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REVIEW

Recent Development on the Chemical Composition and Phenolic Extraction Methods of Apple (*Malus domestica*)—A Review

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Abstract
Apple (*Malus domestica*) is a reliable source of nutrients to increase the bioactive compounds intake in the human body. In the market, the development of apple is still growing, not only in the hybridization of new cultivars, but also in the functional food products, based on apple's bioactive compounds. In this paper, we review the recent studies about the chemical composition of apples, including carbohydrates, proteins, lipids, dietary fibers, minerals, vitamins, phenolics, and other compounds, from various cultivars, grown in different countries or regions. Among the bioactive compounds in apple, the phenolic compounds, including hydroxybenzoic acids, hydroxycinnamic acids, flavanols and their oligo- and polymeric structure, flavonols, dihydrochalcones, and anthocyanins, majorly contribute to beneficial biological impacts. Therefore, the extraction process might be the most critical step to recover all the phenolics from apple and could be used in various food product developments. In this paper, the comparison of conventional and developed phenolic extraction methods is also reviewed in various apple products (flesh, peel, pomace, pulp, etc.). The selection of food grade and green solvents in the optimal phenolic extraction methods could reduce the environmental issues, thus supports sustainability and can be safer for consumers. To sum up, this paper may help the readers, both at general household and industrial levels, to understand the nutritional composition of various apple cultivars from different regions and to select the optimum conditions for apple's phenolic extraction, based on recent studies.

Keywords Antioxidant · *Malus domestica* · Bioactive compounds · Cultivar · Non-conventional extraction · Phenolics

Introduction

Due to its pleasant taste and high nutritional values, apple (*Malus domestica*) becomes one of the most popular fruits consumed worldwide. It is also one of the most cultivated crops in the world due to its ease to grow and harvest. The production of apple in the world increased around 21%, from 77 to 93 million tons from 2011 to 2021, respectively (FAO, 2023). In 2021, China was reported to be the largest producer of apple, contributed 49.37% of worldwide apple production, followed by Turkey (4.82%) USA (4.80%), and Poland (4.37%) (FAO, 2023).

There is a vast number of apple cultivars produced and available all the year-round. Gala (863,000 tons), Red Delicious (625,000 tons) and Honey Crisps (542,000 tons) was reported by the World Apple and Pear Association (WAPA) as the dominant cultivars produced in USA (WAPA, 2021). While in European Union-20 (20 Member States, excluding 7 Member States that are very small producers and for which there is no data) and UK, especially in the period of 2017–2021, the major cultivars produced were Golden Delicious (19.1%), Gala (12.9%), Idared (6.7%), Red Delicious (6.7%), and Shampion (5.9%) (European Commission, 2022). In southern hemisphere (Argentina, Australia, Brazil, Chile, New Zealand, and South Africa), Gala became the most produced cultivar in 2019 with 1,931,000 tons, followed by Fuji (774,000 tons) and Red Delicious (642,000 tons) (WAPA, 2020). In 2019, the worldwide availability of apple was 22 g/capita/day, ranged from 37 g/capita/day (Europe) to 5 g/capita/day (Africa) (FAO, 2022). The abundant apple production and its availability across the world could be an effective way to increase the intake of nutritional compounds in human diet through various apple products.

Regarding apple as one of sources in human diet, it is necessary to ensure the safety of the apples. According to

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European regulation (EC No. 85/2004), to be safely consumed, apple must be intact, clean, free from pests or pathogens, and free from foreign smell and/or taste (European Commission, 2004). The other safety concerns for apple fruit and products are pesticides residues and the secondary metabolite produced by *Penicillium expansum* (most common postharvest pathogen in apple), called patulin, which is a mycotoxin that causes chronic toxic problems, such as genotoxicity, immunotoxicity, and neurotoxicity (Lončarić et al., 2021). According to EC No. 1881/2006, the presence of patulin in apple juice and fermented drink derived from apples is limited to no more than 50 µg/kg, while in solid apple products to 25 µg/kg, and apple products for infant to 25 µg/kg (European Commission, 2006).

Around 70–75% of apples are consumed as fresh fruit (Zhang et al., 2021). It is recommended by the WHO to consume 400 g edible fruits or vegetables in order to improve overall health, and to prevent the nutrient deficiencies and non-communicable diseases (WHO, 2023). With the weight of apple whole fruit around 149–223 g (USDA, 2019), consuming 2–3 apples daily would be enough to fulfill the recommendation. Furthermore, about 25–30% of apples are processed into various products: (a) liquid form, such as apple juice or concentrate, apple cider, and apple vinegar; (b) solid form, such as freeze/dried apple, apple canning, apple sauce/jam, and apple slice. During the apple processing in industry, 25–30% of pomace are generated (Zhang et al., 2021). Each edible part of apple contributes to bioactive compounds, including carbohydrates, fibers, fatty acids, organic acids, polyphenols, carotenoids, and terpenes which provide health-promoting values in human diet (Acquavia et al., 2021). The major phenolics found in apple are hydroxybenzoic acids, hydroxycinnamic acids, flavanols and their oligo- (procyanidins) and polymeric structures (proanthocyanidins), flavonols, dihydrochalcones, and anthocyanins (Kschonsek et al., 2018). Apple shows functional and health promoting effects, such as antioxidant potential, anti-inflammatory activity, cholesterol-lowering effect, cardiovascular protective effect, antidiabetic activity, and anticancer activity due to its major phenolic compounds (Patocka et al., 2020).

In order to perceive the optimum health-promoting benefits from apple's phenolics, the phenolic extraction process for edible parts of apple, such as whole fruit, flesh, peel, and juice, and by-product, such as pomace should be carried out correctly and efficiently. The environmental problems due to high utilization of organic solvent should be reduced by the development of extraction techniques. It is also important to use the solvents which are food grade (Zhang et al., 2021). The obtained phenolic extract could be applied in various food product developments to improve their functional properties. Therefore, this present study focused on the review of apple's chemical compositions, total phenolics, antioxidant potential of edible parts of apples from different cultivars and locations. The optimization of phenolic extraction using conventional and developed extraction methods was also reviewed in this study since different types of apple products from different cultivars might have different optimum extraction conditions. It is worth noticing that, to date, the development of apple fruit in the world is still growing, either in hybridization of new cultivars, or in food product development based on apple's bioactive compounds. Through this review, the readers, especially food industries, could select the extraction methods to produce apple products' phenolics that are suitable for their needs, based on recent studies.

Botanical Characteristics

Apple (*Malus domestica*) belongs to *Rosaceae* family. The botanical characteristics of apple have been described by (Jackson & Palmer, 2011). Depending on the rootstock, the apple tree has 1.5 to 7 m in height, with basal diameter ranges from 1 to 4.5 m. It is a hermaphrodite plant; some cultivars are partly self-fertile, but cross-pollination may also happen. It can also grow in wide range of soil types. The expected yield varied from 20 to 70 tons/ha, in 3–10 years of full production period, depending on the cultivar or rootstock and site. Based on various estimations, there are over 7500 apple cultivars discovered from various regions in the world, but only around 20 cultivars are widely cultivated in horticulture industry (Shlyavas et al., 2019). Several grown cultivars include, but are not limited to Royal Gala, Golden Delicious, Red Delicious, Jonagold, Braeburn, McIntosh, Fuji, Granny Smith, and Cripps Pink (Pink Lady®) (Jackson & Palmer, 2011; Simmonds & Howes, 2016). Apple grows and is adaptable to various climates, but grows better in moderate humidity and cool temperature, in the 35–50° latitude (Hammad & Malik, 2021). Higher humidity may increase the chance of diseases, such as scab and Nectria canker. The symptoms and favorable condition of apple diseases, such as apple scab, black rot, cedar apple rust, flyspeck, powdery mildew, apple mosaic, and core rot were described by Manavalan (2021). Temperature affects the quality of apples. Some cultivars (i.e., Granny Smith) grows better in warmer area, while Cox's Orange Pippin and Red Delicious have better flavor, color, and shape in cooler climates. Apple production can be found in various regions, for example, in Asia (mostly in China), Europe, North America, Africa, Middle East, and also southern hemisphere area (FAO, 2023). In warmer countries, apple is also grown for example in Ethiopia (Jemaneh & Chandravanshi, 2021), South Africa (Tharaga et al., 2021), Indonesia (Efendi, 2014; Utami et al., 2019), Costa Rica (Navarro et al., 2018), Brazil (Pio et al., 2019), and Argentina (Castro et al., 2016). The dormancy-management practices of apple tree should be applied in tropical countries to allow adequate flowering, growth, and productivity (Pio et al., 2019).

Chemical Composition

Apple contains an abundance of chemical compounds with proven health-promoting properties for humans, both by providing nutritional value and preventing various diseases. Bioactive compounds of apple fruit, such as organic acids, fibers, minerals, and vitamins, are shown in Table 1, while phenolic compounds are summarized in Table 2.

Basic Composition

Apple has been known as a beneficial fruit thanks to its balanced nutritional composition. The nutritional value of edible parts of apple is shown in Table 1. The content of each nutrient varied depending on the cultivars. Water dominated the composition of apple, ranged from 81.06 to 85.60 g/100 g. Apple provided moderate calories of

Table 1 Proximate analysis and content of dietary fiber, organic acids, minerals, and vitamins of apple fruit's edible parts per 100 g fresh weight (FW)

Nutrients	National Food Institute of Denmark (2022)[a]	USDA (2019)[b]	Almeida and Gomes (2017)[c]	Feliciano et al. (2010)		**Unit**
				Traditional apples[d]	**Exotic apples**[e]	
Energy	55	52	57	N.A	N.A	kcal/100 g
Water	84.90	85.60	84.50	81.06	81.73	g/100 g
Protein	0.30	0.26	0.35	0.08	0.07	
Total lipid	0.20	0.17	0.10	N.A	N.A	
Ash	0.30	0.19	0.23	N.A	N.A	
Carbohydrate	12.10	13.80	14.80	N.A	N.A	
Sugars	10.90	10.40	N.A	11.86	11.60	
Dietary fiber	2.20	2.40	2.00	2.97	2.29	
Soluble fiber	N.A	N.A	N.A	0.37	0.41	
L-Malic acid	N.A	N.A	N.A	181.80	324.68	mg/100 g
Citric acid	N.A	N.A	N.A	3.25	5.60	
Calcium (Ca)	4.13	6.00	N.A	2.48	3.22	
Iron (Fe)	0.12	0.12	N.A	0.15	0.15	
Magnesium (Mg)	4.49	5.00	N.A	3.90	3.46	
Phosphorus (P)	9.52	11.00	N.A	8.41	10.19	
Potassium (K)	118.00	107.00	N.A	100.90	99.46	
Sodium (Na)	0.60	1.00	N.A	0.82	0.94	
Zinc (Zn)	0.02	0.04	N.A	0.03	0.03	
Copper (Cu)	0.03	0.03	N.A	0.07	0.04	
Manganese (Mn)	0.06	0.04	N.A	0.04	0.04	
Vitamin C	8.26	4.60	N.A	8.88	8.28	
Vitamin E (α-tocopherol)	0.25	0.18	N.A	0.18	0.14	
Thiamin (Vit. B_1)	0.01	0.02	N.A	0.01	0.01	
Riboflavin (Vit. B_2)	0.01	0.03	N.A	0.00	0.00	
Niacin (Vit. B_3)	0.12	0.09	N.A	0.12	0.13	
Pantothenic acid (Vit. B_5)	0.07	0.06	N.A	< 0.10	< 0.10	
Pyridoxine (Vit. B_6)	0.05	0.04	N.A	0.03	0.04	
Folate (Vit. B_9)	9.00	3.00	N.A	< 5.00	< 5.00	µg/100 g
Vitamin A (retinol)	0.00	3.00	N.A	< 4.50	< 4.50	
β-carotene	25.00	27.00	N.A	11.92	10.24	

N.A. data not available

[a]Based on analytical data for all apple cultivars (the cultivars are not described)

[b]Based on analytical data for Red Delicious, Golden Delicious, Gala, Granny Smith, and Fuji apples

[c]Based on analytical data for apples from the Protected Geographical Indication "Maçã de Alcobaça," Portugal: Casa Nova, Fuji, Galaxy, Golden Delicious, Granny Smith, Jonagored, Reinette, and Starking apples

[d]Based on analytical data for traditional apples from Portugal: Bravo de Esmolfe, Malápio Fino, Malápio da Serra, and Pêro Pipo apples

[e]Based on analytical data for exotic apples from Portugal: Fuji, Starking, Reineta Parda, Gala Galaxy, and Golden apples

Table 2 Phenolic composition of flesh and peel of various apple cultivars

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
Hydroxybenzoic acids	Gallic acid	Flesh	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
		Peel	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
	Protocatechuic acid	Flesh	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
		Peel	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
Hydroxycinnamic acids	Chlorogenic acid	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	D in all cultivars	Karaman et al. (2013)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
			Chouka 4, Fuji, JPP35, Nakano no Kirameki, Nakano Shinku, No. 37	D in all cultivars	Sato et al. (2017)
			Big Time, Cripps Pink, Cripps Red, Firm Gold, Fuji, Gala, Galaxy, Golden Delicious, Granny Smith, Hi-Early, Lady Williams, Naga Fu no. 2, Purple Wave, Red Braeburn, Sansa, Splendour, Wandadale, Western Dawn, Western Tang	D in all cultivars	Bondonno et al. (2020)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	N.D. in Ervin Spur and Granny Smith	Karaman et al. (2013)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
	Caffeic acid	Flesh	King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	D in all cultivars	Karaman et al. (2013)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Chouka 4, Fuji, JPP35, Nakano no Kirameki, Nakano Shinku, No. 37	D in all cultivars	Sato et al. (2017)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
		Peel	King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	N.D. in all cultivars	Karaman et al. (2013)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
	p-Coumaric acid	Flesh	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
		Peel	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
	Ferulic acid	Flesh	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
		Peel	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
Flavanols and procyanidins	Catechin	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	D in all cultivars	Karaman et al. (2013)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Chouka 4, Fuji, JPP35, Nakano no Kirameki, Nakano Shinku, No. 37	D in all cultivars	Sato et al. (2017)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	N.D. in all cultivars	Karaman et al. (2013)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
	Epicatechin	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko, (2020)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	D in all cultivars	Karaman et al. (2013)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Chouka 4, Fuji, JPP35, Nakano no Kirameki, Nakano Shinku, No. 37	D in all cultivars	Sato et al. (2017)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	D in all cultivars	Karaman et al. (2013)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Big Time, Cripps Pink, Cripps Red, Firm Gold, Fuji, Gala, Galaxy, Golden Delicious, Granny Smith, Hi-Early, Lady Williams, Naga Fu no. 2, Purple Wave, Red Braeburn, Sansa, Splendour, Wandadale, Western Dawn, Western Tang	D in all cultivars	Bondonno et al. (2020)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Big Time, Cripps Pink, Cripps Red, Firm Gold, Fuji, Gala, Galaxy, Golden Delicious, Granny Smith, Hi-Early, Lady Williams, Naga Fu no. 2, Purple Wave, Red Braeburn, Sansa, Splendour, Wandadale, Western Dawn, Western Tang	D in all cultivars	Bondonno et al. (2020)
	Procyanidins	Flesh	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Chouka 4, Fuji, JPP35, Nakano no Kirameki, Nakano Shinku, No. 37	D in all cultivars	Sato et al. (2017)
		Peel	Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
			Big Time, Cripps Pink, Cripps Red, Firm Gold, Fuji, Gala, Galaxy, Golden Delicious, Granny Smith, Hi-Early, Lady Williams, Naga Fu no. 2, Purple Wave, Red Braeburn, Sansa, Splendour, Wandadale, Western Dawn, Western Tang	D in all cultivars	Bondonno et al. (2020)
Flavonols	Quercetin	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	N.D. in all cultivars	Zielińska and Turemko (2020)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	N.D. in all cultivars	Karaman et al. (2013)
			Ambrosia, Fuji, Golden (Spain), Golden (France), Granny, Pink Lady®, Red, Royal	D in all cultivars	Alarcón-Flores et al. (2015)
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith	D in all cultivars	Karaman et al. (2013)
			Ambrosia, Fuji, Golden (Spain), Golden (France), Granny, Pink Lady®, Red, Royal	D in all cultivars	Alarcón-Flores et al. (2015)
	Isoquercetin	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
	Rutin	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	D in all cultivars	Raudone et al. (2017)
Dihydrochalcones	Phloridzin	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Chouka 4, Fuji, JPP35, Nakano no Kirameki, Nakano Shinku, No. 37	D in all cultivars	Sato et al. (2017)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)

