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Abstract

Apiaceae could be defined as Aroma Tincto Oleo Crops (ATOC), e.g. plants containing both vegetable oil and essential oil. Applying agroreffinery concept to ATOC led to propose a sequential fractionation process coupling co-extraction of vegetal oil and essential oil to a valorization of by-product residues as biosourced active molecules and substrates for designing agromaterials. The aim of this thesis is to determine the biological and technological feasability of application of the ATOC-refinery concept to coriander (*Coriandrum sativum* L.). Chapter I report a bibliographic state of the art study on extraction and characterisation of coriander vegetal oil and essential oil while chapter II describes materials and methods setting up during the thesis for sampling, extraction, analysis and data processing. Chapter III focus on the study of major various biological parameters influencing bioaccumulation of vegetal oil and essential oil in coriander (different plant cultivars, different plant organs, different biological stages) and their impact on anti-oxidant activity of extracts obtained from extraction residues. In chapter IV, coriander fruits are processed by extrusion technology (mono screw and twin-screw extruder) in order to evaluate the feasability of mechanical pressing for extracting flavored vegetal oil. Influence of operating parameters on vegetal oil extraction yields (nozzle diameter and nozzle/screw distance (single-screw extruder) or screw configuration, device's filling coefficient and pressing temperature (twin-screw extruder)) is studied while the feasability of valorization of extraction cake as agromaterial (thermopressing) was stated.

Keywords : ATOC-refinery, coriander, vegetable oil, essential oil, single and twin-screw extruder, antioxidant, agromaterials.

DEDICATION

This dissertation is dedicated to my father, **NGUYEN Xuan Bang**, and to my mother, **DAO Thi Loan**, who always had confidence in me and offered me encouragement and support in all my endeavors. It is also dedicated to my darling wife, **NGUYEN Thi My Dung**, my lovely children **NGUYEN Quang Huy** for their care, love, understanding, and patience.

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Quang Hung NGUYEN

24/09/2015

In Toulouse

List of abbreviations

AAPH	2,2’-azobis (2-amidino-propane) dihydrochloride
ACP	Acyl carrier protein
AOC	Antioxidant capacity
ATP	Adenosine triphosphate
BB	Bilobed paddle screw
B.C	Before Christ
CoA	Coenzyme A
CF2C	Reverse screw
C2F	Conveying double-thread screw
DM	Monolobed paddle screw
DMAPP	Dimethylallyl pyrophosphate
DOXP	1-Deoxy-D-xylulose 5-phosphate
DP	Dry plant
DPPH	2,2-diphenyl-2-picrylhydrazyl radical
EO	Essential oil
FID	Flame ionization detector
FRAP	Ferric reducing/antioxidant power
GAE	Gallic acid equivalent
GC-FID	Gas chromatography-flame ionization detector
HD	Hydrodistillation
HMG	Hydroxy-3-methylglutaryl
HMG-CoA	Hydroxy-3-methylglutaryl-CoA
IPP	Isopentenyl diphosphate
KAS	Ketoacyl- ACP syntheses
MUFA	Monounsaturated fatty acids
NADP	Nicotinamide adenine dinucleotide phosphate
ORAC	Oxygen radical absorbance assay
PUFA	Polyunsaturated fatty acids
RI	Retention index
SD	Steam distillation
SDE	Simultaneous distillation Extraction
SFA	Saturated fatty acids
TBME	Tert-butyl methyl ether
TGA	Thermogravimetric analysis
TMSH	Trimethylsulfonium hydroxide
TE	Trolox equivalent
TPC	Total phenolic content
T2F	Trapezoidal double-thread screw
VO	Vegetable oil

Contents

GENERAL INTRODUCTION 1

Chapter I: LITERATURE REVIEW 5

I. Coriander: Introduction and characteristics 5

I.1 Taxonomy and botanical nomenclature 5

I.1.1 Botanical Classification 5

I.1.2 Common names of Coriander 5

I.1.3 Description 7

I.1.4 Growth conditions 9

I.1.5 Distribution of Coriander 10

I.2 Chemical composition 11

I.3 Composition and uses 16

I.3.1 The essential oil and fatty oil 16

I.3.2 Medical use 17

I.3.3 Use the fruits as a spice 18

I.3.4 Use the green part as a spice and vegetable 19

I.3.5 Use of essential oil of the coriander fruits 20

I.3.6 Use of the fatty oil of the coriander fruits 20

II. Vegetable Oil 21

II.1 Lipids 21

II.1.1 Fatty acid 21

II.1.2 Definition 22

II.2 Biosynthesis of fatty acids 23

II.2.1 Activation phase 23

II.2.2 Phase formation/longation 23

II.2.3 Phase desaturation 24

II.2.4 Transport phase 25

II.3 Functions of Lipids 26

II.4 Oil Extraction 27

II.4.1 The mechanical process to extract oils by the press 27

II.4.2 The physicochemical process with solvent 29

III. Essential Oil 29

III.1 General 29

III.2 Terpenes 31

III.2.1. Terpene biosynthesis 31

III.2.2 Classification 33

III.2.2.1 Monoterpenes 34
III.2.2.2 Sesquiterpenes 34
III.2.2.3 Diterpenes 35
III.2.2.4 Triterpenes 36
III.2.3 Function of Terepnes 36
III.2.4 Extraction 37
III.2.4.1 Hydrodistillation 37
III.2.4.2 Water and Steam Distillation 38
III.2.4.3 Steam Distillation 39
III.2.4.4 Simultaneous Distillation Extraction 39
III.2.4.5 Expression 39
III.3 Antioxidant activities 39
IV. Conclusion 40

Chapter II: MATERIAL AND METHODS 42
I. Plant materials 42
I.1 The materials used for studying vegetable oil and essential oil component 42
I.2 The materials was used for extrusion 42
II. Experiment methods 42
II.1 Extraction of Vegetable Oils 42
II.1.1 Extraction by Solvent 43
II.1.2 Extraction by the extruder 43
II.1.2.1 Extraction by single-screw extruder 44
II.1.2.2 Extraction by twin-screw extruder 46
II.2 Moisture content 49
II.3 Fractionation and Lipid analysis 49
II.3.1 Glyceride profile of Soxhlet extracted oil 49
II.3.2 Fatty acid composition of Soxhlet extracted Oil and pressed Oils 50
II.3.3 Oil quality analysis 50
II.4 Extraction of Essential Oils 51
II.4.1 Hydrodistillation (HD) 51
II.4.2 Simultaneous distillation Extraction (SDE) 51
II.4.3. Analysis of Essential Oils 51
II.5 Antioxidant assay 52
II.5.1 DPPH radical scavenging assay 52
II.5.2 Ferric Reducing/Antioxidant Power (FRAP) assay 53
II.5.3 Oxygen Radical Absorbance Capacity (ORAC) assays 54
II.5.4 Analysis of total phenolic compounds (TPC) 55

II.6 Agromaterials 55
II.6.1 Mechanical properties in bending of the panels produced 55
II.6.2 Surface hardness 57
II.6.3 TGA measurements 57
II.6.4 Impact strength 58
II.7 Statistical analysis 58

Chapter 3: BIOACCUMULATION 59
I. Plant material, crop management and experimental conditions in Auch and Vietnam 59
I.1 Plant material, crop management and experimental conditions in Auch 59
I.2 Plant material, crop management and experimental conditions in Vietnam 61
II. Vegetable Oils Analysis 64
II.1 Oil content and fatty acid composition in different part of Coriander 64
II.2 Effects of sowing time on yield and fatty acid composition of coriander 68
II.3 Fatty acid accumulation in maturity of coriander 71
II. 4 Effects of different location on yield and fatty acid composition of coriander 82
II.5 Fatty acid composition of different cultivars 84
III. Analysis of Essential Oil 89
III.1 Effects of sowing time on yield and fatty acid composition of coriander 89
III.2 Essential oil components of different cultivars of coriander 92
IV. Antioxidant Properties 95
V. Conclusion 98

Chapter 4: BIOREFINERY OF CORIANDER FRUITS, A FIRST INVESTIGATION 101
I. Extraction of coriander oil using the single-screw extrusion technology 103
I.1 Effect of operating conditions on oil yield in the single-screw extruder 105
I.2 Influence of the coriander cultivars on oil extraction efficiency 110
I.3 Potential uses of the press cakes produced in the single-screw extruder 116
II. Extraction of coriander oil using the twin-screw extrusion technology 120
II.1 Chemical composition of the coriander fruits used 124
II.2 Influence of the operating conditions on oil extraction efficiency 125
II.3 Influence of the operating conditions on oil quality 132
II.4 Potential uses of the press cakes produced in the twin-screw extruder 134
III. New renewable and biodegradable fiberboards from a coriander press cake 137
IV. Conclusion 140

GENERAL CONCLUSION 144

REFERENCES 148

List of Figures

Figure 1: (a) Petroselinic acid; (b) linalool....1
Figure 2: Coriander (Coriandrum sativum L.); Origin: www.botanical.com....8
Figure 3: (a) Coriander fruits; (b) Dried fruit of Coriander....13
Figure 4: Structures of the major compounds identified in the essential oil of coriander fruits in previous investigations (Ramezani et al., 2009; Shavandi et al., 2012) 14
Figure 5: (a) Coriander leaves (right); (b) Coriander roots (left) 15
Figure 6: Structures of the major compounds identified in the essential oil of coriander leaves (Shahwar et al., 2012)....15
Figure 7: Structure of some common lipids 22
Figure 8: Biosynthesis of fatty acids by fatty acid synthesis (Salas et al., 2000). 26
Figure 9: (a) Twin-screw extrusion scheme (left); (b) soxhlet extraction (right)....28
Figure 10: Industries and product categories that use essential oil (Brand-Williams et al., 1995)....30
Figure 11: The diagram of the mevalonate pathway (Lange et al., 2000)....33
Figure 12: Monoterpene structures....34
Figure 13: Sesauiterpenes structures 35
Figure 14: Diterpenes structures....35
Figure 15: Triterpens structures....36
Figure 16: Extraction processes used and products from spice, herb and aromatic plants (Brand-Williams et al., 1995)....37
Figure 17: Hydrodistillation process (left) and Clevenger apparatus (right)....38
Figure 18: Single-screw extruder OMEGA 20....44
Figure 19: Functional zones of pressing (single-screw extruder OMEGA 20)....45
Figure 20: Twin-screw extruder type Clextral BC 21 46
Figure 21: The Screws for extraction of vegetable oil from coriander fruits....47
Figure 22: Schematic modular barrel of the twin-screw extruder Clextral BC 21 used for extraction of vegetable oil from coriander fruits....48
Figure 23: Screw configurations for extraction of vegetable oil from coriander fruits....49
Figure 24: Three point flexure test 56
Figure 25: Instron 33R4204 (USA) machine for bending Test....56
Figure 26: The oil contents of coriander flower, leaves, roots and fruits....65
Figure 27: Petroselinic acid composition in different parts of coriander 66
Figure 28: Chromatogram showing fatty acid composition of coriander fruits analyzed by GC....69
Figure 29: The changes in oil yield of coriander during from flowering period to maturity in 2009 72

Figure 30: Variations of major fatty acid from flowering period to maturity (in Auch, 2009) 73
Figure 31: Variations of oil yields of coriander from flowering period to maturity (2009, 2010, 2011) 75
Figure 32: The tendency of petroselinic acid from newly form fruits to maturity (2009, 2010, 2011) 75
Figure 33: Variations of major fatty acid from flowering period to maturity (in Auch, 2010) 76
Figure 34: Variations of major fatty acid from flowering period to maturity (in Auch, 2011) 77
Figure 35: Variations of major fatty acid from flowering period to maturity (in Vietnam, 2012) 77
Figure 36: Typical GC chromatogram of essential oil of coriander cultivars 92
Figure 37: The total phenolic contents in different coriander cultivars 96
Figure 38: Scheme of preparation and analysis of coriander extracts 101
Figure 39: Schematic representation of oil expression of coriander fruits 104
Figure 40: Variation of oil extraction yield (R_S) on different screw configuration 106
Figure 41: Variation in oil extraction yields (%) for the different coriander cultivars tested 111
Figure 42: Comparison between oil extraction yields in the single-screw extruder and using the Soxhlet method with *n*-hexane as extracting solvent. 112
Figure 43: Total phenolic contents (TPC) for the different coriander cultivars used..... 118
Figure 44: Screw configurations for extraction of vegetable oil from coriander fruits..... 121
Figure 45: Schematic representation of oil expression of coriander fruits using a twin-screw extruder (type Clextral BC 21). 122
Figure 46: Variation in oil extraction yields for trials 1 to 10 126
Figure 47: Cumulative number of particles as a function of particle size for the press cakes from trials 2 and 10 135

List of Tables

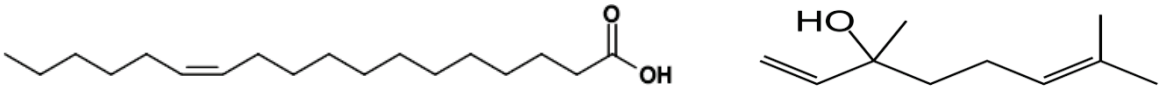
Table 1: Some common names of Coriander (*Coriandrum savitvum* L.) 6
Table 2: Structure of Terpens 33
Table 3: The prevailing weather condition during the plant cycle in three years (2009, 2010 and 2011) in Auch 60
Table 4: The mean air temperature and monthly rainfall in three location (Ha Noi, Thai Binh and Ha Tinh) in 2012 62
Table 5: The fatty acid composition and oil yield in different parts of coriander 67
Table 6: Fatty acid composition of coriander fruit on different sowing time (2011) 70
Table 7: Changes on fatty acid and oil yield during fruits maturation of Coriander in 2009 74
Table 8: Changes on fatty acid and oil yield during fruits maturation of Coriander in 2010 78
Table 9: Changes on fatty acid and oil yield during fruits maturation of Coriander in 2011 79
Table 10: Changes on fatty acid and oil yield during fruits maturation of coriander in 2012 81
Table 11: Effects of different locations on yield and fatty acid composition of coriander fruits (Vietnamese coriander - 2012) 83
Table 12: Variation in oil content and fatty acid composition of coriander cultivars (cultivated in Auch - 2011) 87
Table 13: Comparison oil content and fatty acid composition between origin and coriander cultivars cultivated in Auch 2011 88
Table 14: Composition of essential oil in coriander fruits at different sowing times (DS1, DS2 and DS3) 91
Table 15: Composition of essential oil of coriander cultivars 94
Table 16: Antioxidant capacity and TPC extracts from residue after hydrodistilltaion of cultivars 97
Table 17: Effect of screw configuration conditions on oil extraction in single-screw extruder OMEGA 20 107
Table 18: Variation in fatty acid composition of oil extracted from coriander fruits in a single-screw extruder OMEGA 20 108
Table 19: (following): Variation in fatty acid composition of oil extracted from coriander fruits in a single-screw extruder OMEGA 20 109
Table 20: Chemical composition of the different coriander fruits 110
Table 21: Effect of the coriander cultivars on the oil extraction efficiency in a single-screw extruder OMEGA 20 114
Table 22: Fatty acid composition of pressed oils from different coriander cultivars and produced in a single-screw extruder OMEGA 20 115

Table 23: Essential oil composition in press cakes from different coriander cultivars and produced in a single-screw extruder OMEGA 20 117
Table 24: Antioxidant activity and TPC of press cakes produced by single-screw extrusion depending on the coriander cultivars. 119
Table 25: Results of the expression experiments conducted with the Clextral BC 21 twin-screw extruder 127
Table 26: Quality and fatty acid composition of Soxhlet extracted oil, and pressed oils from coriander fruits (trials 2, 4, 6, 8 and 10) 133
Table 27: Essential oil composition in coriander fruits and press cakes from trials 2, 4, 7, 8 and 10 136
Table 28: Essential oil in coriander fruits and press cakes from trials 2, 4, 7, 8 and 10, and antioxidant capacity of press cakes 2, 4, 8 and 10 after hydrodistillation (DPPH assay). 137
Table 29: Thermo-pressing conditions, mechanical properties, thickness swelling (TS) and water absorption (WA) of the fiberboards manufactured. 139

GENERAL INTRODUCTION

Coriander (*Coriandrum sativum* L.) is an annual herb belonging to the Apiaceae family and indigenous to the Mediterranean basin areas and is nowadays mostly grown in temperate areas around the Mediterranean basin, India, China, Thailand and Eastern Europe. It is used as herbal condiment for many Asian and Mediterranean culinary preparations. This plant is widely distributed and mainly cultivated for its fruits which are used for different purposes such as aromatherapy, food, drugs, cosmetics and perfumery. The fruits also have medicinal uses, treating rheumatism and pain in the joints, gastrointestinal complaints, flatulence and gastralgia worms, indigestion, insomnia, convulsions, anxiety and loss of appetite, glycaemia as well as sleep prolonging. The interest of coriander fruits oil becomes more important since the European Union authorized the use of coriander oil for use as a food supplement.

The coriander fruits contain Vegetable oil (VO) and Essential oil (EO) fraction, both this fraction are the most important sustainable natural resources for the preparation of various foods ingredient, food additives, function component for nutraceuticals, natural remedies, cosmetiques and other applications. The vegetable oil contain petroselinic acid (Figure 1.a), it is an unusual fatty acid that occurs primarily in fruits of coriander. This compounds represent about 70-80 % of the respective oil fractions and it can be oxidative cleaved to produce a mixture of lauric acid, a compound useful in the production of detergents, and adipic acid, a C6 dicarboxylic acid which is utilized in the synthesis of nylon polymer. Moreover, petroselinic acid is an uncommon isomer of oleic acid which is found in high levels in a restricted range of fruits oils, mostly from the Apiaceae family. It forms a unique oleochemical with high potential for food, cosmetic and pharmaceutical industries and may further allow the synthesis of a large number of interesting platform molecules. The essential oil contain linalool (up to 60 - 70%) (Figure 1.b) and Linalool is widely used in the flavoring industry such as cosmetics, lotions, creams, shampoos, perfumes, etc.



(a) (b)

Figure 1: (a) Petroselinic acid; (**b)** linalool

Aroma Tincto Oleo Crops (ATOC) concept are plants contain both a Vegetable Oil (VO) and Essential Oil (EO). Presently, depending on the industrial sector concerned (oil or aromatic industry), only one of these two fractions is valorised, the other being considered as waste. The development of an integrated valorisation of ATOC allowing co-extractions of VO and EO constituted the basics of the ATOC-refinery concept, e.g. a sequential valorization process allowing co-extractions of VO and EO while not penalizing the subsequent valorization of residual by products as biosourced molecules (antioxidant or biocides ones) or as substrate for designing odorous agromaterials. The technology approach was based on the coupling of an extruder (single or twin-screw extruders) to an hydrodistillator/extractor in order to extract first virgin VO from fruits and EO from extruded cake, then antioxidant and biocide molecule cocktails from distillated cake and finally to valorize the ultimate solid residue as substrate for either agromaterials design (by thermopressing or injection-moulding) or bio-energy production (by combustion of formulated pellets). Experiments were conducted with a selected model plant commonly used as a condiment or a spice in the Mediterranean and Sout-East Asia areas : Coriander (*Coriandrum sativum* L.). Results obtained at lab and pilot-scale levels with coriander fruits reported extraction of both VO and EO with high amounts (up to 75%) of valuable components, respectively linalool and petroselinic acid, the refinery approach was then extent to the others valuable parts of the plant, i.e. leaves and roots, in order to reach to valorization of the overall chemical potential of coriander. Based on this concept, biorefinery was developed in this thesis. Plant materials of coriander were processed into four parts: vegetable oil, essential oil (or volatile extract), methanolic extract and final residue (biomaterial). By this way, the molecules in the plants can be sequentially extracted and used, thus avoiding a great waste of natural resources.

Aims and Objectives

The main aim of our object was to comprehensively assess the yield, vegetable oil and essential oil composition and antioxidant properties of coriander and the initial research on waste treatment of coriander into potential sources of raw materials in industrial service to improve the effectiveness of the production and processing of coriander and their products.

- To determine composition of VO and EO of different cultivars of coriander (from different geographic origins). These coriander cultivars was cultivated with organic condition in Auch (near Toulouse, south-west France, 43°38'47"N, 0°35'08"E).
- To determine composition of VO and EO of different parts of coriander (Fruits, Leaves, Stem and Roots).
- To evaluate bioaccumulation of VO and EO during fruit ripening of French and Vietnamese coriander in France and Vietnam.
- To determine the antioxidant properties of extracts by different methods (DPPH, ORAC, FRAP and Analysis of Total Phenolic Compounds) from different parts of their distillation residues of coriander.
- Optimization of operating conditions of extrusion by single and twin screw extruder (single crew OMEGA 20 and twin screw Clextral BC 21) in order to evaluate the feasibility of a processing of extraction fruits oil.
- Feasibility study and influence of operating conditions to assess the flexural properties of the test specimens according to the French standard, including breaking load, flexural strength at break, and elastic modulus.

Thesis outline

This thesis is organized into four chapters and a general conclusion. The mail work of each chapter is recapitulated as follows:

In the first chapter: The first section of this chapter will describe the biological characteristics, distribution, cultivation conditions and industrial processing of coriander. The literature overview of the chemical composition, the valuable used of vegetable oil and essential oil of coriander in the previous studies. This chapter provide a general introduction to studies on vegetable oil and essential oil. Based on knowledge about vegetable oil and essential oil coriander and some basis of methods existing, we have proposed the design of new fractionation process of coriander whole my experiments.

The **second chapter** describes the design, synthesis, characterization and evaluation of overview of different methods in previous studies, and then to give reasons why to select our methods is applied to do experiment. The method, experimental conditions and the processing and analysis of data will be present in this chapter.

The **third chapter**, the results was obtained: (i) composition of vegetable oil and essential oil of different parts of coriander (Fruits, Leaves, Stem and Roots); (ii) vegetable oil and essential oil component of the different coriander cultivars; (iii) the bioaccumulation of VO and EO in maturity stages of French and Vietnamese coriander; (iv) antioxidant properties of extracts prepared by different methods (DPPH, ORAC, FRAP) will be present this chapter. Based on the results achieved, the explanations, argumentation and discussion will be presented.

The last chapter will be devoted to the characterization of coriander fruits and the extraction of the products will be analyzed. This chapter we show that optimization of operating conditions of extrusion by single and twin screw extruder (single crew OMEGA 20 and twin screw CLextral BC 21) in order to evaluate the feasibility of an processing of extraction fruits oil. Finally, feasibility study and influence of operating conditions to assess the flexural properties of cakes from the fractionation of coriander fruits in single and twin-screw extruder by thermopressing and instron testing machine. The characteristics of biomaterials thus obtained will be described.

In conclusion, is a summary of the most important contribution of this research. we point out the major perspectives of our approach and give some idea to prospects.

Chapter I: LITERATURE REVIEW

I. Coriander: Introduction and characteristics

I.1 Taxonomy and botanical nomenclature

I.1.1 Botanical Classification

Kingdom	*Plantae*
Subkingdom	*Trachaeobionta* (vascular plants)
Superdivision	*Spermatophyta* (Seed plants)
Division	*Magnoliophyta* (Flowering plants)
Class	*Magnoliopsida* (Dicotyledons)
Subclass	*Rosidae*
Order	*Apiales*
Family	*Apiaceae* (Umbellifera Juss.)
Genus	*Coriandrum* L.

Accepted species name: ***Coriandrum sativum* L.** , Sp. Pl., 256. 1753.

Synonyms:

Coriandrum globosum Salisb. , Prod. stirp. Chap. Allerton, 166. 1796.

Coriandrum melphitense Ten. & Guss. , Ind. sem. hort. Neap., 3. 1837.

Bifora loureirii Kostel. , Allg. med.- pharm. Fl. vol. 4, 1183. 1835.

Selinum coriandrum E.H.L. Krause in Sturm, , Deutschl. Fl. ed. vol. 2, 12, 163. 1904.

Coriandrum testiculatum auct. , Fl. cochinch., 180. 1790., non non L.

Coriandrum diversifolium Gilib. , Fl. lit. inch. vol. 2, 26. 1782.

Coriandrum majus Gouan , Hortus monsp., 145. 1762.

Atrema testiculatum Miq. , Fl. Ned. Ind. vol. 1 (1), 5, 744. 1856.

Typus: *Coriandrum sativum* L. Described from Italy, Herb. Linn. No. 363/1 (LINN) (Jafri, Fl. Libya 117 (1985) 23).

I.1.2 Common names of Coriander

Coriander (*Coriandrum sativum* L.), also known as **cilantro**, **Chinese parsley** or **dhania**, has several common names according to geographical regions (Table 1).

Table 1: Some common names of Coriander (*Coriandrum savitvum* L.)

Location	Common name of the species
Asian	
Indonesian	Ketumbar (fruits), Daun ketumbar (herb)
Malayalam	കൊത്തമല്ലി, കൊത്തമ്പാല്, കൊത്തമ്പാ, മല്ലി, കൊത്തമ്പാലാരി (Kothamalli, Kotthampal, Malli, Kottampalari)
Malay	Ketumbar (fruits), Daun ketumbar, Wansui (herb), Penjilang
Lao	ຫອມປ້ອມ, ຜັກຫອມປ້ອມ (Hom-pom, Pak hom pom)
Japanese	コリアンダー, コエンドロ (Korianda, Koendoro)
Khmer	Vannsui, Chi van-suy
Korean	고수, 고수풀, 코리앤더, 코리안더, 코리엔더 Kosu, Kosu-pul, Koriaendeo, Koriandeo, Koriendeo
Vietnamese	Cây rau mùi, Hồ tụy, Mùi, Ngò, Ngò ta
Thailand	ผักชี, เมล็ดผักชี, ลูกผักชี Pak chi, Pak chi met (herb); Mellet pak chi, Luk pak chi (fruits
Mongolian	Ycyy (Üsüü)
Chinese	Fan Yan Sui, Wan-Swee, Yan Shi, Yuen sai
Chinese (Mandarin)	番芫荽 [fān yuán suī], 胡荽 [hú suī], 香菜 [xiāng cài], 香荽 [xiāng suī], 芫荽 [yuán suī], 芫茜 [yuán xī] Hu sui, Yuan sui, Yuan xi, Yan shi, Fan yuan sui, Xiang cai, Xiang sui
Chinese (Cantonese)	番芫荽 [fàan yùhn sài], 胡荽 [wùh sèui], 香菜 [hēung choi], 香荽 [hēung sèui], 芫荽 [yùhn sèui], 芫茜 [yùhn sāi] Wuh seui, Yuhn seui, Yuhn sai, Faan yuhn sai, Heung choi, Heung seui
Europe	
Latvian	Kinzas, Koriandrs
Lithuanian	Kalendra, Blakinė kalendra
Italian	Coriandolo
Hungarian	Koriander, Cigánypetrezselyem, Beléndfű, Zergefű
Polish	Kolendra siewna

Portuguese	Coentro
Spanish	Coriandro, Cilantro
Slovenian	Koriander
Turkish	Kişniş, Kişnic
Ukrainian	Коріандр посивний (Koriandr posyvnyj)
Swedish	Koriander
Russian	Кинза, Кишнец, Кориандр Kinza, Kishnets (herb); Koriandr (fruits)
Norwegian	Koriander
Macedonian	Кориjандер (Korijander)
Greek	Κόλιανδρο, Κορίαντρο, Κορίανδρο, Κόλιαντρος, Κολίανδρο Koliandro, Kolianthro, Koriantro, Koriandro, Koliantros
German	Koriander, Wanzenkümmel, Chinesische Petersilie, Indische Petersilie (herb)
French	Coriandre, Punaise mâle, Persil arabe
Estonian	Aedkoriander, Koriander
English	Coriander, Chinese parsley, Indian parsley (herb)
Czech	Koriandr, Koriandr setý
Danish	Coriander
Bulgarian	Кориандър (Koriandur)
Arabic	Kusbarah, Kuzbarah

Origin: http: http://gernot-katzers-spice-pages.com/engl/Cori_sat.html?spicenames=ja#fam

I.1.3 Description

Coriander is both an annual and a perennial herb. It is flowered herb and is usually distinguished with vertical, glabrous, and profusely branching plant. The height of the coriander's plant is in the range from 0.2 to 1.4 m with a well-developing taproot. The stem of coriander usually erect, sympodial and monochasial-branched, sometimes it has several side branches at the basal node. Each branch is finished with an inflorescence. The ribbed stem has the green color, but it turns to red or violet when it is in the flowering period. The stem of full-grown plant is hollow, its basal parts diameter can reach to 2 cm. The coriander's leaves are variable in shape, size and number. They have yellow-green color and sheath surrounding and is scariously margined. The supporting stem is up to three quarters of its circumference.



Figure 2: Coriander (*Coriandrum sativum* L.); Origin: www.botanical.com

The petiole and rachis has light green color and subterete, sulcate appearance; the blade is white waxy, shiny green and often appears with dark green veins; basal 1-3 leaves usually simple, withering early, often in a rosette, blade ovate in outline, deeply cleft or parted into

usually 3 incised-dentate lobes; next leaves decompound, petiole 0-15 cm long, blade ovate or elliptical in outline, up to 30 cm x 15 cm, usually pinnately divided into 3-11 leaflets, each like the blade of the simple lower leaves or again pinnately divided into 3-7 simple leaf-like lobes; all higher leaves compound, petiole restricted to the sheath, blade divided into 3 leaflets of which the central one is largest, each often variously divided into ultimately sublinear, entire, acute lobes. Inflorescence an indeterminate, compound umbel; peduncle up to 15 cm long; bracts sublinear, 0-2, up to 11 mm long; primary rays 2-8, up to 4.5 cm long; bracteoles 0-6, linear, up to 1 cm long; secondary rays up to 20, up to 5 mm long; usually each umbellate has bisexual peripheral flowers, and the central flowers are sometimes male; calyx in all flowers represented by 5 small lobes; corolla with 5 white or pale pink petals, heart-shaped, very small (1 mm x 1 mm) in male flowers, in bisexual peripheral flowers usually 3 petals are larger: 1 petal develops 2 ovate lobes of about 3 mm x 2 mm and the 2 adjacent petals develop each one lobe; stamens 5, filaments up to 2.5 mm long, white; pistil rudimentary in male flowers, in bisexual flowers with inferior ovary, a conical stylopodium bearing 2 diverging styles up to 2 mm long, each one ending in a minutely papillate stigma.

The fruits are an ovoid to globose schizocarp, up to 6 mm in diameter, yellow-brown with 10 straight longitudinal ribs alternating with 10 wavy longitudinal ridges, often crowned by the dry persistent calyx lobes and the stylopodium with styles; fruit does usually not split at maturity; it contains 2 mericarps which each bear on their concave side 2 longitudinal, rather wide lines (vittae), containing essential oil. Seed 1 per mericarp, with testa attached to the fruit wall. Seedling with epigeal germination; taproot thin with many lateral roots; hypocotyls up to 2.5 cm long; cotyledons opposite, oblanceolate, up to 3 cm x 4 mm, pale green (Diederichsen, 1996; Maroufi et al., 2010) (Figure 2).

I.1.4 Growth conditions

In generally, coriander is normally grown in a wide range of conditions. Its grown depends on different factors as the type of soil, geographical areas, effect of sowing time, climatic and environmental conditions. In Mediterranean region, Carrubba et al. showed that the most commonly recommended cropping technique is spring sowing (March–April), since the optimum soil temperature for fruits germination ranges between 20 and 23^{o}C, and the crop shows a remarkable sensitivity to frost and cold. (Carrubba et al., 2006). But in Asia, the best condition for cultivation is from October to February. Because of this period is

favorable for the growth of plants. For the good crop, it requires a cool climate in the early stages and warm dry weather at maturity of growth. Because this period is favorable for the growth of plants. The best sowing density require about 12 to 15 kg/ha-1. Before sowing, the fruits germination is a earlier than the fruit intact, so the fruits should be rubbed into splits fruits. To get the best conditions for the germination of fruits, fruits are soaked in water from 12 to 14 hours and drying them in shade for 12 hours also help in quicker germination. The fruits are sown in lines at a spacing of 25 cm between rows and 15 cm between plants. Sometimes the fruits are sown broadcast and then mixed with land use scratch. Depending on the temperature, germination occurs in 10-15 days (Karan singh, 2007; Khare, 2007; Kirtikar and Basu, 1918; Pathak et al., 2011).

In generally, coriander could be grown on different types of soils, but prefer deep and fertile soils with good water holding capacity, pH from 6.5 to 7.5, and good drainage. Thus, in order to get the best condition for growing, coriander should sown in type of soils where can provided sufficient moisture and organic manure (Karan singh, 2007; Khare, 2007). At soil temperatures above 15^{o}C, fruits could be germinated within two week after being sown and at soil temperature near 25^{o}C are ideal for faster germination. Before the planting, the soils are prepared with fertilizer rate of 30 kg Phosphorus, 20 kg Potassium and 20 kg Nitrogen per hactor. In the irrigated areas, application of N is increased to 60 kg/ha, half of the dose of N is applied as basal dose, and the remaining half is applied 30-45 days after sowing. For a good condition growing, generally hoeing and weeding is necessary in order to provide better soil aeration to the crop. For irrigated crops, first hoeing and weeding are done in about 30 days after sowing and depending upon the weed growth one or two more weeding are also done. Besides, some diseases could be harmful to the crop such as stem-rot, wilt, powdery mildew and stem-gall. Correspondingly, all disease is effectively controlled during the crop (E., 2002; Khare, 2007; Pathak et al., 2011)

I.1.5 Distribution of Coriander

From the 18th century, some authors described coriander (*Coriandrum sativum* L.) as common weeds spreading from Europe to southern Russia (Alefeld, 1866). Until now, coriander has been widely cultivated and used in many parts around the world such as in Russia, Central Europe, Asia, and Africa... According to Ravi, India is the world's largest

producer and exporter of coriander and refined products (Ravi et al., 2007). Others main producers of commercial coriander are Hungary, Poland, Romania, Czech Republic, Slovakia, Morocco, Canada, Pakistan, Iran, Turkey, Guatemala, Mexico, and Argentina (Agri-facts, 1998; Kiehn FA, 1992; López et al., 2008).

For coriander origin, Diederichsen (1996) also mentioned in his report that the origin coriander is derive in the coastal areas North and East of the Mediterranean Sea and from there the plant spread to Europe, Africa and Asia in ancient time. There is evidence that coriander was grown in Assyria (Iraq) around 9,000 years ago, and remains of coriander fruits were found in Tombs in Israel dated 8,000 years old. In India, coriander has been grown for the last 5,000 year. It was used as a medicinal plant and as a condiment by the Egyptians since at least 4,500 year ago (Diederichsen, 1996). In a recent report also pointed out that coriander was grown in England as far back as the 1st century A.D (introduced by the Romans), and was known as a vegetable in China in the 5th century A.D (introduced through Persia). The crop was brought to Hispaniola by Spanish conquerors late in the 15th century, and then to other European colonier in the American continent, Australia and the Philippines (Morales-Payan and Stall, 2005). Besides, there are some notes mentioned about the existence of coriander in wild nature with the scientific name "*Coriandrum tordylium*", and this wild coriander can already be found in Russia for use as a genetic resource. Herbarium specimens of wild of coriander (*Coriandrum tordylium*) from Edinburgh (Scotland) and Turku (Finland) demonstrate that this annual species is very similar to the cultivated *Coriandrum sativum* L. (Diederichsen, 1996). However, up to now there is not any project to study on essential oils, fatty acid and valuable use of wild species coriander and no certain information about the wild species exists. The wild species might be interesting for coriander breeding, but it has not been reported whether crossing *Coriandrum sativum* L. and *Coriandrum tordylium* may be possible. Thus, this is an important question for future research, and may help to shed light on the evolution of the crop. The evidence above indicated coriander had widely distributed in many parts of the world.

I.2 Chemical composition

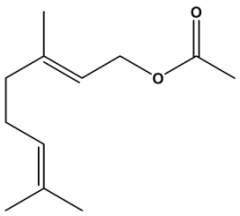
In recent years, a considerable amount of literature has been published on biochemical composition of coriander. These studies determine not only the vegetable oil composition but also could be recognized the essential oil of coriander. Nowadays, the raw material of coriander became important for industrial use and further processing. Therefore, coriander fruits are one of the most popular raw materials in flavoring and oil industry. Besides, there exist very different uses of coriander and these are based on different parts of the plant. So, many studies so far investigate the biochemical composition of different parts of coriander. Currently, lot of published studies determine of chemical composition of coriander fruits (Figure 3.a,b). According to Asgarpanah et al (2012), the biochemical composition of ripe fruits of coriander including contents vary from 11.5 to 21.3% (protein); 17.8 to 19.15% (fat); 28.4 to 29.1% (crude fibers); 4.9 to 6.0% (ash); starch (10.53%), sugars (1.92%), minerals (4.98%) and water (11.37%.) essential oils (0.84%) and pentosans (10.29%) such as water (11.37%) (Asgarpanah and Kazemivash, 2012; Diederichsen, 1996). For the study of vegetable oils and essential oil of coriander, there is a large volume of published studies have been done. Surveys such as that conducted by some authors have shown that the essential oil content in dried coriander fruits is in the range of 0.03 – 2.6 %. Linalool varies from (40.9 to 79.9%), generyl acetate (2.3 to 14.2%), γ-terpinene (0.1 to 13.6%), and α-pinene (1.2 to 7.1%) were identified as the main components in the oil of the coriander fruits (Bhuiyan et al., 2009; Coleman and Lawrence, 1992; Eikani et al., 2007; Guenther, 1948; Guenther, 1950; Lassak, 1996; Nejad Ebrahimi et al., 2010; Pino et al., 1996; Rastogi RP, 1993; Taskinen and Nykanen, 1975). Many phytochemical studies are done to investigate and compare the chemical composition of the coriander fruits essential oil grown in different regions (Anitescu et al., 1997; Bandoni et al., 1998; Ramezani et al., 2009). The presence of other minerals in fruits of coriander such as Mg, Al, Si,P, S, Cl, K, Ca, Ti, Mn, Fe, Cu, an Zn are reported using X-ray fluorescence analysis (Al-Bataina et al., 2003; Diederichsen, 1996). Other researches mainly focused on the fruit oil fatty acid composition, triacylglycerols and glycerophospholipids (Ramadan and Mörsel, 2002; Sriti et al., 2010b), tocopherols and tocotrienols (Horvath et al., 2006), or effects on plasma lipids (Tahvonen et al., 2005). According to Millam et al. (1997) and Kim SW (1996) about 13 – 18% dry weight of the fruit is fatty oil, of which up to 75% is petroselinic acid (C18:1) which is largely used in industry to form lauric acid in soaps and detergents and also C6 dicarboxylic acid for use as a feedstock in the manufacture of nylon.



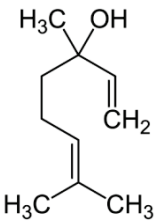
(a) (b)

Figure 3: (**a)** Coriander fruits; **(b)** Dried fruits of Coriander

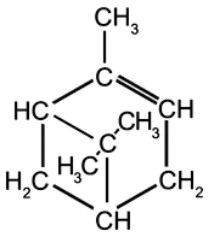
However, some recent reports indicate that the percentage of essential oil components and even the chemical composition of essential oil can be changed by many different factors such as under saline conditions (Karray-Bouraoui et al., 2009; Neffati et al., 2011); climates, geographical regions, sowing time and harvesting stages will also affect the amount and composition of the essential oil of coriander (Mohammadi and Saharkhiz, 2011; Msaada et al., 2009c; Msaada et al., 2009d; Yildirim and Gok, 2012). The structures of the major compounds identified of the essential oil of coriander are represented in Figure 4.



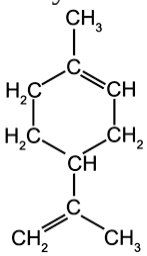
Geranyl acetate



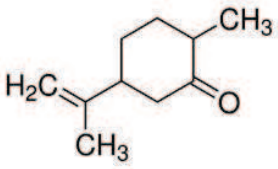
Linalool



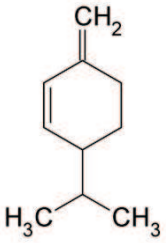
α - Pinene



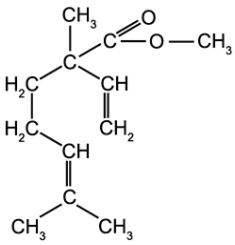
Linmonene



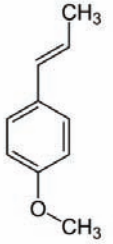
Dihydrocarvone



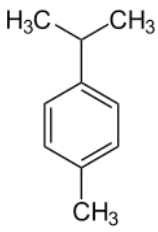
Phellandrene



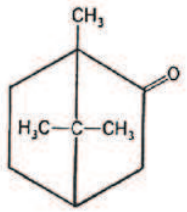
Linalyl acetate



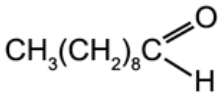
Anethole



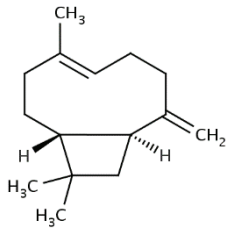
Para-cymene



Camphor



Decanal



β- Caryophyllene

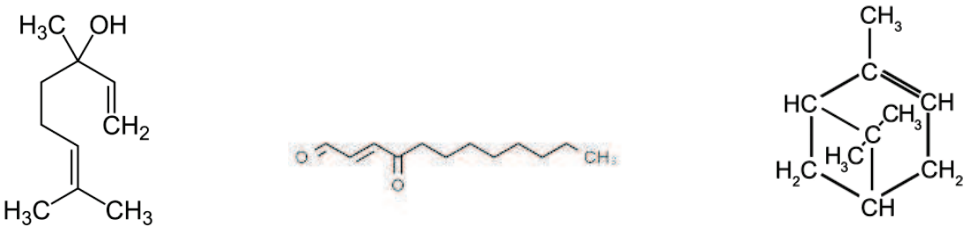
Figure 4: Structures of the major compounds identified in the essential oil of coriander fruits in previous investigations (Ramezani et al., 2009; Shavandi et al., 2012)

Up to now, not only studied essential oil and vegetable oil in coriander fruits, but also studied in the leaves and roots of coriander (the leaves and roots of coriander see in Figure 5.a,b). However, not many studies on the leaves and roots, because of essential oil and vegetable oil content in these parts of coriander to reach very low levels. So it is not really interesting in research and exploitation of them. According to Bhuiyan et al. (2009) analysed the leaf oil contained mostly aromatic acids, including 2- decenoic acid (30.8%), E-11-tetradecenoic acid (13.4%), capric acid (12.7%), undecyl alcohol (6.4%), tridecanoic acid (5.5%), and undecanoic acid (7.1%) as major constituents (Bhuiyan et al., 2009). Matasyoh et al. (2009) showed that essential oil of coriander leaves of Kenya is 2E-decenal (15.9%), decanal (14.3%), 2E-decen-1-ol (14.2%), and n-decanol (13.6%), this componse such as the main ones (Matasyoh et al., 2009). Data from several sources have identified that two new isocoumarins, coriandrone A and B were isolated from the aerial parts of C. sativum together with two known isocoumarins, coriandrin and dihydrocoriandrin (Baba et al., 1991). Three new isocoumarins, coriandrones C, D, and E were also isolated from coriander whole plants (Taniguchi et al., 1996). There is only one volume of published studies describing the vegetable oil and essential oil of coriander roots. The author was conducted to investigate the effect of saline stress on the essential oil and fatty acid composition of Tunisian coriander roots in hydroponic culture condition. Coriander seedlings were treated during 3 weeks with different NaCl concentrations (0, 25, 50 and 75 mM). Neffati et al. (2009) showed that essential oil yield was 0.06% in the control increased significantly with increasing NaCl concentrations to reach 0.12 and 0.21% at 50 and 75 mM NaCl, respectively. The major volatile component was (E)-2-dodecenal with 52% of total essential oil constituents, followed by decanal, dodecanal, (E)-2-tridecenal and (E)-2-dodecenal. Total fatty acid amount of coriander roots increased significantly only with 50 and 75 mM NaCl. Three major fatty acids: linoleic (43%), oleic (25.5%) and

palmitic (21.6%) were identified (Neffati and Marzouk, 2009). Structures of the major compounds identified in the essential oil of coriander leaves are presented in Figure 6.



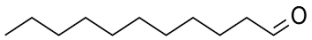
Figure 5: (a) Coriander leaves (right)**; (b)** Coriander roots (left)



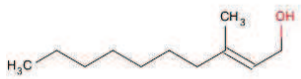
Linalool

2-Dodecenal

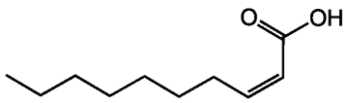
α - Pinene



Undecanal



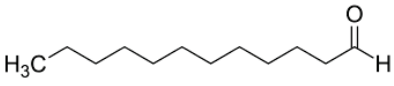
2-Decen-1-ol



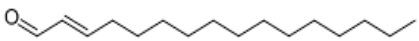
2-Decenoic acid



(E)-2-Tetradecenal



Citronellyl acetate



(E)-2-hexadecenal

Figure 6: Structures of the major compounds identified in the essential oil of coriander leaves (Shahwar et al., 2012)

I.3 Composition and uses

The main products obtained from coriander are green herb and dried spice. In industry, the main product from coriander are distilled oil and solvent extracted oleo-resin for the aroma and flavor producing (Islam et al., 2009). It is interesting to note that, coriander fruits'oil is one of the 20 major essential oils in the world market (Coleman&Lawrence, 1992). Its commercial value depends on its physical properties, chemical composition and aroma (Smallfield et al., 2001). Coriander's oils are familiar not only in perfumery, food, beverage and pharmaceutical industry, but also in medicine. They could be used as antioxidant, treatment of nervous disorder, gut modulatory, blood pressure lowering and diuretic activities, anti diabetic and antimicrobial agent (Isabelle et al., 2010; Jabeen et al., 2009). Coriander produces a considerable quantity of nectar to draw many insects for pollination. Correspondingly, the bees could be obtained in large quantities honey (about 500 kg per hectare) from coriander (Diederichsen, 1996).

I.3.1 The essential oil and fatty oil

The most important constituents in coriander fruits are the essential oil and the fatty oil. Both are used in industry, either in separating or in combining. After extraction of the essential oil, the fatty oil is obtained from the extraction residues either by pressing or by extraction. The essential oil content of the dried fruits varies from very low (0.03%) to a maximum report of 2.7% (Bandara et al., 2000; Purseglove, 1981). Linalool is the main volatile compound in fruits, typically constituting more than 70% of the total essential oil. Fatty acids are also important components of coriander fruits. The content of fatty oil varies between 9.9 and 27.7% and the main fatty acids detected in coriander, in decreasing order, are petroselinic, linoleic, palmitic, and stearic acids (Diederichsen, 1996; Ramadan and Mörsel, 2003). Furthermore, cleavage of the unusual double bond in petroselinic acid leads to the production of lauric acid, to obtain surfactants and edible products, and adipic acid for nylon synthesis (Isbell et al., 2006; Kleiman R, 1982). Beside, residues from the distillation could be used for livestock feed, and the fatty acids also have potential uses as lubricants (Purseglove, 1981).

I.3.2 Medical use

From the ancient time, people had known to use coriander as a medicine. The ancient Egyptians had used coriander for medicinal purposes for thousands years (Mathias, 1994). Acording to Harter, coriander is named in an Egyptian papyrus dating that lists medicinal plants since 1550 B.C. (Harten, 1974). Recent evidence suggests that coriander fruits were found in the tomb of Tutankhamun, and were common in other graves in ancient Egypt at that time. The ancient Greek and Latin also recorded the coriander's medical uses in literature (Manniche, 1989).

From countries to countries, we can find the coriander in many traditional remedies for different diseases (Azhar and Mazhar, 2003; Duke, 1992; Eddouks et al., 2002; Grieve, 1971; Nadkarni, 1976; Usmanghani et al., 1997). Generally, people use coriander as a diuretic (Azhar & Mazhar, 2003; Duke, 1992; Eddouks, Maghrani, Lemhadri, Ouahidi, & Jouad, 2002; Grieve, 1971; Nadkarni, 1976; Usmanghani, Saeed, & Alam, 1997) or to cure the diseases related to the digestive system. For example, in India, due to the diaphoretic, diuretic, carminative and stimulant activity of coriander, people use it to cure the disorders of digestive, respiratory and urinary systems. It is also recommended to treats other diseases like urethritis, cystitis, urinary tract infection, urticaria, rash, burns, sore throat, vomiting, indigestion, nosebleed, cough, allergies, hay fever, dizziness and amoebic dysentery (Grieve, 1971). In Morocco, locally known as "Maadnouss," coriander has been documented as a traditional treatment of diabetes, indigestion, flatulence, insomnia, renal disorders and loss of appetite, and as a diuretic (El-Hilaly et al., 2003; Hassar, 1999; Hmamouchi, 1999).

The different parts of coriander are still used for purpose in folk medicine. In Iranian folk medicine, Coriander fruits have been used as a folk medicine for the relief of anxiety and insomnia, coriander leaves is to mask or disguise the tastes of other medicinal compounds or to calm the irritating effects on the stomach that some medicines cause, such as their tendency to cause gastric or intestinal pain. Coriander is a commonly used domestic remedy, valued especially for its effect on the digestive system, treating flatulence, diarrhea and colic. It settles spasms in the gut and counters the effects of nervous tension. Beside, Coriander fruits are also used as a diuretic by boiling equal amounts of coriander fruits and cumin seeds, then cooling and consuming the resulting liquid. In holistic and some traditional medicine, it is used as a carminative and for general digestive aid. In

India, the fruits are also considered carminative, diuretic, tonic, stomachic, antibilious, refrigerant and aphrodisiac. They are used chiefly, however, to conceal the taste or smell of other ingredients in pharmaceutical preparations (Jansen and Wageningen, 1981), and to correct the gripping qualities of rhubarb and senna (Bhatnagar, 1950). Chinese folk medicine uses coriander leaves and seeds to help remove unpleasant odors occurring in the genital areas of men and women, as well as halitosis or bad breath. It also has been credited in Chinese herbalists to treat indigestion, anorexia, stomachache and treat influenza. In Vietnam, the coriander is also used as traditional treatment of stomach, digestive troubles and cough in folk medicine.

I.3.3 Use the fruits as a spice

Coriander is widely used as flavoring spice from South Asian cuisine to Scandinavian cuisine. The use of coriander in cooking is mentioned from ancient time. It is interesting to note that the ancient Egyptian hieroglyphs "venshivu" and "ounshavu" refer to the coriander plant, and "ounshi" is used to describe the fruits of this species (Sinskaja, 1969). The use of coriander of ancient Egyptians cannot be determined from literature, but the ancient Greek and Latin later recorded the use coriander's fruits in some dishes (Diederichsen, 1996). In such countries like Vietnam, China, coriander is an indispensable spice in the some traditional dishes. With it is highly flavor, it is highly regarded and then is widely spread from country to country. Due to this spread, in some countries, it is named after the region where people learn the way to use it in cuisine. For example, English calls coriander is "Chinese parsley", or in French it is named "persil arabe".

In Russia and Scandinavia, coriander fruits is wildly use in whole of plant or power for flavouring purpose. The most coriander goes into curry powder, of which it forms 25-40% (Purseglove, 1981), and this powder is used to flavor liqueurs as being an important flavouring agent in gin production (Jansen and Wageningen, 1981). The spice is also employed for the preparation of either the steam-distilled essential oil or the solvent-extracted oleoresin. Both products could be used in the flavouring and aroma industries. Essential oils could be fractionated to provide linalool (usually 60 to 70 per cent), which could be used as a starting material for synthetic production of other flavouring agents, such as citral and ionone. Beside, the fruits are also used in the preparation of baking, sausages, pickles, candies, sauces, soups, cookies, buns and cakes, and tobacco products. In Russian, it is easy to find spice of coriander in the famous bread. In Ethiopia, a spice of

coriander is widely used to add flavor for meat and vegetarian dishes. In Germany, coriander spice combined with a few other ingredients to be used as a flavor beer which increased its inebriate effect. The ancient Egyptians made the same use of the plant in wine-making, and candied coriander fruits were once popular (Diederichsen, 1996).

The dried fruits have different taste compare to the fresh leaves. When crushed, the fruits have a lemony citrus flavor described as warm, nutty, spicy, and orange-flavored. In cuisine, fruits could be used both in dried fruits and in ground form. Because of the quick lost of aroma after grinding, the fruits are usually stored in the dried form without any processing (Ivanova, 1990; Prakash, 1990). Today, the use of coriander's fruits as a spice is very popular in many of area of world. In some regions, the use of the word coriander in food preparation always refers to these fruits (as a spice), rather than to the plant itself, and is an important item of international trade. In Russia and Scandinavia, coriander's fruits are wildly use in whole of plant or powder for flavoring purpose. The most coriander goes into curry powder, of which it forms 25-40% (Purseglove, 1981), and this powder is used to flavor liqueurs as being an important flavoring agent in gin production (Jansen and Wageningen, 1981). The spice is also utilized for the preparation of either the steam-distilled essential oil or the solvent-extracted oleoresin. Both products could be used in the flavoring and aroma industries. Essential oils could be fractionated to provide linalool, which could be used as a starting material for synthetic production of other flavoring agents, such as citral and ionone. Moreover, people also use coriander's fruits in the preparation of baking, sausages, pickles, candies, sauces, soups, cookies, buns and cakes, and tobacco products.

I.3.4 Use the green part as a spice and vegetable

All parts of the coriander plant are edible, for example in Thailand, even the coriander's root is used as a vegetable in a cultivar of cuisine (Wikipedia), but the fresh leaves and the dried fruits are the parts most traditionally used in cooking. Because the flavor is diminishes when heated, coriander fresh leaves are usually used raw or added to the dish just before serving. In Indian and Central Asian recipes, coriander leaves are used like vegetable and cooked until the flavor diminishes. The leaves spoil quickly when removed from the plant, and lose their aroma when dried or frozen (source: Wikipedia - Coriander), but luckily nowadays people can easily find fresh coriander in the market, even with large scale, especially in ethnic market which day by day becomes more popular (Simon, 1990).

