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Review

Extraction Methods of Virgin Coconut Oil and Palm-pressed Mesocarp Oil and their Phytonutrients

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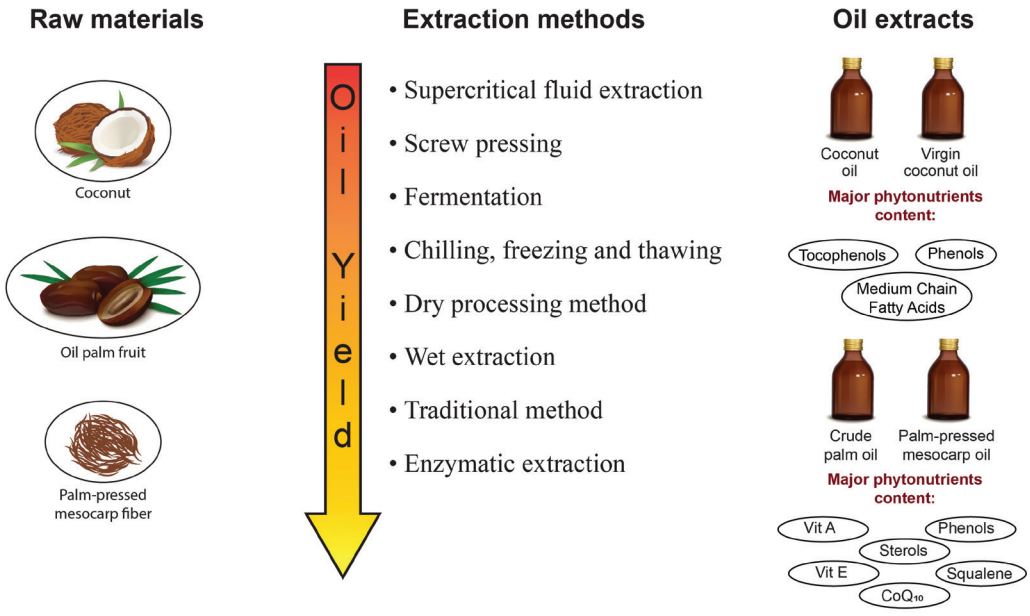
ABSTRACT

The Refined, Bleached and Deodorized (RBD) Coconut Oil (CO) and RBD Palm Oil (PO) are heavily consumed as cooking oils by Asians. Recently, Virgin Coconut Oil (VCO) and Palm-pressed Mesocarp Oil (PPMO) emerged into the market as functional oils. A number of studies were conducted to extract these edible oils by using different methods and conditions. Thus, this review focuses on the extraction methods of CO, VCO, CPO and PPMO, together with their yields and phytonutrient profiles. Phytonutrients such as carotenoids, tocopherol and tocotrienols, squalene, phytosterol and coenzyme Q_{10} were found in all the oils with various concentrations depending on their respective extraction methods. Supercritical Fluid Extraction (SFE) showed the highest efficiency in extracting VCO and PPMO among the extraction methods, in terms of yield and phytonutrient contents. VCO extracted by SFE contains higher medium chain fatty acids which promotes good digestibility when compared to other extraction methods. PPMO extracted by SFE is clean with a pleasant aroma without the presence of the gums and contains a lower amount of phospholipids. Furthermore, water soluble-phenolics were found to be present in the PPMO that contribute to its antioxidant activity. Thus, SFE is highly recommended for the extraction of VCO and PPMO.

GRAPHICAL ABSTRACT



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1. INTRODUCTION

Natural products are the primary source of antioxidants which include polyphenols, phytosterols, alkaloids, terpenoids and organosulphur compounds due to the presence of hydroxyl (–OH) group [1]. These compounds are also known for their anti-inflammatory, blood glucose lowering [2], antimicrobial [3] and protein modulation properties [4]. Majority of these compounds are the secondary plant metabolites that are naturally synthesized by the plants or vegetables for specific functions. These functions include protection from biotic and abiotic stresses and plant interactions with several types of evolved pathogens, resulting in the formation of more than 100,000 metabolites which are involved in the plant's defence system [5]. Besides that, plant secondary metabolites are also known as phytonutrients due to their important role in promoting human health benefits. Fruits, especially coloured fruits contains the richest source of phytonutrients [4] such as carotenoids, tocopherols, tocotrienols, phenolics and phytosterols. For instance, carrots are rich with carotenes while tomatoes with lycopenes. Several types of carotenoids such as α-carotene, β-carotene and β-cryptoxanthin are involved in pro-vitamin A activity and will be transformed into vitamin A in the human body [6]. On the other hand, lycopene is known as effective quencher of free radicals [7]. Vitamin E, especially tocopherols and tocotrienols are found in abundance in oil palm fruits as natural antioxidants and have been reported to possess *in vivo* anti-cancer properties [8]. Furthermore, the consumption of β-sitosterol, a phytosterol present in the vegetables or fruits showed hypocholesterolemic effect by reducing body cholesterol level [9].

The widespread of phytonutrients in plants and vegetables contribute to their phytonutrient profiles. The processing of raw fruits and vegetables are usually performed for the ease of consumptions and to increase their shelf-lives. Thus, the phytonutrients content in the food products are significantly dependent on their processing methods [4]. For edible oil extraction industry, many factors are needed to be taken into consideration to ensure the desired quality of the end-product can be achieved. One of the most important factors is the method and technology used in the oil extraction [10]. The extraction yield of vegetable oils varies based on the types of plant tissues bearing the oils. The oils are usually concentrated in the pulp, seeds, tubers and stone fruits. Examples of plants with oleaginous fruits are oil palm, coconut and olive while soybean, sunflower and rapeseed are examples of plants with oleaginous seeds. Peanuts belong to plants with tubers and corn belongs to plants with germs [11]. Among them, the major oil-bearing plants are oil palm, coconut, sunflower and soybean [12]. This review will be focused on the two major plants with oleaginous fruits from Southeast Asia, which are oil palm and coconut.