Table 2 (continued)

Phenolic group	Name	Part of fruit	Cultivars	Notes (D or N.D.)	References
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Golden Delicious, Red Delicious, Royal Gala	D in all cultivars	Chen et al. (2012)
			Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	D in all cultivars	Kschonsek et al. (2018)
			Big Time, Cripps Pink, Cripps Red, Firm Gold, Fuji, Gala, Galaxy, Golden Delicious, Granny Smith, Hi-Early, Lady Williams, Naga Fu no. 2, Purple Wave, Red Braeburn, Sansa, Splendour, Wandadale, Western Dawn, Western Tang	D in all cultivars	Bondonno et al. (2020)
Anthocyanins	Cyanidin-3-galactoside	Flesh	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Chouka 4, Fuji, JPP35, Nakano no Kirameki, Nakano Shinku, No. 37	N.D. in Fuji	Sato et al. (2017)
		Peel	Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	D in all cultivars	Zielińska and Turemko (2020)
			Lještarka, Božićnica, Kolerova, Srčika, Ivanlija, Duga, Zimnjara, Citronka, Zlatica, Paradija, Adamova Zvijezda, Slavonska Srčika, Cortland, Russet, Wild	D only in Lještarka, Božićnica, and Cortland	Jakobek and Barron (2016)

D detected, *N.D.* not detected

52 kcal/100 g (USDA, 2019). Protein and lipid did not contribute much to the energy value of apple due to the relatively small content. However, the compositions of fatty acids and amino acids found in various apple cultivars were reported by previous studies. Salas et al. (2016) reported that fatty acids are the main precursors of volatile compounds in apples which concentration mostly increased during 210 days of cold storage, especially Golden Delicious from Chihuahua, Mexico, which were dominated by linoleic acid (85–125 mg/kg), followed by palmitic acid (27–45 mg/kg), linolenic acid (5–17 mg/kg), oleic acid (3–12 mg/kg), and stearic acid (5–8 mg/kg). Similar results were found as among 16 analyzed fatty acids, palmitic, stearic, and linoleic acids were dominant in Bravo de Esmolfe apple from Portugal (Pires et al., 2018). Additionally, fatty acids (linoleic, oleic, and palmitic acid were abundantly found), carotenoids,

and α-tocopherol were also found in the seeds of apples grown in Norway (Akšić et al., 2021). For amino acids, Feng et al. (2014) found that most amino acids, except histidine, lysine, threonine, and glycine, were found in higher level in the apples from inner-tree canopy. Total protein content of Eastern Himalayan apples, especially Dorsett Golden and Anna cultivars, was higher than that of Western Himalayan ones (Raj et al., 2021). This phenomenon can be explained as the higher annual temperature of Eastern Himalayan region positively affect the nutritive composition of the apples, while higher precipitation in West Himalayan region affected it negatively. Additionally, the composition and concentration of amino acids varied among cultivars depending on location, season, and crop load (Peck et al., 2016).

The sugars (10.40 to 11.86 g/100 g) were also found in apple and dominantly contributed to the energy value and the taste of apple (Table 1). Most fruits contain higher content of glucose and fructose than sucrose (Li et al., 2020a). The content of fructose and glucose of dried Bravo de Esmolfe apple flesh was around 14 times and 6 times higher than sucrose content, respectively (Pires et al., 2018). In Norway, a study also compared 74 apple cultivars grown in Ullensvang area to 29 cultivars in Njøs; it was found that the apples in Ullensvang showed higher average content of sugars (744.54 g/kg dry weight (DW)), sugar alcohol (60.33 g/kg dw), and organic acid (90.81 g/kg DW) than those in Njøs (Akšić et al., 2022). The sugar and major organic acid (malic acid) contents were higher in the sun-exposed apples, compared to the shaded ones, due to increased total photosynthesis in the leaf (Jing et al., 2020). It is worth noticing that organic acids were also found in all apple cultivars from Turkey (Aslik, Cebegirmez, Bey Elmasi, Arapkizi) with following order: malic > fumaric > succinic > tartaric acids (Celik et al., 2018). A study also reported that malic acid, which is more responsible for the sourness of apple (Petkova et al., 2019), predominated the organic acid composition of Bravo de Esmolfe apple, followed by quinic, oxalic, and shikimic acids (Pires et al., 2018). In the analysis of 10 apple cultivars grown in Austria, the average showed that malic acid predominated the organic acid composition, followed by citric (4.6%), pyruvic (0.4%), oxalic (0.3%), maleic (0.3%), and shikimic acid (0.1%) (Oszmiański et al., 2020). While in China, 85.8% of analyzed 106 sour apple cultivars were reported to have organic acids with following order: malic > oxalic > citric > lactic > succinic > fumaric acid (Yan et al., 2018). A comparative analysis of chemical composition of apples cultivated in Western and Eastern Himalaya reported that among the analyzed cultivars, Dorsett Golden apple in Western Himalaya possessed the lowest content of fructose (355.50 mg/g fresh weight (FW)) and glucose (76.85 mg/g FW), which could be then recommended for the diabetic patients (Raj et al., 2021). The sugar content in apple fruits varied due to some factors such as climate conditions, fruit positioning (Feng et al., 2014), genotype (Akagić et al., 2019), growing region, cultivar, maturity, seasons, blooming period, altitude, temperature, storage condition (Raj et al., 2021; Yoon et al., 2020), quality, and intensity of light (Jing et al., 2020).

The dietary fiber content in edible parts of apples from different cultivars and countries is shown in Fig. 1. Dietary fibers were also found in apples in both soluble and insoluble forms. The total dietary fibers of traditional and exotic apples grown in Portugal were 2.97 and 2.29 g/100 g, respectively, while the soluble fiber contents were 0.37 and 0.41 g/100 g, respectively (Feliciano et al., 2010). Another study reported that total dietary fiber of various apple cultivars was about 1.79 g/100 g, with 0.40 g/100 g of soluble, and 1.39 g/100 g of insoluble fiber (Patocka et al., 2020). The higher ratio of insoluble to soluble fiber in Red Delicious apple (4.37), compared to Granny Smith (3.21), might be the reason why more energy is required to remove the water during the drying process of Red Delicious apple, in comparison to that of Granny Smith (Joardder et al., 2015). It is also noteworthy that consuming apple with the peels is better as the dietary fiber content of apple peel is twice to three times higher than the apple flesh (Patocka et al., 2020).

In comparison to other fruits, apple contains higher fiber contents (pectin, cellulose, hemicellulose, and lignin) (Feliciano et al., 2010). Apple's pectin helps in cholesterol and blood glucose reduction (Asale et al., 2021). A study on pectin content in pulp of 35 apple cultivars grown in Czech Republic found that Strymka cultivar possessed the highest content (3.26 g/100 g FW), while the lowest (1.11 g/100 g FW) was found in Lebelovo cultivar that was harvested in Tišnov (Balík et al., 2012). In the analysis of 17 apple cultivars grown in Siena, Italy, the pectin content varied from 4.05 (Rossa Casetta) to 19.72 mg/g FW (Solaio) (Berni et al., 2019). Recent study found that among the new Hungarian apple cultivars, Damra exhibited the highest pectin content (1.15 g/100 g), followed by Feronia (0.82 g/100 g) and Bellona (0.74 g/100 g) (Ebadi et al., 2022). The pectin content of those three cultivars was significantly higher than that of Jonagored (0.59 g/100 g), Idared (0.57 g/100 g), and Fuji (0.56 g/100 g) as controls or commercial cultivars. The authors also added that the picking or harvesting time significantly influenced the pectin content of abovementioned cultivars, as for examples, Damra showed the highest pectin content in third picking time because it is a late ripening cultivar, while the pectin content of Feronia was the highest in the first picking time because it is an early ripening cultivar. With high pectin content as one of the quality parameters, the authors concluded that those new cultivars are suitable for fresh consumption (Ebadi et al., 2022).

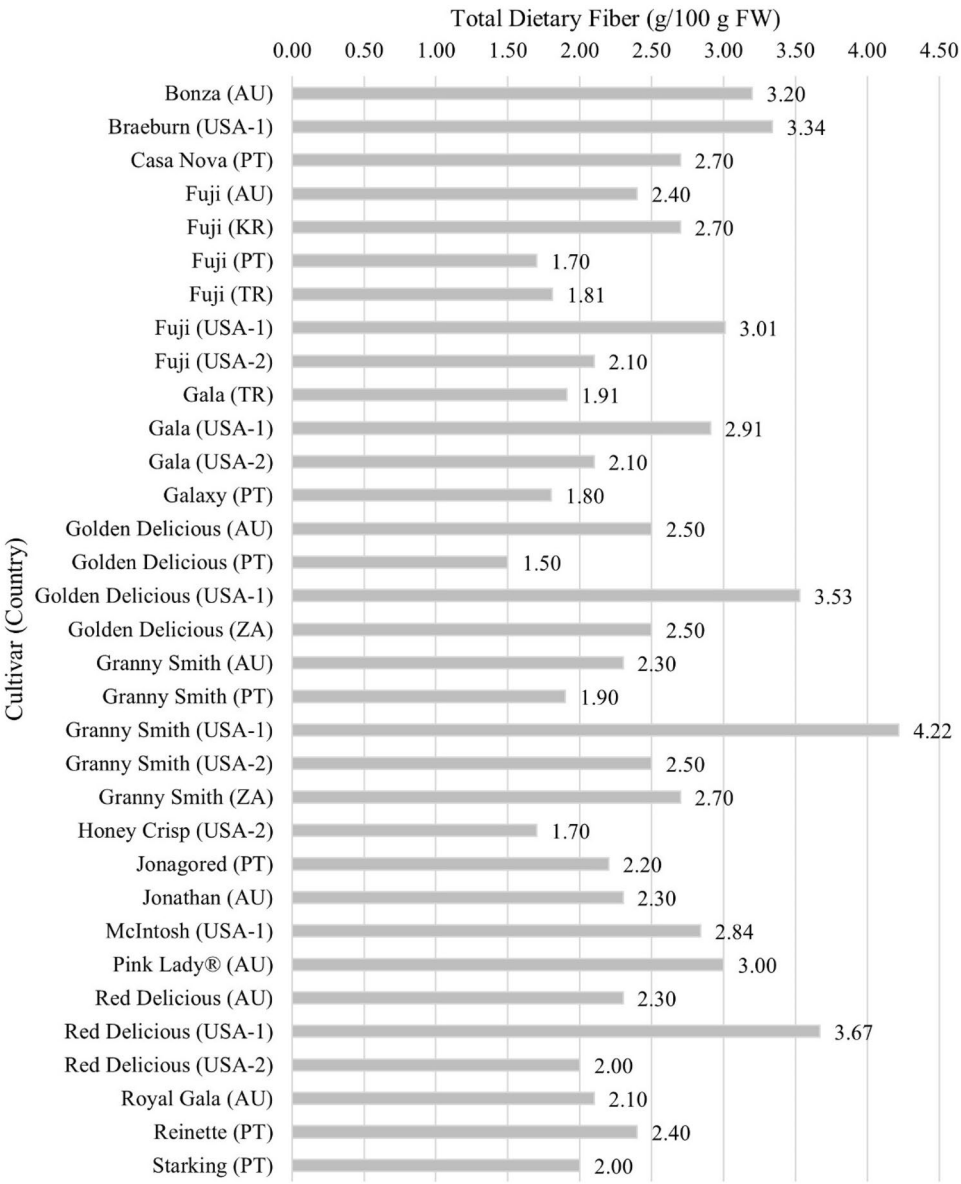

Fig. 1 Total dietary fiber content in edible parts of apples from different cultivars and countries. Abbreviations: FW, fresh weight; AU, Australia (Food Standards Australia & New Zealand, 2021); KR, South Korea (National Institute of Agricultural Sciences of South Korea, 2022); PT, Portugal (Almeida & Gomes, 2017); TR, Turkey (Ministry of Agriculture & Forestry of Turkey, 2022); USA-1, Washington, United States of America (Condezo-Hoyos et al., 2014); USA-2, United States of America (USDA, 2019), ZA, South Africa (South African Medical Research Council, 2018)

Minerals

Minerals such as potassium, magnesium, calcium, sodium, and phosphorus, and trace elements such as zinc, manganese, copper, and iron were found in apple from various regions, reported by some studies (Jemaneh & Chandravanshi, 2021; Petkova et al., 2019). In Table 1, potassium dominates the mineral composition of apple, ranged from 99.46 to 119.00 mg/100 g. This was in agreement with Akšić et al. (2022), who found that potassium and zinc are two dominant minerals in apples grown in Norway. Sachini et al. (2020) reported that potassium from apple cultivars from southern Brazil contributed high to the percentage of daily recommendation value, followed by phosphorus and magnesium, while calcium contributed lower. However, the mineral content varied depending on the cultivars, production cycle, and growing region (Sachini et al., 2020). In apples grown in Ethiopia, and imported apples to Ethiopia from Israel and South Africa, the varied mineral contents between red and green apples were observed (Jemaneh & Chandravanshi, 2021). It was highlighted that green apple fruits had higher calcium and aluminum content (1065–36,275 mg Ca/kg, 77.8–129 mg Al/kg) than red apple fruits (1013–36,143 mg Ca/kg, 52.5–89.6 mg Al/kg). In contrast, nickel content in green apple fruits were lower than in red apple fruits. The authors also reported that the analyzed red and green apples did not contain non-essential toxic metals cadmium and lead. It is worth noticing that according to European regulation (1881/2006/EC), the concentration of lead and cadmium in fruit is limited at 0.10 and 0.05 mg/kg wet weight, respectively (European Commission, 2006).

In apple products, minerals, especially potassium and calcium, together with polysaccharides and proteins contributed to the haze formation of apple cider, juice, and pommeau (alcoholic drink) by taking a role as ligands for polyphenols (Millet et al., 2017).

Vitamins

Various vitamins were present in the apple, including vitamins A, B_1, B_2, B_3, B_5, B_6, B_9, C, and E (Table 1). Those vitamins take a role in human's body metabolism and immune support to protect against infection problems, inflammation, and possibly some cancers (Alpert, 2017). Vitamin A has a role in immune system, particularly in innate and cell-mediated immunity and humoral antibody responses (Alpert, 2017). The carotenoids were found in higher content in the apple peel as it contributes to the fruit coloration together with chlorophyll (Delgado-Pelayo et al., 2014), while in flesh, the concentration of carotenoid is low (Acquavia et al., 2021). Feliciano et al. (2010) found that the β-carotene (provitamin A) content of unpeeled Bravo de Esmolfe apple was almost twice higher than that of the peeled one. B complex vitamins are important to ensure that the body's cells working appropriately; among them, vitamin B_3 is the most abundant vitamin found in apple fruit (Table 1). Vitamin B_3 plays some roles in lipid metabolism, synthesis of HDL (high-density lipoprotein) cholesterol, DNA synthesis, blood glucose regulation, and reducing the risk of cardiovascular disease (Andrade et al., 2018). Vitamin B_9 (folate) was also found in apple (3–9 µg/100 g) (Table 1). Folate deficiency has been associated to some health problems, such as Alzheimer's and cardiovascular disease, osteoporosis, increased breast and colorectal cancer, poor cognitive ability, abnormalities in white and red blood cells, and hearing loss (Alpert, 2017). Vitamin C contained in apples can improve the human immunity, prevent anemia, and act as antioxidant and anti-aging agents (Patocka et al., 2020). Vitamin E (e.g., α-tocopherol) has also been known as a lipid-soluble antioxidant which has the ability to scavenge reactive oxygen species (ROS) (Mutalip et al., 2018). However, in comparison to polyphenols, vitamin C and E contributed less to the biological impact of apple (Feliciano et al., 2010). The vitamin C content of various apple cultivars has been reported by several studies. All analyzed 15 apple cultivars expressed higher vitamin C content in peel than flesh (Kschonsek et al., 2018). Between Granny Smith, Fuji Rose, and Golden Delicious cultivars, the highest content vitamin C content was found in Golden Delicious (31.48 mg/100 g), while the lowest one was found in Granny Smith (14.97 mg/100 g) (Asale et al., 2021). Furthermore, in the study about the effect of location within tree canopy on apples chemical compositions, it was found that the ascorbic acid contents of various apple peels from outer-canopy were 1.6–1.9 times higher than from the inner ones (Feng et al., 2014). It is also worth noticing that apple peels contained higher content of vitamin C than the flesh (Hamadziripi et al., 2014). For vitamin E, the content α-tocopherol, which is the fat-soluble vitamin most abundantly found in apple, was analyzed in the 24 apple cultivars harvested in South Tyrol, Italy (Bianchi et al., 2020). Among them, the authors found that the lowest α-tocopherol content was found in Freiherr von Berlepsch cultivar (0.13 mg/100 g FW), while the highest one was detected in Brixner Plattling cultivar (0.33 mg/100 g FW).

