fresh fruit [7]. Apple pomace in large quantities remains as waste after industrial apple juice production or cider making. It may be utilized in traditional ways, such as composting or low quality animal feed, in particular for horse feeding [8]. Additionally, apple pomace can serve as a source of pectin, dietary fibre of pellets for energy generation purposes [9–11]. Pectin is the most valuable by-product both regarding food technology and human nutrition.

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Even though most plant tissues contain pectin, the major sources for the commercial production of pectin are citrus peel and apple pomace [12]. This is mainly because of the availability of huge amounts of citrus peel and apple pomace from commercial processing operations. However, fresh apple pomace (in the wet state) is very perishable and easily fermented and rotten. Therefore, to allow the storage, transportation and any further processing apple pomace, remained after juice pressing, is usually dried [10]. The dried pomace may be further processed. The new idea presented in this study is to use seeds separated from dried apple pomace for edible oil production, like in the case of grape pomace used for grape seed oil production. Due to high amount of seeds in dried apple pomace, it could be possibly used as a source of edible oil. Similarly to grape seed oil, apple seed oil can be extracted from apple seeds and thus become a new valuable by-product of the production of apple juice or cider. Thus, the aim of the study was to assess the oil content and fatty acid composition of oil extracted form seeds separated from dried apple pomace, and to compare its composition to the canola and grape seed oils. Canola oil, due its favourable fatty acids composition, is recommended in healthy diet and may be considered a nutritional reference for other edible oils. Grape seed oil is an example of a new oil in the food market, which is obtained from grape seeds separated form grape pomace that remain after juice or wine production. This provided the idea to use apple pomace for the same purpose.

MATERIALS AND METHODS

Plant material

The material for the study was dried apple pomace and fresh apples. These were obtained from Agrana Juice Poland Company (Góra Kalwaria, Poland), one of the apple juice concentrate producers. For juice production, apples *(Malus domestica*) of mixed cultivars, mainly Idared and Champion with a minor share of Royal Gala, Jonagold and Golden Delicious, were used. The apple pomace was an industrial waste remained after apple juice production. In the company, the pressed pomace was dried by hot air at 70 °C to protect it against ripening during storage in heaps. Dry mass of apple pomace was determined gravimetrically after drying at 105 °C to achieve constant weight (approximately 3 h).

Grape seed oil and canola oil (the latter also known as rapeseed 00 oil) were used for comparison of oil composition. Both oils, designated for cooking purposes, were obtained from food market in Poland.

Oil recovery

Seeds were separated form dried apple pomace and fresh apples manually. An amount of 1 kg of apple pomace and 5 fresh apples Idared and Champion were used for each analysis. Seeds from fresh apples were separated manually after cutting them into pieces. Samples and separated seeds were weighed and average seed yields were calculated. This procedure was repeated three times.

Usually, fresh apple pomace that remained after juice pressing in the production facility is dried to prolong the shelf life and to stop ripening. Therefore, part of seeds isolated from fresh apples were also dried under typical conditions of the production facility and was also dried to see if this process had an effect on the fatty acid profile. Whole seeds were dried in a tray dryer at 40 °C for 4 h. Heating is known to improve oil recovery from seeds. This procedure is used in particular in small oil mills before pressing native oils from oily seeds [13].

For the recovery of oil, seeds were ground to powder using a laboratory grinder IKA M20 (IKA Werke, Staufen, Germany). The oil fraction was extracted from the apple seed powder (10 g) in a Soxhlet apparatus for 2 h with boiling *n*-hexane (≥95% purity, Sigma-Aldrich, St. Louis, Missouri, USA). Each extraction was repeated three times. Hexane extracts were collected and the solvent was evaporated under vacuum to constant weight using rotoevaporator R300 (Büchi Labortechnik, Flawil, Switzerland). The oil content was determined gravimetrically. The obtained oil samples were again diluted in *n*-hexane and shortly stored at 4 °C under nitrogen gas until methylation.

