Chapter 16
Argan [*Argania spinosa* (L.) Skeels] Oil

Dominique Guillaume, Daniel Pioch, and Zoubida Charrouf

Abstract Argan oil is extracted from the kernels of *Argania spinosa* (L.) Skeels, a tree that almost exclusively grows endemically in southern Morocco. If argan oil was initially only known around its traditional production area, major efforts combining chemical, agronomic and human sciences have led to its international recognition and marketing. In addition, to ensure the sustainable production of a sufficient quantity of argan kernels, a vast and unprecedented program that led to the reforestation of large areas of drylands has been developed in Morocco. Therefore, argan oil production is considered as an economic and ecologic success.

Edible argan oil is prepared by cold-pressing roasted argan kernels. Unroasted kernels afford an oil of cosmetic grade, showing a bitter taste. Both oils, which are not refined and are virgin oils, share a similar fatty acid content that includes oleic and linoleic acids as major components. Additionally, argan oil is rich in antioxidants. Together, these components likely contribute to the oil pharmacological properties that, in humans, traditionally included cardiovascular disease and skin protection. Recent scientific studies have greatly expanded the scope of these pharmacological activities.

Argan oil is now rewarded with a "Geographic Indication" that certifies its exclusive and authentic Moroccan origin and the compliance with strict production rules. In addition, the quality of argan oil can nowadays be ascertained by using an array of physicochemical methods.

By-products, generated in large quantity during argan oil production, are also finding promising development routes.

D. Guillaume (✉)
Faculté de Médecine/Pharmacie, URCA, Reims, France
e-mail: dominique.guillaume@univ-reims.fr

D. Pioch
CIRAD, UR BioWooEB Biorefinery Team, Montpellier, France
e-mail: daniel.pioch@cirad.fr

Z. Charrouf
Université Mohammed V-Agdal, Laboratoire de Chimie des Plantes, Synthèse Organique et Bioorganique, Faculté des Sciences, Rabat, Morocco

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1 Introduction

The argan tree [*Argania spinosa* (L.) Skeels] is a spontaneous, thermophilic and xerophytic tree that belongs to the Sapotaceae family. Before the Pleistocene (Quaternary) glaciations, the argan forest likely extensively covered very large areas in Northern Africa. However, the series of glacial events that characterized the Pleistocene glaciations induced the nearly total extinction of the argan tree from the Maghreb and Sahelian band. Providentially, in the Souss Valley (Morocco) a microclimate simultaneously resulting from temperate oceanic conditions and steepness-related gelid wind protection allowed optimum conditions for the argan tree survival. This environment prevented the argan tree complete extinction (Kenny and De Zborowski 2007). Hence, today the argan tree is an 80-million-year-old relic and is the only species of the family Sapotaceae remaining in the subtropical zone.

The argan tree almost exclusively grows in Morocco on an 870,000–1000,000 ha area referred to as "the argan forest" (Charrouf and Guillaume 2008a). Unfortunately, vast parts of the argan forest present a long history of degraded vegetation (McGregor et al. 2009). This decrepitude is the result of a slowly evolving phenomenon that is now properly addressed but that Moroccan authorities and local people took unfortunately a long time to realize.

The argan forest is a unique flat, hilly, or steep landscape, which constitutes a fragile biotope dominantly populated by an Amazigh agrarian population. Imazighen (singular Amazigh) constitute an ethnic group that brings together the descendants of Northern Africa's original inhabitants before the Romans and the Arab Muslim conquests in the seventh century. In the Souss Valley, as well as other remote areas in Morocco such as the central Rif, Amazigh (sometimes mistakenly called Berber) population makes a socially active group mainly characterized by matriarchal traditions. In the inland, the argan tree must co-exist as harmoniously as possible with ancestral practices allowing domestic young goat, and in its southernmost part stray camels, grazing (Chatibi et al. 2016). On the Atlantic coast, the population is more mixed and urban but the economic activity principally based on tourism is another type of threat for the argan tree. The argan forest was designated by the "UNESCO's Man and Biosphere program" as a Biosphere in 1998.

Hence, argan oil is the finest natural production of a unique type of tree and is itself unique in its composition and properties. Multivariate discriminant analysis of its fatty acid content has shown that it resembles to sesame oil and presents some similarities with high-oleic sunflower oil (Rueda et al. 2014).

Argan oil is extracted from argan kernels. In the South-Moroccan Amazigh culture, argan oil has for centuries been used for its nutritional as well as dermatologic properties. In this latter field, argan oil is particularly recommended to cure skin

pimples, juvenile acne, and chicken pox pustules (Charrouf and Guillaume 1999). As edible oil, argan oil is the main part of the source of the necessary daily lipid fraction of the rural population of the Souss Valley, thus being the basic ingredient of the Amazigh diet (Charrouf and Guillaume 2010).

For cultural and ancestral reasons, women exclusively achieve the preparation of argan oil. Until the 1980s, argan oil preparation was performed following an atavistic process at the family-scale. Most of argan oil properties were unknown or limited to the traditional knowledge of rural women. A vast scientific and multidisciplinary program, both nationally and internationally funded, and named "the Argan Oil Project" came out in the mid-1980s and lasted for almost 40 years (Charrouf and Guillaume 2011, 2018; Charrouf et al. 2011). The Argan Oil Project cumulatively led to the complete reconstruction of the argan oil production, marketing, and delivery chains (Turner 2016), the improvement and optimization of the oil preparative process, the design of distinct cosmetic and edible argan oil grades, the determination of argan oil detailed composition, the design of scientific methods ascertaining argan oil quality, the building of production units based on a woman-only cooperative system, the establishment of an official quality norm, the recognition of a Geographic Indication along with regulatory certification allowing the marketing of argan oil as "endemic argan oil of Morocco", the sustainable management of the argan forest, and finally it allowed to confirm some of the alleged pharmacological benefits of the oil (Charrouf and Guillaume 2014). In short, the Argan Oil Project revolutionized the fate of argan oil by igniting and perpetuating its worldwide popularization. Accordingly, whereas argan oil export was estimated to be 1 ton in 1996, it was 40 tons in 2003, reaching 400 tons in 2009, to boom to 1000 tons in 2014 (Roumane 2017). Recent estimates predict argan oil market to reach almost 20,000 tons by 2022 (Khallouki et al. 2017a). Today, argan oil is found on the North-American, European and Japanese markets and its largest importers are France and Germany with 650 and 100 tons, respectively, in 2014 (Roumane 2017). In parallel, the price of 1 l of argan oil has jumped from a few dirhams in the 1980s (Moroccan market) to 150 euro in 2017 (European market).

In terms of research and development, the Argan Oil Project is often shown as an unprecedented wonderful success story. However, there were some troubles, at least at the start, and sharp questions have been regularly raised regarding some aspects of the project throughout its establishment (Lybbert et al. 2002, 2011; Simenel et al. 2009; Le Polain de Waroux 2013). Presently, some cooperatives must still strive to reach sustainable growth and special attention should be paid so that Amazigh families do not be deprived of argan oil for their own use (Huang 2017). In addition, the wealth produced by the argan oil growing market has not affected identically all argan forest dwellers (Le Polain de Waroux and Chiche 2013). Anyway, the argan oil project frequently illustrates the work to be done to enhance the value of a traditional production and bring it from a local product to a major actor of the global economy. This chapter is aimed at giving a whole picture of the odyssey of argan oil.

2 The Argan Tree

The argan tree is a slow-growing and thorny evergreen tree, gnarly with aging, particularly well adapted to survive extended periods of drought. It can grow as a bush or, if isolated and under sufficiently fertile and favorable conditions, it can reach 7–10 m high. Indeed, the argan tree growth can be strongly modified by its environment. For example, browsing and human use can reduce tree size and leaf production (Ain-Lahout et al. 2013). Natural conditions also affect the tree growth, the proximity of the ocean and its associated high salinity brings some influence, the coastal population of *A. spinosa* being possibly subjected to higher stress than the inland trees. Aridity and anthropogenic environmental changes also influence argan tree growth (Alados and El Aich 2008; Zunzunegui et al. 2017). Accordingly, it has been proposed that observed oil content variations in *A. spinosa* kernels could result from combined genetic and environmental factors (Aabd et al. 2014). This should be taken into account for selecting elite genotypes to be used for reforestation purposes.