Phenolics

The phenolic composition (Fig. 2) of apple flesh and peel from various cultivars have been reported by several studies, summarized in Table 2. The major phenolic groups found in apple are hydroxybenzoic acids (gallic acid, protocatechuic acid), hydroxycinnamic acids (chlorogenic acid, caffeic acid, *p*-coumaric acid, ferulic acid, sinapic acid), flavanols (catechin and epicatechin) and their oligo- (procyanidins) and polymeric structures (proanthocyanidins), flavonols (quercetin, isoquercetin, rutin, kaempferol, astragalin), dihydrochalcones (phloridzin, phloretin-xyloglucoside), and anthocyanins (cyanidin-3-galactoside). However, a study also reported the other phenolic groups, such as flavones and isoflavones contents in apple (Alarcón-Flores et al., 2015). Recently, many studies reported the phenolic composition of various apple cultivars grown in different countries or regions such as Himalaya (Raj et al., 2021), Australia (Bondonno et al., 2020), Austria (Oszmiański et al., 2020), Bosnia and Herzegovina (Akagić et al., 2019), China (Wang et al., 2023), Costa Rica (Navarro et al., 2018; Navarro-Hoyos et al., 2021), Croatia (Lončarić et al., 2021), Italy (Tarola et al., 2019; Wandjou et al., 2020a, b), Japan (Sato et al., 2017), Lithuania (Raudone et al., 2017), Moldova and Romania (Geană et al., 2021), Nepal (Pandey et al., 2020), Norway (Akšić et al., 2022), Poland (Oszmiański et al., 2018), and Portugal (Pires et al., 2018). The phenolic composition of apple may differ from one to another depending on growing location, genetic and cultivars, and environmental factors (Oszmiański et al., 2018). Additionally, the difference in natural antioxidant composition between cultivars, including phenolics, as well as physicochemical properties also affect the effectiveness of enzymatic browning inhibition in apple products (Arnold & Gramza-Michałowska, 2022).

A study also reported that mechanical vibration during transport (28 Hz, 6 h) and storage (14 days, 6 °C) of apple might affect the quality and physicochemical parameters of selected apple cultivars (Gala, Idared, Topaz, and Red Prince), including total phenolic content (TPC), antioxidant capacity, pH value, color, firmness, total soluble solids and dry matter (Walkowiak-Tomczak et al., 2021). Increased

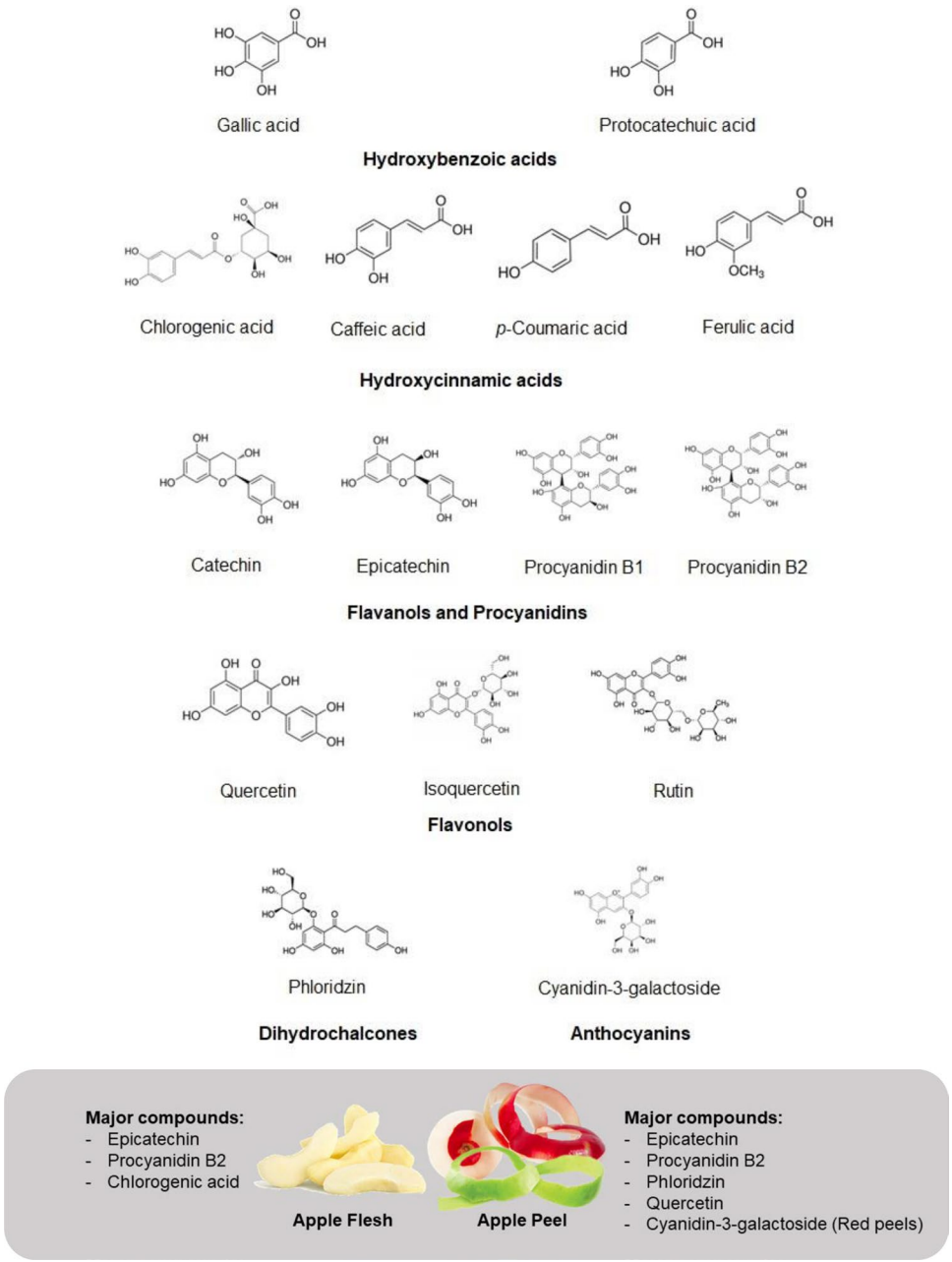

Fig. 2 Chemical structures of major phenolic compounds found in apple fruits

TPC and antioxidant potential of all cultivars were observed after the mechanical vibration and storage, indicating that after fruit transport and storage, the apples still remain a good source of bioactive compounds.

Among all phenolic compounds, in Australian apples, the total procyanidins (including flavanols and procyanidins) predominated the phenolic composition in the flesh (51.5%), followed by hydroxycinnamic acids (39.6%), while in peel, flavonols predominated (66.7%), followed by total procyanidins (24.7%) (Fig. 3A) (Bondonno et al., 2020). For apple cultivars from Southeast Europe, flavanols and procyanidins contributed more than 70% of total polyphenols in flesh and peel; phenolic acids were mainly present in flesh (6–25%); flavonols and dihydrochalcones accounted 1–13% and 1–10%, respectively in the peel; and 1–7% of anthocyanins were found in the red apples peels (Jakobek et al., 2013). Furthermore, in apples grown in Germany, Kschonsek et al. (2018) found that in apple peel, flavonols predominated the composition by more than 70%, followed by flavanols, phenolic acids, and phloridzin, while phenolic acids (more than 40%) predominated in the flesh, followed by flavanols and phloridzin, with missing flavonols (Fig. 3B). It is also worth noticing that apple peels generally had 5 times higher content of total individual phenolics than apple flesh (Kalinowska et al., 2020).

Hydroxybenzoic Acids

In hydroxybenzoic acids group, the gallic acid content in the apples was reported by several studies. Kschonsek et al. reported that there was no difference in gallic acid content between the fleshes of old and new cultivars in Germany, while in the peel, old cultivars possessed higher content than new ones (Kschonsek et al., 2018). Furthermore, based on the finding from Kschonsek et al. (2018) (see Fig. 3B), it was found that the contribution of hydroxybenzoic acids to all analyzed phenolics was 2.3% in the flesh and 7.8% in the peel, but the hydroxybenzoic acids content of the peel was about 3 times higher than the content in the flesh. A study about fruit bagging reported that fruit bagging did not significantly decrease gallic acid content in Red Delicious apple peel ($p > 0.05$), but decreased gallic acid content in Golden Delicious and Red Gala apple peels significantly ($p < 0.05$) (Chen et al., 2012). Besides gallic and protocatechuic acid, other hydroxybenzoic acids such as syringic (0.9 to 2.8 mg/100 ml) and vanillic acid (1.1 to 31.4 mg/100 ml) were reported in apple cultivars grown in Van region, Turkey (Celik et al., 2018).

Hydroxycinnamic Acids

For hydroxycinnamic acids group, as seen in Fig. 3A, it contributed more to the flesh (39.6%) than the peel (1.5%) of Australian apples. Many studies reported the hydroxycinnamic acids composition in apples (Table 2). Generally, chlorogenic acid has been reported by previous studies to predominate the hydroxycinnamic acids composition in apple. The hydroxycinnamic acid composition in the peel of Spanish cultivar Verde Doncella was dominated by chlorogenic acid (around 55%), followed by *p*-coumaric acid (34%) and caffeic acid (11%) (Krawitzky et al., 2014). The total hydroxycinnamic acids of this cultivar was lower than Red Delicious cultivar (Krawitzky et al., 2014). Wang et al. also found chlorogenic acid, together with catechin, phloretin, and quercetin, as the dominant phenolics in core, flesh, and peel of 20 apple cultivars in China (Wang et al., 2023). It is worth noticing that chlorogenic and caffeic acid were found in higher levels in the peels of red to dark-red-peeled apples, while procyanidins and flavanols were found in lower levels compared to the peels of green-peeled apples (De Paepe et al., 2015). Unfortunately, chlorogenic acid, together with flavanols and procyanidins were reported to be a good substrate for polyphenol oxidase (PPO), the enzymatic browning in apple (Kschonsek et al., 2019; Tinello et al., 2018), which can reduce the quality of the apple. The authors indicated that Szampion apple is good to produce light-colored juices and products due to its lower catechin, chlorogenic acid contents, and PPO activity in comparison to other cultivars. An article, however, did not detect the chlorogenic acid in the peels of Ervin Spur and Granny Smith, and caffeic acid in the peels of all analyzed cultivars (King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith) (Karaman

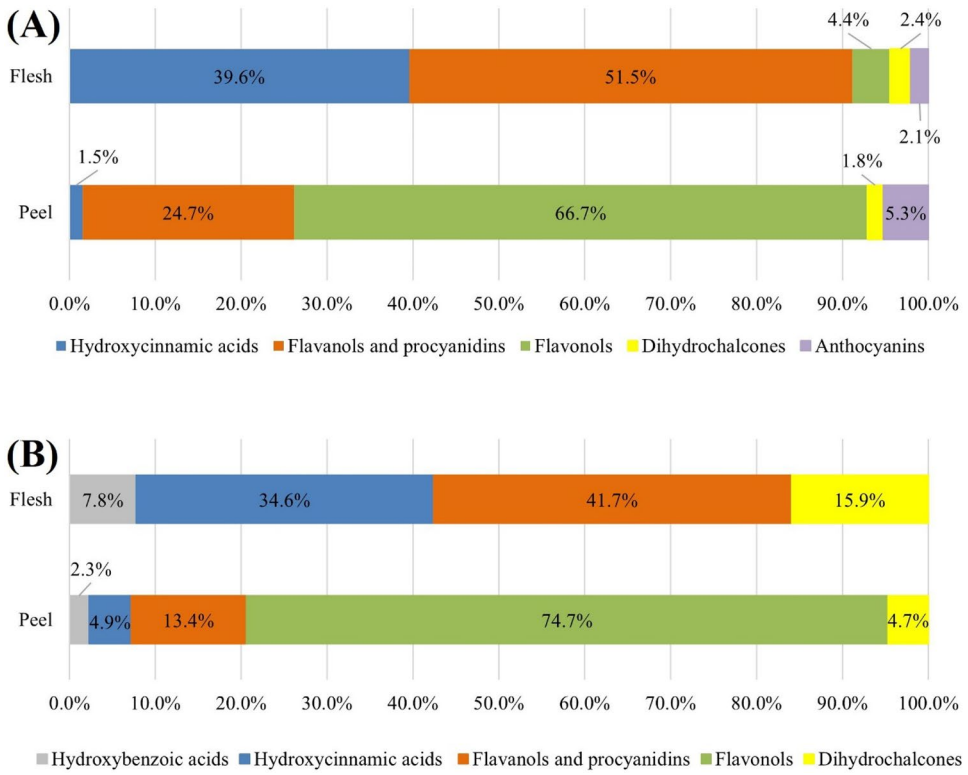

Fig. 3 The contribution of phenolics in flesh and peel of various apple cultivars grown in **A** Australia—average of 91 cultivars, adapted from (Bondonno et al., 2020) (percentage based on fresh weight of apples); **B** Germany—average of 15 cultivars, adapted from (Kschonsek et al., 2018) (percentage based on weight of freeze-dried apples). Notes: Hydroxybenzoic acids content was not evaluated in (**A**). Anthocyanins content was not evaluated in (**B**); Flavonols were not detected in the flesh in (**B**)

et al., 2013). Furthermore, Chen et al. (2012) found that bagging treatment lowered the concentration of hydroxycinnamic acids (chlorogenic, caffeic, *p*-coumaric, gallic, and ferulic acid) in the peels of three apple cultivars (Golden Delicious, Red Delicious, Red Gala), except the chlorogenic acid in Red Delicious apple peel ($p > 0.05$). While in the flesh, the result varied in all cultivars.

Flavanols and Procyanidins

The composition of flavanols such as catechin, epicatechin, and procyanidins in apple flesh and peel have been reported by many studies (Table 2). In Fig. 3A, flavanols and procyanidins contributed 51.5% in the flesh, more than in the peel (24.7%) of Australian apples. However, about the content, Feng et al. (2014) found that higher levels of flavanols and procyanidins were found in the peels compared to the flesh in all analyzed cultivars (McIntosh, Gala, and Mutsu). The authors also reported that McIntosh apple (flesh and peel) from outer-canopy showed higher catechin, epicatechin, and procyanidin B2 contents compared to that from inner-canopy; while procyanidin B1 content was similar, regardless the position (Feng et al., 2014). The average catechin and epicatechin contents were respectively 3.4 and 5.2 times more in the peel than the flesh in old cultivars in Germany. While in new cultivars, the bigger difference was found as the catechin and epicatechin contents were 4.9 and 10.1 times more in the peel than the flesh (Kschonsek et al., 2018). Furthermore, among 11 first-quality grade apple cultivars, a study reported that epicatechin predominated the flavanols and phloridzin composition in both flesh and peel (Zielińska & Turemko, 2020). The authors also reported that the epicatechin content in the peel ranged from 94.79 µg/g FW (Delikates) to 297.77 µg/g FW (Quinte), while in the flesh ranged from 4.99 µg/g FW (Gloster) to 325.04 µg/g FW (Quinte). On the other hand, Panzella et al. (2013) found that procyanidin B2 predominated the flavanols composition of all apple cultivars (whole fruit) from Southern Italy, ranged from 11.87 mg/100 g FW (Gold Chief® Gold Pink) to 83.12 mg/100 g (Cape ‘e Ciuccio). Similar result was also found in all 10 apple cultivars grown in Austria (Oszmiański et al., 2020). In comparison to Red Delicious cultivar, Verde Doncella cultivar from Spain showed lower concentrations of flavanols and quercetin derivates in peel, pomace, and juice (Krawitzky et al., 2014).