In recent years, CO or VCO has attracted the media attention and their consumption by the worldwide population has increased due to their health benefits. Moreover, the medical doctors have endorsed the use of these oils as a cooking media and recommended them as supplementary ingredients in the coffee or vitamin drinks [13]. Overall, functional oil is defined as the oil that has a health beneficial action beyond its nutritional value [14]. VCO is made up of 90% of saturated fatty acids and 62% of its total fatty acid composition is made up of Medium-chain Fatty Acids (MCFA) [15,16]. VCO is gaining popularity as a functional oil due to its high MCFA content, thus a growth of VCO in the market was observed [17]. One of the pharmacologically active components in the VCO is lauric acid, a MCFA which dominates the fatty acid composition with a percentage ranging from 46% to 48%. Oral consumption of VCO causes a transformation of lauric acid into monolaurin in the human body. Monolaurin is a major component of breast milk that enhances the immune system of infants [18] and has the ability to destroy the lipid membrane of the bacteria [14]. Therefore, VCO is used as natural food preservatives via bacterial destruction [14]. Besides that, high MCFA content in VCO exhibits good digestibility in consumers [15] since it has a lower melting and solidification temperature, smaller molecular size and lower energy density compared to other triglycerides that mainly consist of saturated fatty acids [19]. Furthermore, MCFA oils are widely utilised in flavouring industries due to its higher hydrophilicity if compared to conventional fats and oils, thus able to dissolve various polar substances [14]. Due to these benefits, VCO has a wide market throughout South East Asia [17].

For the extraction of Crude Palm Oil (CPO), screw-press process of mesocarp and exocarp of the palm fruitlets in palm oil mills generates a by-product which is known as Palm-pressed Mesocarp Fiber (PPMF) [20]. PPMF contains about 5–6% of residual oil that constitutes of carotenoids, tocopherols, tocotrienols, phytosterol, coenzyme Q_{10} and squalene [7]. Approximately 15% of PPMF will be produced by Fresh Fruit Bunch (FFB) after CPO extraction. PPMF will be used as the fuel for the boilers in the palm oil mills, besides mulching for the plantation field to prevent leaching and to increase soil fertility [7]. Generally, an oil palm mill generates approximately 1.6 tonnes of PPMF per hectare of oil palm annually [21], resulting in a significant loss of phytonutrients. Therefore, several approaches have been studied to recover the residual oil from the PPMF.

The optimization of the extraction methods for VCO and PPMO is an important research area to retain as much of the natural phytonutrients as possible and to obtain a consistent yield of the oil extracts. Thus, the common extraction methods of VCO and PPMO were summarized in this review. The phytonutrients content in these oils were found to differ with the extraction methods used.

2. COCONUT OIL AND VIRGIN COCONUT OIL

RBD CO or copra oil and VCO are extracted from the kernel of coconuts (*Cocos nucifera* L.). Coconut trees belongs to the family Arecaceae and is well known for its hundred uses with limitless application throughout the world, such as sources of timber, fiber and biofuel. There are several varieties of coconuts from dwarf to tall trees, for instance, Indian, Malayan, Jamaica, Ceylon, Java, Laguna, orange, green and Fijian. The top three coconut producers in the world are Indonesia (3.1 million hectares), Philippines (2.7 million hectares) and India (1.5 million hectares). These countries generate three quarters of the total world coconut production with approximately 64 billion coconuts [22]. Besides, other countries including Thailand, Indonesia, Malaysia, Sri Lanka, Vietnam, Fiji and Samoa are considered as large producers of VCO. The largest consumers of the VCO are United States, Europe, Middle East, South Africa, Australia and Asia Pacific [23].

Refined, bleached and deodorized coconut oil and VCO are obtained from the coconut kernel but they are differentiated by

their extraction methods where RBD CO is obtained from the dried copra by using a dry processing method while VCO is obtained from the fresh coconut flesh. The dry processing methods, including wedge presses, screw presses and hot hydraulic presses are widely used in commercial CO production in Asian countries. Subsequently, the extracted CO will go through RBD processes [24]. High temperature is applied during the RBD process, especially during the deodorization process that involves temperatures up to 204–245°C, which deactivates the bioactive components such as tocopherols and polyphenols [25]. Due to the refining process, CO has no taste or fragrance. On the other hand, VCO is obtained from the fresh and mature coconut kernels by mechanical method with or without the use of heat but without any chemical refining [26]. The absence of the chemical treatment and heating retains the taste and fragrance of the oil. Furthermore, the polyphenol fraction of VCO has superior activity in reducing the microsomal lipid peroxidation if compared to CO and groundnut oil [27].

2.1. Extraction Methods

2.1.1. Traditional boiling method

Coconut oil is traditionally extracted by boiling coconut milk to evaporate the water, leaving the oil behind. In order to extract approximately 14 L of coconut milk, the processes last for an hour or until all the oils get separated from the milk. Even though modern technologies are available to achieve higher efficiency in the CO extraction, traditional extraction method is still being practised at the village level in coconut producing countries [28]. The yield of traditional method of CO is about 33%, which is lower compared to other extraction methods such as dry extraction, fermentation and Supercritical Fluid Extraction (SFE). However, the Total Phenolics Content (TPC) study conducted by using Folin–Denis method revealed that the CO extracted by traditional method has a higher TPC value of 618 mg GAE/kg oil if compared to the commercial CO extracted through dry extraction method that gave a TPC value of 91 mg GAE/kg oil. Phenols are polar compounds that easily dissolves in the aqueous phase of coconut milk. During the extraction process, the boiling process of coconut milk causes evaporation of the water and the phenols slowly incorporates into the oil phase. The boiling of coconut milk requires longer time and higher temperature, which is above 100°C and allows longer contact time for CO and phenols. This phenomenon favours the incorporation of the phenols into CO. Thus, the traditional extraction method of CO allows higher amount of phenols to be extracted from the coconut [24]. Although the CO extracted traditionally has higher amount of phenols, its lower yield and higher moisture content leads to a shorter shelf-life compared to commercial CO [28].