Argan tree can easily live 150 or 200 years. In the Souss Valley, the argan tree has always played an important ecological function (Morton and Voss 1987; Ruas et al. 2016) and its participation in biodiversity preservation is essential. Indeed, if the argan forest experiences generally arid conditions, violent rains can sometimes occur and very strong winds can blow over the forest. Because of it's widely spread root-system, the argan tree, which in some places represents the only permanent vegetation, protects the soil against erosion and is a natural dam to slow the desert progression (Le Polain de Waroux and Lambin 2012). The argan tree shades all kind of domestic cultures, as barley or other types of easy-to-grow cereals, and brings moisture to the soil (Morton and Voss 1987). Doing so, its guarantees and perpetuates the soil natural fertility. The rural Amazigh population in the Souss Valley also uses Argan leaves as hanging forage for cattle.

However, due to the combination of several factors including a prolonged over use of argan trees during the nineteenth century and up to the first half of the twentieth century, several recent consecutive arid years, demographic pressure, and ill-management imposed by the very touristy nearby coastal zone, the argan tree has, once again, been threatened with slow extinction (Mellado 1989). Immediate consequences could have been disastrous with an irremediable advance of the desert without the Argan Oil Project that took steps to raise awareness of desertification in South Morocco and, subsequently, the Moroccan government encouraging a vast reforestation program (Charrouf et al. 2008). The argan oil project, based on a sustainable development approach (Charrouf and Guillaume 2009), started progressively and during the year 2005, the infant year of the agroforestry part of the project, only 500 ha were reforested, causing people to doubt about the feasibility of the project (Fazoui 2015). Then, a cumulative area of 100,000 ha was reforested between 2012 and 2017 according to the Moroccan authorities. In addition, and as a proof of awareness by rural people, during the last two decades, the areas dedicated to organic farming significantly increased. In most other areas, trees are labeled as "Wild collection" meaning that inhabitants respect their environment (Azim 2017).

In natural conditions, multiplication of the argan tree is limited because of its slow pace of growth. Propagation from seeds is difficult and argan tree natural renewal is restricted (Lopez Saez and Alba Sanchez 2009). Indeed, germination is influenced by genotypic parameters and environmental conditions, such as light, temperature or soil moisture, this latter being a frequent limiting factor in the argan forest (Alouani and Bani-Aameur 2004). Nonetheless, recent progresses in horticulture science, as well as specific care brought to young plants during the 1st years following plantation, have allowed to get a satisfactory argan tree planting success rate, and as good or better results than those obtained by using complex and alternative multiplication methods such as vegetative propagation or in vitro culture (Bousselmame et al. 2001; Nouaim et al. 2002; Justamante et al. 2017). As a consequence of these horticultural improvements, the argan oil has become the main national organic product, reaching 72% of the Moroccan organic production (Azim 2017).

Argan tree trunk grows gnarly with age. Argan tree leaves are 2 cm long and 0.5 cm wide, on average and generally permanent unless facing strong climatic stress. The argan tree blooms in August, spring or autumn, depending on climatic conditions. Its hermaphrodite flowers are small and yellowish. Argan fruit appears generally after the autumn rains. It ripens in spring and from April to June and argan trees get covered by the fruit that slowly to turn bright yellow between June and September. The fruit is a drupe weighting from 5 to 20 g and consisting of a pericarp and an endocarp (Sandret 1957). The fruit shape may vary depending on each individual tree: oval, spherical, elongated (fusiform), or pointed (apiculate). The pericarp is made of an epicarp (skin) covered by wax and a mesocarp (pulp). The pericarp, traditionally used as feed for goats and sheep, contains soluble sugars, hemicellulose and cellulose, latex, lipids and polyphenols (Kenny and De Zborowski 2007; Chernane et al. 1999; Pioch et al. 2011, 2015a, b). The endocarp contains a nut very rich in lipids whose extraction provides the argan oil.

3 Extraction and Processing of Argan Oil

In the Souss Valley, women have extracted argan oil from argan kernels for centuries following a hand-based method passed down to each generation from mother to daughter. Based on this age-old know-how, in the argan oil project context, a semi-mechanized method was designed 40 years ago to increase the efficiency of the argan oil extraction process and reduce its hardness. Special care was taken to avoid these introduced mechanical improvements altering the fragile equilibrium of the Amazigh society and to fully respect and preserves its traditions. To fulfill these goals the semi-mechanized method was exclusively implanted in woman cooperatives (Charrouf et al. 2011), to continue to acknowledge the ancestral work of women and their specific skill on this subject. This is particularly the case for the breaking of the nutshell without damaging the contained kernels given the shell hardness and variation in shape and size.

3.1 The Argan Fruit Harvest

The argan forest occupies a special place in the Amazigh culture (Simenel 2011) and the property of the fruit is regulated by ancestral and complex rules known by the Amazigh family. In general, fallen fruits belong to everyone (Charrouf et al. 2011). Schematically, traditional argan oil production by Amazigh women culminates between June and September, concomitantly or shortly after the beginning of the argan fruit pick up the season (Charrouf and Guillaume 2011). During this period of time, women (and sometimes kids), daily stride through the argan forest with large wicker baskets and collect ripe fruit that have fallen on the ground, underneath the trees.

3.2 Argan Fruit Processing Prior Oil Extraction

Argan fruit is air-dried for a few days in a ventilated and sunny place. Air drying makes make the initially latex-rich and sticky peel and flesh easier to be discarded, and so the fruit is more prompt to deliver the nuts. Indeed, once the peel is dry, it gets brown and brittle and it can be removed by rubbing it with stones. Nowadays, this step has been mechanized and is efficiently performed by scratching-machines. Need to mention that goat-peeled nuts -whole fruits eaten by goats, then regurgitated without peel and pulp- were sometimes formerly used for argan oil production. This method has been vastly popularized by tourist magazines. However, it is now prohibited in women cooperatives because of the associated bacteriological concerns and quality deterioration of the resulting oil. In the next step, argan nuts have to be broken to free the kernels from their shell. For this, women hold the nuts between thumb and forefinger and violently hit the nut apex with a stone (Charrouf and Guillaume 2011). Nut-breaking is a painstaking process, inexperienced women frequently crushing their fingers with the heavy stone. Nut-breaking is also a thankless task because 40 kg of nuts must be processed to obtain only about 2 kg of kernels.

Collected kernels are then differently processed depending on whether edible or cosmetic argan oil is to be produced. To prepare cosmetic oil, crude kernels are directly processed. To prepare edible oil, kernels have to be roasted for a few minutes in order to temper natural bitterness and provide argan oil with its typical hazelnut flavor. Traditional open-fire roasting was replaced in woman cooperatives with gas- or electric-burners for allowing a more accurate temperature adjustment. Such technology allows the simultaneous roasting of large quantities of kernels and the preparation of an oil of reproducible chemical and organoleptic quality. To prepare cosmetic oil, crude kernels are processed without roasting.

3.3 Argan Oil Extraction

Kernels (roasted or not) need then to be crushed to deliver argan oil. Traditionally, a millstone made up of a bedrock and a rotating cone-shaped grinding stone was used. Such method allowed the formation of an oily dough whose lipid content was extracted after prolonged hand-kneading. Water was added to the dough to facilitate malaxing and the resulting emulsion was decanted to finally deliver argan oil. Using this method, up to half the initial oil content could not be recovered, trapped in the dough. Nowadays, kernel extraction is performed by use of electric endless screw mechanical expellers (Charrouf and Guillaume 2008a). With such optimized instrumentation (Mountasser and El Hadek 1999), the residual amount of oil in the oil cake is very low and the pressing time is reduced by a factor of 3.5. In woman cooperatives where above described mechanized production methods are applied, four liters of argan oil can be obtained from 100 kg of dry fruit, requiring 16–20 h for one operator (Charrouf et al. 2002). Then, argan oil is only filtered after pressing, for clarification. It is not refined thus keeping highly active minor components. Above described improvement of most steps, opened the way to the large-scale production of argan oil, making this technical progress, compared to the traditional method, an important milestone in the Argan Oil Project.