The flavanols content of four new red-fleshed cultivars (RS-1, Redlove Era 107/06, 117/06, and 119/06) and white-fleshed cultivars (Brookfield Gala, Zhen Aztec Fuji, Story, Golden Smoothee, and Granny Smith) was compared by a study (Bars-Cortina et al., 2017). The authors observed that the flavonols content of flesh and peel of red-fleshed cultivars were lower than those of white-fleshed cultivars, which might be due to the competitive synthesis between anthocyanins and proanthocyanidins via flavonoid pathway. With a competitive interaction from substrate between anthocyanidin reductase and anthocyanidin synthase enzymes, it could result in the lower flavanols and proanthocyanidins contents in the red-fleshed cultivars, which accordingly contain higher anthocyanins content than white-fleshed cultivars.

A study in Japan found that pale-red-fleshed cultivars, such as Nakano Shinku and Nakano no Kirameki, possessed high content of catechin and epicatechin; deep-red-fleshed cultivars, such as Chouka 4 and JPP35, were characterized by high accumulation of cyanidin-3-*O*-galactoside; and white-fleshed cultivars (Fuji and no. 37) showed the lowest phenolic compounds compared to others (Sato et al., 2017). The authors suggested to also consider the development of pale-red-fleshed cultivars in functional food although the deep-red-flesh cultivars are the major developed cultivars in Japan.

Regarding food safety, unfortunately, a study found that the apples with higher contents of catechin, epicatechin, and gallic acid were positively correlated to higher content of patulin (Lončarić et al., 2021). The authors added that these results can be explained due to pro-oxidative impact of flavanol, which leads to the formation of ROS in *Penicillium expansum* cells and activated the cellular antioxidant defense system and stimulated the production of patulin to reduce the ROS level inside the cells. However, the presence of phloridzin may reduce the fungal infection in apple (see the “Dihydrochalcones” section), that may also reduce the patulin formation.

Flavonols

Flavonols contributed more to total phenolics in Australian apples peels (66.7% of phenolic composition), while in the flesh only 4.4% (Fig. 3A). The other authors even highlighted that the flavonols were missing in the flesh of various apple cultivars in Germany (Kschonsek et al., 2018). It was in agreement with several studies that did not detected the quercetin in the flesh of any analyzed apple cultivars (Karaman et al., 2013; Zielińska & Turemko, 2020). However, Alarcón-Flores et al. (2015) detected quercetin in the flesh of all apple cultivars, ranged from 2.5 mg/kg DW (Golden (France)) to 4.2 mg/kg DW (Royal). Raudone et al. still detected quercitrin (quercetin-3-rhamnoside) and rutin (quercetin-3-*O*-rhamnoside) in the flesh of six Lithuanian cultivars by using high-performance liquid chromatography (HPLC) method; however, both quantities were very low and did not significantly contribute to the reducing activity (Raudone et al., 2017). De Paepe et al. (2015) studied that overall classic/new cultivars of apple, grown in Belgium, were typified by higher flavonols content, and lower contents of dihydrochalcones, proanthocyanidins, and chlorogenic acid.

Besides quercetin, isoquercetin, and rutin (Table 2), some articles reported the presence of kaempferol derivatives (including kaempferol glucoside or astragalin, kaempferol pentoside, kaempferol rhamnoside, etc.) (Alarcón-Flores et al., 2015; Jakobek et al., 2013), isorhamnetin derivatives (isorhamnetin galactoside, isorhamnetin glucoside) (Oszmiański et al., 2020), and myricetin (Alarcón-Flores et al., 2015) in some apple cultivars. Not only in fruits, quercetin and rutin were also found in peel, bark, and stem bark of apple cultivars originated from Jumla and Mustang, Nepal (Pandey et al., 2020). Among the analyzed samples, Richard apple peel from Mustang expressed the lowest quercetin (2.66 mg/100 g DW) and rutin (3.59 mg/100 g DW), while the highest quercetin and rutin contents were found in Chocolate stem bark from Jumla (171.05 mg/100 g DW) and Jonathan stem bark from Jumla (374.50 mg/100 g), respectively. The peel of Lještarka apple from Croatia were reported to have the highest total flavonols (1222.7 mg/kg FW), in comparison to other analyzed cultivars from Croatia and USA (Jakobek & Barron, 2016). The authors also found that the peel of red-peeled apples generally had higher flavonols content than the peel of yellow-green-peeled apples.

Dihydrochalcones

Dihydrochalcones, e.g., phloridzin, contributed to the total phenolics slightly higher in the flesh than peel of Australian apples (Fig. 3A), but phloridzin was found in higher content in apple peel (8.4 mg/100 g FW; ranged from 1.7 to 31.2 mg/100 g FW) than apple flesh (0.7 mg/100 g FW; ranged from 0.0 to 2.9 mg/100 g FW) (Bondonno et al., 2020). On the other hand, a study found that the phloridzin content was higher in the flesh than the peel, particularly in the Cox Orange, Ontario, and Roter Boskoop cultivars grown in Germany (Kschonsek et al., 2018). Besides phloridzin, which was reported by many studies (Table 2), other dihydrochalcones are also found in apple, for example, phloretin-2′-*O*-xyloglucoside and both of them are the major dihydrochalcones found in Finnish apple juice (He et al., 2022).

Phloridzin is one of the responsible bioactive compounds in apple that exhibits antidiabetic activity as it reduces intestinal sugar uptake by inhibition of Na/glucose cotransporter 1, which then is beneficial for people with type 2 diabetes mellitus (Niederberger et al., 2020). Apples, in the form of juice and fruit, have been the main contributors of phloridzin intake in European consumers (0.7–7.5 mg phloridzin/day) (Niederberger et al., 2020). Other than that, phloridzin can also be the chemotaxonomy marker for apple cultivar identification as well as its products, i.e., to identify the fraudulent admixtures of apple juice (Liaudanskas et al., 2015). In terms of food safety, phloridzin is the major polyphenol responsible for resistance to fungal infection, which is explained by the formation of the hydrolyzed product, phloretin, that oxidizes and forms fungitoxic *o*-quinone (Lončarić et al., 2021).

Anthocyanins

Anthocyanins contributed to total phenolics more in the peels (5.3%) than in the flesh (2.1%) of apples grown in Australia (Fig. 3A). The main anthocyanin found in apple is cyanidin-3-galactoside (Meng et al., 2015). However, cyanidin-hexoside, cyanidin-pentoside, and other cyanidin derivatives were also reported (Jakobek et al., 2013). Anthocyanins content was correlated to the *a**-value (redness) of apple (Sethi et al., 2020). Although most articles reported that anthocyanins were found only in the red peel, several articles reported that anthocyanins were found in the flesh of several cultivars in Australia (Bondonno et al., 2020) and red-fleshed apples, such as Ljubeničarka which is grown in Southeast Europe (318.9 mg/kg FW) (Jakobek et al., 2013), and other apple species from China—*Malus pumila* Niedzwetzkyana (Dieck) (84.28 mg/kg FW) (Katiyo et al., 2018) and. Zielińska and Turemko (2020) also detected cyanidin-3-galactoside in the flesh of all analyzed apple cultivars (low level) and the peel of all analyzed apple cultivars (including the yellow-green-peeled apples—Papierówka and Antonówka, low level). Karaman et al. (2013) did not detect the cyanidin in the flesh of all analyzed cultivars (King Luscious, Amasya, Ervin Spur, Sky Spur, Arap Kizi, Lutz Golden, Granny Smith), but it was detected in the peel of all cultivars, except Lutz Golden and Granny Smith. Furthermore, Jakobek and Barron (2016) only detected the cyanidin-3-galactoside in several red-peeled apple peels (Lještarka, Božićnica, and Cortland), but not in neither the other analyzed red-peeled nor yellow-green-peeled apples peels.

Other Compounds

Other compounds, such as pigments (chlorophyll and carotenoids) in apple were studied (Delgado-Pelayo et al., 2014). The authors found that the most abundant compounds in the flesh and peel of 13 apple cultivars (yellow/red/green-peeled apples) were diesterified xanthophylls and chlorophyll *a*. In the flesh, diesterified xanthophylls ranged from 3.98 µg/g DW (green-peeled apple—Granny Smith) to 25.15 µg/g DW (yellow-peeled apple—Golden Montaňa); while in the peel ranged from 5.04 µg/g DW (green-peeled apple—Granny Smith) to 38.39 µg/g DW (red-peeled apple—Ariane). For chlorophyll *a*, in the flesh, it ranged from 0.81 µg/g DW (red-peeled apple—Fuji from France) to 47.00 µg/g DW (green-peeled apple—Granny Smith); while in the peel ranged from 18.39 µg/g DW (yellow-peeled apple—Golden Rosett) to 1049.26 µg/g DW (green-peeled apple—Granny Smith). Furthermore, the authors also explained that green-peeled apple cultivars possessed high chlorophyll content

in the peel; however, it was also found in high level in some red-peeled apple cultivars, such as Fuji from Italy, Pink Lady®, and Starking Red Chief. While yellow-peeled apple cultivars (Golden Delicious, Golden Montaňa, and Golden Rosett) lack of chlorophyll (Delgado-Pelayo et al., 2014).

To sum up, with the updated knowledge of qualitative and quantitative content of chemical composition in various apple cultivars, the consumers and apple products industries can select the promising apple cultivars that are rich in nutritional values to be consumed as direct consumption or production of apple products. In selecting the promising cultivar, it is also important to consider its availability in consumers' region as it is related to the price of the apples. The apple cultivar with higher availability and affordable price yet is still rich in nutritional values can contribute to wider scale of consumption to promote general health of consumers. For apple products industries, e.g., apple juice industries, the by-products from production process such as apple pomace or peels that still contains valuable bioactive compounds can open a wider range of production instead of just being a waste.

Total Phenolics and Antioxidant Activity

Apples have been studied to possess various bioactive compounds which showed antioxidant activity. Many articles reported the total phenolic content (TPC), total flavonoid content (TFC), and antioxidant activity of the extracts of apple cultivars from different countries (Table 3). TPC is commonly determined using Folin-Ciocalteu assay that is well-known, simple, and reproducible, yet there are some drawbacks that need to be concerned, such as pH sensitivity, temperature, reaction time, and selectivity (Zhong & Shahidi, 2015). Regarding its selectivity, the overestimation of total phenolics may happen as nonphenolic reducing agents, such as reducing sugars and amino acids that present in the extract could affect the result of TPC. Therefore, HPLC could also be conducted to confirm the TPC result.

Generally, studies reported higher TPC and TFC were found in the peel than the flesh. The barrier function of the peels against the external biotic and abiotic stress might be the reason of this phenomenon (Kschonsek et al., 2018). In the study on 13 apple cultivars grown in India, Sethi et al. (2020) reported that the average TPC and TFC in the peel were respectively 2.8 and 2.68 times higher than those in the flesh. The similar result was found by Zhang et al. (2012), who found the order of part of fruits according to their TPC and TFC: peel > core > flesh, in apple cultivars grown in China.

A study on the whole fruit of 104 apple cultivars from Europe (Switzerland, Italy, Netherlands) reported the average TPC of 150 mg catechin equivalents (CAE)/100 g FW, ranged from 52 to 378 mg CAE/100 g FW (Ceymann et al., 2012). The content was comparable and could be more specifically described by Zielińska and Turemko (2020) as they found TPC of the peels of Polish apple cultivars, ranged from 182 mg CAE/100 g FW (Ligol) to 328 mg CAE/100 g FW (Quinte); while the TPC of flesh ranged from 54 mg CAE/100 g FW (Ligol) to 174 mg CAE/100 g FW (Quinte). In terms of TFC, in the peel, it ranged from 55 mg CAE/100 g FW (Ligol) to 130 mg CAE/100 g FW (Paulared); while in the flesh, it ranged from 18 mg CAE/100 g FW (Ligol) to 83 mg CAE/100 g FW (Papierówka). It is also noteworthy that the TPC and TFC are not always higher in peel than the flesh. Several cultivars, such as Topaz (Italy) had higher TPC (Tarola et al., 2019) and Royal Delicious (India) had higher TFC (Sethi et al., 2020) in the flesh than peel. Furthermore, a study reported that the mild heat treatments (40–50 °C) increased 70% TPC of Eva apple fruit, and at the same time reduced the browning activity (Rodríguez-Arzuaga et al., 2019). It is noteworthy that higher TPC might be related to higher browning activity because phenolics are the substrates of PPO. However, in Eva apple, the higher TPC did not increase the browning development as the reduction PPO activity of Eva apple was observed after the mild heat treatment.

Various assays were conducted by recent studies on various parts of fruit and cultivars (Table 3). In brief, the antioxidant activity can be analyzed using (a) reactive oxygen species (ROS) scavenging-based (e.g., 2,2-diphenyl-1-picrylhydrazyl—DPPH, 2,2′-azinobis-(3-ethylbenzothiazoline-6-sulfonic acid—ABTS, oxygen radical absorbance capacity—ORAC, and chemiluminescence), (b) redox potential-based assays (e.g., ferric reducing antioxidant power—FRAP, cupric ion reducing antioxidant capacity—CUPRAC, and cyclic voltammetry method—CV), and (c) metal chelation capacity (e.g., chelating of ferrous ion—CA) (Zhong & Shahidi, 2015). Compared to the other assays or methods, ORAC assay is considered more relevant as a reference for antioxidant effectiveness as it uses biological relevant oxidant—peroxyl radicals, that is generated by azo compounds, commonly the hydrophilic 2,2′-azobis(2-amidinopropane) dihydrochloride (AAPH) (Zhong & Shahidi, 2015). Those different methods or assays were conducted by previous studies in apples as each assay has different hydrophilicity or lipophilicity on natural antioxidants in apples as described in the "Phenolics" and "Other Compounds" sections.

Generally, the antioxidant activity of apple's extract is in accordance with its TPC. The correlation between antioxidant activity and sum of polyphenols or phenolic acids in apple can be analyzed. For examples, using Pearson correlations, the sum of polyphenols of Austrian apple cultivars had high correlation to ABTS, DPPH, and FRAP with r-value of 0.89, 0.95, and 0.93 ($p < 0.05$), respectively; while the total phenolic acids also had high correlation to those respective assays, with r-value of 0.69, 0.70, and 0.68 ($p < 0.05$) (Oszmiański et al., 2020).