I.3.5 Use of essential oil of the coriander fruits

From the 1885s, the first factory to be built in the Vorone district in Russia for the purpose of distilling essential oils of coriander. In this regard, Coriander become an important agricultural crops of this area and it still is a major manufacturer of coriander for commercial purpose (Diederichsen, 1996). In industry, essential oil of coriander is principally used in industry such as aromatherapy, food flavors, and in the liquor, cocoa and chocolate industries. Besides, it is also employed in medicine as a carminative or as a flavouring agent. As far as we know, the concentration of linalool, the major component of essential oil of coriander, varies mostly between 50 to 70% (Coşkuner and Karababa, 2007; Illés et al., 2000; Zoubiri and Baaliouamer, 2010). Linalool is one of the most wildly use as scent in 60- 80% of perfumed hygiene products and cleaning agent including soaps, detergent, shampoos and lotions. Thus, essential oil of coriander will be widely used as a potential source of further technical processing. Today, oleochemically synthesized linalool is usually used in the non-food sector, as it is cheaper at present. The demand for essential oils is rising in Western countries, and the full potential of this use of coriander has not yet been recognized (Simon, 1990).

I.3.6 Use of the fatty oil of the coriander fruits

Fatty Oil is rich in monounsaturated fatty acids and is derived from coriander fruits by pressing or by solvent extraction. Petroselinic is major component of the fatty oil of coriander (up to 70 - 80% at full maturity) (Kiralan et al., 2009; Moser and Vaughn, 2010; Msaada et al., 2009a; Msaada et al., 2009b; Sriti et al., 2010a; Sriti et al., 2010b). Petroselinic acid (C18:1(6c)) is an isomer of the usual oleic acid (C18:1(9c)) and in petroselinic acid, the single double bond has a different position. This acid has peculiar physicochemical properties that make it potentially suitable for many industrial applications, e.g. this acid can be oxidative cleaved to produce a mixture of lauric acid, a compound useful in the production of detergents, and adipic acid, a C_6 dicarboxylic acid which is utilized in the synthesis of nylon polymer or oleochemical raw material (Diederichsen, 1996; Msaada et al., 2009a; Msaada et al., 2009b; Sriti et al., 2010a). Besides, fatty oil from coriander fruits has recently been labeled as a novel food ingredient by the European Food Safety Authority. It is now considered as safe to be used as a food supplement for healthy adults, at a maximum level of 600 mg per day (i.e. 8.6 mg/kg bw per day for a 70 kg person), which

would lead to significantly higher intakes of coriander fruit oil and petroselinic acid than current background intakes. Therefore, the development of a new process for extracting vegetable oil from coriander fruits is a major challenge for the years to come.

II. Vegetable Oil

II.1 Lipids

Lipids are group of molecules that includes fats, oils, waxes, sterols, fat-soluble vitamins (such as vitamins A, D, E, and K), monoglycerides, diglycerides, triglycerides, phospholipids, and some others; Lipids are also important constituent of of the diet because they are a source of high energy value. Lipids play an important role in physiology and pathophysiology of living systems. The structure of some common lipids is represented in Figure 7. Lipids are generally hydrophobic and soluble in organic solvents in nature. However, lipid molecules show a remarkable structural and combinatorial diversity unlike other biological molecules such as nucleic acids and proteins. Chemical structure of the fat will vary if there are changes in the arrangement of chemical space. For example, sterol lipids are characterized by a four fused ring template consisting of three six membered rings and one five membered ring. Alternatively, Glycerolipids typically do not contain any rings and contain radyl chains attached to carbons on glycerol group. The radyl chains may be further unsaturated with varied double bond positions and geometry adding to the structural heterogeneity of lipids. The main biological functions of lipids include storing energy, signaling, and acting as structural components of cell membranes (Subramaniam et al., 2011).

II.1.1 Fatty acid

Fatty acid is a large group of organic acids, especially those found in animal fats and oils and plants. In chemistry, particularly in biochemistry, a fatty acid is a carboxylic acid with a long aliphatic tail (chain), which is either saturated or unsaturated. Fatty acids may be defined as organic acid that occur in a natural triglyceride and is a monocarboxylic acid ranging from C4 to C28 atoms in straight chains and will usually have either a saturated hydrocarbon chain or may contain from one to six double bonds. When they are not attached to other molecules, they are known as "free" fatty acids. Fatty acids are important sources of fuel because, when metabolized, they yield large quantities of ATP. Many cell

types can use either glucose or fatty acids for this purpose. In particular, heart and skeletal muscle prefer fatty acids. Despite long-standing assertions to the contrary, fatty acids can be used as a source of fuel for brain cells, at least in some rodents (Ebert et al., 2003; Marin-Valencia et al., 2013).

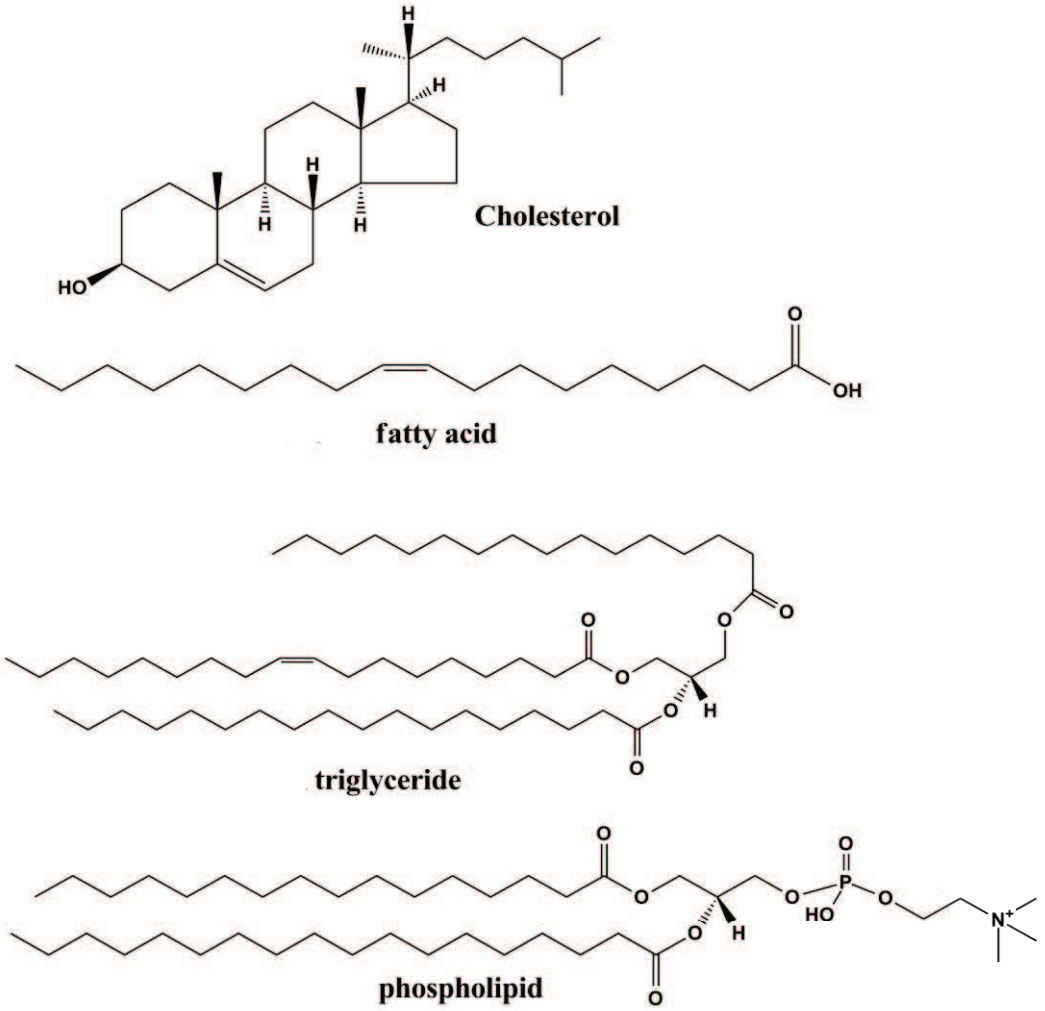


Figure 7: Structure of some common lipids

II.1.2 Definition

Fatty acid is a molecule formed of a hydrocarbon chain terminating with an acid group (COOH). In plant foods, the carbon chain rarely has more than 18 carbon atoms. In animal food and in our bodies, the carbon chain can reach more than 30 carbon atoms. This is may be due to the intervention of sometimes metabolic pathways.

There are three types of fatty acids: (i) Saturated fatty acids; (ii) Monounsaturated fatty acids with one double bond and (iii) Polyunsaturated fatty acids with two or more double

bonds. Fatty acids are the major components of the different classes of lipids that are triglycerides, phospholipids and cholesterol esters. Triglycerides represent 95 to 98% of ingested dietary fats. They consist of a molecule of glycerol esterifies with three fatty acids. In the body, triglycerides, located primarily in adipose tissue, is the principal storage form of energy.

II.2 Biosynthesis of fatty acids

The fatty acid biosynthesis in plants is the level of plastids. It takes place in several phases: activation, phase formation / elongation, desaturation phase and a transmission phase. The formation of fatty acids is initiated in the chloroplasts of vegetative tissues or in the plastids of non-green tissue such seeds (Ohlrogge et al., 1979). Sucrose is the carbon pool used for the synthesis of fatty acids. Converted to glucose molecule, the latter enters glycolysis to give a molecule of acetylCoA, acyl chain at the origin of fatty acids. In effect, there are two sources of production of acetyl-CoA. The first drift Non chlorophyllous cells, fruits plastids that contain all the enzymes necessary for the reactions belonging to the glycolysis, as the pyruvate dehydrogenase (Reid et al., 1977). The second involves the pentose phosphate pathway. Both mechanisms are used to provide the reducing power (NADH, H^+ and NADPH, H^+), energy (ATP) and glucose necessary for the synthesis of fatty acids (Browse and Somerville, 1991).

II.2.1 Activation phase

The first reaction involves acetyl-CoA carboxylase (ACCase) whose substrate is acetyl-CoA to give malonyl-CoA in the presence of CO2 and ATP (Rawsthorne, 2002). The malonyl-CoA is a precursor for the *novo* synthesis of fatty acids. According Roesler et al. this enzyme is activated by the light, and it may be involved in regulating of formation of fatty acids (Roesler et al., 1997).

II.2.2 Phase formation/longation

This phase takes place in cycles, by way of an enzymatic complex multipolypeptidique commonly grouped under the term "fatty acid synthase" (AGS). This reaction requires ATP, malonyl group is transferred to a protein cofactor, the ACP (acyl carrier protein) with a malonyl-CoA: ACP transacylase which allows obtaining of malonyl-ACP. The latter will be used as carbon donor in every other elongation reactions. The first condensation takes place between a acetyl CoA (or acetyl-ACP) and malonyl-ACP by a major condensation of three enzymes (enzymes in the Fatty Acid Synthetase or acyl synthetase): 3-Ketoacyl-ACP syntheses (KAS) III (Jaworski et al., 1989). Then, 3-kétobutyryl-ACP formed various changes (reduction, dehydration and a final reduction) by the action of enzymes AGS. This last step to add two carbons so cyclic until a string of 16 and 18 carbons.

II.2.3 Phase desaturation

This phase allows the introduction of a double bond in the acyl chain fatty acid formation with different enzymes desaturation involved by the use of fatty acids in the plant cell. The location of these enzymes determines the type of substrate used (Murphy et al., 1985). Thus, the soluble desaturases exclusively located in the stroma modify the fatty acids linked to an ACP, while those connected to the membrane acting on the free fatty acid or linked to other complexes of biosynthesis of lipids (phosphatidylcholine, phosphatidylglycerol, monogalactosyl, diacylglycerol).

During this phase stearic acid (C18: 0-ACP) sudden desaturation which converts oleic acid (C18: 1-ACP). This reaction is done with an enzyme stearoyl-ACP Δ9 desaturase (McKeon and Stumpf, 1982) which is responsible for the introduction of a double bond between carbons 9 and 10. On the other hand, this enzyme uses as ferredoxin electron donor (Stumpf, 1984) and requires the presence of molecular oxygen, reducing power via the photosystems I and II (NADPH, H^+) and ferredoxin activity oxidoreductase. Two substrates are preferred for this enzyme: stearoyl-ACP and the palmitoyl-ACP. With a maximum speed of enzyme activity, stearoyl-ACP is the first substrate it uses. This is reason why palmitic acid found in plants triacylglycerol compared to stearic acid (McKeon and Stumpf, 1982). This enzyme is essential for the production of unsaturated fatty acids

membrane lipids as well as those related to triacylglycerol in lipid reserves. According (Kabbaj et al., 1996), there are several genes or isoforms of this enzyme . These isoforms could coexist in the same fabric and be activated independently according to the period of the cycle (Cahoon and Ohlrogge, 1994). These genes have a regulatory environment type and time and their expression depends on the fabric, the stage of development embryo (Slocombe et al., 1994).

The Δ12 desaturase oleoylphosphatidylcholine is the main enzyme desaturation in the fruits and is localized to the membrane of the endoplasmic reticulum. This position involves a movement of the previously localized oleic acid at the flat, into the cytosol. This transfer is done via a thioesterase enzyme which is responsible for the hydrolysis of the ACP of oleic acid to which it is linked. The Δ12 desaturase enzyme catalyzes the formation of a double bond to oleic acid between carbon 12 and 13. It has a important role since it allows the synthesis of polyunsaturated fatty acids in the large majority of oils. The main substrate of the enzyme is oleic acid bound to a lipid phosphatidylcholine cytosolic (Murphy et al., 1985). In soybean, there are two enzyme systems of desaturation involved in the biosynthesis of unsaturated fatty acids (Heppard et al., 1996). One constituent is the whole plant, while the other has a specific expression seed. Biosynthesis of fatty acids is represented in Figure 8.

II.2.4 Transport phase

Transport of saturated fatty acids to the cytosolic compartment is effected by means of the acyl-ACP thioesterase enzyme which is localized in the inner membrane of the plastid (Voelker, 1996). Thus a large portion of the palmitic acid (C16: 0) is converted into C18: 0-ACP desaturated C18: 1-ACP. Stearic acid bound to the ACP (C18: 0-ACP) is very little exported from the chloroplast, palmitic and oleic acids are the major substrates of the reactions taking place in the other compartments. These two fatty acids are supported by acylCoA synthase to form acylCoA-C18: 1 and C16: 0-CoA and enter the synthesis pathway of glycerolipids (Stymne, 1987) allows the formation of all lipid compounds.

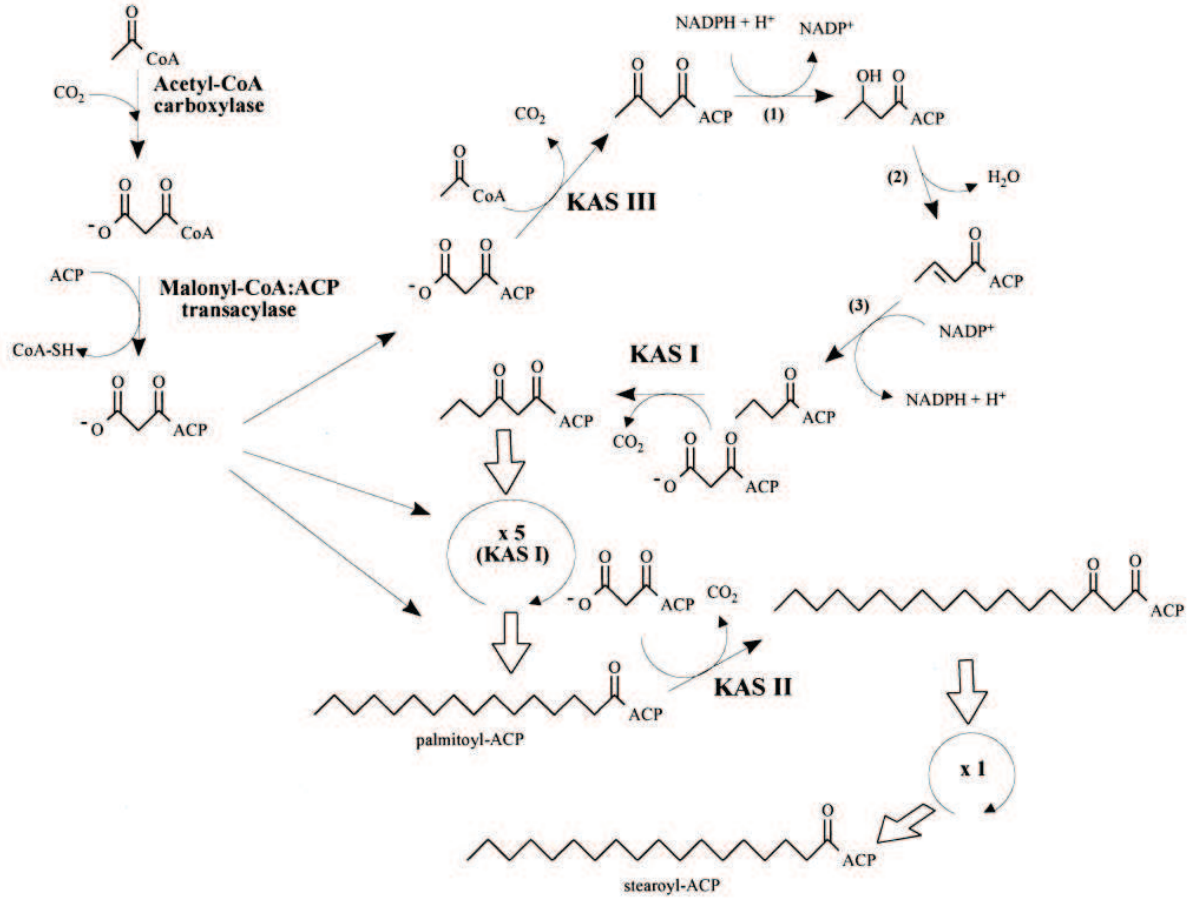


Figure 8: Biosynthesis of fatty acids by fatty acid synthesis (Salas et al., 2000). Abbreviations: ACP, acyl carrier protein; KAS (I, II and III), b-ketoacyl-ACP synthases; (1) b-ketoacyl-ACP reductase, (2) b-hydroxyacyl-ACP dehydrase, (3) enoyl-ACP reductase

II.3 Functions of Lipids

A lipid is a non-soluble molecular organic compound comprised of hydrogen and carbon, lipids has no single common structure. The most commonly occurring lipids are triglycerides and phospholipids. Triglycerides are fats and oils Triglycerides have a glycerol backbone bonded to three fatty acids. If the three fatty are similar then the triglyceride is known as simple triglyceride. If the fatty acids are not similar then the fatty acids are known as mixed triglyceride. Phospholipids contains glycerol and fatty acids, they also contain phosphoric acids and a low-molecular weight alcohol. Common phospholipids are lecithins and cephalins and they are found in membranes of animal and plants. The structure of these molecules determines their function: (i**)** Triglycerides: Triglyceride molecules are made from three molecules of fatty acids and one glycerol

molecule. Triglycerides are able to float in a cell's cytoplasm since they have a lower density than water and are non-soluble, as is the case with all lipids. Triglycerides are crucial in the body for energy storage. (ii) Steroids: Steroids have a structure that resembled four rings fused together which are made from carbon molecules. A few types of common steroids are cholesterol, testosterone, vitamin D2 and estrogen. Steroids benefit the body by helping determine and control the structure of plasma membrane. (iii) Phospholipids: Phospholipids earn their name as their constitution is primarily phosphate groups. They contain molecules that both attract and repel water, playing a key role in constituting cell membranes. (iv) Glycolipids: Short sugar chains form glycolipids, which can be found in a cellular membrane's exoplasmic surface. They play an important role in boosting the body's immune system. (v) Lipoproteins: A lipoprotein is a combination of proteins and lipids found in a cell's membrane, examples being antigens and enzymes. Lipoprotein help fat move around the body in the bloodstream and exist in the form of Low Density Lipoprotein (HDL) and High Density Lipoprotein (LDL). (vi) Waxes: Along with a chain of alcohols, fatty acids are found in waxes. These are extremely common lipids and can be found on animal feathers, in human ears and even on the leaves of plants. Their primary function is one of protection.

II.4 Oil Extraction

Up to now, there are two oil extraction processes: (i) mechanical process to extract oils by the press (e.g. extrusion); (ii) physicochemical process with solvent and then separated.

II.4.1 The mechanical process to extract oils by the press

A typical oil extraction process begins with fruits preparation. The preparation consists of a series of treatments, such as cleaning, breaking, grinding and cooking, which produces a feed material in the optimum condition for subsequent extraction (Evon et al., 2009). In the mechanical process, the pressing is executed an engine driven screw press, so optimization of the extraction procedure is key to the production of vegetable oils. Commonly, the extraction yield is lower when compared to the solvent extraction, because in fruits cakes still contain some oil. However, the obtained oil exhibits higher quality due the low level of compounds such as phospholipids and residual traces of solvent. Henning (Evon et al., 2010b) stated that engine driven screws extract 75-80% of the available oil and manual

ram presses only achieved 60-65%, but a good pretreatment of fruits might increase the yield of screw pressing up to 91%. The latter is schematically represented in Figure 9 (a).

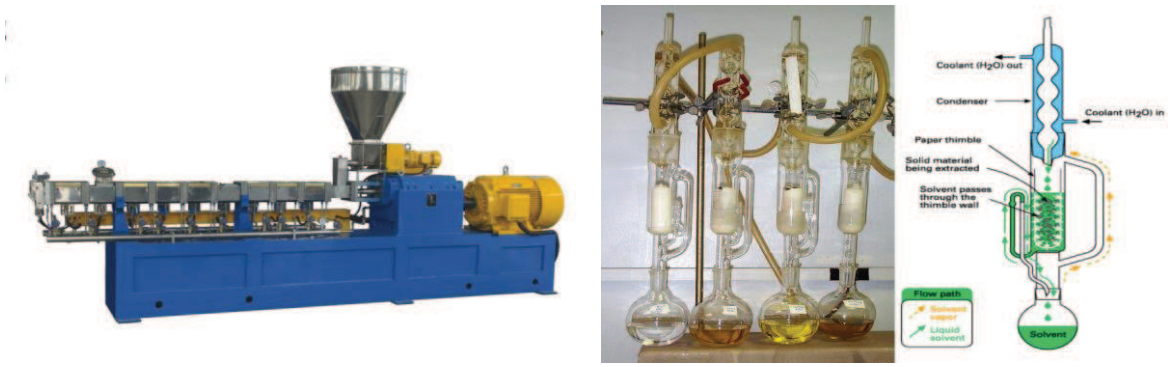


Figure 9: (a) Twin-screw extrusion scheme (left); **(b)** soxhlet extraction (right)

Up to now, only one previous study has dealt with the use of twin-screw extrusion 1 technology for vegetable oil extraction from coriander fruits by mechanical pressing (Sriti et al., 2012a). The single batch used in this study was obtained from Korba area (North East of Tunisia) and exhibited relatively low lipid content (only 21.9% of the dry matter). Both the screw rotation speed and the inlet flow rate of coriander fruits affected the oil extraction yield. The highest oil yield was obtained under operating conditions of 50 rpm and 2.3 kg/h, respectively. Nevertheless, it was never more than 45% and the residual oil content in the press cakes was at least 16.6% of the dry matter. At the same time, the filtrate's foot content, i.e. the solid particles forced through the filter, was always high (from 47.5 to 66.0%). Further, residual essential oil contents in the press cakes and their composition have not been discussed although they may form a valuable application of extrusion cakes. The impact of operating conditions on the fatty acid composition of pressed oils was less important. Ten fatty acids were identified, with petroselinic acid accounting for 66-75%. In conclusion, the use of the twin-screw extrusion technology for coriander oil mechanical pressing appears promising, even if the process efficiency should be improved. This study proposes to evaluate the effects of screw configuration, device's filling coefficient (i.e. the ratio of the inlet flow rate of coriander fruits to the screw rotation speed) and pressing temperature on oil extraction efficiency. It further suggests some potential applications for the obtained press cakes.

II.4.2 The physicochemical process with solvent

Nowadays, the use of n-hexane as the solvent for oil extraction is still the most common method. Higher yields can be obtained compared to mechanical extraction, with a goal of 0.5 % residual oil in the meal, and the operating costs are lower, although solvent extraction brings about a high initial capital cost. Nearly all known oil seed extraction plants are currently using hexane (Conkerton et al., 1995; Evon et al., 2013; Proctor and Bowen, 1996; Wan et al., 1995). This process is represented in Figure 9 (b).

III. Essential Oil

III.1 General

Essential oils are natural, volatile, complex compounds characterized by a strong fragrance and are formed by aromatic plants as secondary metabolites products (Bakkali et al., 2008; Baser and Buchbauer, 2009; Burt, 2004). They have a low solubility in water but are soluble in fats, alcohol, organic solvents and other hydrophobic substances and are generally liquid at room temperature. From the 16^{th} century, the term "essential oil" is thought to derive from the name coined by the Swiss reformer of medicine, he named the effective component of a drug *Quinta essential* (Guenther, 1948).

Since ancient times, essential oils are recognized for their medicinal value and they are very interesting and powerful natural plant products and until now they continue to be of paramount importance. They have been used as perfumes, flavors for foods and beverages, or to heal both body and mind for thousands of years (Adumanya et al.; Baris et al., 2006; Margaris et al., 1982; Wei and Shibamoto, 2010). According to Bakkali et al., they have been largely employed for their properties already observed in nature, i.e. for their antibacterial, virucidal, antifungal and insecticidal activities and cosmetic applications, especially nowadays in pharmaceutical, sanitary, cosmetic, agricultural and food industries (Bakkali et al., 2008). Currently, an estimated 3000 essential oil are known, of which about 300 are commercially important destined chiefly for the flavours and fragrances market (Van de Braak and Leijten, 1999) and some plant families are particularly well known for their oil-bearing species. These include Apiaceae, Asteraceae, Cupressaceae, Clusiaceae, Lamiaceae, Lauraceae, Fabaceae, Liliaceae, Myrtaceae, Pinaceae, Piperaceae, Rosaceae, Rutaceae, Santalaceae, Zingiberaceae and Zygophyllaceae (Baser and Demirci, 2007;

Figueiredo et al., 2008). The tree diagrams for essential oil there are four broad sector - flavor in dustries, personal care, pharmaceutical and industrial (Figure 10)

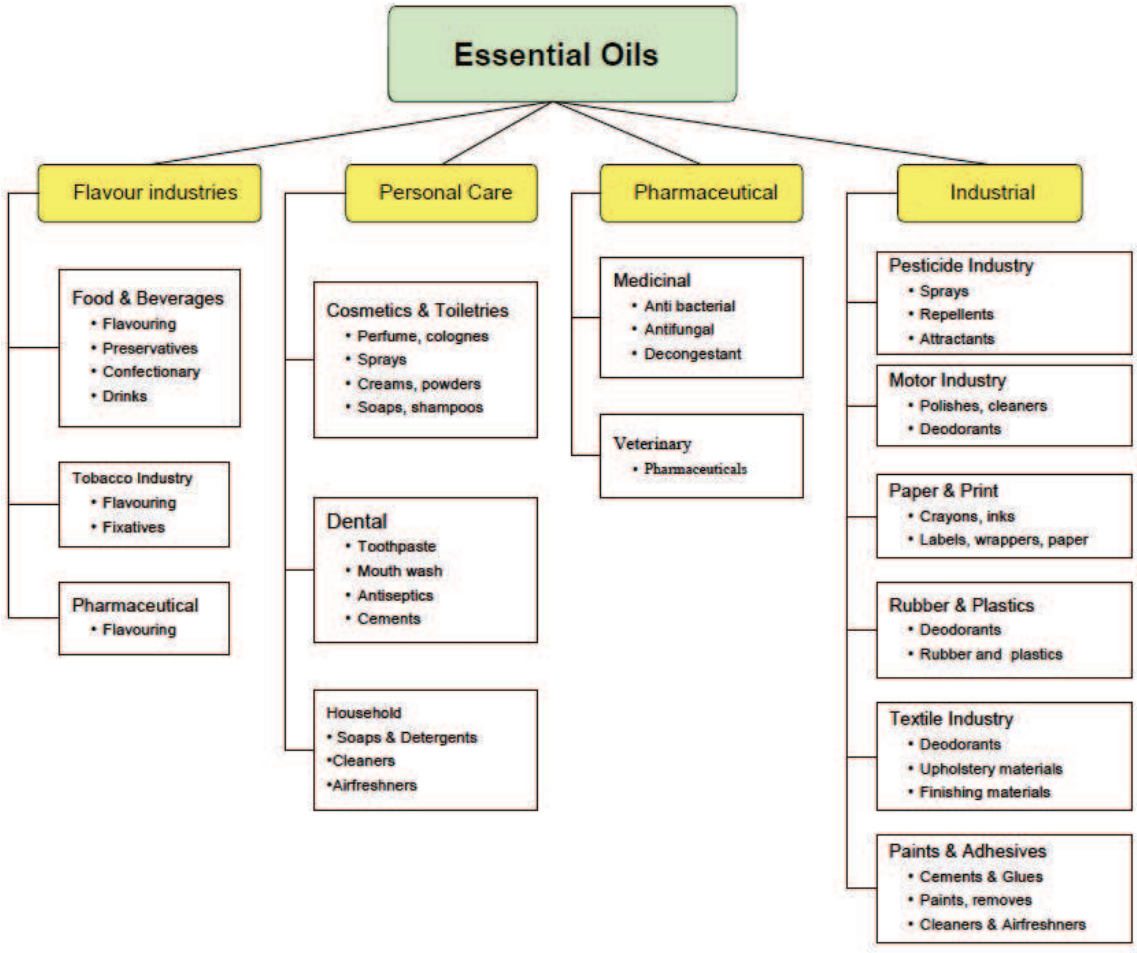


Figure 10: Industries and product categories that use essential oil (Brand-Williams et al., 1995)

In the plant, essential oil can be stored in specialized in special brittle secretoty structures known as secretory cells, secretory cavities, oil cells or ducts, resin ducts, glands or trichomes (glandular hairs) (Baser, 2002; Baser and Demirci, 2007; Pengelly and Herbals, 2004), and they are found in various parts of plant such as buds, flowers (oregano), leaves (lemongrass, eucalyptus), stems, twigs, roots (vetiver), rhizomes (Acorus), fruits (anise) or seeds (caraway); wood (rosewood, sandalwood), bark (cinnamon) (Baser and Demirci, 2007; Burt, 2004; Pengelly and Herbals, 2004). The majority of essential oils are obtained from vegetable raw material in different ways. There are different methods, depending upon the stability of the oil, for the extraction of the oil from the plant materials. Steam distillation and hydrodistillation are still in use today as the most important processes for

obtaining essential oils from the plants (Baker et al., 2000; Kulisic et al., 2004; Surburg and Panten, 2006). Other methods employed for isolation of essential oils include the uses of liquid carbon dioxide or microwaves, low or high pressure distillation employing boiling water or hot steam (Bousbia et al., 2009; Lahlou, 2004; Pengelly, 2004; Van de Braak and Leijten, 1999). The essential oils obtained by steam distillation or by expression are generally preferred for food and pharmacological applications. There is plenty of literature on the characterization of essential oils. In the most cases, capillary gas chromatography (GC), GC with flame ionisation detection (FID), or GC coupled with mass spectrometers (MS) are the methods of choice for quantitative determinations (Msaada et al., 2009d; Neffati et al., 2011; Nejad Ebrahimi et al., 2010; Smallfield et al., 2001; Sriti et al., 2011b; Telci et al., 2006a; Telci et al., 2006b; Yildirim and Gok, 2012). Alternatively, Kovats indices, determined by co-injection of the oil with a homologous series of n-alkanes, are also widely used to identify compounds, where authentic standards are not available (Juliani Jr et al., 2002).

III.2 Terpenes

Essential oils are made up of three elements almost exclusively carbon, hydrogen, and oxygen. By far the most common component class is the terpenes. Terpenes, also known as isoprenes, and a terpene containing oxygen is called a terpenoid. Terpenes are not only the largest group of plant natural products, comprising at least 30,000 compounds (Breitmaier, 2006; Dubey et al., 2003; Hill et al., 1991), but also contain the widest assortment of tructural types.

III.2.1. Terpene biosynthesis

Terpenes are a family of widely used compounds in the reign plant. Their most important structural feature is the presence in them of several skeleton at least two isoprene units to 5 carbon atoms each (C5H8). This isoprene is the basis of the concept of "isoprene rule" in 1953 by Ruzicka (Sriti et al., 2012a) and supplemented by Lynen (Isobe et al., 1992) and Bloch (Wang and Johnson, 2001). This rule considers isopentenyl diphosphate, referred to as isoprene asset name, as the true precursor of the terpene molecule, hence the name under which isoprenoid also designates them. Two paths lead to the formation of

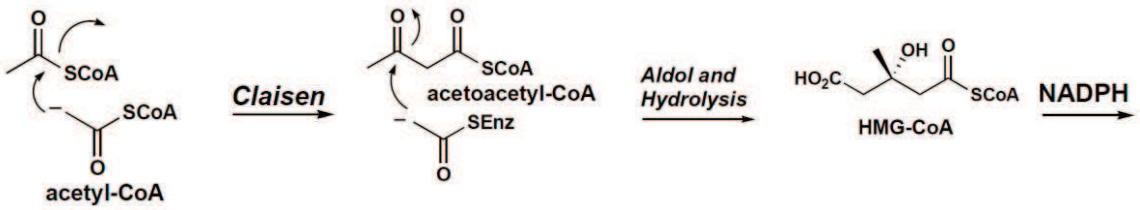
isoprenoid: the mevalonate pathway and route Rohmer (Crowe et al., 2001). The first takes place in the cytoplasm (Dewick, 1999) and the second channel takes place in the plastids (Figure 11).

* Mevalonate pathway

The biosynthesis of isopentenyl diphosphate (IPP) is done by means of the mevalonic acid from acetyl CoA. According to some studies (Dewick, 1999), it appears that the biosynthesis of isopentenyl diphosphate (IPP) takes place in the cytoplasm. Thus, due to the condensation of three acetyl-CoA is formed hydroxy-3-methylglutaryl-CoA (HMG-CoA), which subsequently is converted to mevalonate in an irreversible manner with a HMG-CoA reductase (HMGR). Mevalonate then undergoes phosphorylation by the action of two soluble kinas, mevalonate kinas, and phosphomevalonate kinase, for form mevalonate 5-diphosphate-5. The latter is decarboxylated by Pyrophosphomevalonate decarboxylase to form isopentenyl pyrophosphate (IPP).

* DOXP pathway

The route begins with the condensation of a pyruvate molecule with one molecule of glyceraldehyde-3-phosphate which results in the formation of 1-Deoxy-D-xylulose 5-phosphate (DOXP). The latter undergoes isomerization and reduction leading to 2-C-methyl-4-phosphate-D-erythritol. It then forms the cytidine diphosphate. C2 phosphorylation and cyclization lead to 2-C-methyl-erythritol 2,4-cyclodiphosphate which give the 4-hydroxy-DMAPP and IPP and finally the dimethylallyl diphosphate (DMAPP) (Dufaure et al., 1999). The IPP biosynthesis reactions from DOXP are in plastids. IPP formed is used for the biosynthesis of mono-, di- and tetraterpenes (C10, C20 and C40 respectively) (Kartika et al., 2005, 2006).



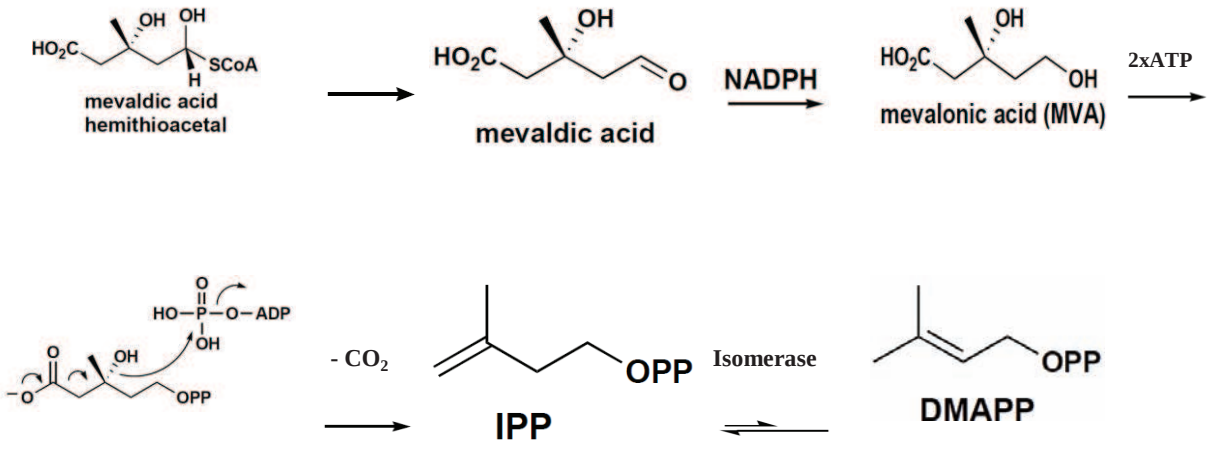


Figure 11: The diagram of the mevalonate pathway (Lange et al., 2000)

III.2.2 Classification

The name 'terpene' comes from the fact that the first described members of this class were isolated from turpentine, the monoterpene rich liquid obtained from the resin of various *Pinus* spp (Breitmaier, 2006). Terpenes are made from combinations of several 5-carbon-base (C5) units called isoprene. Terpenes can form building blocks by joining together in a "head-to-tail" configuration to form mono-, sesqui-, diterpene and larger sequences (Pinder, 1960) . The chief terpenes are the monoterpenes (C10) and sesquiterpenes (C15) and in some cases hemiterpenes (C5), diterpenes (C20), triterpenes (C30) and tetraterpenes (C40) also exist as Table 2 list below. At the present time, hundreds of different monoterpene (Dewick, 1999), sesquiterpene (Fraga, 2006), diterpene (Hanson, 2000) and triterpene (Connolly and Hill, 2008) carbon skeletons are known.

Table 2: Structure of Terpens

Terpene Classification	Number of Isoprene Units	Number of Carbon Atoms
Monoterpenes	2 Isoprene	10 carbon atoms
Sesquiterpenes	3 Isoprene	15 carbon atoms
Diterpenes	4 Isoprene	20 carbon atoms
Triterpenes	6 Isoprene	30 carbon atoms
Tetraterpenes	8 Isoprene	40 carbon atoms

III.2.2.1 Monoterpenes

Monoterpene compounds are presented in nearly all essential oils and are formed from two isoprene units with at least a double bond (structure of ten carbon atoms). While hemiterpenes only contain one such unit. They quickly react to air and heat and consequently lack stability and long shelf life as they are quickly oxidized. Monoterpenes are found in conifers, citrus, herbaceous plants as well as vegetables and fruits (Figure 12)

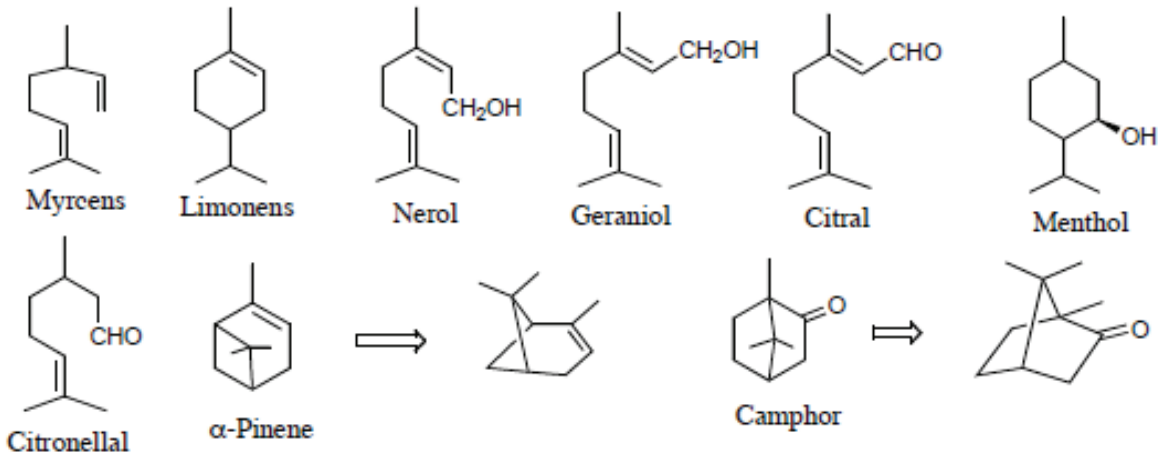


Figure 12: Monoterpene structures

III.2.2.2 Sesquiterpenes

Sesquiterpenes consist of 15 carbon atoms or 3 isoprene units linked to each other, head to tail. Mono- and Sesquiterpenes are the most abundant classes in essential oils. This formation can produce more than 300 different hydrocarbon sesquiterpenes. Sesquiterpenes have great diversity in construction containing up to four carbocyclic rings. are found in a variety of plants and fruits, especially berries (Figure 13).

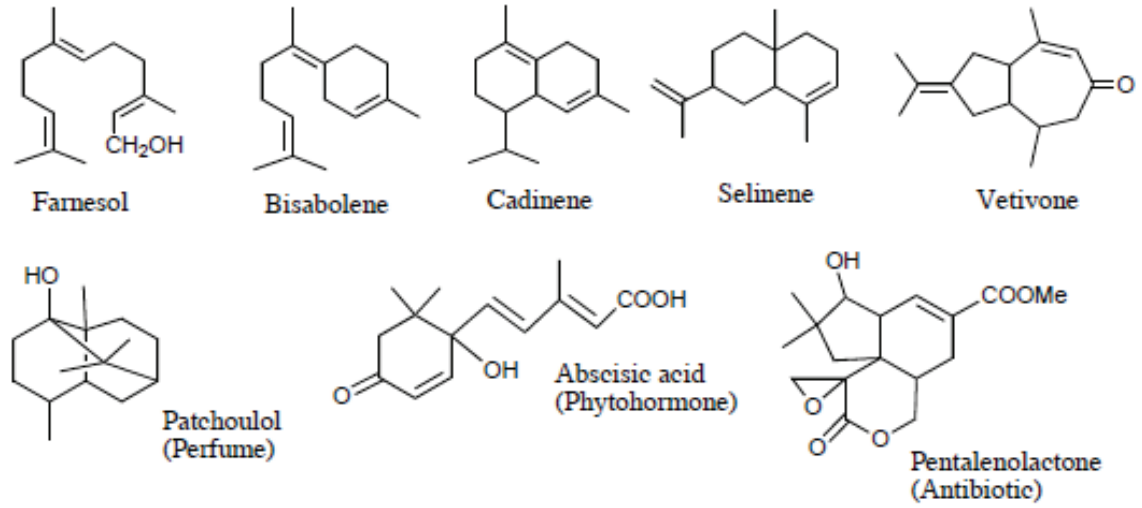


Figure 13: Sesauiterpenes structures

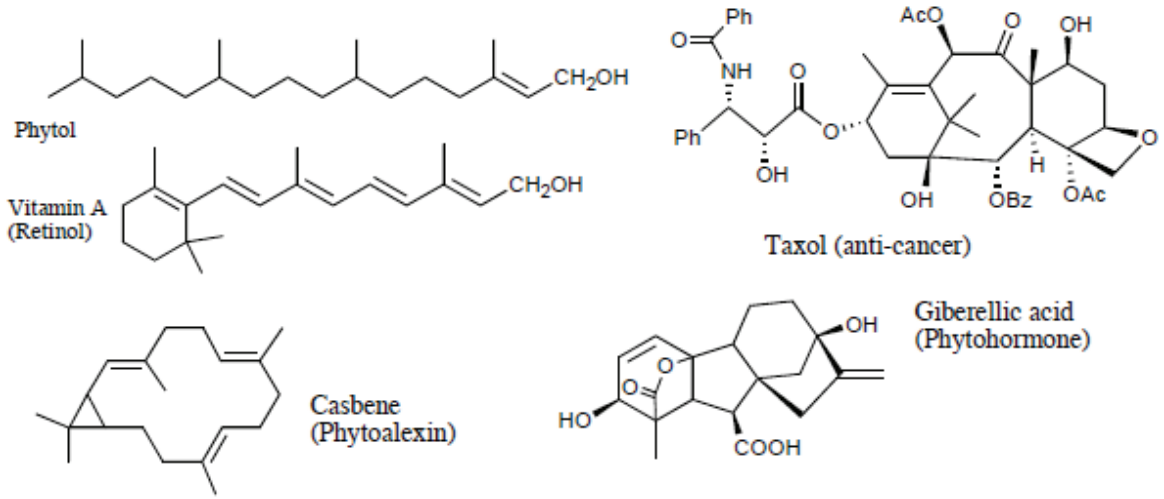


Figure 14: Diterpenes structures

III.2.2.3 Diterpenes

Diterpenes are less commonly found in essential oils, although there are some compounds that are more prominent. They have 20 carbon atoms (4 isoprene units) and are derived from geranylgeraniol pyrophosphate. They are of fungal or plant origin and are found in resins, gummy exudates, and in the resinous high-boiling fractions remaining after distillation of essential oils (Figure 14).

III.2.2.4 Triterpenes

Triterpenes consist of 30 carbon atoms or six isoprene units and have the molecular formula $C_{30}H_{48}$. More than 20,000 triterpenes have been isolated thus far. Among them, tetracyclic and pentacyclic triterpenes are the most abundant. Pentacyclic triterpenes are divided into many subgroups: gammaceranes, hopanes, lupanes, oleananes, ursanes, etc. based on their carbon skeleton. Triterpenes can be found in their free form (sapogenins), or bound to glycosides (saponins). Pentacyclic triterpenes are often bioactive (antitumor, antiviral, antidiabetic, antiinflammatory...) and present a huge therapeutic potential. A few compounds like corosolic acid (dietary supplement against diabetes) are already on the market and several others are under clinical trials or ready to be launched on the market. Triterpenes are also extremely common and are found in animals, plants, and fungi (Figure 15).

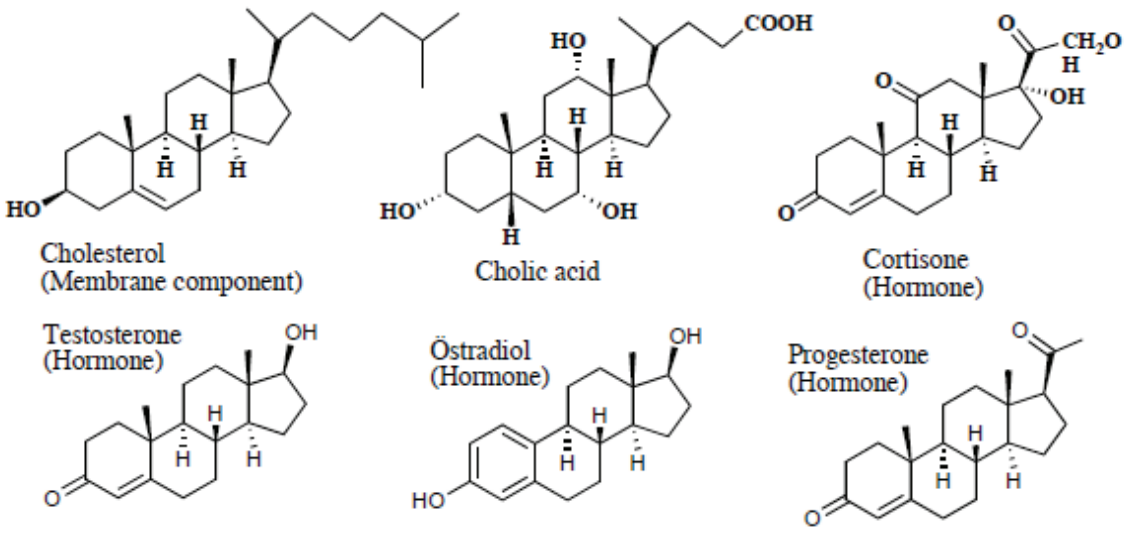


Figure 15: Triterpens structures

III.2.3 Function of Terepnes

Most terpenoids not have a direct fundamental role in the development of plant, but they play purely ecological roles, namely: (i) Attract insects for pollination and seed dispersal of pollen flowers; (ii) Protect against pathogens such as bacteria, fungi and viruses (Bourgaud et al., 2001); Inhibition of germination of neighboring plants (Langenheim, 1994; Moosavi et al., 2012; Yagoub and Hamed, 2013). (iii) Besides, terpenes are used as raw materials in

the manufacture of adhesives, emulsifiers and specialty chemicals on an industrial scale (Ramadan and Mörsel, 2002). (iv) Many monoterpenes are used in the prevention cancer in medicine (Horvath et al., 2006; Sriti et al., 2010b). This is due to the inhibition by monoterpenes of the proliferation of tumor cells, accelerate their rate of mortality and induction of tumor cell (Tahvonen et al., 2005).

III.2.4 Extraction

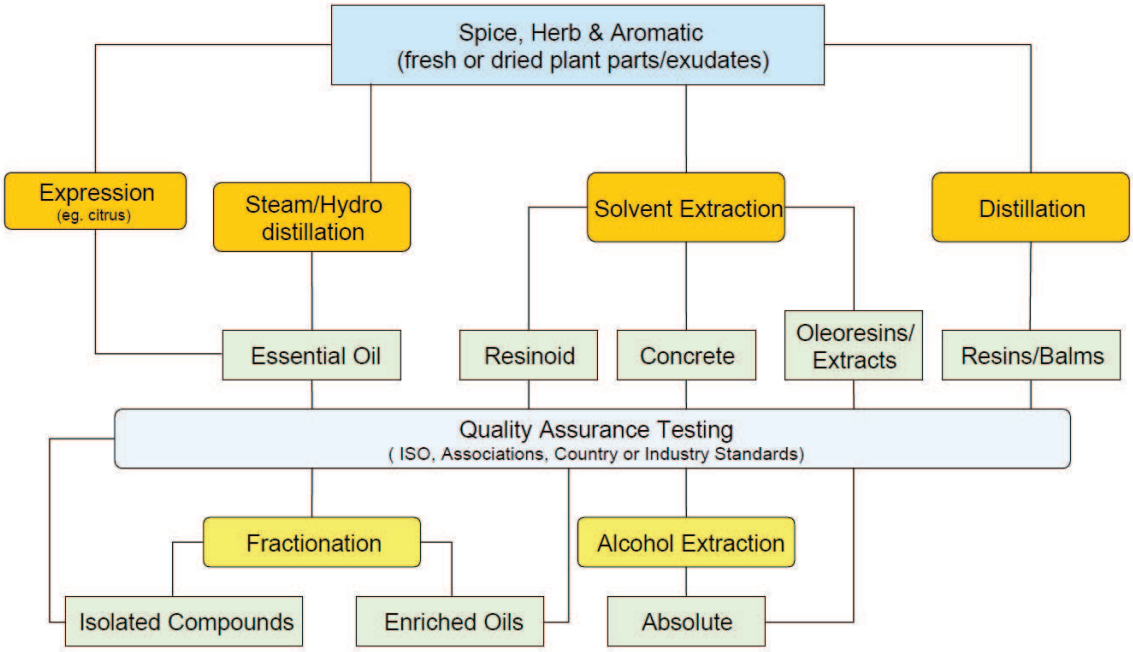


Figure 16: Extraction processes used and products from spice, herb and aromatic plants (Brand-Williams et al., 1995)

Essential oil are generally derived from plant material. Normally, several types of methods can be applied to extract essential oil: (i) Hydro-Water distillation; (ii) Water and Steam Distillation; (iii), Steam Distillation; (iv) Simultaneous Distillation Extraction and (v) Expression. These methods have become established in the essential oil industry (Figure 16). However, some volatile oils cannot be distilled without decomposition and thus are usually obtained by expression (lemon oil, orange oil) or by other mechanical means.

III.2.4.1 Hydrodistillation

Hydrodistillation is a widely used method and is still one of the most economical methods for extraction on essential oil, because of its simplicity, easy construction and more

complete productions. Hydrodistillation can be subdivided in three processes; water, water-steam and steam distillation, differing in the way water is presented to the sample. In order to isolate essential oils by hydrodistillation, the plant material are completely immersed in water and brought to a boil. Due to the influence of hot water and steam, the essential oil is freed from the oil glands in the plant tissue. At present, the most commonly equipment of essential oil extraction is Clevenger apparatus (Figure 17). Whish (Whish and Williams, 1996) found no difference between oil yields, when oils from Melaleuca alternifolia (tea tree) were produced via steam distillation and hydrodistillation, on both macro-, and semimicro-scales, while, Khanavi et al. reported better essential oils yield of *Stachys persica* and *S. byzantine* from hydrodistillation than steam distillation (Khanavi et al., 2004). Sefidkon et al. isolated the essential oils from the aerial parts of *Satureja rechingeri* by steam, hydro- and water steam distillation. The highest oil yield was obtained with hydrodistillation and the lowest with steam distillation (Sefidkon et al., 2007).

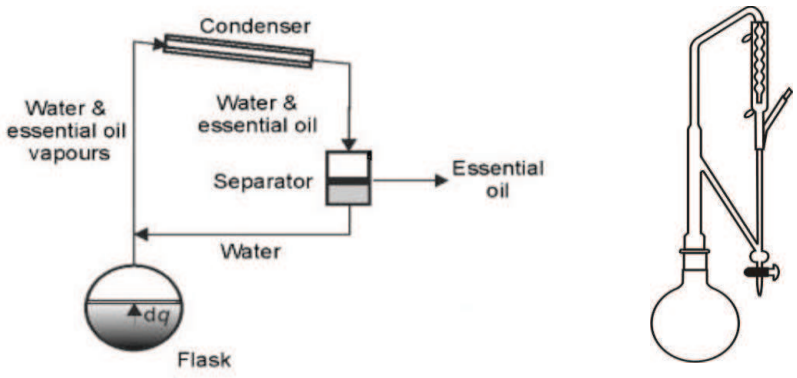


Figure 17: Hydrodistillation process (left) and Clevenger apparatus (right)

III.2.4.2 Water and Steam Distillation

As the name suggests, water and steam distillation is the process of distilling plant material with steam generated outside the still in a satellite steam generator generally referred to as a boiler. This method is frequently applied to produce essential oils and resolves some problems that occur with water distillation, such as thermal degradation of some compounds. It differs slightly from hydrodistillation, the plant material is placed at some distance above the water level on a perforated grid so it only comes into contact with the water vapor or steam arising from the till in during water-steam distillation. This process also exhibits a very low capital cost and a simple design and usually results in higher

quality oil than water distillation. However, water-steam distillation presents a lower capacity of plant.