A critical step in the validation of the large scale and semi-mechanized production of argan oil was the certification that press extraction was fully preserving the nutritional and dietary properties traditionally attributed to hand-extracted argan oil. This was achieved in 2005 by analyzing the composition and physico-chemical properties of 21 randomly selected samples of semi-mechanically or traditionally prepared (including goat-peeling) argan oil through the argan forest (Hilali et al. 2005). This study clearly established the similarity of the composition, in terms of fatty acids and minor components, between hand-extracted and semi-mechanically prepared and the superior quality the latter over the former. Consequently, cold-pressed argan oil, that had already been shown to present quality characteristics similar or better than non-linolenate oils (Yaghmur et al. 2000), was found perfectly suitable for shallow or pan-frying as long as it is not repeatedly used (Gharby et al. 2013b). Moreover, these studies showed the enhanced stability of argan oil during deep fat frying, compared to other oils, significantly contributing to improving the shelf life of culinary fried food products (Yaghmur et al. 2000).

4 Fatty Acid Composition and Acyl Lipid Profile of Argan Oil

Argan oil chemical composition has been investigated. Minute variations could exist depending on the tree genotype (El Adib et al. 2015). Early and pioneering chemical work on argan oil was done by Maurin (1992). Acylglycerols (glycerides), 95% of which being triglycerides, constitute 99% of argan oil. Unsaturated fatty

Table 16.1 Distribution of triglycerides in argan oil (%)

Major oleic acid-containing triglycerides		Minor oleic acid-containing triglycerides		Oleic acid-deprived triglycerides	
O, O, L	19.5	S, O, O	3.4	L, L, L	7.4
O, L, L	13.6	P, P, O	3.2	P, L, L	6.3
P, O, L	13.6	S, O, L	3.0	S, L, L	1.8
O, O, O	12.8	P, S, O	1.8	P, P, L	1.6
P, O, O	11.5	S, O, O	3.4	P, S, L	1.6

acids (UFAs) make 80% of the fatty acid fraction. Oleic (monounsaturated) and linoleic (diunsaturated) acids (46–48%, and 31–35%, respectively) are the two main UFAs. Linolenic acid, a triunsatutrated fatty acid, is only present as traces. Saturated fatty acids found in argan oil are palmitic (11–14%) and stearic acid (5–6%). Other fatty acids frequently found in the composition of most seed oils, although in low amounts like myristic, palmitoleic, arachidic, gadoleic, or behenic acids are here only as trace amount in argan oil.

Regarding the influence of the fruit shape, contrasting results were reported. Oil prepared from spherical and elongated fruits would present higher levels of linoleic or oleic acids, respectively than the average value (Belcadi-Haloui et al. 2008; Gharby et al. 2013a). However, because several factors can act on the fatty acid profile of seed oils, the statistical value of these results over several years and places must be confirmed.

The most abundant triglycerides encountered in argan oil constitutes 19.5% of all triglycerides (Table 16.1). It is composed of one linoleic acid and two oleic acid residues (O, O, L). The four others most abundant triglycerides of argan oil all include at least one oleic acid residue in their composition (Maurin et al. 1992). They are composed of three oleic acid residues (O, O, O), two linoleic and one oleic (O, L, L), one palmitic, one oleic, and one linoleic (P, O, L), two oleic and one palmitic (O, O, P). Minor triglycerides also incorporating an oleic acid residue include a saturated acid (palmitic or stearic) located at the *sn*1,3 positions of glycerol. In triglycerides incorporating a palmitic or stearic acid residue, these later generally esterify the glycerol extremities (*sn*1,3 positions) (Khallouki et al. 2008). Therefore, unsaturated fatty acids are likely to be the ones which are available for biosynthesis, being the major acids in the *sn*2 position (oleic, 67% and linoleic, 29%), which is well suitable in the diet.

5 Minor Bioactive Components in Argan Oil

In addition to glycerides, argan oil is composed of 1% of molecules grouped under the generic name of unsaponifiable-matters (Huyghebaert and Hendrickx 1974). This category includes carotenes (37%), triterpenic alcohols (20%), sterols (20%), phenols [polyphenols, tocopherols, coenzyme Q_{10}, and melatonin] (8%), xanthophylls (5%), wax, and traces of aroma compounds (Farines et al. 1984). Despite the

low content of unsaponifiable-matters in argan oil, the presence of highly pharmacologically active compounds like tocopherols or CoQ_{10} (Venegas et al. 2011) has been frequently used to justify, however without real demonstration, its nutritional value and health properties (Charrouf and Guillaume 1999; Khallouki et al. 2005, 2017b; Cabrera-Vique et al. 2012; Lopez et al. 2013).

5.1 Tocopherols

Tocopherols are of particular importance in argan oil owing to their vitamin E activity. Together with polyphenols and CoQ_{10}, tocopherols act as free radical scavengers, and contribute to the total strong antioxidant capacity of argan oil (Marfil et al. 2011). The good preservation properties of argan oil are also likely to result from the presence of these molecules that reduce argan oil oxidative degradation during storage (Chimi et al. 1994). Compared to most edible vegetable oils, argan oil is particularly rich in tocopherols (Madawala et al. 2012). Such specificity is incorporated into the Moroccan official norm, which specifies that argan oil must contain between 60 and 90 mg of tocopherols per 100 g (Snima 2003). In two independent studies, argan oil produced from kernels originating from pointed fruits was found particularly rich in γ-tocopherol (Belcadi-Haloui et al. 2008; Gharby et al. 2013a). If no significant influence of the extraction method on the tocopherol content has ever been observed (Marfil et al. 2011), storage conditions influence the tocopherol content and prolonged storage at 60 °C halves the total tocopherol content. γ-Tocopherol has been proposed as the most pharmacologically active molecule of the tocopherol group (Christen et al. 1997). Interestingly, γ-tocopherol is in the greatest proportion (above 70% of tocols) in argan oil whereas α-, β- and δ-tocopherols represent 10% or less (Cayuela et al. 2008). Comparing argan oil tocopherol distribution with that of eight other vegetable oils using chemometric methods, it was shown that argan oil is close to walnut oil, followed by sesame and linseed oils (Rueda et al. 2016).

5.2 Phenolics

Phenolic content in argan oil is low, in the range of 56 ppm (Charrouf and Guillaume 2007). Major identified phenolic compounds are oleuropein and caffeic, vanillic, ferulic, and syringic acids (Khallouki et al. 2003; Rojas et al. 2005). Identified polyphenols are ubiquitous molecules like resorcinol, epicatechin, and catechin (Charrouf and Guillaume 2007). Phenolic composition of argan kernels has also been investigated. Unroasted contained (−)-epicatechin and (+)-catechin as major polyphenols (0.6 and 0.4 mg/100 g, respectively). Roasted kernels contain a lower amount of polyphenols than unroasted kernels. This reduction was attributed to a temperature-assisted polyphenol oxidation during the heating step (El Monfalouti et al. 2012).

5.3 Phospholipids

Phospholipids are another class of molecules found as minor constituents in argan oil as a consequence of the absence of refining step. Their level is much higher in food grade oil (0.27 mg/100 mg oil) than in cosmetic oil (0.022 mg/100 mg oil). Because the former presents a better profile in terms of preservation capacity, phospholipids have been suggested to be important compounds for this superior oxidative stability, likely acting indirectly, by extending in time the full antioxidant activity of the tocopherol fraction (Gharby et al. 2012a, b).

5.4 Sterols

As phospholipids, sterols are present in very minute quantity in argan oil. Identified sterols are campesterol, stigmasterol, sitosterol, stigmast-7-en-3β-ol, schottenol, and spinasterol (Farines et al. 1981; Madawala et al. 2012). The implication of the sterol fraction in the physiological properties of argan oil is often suggested (Khallouki et al. 2003). However, the ubiquity of the sterols found in argan oil within the vegetal kingdom is not a favorable factor to support a real physiological interest in argan oil unless it is the consequence of a synergistic effect that still needs to be identified.