Table 3 Content of total phenolics, total flavonoids, total anthocyanins and antioxidant potential of various apple cultivars

Analyzed cultivars	Country	Part of fruit	Assay	Findings	References
Fuji Rose, Golden Delicious, Granny Smith	Ethiopia	Whole fruit	TPC	H: Granny Smith ↔ Fuji Rose; L: Golden Delicious	Asale et al. (2021)
			TFC	H: Fuji Rose ↔ Granny Smith; L: Golden Delicious	
			DPPH	H: Fuji Rose ↔ Granny Smith; L: Golden Delicious	
			FRAP	H: Fuji Rose; L: Golden Delicious	
Gala, Golden Delicious, Granny Smith, Red Delicious, Starking	Mexico	Whole fruit	TPC	H: Granny Smith; L: Golden Delicious	Corona-Leo et al. (2021)
			TFC	H: Starking; L: Gala	
			TAC	H: Red Delicious; L: Golden Delicious ↔ Granny Smith (not detected)	
			ABTS	H: Red Delicious; L: Granny Smith	
			DPPH	H: Red Delicious; L: Starking	
Beliy Naliv, Crestesc, Florina, Gloster, Golden Delicious (Moldova), Golden Delicious (Romania), Idared, Jonathan (Moldova), Jonathan (Romania), Montuan, Renet Simirenco, Richard, Spartan, Starkrimson	Moldova and Romania	Whole fruit	TPC	H: Starkrimson; L: Gloster ↔ Montuan	Geană et al. (2021)
			TFC	H: Beliy Naliv ↔ Montuan ↔ Jonathan (Moldova); L: Idared ↔ Gloster ↔ Golden Delicious (Romania)	
			DPPH	H: Starkrimson ↔ Jonathan (Romania) ↔ Beliy Naliv; L: Montuan ↔ Gloster	
Anna, Jonagold	Costa Rica	Flesh	TPC	H: Jonagold; L: Anna	Navarro-Hoyos et al. (2021)
			DPPH	H: Jonagold; L: Anna	
			ORAC	H: Jonagold; L: Anna	
		Peel	TPC	H: Anna; L: Jonagold; Peel > Flesh for Anna; Peel < Flesh for Jonagold	
			DPPH	H: Anna; L: Jonagold; Peel > Flesh for Anna; Peel < Flesh for Jonagold	
			ORAC	H: Anna; L: Jonagold; Peel > Flesh for Anna; Peel < Flesh for Jonagold	
Berner Rose, Brünnerling Type 1, Brünnerling Type 2, Brünnerling Type 4, Croncels Type 2, Galloway Pepping, Goldgelbe Sommerrenette, Salzburger Type 1, Schmidberger Renette, Transparente de Croncels	Austria	Whole fruit	ABTS	H: Brünnerling Type 1; L: Brünnerling Type 2	Oszmiański et al. (2020)
			DPPH	H: Brünnerling Type 1; L: Schmidberger Renette	
			FRAP	H: Brünnerling Type 1; L: Schmidberger Renette	

Table 3 (continued)

Analyzed cultivars	Country	Part of fruit	Assay	Findings	References
Gale Gala, Oregon Spur II, Red Chief, Royal Delicious, Royal Gala, Scarlet Gala, Scarlet Spur I, Silver Spur, Starkrimson, Super Chief, Top Red, Vance Delicious, Well Spur	India	Flesh	TPC	H: Scarlet Spur I; L: Royal Delicious	Sethi et al. (2020)
			TFC	H: Royal Delicious; L: Royal Gala ↔ Gale Gala ↔ Red Chief ↔ Vance Delicious	
			CUPRAC	H: Silver Spur ↔ Oregon Spur II ↔ Well Spur ↔ Starkrimson ↔ Scarlet Spur I ↔ Scarlet gala; L: Red Chief ↔ Royal Delicious ↔ Royal Gala ↔ Super Chief ↔ Vance Delicious ↔ Top Red	
			DPPH	H: Oregon Spur II; L: Royal Gala ↔ Top Red ↔ Scarlet Gala ↔ Vance Delicious ↔ Super Chief ↔ Well Spur ↔ Starkrimson	
			FRAP	H: Silver Spur ↔ Oregon Spur II; L: Scarlet Gala ↔ Royal Gala ↔ Vance Delicious ↔ Well Spur ↔ Super Chief	
		Peel	TPC	H: Well Spur ↔ Oregon Spur II; L: Top Red ↔ Red Chief ↔ Scarlet Spur I; Peel > Flesh	
			TFC	H: Well Spur; L: Royal Delicious; Peel > Flesh (except Royal Delicious: Flesh > Peel)	
			CUPRAC	H: Top Red; L: Royal Delicious ↔ Scarlet Gala; Peel > Flesh	
			DPPH	H: Well Spur ↔ Royal Gala; L: Silver Spur ↔ Royal Delicious ↔ Vance Delicious; Peel > Flesh	
			FRAP	H: Oregon Spur II; L: Silver Spur ↔ Vance Delicious ↔ Scarlet Gala ↔ Top Red ↔ Starkrimson ↔ Super Chief ↔ Gale Gala; Varied results of Peel vs. Flesh	

Table 3 (continued)

Analyzed cultivars	Country	Part of fruit	Assay	Findings	References
Annurca, Mela Rosa dei Monti Sibillini (MR1—good shape, color, size; MR2—with malformations)	Italy	Whole fruit	TPC	H: MR1; L: Annurca	Wandjou et al. (2020b)
			ABTS	H: MR1; L: Annurca	
			DPPH	H: MR1; L: Annurca	
Antonówka, Delikates, Early Geneva, Gloster, Jonagored, Ligol, Paulared, Papierówka, Quinte, Rubinola, Sunrise	Poland	Flesh	TPC	H: Quinte; L: Ligol ↔ Gloster	Zielińska and Turemko (2020)
			TFC	H: Papierówka; L: Ligol	
			CA	H: Antonówka; L: Ligol	
			CV	H: Quinte; L: Ligol ↔ Gloster	
			DPPH	H: Quinte; L: Gloster	
			FRAP	H: Quinte; L: Gloster	
		Peel	TPC	H: Quinte; L: Ligol; Peel > Flesh	
			TFC	H: Paulared; L: Ligol; Peel > Flesh	
			CA	H: Antonówka; L: Ligol; Peel > Flesh (except Antonówka and Rubinola: Flesh > Peel)	
			CV	H: Paulared ↔ Jonagored; L: Antonówka ↔ Ligol; Peel > Flesh	
			DPPH	H: Jonagored; L: Antonówka; Peel > Flesh	
			FRAP	H: Quinte; L: Ligol; Peel > Flesh	
Ancaiano, Campo Pianacce, Casolana, Ficareto, Filare Delle Pianacce, Gialla Pianacce, Golden Delicious, Piatta Cantine, Podere Pianacce, Red Delicious, Rossa Casetta, Rugginosa Delle Pianacce, Solaio, Sotto Muro Casetta. Strada Pianacce, Tocchi, Tre Colli, Vecchio Pollaio, Vialle Casetta	Italy	Whole fruit	TPC	H: Solaio; L: Casolana	Berni et al. (2019)
			FRAP	H: Solaio; L: Red Delicious ↔ Golden Delicious ↔ Casolana ↔ Sotto Muro Casetta ↔ Strada Pianacce ↔ Rossa Casetta	
Braeburn, Cerina, Florina, Gelato Cola, Gold Rush, Jonagored, Limoncella, Renetta Osiris, Rosada, Topaz	Italy	Flesh	TPC	H: Topaz; L: Rosada	Tarola et al. (2019)
			DPPH	H: Jonagored; L: Rosada	
		Peel	TPC	H: Florina; L: Topaz; Peel > Flesh (except Topaz: Flesh > Peel)	
			DPPH	H: Topaz; L: Gold Rush; Peel > Flesh	
Braeburn, Berlepsch, Cox Orange, Dülmener Rosenapfel, Elstar, Golden Delicious, Goldparmäne, Granny Smith, Gravensteiner, James Grieve, Jonagold, Jonathan, Oldenburger, Ontario, Roter Boskoop	Germany	Flesh	TPC	H: Jonathan; L: Golden Delicious	Kschonsek et al. (2018)
			ABTS	H: Jonathan; L: Golden Delicious ↔ Braeburn	
			ORAC	H: Jonathan; L: Golden Delicious	
		Peel	TPC	H: Oldenburger; L: Golden Delicious; Peel > Flesh	
			ABTS	H: Oldenburger; L: Golden Delicious; Peel > Flesh	
			ORAC	H: Oldenburger; L: Golden Delicious; Peel > Flesh	

Table 3 (continued)

Analyzed cultivars	Country	Part of fruit	Assay	Findings	References
Altländer Pfannkuchenapfel, Boikenapfel, Booskop, Charlamowsky, Dulmener Rosenapfel, Geflammter Kardinal, Gelber Richard, Horneburger Pfannkuchenapfel, Kaiser Alexander, Kaiser Wilhelm, Landsbergen, Lausitzer Nelkenapfel, Renette, Parkers Pepping, Riesenboiken, Rote Sternrenette, Roter Delicious Type Starkinson, Roter Eiserapfel, Roter Herbstkalvill, Roter Trier Weinapfel, Weisser Winterkalvill, Wintergoldparman, Wintergoldparmane	Poland	Whole fruit	ABTS	H: Wintergoldparmane; L: Landsbergen Renette	Oszmiański et al. (2018)
			FRAP	H: Wintergoldparmane; L: Landsbergen Renette	
Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka	Lithuania	Flesh	TPC	H: Lodel ↔ Auksis ↔ Aldas; L: Rajka	Raudone et al. (2017)
			FRAP	H: Aldas ↔ Auksis; L: Ligol	
		Peel	TPC	H: Lodel ↔ Auksis ↔ Aldas; L: Rajka; Peel > Whole fruit > Flesh	
			FRAP	H: Aldas ↔ Auksis; L: Ligol; Peel > Whole fruit > Flesh	
		Whole fruit	TPC	H: Lodel ↔ Auksis ↔ Aldas; L: Rajka	
			FRAP	H: Aldas ↔ Auksis; L: Ligol	
Albrechtovo, Bernské Růžové, Hvězdnatá Reneta, Jadernička Moravská, Lebelovo, Matčino, Starkrimson, Vilémovo	Czech Republic	Flesh	PCL-TAC	All cultivars ↔	Balík et al. (2012)
		Peel	PCL-TAC	H: Bernské Růžové; L: Jadernička Moravská; Peel > Flesh	
Fuji, Golden Delicious, Guoguang, Wanglin	China	Core	TPC	H: Guoguang; L: Golden Delicious ↔ Fuji	Zhang et al. (2012)
			TFC	H: Guoguang; L: Golden Delicious ↔ Fuji	
			DPPH	H: Guoguang ↔ Wanglin; L: Golden Delicious	
			FRAP	H: Guoguang; L: Golden Delicious ↔ Fuji	
		Flesh	TPC	H: Guoguang; L: Golden Delicious	
			TFC	H: Guoguang; L: Golden Delicious	
			DPPH	H: Wanglin ↔ Guoguang ↔ Fuji; L: Golden Delicious	
			FRAP	H: Guoguang; L: Golden Delicious	
		Peel	TPC	H: Fuji; L: Wanglin ↔ Golden Delicious; Peel > Core > Flesh	
			TFC	H: Guoguang ↔ Fuji; L: Golden Delicious; Peel > Core > Flesh	
			DPPH	H: Fuji ↔ Guoguang ↔ Wanglin; L: Golden Delicious; Peel > Core > Flesh (Fuji and Golden Delicious); Core > Peel > Flesh (Guoguang and Wanglin)	
			FRAP	H: Fuji ↔ Guoguang; L: Golden Delicious; Peel > Core > Flesh (except Fuji: Peel > Flesh > Core)	

ABTS 2,2′-azinobis-(3-ethylbenzothiazoline-6-sulfonic acid), *CA* chelating of ferrous ion activity, *CUPRAC* cupric ion reducing antioxidant capacity, *CV* cyclic voltammetry, *DPPH* 2,2-diphenyl-1-picrylhydrazyl, *FRAP* ferric reducing antioxidant power, *H* the highest content (TPC and TFC) or the highest activity (antioxidant activity), *L* the lowest content (TPC and TFC) or the lowest activity (antioxidant activity), *ORAC* oxygen radical absorbance capacity, *PCL-TAC* photochemiluminescene-total antioxidant capacity, *TAC* total anthocyanins content, *TFC* total flavonoid content, *TPC* total phenolic content, ↔ not significantly different

CV method and different assays, such as CA, FRAP, and DPPH assays were conducted on apples from Poland (Zielińska & Turemko, 2020). It was observed that the peels of all cultivars had higher antioxidant potential than the flesh in CV, DPPH, and FRAP assays. By DPPH assay, the intermediate radical-scavenging activity of flesh was observed, ranged from 2.23 to 4.65 µmol Trolox equivalent (TE)/g FW for Gloster and Quinte, respectively. While in the peels, it ranged from 5.27 µmol TE/g FW (Antonówka) to 8.65 µmol TE/g FW (Jonagored). In FRAP assay, the peels of all cultivars expressed at least twice higher value than the flesh. Similar result was also found in CV assay, which is the rapid, simple, low-cost, and sustainable method to analyze the reducing capacity of antioxidants, in comparison to FRAP and DPPH assays. The CA of peels was also higher than that of fleshes in all cultivars, except Antonówka and Rubinola. The authors also discussed the correlation of phenolic compounds and antioxidant activity. It was reported that in the peels, TPC and TFC are positively correlated to the results of FRAP and CV, while cyanidin-3-galactoside was highly correlated to the results of DPPH. While in the flesh, TPC and TFC had strong correlation with FRAP, DPPH, CV, and CA. Additionally, epicatechin, chlorogenic acid, and cyanidin-3-galactoside had high and positive correlations with results of FRAP, DPPH, and CV; while with CA, the correlations were weaker.

Another study reported the antioxidant potential of apples cultivar in Germany using ABTS and ORAC assays (Kschonsek et al., 2018). Among 15 analyzed cultivars (5 new cultivars and 10 old cultivars), Oldenburger and Jonathan possessed the highest TPC and antioxidant potential by both ABTS and ORAC assays in the peel and flesh, respectively. Golden Delicious in contrast showed the lowest TPC and antioxidant potential by both mentioned assays. The other findings reported by the authors were about the comparison between old and new cultivars. The flesh of old cultivars had higher TPC and vitamin C content than of the new cultivars which led to the higher antioxidant capacity determined by ABTS and ORAC. While in the peel, higher content of vitamin C was found in the old cultivars, but there was no significant difference of TPC and antioxidant potential found between the old and new cultivars. This phenomenon could be explained as new cultivars possessed lower content of flavanols (in flesh), total phenolic acids (in flesh and peel), and phloridzin (in flesh and peel) than the old cultivars, whereas monomeric and oligomeric flavanols and phloridzin are strong antioxidants (Kschonsek et al., 2018). It is also worth noticing that vitamin C explains only around 0.4% of the total antioxidant capacity of apples (Feliciano et al., 2010). Similar result was also found by Asale et al. (2021), as the whole fruit of Golden Delicious apple possessed the lowest antioxidant potential by DPPH and FRAP assays. Although it contained the highest vitamin C content, but it contained the lowest TPC and TFC among analyzed cultivars grown in Southern Ethiopia.

Wandjou et al. explored the antioxidant composition and activity of ancient apple cultivar Mela Rosa dei Monti Sibillini (MR) from Sibillini Mountains, central Italy (Wandjou et al., 2020b). The authors compared two choices of MR: first choice which has good shape, color, and size, while second choice is normally discarded due to unpleasant shape and color. Interestingly, the second choice showed majorly higher polyphenols and triterpenes when HPLC–DAD analysis was performed, yet possessed a bit lower TPC (Folin-Ciocalteu method) and antioxidant activity (ABTS and DPPH assays) than the first choice, indicating that the MR apples with malformation are also potential to be reused and recovered from nutraceutical and cosmeceutical properties. In another publication, the same authors also compared the antioxidant composition and activity of MR (pulp and peel) in freeze-dried and dried (45 °C) forms (Wandjou et al., 2020a). Generally, the lyophilized apple showed higher antioxidant potential than the dried materials. Also higher levels of phenolics, especially catechin, epicatechin, procyanidin B2, phloridzin, and triterpenes are found the peel extract than pulp extract.