2.1.2. Fermentation

Fermentation involves the use of pure culture of bacteria such as *Lactobacillus plantarum*, *Lactobacillus delbrueckii* and *Lactobacillus casei*. Among the strains, *L. plantarum* is preferred due to its fast-growing rate in the coconut milk at the temperature range of 40–50°C, with considerable production of lactic acid that indicates a rapid breaking of the emulsion and liberation of the oil. Based on a previous study, the most efficient coconut cream separation was obtained through the incorporation of 5% *L. plantarum* into the coconut milk with settling time of 10 h and the total oil recovery obtained is up to 95% [29].

Besides, the coconut milk emulsion can be separated by adjusting the pH in between 3.0 and 5.6 with the inoculation with bacterial cultures. In a study conducted by Che Man et al. [30], 25% acetic acid was used to destabilize the coconut cream in the VCO extraction at room temperature. The reaction time is in between 10 and 14 h and resulted in a yield of 60% [30]. However, the incorporation of acid treatment with the inoculation of bacteria cultures in the extraction process has a lower recovery rate of good quality oil if compared to inoculation of bacteria alone during the extraction but both approaches has higher CO yield compared to the traditional extraction method.

2.1.3. Enzymatic extraction

The coconut copra is made up of 10% carbohydrate which contains about 50% cellulose. Approximately 75% of the cellulose is made up of α-cellulose [31]. The oil present inside the plant cells links with the proteins and a wide range of carbohydrates such as starch, cellulose, hemicellulose and pectins. Therefore, cell wall degrading enzymes are used to extract the oil by solubilizing the structural cell wall components.

A previous study was conducted requiring the use of the enzymes of cellulase, α-amylase, polygalacturonase and protease, either individually or in a mixture, to degrade the structural cell wall components including mannan, galactomannan, arabinoxylogactan and cellulose. The study was conducted at 25°C and pH 6.5 with 0.1% (w/w) enzyme concentration. Individual enzyme and combinations of enzymes were used in this study. The grated coconut and water were mixed with a 1:4 ratio and incubated for 30 min with different enzyme combinations. The yield of the oil extracted by using protease and polygalacturonase are 32% and 36%, followed by cellulase and α-amylase with the same yield at 28%. The yields of the oil extraction using a combination of enzymes were found to be higher than that of individual enzymes. The yields for the cellulase and α-amylase combination is 36%, cellulase, α-amylase and polygalacturonase is 37% and cellulase, α-amylase, polygalacturonase and protease is 42%. The yields of the oil extracted with enzymes are higher than that of extraction without the enzyme at only 19%. The oil extraction using a combination of four enzymes gave a higher yield is due to the high hydrolysis rate of the cell wall components [28].

2.1.4. Chilling, freezing and thawing

A chilling, freezing and thawing method has been used to break the water–oil emulsion where the coconut milk will be centrifuged at 3220*g* for 10 min and the upper layer of the cream, supernatant was removed [32] prior to a chilling and freezing process to allow better packing of the oil globules [17]. Generally, the temperature used for chilling and freezing are 10 and −4°C respectively, for 6 h in total. Then, the thawing process is carried out in a water bath at 40°C until the centrifuged cream reaches the room temperature of 25°C. The cream is then centrifuged twice at 4000*g* for 30 min to obtain the VCO. During the thawing process, the oil droplets coalesced and form large droplets of various sizes, in turn giving a total oil extraction yield of 69% [32].

2.1.5. Wet extraction

Wet processing method, also known as the domestic method of extraction, involves the separation of water from the coconut milk, which the water extracts of the coconut sap that contains the oil–water (o/w) emulsion. The breaking of the emulsion is difficult due to the high stability of the coconut milk emulsion. The emulsion breaking process involves the steps of creaming, flocculation or clustering and coalescence. The creaming stage is done by the action of gravitational force that results in two phases where the higher specific gravity phase moves downward and lower specific gravity phase moves upward. It is followed by the flocculation or clustering where the oil phase aggregates without breaking the interfacial film that surrounds each oil globules. The last stage, coalescence is carried out by rupturing the interfacial phase among the oil globules so that the oil globules joined together and can be collected [33]. The wet extraction process of VCO is more desirable than the dry processing and organic solvent extraction methods due to its ability to preserve the natural compositions and fragrance of the oil, as well as being free from any forms of chemicals and/or organic solvents. Therefore, this process is more environmental friendly [17]. However, this extraction method has some drawbacks where the extraction process requires 24–48 h and the yield of oil is lower, which is approximately 40%.

2.1.6. Dry processing method

Dry processing method is used commercially to produce VCO by mechanical force because the oil extracted is low in moisture content, thus preventing the microbial contamination and facilitates in upscaling the extraction process to meet the market demand. The fresh grated copra meat is sun-dried or oven-dried at 40–50°C until the moisture is reduced from approximately 50% to the range of 2–5%. The yield of VCO was found to be corresponding to the moisture content, where the dehydrated grated coconut with lower moisture content will produce a higher yield of VCO. After drying the grated coconut, the oil is collected by cold-pressing, which gives a yield of approximately 46–49% [34].

2.1.7. Supercritical fluid extraction

A previous study indicated that 100% of oil could be extracted from the coconut copra within 1 h by using supercritical carbon dioxide ($SC\text{-}CO_2$) as the extraction solvent with the condition of 120°C and 517 bar. $SC\text{-}CO_2$ is selected in the VCO extraction due to its lower critical temperature and pressure that can prevent the degradation of minor and heat-sensitive components. Besides that, the non-polarity nature of $SC\text{-}CO_2$ is able to solvate the oil droplets and subsequently enhance the extraction process. During the vegetable oil extraction, the solubility of triglycerides in the $SC\text{-}CO_2$ is depending on the carbon number of the fatty acids on the triglycerides, where the shorter the chain, more soluble it is in the $SC\text{-}CO_2$ [35]. VCO is rich in MCFA, which constitutes of fatty acid chains with 16 carbons or less, makes it more soluble in $SC\text{-}CO_2$ compared to other vegetable oils such as canola oil, rapeseed oil and corn oil [36]. Besides that, the solubility of VCO in the $SC\text{-}CO_2$ increases with increasing pressure and temperature [36]. The optimum temperature and pressure obtained are 80°C and 345 bar with to the VCO extraction yield of 99% [27]. The extracted VCO can then be used directly without any further refining process. Even though this method is able to obtain higher yield of VCO compared to other conventional extraction methods, this emerging extraction technology is not as popular as others extraction methods mentioned above due to the costly set-up of extraction system.