5.5 Wax

The wax content -a mixture of fatty acids and alcohols- in argan oil ranges between 7 and 95 mg/kg. The average value is 26.4 mg/kg (Cabrera-Vique et al. 2012). Wax does not alter argan oil nutritional properties but they are disfavorably perceived by consumers. Because argan oil is generally filtered after being pressed, wax content fluctuates as a function of the filtration efficiency; it could also be influenced by the extraction method (Cabrera-Vique et al. 2012). Wax formation generally increases above 20 °C, so for a better preservation, storage of argan oil should be performed in the dark, at temperatures below 20 °C and in an oxygen-poor environment (Harhar et al. 2010a). It should be noted that argan oil filtration could lead to a decrease of its oxidative stability (Kartah et al. 2015). Consequently, exessive filtration is not recommended.

5.6 Miscellaneous Organic Compounds

Large variations in CoQ_{10} and melatonin contents have been observed for traditionally prepared argan oil, whereas press-extracted argan oil presents a more stable content. The concentration of CoQ_{10} ranges between 10 and 30 mg/kg oil. The average content of melatonin is 60.5 ng/kg of oil (Venegas et al. 2011).

5.7 Mineral Content

Mineral content in plants or plant-extracted derivatives depends on a large number of factors among which are the growing site conditions, the soil chemical characteristics, anthropogenic pollution induced by industries or the use of fertilizers, and the genotype of the species (Cataldo and Wildung 1978). Because the argan tree is spontaneously growing and no industry is present in the argan forest, the influence of anthropogenic factors on the element content of argan oil can be discarded. Therefore, the element content in argan oil is considered primarily strictly dependent on the argan tree genotype and its growing area. Consequently, argan oil elements content has recently received a lot of attention. This sudden interest is stimulated by the possible implication of some elements in deleterious oxidative reactions as well as their possible use to detect adulteration by precisely identifying the geographical origin of the analyzed samples. Using oil samples prepared in four of the main area, of production of argan oil (Agadir, Ait Baha, Essaouira, and Tarroudant) little correlation has been found between soil element composition and argan oil element content (Mohammed et al. 2013a) suggesting that the argan forest is globally a homogenous area in terms of soil chemical characteristics influencing the metal content. Metal content in argan oil could also be influenced by its preparative process. Variation in element content in press-extracted or traditionally prepared argan oil has also been shown to not be significantly different (Ennoukh et al. 2017) or eventually semi-mechanized argan oil presented a slightly lower element content (Marfil et al. 2008). Element content in argan oil has been found to be remarkably stable over time suggesting its controlled by the tree genotype. Consequently, precise quantitive profile determination of eight elements (Cd, Cr, Cu, Zn, Fe, K, Mg, and Ca) has been proposed as sufficient to ascertain argan oil authenticity (Mohammed et al. 2013a, b).

6 Composition of the Argan Fruit Essential Oil

Seven volatile substances were primarily isolated from the pulp of the argan fruit. Resorcinol was found to represent 73.5% of the isolated products followed by *E*- and *Z*-but-2-enol (12.5% and 6%, respectively). Minor products include 3-methylbutyric acid, and n-octan (Tahrouch et al. 1998). The composition of the essential oil of *A. spinosa* fruit fresh and dried pulp has been investigated. Extraction of essential oils was performed by hydro-/steam-distillation and microwave-assisted extraction. In both cases, camphor was found to be the major (i.e.: 34%) component of the essential oil. In fresh pulp, 1,8-cineole (eucalyptol), endo-borneol, and cyclohex-3-en-ol were found to be in high concentration (16%, 11.8%, and 11.1%, respectively). In the dried pulp, cyclohex-2-ene-1-one was found to be the second most abundant compound (12.6%), and 1,8-cineole (7.8%) the third. Essential oil of argan dried pulp contains more oxidized compounds (Harhar et al. 2010a). In

addition, essential oil of *A. spinosa* leaf extracted by steam distillation has evidenced that essential oil is mainly composed of 1,10 di-epi-cubenol (El Kabouss et al. 2002). A recent study in which essential oil was obtained by microwaved or supercritical fluid extraction indicated that cubenol derivatives are the principal components of argan leaf essential oil (El Amrani et al. 2015).

7 Contribution of Bioactive Compounds in Argan Oil to Organoleptic Properties

Food-grade argan oil is a tasty vegetable oil that is not refined. Its organoleptic properties are consequently only set by the quality of the raw material and the production process (Matthäus et al. 2010). Both parameters have undergone intensive studies that have ultimately led to drafting mandatory to follow specifications included in the Geographical Indication in 2010.

Kernel roasting not only removes argan oil bitterness, but it also induces the formation of odorants since cosmetic and dietary oil present a different content. Therefore, edible argan oil contains in very minute quantity some roasting-induced odorants that are essential for the specific flavor of edible argan oil. Identified odorants belong to various chemical groups as alcohols, aldehyde/ketones, ester/lactones, pyridines, pyrazines, sulfurs, terpenes, and ethers (Charrouf et al. 2006). Odorant composition in high-quality argan oil and that prepared from goat-regurgigated kernels presents some differences, confirming the traditional claim that regurgigation gives a special flavor to the resulting oil (Charrouf et al. 2006). A kernel roasting time of 25 min at 110 °C has been found to be optimum for aroma compound formation in dietary oil (El Monfalouti et al. 2013b).

8 Preservation of Argan Oil

As indicated for its organoleptic properties, argan oil shelf life and stability upon storage and use also depend on that of the kernels and on the production process. The quality of argan kernels can be assessed by Vis/Nir measurements (Guinda et al. 2015). However, implementation of such a method is today impossible in woman cooperatives. Fortunately, a combination of studies has allowed the establishment of semi-empiric rules.

For example, harvest date of argan fruit is of little influences on the fatty acid composition of argan oil. However, harvest date influences the minor component content of argan oil, especially its tocopherol and phospholipid content, which is known to be positively linked to the oil oxidative stability. Tocopherol and phospholipid are at a maximal concentration in argan oil produced from kernels collected from fully ripe fruit, generally harvested in July (Harhar et al. 2014). This confirms

the ancestral practice according to which fruit must be collected when fallen on the ground, and thus supposedly fully ripe.

Next, fruit is sun-dried to facilitate its peeling, it is subjected to a daily cycle alternating high and low temperatures and exposure to sun light. Therefore, oxidative events that could occur during this period of time must be minimized. A detailed analysis of several oil physicochemical properties has shown that a sun-drying period of 10–14 days is optimum for preparing high-quality oil. Such a time is sufficiently long to allow an easy depulping but short enough to prevent oxidative degradation of argan oil constituents (Harhar et al. 2010b).

Depending on the women cooperative practices, fruit may be stored as raw and depulped later when necessary, or it may be depulped immediately after sun-drying thus enabling to store only clean kernels. Under carefully controlled conditions preventing mold formation, both methods lead to an oil of similar quality (Harhar et al. 2015).

After nut breaking large quantities of argan, kernels are obtained. In some places, those kernels are sometimes stored prior to extraction, for example for extending the cooperative activity out of the harvest time, or for shipping oversea when the oil is to be extracted in a foreign country. Consequently, the question of whether it was better to store argan nuts or argan kernels has also been addressed. If kernel storage is performed at 4 °C, kernel quality is preserved for 1 year, but at room temperature, storage should not be longer than 10 months (Harhar et al. 2010a).

Roasting is the next step of the preparative process. Significant reduction of tocopherols during seed roasting has been previously reported (Gemrot et al. 2006). Therefore, care must be taken to avoid tocopherol degradation during argan kernel roasting. Using a large array of quality attributes like taste, residual moisture, color, peroxide value, peroxidative value, fatty acid composition, tocopherol content, benzopyrene content, and oxidative stability, it was shown that a roasting time of 25–30 min at 110 °C is suitable for optimal taste and preservation (Harhar et al. 2011; El Monfalouti et al. 2013b). The influence of much longer and warmer roasting time has also been studied. Such conditions favor negative attributes as dark coloration (Demnati et al. 2018).