Preparation of fatty acid methyl esters

Fatty acid profile of apple seed oil was determined by gas chromatography (GC) after conversion of fatty acids to fatty acid methyl esters (FAME). FAME were prepared according to a slightly modified AOCS Official Method Ce 1h-05 [14]. The oil fraction 3 $g \cdot kg^{-1}$ was saponified by 4.0 ml of 0.5 $mol \cdot l^{-1}$ NaOH solution in methanol (99% purity, Sigma-Aldrich), under nitrogen gas, at mixing and heating in a waterbath at boiling point 80 °C for 40 min. The saponified sample was transmethylated with 140.0 $g \cdot kg^{-1}$ BF_3 solution in methanol, under nitrogen gas, at boiling point 100 °C for 3 min. After that, the mixture was cooled and 3 ml *n*-hexane was added, covered with nitrogen gas and shaken vigorously for 30 s while still warm. Then, 40.0 ml of saturated water

solution of NaCl was added and the mixture was shaken vigorously. After phase separation, the hexane layer was transferred by a syringe to a thin glass tube, dried over anhydrous Na_2SO_4 and decanted to a clean vial, covered with nitrogen gas and capped. Then, 1.0 μl of the prepared FAME sample was injected into the gas chromatograph under appropriate conditions.

Gas chromatography

The analysis of FAME was performed by Agilent 6890 N gas chromatograph (Agilent, Santa Clara, California, USA) equipped with Rtx 2330 silica capillary column (length 30 m, internal diameter 0.32 mm, film thickness 0.20 μm; Restek, Bellefonte, Pennsylvania, USA). Hydrogen was used as the carrier gas at a flow rate of 0.9 ml·s^{-1}. A split-splitless (50:1) injector at 235 °C and flame-ionization detector (FID) at 250 °C were used. The column temperature was programmed as follows: initial 155 °C, time 55 min, then increased at 1.5 °C·min^{-1} to a final temperature of 210 °C. Each sample was analysed in triplicate. Results were processed by Chem-station software (Agilent). Peaks were identified by comparison with known standards using FAME Mix Supelco 37 (Sigma-Aldrich). Results were reported as peak area percentages.

Data analysis

The results obtained were statistically analysed using one-way analysis of variance (ANOVA) to check the significance of differences in levels of particular fatty acids among measurements and among analysed samples. For this, Statgraphics Plus ver 4.1 software (Statpoint Technologies, Warrenton, Virginia, USA) was used, applying a significance level of 1 %. Results were expressed as mean ± margin of confidence interval at 95%. Additionally, the Tukey's post-hoc test was run to confirm where the differences between fatty acid levels in oil samples occurred.

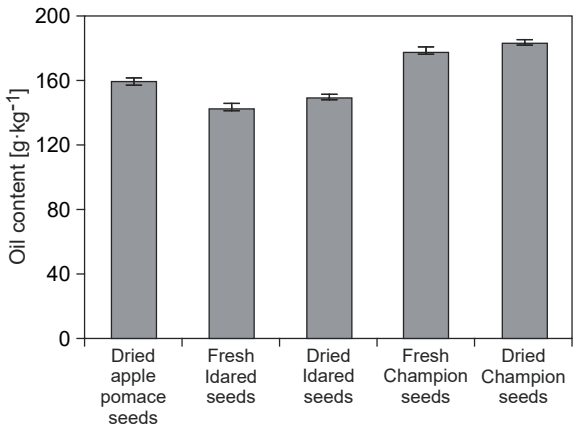

Fig. 1. Oil content in apple seeds separated from apple pomace and fresh apples.

RESULTS AND DISCUSSION

Dry mass, seeds and oil yields

Dry mass of the dried apple pomace used in the study was 922.5 ± 25.0 g·kg^{-1}, while dry mass of freshly pressed apple pomace ranged from 500.0 ± 20.0 g·kg^{-1} to 600.0 ± 25.0 g·kg^{-1}. Average yield of seeds from dried apple pomace was 29.9 ± 1.5 g·kg^{-1} (i.e. 31.3 g·kg^{-1} of dry mass). According to Fromm et al. [15] seeds constitute from 20.0 g·kg^{-1} to 30.0 g·kg^{-1} of apple pomace dry mass, which was very similar to the value obtained in the present study.