In Czech Republic, the antioxidant capacity mean of apple peels was approximately 7 times higher than that of the flesh, using photochemiluminescene (PCL) method (Balík et al., 2012). The value ranged from 19.11 mmol TE/kg FW (Jadernička Moravská) to 56.65 mmol TE/kg FW (Bernské Růžové) in the peel. While in the flesh, it ranged from 4.02 mmol TE/kg FW (Vilémovo) to 6.59 mmol TE/kg FW (Bernské Růžové), but there was no significant difference between them. While in Podlasie province of Poland, Kalinowska et al. (2020) observed the TPC and antioxidant activity (DPPH, ABTS, FRAP, CUPRAC) of the peel's extract of new, scab-resistant cultivar—Gold Milenium, in comparison to that of old, scab-sensitive cultivar—Papierówka. The results showed that the peel's extract of Papierówka possessed higher TPC, antioxidant activity than that of Gold Milenium. This result maybe correlated to the findings of Shah and Gupta, who found that secondary metabolites (including antioxidants) were synthesized in scab-infected apple peel as a defensive response, which resulted in higher TPC and TFC in comparison to non-infected apple peel (Shah & Gupta, 2023).

Corona-Leo et al. (2021) approached the antioxidant potential (ABTS and DPPH assays), TPC, TFC, and total anthocyanins content of five apple cultivars (Gala, Golden Delicious, Granny Smith, Red Delicious, and Starking) in Mexico from their bioaccessibility index after in vitro digestion. The results showed that the antioxidant activity, TPC, TFC, total anthocyanins content after in vitro digestion were significantly lower than those before digestion, indicating that those values before digestion may overestimate the

bioavailability of the antioxidants. The other publications about antioxidant bioaccessibility of apples after in vitro digestion are summarized in Table 4.

Considering the higher TPC, TFC, and antioxidant potential in the peels of most apple cultivars, it is suggested to consume the fruit with the peels. It does not mean that apple peel contributed to phenolic intake more than the flesh as the peel represents only around 10% of the whole fruit and it is not always consumed. The apple flesh then contributes more to the phenolic intake of consumer. Therefore, apple cultivar which has higher phenolic content and antioxidant potential in the flesh is suggested to improve the phenolic contents in consumer's body. Additionally, it is also important to consider the bioavailability of the antioxidants in apples.

Apple Phenolics Extraction

Phenolics are the important metabolites of apple as they could show health-promoting effects when ingested. The phenolic extraction procedure should be accurate and precise to maximize the yield of targeted compounds and their antioxidant activity. Therefore, extraction might be the most critical step in the analysis of apple's metabolites, as the ideal extraction should recover all the targeted metabolites without any chemical addition (Acquavia et al., 2021).

Various apple products (juice, mash, pomace, whole fruit) were extracted using various extraction methods, such as conventional liquid–liquid extraction (LLE) and solid–liquid extraction (SLE), as well as development of SLE, such as microwave-assisted extraction (MAE), radio frequency-assisted extraction (RFAE), ultrasound-assisted extraction (UAE), supercritical fluid extraction (SFE), pulsed electric field (PEF)-assisted extraction, and pressurized liquid extraction (PLE) (Table 5). The independent factors that affect the phenolic extraction, such as type and concentration of solvent, temperature of extraction, sample to solvent ratio, particle size, and preparation or pretreatment of apple samples (grinding, milling, blending) were varied in the research design (Perussello et al., 2017). As the dependent factors, TPC, TFC, and antioxidant activities were then analyzed to find the optimum conditions or to be compared to conventional methods, such as Soxhlet extraction (Ferrentino et al., 2018), or simply compared to control (without developed treatment) (Lohani & Muthukumarappan, 2016; Quang et al., 2014). Some researchers statistically checked the optimization of extraction condition by employing response surface methodology (RSM) (Alberti et al., 2014; Franquin-Trinquier et al., 2014). The result could be confirmed by HPLC–DAD and LC–MS/MS (the sensitive and selective analysis) to check the phenolic compositions (Acquavia et al., 2021).

Conventional Extractions

Various types of products can be produced from apple fruit. The protocol could differ according to the product type. For liquid samples, such as apple juice, it could be analyzed directly after the filtration and/or centrifugation process (Acquavia et al., 2021), or could be extracted by LLE using conventional solvents, such as acetone, ethanol, and methanol (Sharma et al., 2015). From apple juice of Red Delicious and McIntosh apples, Sharma et al. (2015) carried out the phenolic extraction using surfactant mediated LLE. The extraction using surfactant, especially Brij-58 surfactant resulted in higher TPC and antioxidant activity (DPPH assay) of extracted juice than using organic solvents and water. However, further studies on interactional behavior of surfactant mediated assemblies and scale-up procedure at industrial level is still needed.

Solid–liquid extraction (SLE) was conducted by most of the previous studies to extract the phenolics from the solid parts of apple (peel, pomace, and whole fruit) using acetone, methanol, ethanol, or water (Bars-Cortina et al., 2017; Çam & Aaby, 2010; Casazza et al., 2015). Although most studies reported that using methanol resulted in higher phenolic yield, the usage of acetone, ethanol, and water were reported to be safer and more environmentally friendly than methanol (Casazza et al., 2020; Quang et al., 2014). However, the preliminary study on phenolic extraction of whole apple fruit (Ligol cultivar) for 4 h reported that employing ethanol as solvent yielded more flavonoids than employing methanol and acetone (Liaudanskas et al., 2018). Rana et al. (2015) carried out the extraction using 50% acetone (30 min at 60 °C), which was more efficient than 50% methanol and 50% ethanol to extract the apple pomace. By using water as solvent to extract phenolics from pomace, a study found that extraction conducted at 100 °C for 37 min with 100 mL/g of solvent to solid ratio was optimized in terms of TPC yield and limitation of 5-hydroxymethylfurfural (Çam & Aaby, 2010). This compound is an intermediate Maillard reaction product and was limited in some foods, including apple juice (max. 20 mg/kg) according to European Fruit Juice Association (Li et al., 2019). Furthermore, a study investigated that SLE of apple pomace (Champion) using Tween 80 as surfactant was optimum to obtain the highest TPC at concentration of 1.14% in water, 104 mL/g of solvent to material ratio, pH of 3.8, for 65 min of extraction (Skrypnik & Novikova, 2020). Using these optimum conditions, the predicted TPC was 7.75 mg gallic acid equivalent (GAE)/g dry weight (DW), while the experimental value was 7.68 mg GAE/g DW.

The SLE should be done efficiently and selectively. For example, the extraction using only one type of solvent is not very efficient to extract all the polyphenols as other hydrophilic compounds could be also extracted, and less polar phenolics could remain inside the matrix (Acquavia

Table 4 Bioaccesibility of natural antioxidants in apples during in vitro digestion model

Apple products	Cultivar (Country)	Type of analysis	Bioaccessibility (%) during in vitro digestion			Notes	References
			Oral digestion	Gastric digestion	Intestinal digestion		
Whole fruit	Gala (Mexico)	TPC	-	24.98	57.45	-	Corona-Leo et al. (2021)
		TFC	-	41.63	73.15		
		TAC	-	204.23	83.09		
		ABTS	-	14.59	18.22		
		DPPH	-	16.10	33.07		
	Starking (Mexico)	TPC	-	7.23	35.02		
		TFC	-	23.14	50.08		
		TAC	-	152.81	46.07		
		ABTS	-	14.93	22.44		
		DPPH	-	15.57	28.45		
	Red Delicious (Mexico)	TPC	-	26.61	81.43		
		TFC	-	44.28	83.41		
		TAC	-	126.49	82.70		
		ABTS	-	15.53	23.38		
		DPPH	-	10.80	20.76		
	Golden Delicious (Mexico)	TPC	-	25.52	29.63		
		TFC	-	16.62	48.30		
		TAC	-	-	-		
		ABTS	-	8.08	18.91		
		DPPH	-	4.84	12.89		
	Granny Smith (Mexico)	TPC	-	18.78	20.79		
		TFC	-	37.62	43.08		
		TAC	-	-	-		
		ABTS	-	20.20	21.98		
		DPPH	-	15.32	24.05		
Oven-dried apple	Fuji (Chile)	TPC	28.09	31.65	27.11	a, b	Pavez-Guajardo et al. (2020)
Freeze-dried pomace	Pink Lady (Australia)	Total flavanols	-	161.50	24.70	b	Liu et al. (2019)
		Total flavonols	-	352.20	156.60		
		Total phenolic acids	-	227.20	407.60		
		Total dihydrochalcones	-	182.80	6.30		
Freeze-dried extruded pomace	Pink Lady (Australia)	Total flavanols	-	80.20	48.60	b, c	Liu et al. (2019)
		Total flavonols	-	258.60	136.00		
		Total phenolic acids	-	245.60	190.30		
		Total dihydrochalcones	-	311.80	255.50		
Whole fruit	Jonaprinz (Luxembourg)	Total chlorogenic acid	-	109.47	31.61	-	Bouayed et al. (2012)
	Jonagold (Luxembourg)	Total chlorogenic acid	-	103.67	34.19		
	Mutzu (Luxembourg)	Total chlorogenic acid	-	92.07	56.52		
	Golden Delicious (Luxembourg)	Total chlorogenic acid	-	83.93	33.31		

a, The bioaccessibility was calculated based on data obtained in referred source. Bioaccessibility (%) = (value after digestion/value before digestion)*100. b, Final intestinal digestion (ileum) is considered. c, Extruded apple pomace at 15% barrel moisture is considered (Liu et al., 2019)

ABTS 2,2′-azinobis-(3-ethylbenzothiazoline-6-sulfonic acid), *DPPH* 2,2-diphenyl-1-picrylhydrazyl, *TAC* total anthocyanins content, *TFC* total flavonoid content, *TPC* total phenolic content

Table 5 Methods used for phenolics extraction from various apple products

Apple products	Sample form	Reported optimum conditions							Analyses conducted	Notes	References
		Extraction method	Solvent	Concentration	Temperature (°C)	Time	Sample to solvent ratio	Other parameters			
Juice (Red Delicious, McIntosh; India)	Fresh	Surfactant-mediated LLE	Brij-58	7 mM (aqueous solution)	RT	10 min	n.i	2 mL of BHT (0.8% in methanol) was added to prevent oxidative stress	TPC, DPPH	Brij-58 is better than Brij-35, SDS, Span-40, Trixton X-10 or using acetone, ethanol, methanol, water	Sharma et al. (2015)
Mash/juice (Envy; New Zealand)	Mashed (juice for analysis)	UAE	Water	-	30	7.3 min	1:1 (w/w)	20 W/g	TPC, TAA, ABTS	The optimum conditions were based on ABTS	Quang et al. (2014)
Peel (Ruby S; South Korea)	Freeze dried powder	UAE	Ethanol	50% in water	20	25.3 min	0.05 g/mL	40 kHz	TPC, TFC, DPPH	-	Park et al. (2022)
Peel (Jonagold; Italy)	Dried powder	MAE	Ethanol	68% (v/v) in water	150	90 min	0.1 g/mL	Inert atmosphere	TPC, TFC, ABTS	-	Casazza et al. (2020)
Peel (McIntosh; Canada)	Microwave-hot air dried powder	RFAE	Ethanol	42.67% in water	50	10 min	0.0114 g/mL	400 W, 320 mL N_2/min of mixing speed, inert atmosphere	TPC	-	Jusoh et al. (2017)
				37.33% in water	50	10 min	0.02 g/mL	400 W, 165 mL N_2/min of mixing speed, inert atmosphere	DPPH	-	
Peel (Golden Delicious, Jonagold, Renetta Canada, Raventze; Italy)	Oven dried powder	HPTE	Methanol	Pure	150	150 min	0.2 g/mL	Under N_2 atmosphere	TPC, DPPH	It is better than MAE and SLE HPTE of Jonagold = the highest TPC; HPTE of Renetta Canada = the highest antioxidant activity (DPPH)	Casazza et al. (2015)
Pomace (Champion, Gloster, Imant, Jonagored, Ligol; Russia)	Dried powder	Surfactant-mediated SLE	Tween 80	1.14% in water	RT	65 min	0.0096 g/mL	pH 3.8	TPC, ABTS, DPPH, FRAP	Tween 80 is better than Triton X-100, Span 20, and Tween 20, For all cultivars, it is better than using ethanol 70%	Skrypnik and Novikova (2020)

Table 5 (continued)

Apple products	Sample form	Reported optimum conditions							Analyses conducted	Notes	References
		Extraction method	Solvent	Concentration	Temperature (°C)	Time	Sample to solvent ratio	Other parameters			
Pomace (Fructus Meran S.p.A.; Italy)	Fresh / freeze dried powder / oven dried powder	SFE	Ethanol (as co-solvent)	5% (w/w)	45	120 min	-	30 MPa, freeze dried powder is better than fresh or oven dried powder	TPC, DPPH	It is better than Soxhlet with ethanol and boiling water maceration methods	Ferrentino et al. (2018)
Pomace (Tree Top, Inc.; USA)	Naturally fermented powder	PEF-assisted extraction	Water	-	25	500 µs	0.125 g/mL	2 kV/cm	TPC, DPPH	Followed by extraction: methanol (80), sample to solvent ratio (2:10 g/mL), 25 °C, 1 h TPC and antioxidant activity (DPPH) increased 37.4% and 86% than control	Lohani and Muthukumarappan (2016)
Pomace (Appledale Processors Co-op. Ltd.; Australia)	Homogenized	SLE	Water	-	85	30 min	0.05 g/mL	-	TPC, ABTS, DPPH, FRAP	-	Candrawinata et al. (2015)
Pomace (Conde-sur-Vire; France)	Dried	UAE	Malate buffer	50 mM	40	40 min	0.15 g/mL	0.764 W/cm^2, pH 3.8	TPC	It is better than conventional SLE	Pingret et al. (2012)
Whole fruit (without seed, Ligol; Lithuania)	Freeze dried powder	UAE	Ethanol	70% (v/v) in water	44.61	26.90 min	0.083 g/mL (+ 20 mL for rinsing)	480 W	TFC	-	Liaudanskas et al. (2018)
Whole fruit (without seed, Gala; Brazil)	Freeze dried powder	SLE	Acetone	65% in water	10	20 min	0.017 g/mL	-	TPC, TFC, DPPH, FRAP	-	Alberti et al. (2014)
			Methanol	84.5% in water	28	15 min	0.017 g/mL	-	TPC, TFC, DPPH, FRAP	-	

Table 5 (continued)

Apple products	Sample form	Reported optimum conditions						Analyses conducted	Notes	References
		Extraction method	Solvent	Concentration	Temperature (°C)	Time	Sample to solvent ratio	Other parameters		
Whole fruit (unpeeled, Braeburn; France)	Freeze dried powder	PLE	Methanol	Pure	RT	15 min	0.007 g/mL	3 extraction cycles, using pressurized N_2 controlled at 1 MPa for 30 s	TPC	It is better than using acetone 70% In comparison to SLE, PLE is better	Franquin-Trinquier et al. (2014)
		SLE	Methanol:BHT	99:1 (v/v)	RT	20 min	0.01 g/mL	-	TPC	It is better than using acetone:water:BHT 79:30:1	

ABTS 2,2′-azinobis-(3-ethylbenzothiazoline-6-sulfonic acid), *BHT* butylated hydroxytoluene, *DPPH* 2,2-diphenyl-1-picrylhydrazyl, *FRAP* ferric reducing antioxidant power, *HPTE* high pressure and temperature extraction, *MAE* microwave-assisted extraction, *n.i.* not informed, *PEF* pulsed electric field, *PLE* pressurized liquid extraction, *RFAE* radio frequency-assisted extraction, *RT* room temperature, *SFE* supercritical fluid extraction, *SLE* solid–liquid extraction, *TAA* total ascorbic acid content, *TFC* total flavonoid content, *TPC* total phenolic content, *UAE* ultrasound-assisted extraction

et al., 2021). To minimize this issue, a multi-step extraction process could be conducted. Firstly, hexane could be used to discard the lipids, carotenoids, and chlorophyll from the apple. Secondly, extraction using methanol could dissolve the sugars, organic acids, and low molecular weight phenolics. Lastly, aqueous acetone (4:6) could be used to extract the polymerized polyphenols from the residue of previous step (Acquavia et al., 2021).