For many years, argan oil preservation capacity has been considered to be very low. This was a consequence of its traditional preparative process that required the use of water, often of poor bacteriological quality. In addition, unsatisfactory storage conditions favored accelerated fatty acid oxidation. However, argan oil prepared by cold pressing kernels suitably stored according to the Geographic Indication recommendations, displays an excellent preservation capacity that is even better than that of ordinary olive oil (Gharby et al. 2012a, b). Interestingly, edible and cosmetic argan oils present different preservation properties, even though product specifications of the cosmetic or food industry are different, and measured through different laboratory standard.

Cosmetic argan oil is more sensitive to oxidation than edible argan oil (Zaanoun et al. 2014). Nevertheless, to appreciate the oxidative degree, metrics used in the cosmetic industry are acid and peroxide values and the oil specific absorbance. Stored at 25 °C and protected from sunlight, cosmetic argan oil quality is still

satisfactory after 12 months according to the official Moroccan norm, but storage should not exceed 6 months to fulfill industrial standards (Gharby et al. 2014).

Regarding edible argan oil, extensive studies have led to identifying odor and taste (Matthäus 2013) as the most important quality parameters to satisfy consumers. Therefore, quality argan oil must be free from foreign and rancid odor and taste. Sensory attributes typical for high-quality argan oil are "nutty" and "roasty". These attributes result from the fruit itself from the roasting process, respectively. Negative attributes are rancid, Roquefort-cheese, bitter, wood-like, burnt, musty, yeast-like, fusty. Rancid can come from improper storage. Burnt comes from an excess of roasting. Roquefort-cheese and fusty come principally from goat-peeled fruit. Negative attributes are very rarely mentioned for cold pressed argan oil. However, they appear more rapidly for traditionally prepared oil confirming its low preservation capacity. Except for the burning taste that results from a roasting performed at an elevated temperature, or prolonged for a too long time, most of the negative attributes of argan oil come from an excessive and uncontrolled oxidative process. Therefore, tocopherols, polyphenols and molecules presenting an anti-oxidative activity actively participate to the taste of edible argan oil by slowing the formation of products bringing the rancid attribute (Matthäus et al. 2010).

Processing also influences the preservation properties of edible argan oil. Mild roasting enhances the preservation properties of argan oil since food grade argan oil is significantly less susceptible to oxidation and to develop negative attributes during storage than cosmetic argan oil (Matthäus et al. 2010). Furthermore, in cold-pressed extracted edible oil, no negative organoleptic attributes were detected even after 20 weeks of storage whereas for traditionally extracted edible oil, negative attributes were reported after 12 weeks of storage at 20 °C. In cosmetic oil the development of the unpleasant fusty and Roquefort cheese attributes was quick, covering the perception of the nutty attribute that was only slightly perceivable at the beginning of the experiment (Matthäus et al. 2010).

Argan oil preservation properties are satisfactory as long as the oil is not stored at a temperature above 64 °C (Alaoui et al. 2001). But argan oil is also sensitive to UV irradiation and the combined effect of elevated temperature and irradiation results in a shorter shelf life (Kondratowicz-Pietruszka and Ostasz 2017). However, after 2 years of storage at a maximum temperature of 25 °C, cold pressed edible argan oil protected from sunlight still displays physicochemical and preservation properties similar to those of freshly prepared argan oil. The shelf life of edible argan oil has consequently been set at 2 years when protected from sunlight (Gharby et al. 2011). Because the exact date of preparation of argan oil might be difficult to know, a storage time of 1 year at home is recommended (Matthäus and Brühl 2015).

9 Health Promoting Traits of Argan Oil

Edible argan oil worldwide success is undoubtedly due to its special taste but its pharmacological properties are also essential to explain its appeal to consumers. In parallel, cosmetic argan oil presents important to skin and hair improvement

properties. Traditionally claimed properties of edible and cosmetic argan oil and several reviews have been devoted to this topic (Cherki et al. 2006; Charrouf and Guillaume 2008b, 2010, 2012; Rammal et al. 2009; El Monfalouti et al. 2010; Guillaume and Charrouf 2011a, b, 2013; Cabrera-Vique et al. 2012; El Abbassi et al. 2014).

In addition to the traditionally claimed properties of argan oil, several pharmacological properties are attributed to argan oil without bringing a proof of activity. Alleged properties simply come from the presence of above-detailed components found in its chemical composition (Khallouki et al. 2003). This is particularly regretful since the presence of a bioactive compound within a natural product should not be considered as a proof of its pharmacological properties. Such affirmation could result in undue expectations from argan oil consumers and lead to disappointment. Indeed, criteria for absorption, distribution, and toxicity must be considered prior to evoking a pharmacological activity.

Using an in vitro digestion method, the phenolic fraction of argan oil, which is often presented as the probable promotor of most of argan oil properties, has recently been shown to be strongly affected by the digestive process (Rueda et al. 2017). Even though, polyphenols present in argan oil would possess enough bioavailability to induce a high antioxidant potential and health-giving in vivo properties (Seiquer et al. 2015), a direct correlation between the presence of some amount of phenols and a pharmacological activity is impossible to draw, as is a correlation between the quantity of ingested phenols and the intensity of the pharmacological activity.

In addition, some pharmacological properties attributed to this oil have been evidenced only on animals and their relevance in human has never been clearly demonstrated, yet. Consequently, in this chapter, only well-established effects observed on humans or human cells are detailed.

9.1 Hypolipemiant and Antioxidant Effects and the Cardiovascular Risk

Hypolipemiant properties of argan oil were evidenced in a study including a group composed of 96 healthy subjects, 62 of which being regular consumers of argan oil on a daily basis of 15 g (Drissi et al. 2003). Careful analysis of fasting plasma lipids sampled after overnight dietary restriction excepting water, antioxidant vitamins and LDL oxidation susceptibility indicated that subjects consuming argan oil on a regular basis had significantly reduced levels of plasma LDL cholesterol (12.7%) and lipoprotein-a (25.3%), compared with the non-consumer group. Argan oil consumers also presented a significantly lower level of plasma lipoperoxides (58.3%). This important study brought the first evidence that regular consumption of argan oil induces a lowering of LDL cholesterol and has potent antioxidant properties in human. Hence, it scientifically confirmed the traditionally claimed preventive properties of argan oil on the cardiovascular risk. Another nutritional study performed on

60 volunteers reinforced these results and additionally demonstrated that consuming 25 g/day of argan oil for 3 weeks leads to a lower plasma triglyceride level in men (Derouiche et al. 2005). Additional cohort studies carried out on human consuming either 15 g/day of argan oil for 4 weeks (Sour et al. 2012), 22.5 g/day for 3 weeks (Haimeur et al. 2013), or 27 g/day for 4 weeks, confirmed the above findings (Eljaoudi et al. 2015). So, it is now accepted that argan oil supplementation reduces total cholesterol, low-density lipoprotein cholesterol, and triglycerides and increases high-density lipoprotein cholesterol levels in human (Ursoniu et al. 2017).

Concomitantly to observe the hypolipemiant and antioxidant properties of argan oil, Drissi et al. (2003) noticed a strong positive correlation between increasing phenolic-extract, sterol, and tocopherol concentrations and the LDL-Lag phase. The importance of the phenolic-extract of argan oil on the prevention of cardiovascular diseases was confirmed by observing that this extract inhibits human low-density lipoprotein oxidation and enhances cholesterol efflux from human THP-1 macrophages (Berrougui et al. 2006).

9.2 Antiproliferative Effect

The polyphenol-sterol fraction of argan oil was also shown to induce an antiproliferative effect on human prostate cancer cell lines (Bennani et al. 2007). An interventional study performed by comparing 2 groups of 30 men fed either with 25 mL/day of argan oil or with 25 mL/day of olive oil for 3 weeks. Biochemical measurements suggested that the antiatherogenic effect and antioxidant status of argan oil in human could result from improving the activity of a group of enzymes, namely paraoxonases, involved in anti-inflammatory, anti-oxidative, anti-atherogenic, anti-diabetic properties (Cherki et al. 2005). Modification of the peroxisomal acyl-CoA oxidase type1 (ACOX1) activity has also recently been proposed to be a key protein involved in some of argan therapeutic properties (Vamecq et al. 2018). However, a study with a cohort of 125 healthy elderly subjets (Ostan et al. 2016) has failed to evidence any improvement on inflammation when argan oil (25 mL/day for 8 weeks) was associated with RISTOMED diet, a diet built to reach the recommended daily requirement of nutrients, vitamins and minerals complying with different cultural patterns (Valentini et al. 2014).