As a summary, the most commonly used in the extraction method of VCO in the current market is the dry processing method. The other extraction methods are still being practised based on the production size. The traditional processing of CO is usually being practised for personal uses and ayurvedic treatment due to its small scale processing. In contrast, SFE has emerged into the VCO production industry recently due to its high extraction efficiency even though it is an expensive technology. Figure 1 presents the extraction methods of CO and VCO while Table 1 summarizes the advantages and disadvantages of the extraction methods.

2.2. Phytonutrients of CO and VCO

Virgin coconut oil and CO comprise of over 90% of saturated fatty acids and an extremely low percentage of unsaturated fatty acids. According to a previous study, VCO and RBD CO composed of 86.7–96.1% of saturated fatty acids and 6.6–8.3% of unsaturated fatty acids [37]. During late 1930s and in between 1980 and 1990, the commercial interest of domestic fats and oils industries in United States of America (U.S.A.) dropped due to the strong negative influence of CO from the U.S. and caused a decrease in the usage of CO in U.S. and other countries. In 1950, the composition of CO has resulted in various negative perceptions when a

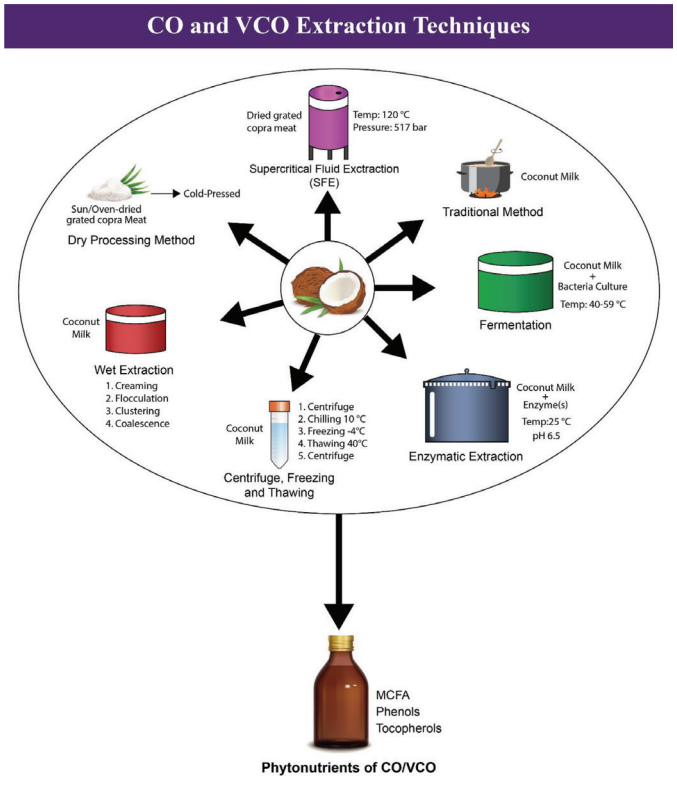

Figure 1 | Extraction methods of Coconut Oil (CO) and Virgin Coconut Oil (VCO).

Table 1 | The advantages and disadvantages of the extraction methods of CO and VCO

Extraction methods	Advantages	Disadvantages	Yield (%)	References
Traditional method	• Retains the pleasant odour • Has higher phenolics content	• Heat used may cause degradation of the minor compounds	33	[24,28]
Fermentation	• High recovery in shorter time	• Application of bacterial culture may subject to user safety in oral consumption	60–95	[29,30]
Enzymatic extraction	• High recovery	• A proper condition is required to maximize enzymes reaction and prevent degradation of enzymes	19–42	[28,31]
Chilling, freezing and thawing	• Prevent degradation of minor compounds as no heat is employed	• The oil droplets formed made up of various sizes	69	[17,32]
Wet extraction	• Able to preserve the oil's natural properties. • Free from chemical treatments and solvents	• The process requires 24–48 h with lower yield	40	[17,33]
Dry processing method	• Longer shelf life	• Prone to microbial contamination during the drying process	46–49	[34]
Supercritical fluid extraction	• High recovery of oil and phytonutrients • Preserve the oil's natural properties	• Large SFE set-up seems to be expensive for up-scaling	99	[27,35,36]

Table 2 | Phytonutrients of CO and VCO

Phytonutrients	Oil	Extraction methods	Contents	References
Medium Chain Fatty Acids (MCFA) (dominated by lauric acid of 46–48%)	VCO	SFE Chilling, freezing and thawing Enzymatic	56.0–67.0%	[17,27,36]
		Fermentation	58.5–61.7%	[41]
	RBD CO	Heated mechanical pressing		
	CO	Traditional boiling		
Total phenolics content	CO	Traditional boiling	618 mg GAE/kg oil	[24]
	RBD CO	Heated mechanical pressing	61.4 mg GAE/kg oil	[37]
	VCO	Fermentation	220 mg GAE/kg oil	[42]
		Dry extraction	91 mg GAE/kg oil	[24]
		Chilling, freezing and thawing Enzymatic	30 mg GAE/kg oil	[42]
Tocopherols (lowest if compared to other vegetable oils)	VCO	Enzymatic Fermentation	0.09 mg/kg oil	[41]
		Chilling, freezing and thawing Dry processing method	0.05 mg/kg oil	[41]
	RBD CO	Heated mechanical pressing		
	CO	Traditional boiling		