A study compared different modified SLE methods to extract the pomace from Australian Pink Lady cultivars (Li et al., 2020a, b). The authors used aqueous methanol to extract the phenolics of the pomace, and after which, the residue was subjected to either sequential base and acid hydrolysis or sequential acid and base hydrolysis and extraction with 1:1 (w/w) diethyl ether/ethyl acetate (DE/EA) solution in order to liberate the bound phenolics from the pomace. As comparison, direct base or acid hydrolysis followed by DE/EA extraction was evaluated. Among the methods conducted, the authors suggested the direct base hydrolysis using NaOH, followed by DE/EA extraction to extract high content of polyphenols (10.62 mg GAE/g DW), considering the simplicity and efficiency.

Despite the modifications of SLE have been studied as above, generally, conventional extraction has some drawbacks to be concerned, such as long extraction time (19 h at 25 °C) (Casazza et al., 2015), degradation of phenolics due to excessive extraction time, high solvent requirement (leading to corrosion of equipment and extra waste treatment), flammability problem, poor selectivity (Perussello et al., 2017), and food safety issues (Zhang et al., 2021). Therefore, the development of extraction, including MAE, RFAE, UAE, SFE, PEF-assisted extraction, and PLE have been studied, especially to reduce the extraction time, organic solvent consumption, as well as to increase the phenolic yield.

Non-conventional Extractions

Microwave-Assisted Extraction

In comparison to conventional extraction method, microwave-assisted extraction (MAE) has been known as an eco-friendly and rapid-heating method with lower solvent utilization. It also produced more yield with higher purity, has higher level of automation, reduced wastes, and has suitable reproducibility (Rezaei et al., 2013). The principle of this method is the fast microwave energy transfer to the materials via molecular interaction with the electromagnetic field, which then weakens the apple tissue and subsequently increasing the phenolic yield during solvent extraction (Perussello et al., 2017).

The MAE was applied for extracting the phenolics from apple pomace (Chandrasekar et al., 2015; Rezaei et al., 2013) and peel (Casazza et al., 2015, 2020). Casazza et al.

(2015) have reported that MAE (using methanol, solid to liquid ratio 0.2 $g_{DM}/mL_{solvent}$, at 110 °C for 60 min) yielded higher TPC in Raventze peel than conventional SLE (using methanol, solid to liquid ratio 0.2 $g_{DM}/mL_{solvent}$, at 25 °C, for 19 h, dark condition). The faster extraction time could be also highlighted as an advantage as MAE was 19 times faster than conventional SLE. In another study, Casazza et al. (2020) used ethanol to extract the phenolic from Jonagold peel, and found that the optimum conditions based on TPC, TFC, and antioxidant activity (ABTS assay) were using ethanol (32% water concentration), 90 min extraction time, and found that the degradation due to high temperature (up to 150 °C) could be limited by inert atmosphere. The authors also paid attention on the solid residue after the extraction, and concluded that that extraction process allowed to obtain solid residue less pollutant and higher caloric value than the unprocessed apple skin, manifesting that it could simplify the waste handling, increasing the efficiency of its potential use in waste-to-energy plants (Casazza et al., 2020).

By employing RSM with central composite design (CCD), Chandrasekar et al. (2015) analyzed the optimum conditions to extract the phenolics from Red Delicious and Jonathan apple pomace using MAE with acetone 70% and ethanol 60%. The authors varied the microwave powder (100–900 W), solvent to sample ratio (4–12 mL/g dry pomace), and extraction time (30–180 s). They found that using acetone 70%, the increasing microwave power (265 to 735 W) significantly increased the TPC of both cultivars, additionally longer extraction time (61 to 149 s) increased the phenolics yield of Jonathan pomace. On the other hand, the solvent volume did not significantly affect the extraction process. When ethanol 60% was employed, the solvent to sample ratio, microwave power, and extraction time significantly affected the TPC of both Red Delicious and Jonathan pomace. In conclusion, the highest TPC (15.8 mg GAE/g) was found in Red Delicious pomace, extracted under optimum conditions (735 W of power, 149 s of extraction time, and 10.3 mL of ethanol 60% per gram dry sample (Chandrasekar et al., 2015).

It is noteworthy that the microwave power and extraction time are the two most fluctuating parameters in MAE, which keep changing depending on plants, bioactive compounds, and different plant part of the same plant (Kala et al., 2016). Furthermore, excessive microwave power and exposition time might result in higher solution temperature, which then degrade the phenolics (Perussello et al., 2017). It was confirmed by Rezaei et al. (2013) that the TPC of Red Delicious pomace decreased with excessive power (more than 90 W) and extraction time (more than 15 min).

Radio Frequency-Assisted Extraction

The radio frequency-assisted extraction (RFAE) mechanism is similar to MAE. In comparison to MAE, RFAE is more advantageous due to longer wavelength and lower frequency that result in greater penetration depth and suitable for bulk material with better uniform heating (Jiao et al., 2018). RFAE for 10 min at 50 °C was used to recover the phenolics from McIntosh apple peel (Jusoh et al., 2017). The authors studied the effects of ethanol concentration (10–70%), mixing speed (10–320 mL N_2/min), solid to liquid ratio (0.002–0.02 g/mL), and radio frequency power (200–400 W) on TPC and antioxidant activity (DPPH assay) (Table 5). By performing RSM, the highest TPC value (121.87 mg GAE/g DW) was achieved after the extraction using 42.67% ethanol, 230 mL N_2/min of mixing speed, 0.0114 g/mL solid to liquid ratio, and 400 W of power. While to achieve the highest antioxidant activity by DPPH assay, 37.33% ethanol, 165 mL N_2/min of mixing speed, 0.02 g/mL solid to liquid ratio, and 400 W of power were used. Besides phenolic, RFAE was reported to increase the yield of pectin from apple pomace, better than MAE and conventional extraction (Zheng et al., 2021). Further investigation on apple's phenolic extraction using RFAE method is still needed, especially from various apple cultivars.

It is also important to know that when the plant matrix is exposed to radio frequency for a longer time (i.e., more than 10 min), the phenolics might degrade, resulted in lower TPC, and some impurities might be released which affected the yield (Kochadai et al., 2022).

Ultrasound-Assisted Extraction

The mechanism of ultrasound-assisted extraction (UAE) is based on the application of acoustic waves to the apple products, which can rupture the plant cells and then releasing more phenolics content to the medium (Perussello et al., 2017). UAE has been studied to have lower extraction time, higher process efficiency, and more sustainable, which has potential for industrial application for phenolic extraction (Acquavia et al., 2021; Perussello et al., 2017).

UAE was carried out in phenolic extraction from apple pomace (Pingret et al., 2012; Pollini et al., 2021; Wang et al., 2018a), mash (Quang et al., 2014), whole fruit (Liaudanskas et al., 2018), flesh (Wiktor et al., 2016), peel (Park et al., 2022; Wang et al., 2022), leaves (Stefova et al., 2019), and wood or bark (Withouck et al., 2019), using various solvents. The ultrasound could be applied during extraction, which was reported by most of the studies, but sometimes applied to pretreat the samples before the extraction (Wang et al., 2018a; Wiktor et al., 2016; Withouck et al., 2019). Wiktor et al. (2016) pretreated the flesh of Ligol apple (without peel) using contact (direct soundwave irradiation) and immersion (indirect soundwave irradiation) ultrasound treatment. It was found that the highest TPC and antioxidant activity (DPPH assay) was found in extract using immersion method (40 kHz, 5 min). In contrast, the contact ultrasound

treatment, particularly for 30 min (which was the longest treatment time analyzed), decreased around 30% TPC and 92% of antioxidant activity in comparison to control. This might be explained as the contact ultrasound may magnify the enzymes activity that degrades the phenolics. Furthermore, the immersion method affected the color of apple in less extent in comparison to contact one. Another study pretreated apple pomace using ultrasound (24 kHz, 400 W, varied pretreatment time of 0–30 min, in 0–50% ethanol solution) under controlled moderate temperature of pretreatment (initial temperature 20 °C, never exceeded 37 °C) to prevent thermal degradation, and continuously was extracted conventionally using the same medium for another 30 min (total 60 min including the 30 min of UAE pretreatment) at 20 °C (Wang et al., 2018a). The highest TPC was obtain in the pomace pretreated with ultrasound for 30 min, and subsequently extracted using 50% of ethanol. The same authors in other publication recovered the catechin from flesh and peel of Granny Smith and Red Delicious with UAE (Wang et al., 2018b). Besides the flesh and skin, apple cultivars could also affect the relative content of catechin in TPC that indicates the selectivity of catechin extraction compared to other phenolics. However, the mechanism of the observed extraction selectivity remained unclear.

The selectivity to extract anthocyanins in Idared and Lještarka peels could also be improved by acidifying the methanol using 0.1% HCl as the solvent, with optimized UAE parameters (15 min, 0.1 g/mL of sample to solvent ratio), noting that anthocyanins are more stable in acidic environment (Jakobek et al., 2015). While to extract phenolics in general, such as flavonols, anthocyanins, dihydrochalcones, flavanols from peels, or flavanols, procyanidins, dihydrochalcones, and hydrocycinnamic acids from fleshes of those apples, UAE with 80% methanol was recommended.

An aqueous buffer—50 mM malate buffer (pH 3.8) was employed by Pingret et al. (2012) as the solvent in phenolic UAE of pomace. The authors varied the temperature (10–40 °C), sonication time (5–55 min), and ultrasonic intensity (0.431–0.764 W/cm^2) as the independent factors, and as dependent one, TPC was analyzed. The UAE conducted at 40 °C for 40 min, with 0.764 W/cm^2 resulted in 30% higher TPC, in comparison to conventional SLE. The authors also explained that temperature and sonication time are the most influential variables, as TPC linearly increased as temperature and sonication time increase. Ultrasonic intensity also showed the same trend, but it was less predominant. However, it is worth noticing that excessive sonication time might degrade the phenolics, and hence might decrease the yield (Perussello et al., 2017).

In whole fruit of Ligol apple, the optimization of UAE using RSM was carried out to extract the phenolics (Liaudanskas et al., 2018). The ethanol 70% was used as the solvent. The effect of optimization of temperature, extraction time, and ultrasonic power was analyzed to find the highest TFC content of the extract. The optimum UAE was fulfilled at 44.61 °C for 26.90 min, with 480 W of ultrasonic power. With this optimum condition, the experimental TFC (6.58 mg rutin equivalent (RE)/g was near to the predicted one (6.69 mg RE/g). This optimum condition was then applied to 6 apple cultivars grown in Lithuania (Aldas, Auksis, Connel Red, Ligol, Lodel, Rajka), and HPLC analysis showed that Aldas apple possessed the highest total phenolics (Liaudanskas et al., 2018).

In comparison to other non-conventional phenolic extraction methods (ultraturrax extraction, accelerated solvent extraction or PLE, and pulsed electric field), UAE method applied during extraction for 60 min at 60 °C was observed as the most effective method to extract the TPC from Red Delicious pomace, especially when 50% ethanol was used as the solvent (Pollini et al., 2021). However, a study also reported that PLE with solid-phase extraction absorbent (da Silva et al., 2020) extracted more apple's TPC than UAE.

Recently, instead of using ethanol or other organic solvents, a study optimized the UAE (0.242 kW.h/kg of energy input) of Gala peel's phenolics using CO_2 water (0–7.05 mmol/L) (Wang et al., 2022). The idea of using CO_2 water is because the dissolved CO_2 could act as nucleus for new cavitation bubbles generated during UAE, then the resulting cavitation effect could eventually improve the phenolic extraction. Among the analyzed concentration of CO_2 in water, UAE using 5.28 mmol CO_2/L extracted the highest TPC, total proanthocyanidins, and antioxidant activity (DPPH assay), and additionally high TFC. This technique can be considered by the industry as CO_2 water is more economically affordable than organic solvents.

According to our knowledge, the comparison between ultrasound treatment applied before and during extraction of phenolics, especially in apples, has not been conducted, and could be explored in future studies. Besides their effects on the phenolics yield and composition, the time and energy consumed by both applications should be studied and considered to be economically beneficial. However, either before and during extraction, it is advisable to maintain the temperature low or moderate (50 °C or lower) to prevent the degradation of phenolics during UAE. The short time (50–100 s) but several pulses of ultrasound can also be applied to maintain the temperature (Wang et al., 2018b).

Supercritical Fluid Extraction

In supercritical fluid extraction (SFE), the nontoxic and safe gas CO_2 (with low critical temperature, around 31 °C) was used as the main solvent to extract the phenolics of apple products (Fernandes et al., 2022). The organic co-solvents (acetone, ethanol, methanol) could be applied to increase the solvating power of CO_2, noting that the CO_2 alone is

selective to non-polar compounds, which might be the limitation to extract the phenolics with higher polarity. In comparison to conventional extractions, the degradation of phenolics could be decreased and the quality of phenolics could be maintained as SFE was conducted in lower temperature, without light and air exposure (Perussello et al., 2017). Additionally, apart from phenolics, the SFE method was also selective to extract and isolate triterpene acids, which also possess antioxidant, anti-inflammatory, and anticancer activities (Zhang et al., 2021).

The SFE method was used to recover the phenolic of apple pomace (Ferrentino et al., 2018) and peel (Massias et al., 2015). Furthermore, a study also analyzed the total phenolics and antioxidant activity of phenolics in the oil extracted from apple seeds by SFE (Ferrentino et al., 2020). Ferrentino et al. (2018) optimized the SFE of apple pomace (fresh, freeze dried, and oven dried) with controlled time (2 h) and various pressure (20 and 30 MPa), temperature (45 and 55 °C), with and without addition of ethanol 5% as co-solvent. The authors found that freeze dried pomace extracted at optimum conditions (45 °C, with 30 MPa of pressure, and applying ethanol 5% as co-solvent) resulted in higher TPC and antioxidant activity (DPPH assay). The TPC, antioxidant activity by DPPH assay, and yield of extract obtained by performing SFE at this optimum condition was compared with conventional Soxhlet extraction and boiling water maceration. The yield obtained by SFE was lower than both conventional extractions, but interestingly, the TPC and antioxidant activity of SFE extract were higher than by both conventional methods. This result indicated that SFE selectively extracted less but more active polyphenols, in comparison to analyzed conventional methods. The result was then confirmed as HPLC analysis (between SFE and Soxhlet) showed that higher signals intensity and relative areas of SFE (in the retention time between 20 and 25 min, in which phloridzin, epicatechin, quercetin, and phloretin are detected) were obtained than of Soxhlet extraction (Ferrentino et al., 2018). Additionally, Ferrentino et al. (2020) extracted the oils from apple seeds using SFE (24 MPa, 40 °C, 1 L/h of flow rate, 140 min), which resulted in higher TPC and antioxidant activities (DPPH and FRAP assays) of oil than that using Soxhlet extraction (using n-hexane, 10/150 g/mL of sample to solvent ratio, 6 h, at boiling temperature).

Massias et al. (2015) investigated the SFE (25 MPa, 50 °C, 75:22:3 mol ratio of CO_2: ethanol: water) of phenolics in dried (lyophilized) and ground Golden Delicious peels. The use of ethanol as cosolvent was adapted due to the fact that phloridzin and quercetin glycoside that are abundant in apple peels, contain a sugar component that is too polar to be soluble in only CO_2. As comparison, maceration using ethanol and conventional solvent extraction using methanol + acetone 70% (v/v) were conducted. The result was higher total phenolics extracted by SFE ranged from 550 to 800 mg/100 g dry peel (depending on solvent to peel ratio—37 to 73 wt basis, mass of dried peel—15 or 30 g, and SFE procedure—static or dynamic) than maceration (177 mg/100 g dry peel). While total phenolics of 792 mg/100 g dry peel were observed after extraction by conventional solvent extraction.

Pulsed Electric Field-Assisted Extraction

PEF (pulsed electric field-assisted extraction) application to plant materials, such as apple, causes the cell disruption due to the short and high voltage pulses that can increase the yield and phenolic extraction efficiency. It is an eco-friendly method as the solvent consumption is reduced. As PEF is non-thermal method, it also minimizes the damage of the nutrients in the foods. The problem of this method is the difficulty to scale-up (Perussello et al., 2017; Zhang et al., 2021).