The insulin-sensitizing and anti-proliferative effects of argan oil unsaponifiable fraction were reported using the human HT-1080 fibrosarcoma cell line. After confirming that argan oil unsaponifiable matters does not exhibit any cytotoxic activity toward HTC cells, it was shown that, at a concentration as low as 25 μg mL^{-1}, argan oil unsaponifiable reduces specifically the ability of extracellular signal-regulated kinases 1 and 2 (ERK1/2) to respond to increasing doses of insulin. Interestingly, the protein kinase B (Akt) response, and the insulin-induced activation of mitogen-activated protein kinase 1 and 2 (MEK1/2) remained undisturbed. The different action of argan oil unsaponifiable on MEK1/2 and ERK1/2 activity in response to insulin in HTC cells means that its strong anti-proliferative activity could be

mediated by the interruption of signaling cascades at the MEK1/2–ERK1/2 interface (Samane et al. 2006).

The antiproliferative effect of argan oil tocopherols was evaluated using the two classical human prostatic cell lines (DU145 and PC3) and the androgen-sensitive LNCaP cell line (Drissi et al. 2006). The best antiproliferative effect of tocopherols was observed with DU145 and LNCaP cell lines with tocopherol concentration inhibiting growth by 50% compared to the controls of 28 μg/mL and 32 μg/mL, respectively. Due to the promising therapeutic activity evidenced for argan oil, its use to prepare nanoemulsions as vehicles possessing anticancer activity has been investigated. If the concept has been validated, its application in human medicine still needs to be established (Jordan et al. 2012).

9.3 Hypoglycemiant Effect

Not surprisingly, considering the above-mentioned findings, it has also been shown that consumption of 25 mL/day of argan oil for 3 weeks may have an antiatherogenic effect by improving lipids, and the susceptibility of LDL to oxidation in type 2 diabetes patients with dyslipidemia (Oud Mohamedou et al. 2011). Argan oil consumption is, therefore, recommended in the nutritional management of type 2 diabetes. Several studies performed on rats support this recommendation.

9.4 Endocrinal Effect

Some studies on human strongly support the idea that, in addition to cardiovascular protection, argan oil possesses endocrinological properties. Hence, a study performed on 60 young men, one half of each receiving an argan oil diet, and the other an olive oil diet, has shown that argan oil consumption at a 25 mL/day dose for 3 weeks induces a positive action on the androgen hormonal profile of men. The effect, which is similar to that of olive oil, could result from an activation of the hypothalamo-pituitary-testicular axis and/or an induction of steroidogenic proteins by argan oil tocopherols (Derouiche et al. 2013). An independent study performed on 151 postmenopausal women has evidenced that an argan oil supplement at a daily dose of 25 mL for 8 weeks reduces postmenopausal symptoms and significantly increases vitamin E concentration (El Monfalouti et al. 2013a).

9.5 Pain Relief and Burn Healing Effects

A randomized controlled clinical trial performed on 100 patients presenting knee osteoarthritis has permitted to evidence the benefits of argan oil consumption at a dose of 30 mL/day for 8 weeks on the relieve of knee osteoarthritis associated pain

(Essouiri et al. 2017). Pain reduction was evaluated by use of the visual analog scale for pain, the determination of the walking perimeter, and the WOMAC index (Bellamy and Buchanan 1986) and Lequesne indexes (Lequesne et al. 1987). Once again, tocopherols have been supposed to actively participate in this improvement (Essouiri et al. 2017). Topical application of argan oil is traditionally recommended to cure skin burns (Charrouf and Guillaume 1999). Evidence that argan oil would accelerate healing of burn injuries have only been observed in rats, so far (Avsar et al. 2016).

9.6 Animal Health

Out of the human therapeutic domain, argan oil can also be used in veterinary medicine. Antibacterial activity against fish pathologic bacteria has been investigated. Argan oil exhibited a MIC value of 62.5 µL/mL against *Yersinia ruckeri,* the causative agent of enteric redmouth disease in fish (Kumar et al. 2015). Against other fish pathologic agents, a MIC value of 125 µL/mL was calculated against *Vibrio anguillarum*, *Aeromonas hydrophila* and *Citrobacter freundii*. Against *Edwardsiella tarda* and *Lactococcus garvieae*, the MIC was 250 µL/mL (Öntas et al. 2016). Food-grade argan oil, at a concentration of 1% and 2%, increases the survival rate of Nile tilapia (*Oreochromis niloticus* L.) against *Lactococcus garvieae*. No associated negative effect on fish growth or feed efficiency was found (Baba et al. 2017). Cosmetic argan oil can be used to cure lice infestation in cats, when mixed with apple cider has been shown by Hassan et al. (2017).

9.7 Dermocosmetic Properties of Argan Oil

The role of argan oil in dermocosmetology is also well documented (Guillaume and Charrouf 2011b; Suggs et al. 2014; Vaughn et al. 2018; Lin et al. 2018). In just a few years, cosmetic argan oil has managed to occupy an important place in the list of raw materials used in cosmetics and its rapid rise has even made the argan tree often nicknamed "*Argania cosmetosa*" (Guillaume and Charrouf 2011b). Cosmetic grade argan oil (INCI name: *Argania spinosa* kernel oil, CAS: 223747-87-3) currently prepared by cold-pressing unroasted kernels may also be obtained using supercritical CO_2 (45 °C, the pressure of 400 bar) as a green solvent, without alteration of its quality parameters (Taribak et al. 2013). Cosmetic argan oil can be found in the composition of commercial shampoos, hair conditioners, hand-wash lotion, or repair serum. Cosmetic argan oil safety has been assessed (Charrouf and Guillaume 2018) and validated by the Cosmetic Ingredient Review Expert Panel (Burnett et al. 2017). But a few cases of contact allergy have nevertheless been reported (Astier et al. 2010; Foti et al. 2014; Veraldi et al. 2016; Barrientos et al. 2014; Lauriola and Corazza 2016). It should also be mentioned that one case of hypersensitivity

pneumonitis has been reported in cosmetic-industry workers exposed to non-sterile argan cake powder (Paris et al. 2015).

Traditionally, the dermatological use of argan oil was mainly for curing skin pimples, juvenile acne, and chicken pox pustules, for reducing dry skin matters and slow down the appearance of wrinkles, brittle fingernails, but also more specifically in the biomedical field for treating psoriasis, eczema, joint pain, skin inflammation, and scabies, and for healing burns and wounds (Charrouf and Guillaume 1999; Lin et al. 2018). It was also said to have a favorable impact on hair loss and dry hair (Karabacak and Dogan 2014). Recent scientific findings confirm some traditional uses. Skin-protecting properties of argan likely come from its main component palmitic and linoleic acids that have emollient and hydration properties for skin (Chelaru et al. 2016). However, recent studies that have confirmed the activity in the dermocosmetic field have focused on the oil antioxidant, hydration, antiaging, and protection properties on the skin.

The efficacy to reduce the greasiness of oily facial skin has been demonstrated with 20 healthy volunteers who applied argan oil on their forehead and cheeks twice daily for a period of 4 weeks (Dobrev 2007). Skin elasticity resulting from the hydration properties of dietary argan oil has also been shown to occur after repeated applications of cosmetic argan oil in the left volar forearm during 60 days (Boucetta et al. 2014). After 2 months of once a day topical application, argan oil also improved skin moisture (Boucetta et al. 2013). On a panel of 60 postmenopausal women, it was shown that argan oil consumption (25 mL/day for 8 weeks) improve skin hydration due to a reduction of transepidermal water loss and an increase water content of the epidermis (Boucetta et al. 2014).

These hydration properties are also expressed by nanostructured lipid carriers (NLCs) that use argan oil as the liquid lipid phase. Such nanostructures present the advantage to synergistically combine the NLC occlusion and argan oil hydration properties (Tichota et al. 2014). The skin-favorable properties in terms of dermophilicity and moisturizing power have made of argan oil a key component of some liposomes in facilitating the drug accumulation in the dermis (Manca et al. 2016). Accordingly, argan oil-nanoparticles designed for local and cosmetic application containing the anti-inflammatory drugs indomethacin (Badri et al. 2015) or diclofenac (Lococco et al. 2012) have been successfully prepared. Microbiological spoilage of argan oil-containing nanostructures can be performed with 10% w/w propylene glycol or 5% pentylene glycol, these two chemicals not altering nanoemulsion stability (particle size or in Zeta potential) for 120 days (Hommoss 2011).