III.2.4.3 Steam Distillation

Steam distillation is considered the most commonly applied extraction method for the production of essential oils on a large scale. This is gentle and productive method of essential oil extraction. The plants are not directly submerged in boiling water because at high temperatures the most subtle aromas might be altered. This provides some advantages such as a controllable flow and quantity of steam, making faster and thus more energy efficient distillation possible. However, commercial steam distillation apparatus is more expensive than hydrodistillation apparatus, limiting the wide application of steam distillation.

III.2.4.4 Simultaneous Distillation Extraction

This method is commonly to produce essential oil and resolves problems that occur with some plant materials have very low essential oil yield and it is rather difficult to collect real essential oil using hydrodistillation or steam distillation. In this case, the classic Likens-Nickerson apparatus is often used. This method is one popular method for preparation of volatile compounds and it can be applied in the fields of food, flavor and fragrance.

III.2.4.5 Expression

Expression or cold pressing, as it is also known, is only applied to obtain essential oils from orange or citrus peel. During this process, the term expression refers to any physical process in which the essential oil glands in the peel are crushed or broken to release the oil. This classical method is still frequently executed by means of rather primitive techniques, but the essential oil produced this way contains more of the fruit odor character than oil produced by any other method.

III.3 Antioxidant activities

Up to now, for antioxidant capacity measurement, many in vitro evaluation methods have been developed. The common methods as: DPPH radical scavenging assay, ABTS cation radical decolourization assay, Ferric reducing/antioxidant power (FRAP) assay, Oxygen radical absorbance capacity assay (ORAC) and Folin-Ciocalteu (F-C) assay is commonly

used to measure antioxidant capacity. For the best selection of methods to evaluate antioxidant capacity. According to Prior et al. proposed that proposed that three methods (Oxygen radical absorbance capacity assay - ORAC, Folin-Ciocalteu assay and Trolox equivalent antioxidant capacity assay) should be standardized for use in the routine quality control and measurements of antioxidant capacity of dietary supplements and other botanicals (Prior et al., 2005). In accordance with this proposal, antioxidant capacity of extract from coriander in our work will be employed for a comprehensive evaluation by ORAC assay, DPPH radical scavenging assay and Ferric reducing/antioxidant power (FRAP) assay.

IV. Conclusion

- ✓ Through research overview of coriander, the biological characteristics and ecology of coriander, the general concept of vegetable oil and essential oil is presented in this chapter. Besides, the previous studies on vegetable oil and essential oil in different parts of the coriander have also been mentioned. However, we recognize that the most previous studies have been made on one genotype in controlled condition, While our work is applied for eight genotype of different countries (coriander of France, Canada, Tunisia, Lithuania, Algeria China and Vietnam), in addition to studies of vegetable oil and essential oil of Vietnam and Lithuania coriander fruits not found before. For the studies the accumulation of vegetable oil and essential oil in the maturity stages, there are some studies have done e.g. (Msaada et al., 2009a; Ramezani et al., 2009; Telci et al., 2006a), but these studies of kinetic is only applied two years for their research while our results is applied for three year in conventional agriculture. That is an important results for research bioaccumulation of vegetable oil and essential oil in maturity stage in the future. Over the year, Sriti et al. (Sriti et al., 2012a; Sriti et al., 2012b; Sriti et al., 2011a) studied the optimization of operating conditions of extrusion by single and twin screw extruder (single crew OMEGA 20 and twin screw Clextral BC 21) in order to evaluate the feasibility of an processing of extraction fruit oil, but her works is only applied for Tunisia cultivar while in our work are studied in different cultivar (France, Tunisia, Lithuania, Vietnam), the results are compared in the same optimal

conditions for different cultivars of coriander (France, Tunisia, Lithuania and Vietnam), perhaps this results is important for scientific. That is the reason why we selected coriander in our research.

Chapter II: MATERIAL AND METHODS

I. Plant materials

I.1 The materials used for studying vegetable oil and essential oil component

The coriander (*Coriandrum sativum* L.) fruits were obtained from supermarket, seed company, retail shops of different European and Asian countries (eight cultivars - Canada, Tunisia, Lithuania, Algeria, China, French (FR1 - Dourou) and Vietnam (two sample - VS1 and VS2). After collected, these cultivars was cultivated in south-western France at the Regional Centre of Experimentation in Organic Agriculture at Auch 2011 (near Toulouse, south-west France; 43°38'47"N, 0°35'08"E). In addition to evaluate accumulation fatty acids of coriander during fruit ripening, the French coriander cultivar (Dourou) (GSN, Riscles, France) were conducted planted during the cropping season (2009, 2010 and 2011) can also be used for this studies. Furthermore, in order to assess the impact of different locations for vegetable oil and essential oil of coriander, we conducted planted coriander fruits of Vietnam at three location in 2012 (Ha Noi: 21° 4' 21''N, 105° 43' 45''E; Thai Binh: 20^o 25' 25''N, 106^o 20'29''E (Northern of Vietnam - Red River Delta) and Ha Tinh: 18^o 20' 120''N, 105^o 53'345''E). The plant materials harvested will be used to study essential oils and vegetable oil in this thesis.

I.2 The materials was used for extrusion

All trials were carried out using a single batch (200 kg) of coriander fruits 1 (GSN maintenaire cultivar), cultivated in the South West part of France and supplied by GSN Semences company (Le Houga, France). The moisture content of coriander fruits was 9.77±0.10% (French standard NF V 03-903). All solvents and chemicals were analytical grade and were obtained from Merck (Germany), Macherey-Nagel (Germany), Sigma-Aldrich (USA) and Prolabo (France).

II. Experiment methods

II.1 Extraction of Vegetable Oils

In our studies, the oil extraction from oil fruits is usually carried out through mechanical pressing with single-screw extruder (Model OMEGA 20, France) and twin-screw extruder (type Clextral BC 21), followed by solvent extraction with *n* hexane or cyclohexane.

II.1.1 Extraction by Solvent

For the vegetable oil content determination, the coriander oil was extracted from the initial material using the Soxhlet extraction apparatus with *n*-hexane or cyclohexane as extracting solvent. Before extraction, the dry materials (fruits, leaves, stem and roots) were ground in the electric grinder. Triplicates samples of 20 - 30g were subjected to conventional extraction for 5h. The solvent was removed by a rotary evaporator at low pressure and at 35 ^{o}C. The vacuum system was used to help to dry vegetable oil at 35 ^{o}C (overnight). The vegetable oil yield was determined.

II.1.2 Extraction by the extruder

Conventional industrial oil extraction from oil fruits is usually carried out through mechanical pressing with a hydraulic or single expeller press, followed by solvent extraction with *n* hexane. The hydraulic press is highly effective for industrial oil extraction from oil fruits by mechanical pressing, but this represents a discontinuous process. The last twenty years research has been focused on continuous oil extraction using extrusion technology (Crowe et al., 2001; Isobe et al., 1992; Singh et al., 2002; Wang and Johnson, 2001; Zheng et al., 2003). This process in a single-screw press is widely used for oil seeds, with a single-screw of variable pitch and channel depth, slowly rotating in a cage type barrel (Isobe et al., 1992). However, transport of material in this type of press mainly depends on friction between the material and the barrel's inner surface and screw surface during screw rotation. Thus, a solid, core component is often necessary to produce this friction, causing overheating, high energy consumption and oil deterioration. Furthermore, single screw presses provide insufficient crushing and mixing if they are not equipped with breaker bars or other special equipment. A twin-screw oil press can be expected to solve these problems because of its higher transportation force, similar to a gear pump, and better mixing and crushing at the twin-screw interface, thus improving mechanical lyses of the cells. In addition, twin-screw extruders have been shown to be significantly more energy efficient (Crowe et al., 2001; Isobe et al., 1992; Singh et al., 2002; Wang and Johnson, 2001; Zheng et al., 2003). Twin-screw extrusion technology has been increasingly used successfully to undertake mechanical pressing of various oil seeds (Amalia Kartika et al., 2006; Dufaure et al., 1999; Evon et al., 2007, 2009; Evon et al., 2010b; Guyomard, 1994; Isobe et al., 1992; Kartika et al., 2005). Therefore, it was applied for the extraction of

vegetable oil from coriander fruits, while aiming to obtain a high level of oil quality, extraction efficiency and feasibility. For this reason, the use of the twin-screw extrusion technology for coriander oil mechanical pressing appears promising, even if the process efficiency should be improved. This study proposes to evaluate the effects of screw configuration, device's filling coefficient (i.e. the ratio of the inlet flow rate of coriander fruits to the screw rotation speed) and pressing temperature on oil extraction efficiency. It further suggests some potential applications for the obtained press cakes.

II.1.2.1 Extraction by single-screw extruder

Extrusion was conducted by a single screw extruder (Model OMEGA 20, France) with a motor (0.75 kW of puissance, 230 V of maximal tension, 5.1 of maximal intensity), a screw length was 18 cm, a pitch screws of 1.8 cm, internal diameter was 1.4 cm and 0.5 cm deep channel and a sleeve of 2.5 cm of internal diameter equipped with a filter pierced outlet of liquid at the end the screw and a surface of nozzles.

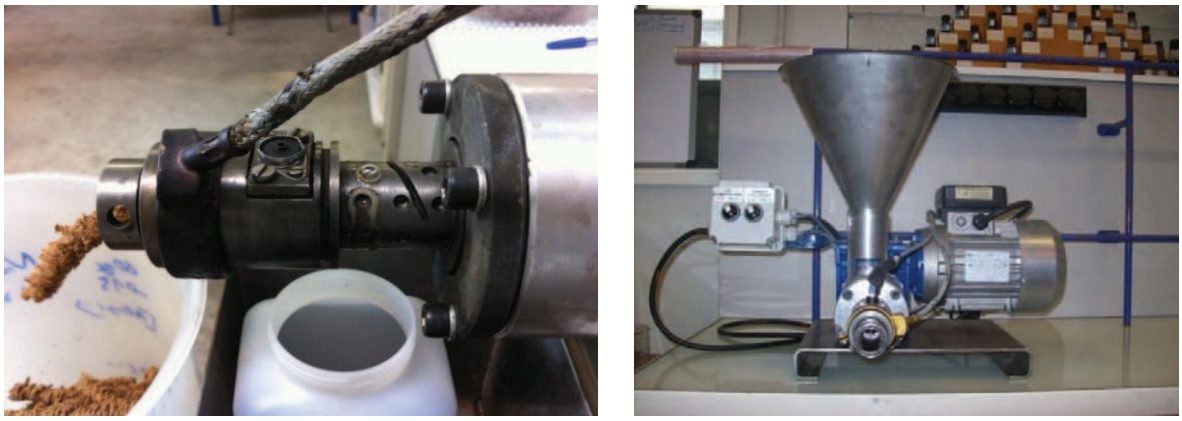


Figure 18: Single-screw extruder OMEGA 20

The filter section was 2 mm in diameter to separate extracted oil. The feed rate and the screw rotation speed maintained constant to 15g/min (0.9 kg/h) and 40 rpm, respectively. The different nozzles and distance nozzle/screw was devoted for pressing in experiments. The screw press was first run for 15 min without material but with heating via an electrical resistance heating ring attached around the press barrel, to raise the screw press barrel temperature to the desired temperature. Running temperature was adjusted with a thermocouple (Figure 18).

The configuration of single-screw system allows defining four zones (Figure 19)**:**

1 Feeding zone: The feeding of the screw by gravity, the fruits contained in the conical hopper are fed through a column until the screw which drives the forcibly inside the sheath.

2 Transport zone: The transport of material is provided by the screw turns inside the sleeve. The driving of screw is effected by an electric motor. In all tests, the speed of screw rotation, which is conditions the transport flow of fruits, is fixed. The gradual reduction of the diameter of the sheath of 4.5 to 2.5 on a length of 4 cm of the sheath causes grinding of the fruits.

3 Filtration zone: The located between the head of screw and nozzle pressing zone, this part allows to separate oil liquid (extract) and extruded meal (cake).

4 Pressing zone: This area is fitted with nozzles in which mill materials are pressed into solid residue. There are six types of nozzles with diameter arranging from 5 to 10 mm are available. The efficiency of the compression exerted on the solid depends partly on the diameter of the nozzle (the cake outlet) and distance between the screw head and the location of the nozzle. The materials transported by the screw builds up in this area and go out under the effect of compression through the nozzle. the screw continue to supply the materials to this area, ensures its pressure on the fruits. Thus, It exerts a strong pressure that expresses the liquid contained in the fruits (oil) and densified the solid. This compression results in a self-heating of the material by friction of fruits.

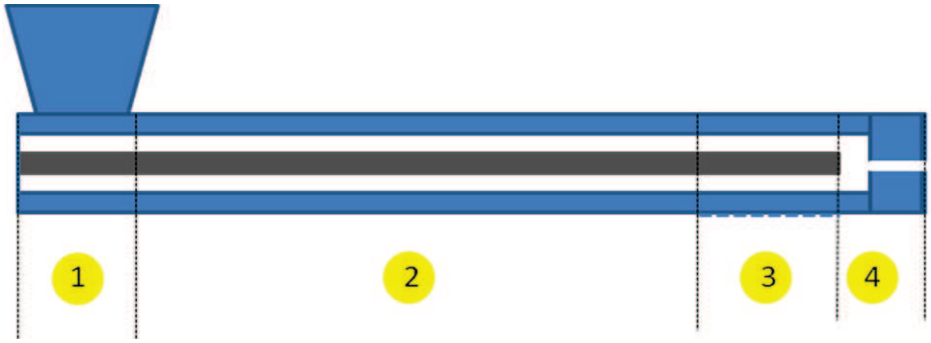


Figure 19: Functional zones of pressing (single-screw extruder OMEGA 20)

II.1.2.2 Extraction by twin-screw extruder

As our mentioned above, the application of technological processes for extracting oil from plant material got some attention from researchers (Crowe et al., 2001; Isobe et al., 1992; Singh et al., 2002; Vadke and Sosulski, 1988; Wang and Johnson, 2001). This process have been successfully carried out with using a twin screw extruder (Evon et al., 2013; Evon et al., 2007, 2009; Evon et al., 2010b; Evon et al., 2012; Kartika, 2008; Kartika et al., 2005, 2006, 2010; Sriti et al., 2012a). According to Dziezak (1989), the great capability of twin-screw extruder offers many advantages: (i) ability to provide better process control and controlling residence time distribution and uniformity of processing; (ii) ability to process specialty formulations which the single-screw extruder cannot handle and (iii) flexibility to design a machine which permits self-cleaning and rapid changeover of screw configuration without disassembling the extruder (Dziezak, 1989; Kartika et al., 2006).

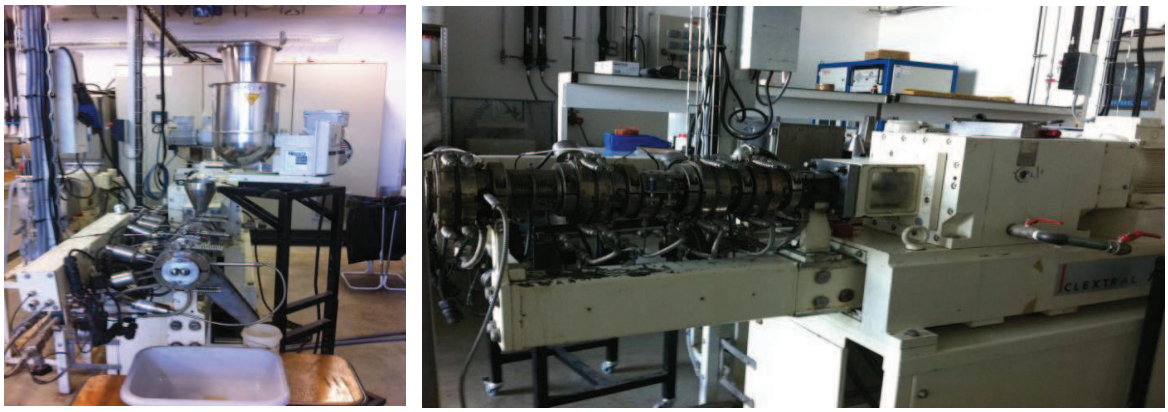


Figure 20: Twin-screw extruder type Clextral BC 21

The twin-screw extruder is mainly built by elements, namely screws (Figure 21). The arrangement of screw elements is the main factor influencing performance during extrusion processing (Gautam, 1999; Gogoi et al., 1996; Kartika et al., 2006). Therefore, this study purposed to evaluate the effects of screw configuration and operating parameters such as temperature pressing, screw rotation speed and fruit input flow rate on oil extraction of coriander fruits using a twin-screw extruder. The characterization of extraction performance was observed by the determination of oil extraction yield and oil quality.

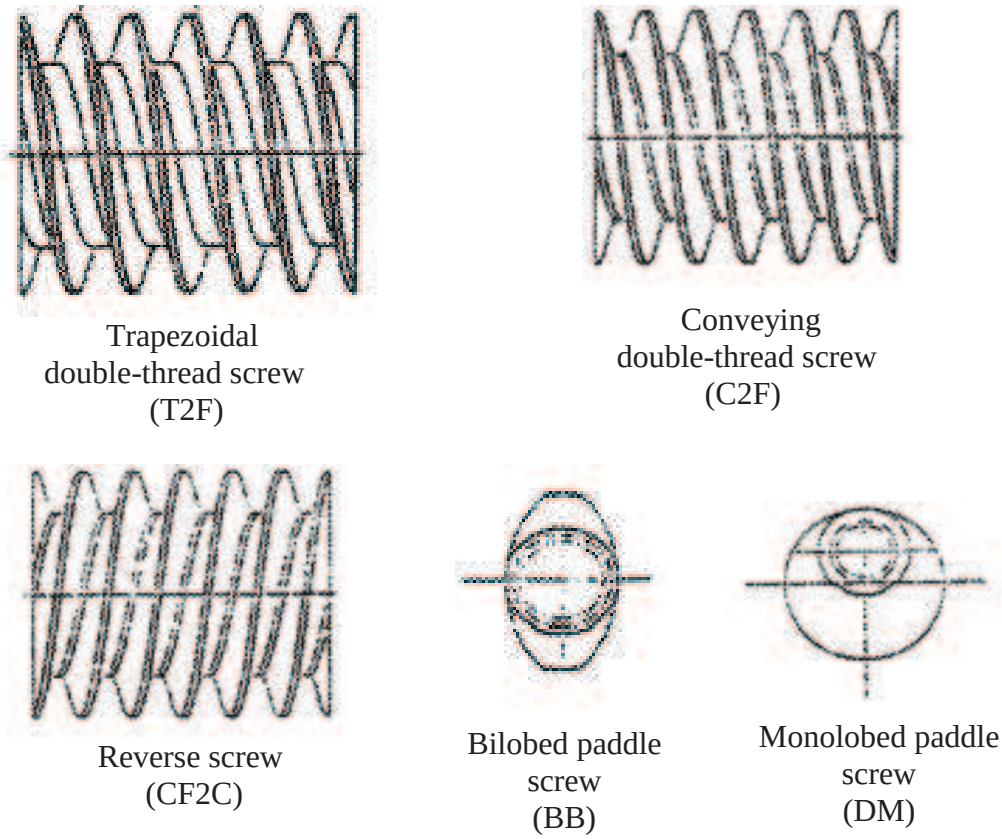


Figure 21: The Screws for extraction of vegetable oil from coriander fruits

Our experiments were conducted with a Clextral BC 21 (France) co-rotating and co-penetrating twin-screw extruder. This consists of two identical, co-rotating and intermeshing screws. The two shafts turn in the same direction and the various paired (twin) screw setups, opposite to each other in the different modules, thus co-rotate and intermesh (co-penetrate) (Figure 20). It had seven, 100 mm long modular barrels and different twin-screws made up of segmental screw elements, 25 and 50 mm in length. Modules 2, 3, 4, 5 and 7 were temperature controlled (electrically heated and water cooled) (Figure 22). Coriander fruits were fed into the extruder inlet port using a volumetric screw feeder (K-Tron Soder KCL-KT20, Switzerland). They entered a sequence of four operations: conveying (forward pitch screws) in module 1, trituration (a succession of 10 monolobe and 10 bilobe paddles) in modules 2 to 4, further conveying in modules 5 and 6, and pressing (reverse pitch screws) in module 7. A filter section consisting of four hemispherical dishes with 500 μm diameter perforations was positioned in module 6 to enable the filtrate (oil containing the 'foot', i.e. the solid particles forced through the filter)

to be collected separately from the press cake. Screw rotation speed (SS), fruit feed rate (QS), and barrel temperature (θc) were monitored from a control panel.

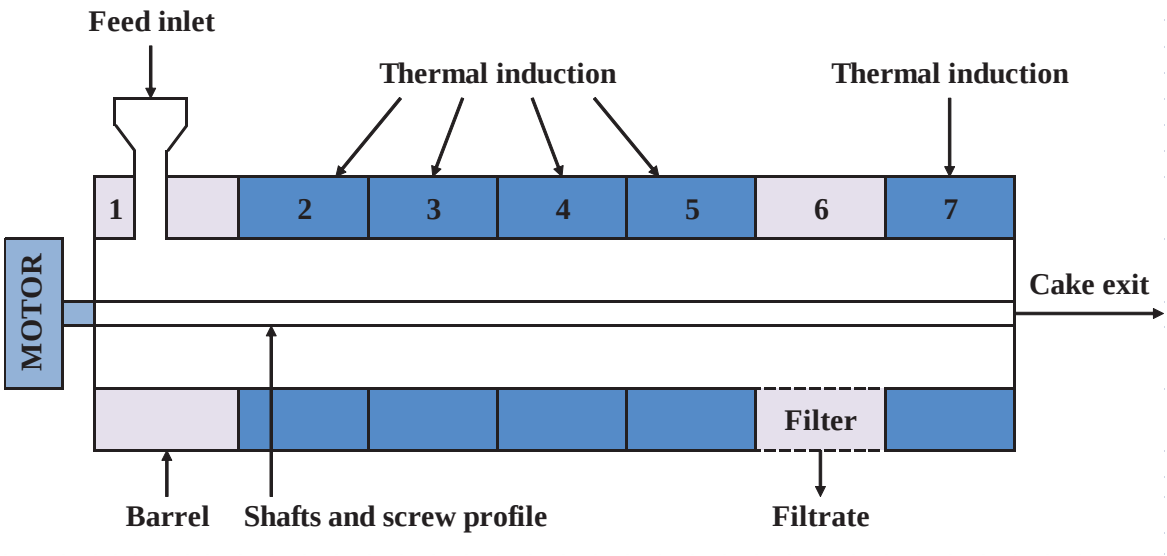


Figure 22: Schematic modular barrel of the twin-screw extruder Clextral BC 21 used for extraction of vegetable oil from coriander fruits

The four screw profiles (1 to 4) tested in this study (Figure 23) were based on those used in (Evon et al., 2013) for jatropha oil extraction. The differences between them concerned their pressing zone, situated in module 7. The reverse pitch screws used in profiles 1 and 2 were 50 mm long, with a pitch of -25 mm. They were positioned immediately after the filtration module for profile 1, and 25 mm from the end of module 6 for profile 2. The reverse pitch screws used in profiles 3 and 4 had the same length (50 mm), but their pitch was greater (-33 mm instead of -25 mm). They were positioned 25 mm from the end of module 6 for profile 3 and immediately after the filtration module for profile 4.

	1		2			3		4		5		6			7		
Profile 1	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -25	C2F 25	C2F 33

	1		2			3		4		5		6			7	
Profile 2	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -25	C2F 33
Profile 3	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -33	C2F 33
Profile 4	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -33	C2F 25	C2F 33

Figure 23: Screw configurations for extraction of vegetable oil from coriander fruits

II.2 Moisture content

The moisture content was determined through drying until a constant weight was obtained. Samples of 3 g were dried for 2 hours in an oven at 135 °C. Equally, moisture contents were analyzed through overnight drying of samples of 1 g in an oven at 103 °C. Cups were dried in the oven for about one hour prior to the addition of material.

II.3 Fractionation and Lipid analysis

II.3.1 Glyceride profile of Soxhlet extracted oil

The glyceride profile of Soxhlet extracted oil was determined through gas chromatography (GC). Samples of about 0.10 to 0.15 g analyzed were weighed and 0.5 mL of BSTFA (N,O-bis (trimethylsilyl) trifluoroacetamide) with a derivatizing agent, i.e. trimethylchlorosilane, was added. Two internal standards were added (0.5 mL). The first one was composed of 8.068 mg/mL betulin in pyridine, while the second one was a 8.087 mg/mL tricaprin solution. Next, samples were heated to 80 °C for 30 min.

They were then analyzed through GC using an Agilent Technologies 7890A (USA) gas chromatograph. The different compounds were separated in a J&W DB-5HT GC (Agilent, USA) column (15 m, 0.32 mm i.d., 0.10 μm film thickness) under the following

conditions: oven temperature: 50 °C; 50-200 °C (15°C/min); 200-290 °C (3 ^{o}C/min, held 10 min); 290- 360 °C (10 °C/min, held for 15 min); FID 380 °C; carrier gas helium (66 kPa).

II.3.2 Fatty acid composition of Soxhlet extracted Oil and pressed Oils

The fatty acid composition of Soxhlet extracted oil and pressed oils was determined through gas chromatography (GC). Samples were diluted with *tert*-butyl methyl ether (TBME) with a concentration of around 20 mg/mL. 100 µL aliquots of the prepared samples were then converted to methyl esters according to the French standard NF ISO 5508 using 50 µL of 0.5 mol/L trimethylsulfonium hydroxide (TMSH) in methanol. All experiments were done in triplicate.

Fatty acid analysis was carried out by using a gas chromatography (GC – 3900) with a flame ionization detector (FID) with Column (CP-select CB for FAME fused silica WCOT; length 50m, internal diameter 0.25mm, film thickness 0.25µm). The carrier gas was helium with flow rate of 1.2 ml/min; split ratio was 1:100. The initial oven temperature was programmed at 185 ^{o}C for 40 minutes, then increased at a rate of 15 ^{o}C/min to 250 ^{o}C, and 250 ^{o}C for 10.68 min. (analysis time: 55.0min). The injection and detector temperature were help at 250 ^{o}C for 55min. Analyses were done in triplicate.

II.3.3 Oil quality analysis

The amount of free fatty acids contained in the oil was determined through titration. A 50 ml 1/1 solvent mixture of hexane and isopropyl alcohol with phenolphthalein as a pH indicator was heated. Sodium hydroxide was added until the transition point of the indicator was reached and the mixture was added to an oil sample of 2.5 g. The oil solution was titrated with a 0.1002 N solution of sodium hydroxide until the transition point of phenolphthalein was reached. Likewise, 50 ml of ethanol may be applied and titration may be executed by means of a 0.1 N potassium hydroxide in ethanol solution. The amount of free fatty acids can be determined from the following equation:

$$\%\text{FFA} = \frac{\text{V x c x M}}{\text{10 x m}}$$

In this equation, V is the titrated volume in ml, c is the exact molarity of the applied base, M is the molar mass of the fatty acid used for expression (here 282 g/moles) and m is the sample mass in grams. Results were reported as mass % FFA as petroselinic acid.

II.4 Extraction of Essential Oils

II.4.1 Hydrodistillation (HD)

Hydrodistillation (or water distillation) was applied to extract the essential oil from the coriander material. i.e. the different parts of coriander or the press cakes: from 30 - 100 g of dry material (leaves, stem or fruits) were mixed with 1 L of water; 200 g milled material (press cake) was mixed with 2 L water (1:10 ratio) and positioned inside a distillation flask where it was refluxed for 5 h. The installation consisted of a Clevenger apparatus. Volatilized compounds and water vapor were condensed through a cooling system, collected, and separated in a separator tube. An overflow system avoided that the material in the flask became dry and underwent thermal degradation. The obtained oil was separated from the water, isolation of oil was performed in duplicate and the samples were stored in a freezer prior to further analysis.

II.4.2 Simultaneous distillation Extraction (SDE)

Simultaneous Distillation Extraction of volatile extracts from flowers and roots of coriander were executed by classic Likens-Nickerson apparatus. This materials arranging from 5 g to 20 g were put into one flask for distillation, followed by addition of 300 mL of water. Then 50 mL of pentane was put into the other flask for extraction of volatile extracts. The SDE process lasted for 3 h. After that, solvents containing volatile extracts in the Likens-Nickerson apparatus were combined and separated from water by separatory funnel. The solvent obtained was then concentrated to about 10 mL using rotary evaporator at reduced pressure and finally concentrated to about 1 mL under the gentle flow of nitrogen gas. The obtained solvent with volatile extract was put into a bottle and stored in the refrigerator for the following analysis.

II.4.3. Analysis of Essential Oils

The composition of essential oils extracted from the coriander fruits and the press cakes was determined through gas chromatography (GC). The essential oils were analyzed using

a HP 5890 Series II (USA) gas chromatograph equipped with a flame ionization detector. The carrier gas was helium with a flow rate of 150 mL/min. Compounds of the essential oils were separated in a Agilent VF-5ms (USA) apolar column (30 m, 0.5 mm i.d., 1 μm film thickness). The injected volume was 0.5 μL. The initial oven temperature was 110°C and increased at a rate of 7°C/min to 220°C. The injector and detector temperatures were 220°C. All determinations were carried out in triplicate.

* Kovats retention index

Retention parameters, preferably Kovats indices (Kovats, 1958; Wehrli & Kováts, 1959), have become an accepted procedure for the identification of Gas Chromatographic peaks using isothermal retention data. The retention value is compared the relative position of a peak with respect to range of hydrocarbons known (from C5 to C14) injected in the same conditions experimental.

The formula for the calculation of the retention index as follows:

$$\mathrm{KI} = 100\,z + 100\frac{\log \mathrm{tR}'(\mathrm{x}) - \log \mathrm{tR}'(\mathrm{nPz})}{\log \mathrm{tR}'(\mathrm{nPz}+1) - \log \mathrm{tR}'(\mathrm{nPz})}$$

Where **x** is the compound of interest which eluated between nPz and nPz+1 n-alkanes, tR' is the adjusted retention time of each component and nPz and nPz+1 are n-alkane standards which eluated before and after the compound of interest and consist of z and z+1 carbon atoms, respectively.

II.5 Antioxidant assay

After essential oils (or volatile extracts) were collected, the mixtures of solid residue were filtered. The solid residues were collected and dried by using air-drying machine at around 60°C. The Soxhlet extraction using methanol as solvent was executed for 5h. the methanolic extracts obtained will be tested antioxidant activity.

II.5.1 DPPH radical scavenging assay

The antioxidant capacity was measured by means of the DPPH radical scavenging assay, through which the radical scavenging activity of an extract against the stable DPPH (2,2-diphenyl-1-picrylhydrazyl) radical was determined. The applied method was based on the one used by (Brand-Williams et al., 1995) and comprises methanolic extraction and DPPH scavenging assessment through UV spectrophotometry. Methanolic extracts were obtained

through methanol Soxhlet extraction for 5 h and subsequent concentration by rotary evaporation. Aqueous extracts were prepared through rotary evaporation of water solutions. Inhibition (%) was calculated from the following formula:

$$\%\text{Inhibition} = \frac{\text{Ab} - \text{Aa}}{\text{Ab}} \text{ x } 100$$

In which: Ab = absorbance of DPPH solution; Aa = absorbance of tested extract solution.
A 6×10^{-5} M DPPH in methanol solution was prepared daily, protected from the light and stored at low temperatures. 10 mg extract was dissolved in 1 mL methanol or a 1/1 water/methanol solution for aqueous extracts. The samples were subjected to sonication to ensure complete dissolution. 50 µL of this extract solution was added to 2 ml of DPPH solution, and the samples were put in the dark for 30 min to react. Absorption was measured at 515 nm through UV spectrophotometry. Further, a calibration curve was set up using methanolic Trolox solutions with known concentrations ranging from 1 100 to 750 µmol/L.
Results were expressed as µmol of Trolox equivalents (TE) per g of press cake. All determinations were carried out in triplicate.

II.5.2 Ferric Reducing/Antioxidant Power (FRAP) assay

The ability of plant extracts to reduce ferric ion to the ferrous one, (FRAP assay) is another indicator frequently used for assessing antioxidant power (Benzie and Strain, 1996). Ferrous ion (Fe^{2+}) produced in this assay forms a blue complex (Fe^{2+}/TPTZ) absorbing at 593 nm. Briefly, the reagent was prepared by mixing acetate buffer (300 mM, pH 3.6), a solution of 10 mM TPTZ in 40 mM HCl, and 20 mM $FeCl_3 \times 6H_2O$ at 10:1:1 (v/v/v). Firstly 300 µl of freshly prepared FRAP reagent was heated to 37 °C and an absorbance (A_0) of a blank reagent was read at 593 nm in a Biotek EL808 micro plate reader (Vermont, USA). Then 10 µl of 0.1% extract solution in water and 30 µl H_2O were added (final dilution of samples in the reaction mixture was 1:34) and the absorbance (A) was recorded every 1 min during the whole monitoring period which lasted up to 30 min. The change in the absorbance (ΔA_{593} nm) between the final reading and A_0 was calculated for each sample and related to the ΔA_{593} nm of a Fe^{2+} reference solution which was measured simultaneously.

II.5.3 Oxygen Radical Absorbance Capacity (ORAC) assays

Oxygen Radical Absorbance Capacity (ORAC) assays were performed according to the methods by Ronald Prior (Prior et al., 2003). It is one of the standardized methods for determining the antioxidant capacity of substance. It uses flourescein as the probe and reactive oxygen species (ROS) generated by the thermal decomposition of AAPH [2,2'-azobis (2-amidino-propane) dihydrochloride] can quench the flourescent signal by flourescein. The addition of antioxidant substances will produce a more stable flourescent signal which can reflect the antioxidant capacity.

Firstly, PBS (Phosphate Buffered Saline) solution was prepared by dissolving 8.18g NaCl, 0.27 g KH_2PO_4 $12H_2O$, 3.58g Na_2HPO_4 and 0.15g KCl in 1 L of deionized water. The pH value of PBS solution was determinded and adjusted to 7.4. The stock flourescein solution (Stock #1) was then prepared by dissolving 0.0313g of FL in 50mL of PBS solution. A second stock solution #2 was prepared by diluting 50µL of stock solution #1 in 10 mL of PBS solution, 800 of stock solution #2 was added to 50 mL of PBS to obtain the flourescein solution of 95.68 nmol/L for the subsequent assays. 240 mM of AAPH solution was prepared by dissolving 0.651g of AAPH in 10mL of PBS solution.

The ORAC assays were performed ny use of FLOUstar Omega microplate reader (BMG LABTECH, Germany). On a 96-wel plate, 25 µL of samples, Trolox solution and PBS solution for blank, were added into the well, followed by 150µL of flourescein solution. Each assay was performed in triplicated. After the gain adjustment, the microplate was covered and incubated in the microplate reader for 15 min at 37 ^{o}C. Then flourescence measurements (Ex. 485 nm, Em. 520 nm) were taken every 66 sec. After 3 cycles, 25µL of AAPH was quickly injected into the well by use of multi-channel pipette and the test was resumed. The whole measurements will be finished in about 120 min.

The final $ORAC_{FL}$ values were calculated by using a regression equation (y=ax+b) between Trolox concentration and the net area under the flourescein decay curve. The PBS solution of Trolox with known concentration ranging from 12.5 to 200 µmol/L were used for calibration. The antioxidant capacity of each sample is expressed as µmol of Trolox equivalent (TE) per gram of plant extract.

The area under the curve (AUC) was calculated as:

$$AUC = (0.5+f_5/f_4+ f_6/f_4+ f_7/f_4 +\ldots+ f_i/f_4) \times CT$$

Where f_4 = initial flourescence reading at cycle 4

f_i = flourescence reading at cycle i.

CT = cycle time in minutes.

The net AUC was obtained by subtracting the AUC of the blank from that of each sample.

II.5.4 Analysis of total phenolic compounds (TPC)

The content of total phenolic compounds in extracts was determined with Folin-Ciocalteu reagent (Folin and Ciocalteu, 1927). The assay was performed according to the slightly modified method by (Medina, 2011). Calibration curve was prepared by using 1 mL reference gallic acid solutions in ethanol (0.025, 0.075, 0.100, 0.175 and 0.350 mg/mL), which were mixed with 5 mL of a standard Folin-Ciocalteu reagent and diluted with distilled water (1:10) and 4 mL of 7.5% sodium carbonate solution in distilled water. The absorption was read after 30 min at 765 nm. The concentration of TPC was expressed in mg of gallic acid equivalents (GAE) per g of plant extract. The TPC was calculated by the following formula:

$$C = \frac{c \times V}{m}$$

Where: C is concentration of the total phenolics, in gallic equivalents (GAE); c is the concentration of gallic acid, determined form the calibration curve (mg/mL); V is the volume of plant extract (100 mL); m is the weight of pure plant extract (g).

II.6 Agromaterials

II.6.1 Mechanical properties in bending of the panels produced

The flexural strength of a material is defined as the ability to resist deformation under load and it is also known as modular broken, bent or broken power strength. Flexural strength of an object also correlates with its tensile strength, or the ability of the object to be extended without significantly changing its shape. The flexural strength represents the highest stress experienced within the material at its moment of rupture. In materials science, the properties of materials such as bending, tensile strength is very important to ensure that the material is strong enough to be used in the structure. For this reason, it is important to assess the breaking load, flexural strength at break and elastic modulus of materials. Flexural Strength is calculated by the formula: $\sigma = 3FL/2bh^2$ where the F is the load that breaks a rectangular specimen of length L, b – specimen width; h – specimen thickness. In a three point bend test arrangement shown here (Figure 24).

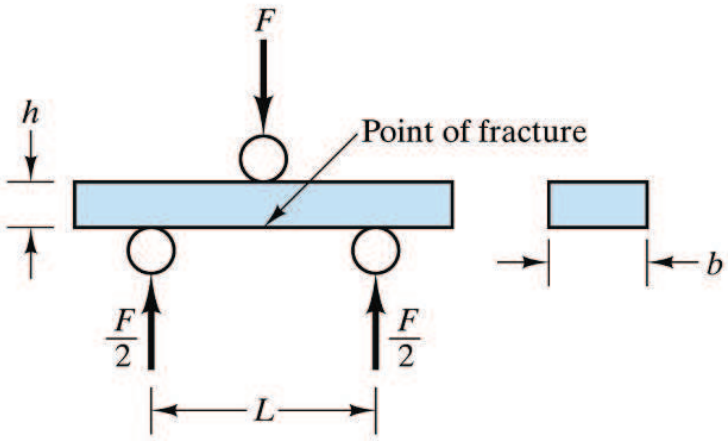


Figure 24: Three point flexure test

For this study, The Instron 33R4204 (USA) (Figure 25) universal testing machine fitted with a 500 N load cell was used to assess the flexural properties of the test specimens according to the French standard NF EN 310, including breaking load (F), flexural strength at break (sf), and elastic modulus (Ef). The test specimens were 150 mm long and 30 mm wide. Their thickness was measured at three points with an electronic digital sliding caliper having a 0.01 mm resolution, and the mean value (t) was recorded to calculate their volume and section. All specimens were weighed to calculate their mean apparent density (d). The test speed was 3 mm/min and the grip separation was 100 mm. Test specimens were cut, and equilibrated in climatic chamber (60% RH, 25°C) during three weeks before being tested. All determinations were carried out four times.

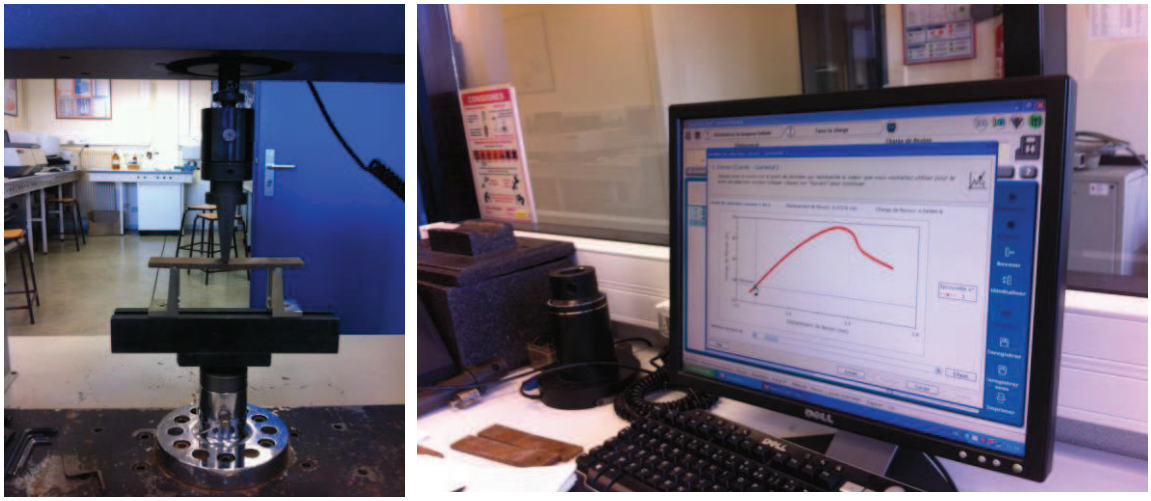


Figure 25: Instron 33R4204 (USA) machine for bending Test

II.6.2 Surface hardness

Hardness is the property of a material that enables it to resist plastic deformation, usually by penetration. However, the term hardness may also refer to resistance to bending, scratching, abrasion or cutting. Some materials, such as metal, are harder than others. Macroscopic hardness is generally characterized by strong intermolecular bonds, but the behavior of solid materials under force is complex. In this study, A Bareiss (Germany) durometer was used to assess the Shore D surface hardness of the fiberboards according to the French standard NF EN ISO 868. Fiberboards were equilibrated in climatic chamber (60% RH, 25^{o} C) during three weeks before being tested. All determinations were carried out ten times.

II.6.3 TGA measurements

Thermogravimetric analysis (TGA) is a method of thermal analysis to determine changes in the physical properties and chemistry of materials. The material is measured as a function of temperature increases (with a constant heating rate), or as a function of time (with constant temperature or weight loss does not change) (Coats and Redfern, 1963). TGA can provide information about the physical phenomena of the material, such as phase transitions, including evaporation, sublimation, absorption, adsorption, and desorption. Similarly, TGA can provide information on the chemical phenomena including chemisorptions, soluble (especially dehydration), decomposition, and solid-gas reactions (e.g., process oxidation or reduction) (Coats and Redfern, 1963). Common applications of TGA is (i) Material properties by analyzing the decay characteristic form, (ii) research on the mechanism of degradation and reaction kinetics, (iii) determine the amount of organic in form, and (iv) determining inorganic (e.g. ash) content in a sample, which may be useful for corroborating physical structure prediction or simply used as an analytical chemistry. This is a particularly useful technique for the study of polymeric materials, including thermoplastics, thermosets, elastomers, composites, plastic films, fibers, coatings and paint.

In study, The Shimadzu TGA-50 (Japan) analyzer was applied for Thermogravimetric analysis (TGA) of fiberboards. Dynamic analysis was carried out under air at a heating rate of 10 ^{o}C/min, from 20 to 650 ^{o}C. Fiberboards were previously crushed using a Foss Cyclotec 1093 (Denmark) mill fitted with a 1 mm screen. Before analysis, the crushed

materials were equilibrated in a climatic chamber (60% RH, 25 ^{o}C) during three weeks. For all measurements, the mass of the test sample was around 10 mg. The weights of samples were measured as a function of temperature and stored. These data were later used to plot the percentage of undegraded sample (1 - D) (%) as a function of temperature, where D = (Wo - W)/Wo; and Wo and W were the weights at the starting point and during scanning (mg). All measurements were carried out in duplicate.

II.6.4 Impact strength

A 0-40 daN cm Testwell Wolpert (France) Charpy machine was used to assess the impact strength of the unnotched test specimens according to the French standard NF EN ISO 179, including absorbed energy (W), and resilience (K). The test specimens were 70 mm long and 10 mm wide. Their thickness was measured at three points with an electronic digital sliding caliper having a 0.01 mm resolution, and the mean value (t) was recorded to calculate their section. Impact strength measurements were realized at 23 °C according to the three points bending technique and the grip separationwas 29.5 mm. Test specimens were cut, and equilibrated in climatic chamber (60% RH, 25°C) during three weeks before being tested. All determinations were carried out ten times.

II.7 Statistical analysis

Data are presented as mean values ± standard deviation calculated from triplicate (in some cases duplicated) determinations. Standard deviations (SD) did not exceed 3% in the major of the values obtained. The DPPH, FRAP, ORAC test and content of TPC were calculated using MS Excel software. Statistical analysis of the data was performed by using one-way analysis of the variance (ANOVA), followed by the Duncan's post hoc test to compare the means that showed significant variation ($p < 0.05$). All the data were subjected to variance analysis using the GLM procedure of SAS (SAS Institute, 1987, Cary, NC, USA).

Chapter 3: BIOACCUMULATION

I. Plant material, crop management and experimental conditions in Auch and Vietnam

I.1 Plant material, crop management and experimental conditions in Auch

As mentioned in Chapter 2, the coriander cultivars (*Coriandrum sativum* L.) fruits were obtained from supermarket, seed company, retail shops of different European and Asian countries (eight samples - Canada, Tunisia, Lithuania, Algeria, China, French (FR1 - Dourou) and Vietnam (2 sample - VS1 and VS2). After collected, The cultivars was cultivated in south-western France at the Regional Centre of Experimentation in Organic Agriculture at Auch (near Toulouse, south-west France, 43°38'47"N, 0°35'08"E).
These coriander genotypes were grown on 25th March, 2011 into the field at a rate of 12 kg ha^{-1} to a depth of 3 cm. The crops were completely managed under organic and rained conditions without any chemical supply. Crushed feathers were applied as an organic fertilizer at a rate of 60 units.ha^{-1} in April and May. Weeds were mechanically eliminated. The soil was a clay-loam (organic matter content: 3.2%; pH 8.1) with a depth of about 1.2m. Maturity took place at the end of July to beginning of August.

In order to evaluate oil content, fatty acids composition and accumulation during fruit ripening, coriander fruits of the French cultivar (FR1 - Dourou) (GSN, Riscles, France) were cultivated during the cropping season (2009, 2010 and 2011). Sowings were made on 23rd March, 9th April and 25th March in 2009, 2010 and 2011, respectively. Fruits samplings were performed each five (average) days between beginning of flowering and physiological maturity. Harvest period was started from 2 days after flowering (DAF) to 59 DAF. The fruit's color and relative moisture content were adopted as a ripening criterion. Fruits water content (SWC in % of the fruits dry matter) was measured on each sample as an indicator of stage of physiological maturity to make possible comparison between years. Moisture contents were determined by heating in an air-oven at 60°C to constant weight. Furthermore, the French cultivar (Dourou) was also selected in order to study the effects of planting date on oil yield and fatty acid composition. This experiment trial was carry out at different sowing time (Sowing Date - DS1, DS2, DS3) with the same organic condition in the field (in 2011). The sowing date (DS1, DS2, DS3) was on 25th March , 14th April and 30th April, respectively. The raw material harvested on the fully maturity stage (September 2011) was naturally dried in the shadow and the fruits were removed by hand.

Table 3: The prevailing weather condition during the plant cycle in three years (2009, 2010

Month	Rainfall (mm)				Ter	
	2009	2010	2011	55ans	2009	2
January	95.4	45.8	17.4	65.0	4.9	3
February	40.4	43.6	58.7	55.9	5.8	5
March	23.6	42.6	36.4	57.1	8.7	8
April	105.4	48.9	13.2	66.6	11.7	1
May	44.4	118.1	46.6	77.2	16.8	1
June	39.4	52.6	35.1	61.1	20.1	1
July	41.2	62.3	92.3	47.1	21.7	2
August	20.0	23.2	27.3	58.3	22.7	2
September	46.8	31.0	13.4	55.9	18.9	1
October	35.6	104.8	18.6	56.8	15.0	1
November	102.2	91.5	41.1	57.2	10.9	9
December	34.6	21.1	75.7	68.1	5.9	4
Mean					13.6	1
Sum year	629	685.5	475.8	726.3		
Mean Mar-Sept					17.2	1
Sum Mar-Sept	320.8	378.7	264.3	423.3		
Mean May-July					19.5	1
Sum May- July	125.0	233.0	174.0	185.4		

In Agriculture, the sowing date is determined to provide the conditions for the plants to maximally use environmental parameters, especially temperature, water and radiation. If environmental factors are appropriate for the greening, establishment and survival of seedlings, then the maximum yield can be realized by appropriate sowing date. Therefore, this study was performed to investigate the effect of sowing date for vegetable oil and essential oil content, yield and quality of coriander, from which will apply the appropriate measures to cultivation in conventional conditions in Auch.

Table 3 shows temperatures and rainfall during the three plant cycles and were compared to the weather data of the last 55 years. Indeed, rainfall was at least 50 mm lower than the half century precipitations observed for the same period in our area. The three growing seasons were less rainy and hottest than the mean values of 55 years. Along the cropping cycle, the driest and hottest year was 2011 and the rainy one was 2010. In contrast, during grain filling period, 2009 was less rainy and hotter than other years and half-century average. Flowering started from end of May to mid-June depending on year. Maturity took place at the end of July to beginning of August.

I.2 Plant material, crop management and experimental conditions in Vietnam

Coriander fruits were obtained from Vietnam National Seed Joint Stock Company (Vinaseed) and then were directly sown by hand into the field at rate of 10 - 12 kg ha^{-1} to a depth of 3 cm at three locations in Vietnam and the crops were completely managed under organic and rained conditions without any chemical supply. This genotype were planted in the first week of March in 2012 and randomly collected at different ripening stages in September 2012 at three locations: Ha Noi (latitude N 21° 4’ 21’’; longitude E 105° 43’ 45’’); Thai Binh (latitude N. 20^o 25’ 25’’; longitude E. 106^o 20’29’’, Northern of Vietnam - Red River Delta) and Ha Tinh (latitude N. 18^o 20’ 120’’; longitude E. 105^o 53’345’’, North central coast of Vietnam). Plants germinated 10 to 15 days after sowing, flowering began 45 to 50 days after germination and then fully maturity began 50 to 60 day after flowering. Harvest of coriander in all treatments occurred when 100% of the fruits in the umbels were matured. Whole plants were harvested by hand, the plants were naturally dried in the shadow and the fruits were removed by hand.

Table 4: The mean air temperature and monthly rainfall in three location (Ha Noi, Thai Binh

Month	Rainfall (mm)			
	Ha Noi	Thai Binh	Ha Tinh	Ha Noi
January	20.9	28.9	37.9	14.6
February	16.5	20.5	47.8	16.1
March	16.9	28.2	19.9	20.2
April	31.8	77.6	83.3	26.2
May	387.7	291.3	88.2	28.9
June	268.9	139.5	137.3	30.3
July	388.3	267.8	99.3	29.6
August	478.1	367.0	65.5	29.3
September	54.7	504.6	547.1	27.9
October	77.5	589.8	235.9	26.8
November	34.8	21.8	381.6	23.4
December	25.7	18.1	54.7	18.7
Mean				24.3
Sum year	1801.8	2355.1	1798.5	
Mean Mar-Sept				27.5
Sum Mar-Sept	1626.4	1676.0	1040.6	

Meteorological data during coriander growing season were obtained from the gauging stations in each province (Ha Noi, Thai Binh and Ha Tinh meteorological service) (Table 4). Thai Binh is located in the humid subtropical climate, hot summers and rainy from May to September, the winter from November to March of next year. Average temperature: 23.6 ° C. The number of hours of sunshine in the year: 1600 - 1800 hours. The average relative humidity: 85 - 90 and the annual rainfall received a large 1700 - 2300 mm, the region is divided by the rivers, which are tributaries of the Red River. The coriander was cultivated in the soil which contains mechanical components usually moderate rich soil and reddish-brown, the pH reaction of 4.5 - 5. These soils are quite nutritious. The shallow layer's underground water regime is relatively stable, less saline.

Ha Tinh has monsoon tropical climate of a coastal province which totally lies in the internal tropic. Every year, there are 1,350-1,700 sunshine hours on average. Sunny season lasts from April to September, and early October. Average annual temperature is about 23.5 - 24.6^{o} C. The highest temperature is over 40^{o} C, and in some places it increases to 42.6^{o} C in April, May or June. The lowest temperature is around 7^{o} C, in some places it decreases to 0.7^{o} C normally in December and January. Average annual rainfall is about 2,300-3,000 mm while the highest rainfall is reported up to 3,800 - 4,400 mm. Rainy season lasts from April-May to November - December. Average annual evaporation amount is within the range of 800 - 1,100mm. Annual humidity index is common from 2.2 to 3.3. Monthly humidity index is mostly over 1. It does even reaches up to 10 - 15 in some months, except for the months when westerly winds are active. The coriander was cultivated in the soil namely sandy soil, alluvium, saline soil, alkaline soil with pH 4.5 - 5.

The coriander are planted in Hanoi where was selected for experimental to evaluation of bioaccumulation of fatty acid in maturity stage. Fruits samplings were performed each 8 - 10 (average) days between beginnings of flowering to maturity. Harvest period was started from 2 days after flowering (DAF) to fully mature 55 (DAF). The fruit's color and relative moisture content were adopted as a ripening criterion. At harvest time, natural drying of fruits performed in a shady place at room temperature. After drying, this material was used for the isolation of oil content.

The field preparation operation including tillage was carried out in early-March. The most of the surface soil are mostly composed of kaolinite, from loamy sand to light loam with

pH 5.6 - 6.1, light texture light and little humus, the cation exchange capacity is also quite low. Most of the research area has a CEC which was lower than 10 cmol (+) kg^{-1} soil. The annual average temperature in Ha Noi (Vietnam) is fairly hot at 24.1 with average precipitation of 1800 mm of rainfall per year, or 150 mm per month and average humidity of 70% and average monthly relative humidity ranges from 67% (December) to 76% (in March).

II. Vegetable Oils Analysis

II.1 Oil content and fatty acid composition in different parts of Coriander

In these experiments, the French coriander cultivar (FR1 - Dourou) was selected in order to study the fatty acid composition and oil content of different parts of coriander (fruit, leaves and stem, roots). The French coriander cultivar was cultivated was on 25th March 2011 under the organic condition.