researcher in Minnesota announced that the heart disease epidemic are being caused by the hydrogenated vegetable fats with supportive response from the edible oil industries. During the same time frame, a study reported that the polyunsaturated fatty acids consumption could lower the serum cholesterol level but the information on the cholesterol lowering which is due to the incorporation of the cholesterols into the body tissues such as arteries and liver, was not mentioned. This fact was vastly accepted by the communities, and the replacement of saturated fats to polyunsaturated fats in the diets were emphasized. The ideas of anti-saturated fat were spread through as book written by Jeremiah Stamler (1963) as the earliest pejorative opinion on tropical oils, particularly CO since it is the only oil entered U.S. market at that time. Although CO was the major source of dietary fats in U.S. since 1890 to 1920 [15], this industry was heavily impacted for more than three decades due to the misinformation provided by the consumer activist groups, Centre for Science in the Public Interest and American Soybean Association, as well as members from edible oil industry, medical and scientific community [38]. The market of the CO industry was recovered in early 2000s due to unveiling of the truth of CO causes neither any alteration in the serum total cholesterol nor coronary heart disease or mortality [39,40]. Table 2 summarises the phytonutrients present in CO and VCO.

2.2.1. Medium chain fatty acids

Various extraction techniques have been studied and the results showed that the phytonutrients profile of VCO is better than CO in terms of shelf life. Unlike CO that is tailor-made for cooking purpose, VCO has been marketed and vastly accepted as a functional oil by the communities [17]. The VCOs extracted has the MCFA content in a range of 51.6–67.0%. The MCFA is dominated by lauric acid with the percentage ranging from 46% to 48%, which is within the limit outlined in the VCO Malaysian Standard 2007 (47–50%). A previous extraction study using SFE method reported that the solubility of VCO in SC-CO_2 increases with an increase of temperature from 40 to 80°C, at the pressure between 310 and 345 bar and contributed to a higher yield of VCO. However, adverse effect was seen if the pressure used is in between 241 and 270 bar where the solubility decreases even though temperature is increased [36]. This phenomenon is supported by another study conducted on the VCO

extracted by SFE that was found to contain about 56–67% of MCFA, depending on the temperature and pressure used in the extraction but independent of the extraction yield. Higher MCFA content with approximately 67% was found in VCO extracted using SFE method at low pressure and high temperature despite of lower yield of VCO with approximately 40% if compared to the extraction condition of high pressure and high temperature with MCFA content of 58% and VCO yield of 96% [27]. On the other hand, the VCO extracted by chilling, freezing and thawing, enzymatic and fermentation methods, the range of MCFA content was found to be 58.5–61.7% [41]. Overall, the SFE method is able to extract VCO with the highest MCFA content if optimum pressure and temperature is applied.

2.2.2. Total phenolic content

Total phenolic content of CO and VCO varies based on the extraction methods. The polyphenols including caffeic acid, *p*-coumaric acid and ferulic acid are responsible for the phenolic-dependent antioxidant capacities for VCO. TPC was found to be much higher in the CO extracted using traditional boiling method than VCO from dry processing method, with 618 and 91 mg GAE/kg oil, respectively. Since traditional method involves water as the extractor, the polar phenolic compounds will be dissolved in the aqueous phase of coconut milk and retained in the CO after evaporation of water [24]. Furthermore, the TPC value of RBD CO is the lowest with 61.4 mg GAE/kg oil since the refining process of RBD CO degrades and removes some of the polyphenols [37]. The TPC values of VCO extracted from different methods were reported where the VCO obtained from fermentation is 220 mg GAE/kg oil, which is higher than that of enzymatic method and chilling, freezing and thawing method with 30 mg GAE/kg oil [42]. Up to date, there is no report on the TPC of VCO extracted by SFE. However, the presence of the modifiers such as ethanol could enhance the polarity of SC-CO_2, in turns increase the TPC of VCO extracted using the SFE method [43].

2.2.3. Tocopherol content

The tocopherol content in VCO is minimal when compared to other vegetable oils such as palm oil and olive oil [44]. According to a previous study, only three types of tocopherols were found to be present in the VCO which are β-tocopherol, γ-tocopherol and δ-tocopherol [41]. Approximately 0.05 mg/kg oil of tocopherol was detected in the VCO extracted by chilling, freezing and thawing method and dry processing method while enzymatic extracted and fermentation extracted VCO gave 0.09 mg/kg oil of tocopherol [41]. Furthermore, α-tocopherol content of VCO extracted by enzymes, fermentation and chilling, freezing and thawing methods were 3, 22 and 3 mg/g oil respectively and reported through the usage of calorimetric assay via a UV/Vis Spectrophotometer and α-tocopherol as the standard to construct the reference curve for the assay to determine the total tocopherol content [42]. So far, there is no report found on the tocopherol content of VCO extracted by SFE method.

2.3. Antioxidant Activity of CO and VCO

Virgin coconut oil is well known for its phenolic contents, which are found in the coconut kernel skin, which is known as coconut testa. A previous study was conducted on the antioxidant properties of VCO extracted from chilling, thawing and freezing and fermentation techniques by using commercial RBD CO as the control through DPPH radical scavenging activity. The EC_{50} values obtained for VCO extracted by fermentation process is 1.24 mg/mL while chilling, freezing and thawing technique is 1.66 mg/mL. The results revealed that VCO possessed stronger scavenging activities with more than 90% of inhibition effect if compared to RBD CO with EC_{50} value of 3.23 mg/mL that gave 81% of inhibition effect. Similarly, the β-carotene bleaching assay showed that VCO exhibited stronger activity if compared to RBD CO at the concentration of 1 mg/mL with the bleaching effect of 65% and 56%, respectively [44]. The findings were further supported by another study that indicated VCO has shown stronger antioxidant activity than RBD CO and this may be due to the higher amount of unsaponifiable components such as polyphenols and vitamin E in VCO [40,41,45]. Therefore, the demand for VCO as functional food oil is rising due to an increased awareness of its benefits [17,46], in turns bringing new opportunities in developing various nutritious edible products rather than consuming the oil solely.