Cosmetic argan oil is a known depigmenting agent for murine cutaneous melanoma cells that could be used against hyperpigmentation troubles. Indeed, it possesses a skin depigmenting effect through the inhibition of melanin biosynthesis, likely via tocopherols or a synergistic effect involving several components. Argan oil causes melanogenesis associated transcription factor (MITF) phosphorylation which subsequently inhibited the transcription of tyrosinase and DOPAchrome tautomerase, two melanogenic enzymes (Villareal et al. 2013). The regulation of melanogenesis by argan oil in uveal melanoma cells would follow a slightly different pathway, likely acting on the ERK1/2 and Akt pathways (Caporarello et al. 2017).

The effect of argan oil on hair is still poorly documented; however, argan oil is frequently used as a reference for testing hair breaking (Del Campo et al. 2017).

10 Miscellaneous Properties of Argan Oils

Observed anti-corrosion properties of argan oil have been attributed to its high antioxidant content (Afia et al. 2011, 2014). However, DFT calculations have demonstrated that under chosenstandard testing conditions, linoleic acid is more stable than oleic acid and hence is likely to be responsible for the corrosion inhibition efficiency in acidic medium (Gece 2017). The continuously increasing demand for energy and the decreasing petroleum resources has led to the search for renewable and sustainable fuels. However, because of its high market price, argan oil is unlikely to be developed as anti-corrosive agent or as fuel (Belgharza et al. 2014). By-products generated during argan oil preparation are much more actively investigated for industrial purposes.

It has also been shown that addition of argan oil to conventional extenders for cryopreservation as tris egg yolk and skim milk may improve the quality of ram semen during liquid storage at different temperatures. This effect is likely due to the argan oil high antioxidant content (Allai et al. 2015). Finally, edible argan oil supplementation with molasses has been shown to enhance fermentative performance and antioxidant defenses of active dry wine yeast possibly by prevention of membrane damage (Gemero-Sandemetrio et al. 2015).

11 Adulteration and Authenticity

Due to its high market price, adulteration of argan oil has become an important issue that needs to be properly considered (Seidemann 1998). Indeed, cases of adulterated argan oil are frequently reported (Momchilova et al. 2014).

Adulteration can result from at least three fraudulent processes. Indeed, adulteration can result from the marketing of a product coming from a country devoid of traditional culture towards this product. Adulteration can also be the blending of something impure with something authentic. It can also result from the mixing of an inferior article with a superior one of the same kind. Both aspects have to be considered in the case of argan oil.

Geographic Indication that certifies the marketing of “endemic argan oil of Morocco”, is aimed at protecting argan oil authenticity by preventing the marketing of argan oil prepared from the fruit of trees grown out of Morocco. This protection is particularly important because several countries have considered, or are currently considering, the culture of argan trees to produce argan oil (Nerd et al. 1994, 1998; Kechairi et al. 2018; Falasca et al. 2018). Identifying the geographical origin is essential for consumers since, for example, argan oils from Morocco and from

Israel present a different content in mono-, di-glycerides, and phospholipids (Yaghmur et al. 1999).

Adulteration of argan oil can also mean marketing a cheap seed oil -like sunflower oil- or a mixture of a cheap seed oil and argan oil while presenting it as high-quality argan oil, or can also mean marketing low-quality argan oil while pretending it is high-quality argan oil. This second aspect is much more complex to tackle particularly considering that, traditional argan oil cannot be removed from the local Moroccan market. To detect adulterations, the Moroccan authorities have edited a norm based on physicochemical parameters (Snima 2003). Selected values do not lead to the market exclusion of traditional argan oil as long as its presents satisfactory quality attributes. In other words, the official norm only certifies that the oil is argan oil, but not cold press-extracted argan oil. Only the respect of Geographic Indication rules certifies the compliance to the strict semi-mechanized preparative process.

Adulteration of argan oil by the addition of another oil can be detected by looking for compounds present in very minute amount of argan oil whereas these compounds are present in high quantity in the contaminating product. The official norm uses campesterol as a chemical marker, a sterol found in very minute quantity in argan oil although relatively abundant in most vegetable oils. Indeed, argan oil contains five sterols: campesterol (0.4%), spinasterol (34–42%), stigma-8,22-dien-3-ol (4–7%), schottenol (42–49%), and stigmasta-7, 24-dien-3-ol (2–7%) (Hilali et al. 2005). By quantitative GC-analysis, it was shown that campesterol quantification enables certifying a 95% purity of argan oil (Guillaume and Charrouf 2007). Other methods have been suggested and other minor compounds have been proposed as markers: stigmastadiene, kaurene and pheophytin-a. A similar detection limit of 5% can be reached (Ourrach et al. 2012).

Triacylglycerol profile analysis, by using HPLC-evaporative light scattering detection, has also been proposed as authentification tool, owing to an improved detection limit (Salghi et al. 2014). Analysis of triacylglycerols by UPHPLC or HPTLC based methods has also been developed for this purpose (Pagliuca et al. 2018).

Certification of the authenticity of edible oils being possible to be based on the singularity of chemical element profile, this method has been successfully applied to ascertain the authenticity of argan oil. The element composition can be determined using ICP-OES (Gonzálvez et al. 2010; Mohammed et al. 2013a). Discriminant analysis can be used to evidence the mixture of argan oil with other vegetable oils (Gonzalvez et al. 2010)

A method, based on the formation of gold nanoparticles and spectrophotometric analysis, has also been proposed to determine total phenolic acids in genuine argan oil samples, and ferulic acid, the main phenolic acid in virgin argan oil, was also used as an adulteration marker (Zougagh et al. 2011). Another proposed method combines midinfrared spectroscopy and chemometrics (Oussama et al. 2012).

Specific methods have also been designed. Electronic nose (e-nose) are systems equipped with an array of chemical sensors. Such instruments can be used to detect oil adulteration (Majchrzak et al. 2018). Accordingly, adulteration of argan oil with sunflower oil by using the combination of a voltammetric e-tongue and an e-nose

based has been successfully applied (Bougrini et al. 2014). Adulteration with olive oil can be detected by fluorescence spectroscopy (Addou et al. 2016). Synchronous fluorescence spectroscopy (SFS) has been used to detect adulteration of argan oil by corn oil and then the determination of the corn oil content. Four percent is deemed the lowest concentration of adulteration detected (Stokes et al. 2018).

Even though the argan forest is a homogenous area and Moroccan argan oil is presented as a single product, a method based on FTIR and chemometric tools has been suggested recently to identify the geographic origin of oil samples within the argan forest, enabling to discriminate samples prepared from fruit harvested in Essaouira, Agadir, Tiznit, Taroudant, or Ait Baha areas (Kharbach et al. 2017).

12 Current and Potential Uses of Co-products Generated During Argan Oil Production

The food industry is known to generate a large amount of wastes or co-products (Helkar et al. 2016) which are now being increasingly evaluated as a source of nutraceuticals, in addition to traditional uses. The same applies to the argan oil production chain, which makes available, starting from the upstream, dry-pulp, shell, and press-cake.

12.1 Argan Pulp

The current processing of argan oil generates large quantities of pulp estimated to reach 44,500 tons per year (Guinda et al. 2011), and which is usually used as food for cattle. However, new outputs actively prospect. The first chemical analysis of argan pulp was reported in 1987 by Fellat-Zarrouk et al. (1987).

The pulp is the main constituent of the fresh fruit (48–59%), its content in water and volatiles is close to 75% (Pioch et al. 2011). Apart of hemicellulose and cellulose polysaccharides (Ray et al. 2004; Habibi and Vignon 2005), which are the cell-wall polymeric components, main pulp constituents are: soluble sugars, non polar fractions like lipids and including wax from fruit skin, polyisoprene, and miscellaneous secondary metabolites (Kenny and De Zborowski 2007; Chernane et al. 1999; Pioch et al. 2011, 2015a, b; Charrouf et al. 1991) whose content is influenced by fruit morphotype and growing location (Zhar et al. 2016).