Mean oil content in apple pomace seeds was 159.0±8.0 g·kg^{-1} (Fig. 1). This level was comparable to grape seeds where, according to other authors, the oil content was from 140.0 ± 7.0 g·kg^{-1} to 160.0 ± 8.0 g·kg^{-1} [1, 3, 16]. The size of apple and grape seeds is also similar. In the present study, the Idared cultivar seeds contained somewhat lower oil level (149.0 ± 7.0 g·kg^{-1}) than seeds of Champion cultivar (183.0 ± 9.0 g·kg^{-1}). However, the apples used for juice production were of mixed cultivars and apple pomace seeds contained oil at the level between those two (Fig. 2). Similar diversification in oil content was found also by other authors, which was dependent on apple cultivar type [15, 17–20].

Fatty acid composition

Illustrative chromatograms of analysed oil samples are shown in Fig. 2. The predominant fatty acid in apple pomace seed oil was linoleic acid with the level of 584.0 ± 20.0 g·kg^{-1}. This content was slightly but significantly lower than in grapeseed oil (679.0 ± 30.0 g·kg^{-1}) but significantly higher than in canola oil (201.0 ± 10.0 g·kg^{-1}; Tab. 1). The level of α-linolenic acid was low (7.0 ± 0.03 g·kg^{-1}), which was only about a half of that in grape seed oil (17.0 ± 0.8 g·kg^{-1}) and even lower than in canola oil (32.0 ± 1.5 g·kg^{-1}). The obtained results on fatty acid composition in apple seed oil were similar to those of other authors [14, 17–20]. However, these authors analysed other cultivars than in the present study, expected from Fromm et al. [14] where one of analysed sample was Idared.

In apple pomace seed oil, erucic acid was detected at the level of 7.0 ± 0.03 g·kg^{-1}. This fatty acid in excess can be harmful for hearth [22]. In edible oils, erucic acid is regulated to a maxi-

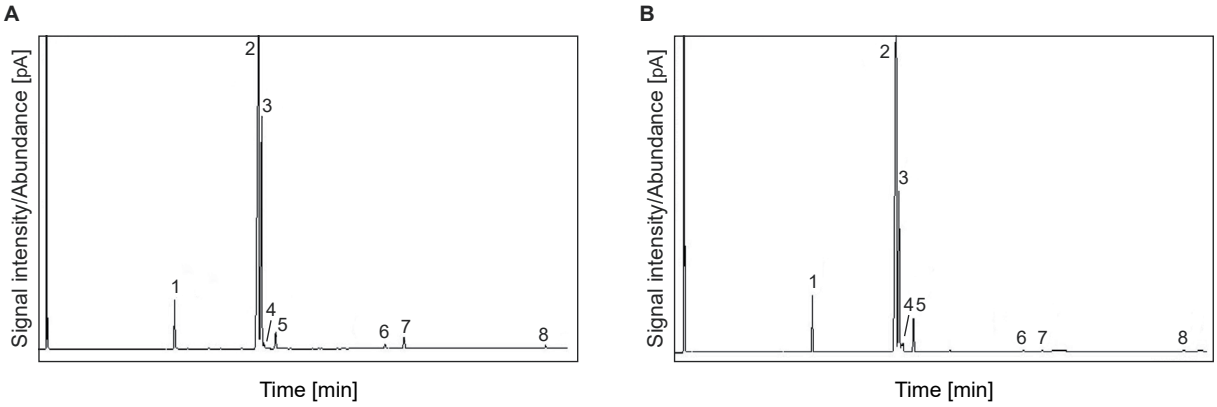

Fig. 2. Chromatograms of fatty acid methyl esters analysis.