3. PALM-PRESSED MESOCARP OIL AND CRUDE PALM OIL

Generally, a palm oil mill extracts only 20% of the oil from fresh palm oil (*Elaeis guineensis*) fruit and produces various by-products including PPMF after the mechanical extraction, which involves screw-pressing of the fresh palm mesocarp to produce mesocarp oil or CPO. Since the extraction is conventionally done with single stage pressing, the remaining oil and phytonutrients in the PPMF is considerably high in amount and worth to be recovered. The oil and phytonutrients recovery could help lower the wastage, besides solving the problems of by-products generated from the palm oil mill. In 2017, the oil palm industry in Malaysia processed about 103.94 million tonnes of FFB into 19.92 million tonnes of CPO and generated 14.55 million tonnes of PPMF with 5–6% of residual oil content, Palm-pressed Mesocarp Oil (PPMO). Based on the amount of FFB processed in 2017, it is estimated that 728,000 tonnes of PPMO can be recovered from 454 palm oil mills per year [47]. The utilization of the PPMO is able to increase the revenue of palm oil industry annually, especially in Indonesia and Malaysia which are the world's largest palm oil producer by processing the FFB to increase the PPMO recovery.

Palm-pressed mesocarp fiber is mainly used as the burning fuel in the boilers [48] and to generate electricity in the palm oil mills. The piling up of PPMF under high humidity conditions may cause environmental issues such as microorganism incubation and release of methane gas and odour. PPMO is an emerging oil that attracts the interest of many researchers due to its valuable phytonutrients besides increasing the yield of palm oil. The amount of oil entrapped in the PPMF is influenced by the condition of sterilization of FFB, condition of the sterilised fruits in the digester, and the pressure exerted on the palm mesocarp during the mechanical pressing for CPO extraction [21]. PPMF contains about 5–6% of residual oil which constitutes of valuable phytonutrients such as carotenoids (vitamin A), tocotrienols and tocopherols (vitamin E), squalene, phytosterols and coenzyme Q_{10} [7]. The phytonutrients content of CPO extracted from fresh palm fruit mesocarp and PPMO extracted from PPMF is different and this may be due to the utilization of the mesocarp at different stages.

Previous studies reported that the amount of minor components in PPMO is higher if compared to CPO, resulted from the removal of longer fatty acid chains, especially C16–C20 in the first press of CPO extraction. The total vitamin E concentration in PPMO was found to be 2000–2600 mg/kg, which is twofold higher than CPO with 600–1000 mg/kg. The vitamin E profile of PPMO is also different from CPO in terms of the percentage of tocopherol and tocotrienols. PPMO contains 63.7% α-tocopherol and 36.3% tocotrienols while CPO has 22.0% α-tocopherol and 78.0% tocotrienols [20]. Furthermore, the carotenoids content of PPMO ranges between 4000 and 6000 mg/kg, which is fourfold higher than CPO that contains only 500–700 mg/kg [7]. Lycopene, a carotenoid present in PPMO is approximately 14%, which is 10 times higher than CPO, resulting in an intense reddish colour of PPMO. The phytosterols detected in PPMO ranges between 3000 and 4800 mg/kg, which is 10 times higher than CPO (250–650 mg/kg) and the amount of squalene found for PPMO is threefold higher than CPO with 1100–1600 and 250–500 mg/kg, respectively [20]. Coenzyme Q_{10} or ubiquinone, a well-known antioxidant, was found in PPMO with a higher concentration of 1000–1500 mg/kg than CPO with only 10–80 mg/kg [49].

3.1. Extraction Methods

3.1.1. Crude palm oil extraction by screw-pressing

The harvested FFB are processed immediately in palm oil mills to minimize the Free Fatty Acids (FFA) generation, which will affect the quality of CPO. In the first stage, the FFB are sterilized by steaming in the sterilizer for 1–1.5 h at a pressure of 3 bars [50] to denature the lipase enzyme and minimize its triglyceride hydrolysis activity, which prevents FFA generation and facilitating the stripping of the fruitlets from the Empty Fruit Bunch (EFB). FFB sterilization also helps in the palm fruit mesocarp conditioning for oil extraction. The sterilized FFB will be stripped off from the fruitlets in a rotating thresher drum that generates EFB as the by-product. EFB will be used for the mulching purpose in the plantation or composted to be used as organic fertilizer.

The stripped fruitlets are then loaded into the digester where the fruitlets are heated and stirred continuously to loosen and detach the mesocarp from the kernel. This process also helps to break the oil cells and release the CPO even before the screw pressing. The liquor ex-screw press is then diluted with hot water before being screened to remove the remaining fibers and coarse contaminants. Upon completion of the screening, the diluted oil will be pumped into a clarifier tank to separate the CPO from the water/solid fraction. The clarified oil is further cleaned by a high-speed purifier to remove dirt and moisture before being channelled into a vacuum drier. Finally, the dried CPO is stored and despatched to the refineries for the RBD process. The overall CPO recovery is approximately 90–93%, which is subjected to the plant machinery and process control [50].

3.1.2. Soxhlet extraction

This is a commercial extraction method of PPMO [51] and has been practised in some of the palm oil mills. The freshly collected PPMF from palm oil mill is oven-dried at 50–60°C for 1 h. Then, the dried PPMF will be extracted with *n*-hexane by using Soxhlet set-up, which is a solvent reflux system that continuously extracts the PPMF with the solvent for 4–6 h. The extracted residual oil will be evaporated to remove the solvent [7]. Then, the oil will be vacuum-dried or oven-dried to remove the remaining solvent. The solvent extracted PPMO usually contains gums and phospholipids that need to be removed by undergoing refining process such as degumming and bleaching, which is similar to CPO [51].

3.1.3. Supercritical fluid extraction

Supercritical fluid extraction is considered as an emerging method to extract PPMO by using fresh or oven-dried PPMF. The fiber is loaded into the extraction vessel and extracted with SC-CO_2 at the temperature in between 50 and 80°C and pressure of 300 bar for 2 h [7,20]. The flow rate of CO_2 into the vessel is in the range of 3 and 10 mL/min. After completion of extraction, the residual oil extract will be collected and the CO_2 can either be released from the chamber or recycled [20]. Previous study reported that the fresh PPMF extracted by SC-CO_2 at 60°C and 300 bar could extract the water-soluble phenolics through the steam condensation during sterilization of FFB, and the water injected in the screw-press during mechanical pressing for CPO extraction [52]. SFE is a single-step process and does not require any further refining or degumming process, since the extracted PPMO is free from any solvent or gum contamination.