The total water-soluble sugars amount 16–24% (relative to pulp dry weight) depending on fruit shape (Pioch et al. 2011). This content showed a fourfold increase during the last month of ripening, just before fruit fall to the ground. Proteoglycans, whose properties and industrial interest need to be addressed, were also isolated from argan pulp (Aboughe-Angone et al. 2008).

The content of neutral lipids in pulp varies from 4.8% to 5.7% (dry weight), and the most abundant fatty acids identified were linoleic (28–31%), palmitic (24–26%) and linolenic (12–13%) acids, oleic acid amounting only 13–15% (Pioch al. 2011).

Polyisoprene, another pulp component, has been investigated in details (Palu et al. 2011). This elastomer, which amounts up to 3.6% in the dry pulp is a mixture of *trans* and *cis*-1,4 polyisoprene, could find uses as chewing-gum or sealing. Both polyisoprene and lipids were at a maximum concentration just before fruit fall to the ground with a total of 11.9% and 7.4%, respectively. This suggests that to value these compounds, fruit should be harvested on the tree, which is not the current practice.

A total polyphenolic content of 1.5% (dry weight) in pulp has been measured (Khallouki et al. 2015). Its composition includes, in addition to catechin, epicatechin, and rutin that were identified early (Chernane et al. 1999), 13 other phenolics that were more recently identified by LC-tandem MS/MS analysis (Charrouf et al. 2010). In a quantitative study, isoquercitrin, hyperoside, and rutin were identified as the major phenolic compounds with a content of 28.4, 21.1, and 9.8 mg/100 g; respectively (El Monfalouti et al. 2012). Using a HPLC-ESI based method, 32 compounds including catechins (39%), flavonoids (28%), procyanidins (26%), and phenolic acids (7%) were identified in argan fruit pulp, together with aminophenols named arganimide A and argaminolics A–C (Klika et al. 2014, 2015; Khallouki et al. 2015).

A large number of parameters including growing conditions, geographical origin, ripening process, ripening level, storage conditions, and tree genotype can influence phenolic content in argan tree parts (Chernane et al. 1999; El Monfalouti et al. 2012; Khallouki et al. 2017b). Antioxidant properties of these phenolics found in argan pulp could be useful in the colorant industry (Chemchame et al. 2015).

Ripening stage and environmental conditions influence the profile and concentration of pentacyclic triterpenes, another class of compounds identified inargan fruit pulp (Guinda et al. 2011). High levels of oleanolic acid were found in unripe fruit pulp originating from the coastal region whereas those from the inland presented high levels of betulinic acid, in addition to ursolic acid present in both cases.

Other compounds of potentially high economic value were found in solvent extracts of argan fruit pulp (Pioch et al. 2015a, b). In addition to pyrocathecol and amyrin, the CH_2Cl_2 extract from the defatted pulp of unripe fruits was found rich in lupeol (48%).

Hence, argan fruit pulp could be better valorized and bring a substantial contribution to farmer's income. Trials at pilot scale have shown that key steps (drying, mechanical depulping and fractionation) can be carried out efficiently with simple hardware (Pioch et al. 2011, 2015a, b). Interestingly, after extraction of the marketable products, the bagasse, rich in cellulose, sugars, and proteins, would remain available as feed for cattle, hence preserving the traditional pulp use.

The argan nutshell is the next by-product generated during argan oil preparation. It is estimated that 60,000 of tons of argan nutshell are annually discarded in Morocco (Tatane et al. 2018). Traditionally this wood-like material has been burnt, including for roasting kernel. Nowadays, several outputs are identified to use argan

shell as a cheap, low-density, and eco-friendly material, either simply under a powder form or after pyrolysis. In the dermocosmetic domain, after being finely crushed, argan shell powder can be introduced in peel-off mask formulations and used as exfoliating cleanser owing to its gentle abrasive properties. Argan shell has also been evaluated as a source of saponins, a group of natural products presenting a large variety of pharmacological properties (Güçlü-Üstündag and Mazza 2007) but its low content (0.01%) doe not pay in favor of uses in the food, dermocosmetic, pharmaceutical industry (Alaoui et al. 2002).

In the agricultural domain argan shell powder can also be used to prepare valuable biochar because of is content in major nutrients (Bouqbis et al. 2016, 2017). The use of argan shell to prepare ultra microporous active carbons useful for CO_2 capture is presently actively investigated (Boujibar et al. 2018); but it could simply be used for any other purposes (i) on the market activated carbon (Ennaciri et al. 2014), and (ii) as natural bio-filler in high-density polyethylene composites of which it decreases the decomposition temperature (Essabir et al. 2015, 2016). Carbonization of argan shell can also yield hard carbon that is an attractive material for negative electrodes in sodium-ion batteries (Dahbi et al. 2017). Improvement of earthen construction materials can also be successfully realized using argan nutshell powder whose addition to compressed earth blocks considerably leads to an increase of the resulting blocks physical and thermal properties, and an increase of their mechanical strength (Tatane et al. 2018).

The residue remaining after argan oil extraction is called the "press-cake". Traditionally-obtained, press cake was used as cattle feed (Rojas et al. 2005). However, the shift to screw-press instead of traditional extraction, resulted in huge amounts of press-cake, being now an abundant biomass whose optimized use is currently investigated.

Chemical composition analysis of argan press cake has confirmed the validity of its traditional use as animal food since it possesses a high-energy content due to the presence of glucides and proteins (Rojas et al. 2005). The bitterness of argan press cake results from the presence of large amounts of saponins, whose main component structure has been investigated and elucidated (Charrouf et al. 1992; Chafchaouni-Moussaoui et al. 2013; Henry et al. 2013). Interestingly, some isolated saponins exhibit cytotoxic activity and inhibit proliferation of LNCaP, DU145, and PC3 human prostatic cell lines. This antiproliferative effect on PC3, the most sensitive cell line, is dose-dependent (Drissi et al. 2006).

Additional chemical investigations of argan press cake have evidence that it is also rich in phenolic compounds (El Monfalouti et al. 2012). Press cake from roasted kernels presents even a sevenfold higher phenolic content than press cake from unroasted kernels (173.4 vs 25.4 mg of gallic acid/kg). Sixteen phenols have been identified in press cake from roasted kernel, epicatechin being the most abundant (110 mg/kg) (Rojas et al. 2005). Phenols are well-known antioxidant molecules. Therefore, the high phenolic content of argan press cake has generated several studies aimed at evaluating its possible use as green steel anticorrosive agent. The results look promising (Afia et al. 2012, 2013).

Argan press cake could also be a cheap source of cellulose for the pharmaceutical and and food domain (Hu al 2016), and even magnetic nanocellulose for the food industry (Benmassaoud et al. 2017).

13 Conclusion

A. spinosa, an endangered species endemic to South-West Morocco, makes a specific and complex agro-sylvo-pastoral system, from financial, environmental and social standpoints. The Argan Oil Project, especially the marked and long-lasting research efforts from national and international teams, has brought a tremendous amount of knowledge on all aspects of argan related topics. This ultimately facilitated the commercialization of argan oil now produced in a sustainable way, on various markets with certified quality and origin.

The cosmetic industry has made argan oil a major ingredient in the composition a range of products, and it famous worldwide as a food ingredient whose use is not restricted to Moroccan cuisine. This quick and rather exceptional development resulted in strong changes in the agricultural sector and within the rural population, argan oil now being devoted to exportation owing to its high price on international markets.

Interestingly this oil extracted from the kernels, and making less than 4% of fresh fruit, provides several by-products now available in large amounts, opening a range of potential uses in agriculture, food and non-food industry, even in human health. Taking advantage of these by-products under the frame of a multiple-product biorefinery would further increase the value of the argan production chain, and reduce its dependency on argan oil market in case of breakdown. Avoiding further degradation of the argan forest and protecting this endangered species when climate evolution is uncertain while meeting the increasing demand of argan products is a challenge that already begins to be addressed (Moukrim et al. 2018).

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