The samples collected from the different parts of coriander (*Coriandrum sativum* L.) were analyzed to determine fatty acid and oil yield, the results of analyses were presented in Table 5. As can be seen in Table 5, there are big differences of oil yield content between fruits, leaves and stem, roots. This results show that the oil content of coriander fruits is highest (up to 24.26 ± 0.14%), similar results were found by some previous studies (Msaada et al., 2009a; Msaada et al., 2009b). Followed by oil content of flower (5.0 ± 0.08%), oil content of leaves and stem (1.48 ± 0.1%), and oil content of coriander roots (0.85 ± 0.08%). Currently, there are very few researches on oil composition and oil yield of flowers, leaves and roots of coriander. Only one previous publish has studied the roots volatiles and fatty acids of coriander grown in saline medium (Neffati and Marzouk, 2009) and one other has studied changes in essential oil and fatty acid composition in coriander leaves under saline conditions (Neffati and Marzouk, 2008). The base on this results, it can be recognize the oil contents of coriander flower, leaves and roots is very low compared with the oil contents of coriander fruits (Figure 26). Therefore, the study of vegetable oil and essential oil of flower, leaves and coriander roots are not really attractive to agricultural scientists. This is reason why our study will be only focus coriander fruits because of its promising in future.

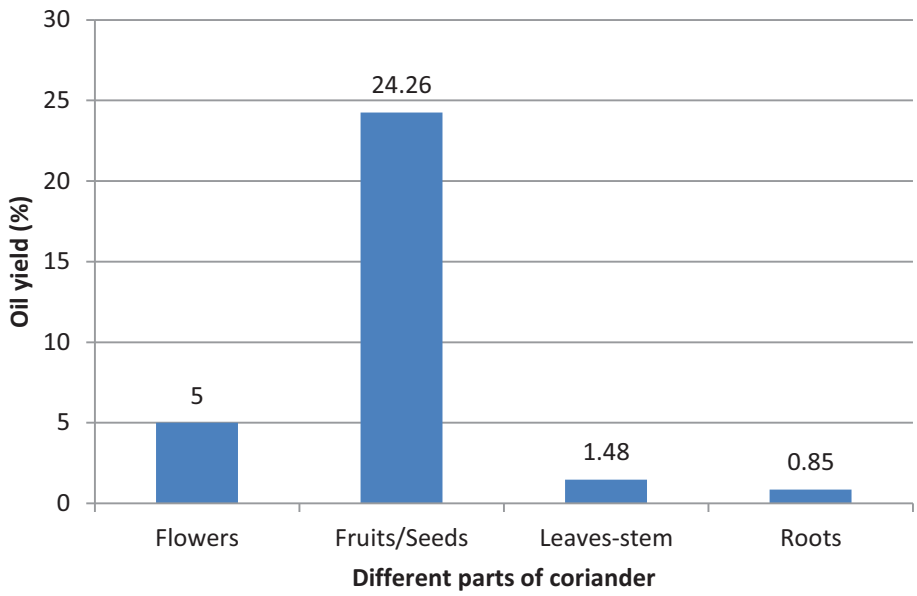


Figure 26: The oil contents of coriander flower, leaves, roots and fruits

Considering the main components, there are substantial differences between the different parts of coriander in a quantitative sense. In the fruits, nine fatty acids were identified, of which the major components is petroselinic acid (74.33 ± 1.42%), linoleic acid (14.67 ± 0.16%) and oleic acid (5.86 ± 1.03%). The same result was also found in major components of coriander fruits in previous studies, the fatty acid profile is a main determinant of the oil quality in coriander fruit mainly with percentage of oleic (5–7%), linoleic (13–16%) and petroselinic acids (65–76%) (Ramadan and Mörsel, 2002; Sriti et al., 2011a; Sriti et al., 2009). Recent studies that have described the composition of coriander fruits in essential oil and fatty acid composition of fruit's different parts (Sriti et al., 2009) and the changes of fatty acid composition of coriander ripening had also defined the influence of maturity stages and growing region on oil content and fatty acid composition (Msaada et al., 2009a; Msaada et al., 2009b). In the flowers, ten fatty acids were identified, of which the major components is linoleic acid (35.59 ± 0.20%), palmitic acid (27.60 ± 0.16%) and linoleic acid (18.72 ± 0.05%). in which linoleic acid (omega-6), and linolenic acid (omega-3) are the two essential fatty acids. They are termed as essential because the body cannot synthesize them and needs to obtain them from the diet. This composition was different from that of the fruits where petroselinic acid (C18:1n12), was the main fatty acid (Peiretti et al., 2004). In the leaves - stem and roots, twelve and eleven fatty acid were also identified, respectively. The major components of leaves - stem is

linoleic acid (27.45 ± 0.35%), palmitic acid (26.94 ± 0.14%), behenic acid (7.96± 0.11%), stearic acid (7.95 ± 0.03%), linolenic acid (7.92 ± 0.40%); and the major components of roots include as linoleic acid (22.81 ± 2.0%), palmitic acid (19.17 ± 0.42%), oleic acid (18.35 ± 0.11%), behenic acid (17.91 ± 2.29%), stearic acid (9.17 ± 0.42%). The fatty acids profile is a main determinant of the mainly oil quality of coriander with percentage of petroselinic acids. However, comparisons between the different parts of coriander were made, petroselinic acid appear on the leaves and flowers with very low, and absence in roots, perhaps this is main reason that there are very few research on fatty acid composition from flowers, leaves and roots of coriander.

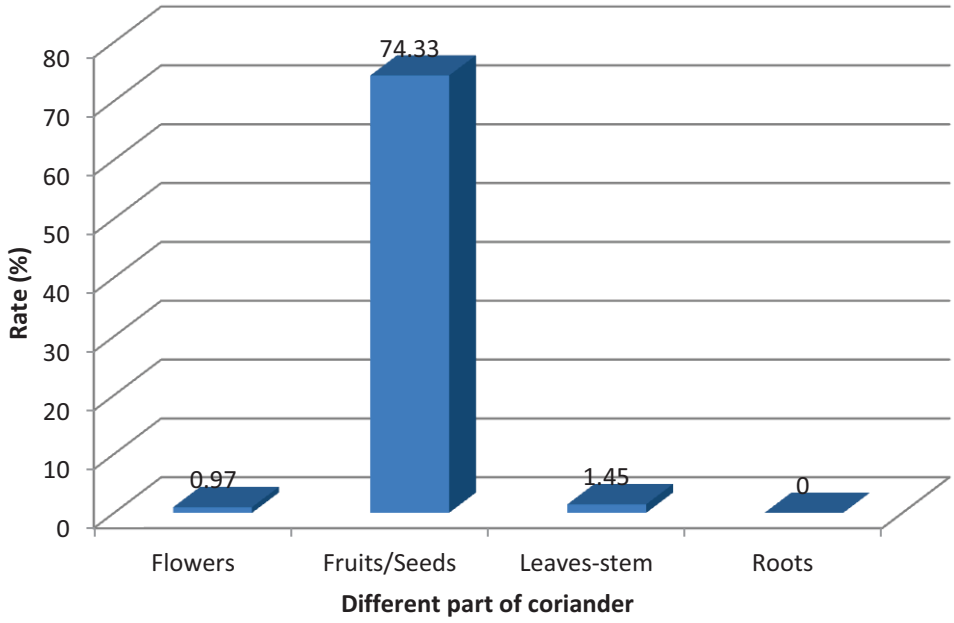


Figure 27: Petroselinic acid composition in different parts of coriander

From these results of the present investigation, it appears that the studied fruits are a valuable source of essential unsaturated fatty acid. Currently unsaturated fatty acids are well known for their value in nutrition and pharmaceutical industrial uses. Thus producers of nutraceuticals and dietary products are interested in natural sources of beneficial unsaturated. Petroselinic acid is an uncommon isomer of oleic acid which is found in high levels in a restricted range of fruits oils, mostly from the Apiaceae family. It forms a unique oleochemical with high potential for food, cosmetic and pharmaceutical industries and may further allow the synthesis of a large number of interesting platform molecules.

Table 5: The fatty acid composition and oil yield in different parts of corian

Fatty acid (%)	Different parts of coria		
	Flowers	**Fruits**	**Leave**
Saturated fatty acid (SFA)			
C14:0 (Myristic acid)	5.85 ± 0.01	-	4.0
C15:0 (Pentadecylic acid)	-	-	1.1
C16:0 (Palmitic acid)	27.60 ± 0.16	3.81 ± 0.19	26.9
C17:0 (Margaric acid)	-	-	2.4
C18:0 (Stearic acid)	3.93 ± 0.05	0.74 ± 0.06	7.9
C20:0 (Arachidic acid)	2.42 ± 0.31	0.13 ± 0.01	4.2
C22:0 (Behenic acid)	2.27 ± 0.03	0.13 ± 0.01	7.9
Total	42.07 ± 0.08	4.81 ± 0.26	54.8
Monounsaturated fatty acid (MUFA)			
C16:1n7 (Palmitoleic acid)	0.36 ± 0.12	0.19 ± 0.01	0.9
C18:1n12 (Petroselinic acid)	0.97 ± 0.25	74.33 ± 1.42	1.4
C18:1n9 (Oleic acid)	2.29 ± 0.16	5.86 ± 1.03	7.4
Total	3.62 ± 0.32	80.38 ± 0.41	9.8
Polyunsaturated fatty acid (PUFA)			
C18: 2n6 (Linoleic acid)	35.59 ± 0.20	14.67 ± 0.16	27.4
C18:3n3 (Linolenic acid)	18.72 ± 0.05	0.15 ± 0.01	7.9
Total	54.31 ± 0.25	14.82 ± 0.16	35.3
SFA/PUFA	0.77 ± 0.01	0.32 ± 0.01	1.5
Oil yield (%)	5.00 ± 0.08	24.26 ± 0.14	1.4

Data presented as Mean ± SD of three replicates; (-): not detected.

Although the percentages of palmitic and linoleic acid in flowers, leaves-stem and roots is higher than in the coriander fruits. But percentage of petroselinic acid is very low from them (< 1% in flower and < 1.4% in leaves and stem), even the petroselinic acid is virtually absent from coriander roots (Figure 27). On the other hand, the oil yield from leaves - stems (< 1.5%) and from roots (< 1%) are much lower than in the fruits (up to 25%). Therefore, this is the main reason for us to focus on study the coriander fruits instead of study on vegetable oil from leaves and roots of coriander in the next studies of this thesis.

II.2 Effects of sowing time on yield and fatty acid composition of coriander

In this study, the French coriander cultivar (FR1 - Dourou) was selected in order to study the effects of planting date on oil yield and fatty acid of coriander fruits. These trials were cultivated at different sowing time under organic conditions. Fatty acid of coriander fruits were analyzed for qualitative composition by using GC-FID. Typical GC chromatogram of the fatty acid is presented in Figure 28.

The results of oil yield and fatty acid composition of coriander fruits shown in Table 6. In this results, the highest oil yield of fruits was obtained in the first trial DS1 (24.26 ± 0.14%), followed by DS2 (20.19 ± 0.69%) and DS3 (17.36 ± 0.23%), respectively. According to the results have also shown that a significant difference between the oil content in different sowing time. Oil yields fell significantly as sowing times were delayed, the lowest fruits yield was obtained from the third sowing time (in third trial DS3), whereas the highest fruits yield was obtained from the first sowing times (in the first trial DS1). The oil content of this study are also consistent with some of the previous studies, appropriate sowing date is important factor because of it ensures the better fruits germination situation and fruits can maximize the plant to make the best use of natural resource, as well as the emergence of seedlings timely and optimal development of the root system (Lakshminarayana et al., 1981). Gross (1964) reported that the oil content and fatty acid components show variation depending upon sowing time in the most crops and sowing time significantly affected growth of plant, furthermore, days to flowering and flowering duration, while fruits yield reduced with delayed sowing time (Gross, 1964). The same trend was also determined by other researchers (Chen et al., 2011; Hocking and Stapper, 2001; Trémolières et al., 1978; Uzun et al., 2009)

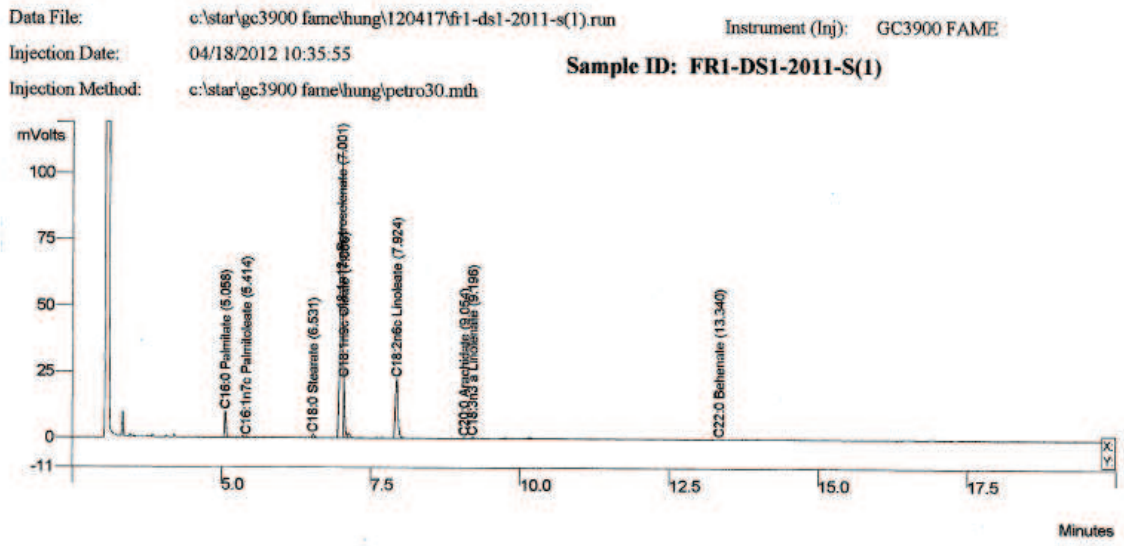


Figure 28: Chromatogram showing fatty acid composition of coriander fruits analyzed by GC (Gas chromatography)

The results indicated that nice fatty acid of coriander fruits was identified as C16:0 (Palmitic acid), C16:1n7 (Palmitoleic acid), C18:0 (Stearic acid), C18:1n12 (Petroselinic acid), C18:1n9 (Oleic acid), C18: 2n6 (Linoleic acid), C18:3n3 (Linolenic acid), C20:0 (Arachidic acid), C22:0 (Behenic acid), of which more than 85% of the fatty acid were unsaturated. The unsaturated fatty acid ratio is important in medicinal and nutritional aspects (Roche et al., 2010). Petroselinic acid was the major fatty acid in both three sowing dates (DS1, DS2 and DS3). In first trial (DS1), the petroselinic acid reach a highest account 74.33 ± 1.42%, followed by linoleic, oleic and palmitic acids, accounting for 14.67 ± 0.12%; 5.86 ± 1.03% and 3.81 ± 0.19, respectively, of the total fatty acids were identified. In second and third trial (DS2 and DS3), this results showed that delaying sowing date obtained percentages of petroselinic acid significant reduction compared with the first sowing day (DS1). Petroselinic acid decrease significantly from 74.33 ± 1.42% in DS1; 71.30 ± 0.56% in DS2 and to 67.83 ± 0.42% in DS3. In contrary, the percentage of palmitic, oleic and linoleic acid have trend increase at DS2, DS3, respectively. Oleic acid increase from 5.86 ± 1.03% on DS1 to 6.64 ± 0.65% on DS2 and to 7.21 ± 0.67% on DS3, respectively. Similarly, linoleic acid increase from 14.67 ± 0.12% on DS1 to 16.23 ± 0.15% on DS2 and to 18.07 ± 0.31% on DS3, respectively. From this results, we can see that this study revealed that different environmental parameters, humidity, rainfall will be significant impacted in both fruits oil content and fatty acid composition in coriander. Besides, this results can be explained by high day temperatures can cause damages to

components of leaf photosynthesis, reducing carbon dioxide assimilation rates and plant height compared to environments having closer to optimal temperatures.

Table 6: Fatty acid composition of coriander fruit on different sowing time (2011)

Fatty acid (%)	Sowing dates		
	DS1	SD2	DS3
Saturated fatty acid (SFA)			
C16:0	3.81 ± 0.19[a]	4.11 ± 0.45[a]	4.57 ± 0.56[a]
C18:0	0.74 ± 0.06[a]	0.81 ± 0.11[a]	1.01 ± 0.16[a]
C20:0	0.13 ± 0.01[a]	0.17 ± 0.04[a]	0.22 ± 0.05[a]
C22:0	0.13 ± 0.01[a]	0.20 ± 0.06[a]	0.25 ± 0.07[a]
Total	4.81 ± 0.26[a]	5.29 ± 0.66[a]	6.05 ± 0.84[a]
Monounsaturated fatty acid (MUFA)			
C16:1n7	0.19 ± 0.01[c]	0.21 ± 0.01[b]	0.23 ± 0.01[a]
C18:1n12	74.33 ± 1.42[a]	71.30 ± 0.56[b]	67.83 ± 0.42[c]
C18:1n9	5.86 ± 1.03[c]	6.64 ± 0.65[b]	7.21 ± 0.67[a]
Total	80.38 ± 0.41[a]	78.16 ± 0.50[b]	75.28 ± 0.55[c]
Polyunsaturated fatty acid (PUFA)			
C18: 2n6	14.67 ± 0.12[c]	16.23 ± 0.15[b]	18.07 ± 0.31[a]
C18:3n3	0.15 ± 0.01[c]	0.36 ± 0.01[b]	0.61 ± 0.01[a]
Total	14.82 ± 0.12[c]	16.60 ± 0.15[b]	18.67 ± 0.31[a]
SFA/PUFA	0.32 ± 0.01[a]	0.32 ± 0.04[a]	0.32 ± 0.04[a]
Water content (%)	10.36 ± 0.35[a]	11.66 ± 0.28[a]	11.81 ± 0.23[a]
Oil yield (%)	24.26 ± 0.14[a]	20.19 ± 0.69[b]	17.36 ± 0.23[c]

Data presented as Mean ± SD; Numbers on the same line with differing superscripts are significantly different at $P<0.05$; C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (Stearic acid); C18:1n12 (Petroselinic acid); C18:1n9 (Oleic acid); C18: 2n6 (Linoleic acid); C18:3n3 (Linolenic acid); C20:0 (Arachidic acid); C22:0 (Behenic acid).

In summary, sowing date is one of the important factors influencing fatty acid composition and oil yield of coriander fruits. There were significant differences of oil content between three sowing times, DS1 showed the highest values of oil yields as compared with significant differences in yield of DS2 and DS3. The sowing date effect with yield and fatty acid showed there was meaningful effect. The sowing date is determined to provide the conditions for the plants to maximally use environmental parameters, especially temperature and radiation. If environmental factors are appropriate for the greening, establishment and survival of seedlings, then the maximum yield can be realized by

appropriate sowing date. Moreover, establish of appropriate sowing date will be useful for healthy plants in a farm surface is a basic element of a successful agricultural system.

II.3 Fatty acid accumulation in maturity of coriander

French coriander cultivar (FR1 - Dourou) was cultivated under the organic agriculture during the cropping season (2009, 2010 and 2011). Sowing were made on 23rd March, 9th April and 25th March in 2009, 2010 and 2011, respectively. Fruits samplings were performed each five (average) days between beginning of flowering and physiological maturity. Harvest period was started from 2 days after flowering (DAF) to 59 DAF to evaluate fatty acids accumulation in maturity of coriander.

The changes in oil yield of coriander during from flowering period to maturity in 2009 (53 days) were showed in the Table 7. As shown in Table 7, water content decreased markedly from flowering to maturity and the oil yield has been found to vary between 4.6% - 25.1% at different stages of fruit ripening. Oil content increased gradually from flowering to maturity. Nevertheless, there were significantly marked changes in this trait. Indeed, oil content increased three times from 2 DAF to 12 DAF. At the time of fruit maturity stage, oil content reached the highest (25.1%) (Figure 29). The value of oil yield in the mature stage of ripening was slightly lower than those previously reported for coriander under conventional agriculture (Msaada et al., 2009a; Ramadan and Mörsel, 2002; Sriti et al., 2010b) but quite higher than values of study of Angelini (Angelini et al., 1997). This result was expected since organic agriculture was considered as stressed conditions (Alignan et al., 2009). Moreover, it is well known than oil seed produce and accumulate less oil under drought than under favorable conditions (Carrubba, 2014; Roche et al., 2006). The oil yield increased very rapidly from the 10th to 34th day after the flowering sampling period and reached its maximum level at maturity, the oil yield was maximal and its value was similar to others reports dealing with mature coriander fruit (Lakshminarayana et al., 1981; Msaada et al., 2009a). The evolutionary trend in oil accumulation in coriander fruit was similar to results reported in Brassicaceae (Chen et al., 2011; Trémolières et al., 1978), Asteraceae (Lardans and Trémolières, 1991; Roche et al., 2010; Roche et al., 2006).

Important accumulation fatty acids initiated from the 2 DAF. Different trends were observed for the detected fatty acids. Content of saturated fatty acids was high from 2 DAF until 10 DAF and fell over twice at 12 DAF. This decline continued until full maturity. The representative fatty acid of this category in coriander was palmitic acid which followed the

same trend as saturated fatty acids. Myristic and stearic acids were also observed with a higher amounts noted at 2 DAF and decreased after 10 DAF (Table 7). In contrast, Monounsaturated fatty acids represented mainly by petroselinic acid (50% at 2 DAF and reached more than 92% at maturity) were present at low amount in the beginning of fruits formation (2 DAF) and increased from 10 DAF. Indeed, petroselinic acid amount increased ten times from 2 DAF to 12 DAF and continued this rise reaching its highest level at 18 DAF. Polyunsaturated fatty acids content was higher at 2 DAF and decreased until 18 DAF reaching a stable amount from this period to maturity. Our results were in agreement with those reported in literature. Higher levels of polyunsaturated and saturated fatty acids have been already reported at earlier stages of fruit ripening (Lakshminarayana et al., 1981; Msaada et al., 2009a). The ratio of saturated fatty acids to polyunsaturated fatty acids significantly decreased during fruit maturation. Similar results have been reported in other oilseed species (Chen et al., 2011; Peiretti et al., 2004; Roche et al., 2006; Vuorinen et al., 2014). The level of petroselinic acid was in accordance with results already reported in coriander ranging from 51.6 to 90.7% (Angelini et al., 1997; Guidotti et al., 2006; Lakshminarayana et al., 1981; Msaada et al., 2009a). The highest amount of petroselinic acid was reached between 18 and 35 DAF. This result agreed with report of Msaada (Msaada et al., 2009a) which emphasized that a period of 32 DAF was sufficient to use of coriander fruits. In our study, an opposed evolutionary trends was observed between palmitic acid (decrease with fruit ripening) and petroselinic acid (increase with fruit ripening), the tendency of increasing or decreasing of major fatty acid was represented in Figure 30. This fact, as reported by Cahoon et al. (1992) may be explained by the fact that palmitic acid served as precursor of petroselinic acid (Cahoon et al., 1992).

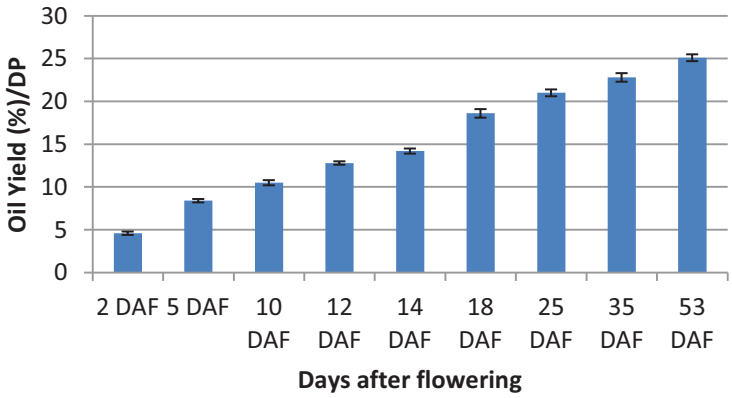


Figure 29: The changes in oil yield of coriander during from flowering period to maturity in 2009

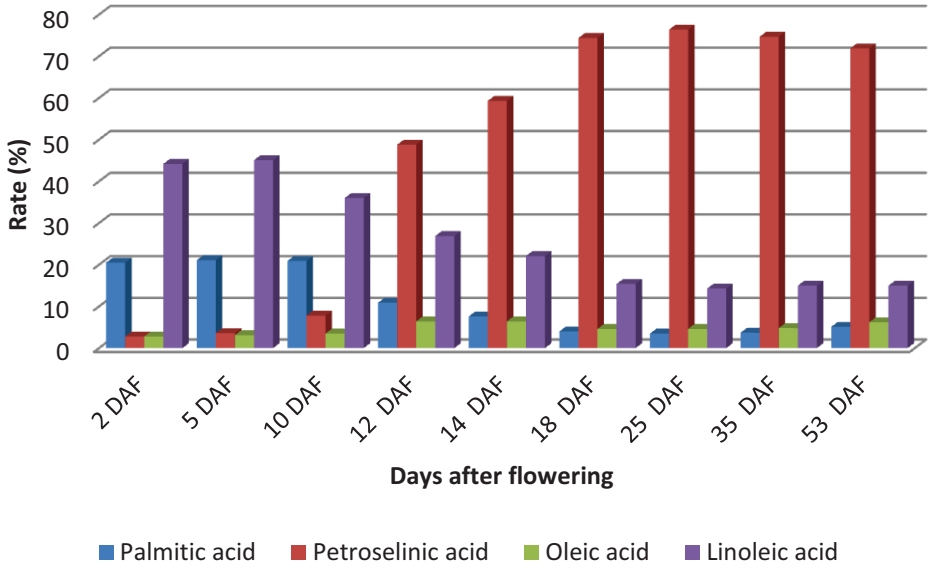


Figure 30: Variations of major fatty acid from flowering period to maturity (in Auch, 2009)

The changes in oil content and fatty acid composition during coriander fruits development and maturation cultivated in 2010 and 2011 are shown in Table 8 and Table 9. Significant changes ($P < 0.05$) were observed among the studied at different stages of fruit ripening for oil content during fruit development. Similarly as observed 2009, water content decreased markedly from flowering to maturity and oil content increased gradually from flowering to maturity, the oil yield increased very rapidly from the 10th to 35th day after the flowering sampling period and reached its maximum level at maturity, the oil yield has been found to vary between 4.6% - 25.1% , 4.79% - 24.34% and 5.0% - 24.26% at different stages of fruit ripening in 2009, 2010 and 2011, respectively. Oil content were similar in amounts and little change was observed between each year and oil content obtained the highest at full maturity in 2009, 2010 and 2011: 25.1 ± 0.4% (53 DAF), 24.34 ± 0.02% (55 DAF); 24.26 ± 0.14% (59 DAF), respectively (Figure 31). Similar this results were found in some previous studied (Msaada et al., 2009b; Ramadan and Mörsel, 2002).

Considerable variation was observed among the fatty acid profiles for the fourteen stages of maturity in 2010, and six stages in 2011 are presented in Table 8 and Table 9. Total of 10 different fatty acids were identified in percentages of the total fatty acid of the fruit oil. Similar fatty acids composition in 2009, the early stages (2 DAF), the content of saturated fatty acid was highest in early stages (2th DAF) and the content of this saturated was dramatically decreased until maturity. In contrary, the monounsaturated fatty acids increased gradually from early stages to full maturity (55 - 59 DAF).

Table 7: Changes on fatty acid and oil yield during fruit maturation of Coriander

Fatty acid (%)	Days after flowering						
	2	5	10	12	14	18	
Saturated fatty acid (SFA)							
C14:0	4.6 ± 0.9^{a}	4.2 ± 0.7^{a}	3.6 ± 0.4^{a}	1.6 ± 0.3^{b}	1.6 ± 0.4^{b}	0.9 ± 0.2^{c}	0.3
C16:0	20.5 ± 0.7^{a}	21.1 ± 1.1^{a}	20.9 ± 1.3^{a}	10.9 ± 0.8^{b}	7.6 ± 0.9^{c}	4.0 ± 0.6^{de}	3.5
C18:0	5.8 ± 0.5^{a}	$5.1^{a} \pm 0.3^{a}$	$6.2^{a} \pm 0.3^{a}$	$1.7^{b} \pm 0.2^{b}$	$1.4^{b} \pm 0.1^{b}$	$0.6^{c} \pm 0.1^{c}$	0.8
C20:0	0.1 ± 0.0^{a}	0.2 ± 0.2^{a}	0.3 ± 0.1^{a}	0.3 ± 0.1^{b}	0.3 ± 0.1^{b}	0.2 ± 0.0^{c}	0.1
C22:0	1.6 ± 0.5^{b}	1.5 ± 0.3^{b}	4.0 ± 0.6^{a}	-	-	-	
Total	32.6 ± 1.4^{a}	32.0 ± 1.5^{a}	34.9 ± 1.3^{a}	14.4 ± 0.7^{b}	10.8 ± 0.7^{c}	5.7 ± 0.3^{d}	4.6
Monounsaturated fatty acid (MUFA)							
C18:1n12	2.8 ± 0.3^{e}	3.6 ± 0.4^{e}	7.8 ± 0.9^{d}	48.8 ± 1.5^{c}	59.3 ± 1.7^{b}	74.4 ± 2.1^{a}	76.4
C18:1n9	2.8 ± 0.2^{d}	3.1 ± 0.2^{cd}	3.5 ± 0.5^{c}	6.4 ± 0.9^{a}	6.4 ± 0.7^{a}	4.6 ± 0.5^{b}	4.6
Total	5.6 ± 0.6^{e}	6.7 ± 0.6^{e}	11.3 ± 0.8^{d}	55.2 ± 1.2^{c}	65.7 ± 1.9^{b}	79.1 ± 1.8^{a}	81.1
Polyunsaturated fatty acid (PUFA)							
C18: 2n6	44.2 ± 2.1^{a}	45.1 ± 1.9^{a}	36.0 ± 1.8^{b}	26.9 ± 1.5^{c}	22.1 ± 1.6^{d}	15.4 ± 1.0^{e}	14.3
C18:3n3	17.6 ± 0.9^{b}	20.3 ± 1.1^{a}	21.4 ± 1.6^{a}	5.2 ± 0.5^{c}	3.0 ± 0.1^{d}	0.7 ± 0.0^{e}	0.3
Total	61.8 ± 2.2^{a}	65.4 ± 2.3^{a}	57.4 ± 1.9^{b}	32.0 ± 1.6^{c}	25.1 ± 1.5^{d}	16.1 ± 2.1^{e}	14.6
SFA/PUFA	0.5 ± 0.1^{a}	0.5 ± 0.1^{a}	0.6 ± 0.1^{a}	0.5 ± 0.1^{a}	0.4 ± 0.1^{ab}	0.4 ± 0.0^{ab}	0.3
Water content (%)	95.3 ± 3.4^{a}	90.3 ± 4.5^{a}	80.6 ± 3.9^{b}	74.8 ± 3.4^{b}	74.0 ± 5.6^{b}	63.9 ± 2.9^{c}	55.1
Oil yield (%)	4.6 ± 0.2^{i}	8.4 ± 0.2^{h}	10.5 ± 0.3^{g}	12.8 ± 0.2^{f}	14.2 ± 0.3^{e}	18.6 ± 0.5^{d}	21.0

Data were means ± S.D of three replicates: Numbers on the same line with differing superscripts are signi
detected. C14:0 (Myristic acid); C16:0 (Palmitic acid); C18:0 (Stearic acid); C18:1n12 (Petroselinic aci
(Linoleic acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22:0 (Behenic acid).

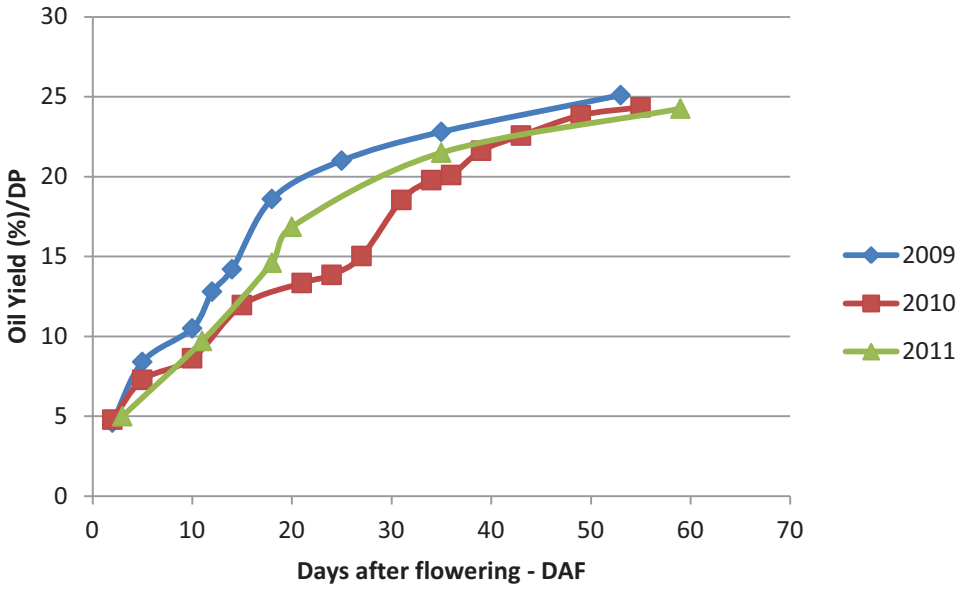


Figure 31: Variations of oil yields of coriander from flowering period to maturity (2009, 2010, 2011)

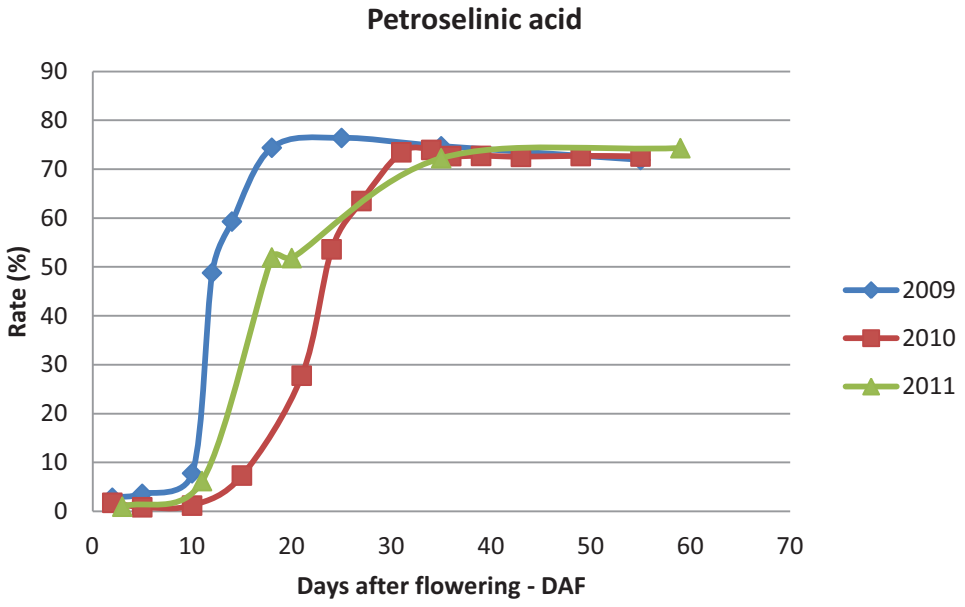


Figure 32: The tendency of petroselinic acid from newly form fruits to maturity (2009, 2010, 2011)

During fruit developments, the fatty acid composition of all samples is shown in Table 7, 8 and 9. Tendency of increasing or decreasing of the major fatty acid components are relatively similar in three year (2009, 2010 and 2011), as can be seen in Figure 30, Figure 33 and Figure 34. In which petroselinic acid is the highest at full maturity fruits, as 76.4 ± 2.1%, 73.95 ± 0.35% and 74.32 ± 1.43% in 2009, 2010 and 2011 respectively and trend of

petroselinic acid increase continuously from newly form fruits to fruit ripening (Figure 32). Petroselinic acid amount detected in this our work of three year is quite similar to that previous studies (Angelini et al., 1997; Guidotti et al., 2006; Lakshminarayana et al., 1981; Msaada et al., 2009a). Petroselinic acid is metabolized and accumulated in the developing endosperm of some Apiaceae species, including coriander and carrot. This fatty acid is the product of the acyl carrier protein Δ^6 desaturase (ACP Δ^6 desaturase) activity. This polypeptide is highly expressed in seed but is absent in tissues that do not biosynthesize petroselinic acid, including leaves and roots of coriander (Cahoon and Ohlrogge, 1994; Cahoon et al., 1992). Furthermore, secondary metabolites in plants are under the influence of synthesis and biotic factors, and variations are observed depending on the different periods (in newly formed fruit to full maturation). In the same study of three year 2009, 2010 and 2011 have shown that the effect according to the harvest periods between the harvest dates of the studied samples despite the difference of about minimum 3 days. From the results obtained, it can be said that the most abundant fatty acid petroselinic acid contents tend significant increase and reached the highest rate in the mature stage of fruit while myristic, palmitic, linoleic, linolenic acid continuous decrease from early stage to full maturity of fruits, increasing or decreasing in the fatty acid amounts is dependent on maturity fruits.

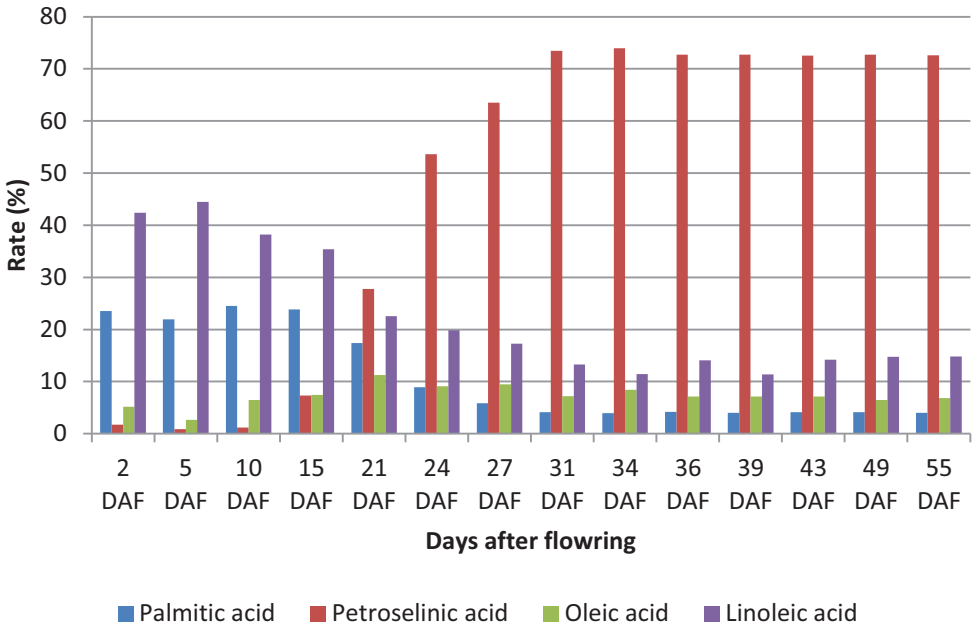


Figure 33: Variations of major fatty acid from flowering period to maturity (in Auch, 2010)

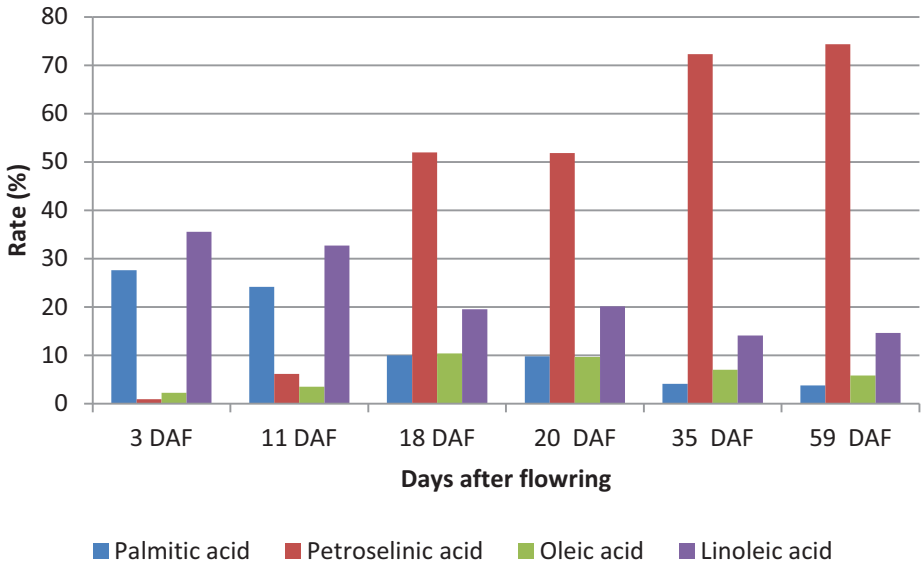


Figure 34: Variations of major fatty acid from flowering period to maturity (in Auch, 2011)

Besides, the Vietnamese coriander cultivar (Vinaseed - VS1) were selected in order to study the fatty acid accumulation in maturity of coriander that was cultivate under the organic agriculture during the cropping season 2012 in Ha Noi (Viet Nam). Sowing was made on the first week of March, 2012. Fruits samplings were performed each ten (average) days between beginning of flowering and physiological maturity. Harvest period was started from 2 days after flowering (DAF) to 55 DAF to evaluate fatty acids accumulation in maturity of coriander.

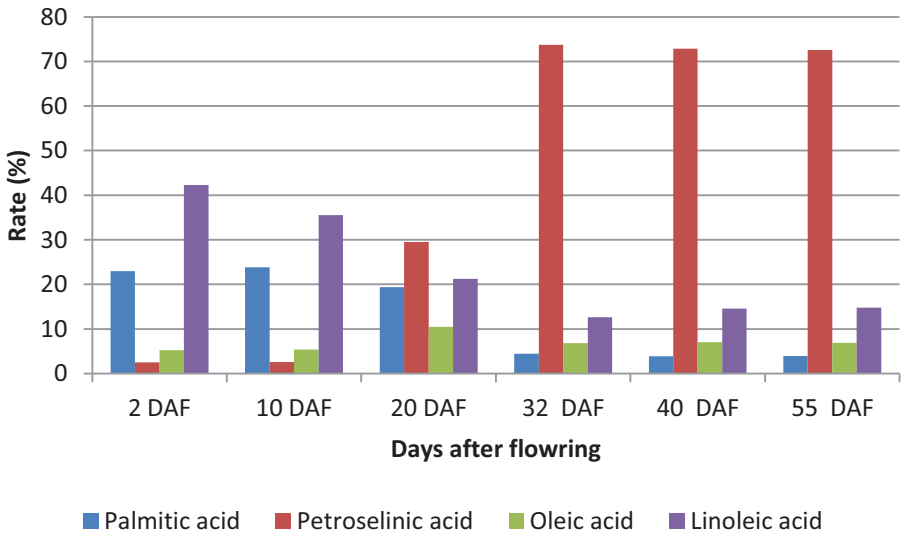


Figure 35: Variations of major fatty acid from flowering period to maturity (in Vietnam, 2012)

Table 8: Changes on fatty acid and oil yield during fruits maturation of Coriand

Fatty acid (%)	Days after flowering									
	2	**5**	**10**	**15**	**21**	**24**	**27**	**31**	**34**	**36**
Saturated (SFA)										
C14:0	3.78±0.48^{e}	4.44±0.38^{d}	7.36±0.24^{c}	8.60±0.16^{b}	11.73±0.06^{a}	4.58±0.03^{d}	0.99±0.03^{f}	0.14±0.01^{g}	0.11±0.01^{g}	0.47±0.01
C16:0	23.52±0.42^{b}	21.93±0.30^{c}	24.52±0.14^{a}	23.84±0.25^{b}	17.39±0.15^{d}	8.92±0.10^{e}	5.83±0.03^{f}	4.15±0.01^{g}	3.97±0.01^{g}	4.18±0.01
C18:0	2.62±0.03^{d}	2.76±0.02^{c}	2.74±0.02^{c}	3.42±0.04^{a}	3.17± 0.02^{b}	1.45±0.05^{e}	1.03±0.03^{f}	0.91±0.00^{g}	0.77± 0.01^{g}	0.71±0.01
C20:0	0.86±0.01^{d}	1.28±0.07^{c}	1.51±0.01^{a}	1.55±0.07^{a}	1.41±0.05^{b}	0.54±0.00^{e}	0.36±0.00^{f}	0.14±0.01^{g}	0.13±0.01^{g}	0.16±0.00
C22:0	1.49±0.04^{b}	1.89±0.03^{a}	1.44±0.05^{b}	1.27±0.05^{c}	0.90±0.01^{d}	0.37±0.01^{e}	0.37±0.01^{e}	0.26±0.01^{f}	0.22±0.01^{f}	0.15±0.01
Total	32.27±0.11^{d}	32.30±0.19 ^{d}	37.57±0.03^{b}	38.69±0.13^{a}	34.59±0.23^{c}	15.86±0.12^{e}	8.58±0.05^{f}	5.59±0.01^{gh}	5.20±0.02^{i}	5.67±0.01
Monounsaturated fatty acid (MUFA)										
C16:1n7	-	-	-	-	-	-	-	0.20±0.00^{b}	0.40±0.00^{a}	0.2±0.01^{b}
C18:1n12	1.76±0.94^{g}	0.85±0.28^{g}	1.19±0.07^{g}	7.34±0.03^{f}	27.77±0.20^{e}	53.62±0.15^{d}	63.50±0.25^{c}	73.48±0.51^{ab}	73.95± 0.35^{a}	72.70±0.38
C18:1n9	5.15±0.21^{d}	2.64±0.27^{e}	6.45±0.10^{c}	7.44±0.23^{c}	11.27±0.11^{a}	9.09±0.17^{b}	9.45±0.30^{b}	7.20±0.49^{c}	8.46± 0.37^{b}	7.17±0.38
Total	6.91±1.07^{h}	3.49±0.52^{i}	7.64±0.17^{h}	14.78±0.21^{g}	39.04±0.29^{f}	62.71±0.06^{e}	73.38±0.08^{d}	80.88±0.04^{b}	82.81±0.02^{a}	80.07±0.01
Polyunsaturated fatty acid (PUFA)										
C18: 2n6	42.38±0.65^{b}	44.45±0.50^{a}	38.22±0.08^{c}	35.38±0.08^{d}	22.55±0.07^{e}	19.87±0.07^{f}	17.29±0.04^{g}	13.31±0.03^{k}	11.43± 0.01^{l}	14.07±0.02
C18:3n3	18.44±0.37^{b}	19.76±0.16^{a}	16.56±0.11^{c}	11.15±0.04^{d}	3.82±0.04^{e}	1.56±0.01^{f}	0.74±0.01^{g}	0.22±0.01^{h}	0.56±0.01^{g}	0.19±0.01
Total	60.82±1.03^{b}	64.21±0.64^{a}	54.79±0.19^{c}	46.53±0.11^{d}	26.37±0.11^{e}	21.43±0.07^{f}	18.03±0.04^{g}	13.53±0.03^{i}	11.99±0.0^{j}	14.26±0.01
SFA/PUFA	0.53±0.01^{e}	0.50±0.01^{f}	0.69±0.01^{d}	0.83±0.01^{b}	1.31±0.01^{a}	0.74±0.01^{c}	0.48±0.01^{g}	0.41±0.01^{i}	0.43±0.01^{h}	0.40±0.01
Water content (%)	95.11±0.12^{a}	90.73±0.7^{b}	75.74±0.07^{c}	69.92±0.08^{d}	61.81±0.53^{e}	56.33±0.32^{f}	47.29±0.18^{g}	34.23±0.37^{h}	31.12±0.14^{i}	30.83±0.04
Oil yield (%)	4.79±0.06^{k}	7.28±0.01^{j}	8.63±0.04^{i}	11.96±0.04^{h}	13.34±0.01^{g}	13.85±0.01^{g}	15.03±0.12^{f}	18.54±0.27^{e}	19.78±0.36^{d}	20.1±0.03

Data were means ± S.D of three replicatesm. Numbers on the same line with differing superscripts are significant
C14:0 (Myristic acid); C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (Stearic acid); C18:1n12 (Petros
2n6 (Linoleic acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22:0 (Behenic acid).

Table 9: Changes on fatty acid and oil yield during fruits maturation of Coriand

Fatty acid (%)	Days after flowering			
	3	11	18	20
Saturated fatty acid (SFA)				
C14:0	5.85 ± 0.01[b]	8.93 ± 0.41[a]	2.96 ± 0.13[c]	2.93 ± 0.03[c]
C16:0	27.60 ± 0.16[a]	24.16 ±1.05[b]	10.01 ± 0.05[c]	9.81 ± 0.23[c]
C18:0	3.93 ± 0.05[a]	3.31 ± 0.05[b]	1.93 ± 0.11[c]	1.92 ± 0.02[c]
C20:0	2.42 ± 0.31[b]	5.90 ± 0.05[a]	0.81 ± 0.02[c]	0.73 ± 0.03[c]
C22:0	2.27 ± 0.03[a]	1.80 ± 0.09[b]	0.63 ± 0.01[c]	0.57 ± 0.01[c]
Total	42.07 ± 0.08[b]	44.10 ± 1.59[a]	16.33 ± 0.21[c]	15.96 ± 0.28
Monounsaturated fatty acid (MUFA)				
C16:1n7	0.36 ± 0.12[b]	0.74 ± 0.13[a]	0.28 ± 0.03[b]	0.3 ± 0.01[b]
C18:1n12	0.97 ± 0.25[d]	6.21 ± 3.09[c]	51.95 ± 0.60[b]	51.83 ± 0.28
C18:1n9	2.29 ± 0.16[c]	3.54 ± 0.93[c]	10.44 ± 0.44[a]	9.68 ± 0.24[a]
Total	3.62 ± 0.32[d]	10.49 ± 3.92[c]	62.66 ± 0.18[b]	61.81 ± 0.47
Polyunsaturated fatty acid (PUFA)				
C18: 2n6	35.59 ± 0.20[a]	32.70 ± 1.45[b]	19.58 ± 0.02[c]	20.13 ± 0.10
C18:3n3	18.72 ± 0.05[a]	12.71 ± 0.90[b]	1.43 ± 0.04[c]	2.10 ± 0.09[c]
Total	54.31 ± 0.25[a]	45.41 ± 2.34[b]	21.01 ± 0.04[c]	22.23 ± 0.19
SFA/PUFA	0.77 ± 0.01[b]	0.97 ± 0.02[a]	0.78 ± 0.01[b]	0.72 ± 0.01[c]
Water content (%)	94.29 ± 0.12[a]	81.80 ± 1.06[b]	66.16 ± 0.75[c]	58.45 ± 1.13
Oil yield (%)	5.0 ± 0.08[f]	9.71 ± 0.17[e]	14.61 ± 0.07[d]	16.86 ± 0.05

Data were means ± S.D of three replicates: Numbers on the same line with differing superscripts are signi
detected. C14:0 (Myristic acid); C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (Stearic ac
C18:1n9 (Oleic acid); C18: 2n6 (Linoleic acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22:0

Changes in oil content, fatty acid composition in the developing fruits are given in Table 10. The oil content from 4.26 ± 0.03% at early stage to 19.36 ± 0.77% at full maturity was observed in these periods of fruit development. This result showed that the oil content of coriander increased gradually from the first to the last fruits development stages. The accumulation pattern of the oil was quite similar in 2009, 2010 and 2011, and the highest oil content was determined at 55 DAF with a value of 19.36 ± 0.77%. The value of oil yield in the mature stage of ripening was slightly lower than those previously reported for coriander under conventional agriculture (Msaada et al., 2009a; Ramadan and Mörsel, 2002; Sriti et al., 2010b). Flowering period was from the first week of May to the first of July 2012, and plus higher air temperatures and higher rainfall may be caused to obtain of low oil content. In this study, fatty acid accumulation resulting from fruits development duration was observed. In each stage of coriander fruits development and maturation, oil yield and some of fatty acid composition increased gradually from early stage to full maturation. Similar with results of major fatty acid of coriander in Auch, petroselinic acid, linoleic and oleic acid was major fatty acid at maturity. There were a little abrupt fluctuation with petroselinic acid, oleic and myristic acid. Trend of variation of major fatty acid was represented in Figure 35. Petroselinic acid increase gradually from the first until to 32 DAF, and then started to lightly decline from 73.74 ± 0.09% to 72.55 ± 0.07%. Similarly, oleic acid and myristic acid increase significantly to 20 DAF, after which it started to decline gradually from 10.51% to 6.89%; 10.05% to 0.23%, respectively. A rapid increase in the synthesis of petroselinic, oleic acid and a rapid decrease in the synthesis of linoleic, palmitic, arachidic and behenic were observed during the development of coriander fruits.

In summary, this is the preliminary report to study the oil and fatty acid accumulation of coriander during fruit ripening cultivated under organic conditions in three seasons in Auch and one season in Vietnam. Our results demonstrated that the highest oil yield was achieved at the full maturity. Fatty acid profiles varied greatly during fruit ripening. At earlier stages, saturated and polyunsaturated fatty acids were higher and decreased with maturity of fruit. petroselinic acid was the major fatty acid after 12 DAF. This latter showed an inversely evolution with palmitic acid which may support a functional correlation between both fatty acids. This study provided data for use coriander oil and its composition in fatty acids for different industrial applications due its content in petroselinic acid.

Table 10: Changes on fatty acid and oil yield during fruits maturation of coriande

Fatty acid (%)	Days after flowering			
	2	10	20	32
Saturated fatty acid (SFA)				
C14:0	3.97 ± 0.43^{c}	7.68 ± 0.54^{b}	10.05 ± 0.49^{a}	$0.17 \pm 0.$
C16:0	23.01 ± 0.71^{a}	23.85 ± 0.32^{a}	19.39 ± 0.13^{b}	$4.45 \pm 0.$
C18:0	2.59 ± 0.19^{b}	3.71 ± 0.36^{a}	3.25 ± 0.30^{a}	$0.94 \pm 0.$
C20:0	1.31 ± 0.25^{b}	2.90 ± 0.20^{a}	1.40 ± 0.13^{b}	$0.15 \pm 0.$
C22:0	2.14 ± 0.42^{a}	2.21 ± 0.30^{a}	0.83 ± 0.05^{b}	$0.26 \pm 0.$
Total	33.03 ± 0.66^{c}	40.38 ± 0.30^{a}	34.95 ± 0.54^{b}	$5.98 \pm 0.$
Monounsaturated fatty acid (MUFA)				
C16:1n7	0.37 ± 0.15^{bc}	0.88 ± 0.09^{a}	0.29 ± 0.10^{bc}	$0.53 \pm 0.$
C18:1n12	2.49 ± 0.22^{d}	2.58 ± 0.19^{d}	29.50 ± 0.63^{c}	73.74 ± 0
C18:1n9	5.27 ± 0.42^{c}	5.42 ± 0.27^{c}	10.51 ± 0.32^{a}	$6.82 \pm 0.$
Total	8.15 ± 0.64^{d}	8.01 ± 0.38^{d}	40.02 ± 0.34^{c}	81.10 ± 0
Polyunsaturated fatty acid (PUFA)				
C18: 2n6	42.30 ± 0.67^{a}	35.51 ± 0.11^{b}	21.25 ± 0.55^{c}	12.67 ± 0
C18:3n3	17.98 ± 0.93^{a}	15.20 ± 0.15^{b}	3.47 ± 0.19^{c}	$0.22 \pm 0.$
Total	60.28 ± 1.40^{a}	50.71 ± 0.15^{b}	24.73 ± 0.71^{c}	12.90 ± 0
SFA/PUFA	0.54 ± 0.01^{c}	0.79 ± 0.01^{b}	1.41 ± 0.06^{a}	$0.46 \pm 0.$
Water content (%)	93.52 ± 1.18^{a}	80.60 ± 0.87^{b}	64.28 ± 1.64^{c}	33.69 ± 2
Oil yield (%)	4.26 ± 0.03^{e}	5.89 ± 0.04^{de}	6.93 ± 0.34^{d}	12.57 ± 0

Data were means ± S.D of three replicates: Numbers on the same line with differing superscripts are sign
(Myristic acid); C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (Stearic acid); C18:1n12 (Petro
C18: 2n6 (Linoleic acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22:0 (Behenic acid).