A – apple seed oil, B – grape seed oil.
Fatty acids: 1 – palmitic acid C16:0; 2 – linoleic acid C18:2 *n*-6; 3 – oleic acid C18:1; 4 – α-linolenic acid C18:3 *n*-3; 5 – stearinic acid C18:0; 6 – gondoic acid C20:1; 7 – arachidic acid C20:0; 8 – erucic acid C22:1.

mum level of 20.0 g·kg^{-1} by weight in USA and of 50.0 g·kg^{-1} in EU [23, 24]. However, the level of erucic acid found in the samples evaluated in the present study was much below these limits.

The analysed apple pomace seed oil had a low content of saturated fatty acids (78.0 ± 3.5 g·kg^{-1}), significantly lower than grape seed oil (95.0 ± 4.5 g·kg^{-1}) but slightly higher than canola oil (61.0 ± 3.0 g·kg^{-1}; Tab. 2). The generally stable mono-unsaturated fatty acids (mainly oleic acid) were present at a substantial content of 331.0 ± 15.0 g·kg^{-1}, significantly higher than in grape seed oil (208.0 ± 10.0 g·kg^{-1}) and lower than in canola oil (681.0 ± 30.0 g·kg^{-1}).

Tab. 1. Fatty acids profile of sample oils.

Sample	Fatty acids [g·kg^{-1}]							
	16:0 palmitic	18:0 stearic	18:1 oleic	18:2 linoleic	18:3 α-linolenic	20:0 arachidic	20:1 gondoic	22:1 erucic
Dried apple pomace seed oil	38.0±1.5	21.0±1.0	318.0±15.0	584.0±20.0	7.0±0.03	19.0±1.0	6.0±0.3	7.0±0.3
Fresh Idared seed oil	43.0±2.0	19.0±1.0	283.0±14.0	621.0±20.0	6.0±0.03	16.0±0.8	6.0±0.3	6 .0± 0.3
Dried Idared seed oil	42.0±2.0	19.0±1.0	286.0±14.0	624.0±30.0	5.0±0.02	15.0±0.7	5.0±0.25	4.0±0.2
Fresh Champion seed oil	44.0±2.0	20.0±1.0	311.0±15.0	581.0±20.0	6.0±0.03	16.0±0.8	6.0±0.3	12.0±0.6
Dried Champion seed oil	45.0±2.0	20.0±1.0	314.0±15.0	585.0±20.0	5.0±0.02	16.0±0.8	5.0±0.25	10±0.5
Grape seed oil	51.0±2.5	42.0±2.0	206.0±10.0	679.0±30.0	17.0±0.8	2.0±0.1	3.0±0.15	0.0
Canola oil	37.0±1.5	18.0±1.0	681.0±30.0	201.0±10.0	32.0±1.5	6.0±0.3	18.0±0.9	0.0

Content of fatty acids is expressed as grams per kilogram of oil.

Tab. 2. Proportions of fatty acids in evaluated oils.

Sample	SFA [g·kg^{-1}]	PUFA [g·kg^{-1}]	MUFA [g·kg^{-1}]	P/S	Degree of unsaturation [g·kg^{-1}]
Dried apple pomace seed oil	78.0±3.5	591.0±25.0	331.0±15.0	7.5	922.0
Fresh Idared seed oil	78.0±3.5	627.0±30.0	295.0±14.0	8.0	922.0
Dried Idared seed oil	76.0±3.5	629.0±30.0	295.0±14.0	8.2	924.0
Dried Champion seed oil	81.0±4.0	590.0±25.0	329.0±16.0	7.3	919.0
Grape seed oil	95.0±4.5	697.0±30.0	208.0±10.0	7.3	905.0
Canola oil	61.0±3.0	233.0±10.0	699.0±34.0	3.8	932.0

SFA – saturated fatty acids, PUFA – polyuinsaturated fatty, MUFA – monounsaturated fatty acids, P/S - polyunsaturated to saturated fatty acids ratio.