Overall, SFE is a highly recommended technique for the PPMO extraction. Currently, solvent extraction method is commonly practised and has been prioritised due to easy handling in the mills. However, the PPMO extracted needs to be treated in the same way as the CPO to undergo RBD process. The solvent extraction is not encouraged due to the utilization of PPMO as an edible oil. The solvent *n*-hexane used in the extraction process is a health hazard. Moreover, there is less processing time and steps in SFE, resulting in more energy and cost savings in the palm oil industry. Therefore, SFE technique is preferred in the extraction of PPMO. Figure 2 presents the extraction methods of CPO and PPMO while Table 3 summarizes the advantages and disadvantages of the extraction methods.

3.2. Phytonutrients of Palm-Pressed Mesocarp Oil

3.2.1. Carotenoids, vitamin E and sterols

Palm-pressed mesocarp oil, which is extracted from the recovered mesocarp fiber from pressed palm fruitlets, has an excellent phytonutrient profile. A previous study was conducted to extract the PPMF by using solvents such as *n*-hexane and chloroform, as well as SFE method. The results showed that the solvent extracted PPMO seems to contain lower amount of carotenoids (3800–5300 mg/kg) and vitamin E, tocopherol and tocotrienols (1200–2600 mg/kg) but higher amount of sterols (6906–8490 mg/kg), comprised of β-sitosterol (56.0–58.4%), campesterol (19.9–21.2%) and stigmasterol (18.7–19.6%). Conversely, the PPMO extracted from SFE possesses higher content of carotenoids (4100–6000 mg/kg), vitamin E, tocopherol and tocotrienols (2500–3000 mg/kg) but

lower of sterols (4509–5200 mg/kg), comprised of β-sitosterol (56.5%), campesterol (22.0%) and stigmasterol (19.0%). Both PPMO extracted from solvent and SFE has a negligible amount of cholesterol.

For the CPO, the major carotenes present are α- and β-carotenes that comprise of 90% of the total carotenes present. In contrast, the α- and β-carotenes present in PPMO comprise of 50% of total carotenes present, thus indicating other carotenes are present in higher amount.

3.2.2. Fatty acid profile

For the fatty acid profile of PPMO, a higher percentage of lauric acid (C12:0) and myristic acid (C14:0) were detected and the outcome is due to the presence of some broken nuts and kernel shells in the PPMF [7]. For the extraction of MCFA, solvent extraction seems to extract lauric acid (C12:0) and myristic acid (C14:0) in higher concentration than of SFE. The yield of lauric acid and myristic acid extracted by solvent are 13.8% and 6.2%, respectively, while SFE obtained lower yields of 8.0% and 4.2%. The explanation is that the solvents have very low polarity index, thus having higher affinity toward non-polar compounds such as lauric acid and myristic acid if compared to SC-CO_2.

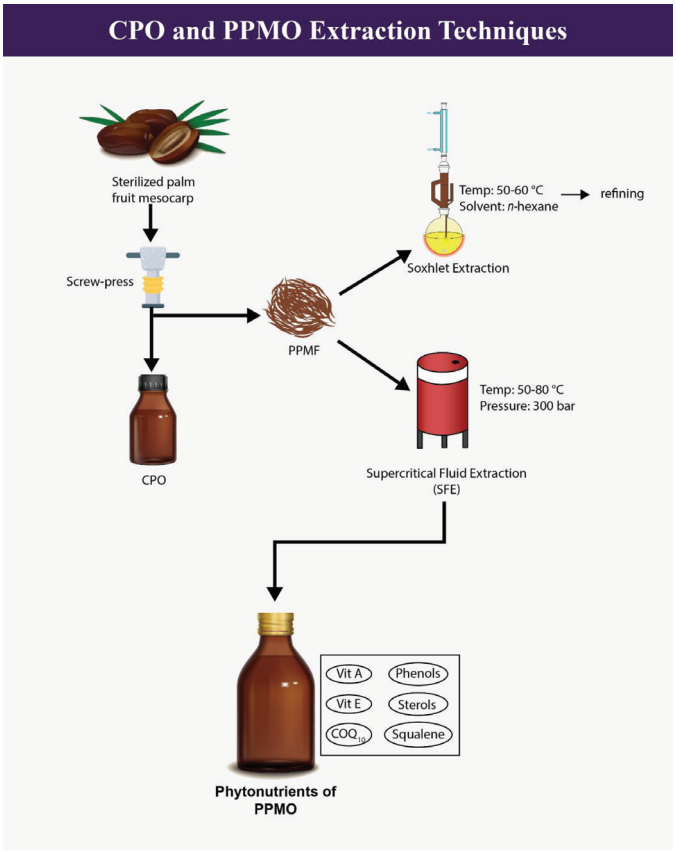

Figure 2 | Extraction methods of Crude Palm Oil (CPO) and Palm-pressed Mesocarp Oil (PPMO).

3.2.3. Squalene

A previous study was conducted to compare the composition of PPMO extracted from dried and fresh PPMF by using solvent *n*-hexane and SFE. The results showed that PPMO contains squalene, which is considered as a minor component in PPMO. The squalene content in PPMO extracted from dried fiber is higher than fresh fibers. PPMO extracted using the SFE method netted 1642–1633 mg/kg using dried fiber, and only 1343–1102 mg/kg using fresh fiber, meanwhile, PPMO extracted using solvent extraction netted 1495 mg/kg using dried fiber, and only 1117 mg/kg using fresh fiber. Besides that, other phytonutrients content were found to be higher in the PPMO extracted from dried fiber by using SFE method at 50–80°C and 300 bar if compared to that from fresh fiber by using solvent extraction method [20]. The finding is further supported by a study that reported some other phytonutrient compositions are higher in SFE extracted PPMO, except for the phytosterol [7]. The lower phytosterol content may be due to lower solubility of the sterols in the liquid CO_2 at lower pressure.