II. 4 Effects of different locations on yield and fatty acid composition of coriander

Coriander is widely cultivated throughout in Vietnam. However, there are no data on the variation of lipid and fatty acid composition among crops from different regions. The aim of this study was to examine the compositions of fatty acids and oil content in coriander fruits grown at different locations. Three locations was selected for planting of coriander, and the fruits harvested from three locations was analyzed to determine effects of different locations on oil yield and fatty acid composition of Vietnamese coriander. Vietnamese coriander (Vinaseed - VS1) was cultivated at three locations: Ha Noi; Thai Binh (Northern of Vietnam - Red River Delta) and Ha Tinh (North central coast of Vietnam). Before planting, the fruits were rubbed for separating the two mericarps and were soaked in water for 24 hours to enhance germination. The fruits were sown in rows 20 cm apart continuously by hand 12 kg ha^{-1}. To allow uniform sowing in rows, fruits were mixed with some loose soil (about four to five times of weight of fruits). The fruits were covered with good pulverized soil just after sowing and gently pressed by hands. The sowing was done on in the first week of March 2012. Harvest of coriander conducted in September 2012 when 100% of the fruits in full maturity stages. The fruits were harvested by hand and were dried naturally in the shadow.

Coriander fruits harvested were analyzed to determine the impact of different location on fatty acid composition and coriander oil. The results of analyses were presented in Table 11. The fatty acids such as C14:0 (Myristic acid), C16:0 (Palmitic acid), C16:1n7 (Palmitoleic acid), C18:0 (Stearic acid), C18:1n12c (Petroselinic acid), C18:1n9c (Oleic acid), C18: 2n6c (Linoleic acid), C20:0 (Arachidic acid), C18:3n3 a (Linolenic acid), C22:0 (Behenic acid) were indentified. According to the analyses of data, oil content of coriander were significantly affected by growth location and oil content varied 17.23% - 19.19% depending on location, the highest oil rate was observed in Ha Noi (Table 11). The temperature Ha Noi was higher than Thai Binh and Ha Tinh in the flower season of coriander (from May to July 2012), this is may be causes high oil yield of the coriander plant in Hanoi.

The results obtained from the preliminary analysis of are presented Table 11, it can be seen that saturated and unsaturated fatty acids in coriander oil changed significantly depending on growing location.

Table 11: Effects of different locations on yield and fatty acid composition of coriander fruits (Vie

Fatty acid (%)	Different locations of	
	Ha Noi	Thai Binh
Saturated fatty acid (SFA)		
C14:0	0.23 ± 0.01[c]	0.33 ± 0.02
C16:0	3.99 ± 0.08[c]	4.30 ± 0.05
C18:0	0.83 ± 0.01[a]	0.87 ± 0.05
C20:0	0.12 ± 0.01[b]	0.19 ± 0.02
C22:0	0.12 ± 0.01[c]	0.35 ± 0.02
Total	5.30 ± 0.08[b]	6.07 ± 0.05
Monounsaturated fatty acid (MUFA)		
C16:1n7	0.19 ± 0.01[a]	0.23 ± 0.02
C18:1n12	72.55 ± 0.07[a]	70.27 ± 0.3
C18:1n9	6.89 ± 0.12[a]	6.72 ± 0.06
Total	79.64 ± 0.11[a]	77.23 ± 0.3
Polyunsaturated fatty acid (PUFA)		
C18: 2n6	14.76 ± 0.05[b]	16.35 ± 0.3
C18:3n3	0.28 ± 0.01[a]	0.34 ± 0.05
Total	15.04 ± 0.07[c]	16.69 ± 0.3
SFA/PUFA	0.35 ± 0.01[a]	0.36 ± 0.01
Water content (%)	10.42 ± 0.47[a]	10.09 ± 0.0
Oil yield (%)	19.19 ± 0.24[a]	18.31 ± 0.7

Data were means ± S.D of three replicates: Numbers on the same line with differing superscripts are sign
(Myristic acid); C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (Stearic acid); C18:1n12 (Petro
C18: 2n6 (Linoleic acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22:0 (Behenic acid).

The saturated fatty acid in this oil was palmitic (ranging from 3.99 to 4.69%). The other four minor fatty acids were myristic acid (ranging from 0.23 to 0.44 %), stearic acid (ranging from 0.83 to 0.97%), arachidic acid (ranging from 0.12 to 0.19 %) and behenic acid (ranging from 0.12 to 0.35%). The two main monounsaturated fatty acids were petroselinic acid (ranging from 68.96 to 72.55 %) and oleic acid (ranging from 6.72 to 6.92 %). There are four essential fatty acids, petroselinic acid, linoleic acid, oleic acid and palmitic acid were the significant fatty acids in terms of quality and quantity of coriander fruits oils, in which most predominant fatty acid was petroselinic acid (68.96 to 72.55 %) of total fatty acid.

This result was in agreements with Msaasa et. al (2009) who studied on effect of oil yield and fatty acid composition during maturation of coriander fruits cultivated in different locations (in Menzel Temime and Oued Beja, Tunisia). They notes that rapid accumulation of oil started at newly formed fruits and continued till the full maturity of fruits. Their result showed that during fruit maturation, fatty acid profiles varied significantly among the growing regions and stages of maturity. Fatty acid profile of fruits at full maturity cultivated in Oued Beja showed that petroselinic acid (80.90%) was the main component followed by oleic (14.79%), palmitic (3.50%) and stearic (0.49%) acids. While in Menzel Temime, at full maturity, the main fatty acids were petroselinic acid (80.86%) followed by oleic (14.83%), palmitic (3.27%) and stearic (0.31%) acids (Msaada et al., 2009b). From this results, we can see that this study revealed that different ecological especially temperature, humidity and rainfall will be significant impacted in both fruit oil content and fatty acid composition in coriander. Therefore, growth location is an important factor for meeting market requirements of coriander fruits in terms of oil quality. Apart from growing condition, one should also consider some other factors such as cultivar, planting time and irrigation, cultivation mode in order to obtain a desire yield and quality.

II.5 Fatty acid composition of different cultivars

This study was performed to evaluate coriander cutivars pertaining to its fruit yield performance in Auch. It is necessary to develop more suitable cultivars for fruits production to fulfill the increasing demand of this spice crop. Selection of better cultivars can be of immense value to the breeder for further improvement and development of the crop. Therefore, the present investigation was undertaken in order to evaluate the appropriate conditions of the collected coriander cultivars and to select the promising

cultivars for higher fruit yield in France. The experiment was conducted under organic condition in Auch, during March to September 2011. The different seven coriander cultivars (Canada, Tunisia, Lithuania, Algeria, French (FR1 (Dourou) - DS1), Vietnam (VS2) and China) was grown and the raw material harvested on the fully maturity stage (September 2011) was studies.

Data for coriander cultivars fruits were analyzed to determine the effect of conventional agriculture on fatty acid composition of coriander oil and the results of analyses were presented in Table 12. According to variance analyses of data, oil content varied 15.28 - 24.26% depending on the different cultivars. Among the seven coriander cultivars, the oil content of the three coriander cultivars (Algeria, Canada and China) was relatively lower, ranging from 15.28 % to 16.87 % (Table 12), French coriander had the highest oil content (up to 24.26%), followed by Lithuanian (19.04%), Tunisian (18.70%) and Vietnamese (17.64 %) cutivars. However, the oil yield in our study was in agreement with the value previous reported by Msaada (Msaada et al., 2009a). Diederichsen (1996) noted that the coriander fruit presented an oil yield ranging from 9.9% to 27.7% (Diederichsen, 1996); others reported values of 28.4% (Ramadan and Mörsel, 2002), 13–18% (Khan et al., 1986) and 17% (Griffiths et al., 1992).

Fatty acids composition of maturity fruits of cultivars are shown in Table 12. For almost fatty acids, the varietal difference was not as significant as the difference among species. However, the cultivars have qualitatively similar fatty acid compositions, quantitative variation in major fatty acids were significant statistically in the research. Similar to oil fruits of coriander studied, all seven coriander cultivars contained major fatty acids including palmitic (C16:0), petroselinic (C18:1n12), Oleic (C18:1n9), linoleic (C18: 2n6) acid. Several other fatty acids were also detected, but at much lower concentrations, including palmitoleic (C16:1n7), stearic (C18:0), linolenic (C18:3n3), arachidic (C20:0), behenic (C22:0) acids. This cultivars are rich in unsaturated fatty acids, and the total unsaturated fatty acid ratio was determined as Canada (93.19%), Tuinisia (94.28%), Lithuania (94.91%), Algeria (94.05%), China (94.14%), Vietnam - VS2 (94.21) and France - DS1(95.2%). Of which petroselinic acid is the most abundant fatty acid in seven cultivars with a rate of 69.65% to 74.33% and the content of French coriander cultivar (74.33%) was statistically highest of all cultivas. Today, unsaturated fatty acids are well known for their value in nutrition and pharmaceutical industrial uses. consequently producers of

nutraceuticals and dietary products are interested in natural sources of beneficial unsaturated. Furthermore, petroselinic acid is an uncommon isomer of oleic acid which is found in high levels in a restricted range of fruit oils. It forms a unique oleochemical with high potential for food, cosmetic and pharmaceutical industries. On other hand, oleic acid can be used as a raw material for fine chemicals. This fatty acid is a useful compound in the production of many food products or for the manufacture of fatty esters, which are used as biofunctional ingredients (emulsifiers and adjuvants) in formulation of bio-products. The total saturated fatty acid ratios of seven cultivars was determined as 4.8% in French, 5.1% in Lithuanian, 5.7 % in Tunisian, 5.8% in Vietnamese, 5.9% in Chinese, 5.9% in Algerian and 6.8% in Canadian coriander cultivar. Palmitic acid was determined as the highest saturated fatty acid in seven cultivars, palmitic acid was found at highest values in Canadian coriander (5.2%), and lowest values (3.8%) in French coriander cultivars among them. Palmitic acid ratios was determined ranging from 3.8% to 5.2%, this is in agreement with the results obtained in previous reported (Msaada et al., 2009a; Msaada et al., 2009b; Sriti et al., 2010a). As can be seen from the Table 12, the fruits were taken from seven cultivars of coriander according to planting organic condition was identified. The results showed that oil content and fatty acid composition of coriander cultivars significant effects dependent on different cultivars under conventional agriculture in Auch. Oil content of coriander fruits varied 15.3 to 24.3 % and percentages of major fatty acids such as petroselinic (69.7 to 74.3%), linoleic (14.6 to 16.3%), oleic (5.1 to 7.5%), palmitic (3.8 to 5.2%) acid were also significantly affected by cultivars. On the other hand, In order to compare the oil content and fatty acid composition between cultivars grown in Auch under organic conditions with the original fruits, the results obtained from the preliminary analysis of can be compared in Table 13. There are considerable variations among the oil content and fatty acid profiles between cultivars grown in Auch compared with the original cultivars. It can be seen from the data in Table 13 that the oil content and fatty acid composition of coriander fruits significantly affected by the ecological conditions in Auch. The oil content of most cultivars in Auch was lower than original fruits, between 16.5 % to 17.9 % of Canadian, between 19 % to 21.1 % of Lithuanian, between 24.3% to 25 % of French and between 17.6% to 21.1 % of Vietnamese coriander cultivar. The differences between the values of the fruits oil content can be probably due to genetic, growing, in climatic factors such as temperature and humidity during the different growth stages, and environmental conditions as well as analytical conditions and localities.

Table 12: Variation in oil content and fatty acid composition of coriander cultivars (cultivat

Fatty acid (%)	Different cultivars of coriander				
	Canada	**Tuinisia**	**Lithuania**	**Algeria**	**China**
Saturated fatty acid (SFA)					
C16:0	5.21 ± 0.01[a]	4.49 ± 0.14[b]	3.93 ± 0.03[c]	4.54 ± 0.04[b]	4.45 ± 0.0
C18:0	1.19 ± 0.01[a]	0.80 ± 0.07[b]	0.84 ± 0.01[b]	1.11 ± 0.01[a]	0.85 ± 0.0
C20:0	0.24 ± 0.01[a]	0.23 ± 0.02[a]	0.15 ± 0.01[c]	0.19 ± 0.02[b]	0.26 ± 0.0
C22:0	0.20 ± 0.01[b]	0.20 ± 0.02[b]	0.15 ± 0.01[b]	0.11 ± 0.01[c]	0.28 ± 0.0
Total	6.84 ± 0.01[a]	5.72 ± 0.19[b]	5.07 ± 0.04[c]	5.96 ± 0.05[b]	5.85 ± 0.0
Monounsaturated fatty acid (MUFA)					
C16:1n7	0.20 ± 0.01[b]	0.28 ± 0.08[a]	0.17 ± 0.03[c]	0.22 ± 0.01[b]	0.31 ± 0.0
C18:1n12	72.1 ± 0.17[b]	72.06 ± 0.49[b]	69.65 ± 0.31[c]	70.07 ± 0.57[c]	71.37 ± 0.
C18:1n9	5.08 ± 0.22[d]	6.39 ± 0.26[c]	8.46 ± 0.18[a]	8.60 ± 0.55[a]	6.77 ± 0.2
Total	77.38 ± 0.06[bc]	78.74 ± 0.29[b]	78.28 ± 0.11[b]	78.90 ± 0.02[b]	78.47 ± 0.
Polyunsaturated fatty acid (PUFA)					
C18: 2n6	15.43 ± 0.05[b]	15.16 ± 0.18[b]	16.35 ± 0.05[a]	14.90 ± 0.04[c]	15.28 ± 0.
C18:3n3	0.38 ± 0.02[a]	0.37 ± 0.04[a]	0.28 ± 0.01[b]	0.24 ± 0.02[b]	0.39 ± 0.0
Total	15.81 ± 0.05[b]	15.54 ± 0.18[b]	16.63 ± 0.06[a]	15.15 ± 0.06[b]	15.67 ± 0.
SFA/PUFA	0.43 ± 0.01[a]	0.37 ± 0.01[b]	0.30 ± 0.01[c]	0.39 ± 0.01[b]	0.37 ± 0.0
Water content (%)	10.94 ± 0.10[a]	10.54 ± 0.20[a]	10.82 ± 0.21[a]	11.47 ± 0.30[a]	11.26 ± 0.
Oil yield (%)	16.48 ± 0.14[d]	18.70 ± 0.17[b]	19.04 ± 0.12[b]	15.28 ± 0.09[d]	16.87 ± 0.
Acidity (% FFA)	1.78 ± 0.02[b]	1.48 ± 0.05[c]	1.20 ± 0.05[d]	1.38 ± 0.04[c]	1.75 ± 0.0

Data were means ± S.D of three replicates: Numbers on the same line with differing superscripts are sign
(Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (Stearic acid); C18:1n12 (Petroselinic acid); C18:1n9 (C
C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22:0 (Behenic acid), FFA: free fatty acid content.

Table 13: Comparison oil content and fatty acid composition between origin and coriander cultivars was cul

Fatty acid (%)	Canada		Lithuania		France - FR1	
	Auch	Origin	Auch	Origin	Auch	Ori
Saturated fatty acid (SFA)						
C16:0	5.21 ± 0.01	4.01 ± 0.28	3.93 ± 0.03	3.72 ± 0.01	3.81 ± 0.19	3.55 ±
C18:0	1.19 ± 0.01	0.89 ± 0.06	0.84 ± 0.01	0.77 ± 0.01	0.74 ± 0.06	0.75 ±
C20:0	0.24 ± 0.01	0.14 ± 0.02	0.15 ± 0.01	0.11 ± 0.01	0.13 ± 0.01	0.17 ±
C22:0	0.20 ± 0.01	0.16 ± 0.03	0.15 ± 0.01	0.13 ± 0.01	0.13 ± 0.01	0.06±
Total	6.84 ± 0.01	5.21 ± 0.40	5.07 ± 0.04	4.73 ± 0.01	4.81 ± 0.26	4.53 ±
Monounsaturated fatty acid (MUFA)						
C16:1n7	0.20 ± 0.01	0.21 ± 0.01	0.17 ± 0.03	0.16 ± 0.01	0.19 ± 0.01	0.22 ±
C18:1n12	72.1 ± 0.17	73.16 ± 1.36	69.65 ± 0.31	71.83 ± 0.35	74.33 ± 1.42	74.13 ±
C18:1n9	5.08 ± 0.22	6.57 ± 0.72	8.46 ± 0.18	6.33 ± 0.36	5.86 ± 1.03	6.12 ±
Total	77.38 ± 0.06	79.95 ± 0.63	78.28 ± 0.11	78.33 ± 0.02	80.38 ± 0.41	80.47 ±
Polyunsaturated fatty acid (PUFA)						
C18: 2n6	15.43 ± 0.05	14.60 ± 0.17	16.35 ± 0.05	16.60 ± 0.01	14.67 ± 0.12	14.77 ±
C18:3n3	0.38 ± 0.02	0.21 ± 0.02	0.28 ± 0.01	0.30 ± 0.01	0.15 ± 0.01	0.23 ±
Total	15.81 ± 0.05	14.81 ± 0.19	16.63 ± 0.06	16.90 ± 0.01	14.82 ± 0.12	15.00 ±
SFA/PUFA	0.43 ± 0.01	0.35 ± 0.26	0.30 ± 0.01	0.28 ± 0.11	0.32 ± 0.01	0.30 ±
Water content (%)	10.94 ± 0.10	9.82 ± 0.17	10.82 ± 0.21	10.18 ± 0.17	10.36 ± 0.35	9.85 ±
Oil yield (%)	16.48 ± 0.14	17.90 ± 0.19	19.04 ± 0.12	21.14 ± 0.08	24.26 ± 0.14	25.02 ±

Data were means ± S.D of three replicates: C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (St
acid); C18:1n9 (Oleic acid); C18: 2n6 (Linoleic acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22

The results indicated that nine fatty acid of coriander fruits was identified as C16:0 (Palmitic acid), C16:1n7 (Palmitoleic acid), C18:0 (Stearic acid), C18:1n12 (Petroselinic acid), C18:1n9 (Oleic acid), C18: 2n6 (Linoleic acid), C18:3n3 (Linolenic acid), C20:0 (Arachidic acid), C22:0 (Behenic acid). As Table 13 shows, there is a clear trend of decreasing monounsaturated fatty acid of cultivars in Auch compare with original cultivars. In this results also showed that there is a significant difference between the petroselinic acid of cultivars in Auch and petroselinic acid of original cultivars. Petroselinic acid of all cultivars used in this study had lower than of original fruits (between 77.4% to 80% of Canadian, between 78.3% to 78.3% of Lithuanian, between 80.4% to 80.5% of French and between 78.5% to 80.3% of Vietnamese coriander varities). In contrast, saturated fatty acid tends to decrease between cultivars in Auch and original fruits. This study was the first to provide comparative information about oil content and fatty acid of seven cultivars (Canada, Tunisia, Lithuania, Algeria, French (FR1 (Dourou) - DS1), Vietnam (VS2) and China) under organic condition. The results of the present study may be helpful for finding the cultivars of coriander, which can suitable cultivate in conventional agriculture conditions in Auch. Moreover, our results contribute to choice of the suitable cultivars reveals a decisive factor for the good performance of the crop and in similar climatic conditions.

III. Essential oil composition

III.1 Effects of sowing time on yield and essential oil composition of coriander

In this study, the French coriander cultivar (FR1 - Dourou) was selected in order to study the effects of planting date on oil yield and essential oil composition of coriander fruits. These trials were cultivated at different sowing time in organic condition. The first trial (DS1) was planted on 25th March, the second one (DS2) was 14th April, the third one (DS3) was 30th April in Auch (2011). In the experiment, the raw material was harvested on the fully maturity stage (September 2011).

The results of analysis of essential oil ratio and essential oil components shown in Table 14. According to the results have shown that a significant difference between the oil content in different sowing time (trial DS1, DS2 and DS3). Oil yield tends to decrease dependence on sowing time delay. As shown in Table 14 the average rate of essential oil yield ranged from 0.41 - 0.47%. The different between the average value are significant,

the highest value was obtained with the first trial DS1 (0.47 ± 0.2%), followed by DS2 (0.44 ± 0.2%) and the lowest value obtained with DS3 (0.41 ± 0.1%). These variations of oil yield could be to the changes in the environmental factors such as temperature, humidity and rainfall during fruits maturation and to the physiological maturation process of coriander fruits (Hornok, 1976; Msaada et al., 2007; Telci et al., 2006a). Its value was similar with Yildirim who has studied on ecological condition in different sowing time to determine the essential oil ratio and components of different cutivars of coriander. They found that the essential oil ratio was influenced by the cultivars and sowing time (Yildirim and Gok, 2012). Furthermore, the oil content of this study are also consistent with some of the previous studies, appropriate sowing date is important factor because of it ensures the better growth situation of plants and fruit can maximize the plant to make the best use of natural resource. However, oil yield values obtained from three trial (DS1, DS2 and DS3) remained quite similar values from the other researchers (Msaada et al., 2009d; Nejad Ebrahimi et al., 2010; Sriti et al., 2012b).

It can be seen from the data in Table 14 that thirteen (in trial DS1 and DS3) and fourteen (in trial DS2) components were identified representing from total oil. Linalool is the most characteristic coriander oil component and in all trial studied linalool represents 74.05 - 76.59 % of the total oil. The other major characteristic components of coriander fruits oils were α-pinene (3.30 - 4.27 %), γ-terpinene (3.15% - 4.13%), linalyl acetate (2.51 - 4.28%), geranyl acetate (2.89 - 3.95%), p-cymene (1.27 - 1.29%). Of which linalool and α-pinene tends to decrease dependence on sowing time delay. In contract γ-terpinene, linalyl acetate, geranyl acetate, p-cymene components tends to increase slightly on sowing time delay. Linalool, an oxygenated monoterpene, was the main component in essential oil of ripened fruits. It constitutes more than two-thirds of coriander fruit oil volatiles and is regarded as one of the flavour-impact compounds for fruit essential oil (Cadwallader et al., 1999). It has been known that climatic factors such as cloudy days and lower temperature during maturation and high amount of rainfall may have adverse effect on the accumulation of linalool (Sangwan et al., 2001). The present report in trial DS1, DS2 and DS3) is in agreement that linalool obtained was quite similar percentage in comparison with some previous reports. Raal et al. analyzed the oil of coriander fruits from different geographical origins of Europe. The major constituent of the oils were linalool (58.0−80.3%), γ-terpinene (0.3 - 11.2%), α-pinene (0.2 - 10.9%), p-cymene (0.1 - 8.1%), camphor (3.0 - 5.1%) and geranyl acetate (0.2 - 5.4%) (Raal et al., 2004). The main

constituents of the essential oil of coriander growing in 6 different zones of Argentina were linalool (68.9 - 83.7%), γ-terpinene (2.2 - 5.1%), camphor (3.2 - 4.8%), α-pinene (1 - 6.5%), geraniol (1.4 - 3.2%) and geranyl acetate (0.8 - 3.8%) (Bandoni et al., 1998). Ebrahimi and his co-worker cultivated coriander in different parts of Iran and analysed the chemical profiles of different accessions of coriander. The result showed that Linalool, neryl acetate, γ-terpinene and α-pinene were the major components in oil of coriander accessions and almost all accessions studied contain 60% linalool and the dried fruits contained the essential oil between 0.1 - 0.36% percent (Nejad Ebrahimi et al., 2010).

Table 14: Composition of essential oil in coriander fruits at different sowing times (DS1, DS2 and DS3)

Compound	Essential oil composition (rel %)				
	KIcalc	**KI**ref	**DS1**	**DS2**	**DS3**
α-Pinene	960	939	4.27 ± 0.5	3.30 ± 0.2	4.24 ± 0.1
Camphene	984	954	0.68 ± 0.1	0.69 ± 0.1	0.47 ± 0.2
β-Pinene	995	979	0.77 ± 0.2	0.47 ± 0.1	0.76 ± 0.1
Myrcene	1011	991	0.46 ± 0.1	0.46 ± 0.2	0.76 ± 0.1
p-Cymene	1046	1025	1.27 ± 0.3	1.29 ± 0.3	1.26 ± 0.3
Limonene	1052	1029	1.21 ± 0.2	0.96 ± 0.3	1.20 ± 0.1
γ-Terpinene	1078	1073	3.15 ± 0.5	3.19 ± 0.1	4.13 ± 0.3
Linalool	1113	1097	76.59 ± 0.3	75.44 ± 0.3	74.05 ± 0.2
Camphor	1189	1146	4.62 ± 0.1	4.27 ± 0.2	4.19 ± 0.2
Borneol	1211	1169	1.20 ± 0.2	1.21 ± 0.3	1.19 ± 0.1
α-Terpineol	1223	1189	0.39 ± 0.1	0.39 ± 0.1	0.38 ± 0.3
Carvone	1231	1243	-	0.11 ± 0.2	-
Linalyl acetate	1259	1257	2.51 ± 0.3	4.28 ± 0.1	3.50 ± 0.1
Geranyl acetate	1381	1381	2.89 ± 0.1	3.95 ± 0.1	3.87 ± 0.1
EO yield (%)			0.47 ± 0.2	0.44 ± 0.2	0.41 ± 0.1

Method: GC, peak identification through Kovats renteion indices. **KI**calc: Retention index calculated with reference to a series of n-alkanes using an apolar column; **KI**ref: Retention index reported in literature. (-) : not detected.

Generally, sowing date is one of the important factors influencing oil yield and essential oil components. The results of oil yield and its components in different sowing times have already mentioned above. There were significant differences of oil content in three sowing dates (DS1, DS2 and DS3). DS1 showed the highest values of oil yields and linalool compound. Therefore, it could be proposed that the suitable sowing time is DS1 (in the end of March) for planting of coriander cultivars under organic condition in Auch.

III.2 Essential oil components of different culitvars of coriander

As mention in chapter 2, the coriander (*Coriandrum sativum* L.) fruits were obtained from supermarket, seed company, retail shops of different European and Asian countries (Seven cultivars - Canada, Tunisia, Lithuania, Algeria, China, French (FR1 (Dourou) - DS1) and Vietnam (VS2). After collected, coriander cultivars was cultivated in organic agriculture at Auch, France. These coriander cultivars are grown on 25th March, 2011. The raw material harvested on the fully maturity stage (September 2011) was used in this analysis. Essential oil of coriander fruits were analyzed for qualitative composition by using Gas chromatography GC. Typical GC chromatogram of the essential oil is presented in Figure 36

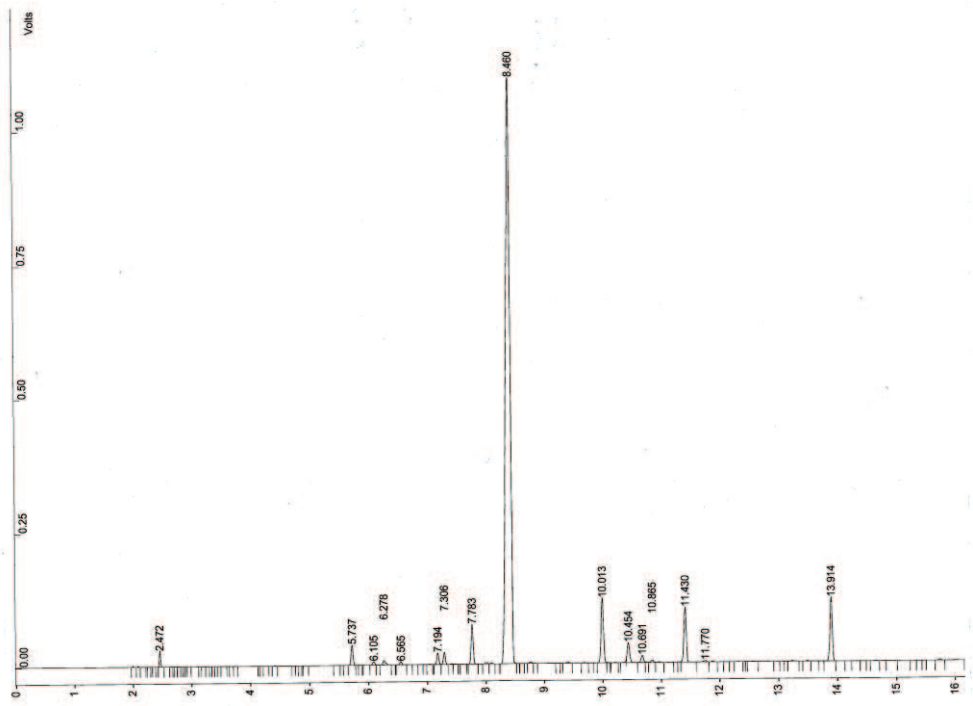


Figure 36: Typical GC chromatogram of essential oil of coriander cultivars

The essential oil obtained by hydrodistillation from the dried fruits of different cultivars was identified. The essential oil yield was varied from 0.43 - 0.95%, in which maximum oil content (up to 1.21%) was observed in Lithuanian coriander cultivar, follow by in Vietnamese (0.65%), in Chinese (0.61%), in Tunisian (0.59%), in Algerian (0.45%), French DS1 (0.44%), Canadian (0.43%) cultivar. Its value was similar to others reports that in the ripe fruits, the content of essential oil is comparably low (typically, less than 1%) (Telci et al., 2006b). In the previous studies, the essential oil content of coriander fruits varies from very low (0.03%) to a maximum report of 2.7% (Purseglove et al.). Dobos et al. also reported a range of variation of oil content between 0.2 and 1.3% among 36 different Coriander accessions from Austria (Dobos and Novak, 2005). Results of chemical composition of coriander essential oil (Coriandrum sativum) are shown on Table 15. Fourteen different compounds were identified in essential oil of five cultivars (Canadian, Tunisian, Lithuanian, Algerian, Vietnamese) and thirteen compounds have been found in essetial oil of the two coriander cultivars (Chinese and French coriander cultivar) (Table 15). Linalool, γ-terpinene, campor, linalyl acetate, geranyl acetate, α-pinene were identified as main components in the oil of all cultivars. Essential oil of the five coriander cultivars including cultivar collected from Canadian (77.67%), from Lithuanian (78.34%), from chinese (75.78%), from French DS1 (76.59%) and Vietnamese coriander cultivar (78.05%) showed a linalool content more than 75%. Two cultivars showed a linalool content less than 75% as Tunisian and Algerian coriander cultivars. Pino et al. (1996) have identified 35 compounds and have found different percentage for linalool (54.57 %), α- pinene (1.14 %), geraniol (6.97 %), γ-terpinene (4.08 %) and limonene (1.55 %) when studied the chemical composition of the seeds oil of Cuban coriander (Pino et al., 1996). Pino & Borges (1993, 1999) have also studied the content and chemical composition of coriander essential oil grown in different geographic regions in Russia, Italy, Albania and India. They have found different percentage for content and chemical composition of the oil for each studied region. The major compounds were linalool, γ-terpinene, camphor and α-pinene, and the highest percentages were found in Russia and Albania (Borges, 1999; Pino et al., 1993). Frank et al. (1995) have found very close values for its major compounds when carried out to evaluate chemical composition of coriander essential oil produced in Russia and Bulgaria. His reports showed that the linalool (65.0 % and 68.4 %), α-pinene (3.0 % and 2.5 %) and limonene (2.0 % and 1.3 %) (Frank et al., 1995).

Table 15: Composition of essential oil of coriander cultivars

Compound			Essential oil composition (rel %)				
	KIcalc	**KI**ref	**Canada**	**Tunisia**	**Lithuania**	**Algeria**	**Ch**
α-Pinene	960	939	2.48 ± 0.3	3.31 ± 0.1	1.27 ± 0.2	3.59 ± 0.1	4.71
Camphene	984	954	0.38 ± 0.1	0.84 ± 0.3	0.39 ± 0.1	0.40 ± 0.1	0.55
β-Pinene	995	979	0.55 ± 0.1	0.53 ± 0.3	0.33 ± 0.3	0.63 ± 0.2	0.69
Myrcene	1011	991	0.27 ± 0.4	0.85 ± 0.1	0.74 ± 0.4	0.84 ± 0.3	0.34
p-Cymene	1046	1025	1.17 ± 0.2	1.15 ± 0.1	0.65 ± 0.1	0.65 ± 0.3	1.22
Limonene	1052	1029	1.21 ± 0.1	1.26 ± 0.1	0.71 ± 0.1	0.71 ± 0.1	1.41
γ - Terpinene	1078	1073	4.46 ± 0.1	3.85 ± 0.2	5.32 ± 0.1	4.34 ± 0.1	2.05
Linalool	1113	1097	77.67 ± 0.1	74.13 ± 0.3	78.34 ± 0.2	74.65 ± 0.3	75.78
Camphor	1189	1146	2.88 ± 0.2	4.64 ± 0.1	3.73 ± 0.5	3.75 ± 0.2	4.30
Borneol	1211	1169	1.53 ± 0.3	1.63 ± 0.2	1.33 ± 0.1	1.34 ± 0.1	1.27
α-Terpineol	1223	1189	0.47 ± 0.2	0.21 ± 0.2	0.97 ± 0.4	0.98 ± 0.1	0.38
Carvone	1231	1243	0.17 ± 0.2	0.21 ± 0.3	0.23 ± 0.1	0.10 ± 0.2	-
Linalyl acetate	1259	1257	3.55 ± 0.4	2.26 ± 0.3	2.73 ± 0.2	3.75 ± 0.1	2.85
Geranyl acetate	1381	1381	3.22 ± 0.3	5.13 ± 0.2	3.26 ± 0.1	4.29 ± 0.2	4.43
EO yield (%)			0.43 ± 0.3	0.59 ± 0.1	1.21 ± 0.2	0.45 ± 0.1	0.61

Method: GC, peak identification through Kovats renteion indices. **KI**calc: Retention index calculated with re
an apolar column; **KI**ref: Retention index reported in literature. (-): not detected.

In this results, all cultivars studied was obtained more that 74% linalool showing high quality of coriander fruits produced which can be used in food, pharmaceutical and other related industries. The results obtained by several authors compared to the ones from this work, show that these compounds present variation, which may be related to climate and soil condition, harvest season and plant development. Linalool as an alifatic terpene was major component of the seven coriander cultivars collected from planting under organic condition. The results of the present study may be helpful for the recommendation of optimum coriander cultivars for planting under organic conditions in Auch. Beside, the results revealed the suitability of seven cultivars for use as initial genetic materials for breeding of homogenous and talent coriander cultivars in France.

IV. Antioxidant Properties

Antioxidants are compounds or substances that can retard the process of lipid oxidation, they can also block the propagation step of the radical chain reaction by destroying or binding free radicals, inhibition of catalysts or stabilization of hydroperoxides. In recent years, the importance of the search for the exploitation of natural antioxidants from plant origin, has greatly increased and natural antioxidants have gained popularity. On the other hand, Agricultural and industrial residues, such as the waste obtained during processing of some raw plant materials, are promising potential sources of natural antioxidants. Production of the essential oils results in a very high content of the waste (up to 99.5 % of the raw material), which remain unused after the distillation of oils, therefore such products should be carefully examined to assess the possibilities of their further processing to various valuable products, e.g. extracts containing phenolic compounds and other useful phytochemicals. Moreover, the recovery process of waste is part of the current existing sustainable development and environmental protection. In these experiments, the residues of coriander after hydrodistillation were separated into the solid and liquid fractions. The solid residue was dried at 30^{o}C by and extracted with methanol, the methanolic extracted was measured by different method assay (FRAP, ORAC, DPPH assay), the extractions were performed in triplicate.

Several analytical methods have been developed to determine the antioxidant capacity of natural substances in vitro. Each evaluation methods of which have their advantages and shortcomings. Currently, more and more analytical methods have been adapted for analysis on the microplate so that high-throughput screening of antioxidants could be made. They

can be categorized into two groups: (i) assays for radical-scavenging ability and (ii) assays for lipid oxidation inhibitory effect. However, the total antioxidant activities of plant extracts cannot be evaluated by using one single method, due to the complex composition of phytochemicals as well as of oxidative processes. Therefore, the use of at least two methods should be employed in order to evaluate the total antioxidant activity (Bohm et al., 2001). Consequently, the methanolic extracts from different coriander cultivars were evaluated by FRAP, ORAC, DPPH assays (Table 16). The aim of our study was to investigate the antioxidant activity of extracts of different polarity from coriander cultivars. According to the recommendations, the antioxidant effects in three different bioassays were studied, besides determination of total phenolics.

As can be seen from the Table 16, the results showed that methanolic extracts were obtained range 7.27 - 9.20%, there no significant differences between the content of the extracts of coriander cultivars. Methanolic extracts contents of Tunisian cultivar is highest (9.61 ± 0.93%), extracts contents obtained from Tunisian coriander were quite similar to Canadian and Tunisia coriander cultivar 9.20 ± 1.33%, and 9.18 ± 0.77%, respectively.

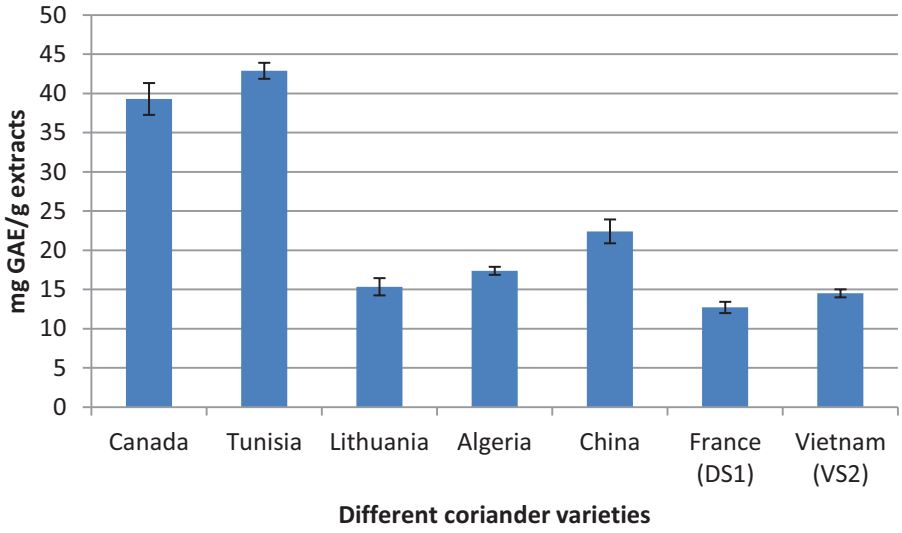


Figure 37: The total phenolic contents in different coriander cultivars

The total phenolic content (TPC) in extract was expressed as mg of gallic acid equivalents (GAE) / g of extract. TPC results was presented that the amount of total phenolics in extracts of seven coriander cultivar ranged from 12.71 ± 0.72 to 42.88 ± 1.02 mg GAE/g, of which extract of Tunisian cultivar showed the highest TPC value of 42.88 ± 1.02 mg GAE/g, followed by Canadian coriander varity (39.29 ± 2.03 mg GAE / g extract), extract

from French coriander cultivar the lowest TPC (Figure 37). Previous studies showed that antioxidant activity is directly related to total phenolic contents. It means the sample having the higher total phenolic content will show the higher antioxidant activity (Conkerton et al., 1995). According to Sriti et al. (2011) reported that total polyphenol contents found in coriander fruit methanolic extracts were 15.16 mg GAE/g DW for Canadian cultivar and 12.10 mg GAE/g DW for Tunisian one (Sriti et al., 2011b). A little differences in the extraction yield could be the result of using different extraction solvents in other works and methanol in ours. The importance of the solvent type used in the extraction has already been mentioned (Liu et al., 2007). Antioxidant analysis shows that all methanolic extract of coriander cultivars were antioxidatively active. However, their radical scavenging capacity (RSC) varied in a wide range Table 16. The RSC of different cultivars extracts was from 0.21 ± 0.02 to 0.93 ± 0.04 mg/mL in FRAP reaction system, from 1224.11 ± 2.36 to 1798.78 ± 0.23 µmol TE/1g extracts in ORAC, and from 13.42 ± 0.55 to 35.72 ± 1.83 µmol TE/1g in DPPH test.

Table 16: Antioxidant capacity and TPC extracts from residue after hydrodistilltaion of cultivars

Coriander cultivars	**Extracts** yield (%)/DP	**FRAP** mg/ml	**ORAC** µmol TE/1g extract (TroloxE/1g extract)	**DPPH** µmol TE/1g Extract	**TPC** mg GAE/g extract
Canada	9.20 ± 1.33	0.36 ± 0.01	1689.23 ± 0.95	14.54 ± 0.24	39.29 ± 2.03
Tunisia	9.61 ± 0.93	0.93 ± 0.04	1798.78 ± 0.23	13.42 ± 0.55	42.88 ± 1.02
Lithuania	9.18 ± 0.77	0.39 ± 0.01	1452.42 ± 0.87	35.72 ± 1.83	15.35 ± 1.10
Algeria	7.27 ± 0.69	0.21 ± 0.02	1284.53 ± 0.89	29.65 ± 0.20	17.38 ± 0.51
China	7.01 ± 1.14	0.38 ± 0.02	1701.66 ± 0.65	33.51 ± 1.87	22.41 ± 1.52
France (DS1)	7.95 ± 0.43	0.52 ± 0.02	1224.11 ± 2.36	25.41 ± 2.88	12.71 ± 0.72
Vietnam - VS2	8.89 ± 0.35	0.32 ± 0.02	1324.59 ± 0.44	25.79 ± 0.68	14.51 ± 0.51

In DPPH test, extract from Tunisian and Canadian coriander shows little better antioxidant activity than those of the same cultivated under organic condition. FRAP test show that Tunisian and French coriander cultivar extracts have better antioxidant activity compared of all cultivars, FRAP assay measures the ability of the extract to donate electron to ferric. The higher the FRAP value, the higher will be the antioxidant activity. In ORAC, the results showed the extracts of cultivars tend similar to the results of DPPH and FRAP test.

Extract from Tunisian cultivar (1798.78 ± 0.23 μmol TE/g extract) gave the highest antioxidant capacity values, followed by Chinese cultivar (1701.66 ± 0.65 μmol TE/g extract) and Canadian cultivar (1689.23 ± 0.95 μmol TE/g extract), which was in accordance with TPC assay.

In these preliminary results, the antioxidant activities of methanolic extract of seven coriander cultivars was studied. Coriander antioxidant activity was relatively high for the plant to be a new and natural source of antioxidant substances for its use as natural additives in food. Tunisian and Canadian cultivar showed the highest amount on total polyphenol and Tunisian one showed the strongest DPPH scavenging activity, FRAP and ORAC assay. In addition, antioxidant capacities varied significantly according to coriander cultivars and depends on phenolic composition of cultivars planting under organic condition in Auch.

V. Conclusion

- ✓ In terms of essential oils and vegetable oils yields and fatty acid profile, we can realize that coriander (*Coriandrum sativum* L.) are a potential class of ATOC resources. According to the described results, different parts of coriander (flower, leaves, stem, fruits and roots) are obtained both essential oil and vegetable oil, of which the fruit will be optimal for efficient exploitation more than other parts of coriander (flower, leaves, stem and roots). This study indicates that these organs are excellent sites for the bioaccumulation of functional secondary metabolites.
- ✓ The changes of fatty acid composition of coriander ripening had also defined the influence of maturity stages, this is the preliminary report to study the oil and fatty acid accumulation of Coriander during fruit ripening cultivated under organic conditions in three seasons (2009, 2010 and 2011). Our results demonstrated that the highest oil yield was achieved at the full maturity. Fatty acid profiles varied greatly during fruit ripening. At earlier stages, saturated and polyunsaturated fatty acids were higher and decreased with maturity of fruit. Petroselinic acid was the major fatty acid after 12 DAF. This latter showed an inversely evolution with palmitic acid which may support a functional correlation between both fatty acids. This study provided data for use coriander oil and its composition in fatty acids for different industrial applications due its content in petroselinic acid.

- ✓ Variation of fatty acid composition of coriander had also defined the influence of growing region. Base on the results, we can see that different ecological areas especially temperature, humidity and rainfall will be significant impacted in both fruits oil content and fatty acid composition in coriander. Therefore, growth location is an important factor for meeting market requirements of coriander fruits in terms of oil quality. Apart from growing condition, one should also consider some other factors such as cultivar oil type, planting time and irrigation in order to obtain a desire yield and quality.
- ✓ Sowing date is one of the important factors influencing fatty acid composition and oil yield of coriander fruits. The results on yield and fatty acid composition of different sowing times have already mentioned above. There were significant differences of oil content between three sowing times, DS1 (25th March) showed the highest values of oil yields as compared with significant differences in yield of DS2 (14th April) and DS3 (30th April). So it could be recommended planting coriander at the end of March is appropriate time for good crops. The sowing date effect with yield and fatty acid showed there was meaningful effect. The sowing date is determined to provide the conditions for the plants to maximally use environmental parameters, especially temperature and radiation. If environmental factors are appropriate for the greening, establishment and survival of seedlings, then the maximum yield can be realized by appropriate sowing date. Moreover, the establishment of appropriate sowing date will be useful for healthy plants and in a farm surface is a basic element of a successful agricultural system.
- ✓ Researches on vegetable oil and essential oil composition of different coriander cultivars (the seven coriander cultivars - Canada, Tunisia, Lithuania, Algeria, French (FR1 (Dourou) - DS1), Vietnam (VS2) and China) was performed to evaluate coriander cultivars pertaining to its fruits yield performance in Auch. From results, Tunisian and Lithuania cultivars could be voted for planting under organic conditions (in Auch) because of its potential for exploitation. It is necessary to develop more suitable cultivars for fruits production to fulfill the increasing demand of this spice crop. Selection of better cultivars can be of immense value to the breeder for further improvement and development of the crop. Therefore, the present investigation was undertaken in order to evaluate the appropriate conditions

of the collected coriander cultivars and to select the promising cultivars for higher fruits yield in France.

- ✓ For the researches on antioxidant capacity, antioxidant activities of methanolic extract of seven coriander cultivars was determined. Coriander antioxidant activity was relatively high for the plant to be a new and natural source of antioxidant substances for its use as natural additives in food. Tunisian and Canadian coriander cultivar showed the highest amount on total polyphenol and Tunisian one showed the strongest DPPH scavenging activity, FRAP and ORAC assay. The FRAP, ORAC and DPPH technique was simple, rapidly performed, thus it would be an appropriate technique for determining antioxidant in coriander extract.

Chapter 4: BIOREFINERY OF CORIANDER FRUITS, A FIRST INVESTIGATION

The biorefinery is a concept that integrates biomass conversion processes and equipments to produce fuels, power, and chemicals. Industrial biorefineries have been identified as the most promising route to the creation of a new domestic biobased industry. The goal is to use the by-products from one process as raw materials to obtain new valuable products, so that the least amount of the biomass is wasted. By producing multiple products, a biorefinery can take advantage of the differences in biomass components and intermediates, and maximize the value derived from the biomass feedstock.

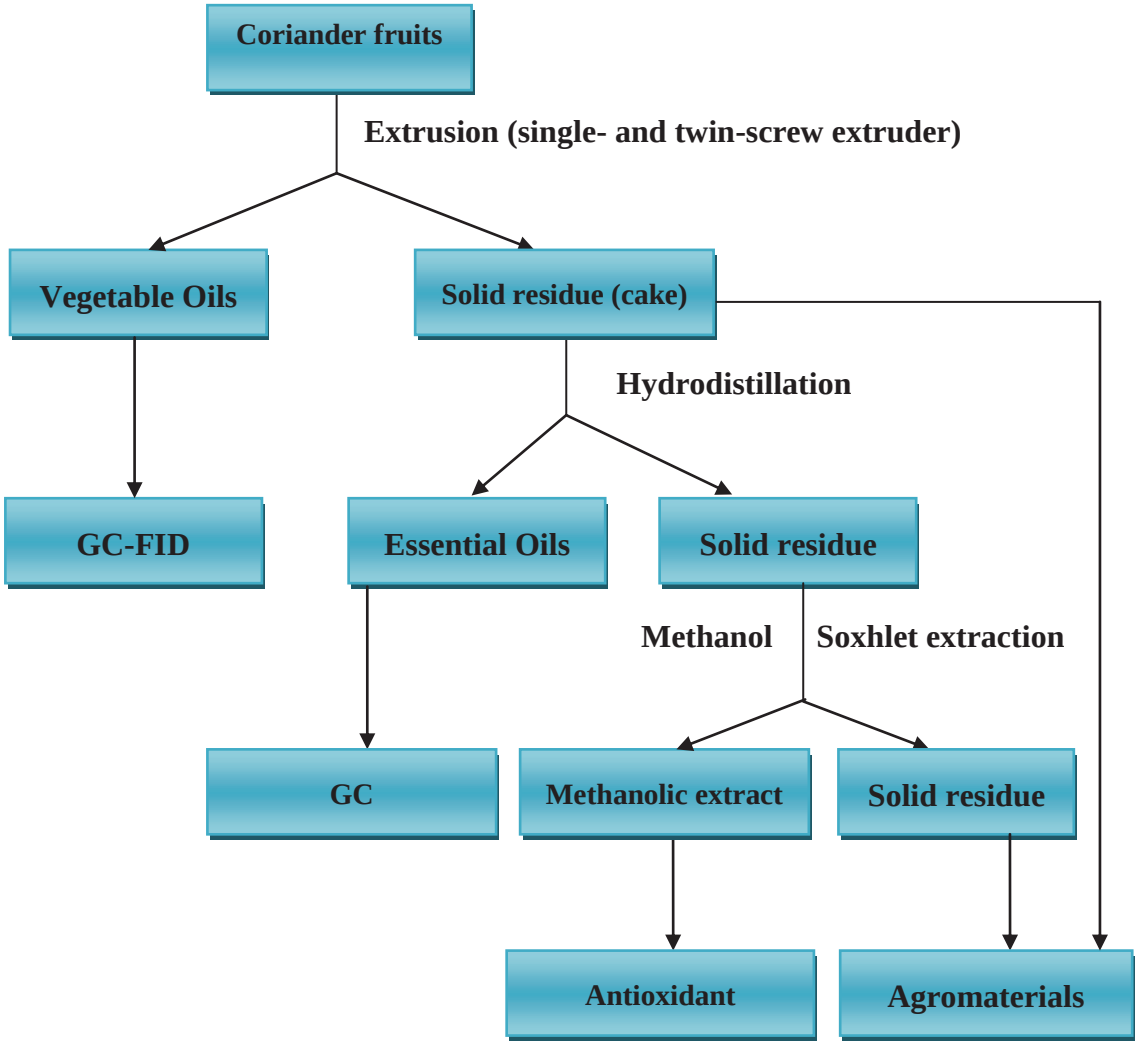


Figure 38: Scheme of preparation and analysis of coriander extracts

At present, some different biorefinery concepts are widely applied, such as conventional biorefineries, green biorefineries, feedstock biorefineries, thermo-chemical biorefineries, marine biorefineries, etc. Because biomass is a renewable resource, the use of the industrial biorefinery has been determined as one potential solution that may meet increasing demand for energy, fuel, chemicals and materials (Diep et al., 2012). Plant materials are renewable biomass. The separation process of biomass into distinct components, which could be individually utilized, i.e. biorefinery, is suitable for a global valorization of different molecules from these plants. Consequently, based on the biorefinery concept, the goal of this chapter is a processing plant that works to achieve the highest possible value out of biomass feedstock, i.e. valorization of the entire plant by sequential extractions of molecules of interest. It can be seen from the (Figure 38) that vegetable oil extract and solid residue (cake) of coriander will be produced through extrusion using the single- or the twin-screw extrusion technologies. Then, essential oil extract will be obtained from cake (residue) through hydrodistillation. For solid residue, it can be treated with methanol to get methanolic extracts. The final solid residue can be processed into agromaterials and biocomposites, used as fertilizer, for energy production, as animal feed or directly as fuel. Methanolic extracts will be evaluated for their antioxidant activity. The plants with high antioxidant activity will be potential sources of natural antioxidants. By modeling studies in this chapter, the molecules in the plant material can be fully used, hence avoiding a great waste of natural resources.

As mentioned in chapter 1, two different extraction techniques are commonly used to obtain vegetable oil from fruits, that is to say the organic solvent extraction and the mechanical pressing. Shahidi showed that the screw pressing can be used for oil recovery up to 90-95%, while solvent extraction is capable of extracting 99% (Shahidi, 2005). Currently, mechanical pressing becomes the most commonly used method for commercial oil extraction. Although the corresponding extraction yield is slightly lower, screw pressing is the most popular oil extraction method. Indeed, such process is simple, continuous, flexible and safe (Singh and Bargale, 2000; Zheng et al., 2005). Screw presses exist with capacities ranging from 10 kg/h up to tens of tons/h. Their principle is rather simple: the fruits are fed in a hopper during the pressing process and then transported and crushed by a rotating screw in the direction of a restriction. As the feeding section of a screw press is loosely filled with fruits material, the first step of the compression process consists of rolling, breaking and the displacement and removal of air from inter-material

voids. As soon as the voids diminish, the fruits start to resist the applied force through mutual contact (Faborode and Favier, 1996). The continuous transport of material causes pressure to increase to a level needed to overcome the restriction. This pressure causes the oil to be expressed from the fruits and oil yield obtained depends on the type of press (in particular the restriction geometry) and input material. For commercial oil extraction, this technology has many advantages, including its versatility, high productivity, low cost and the ability to produce unique product shapes and high product quality (Köksel et al., 2004). Therefore, the application of oil extraction process using extrusion technology will be studied in this chapter using both single- and twin-screw extruders. The aims of this work were to evaluate the feasibility of mechanical pressing to extract vegetable oil from coriander fruits, and to study the influence of operating conditions, i.e. nozzle diameter and nozzle/screw distance (case of single-screw extrusion) or screw configuration, device's filling coefficient and pressing temperature (case of twin-screw extrusion), on oil extraction yield.