Total polyunsaturated fatty acid (PUFA) level in apple pomace seed oil was 591.0 ± 25.0 g·kg^{-1}, significantly lower than in grapeseed oil (697.0 ± 30.0 g·kg^{-1}), and significantly higher than in canola oil (233.0 ± 10.0 g·kg^{-1}). Polyunsaturated to saturated fatty acids ratio (P/S) in apple pomace seed oil was 7.7, similar to grape seed oil (7.3) but twice higher than in canola oil. Edible fats of high P/S value, of 2 and above, are generally recognized as being hypocholesterolemic. Raising the P/S ratio in the diet is recommended for the prevention of cardio-vascular diseases [25]. Apple seed oil due to a high P/S ratio of 7–8, can be considered as hypocholesterolemic. However, higher PUFA content and degree of the oil unsaturation decrease the storage time of oil due to their highest susceptibility towards oxidation [26]. The level of unsaturation degree was similar in all oil samples (approximately 920 g·kg^{-1}).

Nutritionally favourable fatty acid composition of apple seed oil was shown to influence health. In animal studies, Fotschki et al. [27] found that addition of apple seed oil in rats diet had slightly better metabolic effects on organism than canola oil in case of triacylglycerols concentration and atherogenic index of plasma as well as plasma alanine (ALT) and aspartate aminotransferase (AST) activities. Moreover, antioxidant and antimicrobial activities of apple seed oil were reported by Tian et al. [28] with the minimum inhibitory concentration (MIC) preventing visible growth of bacteria, moulds and yeasts of apple seed oil ranging from 0.3 g·ml^{-1} to 0.6 g·ml^{-1}. Walia et al. [29] showed in vitro cytotoxic activity against specific cell lines exhibiting its potential as an anticancer agent. This effect was due to the high level of natural antioxidants [19]. Gornaś et al. [30] found in apple seed oils variability of tocochromanols, where α-tocopherol was predominant and of very high level (587.7 mg·kg^{-1}). Moreover, apple pomace contains also other antioxidants including polyphenolic and triterpenic compounds [31].

In large scale industry, approximately three fourths of processed apples are utilized for juice production and the remaining one fourth constitutes the remaining by-product, i.e. apple pomace [32]. According to the Polish Association of Juice Producers (KUPS, Warsaw, Poland), in 2016 in Poland almost 2 500 000 t of apples (i.e. more than a half of the total harvest) was used for juice, concentrate and cider production, of which 2 150 000 t was used only for concentrate production. These production volumes caused the generation of approximately 500 000 t of pressed apple pomace of high seeds content, which can be used for new edible oil production. The rest of apple pomace and extracted seeds still can be used for pectin, fibre or pellet production. Moreover, attempts have been made to use apple pomace to generate several value-added products, such as enzymes, single-cell protein, aroma compounds, ethanol, organic acids, polysaccharides, or mushrooms [33].

CONCLUSIONS

This study contributes to the knowledge of food industry waste management as well as new assortment of edible oil production. We showed that the oil extracted form apple pomace seeds contained a high level of unsaturated fatty acids, predominantly linoleic acid. Polyunsaturated fatty acids level was significantly higher in apple pomace seed oil than in canola oil but in lower than in grape seed oils. A similar situation was determined in the case of saturated fatty acids. The level of monounsaturated acids was lower than in canola oil but higher than in grape oil. The results obtained in the study suggest that apple pomace may serve as a starting material for new edible oil production of high nutritional value. Hereby, the large quantities of waste material remaining after industrial processing of apples could be utilized in an innovative and profitable way. Due to the growing interest of consumers in new types of vegetable oils, apple oil can be successful in the food market. A limitation of the idea of oil extraction from apple pomace is the separation of seeds from peels, stems and other dried apple pomace particles. However, this limitation can be overcome by separation of dried apple pomace using air separation similar to blower for cereals.

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