3.2.4. Coenzyme Q_{10}

The solvent extraction of PPMO by using ethanol (EtOH) revealed the presence of coenzyme Q_{10} in the range of 1000 and 1500 mg/kg [49], which is tremendously higher than that in CPO with a range of 10 and 80 mg/kg [52]. The high contents of carotene and vitamin E contents in PPMO tend to mask the occurrence of the coenzyme Q_{10} in PPMO. Therefore, PPMO was pre-treated with saponification so the unsaponifiables can be extracted with *n*-hexane. The spectroscopic characterization of coenzyme Q_{10} was conducted by using Supercritical Fluid Chromatography (SFC) where C18 reversed-phase column was used, together with SC-CO_2 as the mobile phase. Prior to separation, the unsaponifiable fraction was subjected to open column chromatography to extract the coenzyme Q_{10}. The silica gel packed column was eluted with pure *n*-hexane, followed by pure EtOH and both fractions were then subjected to SFC separation. The results indicated that coenzyme Q_{10} is present in the EtOH fraction, indicating the polar nature of this phytonutrient [49].

Table 3 | The advantages and disadvantages of extraction methods of CPO and PPMO

Extraction methods	Advantages	Disadvantages	References
Screw pressing of CPO	• High yield of oil (90–93%)	• Leaves some residual oil in the extracted fiber • The extracted oil need to undergo RBD process	[47]
Solvent extraction of PPMO	• The oil extracted found to be stable against oxidation	• Need to undergo RBD process since the extract tend to contain gums and phospholipids	[7,44]
Supercritical Fluid Extraction (SFE) of PPMO	• The PPMO extracted can be used directly without undergoing further purification procedure	• The large scale SFE set-up seems to be expensive for up-scaling	[7,20,48]

Table 4 | Phytonutrients found in CPO and PPMO

Phytonutrients	Oil	Extraction methods	Contents	References
Carotenoids	PPMO	SFE	4100–6000 mg/kg oil	[20]
		Solvent extraction	3800–5300 mg/kg oil	[20]
	CPO	Screw-pressing	700–500 mg/kg oil	[7]
Tocopherol and tocotrienols	PPMO	SFE	2500–3000 mg/kg oil	[20]
		Solvent extraction	1200–2600 mg/kg oil	[20]
	CPO	Screw-pressing	1000–600 mg/kg oil	[7]
Sterols (Comprised of β-sitosterol, campesterol, stigmasterol and a negligible amount of cholesterol)	PPMO	Solvent extraction	6906–8490 mg/kg oil	[20]
		SFE	4509–5200 mg/kg oil	[20]
	CPO	Screw-pressing	650–250 mg/kg oil	[7]
Fatty acids profile	PPMO	Solvent extraction	Lauric acid: 6.2%; myristic acid: 13.8%	[7]
		SFE	Lauric acid: 4.2%; myristic acid: 8.0%	
Squalene	PPMO	Solvent extraction	1117–1495 mg/kg oil	[20]
		SFE	1102–1642 mg/kg oil	[20]
	CPO	Screw-pressing	500–250 mg/kg oil	[7]
Coenzyme Q_{10}	PPMO	Solvent extraction	1000–1500 mg/kg oil	[49]
	CPO	Screw-pressing	80–10 mg/kg oil	[52]
Polyphenols (Water soluble)	PPMO	SFE	1000–2000 mg/kg oil	[7,53]

3.3. Phenolic Compounds and their Antioxidant Activity

The phytonutrients present in PPMO are found to be higher in concentration when compared to CPO, regardless of the extraction techniques [7]. Furthermore, the SC-CO_2 extracted PPMO by using the fresh fiber, contains additional water-soluble phenolics compounds with concentration of 1000–2000 mg/kg oil [53], which are neither found in the dried fiber extract, nor through solvent extraction method. A total of 12 types of water soluble phenolics were identified and evaluated with the antioxidant assays through Ferric Reducing Ability of Plasma (FRAP) and Trolox Equivalent Antioxidant Capacity (TEAC) methods at the concentration of 1 mg/mL. The results of TEAC indicated that the water-soluble phenolics possess superior antioxidant capacity, which is 10 times more effective than the standard compound, α-tocopherol. On the other hand, FRAP results revealed an eightfolds reduction in the antioxidant capacity of PPMO when the water layer is removed from the oil [53]. Table 4 summarises the phytonutrients present in CPO and PPMO. The broad spectrum of phytonutrients present in PPMO will eventually lead to its growth in the health supplement market where various options of oil intake may be available such as oil capsule or PPMO incorporated food products. Besides, the high carotenoids content in PPMO may contribute to the natural food colorant industry.

4. CONCLUSION

Virgin coconut oil and PPMO have been proven to show health-promoting effects, attributed to the presence of MCFA and polyphenols which are one of the contributors to the antioxidant activities. Various extraction techniques have been studied and used in the VCO and PPMO extraction. The choices of extraction methods and their extraction conditions are crucial in the yield of oil, as well as preserving the natural phytonutrients composition of oils. For the edible oil market, solvent free extraction of VCO and PPMO have been focused. SFE technique will eventually become the best choice due to its advantages of being high yield and solvent free, despite its high installation cost. Lastly, the utilization of the mesocarp fibers by recovering the residual oils not only reduces the waste generated from palm oil mills, it recovers valuable phytonutrients. The phytonutrients profile of VCO and PPMO are highly potential in creating new opportunities to functional food industry where related nutritious edible products could be produced. Thus, future studies on the detail identification of the phytonutrients present in the VCO and PPMO extracted from SFE is highly recommended in order to unveil their nutritional values.

CONFLICTS OF INTEREST

The authors declare they have no conflicts of interest.

AUTHORS' CONTRIBUTION

SS contributed in literature search and manuscript drafting while SHM contributed in manuscript conceptualization and editing.

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