I. Extraction of coriander oil using the single-screw extrusion technology

Experiments for the extraction of vegetable oil in the single-screw extruder were conducted from the different five coriander cultivars. Tunisian, Lithuanian and Vietnamese coriander cultivars were obtained from supermarket. The two other cultivars were French coriander supplied by GSN Semences company (France) and French Dourous DS1 cultivated in Auch (France).

For each experiment, the fruits are fed into the hopper of the machine. The transport of material is provided by the screw turn inside the sleeve. The driving of screw is effected by an electric motor. In all tests, the screw rotation speed, which is the condition to a good transport flow of the fruits, is fixed at 40 rpm. The feed rate is also maintained constant at 15 g/min (i.e. 0.9 kg/h). The pressing temperature is raised to the level of the filtration area at the beginning and at the end of experience, when the fruits is pressed. A 500 g weight of fruits is used for each experiment. The two single-screw extrusion conditions studied are the nozzle diameter and the distance between the screw head and the nozzle. For each nozzle diameter (5, 6, 7, 8, 9 and 10 mm), the experiments are performed from a distance between the screw head and the nozzle of 1, 2 and 3 mm. The mechanical shearing in the pressing area generates fine particles of smaller dimension than the diameter of the filtering sieves (i.e. 2 mm). These solid particles, commonly called "foot", are mixed in the

filtrate. A centrifugation step needs to be conducted to eliminate them from the pressed oil. Thus, the oil and foot contents are determined (Figure 39). Oil content (T_L) and foot content (T_F) in the filtrate are expressed relatively to the total mass of the filtrate and the residual oil content of the cake (L_C) is determined by Soxhlet extraction using *n*-hexane as extracting solvent.

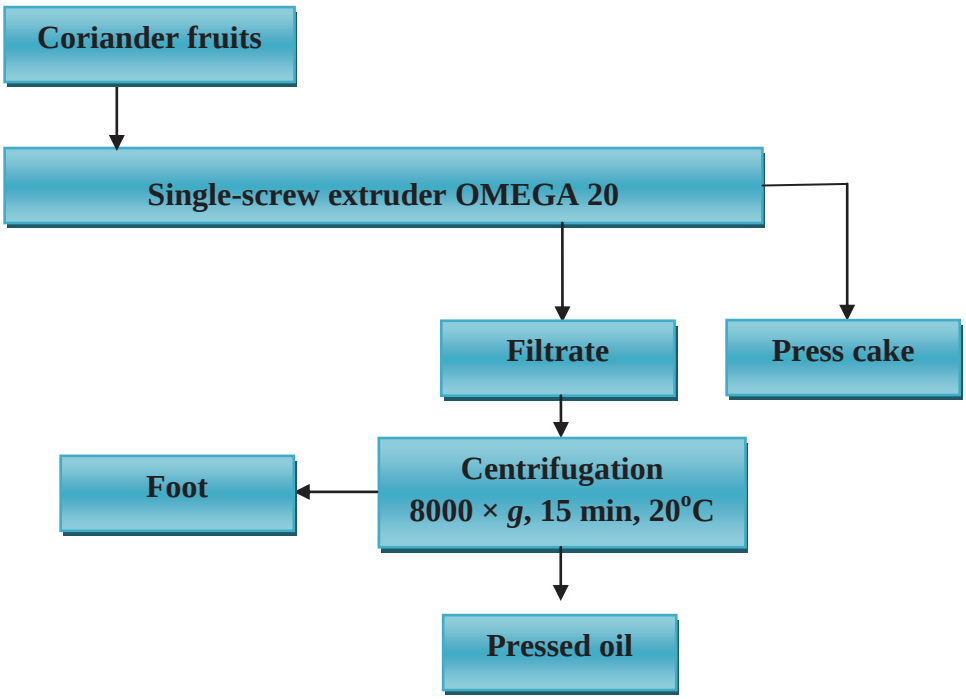


Figure 39: Schematic representation of oil expression of coriander fruits using a single-screw extruder (type OMEGA 20)

After the collection of both filtrate and press cake, the filtrate is centrifuged to separate the foot from the pressed oil (see above). The moisture and residual oil contents of the cake are determined according to French standards NF V 03-903 and NF V 03-908, respectively. The responses investigated in the different trials are calculated from the following equations:

- The oil yield collected after centrifugation (R_S), relative to the weight of dried fruits introduced.

$$R_s(\%) = \frac{m_F \times T_L}{m_S} \times 100$$

where m_F is the mass of the filtrate (g), T_L is the oil content in the filtrate (%) and m_S is the mass of dried fruits introduced (g).

- The oil yield collected after centrifugation (R_L), relative to the oil introduced.

$$R_L(\%) = \frac{m_F \text{ x } T_L}{m_S \text{ x } L_S} \text{ x } 100$$

where L_S is the oil content of the fruits (% of dry matter).

- The total oil yield (R_C), calculated from the residual oil contained in the press cake.

$$R_C(\%) = \frac{(m_S \text{ x } L_S) - (m_C \text{ x } L_C)}{m_S \text{ x } L_S} \text{ x } 100$$

where m_C is the mass of the dried press cake (g) and L_C is the residual oil content in the press cake (% of dry matter).

I.1 Effect of operating conditions on oil yield in the single-screw extruder

In order to evaluate the effects of nozzle diameter and nozzle/screw distance in the extraction of oil from coriander fruits, all trials were carried out using a single batch of coriander fruits: GSN maintenaire cultivar, cultivated in the South West part of France and supplied by GSN Semences Company (Le Houga, France). The moisture content of French coriander (GSN) fruits was 9.77 ± 0.10% (French standard NF V 03-903). The oil content determined by Soxhlet extraction using *n*-hexane as extracting solvent was 27.67 ± 0.57 % of dry matter (NF V 03-908).

Eighteen trials were conducted. The results obtained from the different operating conditions tested (i.e. the nozzle diameter and the distance between the nozzle and the screw) are shown in Table 17, and the obtained oil yields (R_S) are presented in Figure 40. Data in Table 17 show that the different operating conditions used have an influence on vegetable oil extraction from coriander fruits. R_S oil yield ranges from 12.8% to 16.0%, and the oil extraction yield tends to increase from trial 1 to trial 18. Generally, the diameter of the nozzle (from 5 to 10 mm) affects the oil extraction efficiency. The oil yields obtained for nozzle diameters of 8, 9 and 10 mm are higher than oil yields obtained for nozzle diameters of only 5, 6 and 7 mm. The highest oil extraction yield (16.0%) was obtained for nozzle diameter of 9 mm and nozzle/screw distance of 3 mm, while the lowest one (i.e. 12.8%) was obtained for nozzle diameter of only 5 mm and nozzle/screw distance of only 1 mm (Figure 40). Besides, from these results, the oil extraction yield increases as the distance between the nozzle and the screw increases, for all nozzle diameters tested and in particular for nozzle diameters of 8, 9 and 10 mm. For example, for the 9 mm nozzle

diameter, the R_S oil yield varies from 15.3% for the 1 mm distance to 16.0% for the 3 mm one. Thus, the increase in the distance between the nozzle and the screw (from 1 to 3 mm) contributes to the rise of the pressure on the material. In addition, a higher distance between the nozzle and the screw leads to the increase in the raw material residence time in the pressing zone, thus increasing both material pressure and oil extraction yield. On the other hand, the higher the nozzle/screw distance, the lower the foot content in the filtrate (i.e. less solid particles driven through the filter during the liquid/solid separation). The quality of pressed oils was also examined (Table 17). The oil acidity varies from 1.3 to 1.8%. From these results, the operating conditions (i.e. nozzle diameter and distance between the nozzle and the screw) have no influence on the oil quality, the latter being always satisfactory.

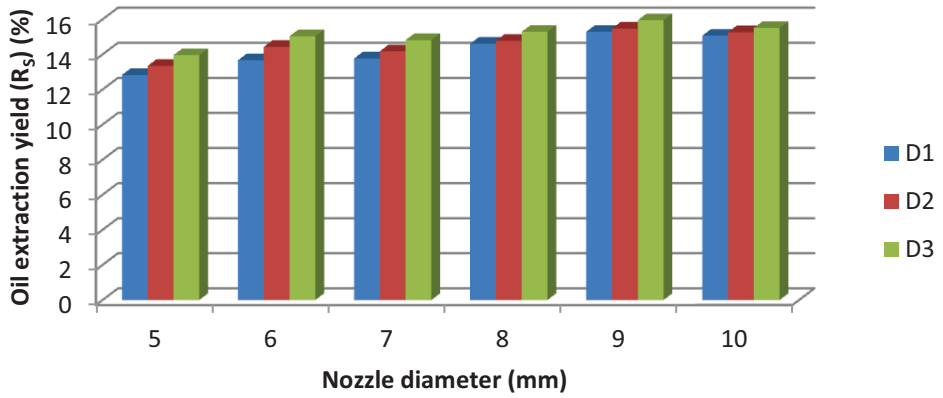


Figure 40: Variation of oil extraction yield (R_S) on different screw configuration

In this study, the different configurations were tested in order to assess the impact of the extrusion operating conditions on oil extraction efficiency. The effect on the oil yield was evaluated through the determination of three different yields: R_S (discussed before), R_L and R_C. The oil yield (R_L) is defined as the ratio of the pressed oil to the total oil contained within the fruit. In Table 17, the oil yield (R_L) varies from 41.8 to 52.0%. The highest oil yield (R_L) for this study (i.e. 52.0%) was obtained under the following operating conditions: 9 mm nozzle diameter and 3 mm nozzle/screw distance. Such value is similar to other results reported in the literature for coriander oil extraction in a single-screw extruder (Sriti et al., 2011a). In addition, the residual oil content in the press cake varies from 14.2 to 16.8% of the dry matter. This leads to an oil yield (R_C), based on the residual oil content in the press cake, of between 49.8 and 57.1% (53.8% for the optimal conditions) (Table 17).

Table 17: Effect of screw configuration conditions on oil extraction in single-screw extru

Trial number	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**
Operating conditions													
Nozzle diameter (mm)	5	5	5	6	6	6	7	7	7	8	8	8	9
Nozzle/screw distance (mm)	D_1	D_2	D_3	D_1	D_2	D_3	D_1	D_2	D_3	D_1	D_2	D_3	D_1
T^o C initial	26	27	25	27	28	26	25	25	28	24	26	25	27
T^o C final	57	62	65	63	60	65	56	58	57	63	65	62	62
Filtrate													
Mass (g)	65.35	67.31	70.72	69.89	72.21	74.35	69.78	71.37	73.65	74.35	74.31	75.89	77.0
T_L (%)	88.54	89.45	89.69	88.25	90.17	91.28	89.01	89.62	90.77	88.66	89.75	90.89	89.5
T_F (%)	11.46	10.55	10.31	11.75	9.83	8.72	10.99	10.38	9.23	11.34	10.25	9.11	10.4
Acidity (% FFA)	1.61	1.65	1.65	1.74	1.53	1.56	1.56	1.63	1.63	1.66	1.66	1.62	1.7(
Press cake													
Mass (g)	418.89	412.54	411.19	412.08	408.32	405.1	416.88	410.26	409.89	413.98	409.54	410.45	405.(
L_C (%)	15.6	15.2	14.2	16.2	15.8	14.8	15.6	15.2	15.0	16.6	16.2	15.6	16.8
H_C (%)	7.81	8.01	8.32	8.3	8.46	8.41	8.0	8.21	8.25	8.67	8.55	8.14	8.1
Oil yields (%)													
R_S	12.83	13.35	13.96	13.67	14.43	15.04	13.77	14.18	14.82	14.61	14.78	15.29	15.2
R_L	41.82	43.52	45.52	44.58	47.06	49.06	44.98	46.23	48.32	47.65	48.20	49.86	49.8
Rc	51.75	53.80	57.12	50.97	52.69	56.02	52.15	54.16	54.82	49.79	51.40	52.91	49.9

T_L: oil content in the filtrate, T_F: foot content in the filtrate, L_C: residual oil content in the press cake, H_C: mo oil yield in proportion to the fruits weight, R_L: oil yield in proportion to the oil that the fruits contains, R_c content in the press cake. FFA: free fatty acid content, D_1: Nozzle/screw distance = 1 mm, D_2: Nozzle/screw distance = 3 mm.

Table 18: Variation in fatty acid composition of oil extracted from coriander fruits in a single-scr

Fatty acid (%)	Nozzle diameter 5 mm			Nozzle diameter 6 mm		
	D_1	D_2	D_3	D_1	D_2	D_3
Saturated fatty acids (SFA)						
C16:0	3.36 ± 0.10	3.62 ± 0.12	3.29 ± 0.05	3.83 ± 0.03	3.78 ± 0.08	3.58 ± 0.08
C18:0	0.79 ± 0.03	0.81 ± 0.02	0.78 ± 0.05	0.70 ± 0.03	0.82 ± 0.05	0.81 ± 0.07
C20:0	0.11 ± 0.02	0.12 ± 0.03	0.18 ± 0.01	0.15 ± 0.01	0.15 ± 0.02	0.19 ± 0.01
C22:0	0.07 ± 0.02	0.07 ± 0.01	0.11 ± 0.02	0.10 ± 0.01	0.10 ± 0.02	0.13 ± 0.01
Total	4.33 ± 0.07	4.62 ± 0.09	4.37 ± 0.04	4.78 ± 0.03	4.85 ± 0.11	4.72 ± 0.12
Monounsaturated fatty acids (MUFA)						
C16:1n7	0.16 ± 0.01	0.16 ± 0.01	0.18 ± 0.01	0.18 ± 0.02	0.17 ± 0.01	0.19 ± 0.01
C18:1n12	74.07 ± 0.20	73.74 ± 0.17	74.22 ± 0.08	73.73 ± 0.17	73.63 ± 0.12	73.86 ± 0.05
C18:1n9	7.19 ± 0.39	7.15 ± 0.23	7.07 ± 0.16	7.11 ± 0.18	7.11 ± 0.13	7.10 ± 0.04
Total	81.42 ± 0.27	81.05 ± 0.12	81.47 ± 0.21	81.01 ± 0.05	80.90 ± 0.03	81.15 ± 0.04
Polyunsaturated fatty acids (PUFA)						
C18:2n6	13.89 ± 0.18	14.08 ± 0.05	13.85 ± 0.19	13.89 ± 0.06	14.01 ± 0.08	13.80 ± 0.11
C18:3n3	0.36 ± 0.04	0.25 ± 0.03	0.31 ± 0.01	0.31 ± 0.01	0.24 ± 0.03	0.33 ± 0.03
Total	14.25 ± 0.17	14.33 ± 0.06	14.16 ± 0.16	14.20 ± 0.05	14.25 ± 0.08	14.13 ± 0.12
SFA/PUFA	0.30 ± 0.00	0.32 ± 0.01	0.31 ± 0.00	0.33 ± 0.00	0.34 ± 0.01	0.33 ± 0.01

C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (Stearic acid); C18:1n12 (Petroselinic acid); C18: acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22:0 (Behenic acid). Data were means ± SD fro distance = 1 mm, D_2: Nozzle/screw distance = 2 mm, D_3: Nozzle/screw distance = 3mm.

Table 19: (following): Variation in fatty acid composition of oil extracted from coriander fruits in a sing

Fatty acid (%)	Nozzle diameter 8 mm			Nozzle diameter 9 mm		
	D_1	D_2	D_3	D_1	D_2	D_3
Saturated fatty acids (SFA)						
C16:0	3.74 ± 0.01	3.85 ± 0.01	3.99 ± 0.09	3.96 ± 0.07	3.85 ± 0.01	3.92 ± 0.04
C18:0	0.83 ± 0.03	0.74 ± 0.02	0.75 ± 0.01	0.84 ± 0.02	0.74 ± 0.02	0.74 ± 0.02
C20:0	0.17 ± 0.01	0.18 ± 0.01	0.19 ± 0.01	0.14 ± 0.01	0.18 ± 0.01	0.20 ± 0.04
C22:0	0.13 ± 0.01	0.11 ± 0.01	0.13 ± 0.01	0.10 ± 0.02	0.11 ± 0.02	0.13 ± 0.01
Total	4.86 ± 0.01	4.86 ± 0.03	5.05± 0.09	5.04± 0.12	4.88 ± 0.03	4.99 ± 0.07
Monounsaturated fatty acids (MUFA)						
C16:1n7	0.22 ± 0.01	0.23 ± 0.02	0.24 ± 0.01	0.23 ± 0.01	0.23 ± 0.02	0.26 ± 0.01
C18:1n12	73.79 ± 0.05	73.40 ± 0.01	72.60 ± 0.01	73.88 ± 0.03	73.40 ± 0.01	72.66 ± 0.04
C18:1n9	7.24 ± 0.02	7.29 ± 0.03	7.76 ± 0.08	7.34 ± 0.01	7.29 ± 0.03	7.80 ± 0.04
Total	81.25 ± 0.02	80.92 ± 0.01	80.59 ± 0.06	81.46 ± 0.11	80.92 ± 0.01	80.71 ± 0.01
Polyunsaturated fatty acids (PUFA)						
C18:2n6	13.65 ± 0.11	13.95 ± 0.01	14.10 ± 0.04	13.29 ± 0.02	13.95 ± 0.01	14.06 ± 0.05
C18:3n3	0.23 ± 0.01	0.24 ± 0.01	0.25 ± 0.01	0.21 ± 0.01	0.24 ± 0.01	0.23 ± 0.01
Total	13.88 ± 0.02	14.20 ± 0.02	14.35 ± 0.02	13.51 ± 0.02	14.20 ± 0.02	14.29 ± 0.05
SFA/PUFA	0.35 ± 0.00	0.34 ± 0.01	0.35 ± 0.01	0.37 ± 0.01	0.34 ± 0.01	0.35 ± 0.01

C16:0 (Palmitic acid), C16:1n7 (Palmitoleic acid); C18:0 (Stearic acid), C18:1n12 (Petroselinic acid), C18: acid), C20:0 (Arachidic acid), C18:3n3 (Linolenic acid), C22:0 (Behenic acid). Data were means ± SD fron distance = 1 mm, D_2: Nozzle/screw distance = 2 mm, D_3: Nozzle/screw distance = 3mm.

The fatty acid compositions of pressed oils are in the Table 18 and Table 19. The results show no real change in the relative proportions of fatty acids according to the extrusion conditions used. In all trials, nine fatty acids were identified. All pressed oils were very rich in petroselinic acid (72.6 - 74.2% depending on the conditions used, i.e. the nozzle/screw distance and the nozzle diameter), followed by linoleic, oleic and palmitic acids, accounting for 13.2 - 14.1%, 7.1 - 7.8% and 3.3 - 4.0%, respectively, of the total fatty acids. In all trials, saturated fatty acids (SFA) represented from 4.3 to 5.0% of total fatty acids, while monounsaturated fatty acids (MUFA) accounted for 80.6 - 81.9% and polyunsaturated fatty acids (PUFA) accounted for 13.5 - 14.3%.

In summary, the operating conditions played an important role to influence the oil extraction yield in the single-screw extruder, and the highest oil yields were obtained from the configuration allowing the stronger material pressure, i.e. a nozzle diameter from 8 to 10 mm. Generally, the modification of the extrusion conditions did not influence the oil quality. Moreover, the best operation condition (3 mm nozzle/screw distance and 9 mm nozzle diameter) led to a 16.0% oil yield (R_S) and to a 52.0% oil yield (R_L).

I.2 Influence of the coriander cultivars on oil extraction efficiency

Experiments were conducted for the extraction of vegetable oil in the single-screw extruder from the different five coriander cultivars (fruits). Tunisian, Lithuanian and Vietnamese cultivars were obtained from supermarket. French coriander was supplied by GSN Semences Company (France) and French Dourous DS1 was cultivated in Auch (France). The various constituents were determined according to the following French standards: moisture contents NF V 03-903, oil contents NF V 03-908. The moisture contents and oil contents are presented in Table 20.

Table 20: Chemical composition of the different coriander fruits used for experiment (% of dry matter).

Coriander cultivars	Moisture content (%)	Oil content (%)
Tunisia	10.04 ± 0.22	22.07 ± 0.35
Lithuania	10.18 ± 0.17	21.14 ± 0.08
France (DS1)	9.85 ± 0.12	25.02 ± 0.10
France (GSN)	9.77 ± 0.10	27.67 ± 0.57
Vietnam (VS2)	10.10 ± 0.01	21.08 ± 0.25

Data are expressed as mean ± SD from three replicates.

When studying the effect of operating conditions on oil extraction from French coriander fruits (GSN) in the single-screw extruder, the highest oil yield was obtained from the next configuration: 3 mm nozzle/screw distance and 9 mm nozzle diameter. This optimizing configuration is the best to be applied for the extraction of vegetable oil, thus leading to the best oil extraction efficiency. Therefore, this screw configuration will be applied from the different coriander cultivars. Fifteen trials were conducted, i.e. three replications from each variety.

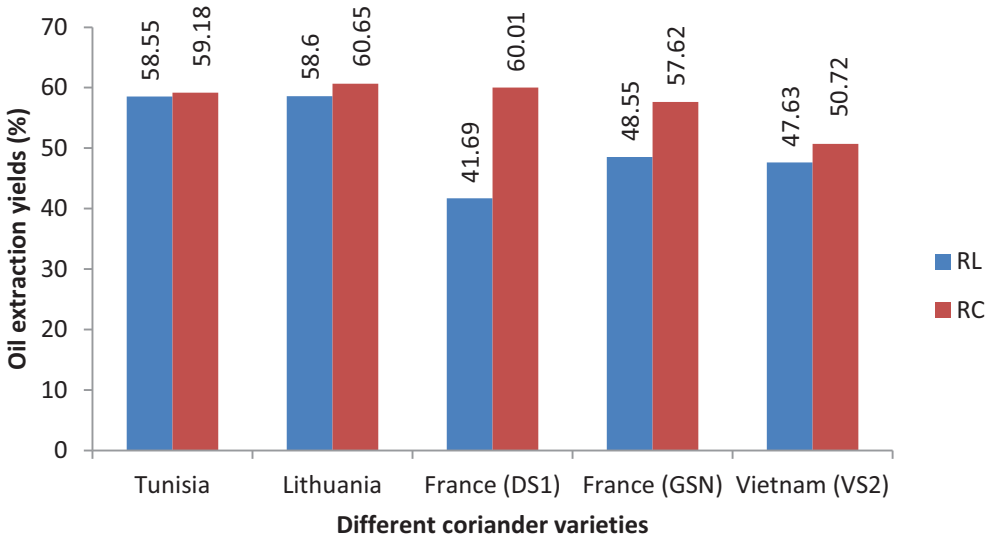


Figure 41: Variation in oil extraction yields (%) for the different coriander cultivars tested

For the five cultivars tested, the effect on oil extraction was evaluated thanks to the determination of three different oil yields: R_S, R_L and R_C. The results obtained are shown in Table 21, and the associated oil yields (R_L and R_C) are presented in Figure 41. Depending on the coriander cultivars, the oil yield (R_S) ranges from 11.1 to 14.9%. The highest oil yield (i.e. 14. 9%) is obtained from the French coriander (GSN), followed by Tunisian, Lithuanian, French (DS1) and Vietnamese corianders, accounting for 14.4%, 13.8%, 11.6% and 11.1%, respectively. In Figure 41, R_C oil yield is higher than R_L oil yield for all trials because it includes all the vegetable oil in the filtrate, i.e. not only the pressed oil but also the oil contained within the foot. For Tunisian coriander cultivar, the oil yield obtained in this experiment (14.4% for R_S yield) is a little lower than in a previous study (15.7%) conducted from the same operating conditions (3 mm nozzle/screw distance and 9 mm nozzle diameter) (Sriti et al., 2011a). This can be explained by variations in climatic

conditions, thus leading to different oil contents within the fruits depending on the year of cultivation and harvesting, and differences in storage conditions. The lowest R_S oil yield (11.1%) was obtained from the Vietnamese cultivar, and the oil quality was then significantly reduced compared with other corianders. The oil was strongly dark-colored and displayed an unacceptable high amount of free fatty acids of over 4% instead of no more than 2.2% for the other cultivars. As the Vietnamese coriander fruits originated from the 2010 harvesting, the strong reduction in both oil yield and oil quality may thus be due to aging. The French (DS1) coriander fruits were cultivated in 2011 and they were subjected to single-screw extrusion as well. This rendered a high yield of lightly colored oil and weakly elevated free fatty acid content (1.9 - 2.2%), discarding the hypothesis of aging causing color changes and oil loss. This leads to the conclusion that, firstly, oil yield and appearance is highly dependent on fruits origin and, secondly, vegetable oil yield varies throughout different years, most likely due to variations in climatic conditions. The strong difference in FFA content between oils coming from French and Vietnamese coriander fruits that were both subject to aging, may be explained by significant differences in storage conditions. While the French fruits were stored in the freezer, being protected from the light, there are no such guarantees regarding storage of the Vietnamese fruits and they may have been subjected to long-term storage at high temperature and moisture conditions.

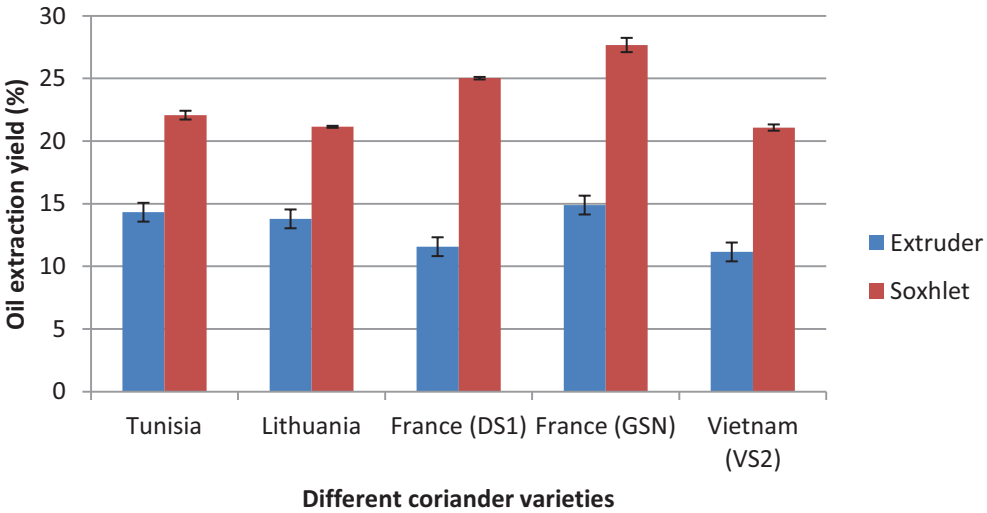


Figure 42: Comparison between oil extraction yields in the single-screw extruder and using the Soxhlet method with *n*-hexane as extracting solvent.

Residual oil contents in the press cakes are reported in Table 22. From the Figure 42, it can be seen that the oil yield extracted by mechanical pressing using the single-screw extruder is always lower than by solvent extraction (Soxhlet method). This is in agreement with opinion of Shahidi (2005) that the screw pressing is used for oil recovery up to 90%, while solvent extraction is capable of extracting 99% (Shahidi, 2005). Although the oil yield obtained using the single-screw extruder is lower compared with a previous research (Sriti et al., 2011a), single-screw pressing is still a popular oil extraction method as the process is simple, continuous, flexible and safe.

The results of fatty acid composition in pressed oils are presented in Table 22. They indicate that nine fatty acids were identified: C16:0 (Palmitic acid), C16:1n7 (Palmitoleic acid), C18:0 (Stearic acid), C18:1n12 (Petroselinic acid), C18:1n9 (Oleic acid), C18:2n6 (Linoleic acid), C18:3n3 (Linolenic acid), C20:0 (Arachidic acid) and C22:0 (Behenic acid). The fatty acid composition of all coriander pressed oils was characterized by substantial amounts of Petroselinic acid, Linoleic acid, Oleic acid and Palmitic acid, the variation in fatty acids depending on the coriander cultivars used. The unsaturated fatty acid (MUFA plus PUFA) contents were always high (> 94%). The unsaturated fatty acid ratio is important in medicinal and nutritional aspects (Roche et al., 2010). Generally, fatty acid compounds have not significantly changed between the two extracting methods (i.e. single-screw extrusion and Soxhlet extraction).

In summary, oil yield achieved depends a lot on the extraction method used (mechanical pressing using a single-screw extruder or Sohxlet extraction using *n*-hexane as extracting solvent). For mechanical pressing, the optimization of oil extraction with respect to oil quality depends on the single-screw extrusion conditions (i.e. nozzle diameter and nozzle/screw distance). The increase in the nozzle diameter (8, 9 and 10 mm) will contribute to higher oil yields, and maximum yield was obtained with 3 mm nozzle/screw distance and 9 mm nozzle diameter. Besides, the fatty acid composition is unaffected by the variation of the extrusion parameters. It is characterized by the abundance of monounsaturated fats (close to 80%), in which petroselinic acid (C18:1n12) is the major compound (varying from 73.6 to 75.8% depending on the coriander cultivars).

Table 21: Effect of the coriander cultivars on the oil extraction efficiency in a single-screw e

Coriander cultivars	**Tunisia**			**Lithuania**			**France (DS1)**			**Franc**
Trial number	1	2	3	4	5	6	7	8	9	10
Operating conditions										
Nozzle diameter	Nozzle diameter: 9 mm			Nozzle diameter: 9 mm			Nozzle diameter: 9 mm			Nozzle dia
Nozzle/screw distance (mm)	D_3	D_3	D_3	D_3	D_3	D_3	D_3	D_3	D_3	D_3
T^oC initial	27	27	30	31	27	28	25	25	30	26
T^oC final	60	62	64	60	58	65	57	58	55	63
Filtrate										
Mass (g)	68.21	75.98	76.35	66.89	72.11	73.35	59.05	64.42	61.07	71.76
T_L (%)	89.40	86.69	87.73	88.17	87.19	87.23	83.22	85.78	85.30	89.01
T_F (%)	10.60	13.31	12.27	11.83	12.81	12.77	16.78	14.22	14.70	10.99
Acidity (% FFA)	1.47	1.42	1.53	1.32	1.47	1.39	2.20	1.97	1.91	1.42
Press cake										
Mass (g)	415.08	406.91	402.56	416.08	415.33	409.56	427.68	407.68	416.2	414.33
L_C (%)	14.2	13.4	13.2	12.8	13.3	12.8	11.5	12.2	11.6	14.6
H_C (%)	8.36	8.17	8.18	8.19	8.14	8.21	7.99	8.20	8.11	8.00
Oil yields (%)										
R_S	13.56	14.64	14.89	13.13	14.00	14.25	10.90	12.26	11.56	14.16
$\overline{R_S}$	14.36 ± 0.70^a			13.79 ± 0.58^a			11.57 ± 0.68^b			14.89
R_L	55.26	59.69	60.70	55.80	59.48	60.53	39.28	44.17	41.64	46.17
$\overline{R_L}$	58.55 ± 2.98^a			58.60 ± 2.48^a			41.69 ± 2.44^b			48.55
Rc	56.74	59.89	60.92	60.84	59.66	61.46	59.87	59.87	60.66	55.42
$\overline{R_C}$	59.18 ± 2.17^a			60.65 ± 0.91^a			60.01 ± 0.58^a			57.62

T_L: oil content in the filtrate, T_F: foot content in the filtrate, L_C: residual oil content in the press cake, H_C: moisture co proportion to the fruits weight; R_L: oil yield in proportion to the oil that the fruits contains, $R_{C:}$ oil yield based on th FFA: free fatty acid content, D_3: Nozzle/screw distance = 3mm. Data were means ± SD from three replicates. Nu superscripts are significantly different at $P < 0.05$.

Table 22: Fatty acid composition of pressed oils from different coriander cultivars and produced in a sin

Fatty acid (%)	Tunisia		Lithuania		France (DS1)		Franc
	Extruder	Soxhlet	Extruder	Soxhlet	Extruder	Soxhlet	Extruder
Saturated fatty acid (SFA)							
C16:0	3.66 ± 0.32	3.74 ± 0.03	4.24 ± 0.04	3.72 ± 0.01	3.62 ± 0.02	3.55 ± 0.10	3.48 ± 0.06
C18:0	0.83 ± 0.23	0.75 ± 0.01	0.66 ± 0.04	0.77 ± 0.01	0.66 ± 0.01	0.75 ± 0.04	0.78 ± 0.03
C20:0	0.23 ± 0.04	0.20 ± 0.01	0.12 ± 0.04	0.11 ± 0.01	0.08 ± 0.01	0.17 ± 0.01	0.09 ± 0.01
C22:0	0.04 ± 0.02	0.10 ± 0.01	0.17 ± 0.01	0.13 ± 0.01	0.06 ± 0.01	0.06± 0.01	0.07 ± 0.01
Total	4.75 ± 0.31	4.79 ± 0.04	5.19 ± 0.03	4.73 ± 0.01	4.42 ± 0.03	4.53 ± 0.15	4.42 ± 0.08
Monounsaturated fatty acid (MUFA)							
C16:1n7	0.20 ± 0.03	0.21 ± 0.01	0.19 ± 0.01	0.16 ± 0.01	0.20 ± 0.01	0.22 ± 0.06	0.19 ± 0.01
C18:1n12	75.28 ± 0.1	76.90 ± 0.26	73.56 ± 0.45	71.83 ± 0.35	74.02 ± 0.29	74.13 ± 0.18	73.72 ± 0.42
C18:1n9	6.41 ± 0.16	4.93 ± 0.20	5.98 ± 0.04	6.33 ± 0.36	7.39 ± 0.31	6.12 ± 0.22	7.31 ± 0.42
Total	81.89 ± 0.07	82.04 ± 0.09	79.72 ± 0.12	78.33 ± 0.02	81.61 ± 0.04	80.47 ± 0.46	81.22 ± 0.05
Polyunsaturated fatty acid (PUFA)							
C18:2n6	13.23 ± 0.26	12.97 ± 0.06	14.81 ± 0.14	16.60 ± 0.01	13.87 ± 0.02	14.77 ± 0.16	14.23 ± 0.08
C18:3n3	0.12 ± 0.04	0.19 ± 0.01	0.28 ± 0.03	0.30 ± 0.01	0.10 ± 0.01	0.23 ± 0.07	0.13 ± 0.01
Total	13.35 ± 0.23	13.15 ± 0.04	15.09 ± 0.09	16.90 ± 0.01	13.97 ± 0.02	15.00 ± 0.23	14.36 ± 0.06
SFA/PUFA	0.36 ± 0.03	0.36 ± 0.01	0.34 ± 0.01	0.28 ± 0.11	0.32 ± 0.01	0.30 ± 0.27	0.31 ± 0.01
Oil yield (%)	14.36 ± 0.70	22.07 ± 0.35	13.79 ± 0.58	21.14 ± 0.08	11.57 ± 0.68	25.02 ± 0.10	14.89 ± 0.63

Data were means ± SD from three replicates. C16:0 (Palmitic acid); C16:1n7 (Palmitoleic acid); C18:0 (S
acid); C18:1n9 (Oleic acid); C18:2n6 (Linoleic acid); C20:0 (Arachidic acid); C18:3n3 (Linolenic acid); C22

I.3 Potential uses of the press cakes produced in the single-screw extruder

Aroma Tincto Oleo Crops (ATOC) are plants containing both a vegetable oil and an essential oil. Presently, depending on the industrial sector concerned (oil or aromatic industry), only one of these two fractions is valorised, the other being considered as waste. The development of an integrated valorisation of ATOC allowing co-extraction of vegetable oil and essential oil constitutes the basis of the ATOC-refinery concept, e.g. a sequential valorization process allowing co-extraction of vegetable oil and essential oil while not penalizing the subsequent valorization of residual by-products as biosourced molecules (antioxidants, biocides, etc.) or the solid raffinate for designing odorous agromaterials. The technology approach was based on single-screw extrusion in order to extract (i) a virgin vegetable oil from fruits, (ii) an essential oil from the extruded cake and then (iii) antioxidant and biocide molecule cocktails from the distillated cake, and finally to valorize the ultimate solid residue as starting material for either agromaterial design (by thermopressing or injection-moulding) or bioenergy production (by combustion of formulated pellets). Based on the biorefinery concept that was developed in this thesis, plant materials of coriander were processed into different parts: vegetable oil, essential oil (or volatile extract), methanolic extract and final residue (biocomposite). By this way, the molecules in the plants can be sequentially extracted and used, thus avoiding a great waste of natural resources.

After collecting the oil and solid residue (press cake) from the pressing method, this cake still contained part of the essential oil, its content varying from 0.26 to 0.35% of the dry matter. Maximal essential oil content of 0.35% was found inside the press cake from Lithuanian coriander, and the lowest one (0.26%) was in the press cake from Vietnamese coriander. During the coriander oil expression using single-screw extruder, the temperature applied was never more than 65 °C, and such temperature did not allow strong evaporation of the essential oil contained within the press cake. Residual essential oil in the press cakes could be extracted by means of hydrodistillation. Results of essential oil composition in the different press cakes are shown in Table 23. Fourteen compounds have been identified and the major ones were as follows: linalool, γ-terpinene, camphor, geranyl acetate, α-pinene, limonene, p-cymene. In which linalool still being the main component for all press cakes: 77.8% for Tunisian coriander, 78.8% for Lithuanian coriander, 75.4% for French DS1 cultivar, 76.6% for French cultivar supplied by GSN Company and 78.1% for Vietnamese coriander.

Table 23: Essential oil composition in press cakes from different coriander cultivars and produced in a si

Compound			Essential oil composition (rel %)		
	KIcalc	KIref	Tunisia	Lithuania	French (DS1)
α-Pinene	960	939	2.06 ± 0.2	2.15 ± 0.1	2.63 ± 0.2
Camphene	984	954	0.54 ± 0.2	0.64 ± 0.2	0.40 ± 0.2
β-Pinene	995	979	0.67 ± 0.1	0.76 ± 0.2	0.56 ± 0.2
Myrcene	1011	991	0.30 ± 0.1	0.32 ± 0.1	0.38 ± 0.1
p-Cymene	1046	1025	1.73 ± 0.1	1.12 ± 0.1	1.74 ± 0.1
Limonene	1052	1029	1.30 ± 0.1	1.49 ± 0.2	1.17 ± 0.1
γ - Terpinene	1078	1073	4.27 ± 0.1	3.79 ± 0.1	3.02 ± 0.1
Linalool	1113	1097	77.83 ± 0.2	78.84 ± 0.2	75.40 ± 0.1
Camphor	1189	1146	4.66 ± 0.1	4.78± 0.5	3.86 ± 0.1
Borneol	1211	1169	0.21 ± 0.1	0.31 ± 0.1	1.40 ± 0.2
α-Terpineol	1223	1189	0.33 ± 0.1	0.33 ± 0.1	1.45 ± 0.1
Carvone	1231	1243	0.15 ± 0.3	-	-
Linalyl acetate	1259	1257	1.60 ± 0.1	1.68 ± 0.2	3.54 ± 0.3
Geranyl acetate	1381	1381	4.36 ± 0.2	3.79 ± 0.1	5.44 ± 0.2
EO yield (%)			0.29 ± 0.1	0.35 ± 0.2	0.28 ± 0.2

Method: GC, peak identification through Kovats retention indices. KIcalc: Retention index calculated wit using an apolar column; KIref: Retention index reported in the literature. (-): Not detected.

Another possible use of the press cakes could be the isolation of some natural molecules in order to test their antioxidant activity. Natural antioxidants have attracted attention owing to their potential good effects in health. Evaluation of the antioxidant activity of natural substances has been of interest in the recent years. Antioxidants scavenge free radicals and reactive oxygen species and can be extremely important in inhibiting oxidative mechanisms that lead to degenerative disease (Cardador-Martínez et al., 2002). Free radicals have been implicated as playing a role in the etiology of cardiovascular disease, cancer, Alzheimer's disease and Parkinson's disease. The antioxidant capacity of most plant food sources is usually associated with their phenolic contents. On the other hand, valorization of residues is an opportunity to obtain profit in a sustainable way. In this study, antioxidants were extracted from the residues (press cakes) of coriander fruits obtained through single-screw extrusion, after extraction of essential oil using hydrodistillation. A solvent extraction with methanol as extracting solvent was used to obtain the antioxidants (i.e. methanolic extracts). Then, a comparison of the results was made in order to know which coriander cultivars delivers a residue with higher concentration of antioxidants. The extract was measured by different method assay (FRAP, ORAC, DPPH assay), and the total phenolic compounds were also determined. All extractions were performed in triplicate.

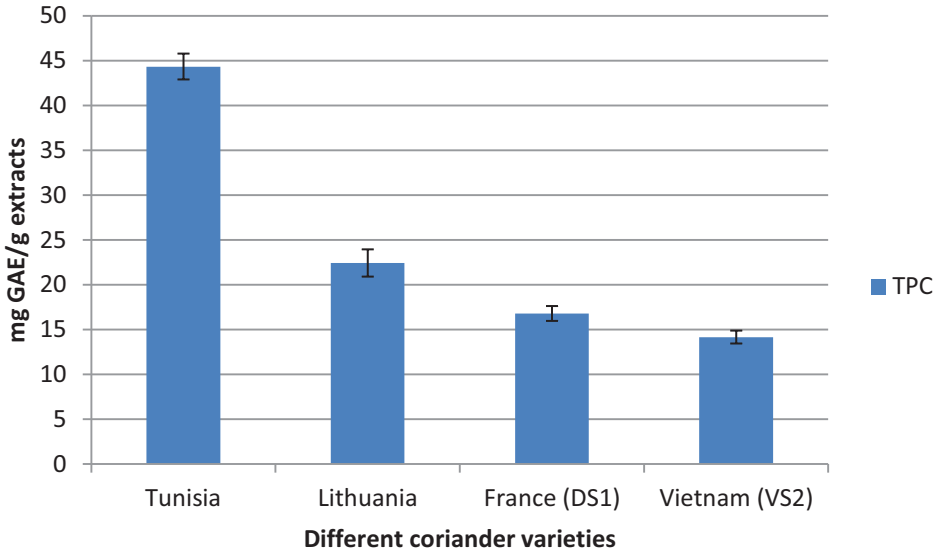


Figure 43: Total phenolic contents (TPC) for the different coriander cultivars used.

The total phenolic content (TPC) of extracts from press cakes is expressed as mg of gallic acid equivalents (GAE) per g of extract. TPC results show that the amount of total

phenolics in extracts of press cakes varies from 14.1 to 44.3 mg GAE/g. The extracts of press cakes from Tunisian coriander reveal the highest TPC value of 44.3 mg GAE/g, followed by extracts of press cakes from Lithuanian coriander (22.4 mg GAE/g extract). On the contrary, the extracts of press cakes from Vietnamese coriander revealed the lowest TPC value (only 14.1 mg GAE/g extract) (Figure 43). Previous studies showed that antioxidant activity is directly related to the total phenolic contents, meaning that the sample having the highest total phenolic content will show the most important antioxidant activity (Kähkönen et al., 1999).

All methanolic extracts of press cakes from the different coriander cultivars were antioxidant active. Their radical scavenging capacity (RSC) varied in a wide range. It was from 0.31 to 0.52 mg/ml in the FRAP reaction system, from 1181 to 2357 µmol TE/g extract in ORAC, and from 22.8 to 52.4 µmol TE/g extract in DPPH test (Table 24).

Table 24: Antioxidant activity and TPC of press cakes produced by single-screw extrusion depending on the coriander cultivars.

Coriander cultivars	**FRAP** mg/ml	**ORAC** µmol TE/g extract (TroloxE/g extract)	**DPPH** µmol TE/g extract	**TPC** mg GAE/g extract
Tunisia	0.41 ± 0.02	2357.23 ± 1.83	22.83 ± 0.54	44.32 ± 1.44
Lithuania	0.52 ± 0.03	1701.66 ± 0.65	30.02 ± 2.59	22.41 ± 1.52
France (DS1)	0.31 ± 0.02	1477.57 ± 1.16	52.37 ± 0.06	16.78 ± 0.83
Vietnam (VS2)	0.38 ± 0.02	1181.36 ± 0.21	49.20 ± 3.13	14.15 ± 0.72

From the Table 24, the extracts of press cakes from Tunisian and Lithuanian cultivars have better antioxidant activity in FRAP assay. FRAP assay measures the ability of the extract to donate electron to iron, and the higher the FRAP value, the higher will be the antioxidant activity. For DPPH assay, is antioxidant activity better when DPPH value is lower. Therefore, the extracts of press cakes from Tunisian and Lithuanian coriander show also a better antioxidant activity (22.8 and 30.0 µmol TE/g extract, respectively) than extracts from France (DS1) and Vietnam. Lastly, in ORAC assay, extracts of press cakes from Tunisian and Lithuanian coriander still reveal a better antioxidant activity. This is directly related to their higher total phenolic contents, thus leading to a higher antioxidant activity.

II. Extraction of coriander oil using the twin-screw extrusion technology

In the mid-1980s, twin-screw extruders got their popularity in the food industry to manufacture specialized food items. At present, the extrusion technology has an important role and is widely used in industry as efficient manufacturing process. Recent years have seen increasing requirements for new products with intricate shapes and small sizes that are beyond the capabilities of single-screw systems. Twin-screw extruders can fill some of these needs. The term 'twin-screw' applies to extruders with two screws of equal length placed inside the same barrel. Twin-screw extruders are more complicated than single-screw ones. But, at the same time, they provide much more flexibility and better control. Comparing vegetable oil extraction from fruits by mechanical pressing using single-screw extruder (SSE) or twin-screw extruder (TSE), the interest of the twin-screw technology is to overcome the problems faced with single-screw extruders. The presence of two screws makes possible to force materials to move forward in the machine, making the propulsion of materials less dependent on friction. The TSE has several advantages over the SSE, i.e. (i) a better feeding and more positive conveyance characteristics, thus allowing the machine to process "hard-to-feed" materials (powders, slippery materials, etc.), (ii) a better mixing and a large heat transfer surface area, thus allowing good control of the feedstock temperature, (iii) a residence time distribution short and narrow, (iv) a good control over residence times and feedstock temperatures along the screw profile, particularly required for thermally sensitive materials, and (v) interchangeable screw and barrel sections for more versatility. Consequently, this study purposed to evaluate the ability of a twin-screw extruder for the extraction of vegetable oil from coriander fruits by mechanical pressing. The effects of both screw configuration (or screw profile) and operating parameters such as pressing temperature, screw rotation speed and fruits input flow rate on oil extraction efficiency were studied. The characterization of oil extraction performance was observed by the determination of oil extraction yields and oil quality. All trials were carried out using a single batch (200 kg) of coriander fruits (GSN maintenaire cultivar) revealing a 9.77 ± 0.10% moisture content.

Experiments were conducted using a Clextral BC 21 (France) co-rotating and co-penetrating twin-screw extruder (Described in Chapter II, section II.1.2.2). In this study, the four screw profiles tested were based on those used in Evon et al. (Evon et al., 2010b) for jatropha oil extraction. The differences between them concerned their pressing zones,

all situated in module 7. The reverse pitch screws used in profiles 1 and 2 were 50 mm long, with a pitch of -25 mm. They were positioned immediately after the filtration module for profile 1, and 25 mm from the end of module 6 for profile 2. The reverse pitch screws used in profiles 3 and 4 had the same length (50 mm), but their pitch was greater (-33 mm instead of -25 mm). They were positioned 25 mm from the end of module 6 for profile 3 and immediately after the filtration module for profile 4 (Figure 44).

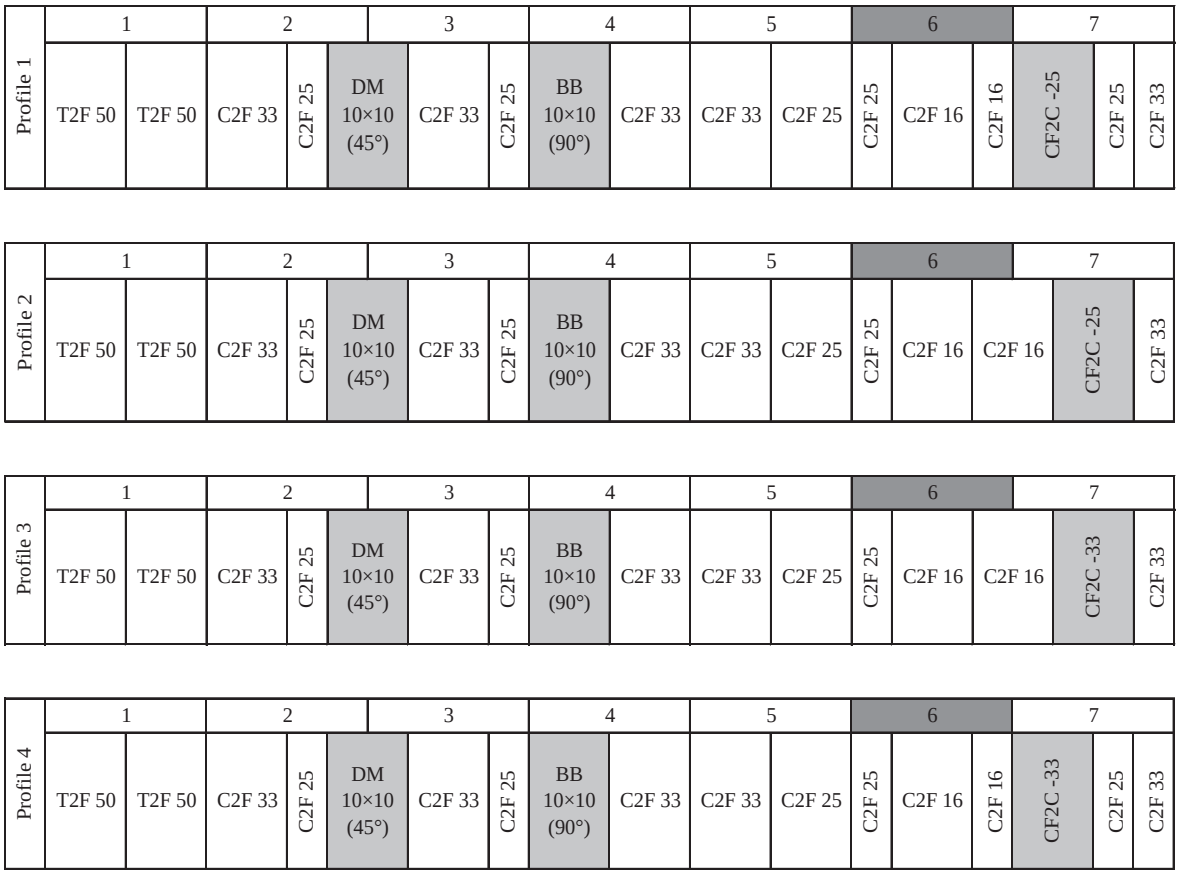


Figure 44: Screw configurations for extraction of vegetable oil from coriander fruits
T2F (trapezoidal double-thread screw); C2F (conveying double-thread screw); DM (monolobe paddle-screw); BB (bilobe paddle-screw); CF2C (reverse screw). The numbers following the type of screw indicate the pitch of T2F, C2F, and CF2C screws and the length of the DM and BB screws.

Ten experiments were conducted (Table 25). Different operating conditions were tested including the screw profile, the device's filling coefficient and the temperature in the pressing zone. Except for trial 4, the screw rotation speed was 100 rpm for all the experiments, meaning that the device's filling coefficient directly depended on the feed flow of coriander fruits.

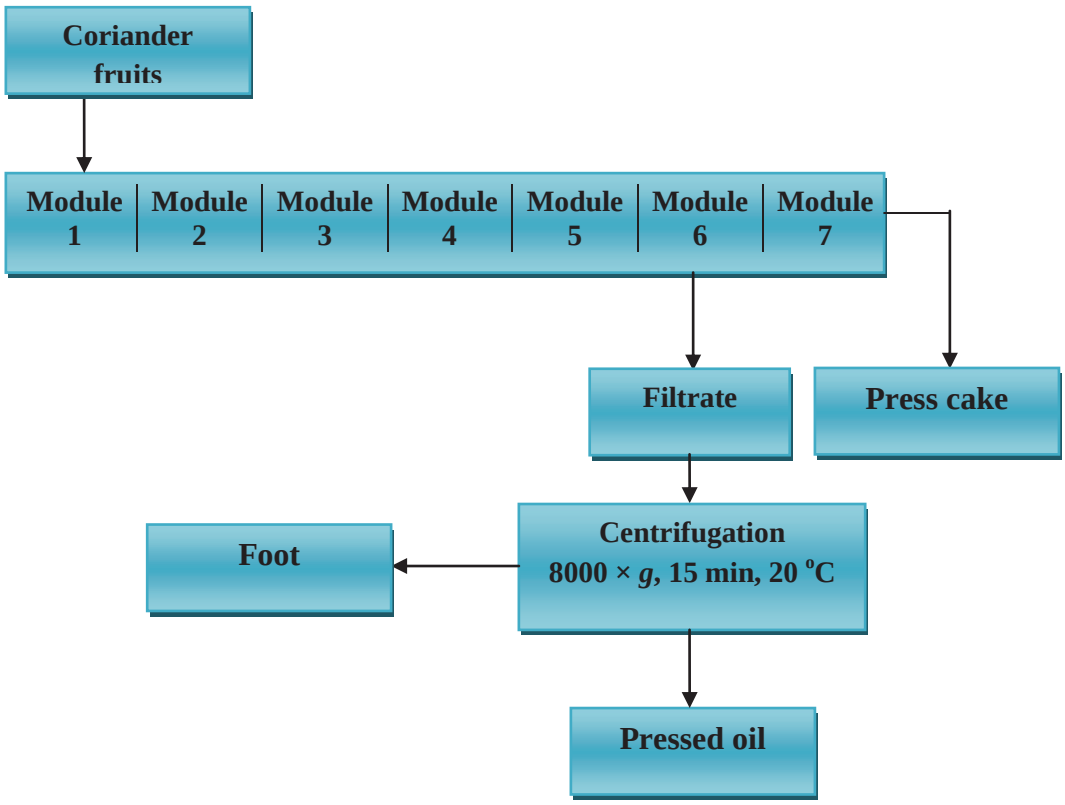


Figure 45: Schematic representation of oil expression of coriander fruits using a twin-screw extruder (type Clextral BC 21).