The study on the effect of PEF treatment on the phenolic extraction from apple was reported in pomace (Lohani & Muthukumarappan, 2016), flesh (Wiktor et al., 2015), mash (Turk et al., 2010), and whole fruit (Ribas-Agustí et al., 2019). Lohani and Muthukumarappan (2016) applied the mild PEF to release the bound phenolics of apple pomace powder. The varied flour (pomace powder) to water ratios (FWR 5–12.5%, w/v), treatment time (500–1250 µs), and electric field intensity (1–3 kV/cm) were optimized based on the results of TPC and antioxidant activity (DPPH assay). The factors affecting the release of phenolics were in following order: FWR > electric field intensity > treatment time. The authors found that the optimized conditions (12.5% FWR, 500 µs, 2 kV/cm) resulted in the increased TPC and antioxidant activity values of 37.4% and 86%, respectively, compared to the control. Those phenolics were dominantly consisted of protocatechuic acid, followed by chlorogenic and salicylic acid, analyzed by UHPLC-DAD.

Wiktor et al. (2015) analyzed the impact of PEF treatment on TPC, antioxidant activity (DPPH assay) and color of Ligol apple flesh, grown in Warsaw, Poland. The authors varied the electric field intensities (0–5 kV/cm) and pulse numbers (0–100 pulses), which corresponded to the energy input of 0–80 kJ/kg (based on calculation). After the PEF treatment, the polyphenols were extracted using 80% ethanol for further analyses. It was reported that the highest TPC and antioxidant activity were observed in the samples treated by 1.13 kJ/kg (1.85 kV/cm and 10 pulses). Excessive energy above 40 kJ/kg (5 kV/cm, 50–100 pulses) negatively impacted the TPC and antioxidant activity of samples up to 35.93 and 32.95%, respectively. The authors explained that this phenomenon might happen as the electric energy was not sufficient for PPO deactivation, which resulted in TPC degradation. The color of samples was analyzed 0 and 60 min after the PEF treatment and was compared to the control (untreated sample). The total color change at 0 min

(ΔE_0) ranged from 0.48 to 8.58, while at 60 min (ΔE_{60}) ranged from 1.25 to 21.87) which could be related to the different chemical compositions of apples. Higher electric field intensity might increase the chance of enzymatic browning. The samples treated at 3 and 5 kV/cm showed decreasing L^*-value (brightness), indicating more intensive browning, which might be caused by the higher release of PPO and its substrates.

The negative impact of PEF on phenolic composition was reported in different study. From the mash of Golden Delicious apple from France, phenolic extraction was conducted using PEF treatment (Turk et al., 2010). The mash size (small and large: 120 and 630 mm^3, respectively) and electric field intensity (0 and 450 V/cm) were varied. The juice was collected from the PEF-treated mash. The juice yield was checked, and the polyphenolic compounds of obtained juice were then analyzed using HPLC. It was found that PEF-treatment increased the juice yield in comparison to control, regardless the mash size. Unfortunately, in comparison to untreated sample, the PEF treatment decreased 53.4 and 17.1% of total polyphenol concentration for small and large mash size, respectively. In comparison to the loss from small size mash, the lower loss from the large size mash could be explained by the decreasing oxidative area, so the oxidation of hydroxycinnamic acids could be lower.

In an in vitro simulated digestion study, the bioaccessible and non-bioaccessible fractions of phenolic compounds of Golden Delicious whole fruit were analyzed after the PEF treatment (0 and 24 h after 0.01, 1.8, and 7.3 kJ/kg of energy input), and was compared to the untreated apple (Ribas-Agustí et al., 2019). By employing UHPLC-DAD-MS, the TPC and individual phenolic content were determined. The phenolic extraction using methanol was conducted only to undigested apples, but not conducted to bioaccessible and non-bioaccessible fractions (after digestion) as there was no significant difference result from direct analysis in the preliminary study. The result showed that 0.01 kJ/kg treatment after 24 h resulted in increased bioaccessible (61%) and non-bioaccessible (35%) 5-caffeoylquinic acid, as well as TPC of bioaccessible (26%) and non-bioaccessible phenolic compounds (19%) in comparison to untreated apple. The increase of bioaccessible phenolic contents could be explained by the increased contents in the undigested apples. In comparison to untreated apple, higher energy inputs (1.8 and 7.3 kJ/kg) were observed to decrease the overall TPC of bioaccessible and non-bioaccessible fractions up to 37% and 22%, respectively after 0 h of treatment, and 44% and 22%, respectively after 24 h of treatment. Furthermore, the bioaccessibility of phenolics was also determined by calculating the ratio of bioaccessible compounds to the compounds of undigested one. The bioaccessibility of overall phenolics increased from 14% (untreated) to 27% (24 h after 7.3%). Therefore, the functional properties of apple could be improved by PEF treatment by either increasing TPC of bioaccessible and non-bioaccessible fractions or the phenolic bioaccessibility (Ribas-Agustí et al., 2019).

To conclude, some factors such as processing parameters, solvent types, and apple's composition affect the efficacy of apple's phenolic extraction. Although some studies reported negative impact of PEF on extracted phenolic composition due to some possible mechanisms, PEF is still a good alternative for phenolic extraction as it reduces the extraction time and solvent usage as reported by other studies. Further studies are still needed to optimize the conditions, especially in various apple cultivars.

Pressurized Liquid Extraction

PLE (pressurized liquid extraction) is another green extraction technique with shorter extraction time, lower solvent requirement, and higher extraction rate and yield in comparison to conventional extractions. The method was based on the high pressure and temperature applied to the solvents (above boiling point of solvent, but it keeps the solvent still in the liquid state), which leads to penetration within the matrix tissue, hence increases the phenolic extraction efficiency. The expensive equipment cost to withstand the high pressure utilized limits the usage of PLE (Deen et al., 2019). In PLE, the solid sample, such as apple, is extracted using solvent under following conditions: 40–200 °C of temperature, 500–3000 psi, and 5–15 min of extraction time (Deen et al., 2019).

In previous studies, the PLE was performed to extract the phenolics from apple fruit (Franquin-Trinquier et al., 2014) and pomace (da Silva et al., 2020, 2023). A study performing RSM to optimize the PLE of phenolics from Braeburn apple fruit (Franquin-Trinquier et al., 2014). At room temperature and 1 MPa, the authors varied the solvent (pure methanol, acetone 70%), sample mass (50–550 mg), extraction time (1–15 min), and number of extraction cycles (1–3 cycles), and compared the resulting TPC with manual solvent extraction. The extraction using PLE was optimum when 50 mg sample was extracted using pure methanol for 15 min and three cycles. While for the manual extraction, the usage of methanol-butylated hydroxytoluene (BHT), 100 mg sample, 20 min, resulted the highest TPC. The TPC resulted from PLE was 19.5 times higher than manual extraction.

High pressure and high temperature extraction (HPTE) was analyzed by Casazza et al. (2015) to extract the phenolics of peels of 4 different apple cultivars (Golden Delicious, Jonagold, Renetta Canada, Raventze), in comparison to MAE and conventional SLE. It was found that the antioxidant activity (DPPH assay) of extract obtained by HPTE was higher than MAE and SLE, in all cultivars.

The development of PLE coupled on-line with solid-phase extraction (SPE) was reported to extract the

phenolics of apple pomace (da Silva et al., 2020). By using solid adsorbents in SPE, the specific phenolic classes and compounds could be separated with the mechanism similar to a chromatographic separation. The PLE-SPE consisted of several steps: (1) activation (using methanol or ethanol) and conditioning (using water) of adsorbents, (2) first extraction stage using water, (3) second extraction stage using a lower polarity solvent (methanol or ethanol) than that in the first extraction stage. During the extraction, the authors used various solid adsorbents (Sepra, Isolute, Strata X, and Oasis), amount of water in the first extraction stage (0–120 mL), temperature (60–80 °C), and activation or elution solvent (methanol or ethanol). HPLC analysis was used to identify the compounds (two phenolic acids, 10 flavonoids) in the sample. Generally, the best adsorbent to recover the phenolics was Sepra. There was a small effect of temperature on the extraction as higher temperature only significantly affected the content of chlorogenic acid (decreased), total phenolic acids (decreased), quercetin derivative (decreased), and phloridzin (increased). Furthermore, the authors included that ethanol could be used as the substitute (or partially substitute) for methanol, as no significant difference was found in total phenolic acids and total flavonoids. Additionally, in comparison to other methods (PLE, UAE, shaker, magnetic stirring), regardless the solvent used for extraction (water, ethanol, methanol), PLE-SPE exhibited higher or similar recovery of total phenolic acids and total flavonoids (da Silva et al., 2020).

Recently, in another article, the same authors also used PLE-SPE method, online with HPLC-photodiode array, so called 2D-PLE-SPE-HPLC–PDA, to extract the apple pomace (da Silva et al., 2023). The authors used Sepra as the adsorbent in SPE column, five different extraction solvent gradients (different concentrations of ethanol and water, and time of each solvent), and also analyzed the effect of temperature (40–80 °C) and static time (0–30 min) in the later steps. The solvent gradient (25 min: 100% water; 100–160 min: 100% ethanol), static time (20 min), and extraction temperature (80 °C) were suggested to obtain higher mass transfer of target analytes, such as furfurals, chlorogenic acids, flavonoids, and phloridzin. This developed method was compared to other methods (PLE only, UAE, and stirring). The developed method extracted similar (furfural, chlorogenic acid) or higher (phloridzin, flavonoids) extraction yield than PLE alone. UAE still showed significantly higher extraction yield of all target analytes than this developed method, except phloridzin (not significantly different). The developed method provides advantages, such as high efficiency (especially for phloridzin), elimination of sample preparation procedure, high automation, low human intervention, real-time process monitoring, and compounds fractionation of samples.

Other Factors Affecting Extractions

The pretreatment using enzyme could accelerate the phenolic extraction by degrading the cell walls that retain the phenolic in the polysaccharide-lignin network via hydrogen or hydrophobic bonding. The enzymatic maceration of Szampion apple pomace using commercial pectinase (Pectinex Yield Mash, Pectinex Smash XXL, Pectinex XXL, Pectinex Ultra-SPL, Pectinex AFP L-4) was carried out (Oszmiański et al., 2011). The obtained purees were mixed with the apple juices and increased the TPC of juices in comparison to the raw juice.

Fermentation pretreatment have been reported for improving the phenolic extraction efficiency. The microbial enzymes, such as pectinase, cellulase, β-galactosidase, naringinase, α-rhamnosidase and hesperidinase could be beneficial in phenolic extraction by rupturing the cell walls to release the phenolics, or deglycosylation of phenolics into their corresponding aglycones (Huynh et al., 2018). In comparison to control, the higher TPC and antioxidant activities were observed after the solid-state fermentation pretreatment of apple peel using four isolated *Aspergillus* spp. (black rot fungi) (Gulsunoglu et al., 2020) have been reported. The natural fermentation (without inoculum) was also reported to pretreat the apple prior to phenolic extraction. Natural fermentation pretreatment resulted in a slight rupture of apple pomace tissue, observed by optical microscopy (Lohani & Muthukumarappan, 2016). On the other hand, Gulsunoglu et al. (2020) did not find any significant differences on antioxidant activities (CUPRAC and DPPH assays) during 7 days of incubation of naturally fermented apple peel, while significant increases were found in the peel fermented by *Aspergillus* spp., except *A. aculeatus* ZGM6 (not significant).

Ferrentino et al. (2018) compared freeze dried, fresh, and oven dried apple pomace and their effects on TPC and antioxidant activity (DPPH assay) of extracted phenolics by SFE. Generally, it was found that freeze drying pretreatment significantly enhanced the TPC and antioxidant activity, followed by oven drying and without drying (fresh). This can be explained as freeze drying pretreatment minimize the thermal degradation of phenolics in comparison to oven drying, leading to higher content of recoverable phenolics. Rana et al. (2015) investigated that the TPC of freeze dried pomace (5.78 mg GAE/g DW) was higher than that oven dried and sun dried pomace, after the extraction using 70% ethanol (60 °C, 30 min). The TFC correspondingly showed similar result with TPC.

It is important to notice that high temperature not only can effect positively by increasing the yield of phenolics, but also negatively due to higher chance of phenolic degradation (Perussello et al., 2017). A negative impact of drying process on phenolics content of apple was reported by

previous study. Prior to phenolic extraction using acetone 80%, Red Delicious apple pomace was blanched and subsequently dried in a cabinet drier at various temperatures, from 50 to 80 °C (Heras-Ramírez et al., 2012). In comparison to undried pomace, the TPC, TFC, and antioxidant activity (ABTS) significantly decreased ($p < 0.05$) after the drying process of blanched and unblanched apple pomace due to thermal degradation. The degradation of phenolics could be reduced by increasing the drying temperature, which also means reducing the drying time at the same time. However, blanching process could help maintaining the stability of remaining phenolics during the drying process. This phenomenon might be explained by the PPO deactivation through blanching (Heras-Ramírez et al., 2012).

It is noteworthy that smaller particle size could enhance the extraction process by promoting better mass transfer, which results in higher bioactive compounds release in the solvent, in a shorter time of extraction. Prior to extraction, most studies prepared the apple samples in powder form by grinding the samples after drying (Table 5). Grinding process not only increases the apple product's surface contact to the solvent, but also to homogenize the particle size from the pomace as different sizes and shapes of stem, seeds, peels were found in it (Perussello et al., 2017).

Conclusions and Future Works

Apple has been known as one of the most consumed fruits in the world and has been investigated by previous studies to be the source of functional ingredients, such as carbohydrates, fibers, minerals, vitamin C, vitamin B complex, and phenolics. The growing location, cultivars, and environmental factors affect the chemical composition of the fruit. Major phenolics found in apple are hydroxybenzoic acids, hydroxycinnamic acids, flavanols and their oligo- and polymeric structures (procyanidins and proanthocyanidins), flavonols, dihydrochalcones, and anthocyanins. More phenolics are generally found in higher concentration in the peel. Therefore, consuming unpeeled apple is recommended to increase daily intake of phenolics. In choosing the apple, it is also necessary to consider the bioaccessibility of antioxidants after the digestion. As different cultivars showed different phenolic compositions, through this review, the consumers may select the suitable apple cultivars that are available in their regions, and affordable, yet still have high nutritional value, in order to achieve wider scale of consumption to promote general health of consumers. In the future, the bioactive compounds composition and their bioaccessibility are still widely open to be explored as the hybridization of new apple cultivars is still developing in different regions.

The antioxidant activity of apple's phenolics majorly contributes to the health promoting impacts. Thus, the phenolic extraction should be conducted correctly to maximize its benefits. This paper reviewed the optimization of phenolic extraction conducted with different methods. The developed methods, including MAE, RFAE, UAE, SFE, PEF-assisted extraction, and PLE have been studied and optimized to shorten the extraction time, decrease the solvent consumption and waste, and increase the phenolic yield and antioxidant activity of apple extracts, in comparison to the conventional extraction. Concerning the food grade solvents, acetone and ethanol were used by several studies for phenolic extraction as opposed to methanol. Besides optimization of non-conventional extraction methods using other new apple cultivars, further studies can be conducted to characterize those extraction processes with kinetic modeling.

The extracted apple phenolics could be beneficial for various food product developments to improve their functional properties. However, some challenges may be found during the adaptation from scientific literatures to industrial scale, either in non-conventional extraction method of apple polyphenols or the development of functional food products based on apple phenolics. Both researchers and professionals in industries need to concern not only about the scale-up cost, but also the safety, effectiveness, and quality of the apple phenolics to be applied as food ingredients. The approval of Generally Recognized as Safe (GRAS) in the food regulations could be crucial to widely commercialize the apple phenolics, both as food ingredients or apple phenolic-based functional foods, which subsequently contributes to better general health in the population. To support the safety concern of apple phenolics, the strategies to limit the presence of harmful substances from apples, such as patulin and pesticide residues can be further studied. Furthermore, it is suggested by researchers and food industries in different countries to study, develop, and formulate the apple phenolics-based functional foods following the common healthy food types, or even healthy traditional foods, consumed in that regions, so that the consumer acceptability on those functional foods would be higher.

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Declarations

Competing Interests The authors declare no competing interests.

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