For each experiment, the extruder was left to function for 30 min before any sampling in order to ensure stabilization of the operating conditions (fruit feed flow, temperature and motor torque). After this, the filtrate and the press cake were immediately collected for a period long enough (30 min) to minimize any variation in the outlet flow rates. Sample collection, timed using a stopwatch, was carried out once for each trial, meaning that the ten experiments were not replicated. As in the case of the single-screw extruder, the filtrate and the press cake were weighed, after which the filtrate was centrifuged (8000 × g, 15 min, 20 °C) to eliminate the foot (Figure 45). The oil yields were calculated from the following formulae:

$$R_L = \frac{Q_F \text{ x } T_L}{Q_S \text{ x } L_S} \text{ x } 100 \quad (1)$$

where R_L is the oil yield relative to the total oil that the fruit contains (%), Q_S the inlet flow rate of coriander fruits (kg/h), Q_F the flow rate of the filtrate (kg/h), T_L the mass content of the pressed oil in the filtrate (%), and L_S the total oil content in the coriander fruits (%).

$$R_C = \frac{(Q_S \text{ x } L_S) - (Q_C \text{ x } L_C)}{Q_S \text{ x } L_S} \text{ x } 100 \quad (2)$$

where R_C is the oil yield based on the residual oil content in the press cake (%), Q_C the flow rate of the press cake (kg/h), and L_C the oil content in the press cake (%). Although R_L and R_C are expressed in terms of the oil in the fruit, R_C is higher than R_L because it includes all oil present in the filtrate (pressed oil and oil contained within the foot). The energy consumed by the motor was determined from the following formulae:

$$P = P_M \text{ x } \frac{S_S}{S_{max}} \text{ x } \frac{T}{T_{max}} \text{ x } \cos\varphi \quad (3)$$

where P is the electrical power supplied by the motor (W), P_M the motor's power rating (P_M = 8300 W), T and T_{max} the test torque and maximum torque (100%) of the extruder motor (%), cos ϕ its theoretical yield (cos ϕ = 0.90), and S_S and S_{max} the test speed and maximum speed (682 rpm) of the rotating screws (rpm), respectively.

$$SME = \frac{P}{Q_S} \quad (4)$$

where SME is the specific mechanical energy consumed by the motor per unit weight of coriander fruits (W h/kg).

$$SME' = \frac{P}{Q_F \text{ x } T_L} \quad (5)$$

where SME' is the specific mechanical energy consumed by the motor per unit weight of pressed oil (W h/kg).

The various constituents were determined according to the following French standards: moisture contents NF V 03-903, mineral contents NF V 03-322, oil contents NF V 03-908, protein contents NF V 18-100. For the vegetable oil content determination, coriander oil was extracted from the initial material (fruits or press cakes) using the Soxhlet extraction apparatus and *n*-hexane as extracting solvent. Oil extraction was carried out in two successive steps, each of 5 h duration, in order to avoid underestimating the oil content. Between these two steps, the material was reground using a Foss Cyclotec 1093 (Denmark) mill fitted with a 300 µm screen. An estimation of the three parietal constituents (cellulose, hemicelluloses, and lignins) contained in the coriander fruits was made using the ADF-NDF method from Van Soest and Wine (Van Soest and Wine,

1968). An estimation of the water-soluble components contained in the coriander fruits was made by measuring the mass loss of the test sample after 1 h in boiling water. All determinations were carried out in duplicate.

II.1 Chemical composition of the coriander fruits used

The vegetable oil content of the coriander fruits used in this study was 27.67 ± 0.57% of the dry matter, and this agrees with the 9.9 - 28.4% results previously reported (Diederichsen, 1996; Griffiths et al., 1992; Khan et al., 1986; Ramadan and Mörsel, 2002; Sriti et al., 2012a; Sriti et al., 2010b). It was higher than the oil content of the raw material used in the only antecedent study that also dealt with the extraction of coriander oil by twin-screw extrusion, which was 21.9% of the dry matter for Tunisian coriander (Sriti et al., 2012a). These variations can be attributed not only to the difference in cultivars but also to some factors like cultivation conditions, especially the use of fertilizers and irrigation (Ravi et al., 2007). Mineral and protein contents were 5.86 ± 0.11% of the dry matter and 14.07 ± 0.43% of the dry matter, respectively. For the three parietal constituents (cellulose, hemicelluloses, and lignin), their contents were 24.38 ± 0.55% of the dry matter, 17.84 ± 0.56% of the dry matter and 11.01 ± 0.25% of the dry matter, respectively. Lastly, the water-soluble components represented 14.32 ± 0.09% of the dry matter in the coriander fruits.

The fatty acid composition of the oil extracted from the coriander fruits using Soxhlet extraction with *n*-hexane as the extracting solvent is presented in Table 26. The oil was rich in petroselinic (72.6%), linoleic (13.8%) and oleic (6.0%) fatty acids, like other coriander oils described in the literature (Ramadan and Mörsel, 2002; Sriti et al., 2012a; Sriti et al., 2010b), and contained only 1.8 ± 0.1% of free fatty acids (FFA) (Table 26). The glyceride profile of Soxhlet extracted oil was determined by GC. The oil contained 96.25 ± 1.05% of triglycerides (TAG), 1.03 ± 0.09% of diglycerides (DAG), and 0.09 ± 0.02% of monoglycerides (MAG). GC analysis also led to a second estimation of the free fatty acid content and the latter was slightly lower (1.02 ± 0.03%) than that obtained by titration (French standard NF T 60-204), i.e. 1.8%. The high amount of TAG and concurrent low FFA content demonstrated that little hydrolysis or enzymatic degradation had occurred in the Soxhlet extracted oil, indicating its good quality.

The coriander fruits displayed an essential oil content of 0.7% of the dry matter through water distillation (Table 28), and this agrees with the 0.3 - 1.2% results reported by Kiralan et al. (Kiralan et al., 2009). Further, the essential oil was particularly rich in linalool (71.7%), a monoterpene alcohol rendering the coriander fruits their characteristic lemony citrus flavor (Anju et al., 2011) (Table 27). Such content was comparable to those (41-80%) mentioned by Asgarpanah and Kazemivash (Asgarpanah and Kazemivash, 2012) and Sahib et al. (Sahib et al., 2013) depending on the cultivars and origin of coriander. Other important compounds were α-pinene and γ-terpinene, representing 5.6% and 5.0% of the essential oil, respectively.

II.2 Influence of the operating conditions on oil extraction efficiency

In order to assess the impact of the extrusion operating conditions on oil extraction, different screw profiles were tested and the device's filling coefficient and pressing temperature were varied. The effect on the extraction efficiency was evaluated through the determination of two oil yields (R_L and R_C), the filtrate's foot content and the extruder's energy consumption. As observed in previous studies dealing with the extraction of sunflower oil (Dufaure et al., 1999; Evon et al., 2007, 2009; Evon et al., 2010b; Kartika et al., 2005, 2006) and jatropha oil (Evon et al., 2013), the extruder was effective for the extraction of coriander vegetable oil because of its capacity to crush the fruits whilst pressing. The trituration zone (monolobe and bilobe paddles) reduced the size of the solid particles significantly, leading to the release of part of the oil, while the compressing action by the reverse pitch screws was essential for liquid/solid separation. Positioned at the beginning of module 7, the CF2C reverse pitch screws pushed part of the mixture upstream against the general movement in the extruder, and this counter pressure ensured the efficiency of the liquid/solid separation above the metal filter.

In the first screw profile tested (profile 1), the CF2C reverse pitch screws were 50 mm long, with a pitch of -25 mm, and they were positioned immediately after the filtration module (Figure 44). With such a screw configuration and a 120 °C pressing temperature, a lot of solid particles were rapidly forced through the filter, preventing the oil from draining freely and thus its separation from a press cake. Then, it simply resulted in the clogging of the twin-screw extruder. This phenomenon persisted even when reducing the device's filling coefficient to less than 10 g/h rpm.

A second screw profile (profile 2) was therefore tested, the same CF2C screws with a -25 mm pitch being then positioned 25 mm from the end of module 6 (Figure 44). However, the same phenomenon, i.e. the clogging of the machine, was observed after only a few tens of seconds. As the CF2C screws with a -25 mm pitch used for screw profiles 1 and 2 were too restrictive, these reverse pitch screws were replaced in screw profiles 3 and 4 by CF2C screws with a higher i.e. less restrictive pitch (-33 mm instead of -25 mm).

For all experiments conducted with these last two screw profiles, filtrate samples and press cake samples were always collected separately. The oil content in the press cake was lower than in the coriander fruits and it varied from 18.1 to 15.0% of the dry matter (Table 25), depending on the applied operating conditions. This led to an oil yield (R_C), based on the residual oil content in the press cake, of between 42.9 and 53.7% (Table 25). As expected, the lower the oil content in the press cake the higher the oil yield (R_C), and this relation was indeed linear. The oil yield (R_L), defined as the ratio of the pressed oil to the total oil contained within the fruit, varied from 28.0 to 46.9% (Table 25). It was lower than the oil yield (R_C) due to the oil retained in the filtrate foot (Figure 46) and the difference between these two oil yields was minimal with the lowest filtrate's foot contents (trials 6 to 8).

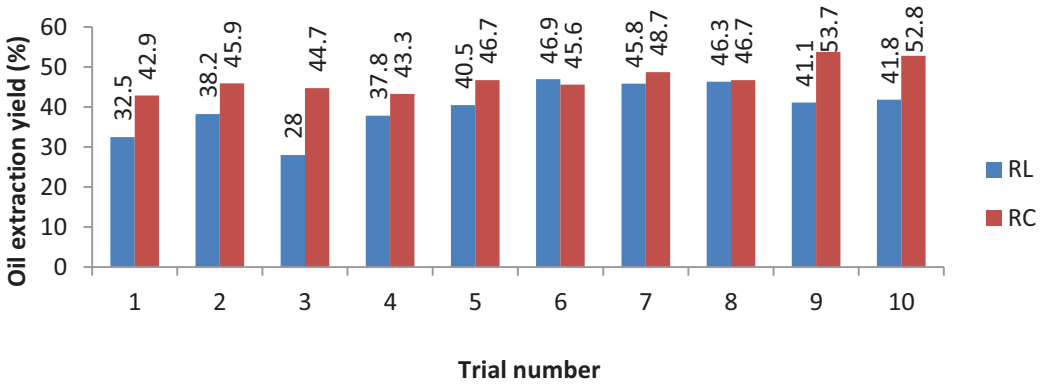


Figure 46: Variation in oil extraction yields for trials 1 to 10

Moreover, the mass content of the foot in the filtrate was never more than 18%, instead of 47-66% for oil pressing from coriander fruits of Tunisian cultivar (Sriti et al., 2012a), meaning that the screw configurations used in this study were much more suited. The use of a filter section with smaller perforations (less than 500 µm in diameter) would probably further reduce the amount of solid particles forced through, without preventing the oil from draining. However, such a filtration module could not be tested in this study. Because the foot is commonly recycled into the press during commercial operations, lowering the filtrate's foot content would also reduce the amount of material for recycling, thus minimizing the impact on material throughput.

Table 25: Results of the expression experiments conducted with the Clextral BC 21 twin

Trial number	1	2	3	4	5	6	7
Operating conditions							
Screw profile	3	3	3	3	4	4	4
S_S (rpm)	100	100	100	133	100	100	100
Q_S (kg/h)	3.12	4.66	6.19	6.19	3.15	3.94	4.71
C_F (g/h.rpm)	31.2	46.6	61.9	46.5	31.5	39.4	47.1
θ_{c6} (°C)	83.0 ± 0.0	79.4 ± 0.8	77.7 ± 0.6	77.9 ± 0.3	95.2 ± 0.8	90.6 ± 0.6	89.1 ± 0.
θ_{c7} (°C)	122.0 ± 4.6	119.0 ± 0.6	118.7 ± 0.5	120.4 ± 0.8	120.1 ± 0.7	118.1 ± 2.1	119.5 ± 0
Filtrate							
Q_F (kg/h)	0.31	0.51	0.49	0.70	0.38	0.52	0.59
T_L (%)	82.8	87.3	88.3	83.5	83.6	89.3	91.5
T_F (%)	17.2	12.7	11.7	16.5	16.4	10.7	8.5
H_F (%)	-	-	-	-	3.32 ± 0.05	3.60 ± 0.01	3.74 ± 0.0
L_F (% of dry matter)	-	-	-	-	59.33 ± 1.71	63.34 ± 2.31	73.48 ± 0.
Press cake							
Q_C (kg/h)	2.58	3.84	5.08	5.07	2.61	3.31	3.92
H_C (%)	2.16 ± 0.02	3.85 ± 0.07	4.60 ± 0.04	4.46 ± 0.06	2.32 ± 0.14	3.73 ± 0.08	4.05 ± 0.0
L_C (% of dry matter)	17.63 ± 0.07	17.06 ± 0.05	17.62 ± 0.03	18.10 ± 0.08	16.48 ± 0.21	16.82 ± 0.59	16.06 ± 0.
Oil yields (%)							
R_L	32.5	38.2	28.0	37.8	40.5	46.9	45.8
R_C	42.9	45.9	44.7	43.3	46.7	45.6	48.7
Energy consumed							
CP (bars)	2.1 ± 0.2	3.6 ± 0.4	5.4 ± 0.5	3.3 ± 0.3	1.4 ± 0.3	1.6 ± 0.2	1.8 ± 0.4
T (%)	16.4	42.7	57.2	32.5	25.0	37.2	45.6
P (W)	180.0	467.9	626.7	473.1	274.2	407.8	499.0
SME (W h/kg fruits processed)	57.7	100.4	101.3	76.5	86.9	103.5	105.9
SME' (W h/kg expressed oil)	710.2	1053.8	1447.2	810.4	859.1	884.1	925.6

C_F is the device's filling coefficient (g/h rpm); it is defined as the ratio of the inlet flow rate of coriander fruits (Q_S) to the temperature measured during sampling at the level of module 6, i.e. the filtration module (°C). θ_{c7} is the barrel temperature at th during sampling), i.e. the pressing zone (°C). Modules 2-5 were heated to 65 °C for all trials. T_F is the mass content of the foot in t the foot of the filtrate (%). L_F is the oil content in the foot of the filtrate (% of the dry matter). H_C is the moisture content in the pres

For all screw profiles tested, the trituration zone was composed of a succession of 10 monolobe paddles and 10 bilobe paddles. It was the same as the one in the optimized screw profile used for the extraction of oil from jatropha seeds by mechanical pressing (Evon et al., 2013). The object of such a configuration was to obtain effective mechanical lysis of the cells and therefore an efficient oil release. In the third screw profile tested (profile 3), CF2C screws with a -33 mm pitch were still positioned 25 mm from the end of module 6 (Figure 44). Four experiments were conducted with this profile (trials 1 to 4). However, when using screw profile 3, there was an accumulation of oil between the end of the filter section and the beginning of the pressing zone which could not drain freely. This has presumably led to a decrease in the filtrate's flow rate as well as in the oil yield (R_L). A fourth screw profile was therefore tested (profile 4), with the CF2C screws with a -33 mm pitch being positioned immediately after the filtration module (Figure 44). This profile was applied for trials 5 to 10. Comparing trials 1 and 5, conducted with screw profiles 3 and 4, respectively, while maintaining the same operating conditions, screw profile 4 was found to be significantly more effective in oil extraction. It further resulted in lower residual oil content in the press cakes and a slight reduction in the filtrate's foot content, leading to an increase in both oil yields (Table 25). However, the increase in the motor's torque resulted in a modest augmentation of the specific mechanical energy per unit weight of pressed oil (+21%). In conclusion, screw profile 4 was considered as the optimized screw configuration for this study.

Next, the influence of the device's filling coefficient on oil extraction efficiency was assessed for screw profiles 3 and 4. When comparing trials 1 to 3, using the third screw profile, the increase in the inlet flow rate of coriander fruits from 3.12 kg/h (trial 1) to 6.19 kg/h (trial 3) directly affected the device's filling coefficient, increasing it from 31 to 62 g/h rpm (Table 25). Because it was more filled, the crushing ability of the trituration zone certainly diminished and the size reduction of solid particles became less significant, thus resulting in a decrease in the filtrate's foot content. Furthermore, the degree of filling of the CF2C screws increased as the device's filling coefficient increased, and this led to a better pressing action on the matter and so to a more efficient liquid/solid separation. This was demonstrated by a significant increase in the motor's torque, resulting in a higher specific mechanical energy per unit weight of pressed oil, and a decrease in the residual oil content of the obtained press cakes. Consequently, the oil yield (R_C) increased with increasing device's filling coefficient. Because the mass content of the foot in the filtrate decreased at

the same time, the oil yield (R_L) also increased, but more significantly. The same tendency has previously been observed for sunflower and jatropha oil pressing (Amalia Kartika et al., 2006; Evon et al., 2013; Kartika et al., 2005).

For trial 3, the device's filling coefficient reached 62 g/h rpm. However, even though the motor's torque still increased, this did not result in a decrease of the press cake's residual oil content, the latter exhibiting a slight increase. With such a device's filling coefficient, the solid particles accumulated more upstream from the pressing zone, obstructing part of the filtering screens and thus reducing the filtration surface. This resulted in the decrease of the filtrate's flow rate, causing a significant reduction in the oil yield (R_L) and an important increase in the specific mechanical energy. With screw profile 3, the machine became in fact too filled with a 62 g/h rpm device's filling coefficient, and higher filling coefficients were not tested to avoid clogging of the twin-screw extruder.

Comparing trials 2 and 4, the device's filling coefficient was the same (47 g/h rpm), but the screw rotation speed and the inlet flow rate of coriander fruits increased in the same proportions for trial 4: from 100 to 133 rpm and from 4.66 to 6.19 kg/h, respectively (Table 25). The increase in the screw rotation speed certainly led to a more effective size reduction of the solid particles at the level of the trituration zone, resulting in an increase of the foot content in the filtrate. Although the press cake's residual oil content was higher due to a less efficient compression of the mixture in the reverse screw elements, there was only a marginal reductive effect on both oil yields (R_L and R_C). Furthermore, the increase in the screw rotation speed significantly reduced the motor's torque, further reducing the specific mechanical energy per unit weight of pressed oil. In conclusion, a device's filling coefficient close to 50 g/h rpm was better suited for such a screw configuration, and higher productivity of the extruder resulted in the production of similar oil yields at a lower cost.

The effect of the device's filling coefficient on oil extraction efficiency was further evaluated for the fourth screw profile through trials 5 to 7. As previously observed with screw profile 3 (trials 1 to 3), a decrease in the filtrate's foot content was observed with the increase of the device's filling coefficient, which is due to a lowering of the crushing ability of the trituration zone, thus leading to larger solid particles at the outlet of this zone. A significant increase in the motor's torque was observed at the same time due to the increase in the degree of filling of the CF2C screws and thus resulting in a better pressing action on the matter in this location. Comparing trials 5 and 6, this further led to an

improvement in the liquid/solid separation, as illustrated by the relative increase in pressed oil quantity, resulting in an increasing oil yield (R_L) without any significant effect on specific mechanical energy. Trial 6 was also the most efficient experiment of the entire study for oil extraction, exhibiting an oil yield (R_L) of 46.9% and producing a press cake with good quality (residual oil content of 16.8% of the dry matter).

For a higher device's filling coefficient (47 g/h rpm, trial 7), the relative quantity of pressed oil slightly decreased compared to trial 6, illustrating the same phenomenon as that observed more significantly for trial 3 with the third screw profile, i.e. the obstruction of part of the filtering screens by an accumulation of solid particles. Despite a slightly excessive filling of the machine, the effect on oil yield (R_L) was quite limited and the corresponding specific mechanical energy only increased by 5% compared to trial 6. At the same time, the press cake was of better quality, resulting in an increase in the oil yield (R_C). Therefore, analogous to what was found for screw profile 3, the 47 g/h rpm device's filling coefficient was considered as a good compromise for screw profile 4 between oil extraction efficiency and the press cake's impoverishment in lipids.

As a second important operating parameter for twin-screw extrusion, the effect of the pressing temperature on oil extraction efficiency was assessed for screw profile 4 using the optimized device's filling coefficient through trials 7 to 10. The applied pressing temperatures were 120 °C, 100 °C, 80 °C and 65 °C, respectively (Table 25). Contrary to what has been reported for sunflower and jatropha seeds (Amalia Kartika et al., 2006; Evon et al., 2013; Kartika et al., 2005), decreasing the pressing temperature from 120 to 65 °C did not substantially improve the oil extraction efficiency. The influence of pressing temperature on the residence time and energy input is significantly correlated with material viscosity and the degree of filling (Gautam, 1999). The highest degree of filling was obtained when the flow of material across the CF2C screws was more viscous, thus the residence time and the energy input increased with decreasing pressing temperature. This led to an increase in both motor's torque and specific mechanical energy per unit weight of fruit processed, rendering the pressed oil slightly more expensive to produce. However, the oil extraction efficiency and filtrate's foot content at 100 °C (trial 8) were quite similar to that obtained at 120 °C (trial 7). At the same time, the press cake's oil contents were comparable between trials 7 and 8, further resulting in similar oil yields (R_C).

Conversely, when pressing temperature was only 80 °C (trial 9) or 65°C (trial 10), the flow of material across the CF2C screws became much more viscous, as illustrated by the high motor's torque values obtained (Table 25). This led to a better compression action on the matter by the reverse pitch screws, and thus to a better impoverishment of the press cakes in lipids. Moreover, press cakes from trials 9 and 10 revealed the two lowest residual oil contents of the entire study: 15.0% and 15.2% of the dry matter, respectively, resulting in the two highest values for the oil yield (R_C): 53.7% and 52.8%, respectively. However, because solid particles accumulated more upstream from the pressing zone for these two lowest pressing temperatures, a significant increase in the foot content in the filtrate was also observed. Therefore, oil yields (R_L) slightly decreased compared to trials 7 and 8, further contributing to an important increase (+ 30% on average) in the specific mechanical energy. In conclusion, the highest oil yield (R_L) for this study was 46.9%, obtained under the following operating conditions (trial 6): profile 4 screw configuration, 39.4 g/h rpm device's filling coefficient and 120 °C pressing temperature (Table 25). In addition, the residual oil content in the press cake was 16.8% of the dry matter. The specific mechanical energy was 103 W h/kg fruit processed or 884 W h/kg pressed oil, which is quite low compared to other samplings in this study. However, these values need to be confirmed when coriander oil is extracted with twin-screw extruders of higher capacity. The amount of foot in the filtrate was only 10.7% under these optimal conditions.

The lowest press cake's residual oil content (15.0% of the dry matter) was obtained under the following operating conditions (trial 9): profile 4 screw configuration, 47.1 g/h rpm device's filling coefficient and 80 °C pressing temperature (Table 25). The oil yield (R_C) was then maximal (53.7%). However, the amount of foot in the filtrate was much more important for these conditions (17.9%). The oil yield (R_L) was slightly lower than for the optimal conditions (41.1% instead of 46.9%). Moreover, the corresponding pressed oil was more expensive to produce (126 W h/kg fruit processed or 1229 W h/kg pressed oil). Reducing the diameter of perforations in the filter section should diminish the filtrate's foot content.

The sudden shutdown and opening of the twin-screw extruder at the end of trial 10 made it possible to observe the location of the matter inside, along the optimized screw profile (profile 4). The monolobe paddles (DM) and especially the bilobe paddles (BB) were filled more than the conveying elements situated in modules 1 to 5. Their crushing ability was

confirmed by the reduction in the size of the solid particles, observed visually after each of these two restrictions even if no accurate measurement of the particle size was carried out. Because of their high pressing action on the matter, leading to the liquid/solid separation, the CF2C screws (beginning of module 7) were logically the elements of the screw profile which were most filled. And, as it was previously observed in the case of Jatropha oil pressing experiments (Evon et al., 2013), the intensive shearing applied to the matter at this location contributed to an additional reduction in the size of solid particles.

In summary, the extraction of vegetable oil from coriander fruits originating from France by mechanical pressing in a twin-screw extruder was improved compared to results obtained in the previous study, i.e. with fruits from the Tunisian cultivar (Sriti et al., 2012a). Indeed, even if the French cultivar was richer in vegetable oil than the Tunisian one (27.7% of the dry matter instead of 21.9%), the lowest residual oil content in the press cake (15.0% of the dry matter) was lower than for the Tunisian cultivar (16.6%). This means that the impoverishment in lipids was much more effective, leading to a better oil yield (R_C): 54% instead of only 45% from the Tunisian cultivar. Furthermore, the filtrate's foot content was never more than 17.9% due to the use of an optimized screw configuration, while it was at least 47.5% in the previous study (Sriti et al., 2012a). This considerably facilitates the pressed oil isolation, i.e. the foot elimination from the filtrate by centrifugation.

II.3 Influence of the operating conditions on oil quality

The quality of pressed oils was examined through two parameters: the acidity and the petroselinic acid content (Table 26). For screw profile 3, the two pressed oils that were analyzed were those from the two most effective trials in terms of oil yield (R_L), i.e. trial 2 and trial 4. For screw profile 4, this included the three pressed oils (trials 7, 8 and 10) that were produced with varying pressing temperatures: 120 °C, 100 °C and 65 °C, respectively. The oil acidity varied between 1.4 and 1.5% for all pressed oils that were analyzed, corresponding to acid values between 2.8 and 3.0 mg of KOH/g of oil (Table 26).

Table 26: Quality and fatty acid composition of Soxhlet extracted oil, and pressed oils from coriander

	Soxhlet extracted oil	**Trial 2**	**Trial 4**	**Trial 7**
Oil quality				
Acid value (mg of KOH/g of oil)	3.58 ± 0.10	2.90 ± 0.08	2.92 ± 0.02	2.80 ± 0.14
Acidity (% FFA as petroselinic acid)	1.80 ± 0.05	1.46 ± 0.04	1.47 ± 0.01	1.41 ± 0.07
Fatty acid composition (%)				
C6:0 (Caproic acid)	0.1 ± 0.1	-	-	-
C14:0 (Myristic acid)	-	0.2 ± 0.1	0.2 ± 0.1	0.1 ± 0.1
C16:0 (Palmitic acid)	2.9 ± 0.1	3.3 ± 0.1	3.3 ± 0.1	3.3 ± 0.1
C16:1 (Palmitoleic acid)	0.4 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1
C17:0 (Margaric acid)	< 0.1	-	-	-
C18:0 (Stearic acid)	< 0.1	0.7 ± 0.1	0.7 ± 0.1	0.7 ± 0.1
C18:1n12 (Petroselinic acid)	72.6 ± 0.1	72.8 ± 0.2	72.9 ± 0.2	73.4 ± 0.2
C18:1n9 (Oleic acid)	6.0 ± 0.2	5.5 ± 0.3	5.4 ± 0.1	4.9 ± 0.2
C18:1n7 (cis-Vaccenic acid)	1.2 ± 0.1	1.3 ± 0.1	1.3 ± 0.1	1.3 ± 0.1
C18:2 (Linoleic acid)	13.8 ± 0.1	13.8 ± 0.1	13.8 ± 0.1	13.8 ± 0.1
C18:3 (Linolenic acid)	0.2 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1
C20:0 (Arachidic acid)	0.1 ± 0.0	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1
C20:1 (Gadoleic acid)	0.2 ± 0.1	-	-	-
SFA	3.2 ± 0.2	4.2 ± 0.4	4.2 ± 0.4	4.2 ± 0.4
MUFA	80.4 ± 0.6	79.7 ± 0.7	79.8 ± 0.5	79.9 ± 0.6
PUFA	14.0 ± 0.2	14.0 ± 0.2	13.9 ± 0.2	14.0 ± 0.2
SFA/PUFA	0.2 ± 0.1	0.3 ± 0.1	0.3 ± 0.1	0.3 ± 0.1
Identified fatty acids	97.6 ± 1.0	97.9 ± 1.3	97.9 ± 1.1	98.0 ± 1.2

SFA: Saturated fatty acids, MUFA: Monounsaturated fatty acids, PUFA: Polyunsaturated fatty acids. (-): Not

Thus, these virgin-type vegetable oils exhibit a higher quality than the Soxhlet extracted one (1.8% acidity). Their acidity was slightly higher than that measured for pressed oils from the Tunisian cultivar of coriander fruits, which was situated between 1.5 and 2.2 mg of KOH/g of oil (Sriti et al., 2012a). However, they were of excellent quality. Next to this, it appeared that screw configuration had no effect on oil quality and that a decrease in the pressing temperature in the twin-screw extruder did not induce an oil quality improvement. This could be due to the fact that pressed oils did not properly come into contact with the pressing temperature, but rather with the temperature at the filter section (i.e. module 6). The latter did not exhibit high temperature variation between the different trials, fluctuating between 76 and 89 °C for the pressed oils analyzed (Table 25).

The fatty acid composition of pressed oils was also largely independent of the operating conditions used for twin-screw extrusion, i.e. screw configuration and pressing temperature (Table 26). As for the oil obtained through solvent extraction, all the analyzed pressed oils were very rich in petroselinic acid, exhibiting contents between 72.8% and 73.4%, while also similar contents of linoleic and oleic fatty acids were found. The minor differences between samples constituted a small variation in the petroselinic/oleic acid ratio. This possibly resulted from a complex integration of GC results, as retention times of both petroselinic and oleic acid were very similar. In conclusion, on the basis of the two quality criteria examined, it was clear that all analyzed pressed oils were high-quality vegetable oils.

II.4 Potential uses of the press cakes produced in the twin-screw extruder

The ten press cakes were fine powders composed of almost spherical particles and their particle size distribution was estimated for press cakes from trials 2 and 10, each corresponding to the highest oil yield (R_C) obtained for the two screw profiles used successfully (i.e. profiles 3 and 4, respectively). It appeared that the particle size distribution of press cakes was independent on the screw profile that was applied (Figure 47). Indeed, the mean diameter of particles in press cakes from trials 2 and 10 was 35 μm and 38 μm, respectively.

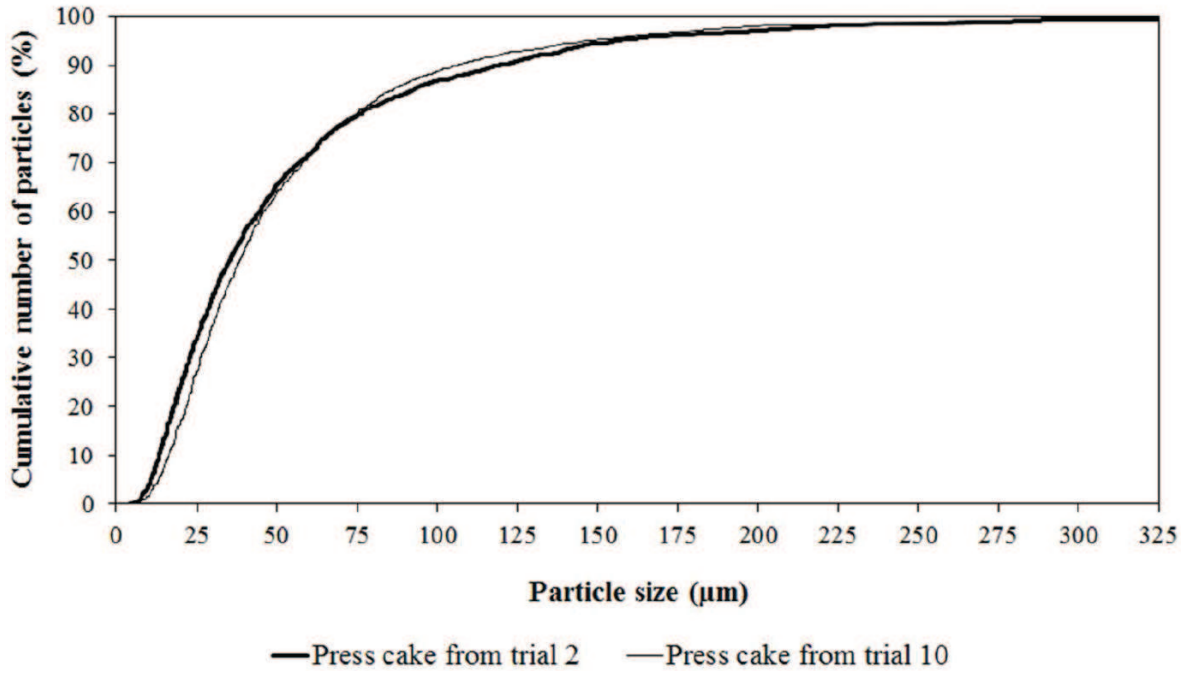


Figure 47: Cumulative number of particles as a function of particle size for the press cakes from trials 2 and 10

Press cakes still contained part of the essential oil from coriander fruits, its content varying from 0.14 to 0.31% of the dry matter (Table 28). Further, the residual essential oil content in the press cake mainly depended on the pressing temperature applied in the twin-screw extruder, increasing with its decrease. A maximal essential oil content of 0.31% was found inside the press cake from trial 10, corresponding to the lowest pressing temperature tested (i.e. 65 °C). Such temperature did not allow strong evaporation of the essential oil contained within the press cake. The residual essential oil content amounted in that case to 40% of the essential oil in the starting material, leading to the conclusion that the remaining 60% was probably co-extracted by mechanical pressing from the coriander fruits with the vegetable oil, rendering it pleasantly scented. Residual essential oil in the press cakes could be extracted by means of hydrodistillation. Its composition was rather similar to that of the essential oil extracted from the fruits (Table 27), linalool still being the main component. Linalool is one of the most commonly applied fragrance ingredients in cosmetics, perfumes, shampoos, soaps and even household detergents due to its fresh and flowery scent and it displays a worldwide use of over 1000 tons per year (Lapczynski et al., 2008). Further applications include its use as a repellent against mosquitoes (Müller et al., 2009). However, because of the low quantities of volatile oil that could be extracted using hydrodistillation and the lack of a strong market for this essential oil, it would

currently not be economical to extract the remaining essential oil from the press cakes. It might therefore be more interesting to leave the volatile oil within other end products such as agromaterials, providing them with an added value.

Table 27: Essential oil composition in coriander fruits and press cakes from trials 2, 4, 7, 8 and 10

Essential oil composition (%)	Coriander fruits	Press cake number 2	Press cake number 4	Press cake number 7	Press cake number 8	Press cake number 10
α-Pinene	5.6 ± 0.1	1.2 ± 0.2	1.2 ± 0.2	2.2 ± 0.1	2.43 ± 0.2	3.73 ± 0.1
Camphene	0.8 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	0.32 ± 0.2	0.37 ± 0.1	0.56 ± 0.1
β-Pinene	1.1 ± 0.2	0.4 ± 0.1	0.4 ± 0.1	0.52 ± 0.1	0.54 ± 0.1	0.70 ± 0.1
Myrcene	0.6 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	0.23 ± 0.1	0.26 ± 0.1	0.34 ± 0.1
p-Cymene	1.6 ± 0.2	0.8 ± 0.1	0.7 ± 0.1	1.12 ± 0.2	1.15 ± 0.1	1.23 ± 0.1
Limonene	1.9 ± 0.2	0.9 ± 0.1	0.8 ± 0.1	1.23 ± 0.1	1.19 ± 0.1	1.43 ± 0.1
γ - Terpinene	5.0 ± 0.5	3.3 ± 0.2	2.8 ± 0.3	3.76 ± 0.1	3.36 ± 0.2	4.13 ± 0.1
Linalool	71.7 ± 1.14	77.0 ± 1.0	77.4 ± 1.1	72.35 ± 1.2	76.10 ± 0.9	74.49 ± 0.4
Camphor	4.2 ± 0.1	4.8 ± 0.1	4.8 ± 0.1	4.52 ± 0.1	4.84 ± 0.2	4.35 ± 0.1
Borneol	1.2 ± 0.1	1.6 ± 0.1	1.5 ± 0.1	1.59 ± 0.1	1.50 ± 0.1	1.29 ± 0.1
α-Terpineol	0.4 ± 0.1	0.5 ± 0.1	0.5 ± 0.1	0.51 ± 0.1	0.46 ± 0.1	0.39 ± 0.1
Carvone	-	0.2 ± 0.1	0.2 ± 0.1	0.21 ± 0.1	0.17 ± 0.1	-
Linalyl acetate	2.9 ± 0.2	4.1 ± 0.2	4.2 ± 0.3	5.0 ± 0.2	3.48 ± 0.2	3.44 ± 0.2
2-(E)-decenal	-	-	0.1 ± 0.1	0.17 ± 0.1	-	-
Geranyl acetate	3.0 ± 0.1	4.8 ± 0.3	4.9 ± 0.3	6.26 ± 0.2	4.16 ± 0.1	3.91 ± 0.2

(-): Not detected.

Another possible use of the press cakes could be the isolation of some natural antioxidants through methanolic extraction, these being potentially interesting due to their beneficial impact on health. The antioxidant capacity of the press cakes was determined through DPPH analysis, and results varied from 11.8 to 77.0 μmol of TE per g of press cake (Table

28). Press cakes from trials 8 and 10 revealed a better antioxidant capacity due to the decrease in the pressing temperature used in the twin-screw extruder.

Table 28: Essential oil in coriander fruits and press cakes from trials 2, 4, 7, 8 and 10, and antioxidant capacity of press cakes 2, 4, 8 and 10 after hydrodistillation (DPPH assay).

	Coriander fruits	**Press cake number 2**	**Press cake number 4**	**Press cake number 7**	**Press cake number 8**	**Press cake number10**
Essential oil content (% of the dry matter)	0.67 ± 0.10	0.14 ± 0.01	0.14 ± 0.01	0.14 ± 0.01	0.17 ± 0.01	0.31 ± 0.02
Antioxidant capacity (µmol TE/g extract (TroloxE/g extract)						
After hydrodistillation	-	65.98 ± 1.15	77.03 ± 0.47	-	11.83 ± 1.27	56.13 ± 1.00

(-): Non determined

In this study, the oil content in the press cakes was at least 15.0% of the dry matter (Table 25). Although this could be a disadvantage for direct utilization of the cakes, they could be converted into usable energy by combustion, gasification or pyrolysis (Gercel, 2002; Yorgun et al., 2001). As a mixture of proteins and lignocellulosic fibers, the press cakes can also be considered as natural composites. Thus, they could be transformed into biodegradable and value-added agromaterials by thermo-pressing (Evon et al., 2014; Evon et al., 2010a; Evon et al., 2012; Evon et al., 2015). Finally, the press cakes could act as reinforcing fillers for biodegradable polymers, i.e. polycaprolactone (PCL) and poly(lactic acid) (PLA), and thus have some potential in biocomposite applications (Diebel et al., 2012; Gamon et al., 2013).

III. New renewable and biodegradable fiberboards from a coriander press cake

As mixtures of proteins and lignocellulosic fibers, the coriander press cakes obtained after mechanical pressing of the fruits using single- or twin-screw extruders can be considered as natural composites. Therefore, they are transformable into biodegradable agromaterials through thermo-pressing (Evon et al., 2014; Evon et al., 2010a; Evon et al., 2012; Evon et al., 2015). This study aimed to evaluate the influence of the thermo-pressing conditions (mold temperature, applied pressure and molding time) on the mechanical properties, the thickness swelling and the water absorption of fiberboards made from a coriander press cake produced in a single-screw extruder.

New renewable and biodegradable fiberboards were manufactured from a coriander press cake by thermo-pressing. The press cake originated from the extraction of vegetable oil from coriander fruits of French origin through mechanical pressing, using an OMEGA 20 (France) single-screw extruder. Its residual oil content was 17.2% of the dry matter instead of 27.7% for the fruits, leading to an oil extraction yield of 47.6% during extrusion. As a mixture of proteins (18.2%) and lignocellulosic fibers (53.8% including cellulose, hemicelluloses and lignins), the press cake was a natural composite. It was crushed before thermo-pressing using an Electra F3 (France) hammer mill fitted with a 15 mm screen. After milling, it consisted of almost spherical particles with a mean diameter of around 170 µm and its apparent and tapped densities were 0.416 and 0.482 g/cm^3, respectively. Lastly, its thermogravimetric analysis (TGA) performed with a Shimadzu TGA-50 (Japan) analyzer revealed that no thermal degradation of the organic compounds in the press cake occurs before 225 °C.

A PEI (France) heated hydraulic press with 400 tons capacity was used for thermo-pressing. The square mold applied was 15 cm × 15 cm, and it was equipped with vents to allow the expression of part of the residual oil during molding. The moisture and the mass of the press cake at molding were 2.1% and 200 g, respectively, for all tested thermo-pressing conditions. These comprised 160-200 °C mold temperature, 24.5-49.0 MPa applied pressure and 60-300 s molding time. The eleven fiberboards resulting from these thermo-pressing conditions (Table 29) were all cohesive, with proteins and fibers acting respectively as a natural binder and reinforcing fillers. When comparing boards from trials 1, 2 and 7, the increase in mold temperature (from 160 to 200 °C) led to a significant reduction in viscosity of the protein-based resin, resulting in the progressive improvement of fiber wetting. This could explain why the board density and its mechanical properties increased as the mold temperature increased, the 200 °C mold temperature (trial 7) leading to the most resistant (1.30 board density, 11.3 MPa flexural strength at break, 2625 MPa elastic modulus, 1.26 kJ/m^2 resilience and 70.8° surface hardness) and the least water-sensitive (51% thickness swelling and 33% water absorption) fiberboard. For this optimal mold temperature, both the applied pressure and the molding time affected the board density, its mechanical properties, its thickness swelling and its water absorption. For the 60 s molding time (trials 3 to 5), the increase in the applied pressure resulted in an increased board density, enhanced mechanical properties and in a decreased water-sensitivity. Next to this, further improvement of the flexural properties, the surface hardness, the thickness swelling and the water absorption was obtained for molding times of at least 180 s (trials 6 to 11) with every tested condition of applied pressure.

Table 29: Thermo-pressing conditions, mechanical properties, thickness swelling (TS) and water absor manufactured.

Trial	Panel 1	Panel 2	Panel 3	Panel 4	Panel 5	Panel 6	Panel 7	Panel 8
Thermo-pressing conditions								
Temperature (°C)	160	180	200	200	200	200	200	200
Pressure (MPa)	36.8	36.8	24.5	36.8	49.0	24.5	36.8	49.0
Time (s)	180	180	60	60	60	180	180	180
Flexural properties (French standard NF IN 310) [1]								
t (mm)	6.36	6.20	6.52	6.24	6.11	6.15	5.93	6.00
d	1.27	1.28	1.24	1.27	1.28	1.29	1.30	1.29
F (N)	36.6	52.0	30.3	49.7	66.2	70.6	79.5	68.4
σ_f (MPa)	4.5	6.8	3.6	6.4	8.9	9.3	11.3	9.5
E_f (MPa)	929	1457	785	1418	1895	2141	2625	2074
Charpy impact strength at 23 °C (French standard NF EN ISO 179, unnotched test specimens)[2]								
W (mJ)	70	67	73	82	86	80	75	82
K (kJ/m^2)	1.09	1.08	1.12	1.32	1.40	1.30	1.26	1.36
Surface hardness (French standard NF EN ISO 868)								
Shore D (°)	60.8	63.6	59.5	65.0	66.2	68.8	70.8	69.7
Thickness swelling and water absorption (French standard NF EN 317)[3]								
TS (%)	157	160	148	137	124	69	51	42
WA (%)	167	112	173	71	67	39	33	42

[1] 30 mm specimen width and 100 mm grip separation. [2] 10 mm specimen width and 25 mm grip separat thickness, d: density, F: breaking load, σ_f: flexural strength at break, E_f: elastic modulus. W: absorbed energy

The best compromise between mechanical properties, thickness swelling and water absorption is represented by the board from trial 7 (Table 29). It was produced from the following thermo-pressing conditions: 200 °C mold temperature, 36.8 MPa applied pressure and 180 s molding time. Moreover, the pressure applied during molding resulted in the expression of part of the residual oil in the press cake. This led to a decrease in the residual oil content inside fiberboards (7.7% of the dry matter for trial 7, and up to 7.0% for trial 10) and to an increase in the total oil yield (from 47.6% after extrusion to 79.7% after thermo-pressing for trial 7, and up to 81.2% for trial 10).

In conclusion, in regard to the mechanical properties of the board 7 and its water-sensitivity, such a fiberboard would be applicable as inter-layer sheets for pallets, for the manufacture of containers or furniture, or in the building trade (floor under-layers, interior partitions or ceiling tiles). Moreover, thermo-pressing not only produces cohesive fiberboards, but also significantly increases the oil extraction efficiency.

IV. Conclusion

- ✓ For the extraction of coriander vegetable oil using the single-screw extrusion technology, oil extraction yield depends on the next operating conditions: the nozzle diameter and the nozzle/screw distance. The increase in the nozzle diameter (up to 8-10 mm) leads to an increase in oil yield and maximum oil extraction yield (i.e. 52%) was obtained from the extrusion conditions allowing the strongest material pressure and oil expression, i.e. 3 mm nozzle/screw distance and 9 mmnozzle diameter. The acidity of the obtained pressed oil is low (less than 2%). Besides, the fatty acid composition of pressed oils is unaffected by the modification of the machine parameters. It is characterized by the abundance of monounsaturated fats (80%), in which petroselinic acid is the major compound (varying from 73.5 to 75.8% depending on the coriander culitvars used as starting material). Finally, the press cakes may be used as sources of both an essential oil with high linalool content (75%) and natural antioxidants. Indeed, all produced press cakes revealed a promising antioxidant activity, especially the press cakes from the Tunisian and Lithuanian cultivars. This is related to the higher total phenolic contents of their methanolic extracts.
- ✓ The extraction of vegetable oil from coriander fruits by mechanical pressing was also carried out successfully using the twin-screw extrusion technology. The

operating conditions investigated in this study (i.e. the screw configuration, the device's filling coefficient and the pressing temperature) had an influence on the oil yield and the specific mechanical energy (per unit weight of pressed oil). The foot contents in the filtrate were always low (8-18%), and best oil extraction efficiencies were obtained with -33 mm reverse pitch screws, 50 mm in length and positioned in the pressing zone immediately after the filtration module. The oil extraction yield was at least 41% with such a screw profile, and it reached 47% under operating conditions of 39.4 g/h rpm device's filling coefficient and 120 °C pressing temperature. The mass content of the foot in the filtrate was only 11% under these conditions and the quality of the corresponding press cake was reasonable, with a residual oil content of less than 17% of the dry matter. Better impoverishment in lipids of the press cake (15%) was obtained with the increase in the device's filling coefficient (47.1 g/h rpm) and the decrease in the pressing temperature (80 °C), leading to a 54% oil yield based on the press cake's residual oil content. However, the filtrate's foot content was much more important (18%) for such conditions. Moreover, the extrusion process systematically produced a pleasantly scented oil of good quality (<1.5% acidity), with high petroselinic acid content (73%). Lastly, the press cakes may be used as sources of an essential oil with high linalool content (77%) and/or natural antioxidants.

- ✓ As natural composites, the press cakes can be transformed into renewable and biodegradable fiberboards by thermopressing, proteins and lignocellulosic fibers acting respectively as a natural binder and reinforcing fillers. The best compromise between mechanical properties (i.e. bending properties, surface hardness and impact strength), thickness swelling and water absorption is represented by the board molded from the next conditions: 200 °C mold temperature, 36.8 MPa applied pressure and 180 s molding time. In regard to its mechanical properties (11.3 MPa flexural strength at break, 2.6 GPa elastic modulus and 71° Shore D surface hardness) and its water-sensitivity (51% thickness swelling and 33% water absorption), such a fiberboard would be applicable as inter-layer sheets for pallets, for the manufacture of containers or furniture, or in the building trade (floor under-layers, interior partitions or ceiling tiles). Moreover, thermo-pressing not only produces cohesive fiberboards, but also significantly increases the oil extraction

efficiency (up to 81% for the total oil extraction yield, i.e. after extrusion plus thermo-pressing).

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GENERAL CONCLUSION

- ✓ In terms of essential oils and vegetable oils yields and fatty acid profile, we can realize that coriander (*Coriandrum sativum* L.) are a potential class of ATOC resources. According to the described results, different parts of coriander (flower, leaves, stem, fruits and roots) are obtained both essential oil and vegetable oil, of which the fruit will be optimal for efficient exploitation more than other parts of coriander (flower, leaves, stem and roots). This study indicates that these organs are excellent sites for the bioaccumulation of functional secondary metabolites.
- ✓ The changes of fatty acid composition of coriander ripening had also defined the influence of maturity stages, this is the preliminary report to study the oil and fatty acid accumulation of Coriander during fruit ripening cultivated under organic conditions in three seasons (2009, 2010 and 2011). Our results demonstrated that the highest oil yield was achieved at the full maturity. Fatty acid profiles varied greatly during fruit ripening. At earlier stages, saturated and polyunsaturated fatty acids were higher and decreased with maturity of fruit. Petroselinic acid was the major fatty acid after 12 DAF. This latter showed an inversely evolution with palmitic acid which may support a functional correlation between both fatty acids. This study provided data for use coriander oil and its composition in fatty acids for different industrial applications due its content in petroselinic acid.
- ✓ Variation of fatty acid composition of coriander had also defined the influence of growing region. Base on the results, we can see that different ecological areas especially temperature, humidity and rainfall will be significant impacted in both fruits oil content and fatty acid composition in coriander. Therefore, growth location is an important factor for meeting market requirements of coriander fruits in terms of oil quality. Apart from growing condition, one should also consider some other factors such as cultivar oil type, planting time and irrigation in order to obtain a desire yield and quality.
- ✓ Sowing date is one of the important factors influencing fatty acid composition and oil yield of coriander fruits. The results on yield and fatty acid composition of different sowing times have already mentioned above. There were significant differences of oil content between three sowing times, DS1 (25th March) showed

the highest values of oil yields as compared with significant differences in yield of DS2 (14th April) and DS3 (30th April). So it could be recommended planting coriander at the end of March is appropriate time for good crops. The sowing date effect with yield and fatty acid showed there was meaningful effect. The sowing date is determined to provide the conditions for the plants to maximally use environmental parameters, especially temperature and radiation. If environmental factors are appropriate for the greening, establishment and survival of seedlings, then the maximum yield can be realized by appropriate sowing date. Moreover, the establishment of appropriate sowing date will be useful for healthy plants and in a farm surface is a basic element of a successful agricultural system.

- ✓ Researches on vegetable oil and essential oil composition of different coriander cultivars (the seven cultivars - Canada, Tunisia, Lithuania, Algeria, French (FR1 (Dourou) - DS1), Vietnam (VS2) and China) was performed to evaluate coriander cultivars pertaining to its fruits yield performance in Auch. From results, Tunisian and Lithuania cultivars could be voted for planting under organic conditions (in Auch) because of its potential for exploitation. It is necessary to develop more suitable cultivars for fruits production to fulfill the increasing demand of this spice crop. Selection of better cultivars can be of immense value to the breeder for further improvement and development of the crop. Therefore, the present investigation was undertaken in order to evaluate the appropriate conditions of the collected coriander cultivars and to select the promising cultivars for higher fruits yield in France.
- ✓ For the researches on antioxidant capacity, antioxidant activities of methanolic extract of the seven coriander cultivars was determined. Coriander antioxidant activity was relatively high for the plant to be a new and natural source of antioxidant substances for its use as natural additives in food. Tunisian and Canadian coriander cultivars showed the highest amount on total polyphenol and Tunisian one showed the strongest DPPH scavenging activity, FRAP and ORAC assay. The FRAP, ORAC and DPPH technique was simple, rapidly performed, thus it would be an appropriate technique for determining antioxidant in coriander extract.
- ✓ For the extraction of coriander vegetable oil using the single-screw extrusion technology, oil extraction yield depends on the next operating conditions: the

nozzle diameter and the nozzle/screw distance. The increase in the nozzle diameter (up to 8-10 mm) leads to an increase in oil yield and maximum oil extraction yield (i.e. 52%) was obtained from the extrusion conditions allowing the strongest material pressure and oil expression, i.e. 3 mm nozzle/screw distance and 9 mmnozzle diameter. The acidity of the obtained pressed oil is low (less than 2%). Besides, the fatty acid composition of pressed oils is unaffected by the modification of the machine parameters. It is characterized by the abundance of monounsaturated fats (80%), in which petroselinic acid is the major compound (varying from 73.5 to 75.8% depending on the coriander cultivars used as starting material). Finally, the press cakes may be used as sources of both an essential oil with high linalool content (75%) and natural antioxidants. Indeed, all produced press cakes revealed a promising antioxidant activity, especially the press cakes from the Tunisian and Lithuanian cultivars. This is related to the higher total phenolic contents of their methanolic extracts.

- ✓ The extraction of vegetable oil from coriander fruits by mechanical pressing was also carried out successfully using the twin-screw extrusion technology. The operating conditions investigated in this study (i.e. the screw configuration, the device's filling coefficient and the pressing temperature) had an influence on the oil yield and the specific mechanical energy (per unit weight of pressed oil). The foot contents in the filtrate were always low (8-18%), and best oil extraction efficiencies were obtained with -33 mm reverse pitch screws, 50 mm in length and positioned in the pressing zone immediately after the filtration module. The oil extraction yield was at least 41% with such a screw profile, and it reached 47% under operating conditions of 39.4 g/h rpm device's filling coefficient and 120 °C pressing temperature. The mass content of the foot in the filtrate was only 11% under these conditions and the quality of the corresponding press cake was reasonable, with a residual oil content of less than 17% of the dry matter. Better impoverishment in lipids of the press cake (15%) was obtained with the increase in the device's filling coefficient (47.1 g/h rpm) and the decrease in the pressing temperature (80 °C), leading to a 54% oil yield based on the press cake's residual oil content. However, the filtrate's foot content was much more important (18%) for such conditions. Moreover, the extrusion process systematically produced a pleasantly scented oil of good quality (<1.5% acidity), with high petroselinic acid

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List of publications

Publications

1. Oil and fatty acid accumulation during coriander (*Coriandrum sativum* L.) fruit ripening under organic cultivation. Quang-Hung Nguyen, Thierry Talou, Mureil Cerny, Philippe Evon, Othmane Merah, (2015). The Crop Journal.

2. Extraction of coriander oil using twin-screw extrusion: Feasibility study and potential press cake applications. E. Uitterhaegen, Q.H. Nguyen, K.A. Sampaio, C.V. Stevens, O. Merah, T. Talou, L. Rigal, Ph. Evon. Journal of the American Oil Chemists - 2015.

Oral communication

Études de la bioaccumulation et du bioraffinage des huiles végétales et huiles essentielle de Coriandre (*Coriandrum sativum* L.). Sixiemes Journees de l'ED, 23 - 24 Mai 2013 - CEMES.

Posters

1. ATOC – Refinery: Sequential Valorization of Agromolecules Extracted from Coriander Seeds, Leaves and Roots. Quang Hung NGUYEN, Jazia SRITI, Ana CODONER MONTOYA, Philippe EVON, Othmane MERAH and Thierry TALOU. 7th International Conference on Renewable Resources & Biorefineries (RRB 7), 8-10 June 2011, Bruges – Belgium.

2. ATOC-refinery: aroma-tincto-oleo-crops as a source of bio-active extracts for biosourced additives. Quang Hung NGUYEN, Othmane MERAH and Thierry TALOU. 8th International Conference on Renewable Resources & Biorefineries (RRB 8), June 4-6, 2012, Toulouse – France.

3. AROMA-TINCTO-OLEO-CROPS: Potential worldwide sources for bioactive extracts, agromaterials, pelleted feeds and green charcoal. Q.H. NGUYEN, O. MERAH, P. EVON, T. TALOU. 9th International Conference on Renewable Resources & Biorefineries (RRB 9), 5-7 Juin 2013, Anvers – Belgium.

4. Anthorefinery for food, feed and non food: dream or reality? T. TALOU, T.M. ZHAO, Q.H. NGUYEN. 9th Baltic Conference on Food Science and Technology (FOODBALT 2014) Jelgava (Letonnie), 8-9 Mai 2014

5. Coriander biorefinery - Reactive extrusion and oleochemistry. E. Uitterhaegen, K. Sampaio, E. Delbeke, Q.H. Nguyen, P.Evon, O. Merah, C. Stevens, T. Talou. 10th International Conference on Renewable Resources & Biorefineries (RRB 10) Valladolid (Espagne), 4-6 Juin 2014.

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Oil and fatty acid accumulation during coriander (*Coriandrum sativum* L.) fruit ripening under organic cultivation

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ABSTRACT

To evaluate the accumulation of oil and fatty acids in coriander during fruit ripening, a field experiment was conducted under organic cultivation conditions in Auch (near Toulouse, southwestern France) during the 2009 cropping season. The percentage and composition of the fatty acids of coriander were determined by gas chromatography. Our results showed that rapid oil accumulation started in early stages (two days after flowering, DAF). Twelve fatty acids were identified. Saturated and polyunsaturated acids were the dominant fatty acids at earlier stages (2–12 DAF), but decreased after this date. After this stage, petroselinic acid increased to its highest amount at 18 DAF. In contrast, palmitic acid followed the opposite trend. Saturated and polyunsaturated fatty acids decreased markedly and monounsaturated fatty acids increased during fruit maturation. It appears that the fruit of coriander may be harvested before full maturity.

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1. Introduction

Coriander (*Coriandrum sativum* L.), an annual herb belonging to the Apiaceae family, is a Mediterranean indigenous plant [1]. The species is grown mostly in temperate areas around the Mediterranean basin and in India, China, Thailand, and Eastern Europe [2]. It is used as an herbal condiment in many culinary preparations. The plant is cultivated for its seeds, which are used for many purposes including aromatherapy, food, drugs, cosmetics, and perfumery. The seeds also have medicinal uses in treatment of rheumatism, gastrointestinal complaints, flatulence and gastralgia, worms, insomnia, anxiety, loss of appetite, and glycemia [3,4]. In industry, the main product from coriander is distilled oil and solvent-extracted oleoresin for aroma and flavor production [5]. Coriander oils are familiar not only in the perfumery, food, beverage, and pharmaceutical industries, but also in medicine. They are used as antioxidants, in treatment of nervous disorders, for gut modulation, blood pressure lowering, and diuretic activity, as an anti-diabetic and antimicrobial agent, and in many traditional remedies for various diseases [6–8]. xThe use of coriander seeds as a spice is widespread. Coriander represents 25–40% of curry powder, and is used to flavor liqueurs, being an important flavoring agent in gin production. Coriander seeds are also used in the preparation of baked goods and tobacco products [1].

Interest in coriander seed oil has increased since the European Union authorized the use of coriander oil as a food supplement. The accumulation of oil in coriander seed is thus of great interest for food use. Knowledge of its seed oil accumulation with the aim of maximizing oil production has become important.

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Petroselinic acid is an unusual fatty acid that occurs primarily in seeds. This fatty acid composes nearly 85% of the total fatty acids of Apiaceae seeds [9]. It can be oxidatively cleaved to produce a mixture of lauric acid, a compound useful in the production of detergents, and adipic acid, a C_6 dicarboxylic acid used in the synthesis of nylon polymer [10].

To our knowledge, aside from two reports [10,11], the accumulation of lipids and fatty acids during the seed development of *C. sativum* L. has been little studied. Moreover, both studies were performed under conventional cultivation or in the greenhouse. No study of the accumulation of fatty acids in coriander seeds under organic conditions has been reported. The aim of this study was the evaluation of oil content and fatty acid composition from flowering to maturity of coriander fruits under organic cultivation.

2. Material and methods

2.1. Location and plant experiment

A field trial was conducted in south-western France at the Regional Centre of Experimentation in Organic Agriculture at Auch (near Toulouse, southwestern France, 43°38′47″ N, 0°35′ 08″ E) during the 2009 cropping season. Sowing was performed on March 23, 2009. Seeds of the French coriander cultivar Dourou (GSN, Riscle, France) were directly sown by hand into the field at a rate of 1.2 g m^{-2} to a depth of 3 cm.

The crops were managed under completely organic and rainfed conditions without chemical addition. Weeds were mechanically removed. The soil was a clay loam (organic matter content: 3.2%; pH 8.1) with a depth of about 1.2 m. Flowering started at the end of May and maturation occurred at the beginning of August.

Table 1 shows temperatures and rainfall during the plant cycle in comparison to weather data for the last 55 years. Indeed, rainfall was at least 50 mm lower than the half-century precipitation observed for the same period in this area. The 2009 growing season was less rainy and hotter than the mean values of 55 years.

Table 1 – The prevailing weather condition during the 2009 plant cycle.

Month	Rainfall		Temperature	
	2009	55 years	2009	55 years
January	95.4	65.0	4.9	5.7
February	40.4	55.9	5.8	6.9
March	23.6	57.1	8.7	9.2
April	105.4	66.6	11.7	11.5
May	44.4	77.2	16.8	15.3
June	39.4	61.1	20.1	18.9
July	41.2	47.1	21.7	21.5
August	20.0	58.3	22.7	21.3
September	46.8	55.9	18.9	18.5
October	35.6	56.8	15.0	14.4
November	102.2	57.2	10.9	9.0
December	34.6	68.1	5.9	6.2
Mean: sum over year	629.0	726.3		
Mean: sum Mar.–Sept.	320.8	423.3		
Mean: sum May–July	125.0	185.4		

2.2. Oil and fatty acid composition measurements

Seed sampling was performed every five (on average) days from flowering to maturity. Harvesting continued from 2 days after flowering (DAF) to 53 DAF. The fruit's color and relative moisture content were adopted as ripening criteria. Seed water content (SWC in % of the seed dry matter) was measured for each sample as an indicator of stage of physiological maturity. Moisture contents were determined by heating in an air oven at 60 °C to constant weight.

Dry coriander seeds were ground in an electric grinder (IKA MF-10-basic Microfine grinder, Sigma Aldich, Frankfurt, Germany). Triplicate samples of 20–30 g were subjected to conventional extraction for 5 h with cyclohexane in the dark. The solvent was removed in a rotary evaporator under low pressure at 35 °C. The vacuum system was used to dry the oil at 35 °C overnight. The oil yield was determined. Oil (20 mg) was extracted in a Soxhlet extractor for 5 h, and then 1 mL *tert*-butyl methyl ether was added. The mixture was filtered through a glass fiber filter (GHP, 0.45 μm, small diameter). At this step 100 μL of filtrate was added to 50 μL of trimethylsulfonium hydroxide 0.5 mol L^{-1} in methanol and stirred gently. Fatty acid analysis was performed by gas chromatography (GC-3900) with a flame ionization detector with a CP-select CB for FAME fused silica WCOT column of length: 50 m, internal diameter: 0.25 mm, and film thickness: 0.25 μm. The carrier gas was helium with a flow rate of 1.2 mL min^{-1} and the split ratio was 1:100. The initial oven temperature was programmed to 185 °C for 40 min, increasing at 15 °C min^{-1} to 250 °C and held for 10.68 min. (analysis time: 55.0 min). The injection and detector temperature were held at 250 °C for 55 min. Analyses were performed in triplicate.

All data were subjected to variance analysis using the GLM procedure of SAS (SAS Institute, Cary, NC, USA). Mean comparisons were performed with a Duncan test at the 0.05 probability level.

3. Results and discussion

The changes in oil yield of coriander from flowering period to maturity (53 days) are presented in Table 2. Water content decreased markedly from flowering to maturity (Table 2). Oil yield varied between 4.6% and 25.1% at different stages of fruit ripening (Table 2). Oil content increased gradually from flowering to maturity. Oil content increased threefold from 2 to 12 DAF. At maturity, the oil content reached its highest level (25.1%). The oil yield in the mature stage of ripening was slightly lower than that previously reported for coriander under conventional agriculture [10,12] but higher than the values reported by Angelini et al. [9]. This result was expected, given that organic cultivation is considered a stress condition [13]. Moreover, it is well known that oilseeds produce and accumulate less oil under drought than under favorable conditions [14,15]. This difference may be explained by the genetic origins of the cultivars used in these studies [13]. The oil yield increased rapidly from days 10 to 34 after the flowering sampling period and reached its maximum level at maturity, when the oil yield was maximal and its value was similar to those of other reports describing

Table 2 – Changes in fatty acid and water content and oil yield during seed maturation in coriander (*Coriandrum sativum* L.) fruit in 2009 from flowering to maturity.

Fatty acid (%)	Days after flowering								
	2	5	10	12	14	18	25	35	53
Saturated fatty acid (SFA)									
C14:0 (myristic acid)	$4.6^{a} \pm 0.9$	$4.2^{a} \pm 0.7$	$3.6^{a} \pm 0.4$	$1.6^{b} \pm 0.3$	$1.6^{b} \pm 0.4$	$0.9^{c} \pm 0.2$	$0.3^{d} \pm 0.1$	$0.2^{d} \pm 0.0$	$0.3^{d} \pm 0.1$
C16:0 (palmitic acid)	$20.5^{a} \pm 0.7$	$21.1^{a} \pm 1.1$	$20.9^{a} \pm 1.3$	$10.9^{b} \pm 0.8$	$7.6^{c} \pm 0.9$	$4.0^{de} \pm 0.6$	$3.5^{e} \pm 0.7$	$3.7^{e} \pm 0.4$	$5.1^{d} \pm 0.9$
C18:0 (stearic acid)	$5.8^{a} \pm 0.5$	$5.1^{a} \pm 0.3$	$6.2^{a} \pm 0.3$	$1.7^{b} \pm 0.2$	$1.4^{b} \pm 0.1$	$0.6^{c} \pm 0.1$	$0.8^{c} \pm 0.1$	$0.8^{c} \pm 0.1$	$1.0^{c} \pm 0.1$
C20:0 (arachidic acid)	$0.1^{c} \pm 0.0$	$0.2^{b} \pm 0.0$	$0.3^{a} \pm 0.1$	$0.3^{a} \pm 0.1$	$0.3^{a} \pm 0.1$	$0.2^{b} \pm 0.0$	$0.1^{c} \pm 0.0$	$0.2^{a} \pm 0.0$	$0.1^{c} \pm 0.0$
C22:0 (behenic acid)	$1.6^{b} \pm 0.5$	$1.5^{b} \pm 0.3$	$4.0^{a} \pm 0.6$	$0.0^{c} \pm 0.0$	$0.0^{c} \pm 0.0$	$0.0^{c} \pm 0.0$	$0.0^{c} \pm 0.0$	$0.0^{c} \pm 0.0$	$0.0^{c} \pm 0.0$
Total	$32.6^{a} \pm 1.4$	$32.0^{a} \pm 1.5$	$34.9^{a} \pm 1.3$	$14.4^{b} \pm 0.7$	$10.8^{c} \pm 0.7$	$5.7^{d} \pm 0.3$	$4.6^{e} \pm 0.5$	$4.9^{de} \pm 0.6$	$6.6^{d} \pm 0.9$
Monounsaturated fatty acid (MUFA)									
C18:1n12 (petroselinic acid)	$2.8^{e} \pm 0.3$	$3.6^{e} \pm 0.4$	$7.8^{d} \pm 0.9$	$48.8^{c} \pm 1.5$	$59.3^{b} \pm 1.7$	$74.4^{a} \pm 2.1$	$76.4^{a} \pm 2.1$	$74.7^{a} \pm 2.3$	$71.9^{a} \pm 2.2$
C18:1n9 (oleic acid)	$2.8^{d} \pm 0.2$	$3.1^{cd} \pm 0.2$	$3.5^{c} \pm 0.5$	$6.4^{a} \pm 0.9$	$6.4^{a} \pm 0.7$	$4.6^{b} \pm 0.5$	$4.6^{b} \pm 0.4$	$4.8^{b} \pm 0.5$	$6.2^{a} \pm 0.5$
Total	$5.6^{e} \pm 0.6$	$6.7^{e} \pm 0.6$	$11.3^{d} \pm 0.8$	$55.2^{c} \pm 1.2$	$65.7^{b} \pm 1.9$	$79.1^{a} \pm 1.8$	$81.1^{a} \pm 1.9$	$79.5^{a} \pm 2.3$	$78.2^{a} \pm 2.8$
Polyunsaturated fatty acid (PUFA)									
C18:2n6 (linoleic acid)	$44.2^{a} \pm 2.1$	$45.1^{a} \pm 1.9$	$36.0^{b} \pm 1.8$	$26.9^{c} \pm 1.5$	$22.1^{d} \pm 1.6$	$15.4^{e} \pm 1.0$	$14.3^{e} \pm 0.9$	$15.0^{e} \pm 1.3$	$15.0^{e} \pm 1.2$
C18:3n3 (linolenic acid)	$17.6^{b} \pm 0.9$	$20.3^{a} \pm 1.1$	$21.4^{a} \pm 1.6$	$5.2^{c} \pm 0.5$	$3.0^{d} \pm 0.1$	$0.7^{e} \pm 0.0$	$0.3^{f} \pm 0.0$	$0.7^{e} \pm 0.0$	$0.6^{e} \pm 0.1$
Total	$61.8^{a} \pm 2.2$	$65.4^{a} \pm 2.3$	$57.4^{b} \pm 1.9$	$32.0^{c} \pm 1.6$	$25.1^{d} \pm 1.5$	$16.1^{e} \pm 2.1$	$14.6^{e} \pm 2.0$	$15.7^{e} \pm 1.6$	$15.6^{e} \pm 2.0$
SFA/PUFA	$0.5^{a} \pm 0.1$	$0.5^{a} \pm 0.1$	$0.6^{a} \pm 0.1$	$0.5^{a} \pm 0.1$	$0.4^{ab} \pm 0.1$	$0.4^{ab} \pm 0.0$	$0.3^{b} \pm 0.0$	$0.3^{b} \pm 0.0$	$0.4^{ab} \pm 0.0$
Water content (%)	$95.3^{a} \pm 3.4$	$90.3^{a} \pm 4.5$	$80.6^{b} \pm 3.9$	$74.8^{b} \pm 3.4$	$74.0^{b} \pm 5.6$	$63.9^{c} \pm 2.9$	$55.1^{d} \pm 2.8$	$30.3^{e} \pm 2.0$	$10.6^{f} \pm 1.8$
Oil yield (%)	$4.6^{i} \pm 0.2$	$8.4^{h} \pm 0.2$	$10.5^{g} \pm 0.3$	$12.8^{f} \pm 0.2$	$14.2^{e} \pm 0.3$	$18.6^{d} \pm 0.5$	$21.0^{c} \pm 0.4$	$22.8^{b} \pm 0.5$	$25.1^{a} \pm 0.4$

For each line (each fatty acid) means followed by the same letter are not significantly different based on Duncan's test at the 0.05 probability level.

mature coriander fruit [10,11]. This developmental trend in oil accumulation in coriander fruit was similar to that reported for other species [14,16].

Large accumulations of fatty acids began at 2 DAF. Different trends were observed for the fatty acids. Contents of saturated fatty acids were high from 2 until 10 DAF but declined by half by 12 DAF. This decline continued until full maturity. The representative fatty acid of this category in coriander was palmitic acid, which followed the same trend as the saturated fatty acids. Myristic and stearic acids were also present in higher amounts at 2 DAF and decreased after 10 DAF (Table 2). In contrast, monounsaturated fatty acids, represented mainly by petroselinic acid (50% at 2 DAF and reaching more than 92% at maturity) were present in low amounts at the beginning of seed formation (2 DAF) and rose after 10 DAF. Indeed, the petroselinic acid amount increased tenfold from 2 to 12 DAF and continued this rise, reaching its highest level at 18 DAF (Table 2). The polyunsaturated fatty acid content was highest at 2 DAF and decreased until 18 DAF, remaining stable from this point to maturity (Table 2). Higher levels of polyunsaturated and saturated fatty acids have been reported at earlier stages of fruit ripening [10,11]. The ratio of saturated to polyunsaturated fatty acids decreased markedly during fruit maturation. Similar results have been reported in other oilseed species [14,17]. The level of petroselinic acid was in accord with values previously reported for coriander ranging from 51.6% to 90.7% [5–7]. The highest amount of petroselinic acid was reached between 18 and 35 DAF, in agreement with results of Msaada et al. [10] which emphasized that a period of 32 DAF was sufficient for use of coriander fruits. In our study, opposite developmental trends were observed for palmitic acid (decreasing with fruit ripening) and petroselinic acid (increasing with fruit ripening). This observation could be explained by the role of palmitic acid as a precursor of petroselinic acid [18].

In summary, this study constitutes the first to investigate oil and fatty acid accumulation in coriander during fruit ripening under organic cultivation. Highest oil yield was achieved at full maturity. Fatty acid profiles varied greatly during fruit ripening. At earlier stages, saturated and polyunsaturated fatty acids were higher and decreased with fruit maturation. Petroselinic acid was the major fatty acid after 12 DAF, showing an inverse relationship with palmitic acid that supports a functional correlation between the two fatty acids. This study provided data for use of coriander oil and its composition of fatty acid, in particular petroselinic acid, for industrial applications.

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ORIGINAL PAPER

Extraction of Coriander Oil Using Twin-Screw Extrusion: Feasibility Study and Potential Press Cake Applications

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Abstract This study presents an assessment of the vegetable oil extraction from coriander fruits through mechanical pressing, more specifically twin-screw extrusion. This comprises an evaluation of the oil recovery obtained and its respective quality, as well as the specific mechanical energy, representing an economical point of view. With regard to the extrusion optimization, the screw configuration, the device's filling coefficient and the pressing temperature were varied. The screw configuration was shown to exhibit a key influence on the extraction efficiency and oil recoveries of at least 40 % were reached when the pressing zone was positioned immediately after the filter and consisted of 50 mm long, reverse screws with a -33 mm pitch. Furthermore, with a device's filling coefficient of 39.4 g/h rpm and a pressing temperature of 120 °C, an oil recovery of 47 %, the highest of this study, was reached with concurrent low energy consumption. Next to this, operating parameters of 47.1 g/h rpm and 80 °C resulted in the production of a press cake with the lowest residual oil content (15 %) in this study, although this also involved a significant increase in the filtrate's foot content. All the produced oils were of acceptable quality (<1.5 % acidity), showed high petroselinic acid content (73 %), and were pleasantly scented.

Keywords Twin-screw extruder · Coriander · Vegetable oil extraction · Oil quality · Press cake · Essential oil · Antioxidant capacity

The first two authors have both participated equally to this work. Therefore, they are both considered as principal authors.

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Introduction

Coriander (*Coriandrum sativum* L.) is an annual herb, native to the eastern Mediterranean and then spread to India, China and the rest of the world. It is commonly used as a condiment or a spice. The fruit has been used as a traditional medicine to treat various medical conditions such as indigestion, worms, rheumatism and joint pain. Indeed, coriander has been shown to exhibit a wide range of biological activities including alterative, antibilious, antispasmodic, aphrodisiac, appetite stimulant, aromatic, carminative, diaphoretic, diuretic, refrigerant, stimulant, stomachic and tonic [1]. The coriander fruits are particularly interesting as they contain both a vegetable oil and an essential oil fraction.

The composition of coriander fruits depends on several factors such as the growing region and maturity stages [2]. Previous research mainly focused on the fruit oil fatty acid composition, triacylglycerols and glycerophospholipids [3, 4], tocopherols and tocotrienols [5], or effects on plasma lipids [6]. The main fatty acid constituent in *Coriandrum sativum* vegetable oil is petroselinic (6Z-octadecenoic) acid, representing between 31 and 75 % of the fatty acid profile. Petroselinic acid is an uncommon isomer of oleic acid and is found in high levels in a restricted range of seed oils, mostly from the Apiaceae family [7]. It presents

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a unique oleochemical with high potential for food, cosmetic and pharmaceutical industries and may further allow the synthesis of a large number of interesting platform molecules.

Moreover, vegetable oil from coriander fruits has recently been labeled as a novel food ingredient (NFI) by the European Food Safety Authority [8]. It is now considered as safe to be used as a food supplement for healthy adults, at a maximum level of 600 mg per day (i.e., 8.6 mg/kg bw per day for a 70 kg person). This will lead to significantly higher intakes of coriander fruit oil and petroselinic acid than current background intakes. Therefore, the development of a new process for extracting vegetable oil from coriander fruits is a major challenge for the years to come.

The extraction of vegetable oil on a commercial scale is usually achieved through the application of a two-step process. As a first step, seeds with considerable oil content are pre-pressed by the use of a hydraulic or expeller press, while the second step comprises solvent extraction of the press cake to obtain sufficiently low residual oil contents of typically less than 1 %. However, the continuous extraction of vegetable oil using extrusion technology as a sole method is becoming of increasing importance [9–13]. This commonly involves a single-screw extruder of variable pitch and channel depth, rotating in a cage type barrel [9]. As friction between the material and the screw or barrel surface is a key factor during single-screw extrusion, this process often leads to high energy consumption, overheating and subsequent oil deterioration. Apart from this, complementary special equipment such as breaker bars is often necessary in order to avoid insufficient mixing.

Twin-screw extrusion could provide a solution to these issues, as it exerts a stronger transportation force and enhances mixing and crushing of the seeds during extrusion. In addition, twin-screw extruders have been shown to be significantly more energy efficient [9, 14]. Recently, twin-screw extrusion has been increasingly applied to achieve efficient vegetable oil extraction from various oil seeds [9, 14–23]. Therefore, it was applied for the extraction of vegetable oil from coriander fruits, while aiming to obtain a high level of oil quality, extraction efficiency and feasibility.

Only one previous study has dealt with the use of twin-screw extrusion technology for vegetable oil extraction from coriander fruits by mechanical pressing [24]. The single batch of fruits used in this study was cultivated in the Korba area (North East of Tunisia) and exhibited a relatively low lipid content (only 21.9 % of the dry matter). Both the screw rotation speed and the inlet flow rate of coriander fruits affected the oil extraction. The highest oil recovery was obtained under operating conditions of 50 rpm and 2.3 kg/h, respectively. Nevertheless, it was never more than 45 % and the residual oil content in the press cakes was at least 16.6 % of the dry matter. At the same time, the filtrate's foot content, i.e., the solid particles forced through the filter, was always high (from 47.5 to 66.0 %). Further, essential oil contents in the press cakes and their composition have not been the subject of any earlier studies although they may present a valuable application of extrusion cakes. The impact of operating conditions on the fatty acid composition of pressed oils was less important. Ten fatty acids were identified, with petroselinic acid accounting for 66–75 %. In conclusion, the use of twin-screw extrusion technology for the mechanical pressing of coriander oil appears promising, even if the process efficiency should be improved.

This study comprises an evaluation of the impact of several operating conditions on the twin-screw extrusion process. These include the screw configuration, the device's filling coefficient, which represents the ratio of the inlet flow rate of coriander fruits to the screw rotation speed, and the pressing temperature. Further, some potential applications for the press cakes obtained are suggested.

Experimental Procedures

Materials

All trials were carried out using a single batch (200 kg) of coriander fruits (GSN maintenaire variety), cultivated in the South West part of France and supplied by GSN Semences (Le Houga, France). The moisture content of the coriander fruits was 9.77 ± 0.10 % (French standard NF V 03–903) [25]. All solvents and chemicals were of analytical grade and were obtained from Merck (Germany), Macherey–Nagel (Germany), Sigma-Aldrich (USA) and Prolabo (France).

Twin-Screw Extruder

Twin-screw extrusion was carried out by the use of a Clextral BC 21 (France) co-rotating and co-penetrating twin-screw extruder, comprising two identical, intermeshing screws. The extruder was composed of seven modular barrels, each 100 mm long (Fig. 1), while the screws consisted of several segmental screw elements that were either 25 or 50 mm long. The temperature of modules 2, 3, 4, 5 and 7 was controlled and adjusted through electrical heating and water cooling. Coriander fruits were fed into the extruder inlet port using a volumetric screw feeder (K-Tron Soder KCL-KT20, Switzerland). Through the application of different screw types, different extrusion zones may be created which subject the fruits to a series of operations. In module 1, the forward pitch screws exert a conveying force, while a succession of 10 monolobe and 10 bilobe paddles

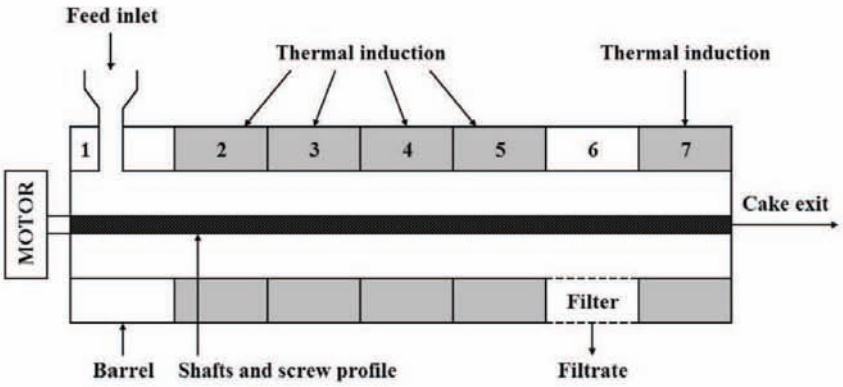


Fig. 1 Schematic modular barrel of the Clextral BC 21 twin-screw extruder used for extraction of vegetable oil from coriander fruits

	1		2			3		4		5		6			7		
Profile 1	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -25	C2F 25	C2F 33
Profile 2	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -25		C2F 33
Profile 3	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -33		C2F 33
Profile 4	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -33	C2F 25	C2F 33

Fig. 2 Screw configurations for extraction of vegetable oil from coriander fruits. *T2F* trapezoidal double-thread screw; *C2F* conveying double-thread screw; *DM* monolobe paddle-screw; *BB* bilobe paddle-screw; *CF2C* reverse screw. The numbers following the type of *screw* indicate the pitch of T2F, C2F, and CF2C screws and the length of the DM and BB screws

ensures thorough trituration in modules 2–4. The fruits are further conveyed in modules 5 and 6, while module 7 represents a pressing zone made up of reverse pitch screws. The filtrate was collected at the filter section in module 6, which is composed of four hemispherical dishes with perforations of 500 µm diameter. A control panel allowed the continuous monitoring of the screw rotation speed (S_S), the fruit feed rate (Q_S) and the barrel temperature (θ_c).

The screw configurations that were applied during this study (Fig. 2) were based upon those used in Evon *et al.* [22] for jatropha oil extraction. Variations concerned the pressing zone of the extruder, situated in module 7. The reverse pitch screws used in profiles 1 and 2 were 50 mm long, with a pitch of −25 mm. They were positioned immediately after the filtration module for profile 1, and 25 mm from the end of module 6 for profile 2. The reverse pitch screws used in profiles 3 and 4 had the same length (50 mm), but their pitch was greater (−33 mm instead of −25 mm). They were positioned 25 mm from the end of module 6 for profile 3 and immediately after the filtration module for profile 4.

Experimental

Ten trials were conducted for the extraction of vegetable oil from coriander fruits in the twin-screw extruder (Table 1). Different operating conditions were tested including the screw profile, the device's filling coefficient and the temperature in the pressing zone. Except for trial 4, the screw rotation speed was 100 rpm for all the experiments, meaning that the device's filling coefficient directly depended on the feed flow of coriander fruits.

In order to ensure stabilization of the extruder and the operating conditions, extrusion was carried out for 30 min prior to sampling. Then, samples consisting of the filtrate and the press cake were collected for a sufficient period of time (30 min) to minimize any variation of the outlet flow rates and to allow a single sampling for each trial. After collection of the filtrate during extrusion, it was subjected to centrifugation (8000 ×*g*, 15 min, 20 °C) in order to eliminate the solid residue, i.e., the centrifugation foot.

The oil recoveries were calculated from the following formulas:

$$R_L = \frac{Q_F \times T_L}{Q_S \times L_S} \times 100, \quad (1)$$

where R_L is the oil recovery relative to the total oil content of the fruits (%), Q_S the inlet flow rate of coriander fruits (kg/h), Q_F the flow rate of the filtrate (kg/h), T_L the mass fraction of oil in the filtrate (%), and L_S the total oil content of the coriander fruits (%).

$$R_C = \frac{(Q_S \times L_S) - (Q_C \times L_C)}{Q_S \times L_S} \times 100, \quad (2)$$

where R_C is the oil recovery based on the residual oil content of the press cake (%), Q_C the flow rate of the press cake (kg/h), and L_C the oil content of the press cake (%).

Although both R_L and R_C are expressed in terms of the oil content of the fruit, R_C is always higher than R_L because it includes all oil present in the filtrate (pressed oil and oil contained within the foot).

The energy consumed by the motor was determined from the following formulas:

$$P = P_M \times \frac{S_S}{S_{\max}} \times \frac{T}{T_{\max}} \times \cos\varphi, \quad (3)$$

where P is the electrical power supplied by the motor (W), P_M the motor's power rating ($P_M = 8300$ W), T and T_{max} the test torque and maximum torque (100 %) of the extruder motor (%), cos φ its theoretical yield (cos $\varphi = 0.90$), and S_S and S_{max} the test speed and maximum speed (682 rpm) of the rotating screws (rpm), respectively.

$$SME = \frac{P}{Q_S}, \quad (4)$$

where SME is the specific mechanical energy consumed by the motor per unit weight of coriander fruits (Wh/kg).

$$SME' = \frac{P}{Q_F \times T_L}, \quad (5)$$

where SME' is the specific mechanical energy consumed by the motor per unit weight of pressed oil (Wh/kg).

Analytical Methods

Analyses of the fruits or the press cakes obtained were carried out according to the following French standards: moisture contents NF V 03–903 [25]; mineral contents NF V 03–322 [26]; oil contents NF V 03–908 [27]; protein contents NF V 18–100 [28]. Determination of the fruit's oil content and the residual oil content of press cakes was performed by means of a two-step Soxhlet extraction, each of 5 h duration, with *n*-hexane as the extracting solvent. The material was further milled between both steps by the use of a Foss Cyclotec 1093 (Denmark) mill fitted with a 300-µm screen. An estimation of the three parietal constituents (cellulose, hemicelluloses, and lignins) of the coriander fruits was made using the ADF-NDF method from Van Soest and Wine [29, 30]. An estimation of the water-soluble components contained in the coriander fruits was made through measurements of the mass reduction of the test sample after 1 h in boiling water. All determinations were carried out in duplicate.

Oil Quality Analysis

In order to assess the quality of the Soxhlet extracted oil (i.e., the oil extracted from coriander fruits using the Soxhlet extraction apparatus and *n*-hexane as the extracting solvent) and the pressed oils, their free fatty acid content was determined through two parameters: the acid value and the acidity (based on petroselinic acid), expressed in mg of KOH/g of oil and in %, respectively (French standard NF T 60–204) [31]. All determinations were carried out in duplicate.

Glyceride Profile

The glyceride profile of the Soxhlet extracted oil was determined through gas chromatography (GC). Samples of about 0.10–0.15 g were weighed and 0.5 mL of BSTFA (*N*,*O*-bis(trimethylsilyl)trifluoroacetamide) with a derivatizing agent, i.e., trimethylchlorosilane, was added. Two internal standards were added (0.5 mL). The first one was composed of 8.068 mg/mL betulin in pyridine, while the

Table 1 Results of the pressing experiments conducted with the Clextral BC 21 twin-screw extruder

Trial number	1	2	3	4	5	6	7	8	9	10
Operating conditions										
Screw profile	3	3	3	3	4	4	4	4	4	4
S_S (rpm)	100	100	100	133	100	100	100	100	100	100
Q_S (kg/h)	3.12	4.66	6.19	6.19	3.15	3.94	4.71	4.71	4.71	4.71
C_F (g/h rpm)	31.2	46.6	61.9	46.5	31.5	39.4	47.1	47.1	47.1	47.1
θ_{c6} (°C)	83.0 ± 0.0^{e}	79.4 ± 0.8^{f}	77.7 ± 0.6^{g}	77.9 ± 0.3^{g}	95.2 ± 0.8^{a}	90.6 ± 0.6^{b}	89.1 ± 0.3^{c}	84.1 ± 0.9^{d}	82.6 ± 0.8^{e}	76.1 ± 1.1^{h}
θ_{c7} (°C)	120 (121.0 ± 0.6^{a})	120 (119.0 ± 0.6^{cd})	120 (118.7 ± 0.5^{d})	120 (120.4 ± 0.8^{b})	120 (120.1 ± 0.7^{b})	120 (118.1 ± 1.1^{e})	120 (119.5 ± 0.7^{c})	100 (101.0 ± 1.1^{f})	80 (79.7 ± 0.9^{g})	65 (64.8 ± 0.5^{h})
Filtrate										
Q_F (kg/h)	0.31	0.51	0.49	0.70	0.38	0.52	0.59	0.59	0.59	0.59
T_L (%)	82.8	87.3	88.3	83.5	83.6	89.3	91.5	91.7	82.1	84.1
T_F (%)	17.2	12.7	11.7	16.5	16.4	10.7	8.5	8.3	17.9	15.9
H_F (%)	n.d.	n.d.	n.d.	n.d.	3.32 ± 0.05^{f}	3.60 ± 0.01^{e}	3.74 ± 0.07^{d}	4.59 ± 0.13^{c}	4.99 ± 0.11^{b}	5.78 ± 0.13^{a}
L_F (% of dry matter)	n.d.	n.d.	n.d.	n.d.	59.33 ± 0.86^{e}	63.34 ± 0.77^{d}	73.48 ± 0.33^{b}	76.21 ± 0.12^{a}	68.74 ± 0.95^{c}	68.90 ± 0.04^{c}
M_F (% of dry matter)	n.d.	n.d.	n.d.	n.d.	3.80 ± 0.01^{a}	3.35 ± 0.05^{b}	2.71 ± 0.13^{cd}	2.61 ± 0.04^{d}	2.78 ± 0.01^{c}	2.71 ± 0.04^{cd}
Press cake										
Q_C (kg/h)	2.58	3.84	5.08	5.07	2.61	3.31	3.92	4.00	3.97	4.00
H_C (%)	2.16 ± 0.02^{g}	3.85 ± 0.07^{f}	4.60 ± 0.04^{d}	4.46 ± 0.06^{d}	2.32 ± 0.14^{g}	3.73 ± 0.08^{f}	4.05 ± 0.06^{e}	4.88 ± 0.22^{c}	8.62 ± 0.27^{b}	8.99 ± 0.19^{a}
L_C (% of dry matter)	17.63 ± 0.07^{b}	17.06 ± 0.05^{c}	17.62 ± 0.03^{b}	18.10 ± 0.08^{a}	16.48 ± 0.21^{e}	16.82 ± 0.29^{d}	16.06 ± 0.08^{f}	16.46 ± 0.11^{e}	14.99 ± 0.05^{g}	15.23 ± 0.14^{g}
M_C (% of dry matter)	6.03 ± 0.01^{c}	6.10 ± 0.01^{b}	6.09 ± 0.03^{b}	5.98 ± 0.04^{c}	6.15 ± 0.03^{b}	6.15 ± 0.01^{b}	6.29 ± 0.01^{a}	6.11 ± 0.02^{b}	6.26 ± 0.09^{a}	6.17 ± 0.08^{b}
Oil recoveries (%)										
R_L	32.5	38.2	28.0	37.8	40.5	46.9	45.8	46.3	41.1	41.8
R_C	42.9	45.9	44.7	43.3	46.7	45.6	48.7	46.7	53.7	52.8
Energy consumed										
T (%)	16.4 ± 2.2^{i}	42.7 ± 1.6^{e}	57.2 ± 1.6^{a}	32.5 ± 2.1^{g}	25.0 ± 1.7^{h}	37.2 ± 1.4^{f}	45.6 ± 1.5^{d}	47.7 ± 2.0^{c}	54.2 ± 2.2^{b}	54.9 ± 1.3^{b}
P (W)	180.0 ± 23.8^{h}	467.9 ± 17.3^{e}	626.7 ± 17.8^{a}	473.1 ± 30.3^{e}	274.2 ± 18.8^{g}	407.8 ± 15.6^{f}	499.0 ± 16.1^{d}	522.9 ± 21.7^{c}	594.1 ± 23.9^{b}	601.0 ± 14.4^{b}
SME (Wh/kg fruit processed)	57.7 ± 7.6^{g}	100.4 ± 3.7^{f}	101.3 ± 2.9^{g}	76.5 ± 4.9^{f}	86.9 ± 6.0^{g}	103.5 ± 4.0^{e,d}	105.9 ± 3.4^{c}	111.0 ± 4.6^{b}	126.1 ± 5.1^{a}	127.6 ± 3.0^{a}
SME* (Wh/kg pressed oil)	710.2 ± 93.8^{h}	1053.8 ± 39.0^{g}	1447.2 ± 41.1^{a}	810.4 ± 51.9^{g}	859.1 ± 58.9^{f}	884.1 ± 33.9^{e,f}	925.6 ± 29.9^{d,e}	960.1 ± 39.9^{d}	1228.9 ± 49.5^{b}	1221.4 ± 29.2^{b}

C_F is the device's filling coefficient (g/h rpm); it is defined as the ratio of the inlet flow rate of coriander fruits (Q_S) to the screw rotation speed (S_S). θ_{c6} is the barrel temperature measured during sampling at the level of module 6, i.e., the filtration module (°C). θ_{c7} is the barrel temperature at the level of module 7 (set value first mentioned, plus temperature measured during sampling in parentheses), i.e., the pressing zone (°C). Modules 2-5 were heated to 65 °C for all trials. T_F is the mass content of the foot in the filtrate (%). H_F is the moisture content in the foot of the filtrate (%). L_F is the oil content in the foot of the filtrate (% of the dry matter). M_F is the mineral content in the foot of the filtrate (% of the dry matter). H_C is the moisture content in the press cake (%). M_C is the mineral content in the press cake (% of the dry matter). Means in the same line with the same superscript letter (a–i) are not significantly different at $P < 0.05$. *n.d.* non determined

second one was a 8.087 mg/mL tricaprin solution. Next, samples were heated to 80 °C for 30 min.

They were then analyzed through GC using an Agilent Technologies 7890A (USA) gas chromatograph. The different compounds were separated in a J&W DB-5HT GC (Agilent, USA) column (15 m, 0.32 mm i.d., 0.10 µm film thickness) under the following conditions: oven temperature: 50 °C; 50–200 °C (15 °C/min); 200–290 °C (3 °C/min, held 10 min); 290–360 °C (10 °C/min, held for 15 min); flame ionization detector (FID) 380 °C; carrier gas helium (66 kPa).

Fatty Acid Composition

Determination of the fatty acid composition of the oils obtained was performed by gas chromatography (GC). For this, oil samples were dissolved in *tert*-butyl methyl ether (TBME) to a concentration of 20 mg/mL. Next, 100-µL aliquots of these solutions were converted to methyl esters by the addition of 50 µL of a 0.2 mol/L trimethylsulfonium hydroxide (TMSH) in methanol solution (French standard NF ISO 5508) [32].

The resulting fatty acid methyl esters were subjected to GC analysis by the use of a Varian 3800 (USA) gas chromatograph equipped with a flame ionization detector. Separation of the methyl esters was achieved in a CP Select CB (Varian, USA) fused silica capillary column (50 m, 0.25 mm i.d., 0.25 µm film thickness). The initial oven temperature was held at 185 °C for 40 min, after which it was increased to 250 °C at a rate of 15 °C/min and maintained at 250 °C for 10 min. The temperature of the injector and the detector was kept at 250 °C. Helium was used as the carrier gas with a flow rate of 1.2 mL/min. All determinations were carried out in triplicate.

Essential Oil Content

Hydrodistillation (or water distillation) was applied to extract the essential oil from the coriander material, i.e., the fruits or the press cakes. 200 g of milled material was mixed with 2 L of water (1:10 ratio) and placed in a distillation flask where it was distilled for 5 h. The installation consisted of a Clevenger-like apparatus. Volatilized compounds and water vapor were condensed through a cooling system, collected, and separated in a separatory tube. All determinations were carried out in triplicate.

Essential Oil Composition

The composition of essential oils extracted from the coriander fruits and the press cakes was determined by gas chromatography (GC). The essential oils were analyzed using a HP 5890 Series II (USA) gas chromatograph equipped with a flame ionization detector. The carrier gas was helium with a constant pressure of 15 psi. Compounds of the essential oils were separated in an Agilent VF-5 ms (USA) apolar column (30 m, 0.25 mm i.d., 1 µm film thickness). The injected volume was 0.5 µL. The initial oven temperature was 110 °C and increased at a rate of 7 °C/min to 220 °C. The injector and detector temperatures were 220 °C. Identification of the compounds was based on a comparison of their retention indices relative to a series of *n*-alkanes (C_9–C_{15}) with those of literature (NIST). All determinations were carried out in triplicate.

Particle Size Distribution of the Press Cakes

The particle size distribution of press cakes was determined by optical microscopy. For this, a Nachet France Z 45 P (France) × 15 binocular magnifier was used and five different photographs were taken of each sample by the use of the Archimed 4.0 (France) software. Next, the particle size distribution was constructed through manual measurement of the diameter of all particles on the five photographs by the use of the ImageJ (USA) software.

Antioxidant Capacity of the Press Cakes

The antioxidant capacity of the press cakes was measured by means of the DPPH radical scavenging assay, through which the radical scavenging activity of an extract against the stable DPPH (2,2-diphenyl-1-picrylhydrazyl) radical was determined. The applied method was based on the one used by Brand-Williams *et al.* [33] and comprises methanolic extraction and DPPH scavenging assessment through UV spectrophotometry. Methanolic extracts were obtained by methanol Soxhlet extraction for 5 h and subsequent concentration by rotary evaporation. Aqueous extracts were prepared through rotary evaporation of water solutions. Inhibition (INH, %) was calculated from the following formula:

$$INH = \frac{AB - AA}{AB} \times 100 \quad (6)$$

where AB and AA are the absorbances of the DPPH solution and the tested extract solution, respectively.

A 6×10^{-5} M DPPH in methanol solution was prepared daily, protected from the light and stored at low temperatures (i.e., at about 5 °C). A 10-mg sample of the extract was dissolved in 1 mL of methanol or a 1/1 water/methanol solution for aqueous extracts. The samples were subjected to sonication to ensure complete dissolution. Then, 50 µL of this extract solution was added to 2 mL of DPPH solution, and the samples were put in the dark for 30 min to react. Absorption was measured with UV spectrophotometry at 515 nm. Further, a calibration curve was set up using

methanolic Trolox solutions with known concentrations ranging from 100 to 750 µmol/L. Results were expressed as µmol of Trolox equivalents (TE) per g of press cake. All determinations were carried out in triplicate.

Statistical Analyses

All determinations were conducted in duplicate or triplicate, and data are expressed as means ±standard deviations. The means were compared by the use of a single-factor analysis of variance (ANOVA) using the GLM procedure of the SAS data analysis software. The comparison between the different individual means was performed using the Duncan's multiple range test at a 5 % probability level.

Results and Discussion

Chemical Composition of the Coriander Fruits

The coriander fruits that were utilized during this study were of French origin and were shown to contain 27.67 ± 0.57 % vegetable oil on a dry basis. This value is in accordance with the oil contents of 9.9–28.4 % reported in the literature [3, 4, 24, 34–36]. It was higher than the oil content of the raw material used in the only antecedent study that also dealt with the extraction of coriander oil by twin-screw extrusion, which was 21.9 % of the dry matter for Tunisian coriander [24]. These variations can be attributed not only to the difference in variety but also to some factors such as cultivation conditions, especially the use of fertilizers and irrigation [37]. Mineral and protein contents were 5.86 ± 0.11 % of the dry matter and 14.07 ± 0.43 % of the dry matter, respectively. The three parietal constituents (cellulose, hemicelluloses, and lignins) constituted 24.38 ± 0.55 % of the dry matter, 17.84 ± 0.56 % of the dry matter and 11.01 ± 0.25 % of the dry matter, respectively. Lastly, the water-soluble components represented 14.32 ± 0.09 % of the dry matter in the coriander fruits.

The fatty acid composition of the solvent extracted oil obtained from coriander fruits is shown in Table 2. Petroselinic acid was found to be the major fatty acid, representing 72.6 % of all fatty acids, while significant amounts of linoleic (13.8 %) and oleic acid (6.0 %) were also detected. This is consistent with earlier reports on the fatty

Table 2 Quality and fatty acid composition of Soxhlet extracted oil, and pressed oils from coriander fruits (trials 2, 4, 7, 8 and 10)

	Soxhlet extracted oil	Trial number 2	Trial number 4	Trial number 7	Trial number 8	Trial number 10
Oil quality						
Acid value (mg of KOH/g of oil)	3.58 ± 0.10^{a}	$2.90 \pm 0.08^{b,c}$	$2.92 \pm 0.02^{b,c}$	2.80 ± 0.14^{c}	$2.90 \pm 0.02^{b,c}$	3.02 ± 0.16^{b}
Acidity (% FFA as petroselinic acid)	1.80 ± 0.05^{a}	$1.46 \pm 0.04^{b,c}$	$1.47 \pm 0.01^{b,c}$	1.41 ± 0.07^{c}	$1.46 \pm 0.01^{b,c}$	1.52 ± 0.08^{b}
Fatty acid composition (%)						
Caproic acid (C6:0)	0.1 ± 0.1	n.dc.	n.dc.	n.dc.	n.dc.	n.dc.
Myristic acid (C14:0)	n.dc.	0.2 ± 0.0^{a}	0.2 ± 0.0^{a}	0.1 ± 0.0^{a}	0.2 ± 0.0^{a}	0.2 ± 0.0^{a}
Palmitic acid (C16:0)	2.9 ± 0.1^{b}	3.3 ± 0.0^{a}	3.3 ± 0.0^{a}	3.3 ± 0.0^{a}	3.3 ± 0.0^{a}	3.3 ± 0.0^{a}
Palmitoleic acid (C16:1)	0.4 ± 0.1^{a}	0.2 ± 0.0^{b}	0.2 ± 0.0^{b}	0.2 ± 0.0^{b}	0.2 ± 0.0^{b}	0.2 ± 0.0^{b}
Margaric acid (C17:0)	<0.1	n.dc.	n.dc.	n.dc.	n.dc.	n.dc.
Stearic acid (C18:0)	<0.1	0.7 ± 0.0^{a}	0.7 ± 0.0^{a}	0.7 ± 0.0^{a}	0.7 ± 0.0^{a}	0.7 ± 0.0^{a}
Petroselinic acid (C18:1n-12)	72.6 ± 0.1^{c}	72.8 ± 0.2^{c}	$72.9 \pm 0.2^{b,c}$	73.4 ± 0.2^{a}	$73.3 \pm 0.1^{a,b}$	$73.3 \pm 0.2^{a,b}$
Oleic acid (C18:1n-9)	6.0 ± 0.2^{a}	5.5 ± 0.3^{b}	$5.4 \pm 0.1^{b,c}$	4.9 ± 0.2^{d}	$5.2 \pm 0.1^{b,c,d}$	$5.0 \pm 0.2^{c,d}$
cis-Vaccenic acid (C18:1n-7)	1.2 ± 0.1^{b}	1.3 ± 0.0^{a}	1.3 ± 0.0^{a}	1.3 ± 0.0^{a}	1.3 ± 0.0^{a}	1.3 ± 0.0^{a}
Linoleic acid (C18:2)	13.8 ± 0.1^{b}	13.8 ± 0.1^{b}	13.8 ± 0.0^{b}	13.8 ± 0.0^{b}	13.8 ± 0.0^{b}	13.9 ± 0.0^{a}
Linolenic acid (C18:3)	0.2 ± 0.1^{a}	0.1 ± 0.0^{b}	0.1 ± 0.0^{b}	0.1 ± 0.0^{b}	0.1 ± 0.0^{b}	0.1 ± 0.0^{b}
Arachidic acid (C20:0)	0.1 ± 0.0^{a}	0.1 ± 0.0^{a}	0.1 ± 0.0^{a}	0.1 ± 0.0^{a}	0.1 ± 0.0^{a}	0.1 ± 0.0^{a}
Gadoleic acid (C20:1)	0.2 ± 0.1	n.dc.	n.dc.	n.dc.	n.dc.	n.dc.
SFA	3.2 ± 0.0^{b}	4.2 ± 0.0^{a}	4.2 ± 0.0^{a}	4.2 ± 0.0^{a}	4.2 ± 0.0^{a}	4.2 ± 0.0^{a}
MUFA	80.4 ± 0.2^{a}	79.7 ± 0.2^{c}	79.8 ± 0.1^{c}	$79.9 \pm 0.0^{b,c}$	80.0 ± 0.0^{b}	$79.9 \pm 0.0^{b,c}$
PUFA	14.0 ± 0.1^{ab}	14.0 ± 0.1^{ab}	13.9 ± 0.0^{b}	14.0 ± 0.0^{ab}	13.9 ± 0.0^{ab}	14.0 ± 0.0^{a}
SFA/PUFA	0.2 ± 0.0^{b}	0.3 ± 0.0^{a}	0.3 ± 0.0^{a}	0.3 ± 0.0^{a}	0.3 ± 0.0^{a}	0.3 ± 0.0^{a}
Identified fatty acids	97.6 ± 0.1^{c}	97.9 ± 0.1^{b}	97.9 ± 0.1^{b}	98.0 ± 0.0^{ab}	98.1 ± 0.0^{a}	98.1 ± 0.0^{a}

SFA saturated fatty acids, *MUFA* monounsaturated fatty acids, *PUFA* polyunsaturated fatty acids. Means in the same line with the same superscript letter (a–d) are not significantly different at $P < 0.05$. n.dc. not detected

acid composition of coriander oil [3, 4, 7, 24]. In addition, the oil was shown to exhibit an acceptable free fatty acid (FFA) content of 1.8 ± 0.1 %.

The glyceride profile of Soxhlet extracted oil was determined through GC. The oil contained 96.25 ± 1.05 % of triglycerides (TAG), 1.03 ± 0.09 % of diglycerides (DAG), and 0.09 ± 0.02 % of monoglycerides (MAG). GC analysis also led to a second estimation of the free fatty acid content and the latter was slightly lower (1.02 ± 0.03 %) than that obtained by titration (French standard NF T 60-204), i.e., 1.8 %. The high amount of TAG and concurrent low FFA content demonstrated that little hydrolysis or enzymatic degradation had occurred in the Soxhlet extracted oil, indicating its good quality.

The coriander fruits displayed an essential oil content of 0.7 % of the dry matter through water distillation (Table 3), and this agrees with the 0.3–1.2 % results reported by Kirilan *et al.* [38]. Further, the essential oil was particularly rich in linalool (71.7 %), a monoterpene alcohol providing the coriander fruits with their characteristic lemony citrus flavor [39]. Such content was comparable to those (41-80 %) mentioned by Asgarpanah and Kazemivash [40] and Sahib *et al.* [41], depending on the variety and origin of coriander. Other important compounds were α-pinene and γ-terpinene, representing 5.6 % and 5.0 % of the essential oil, respectively.

Influence of the Operating Conditions on Oil Extraction Efficiency

In order to assess the impact of the extrusion operating conditions on oil extraction, different screw profiles were tested and the device's filling coefficient and pressing temperature were varied. The effect on the extraction efficiency was evaluated through the determination of two oil recoveries (R_L and R_C), the filtrate's foot content and the extruder's energy consumption. Twin-screw extrusion is capable of combining a crushing operation, located at the trituration zone, with a compressing action, performed by the reverse pitch screws. The former leads to a substantial reduction in the particle size of the material, resulting in a more efficient oil release, while the latter ensures solid/liquid separation through the formation of a counter pressure at the beginning of module 7. The effectiveness of twin-screw extrusion for the extraction of vegetable oil from oil seeds was formerly demonstrated for sunflower oil [16–21], jatropha oil [22] and neem oil [23].

In the first screw profile tested (profile 1), the CF2C reverse pitch screws were 50 mm long, with a pitch of −25 mm, and they were positioned immediately after the filtration module (Fig. 2). With such a screw configuration and a 120 °C pressing temperature, a lot of solid particles were rapidly forced through the filter, preventing the oil from draining freely and thus its separation from a press cake. Then, it simply resulted in the clogging of the twin-screw extruder. This phenomenon persisted even when reducing the device's filling coefficient to less than 10 g/h rpm.

A second screw profile (profile 2) was therefore tested consisting of the same CF2C screws with a −25 mm pitch, but positioned 25 mm from the end of module 6 (Fig. 2). However, the same phenomenon, i.e., the clogging of the machine, was observed after only a few tens of seconds. As the CF2C screws with a −25 mm pitch used for screw profiles 1 and 2 were too restrictive, these reverse pitch screws were replaced in screw profiles 3 and 4 by CF2C screws with a higher, i.e., less restrictive, pitch (−33 mm instead of −25 mm).

For all experiments conducted with these last two screw profiles, solid/liquid separation at the filter section was effective and the oil content of the press cakes had decreased compared to the coriander fruits. This led to residual oil contents of 18.1–15.0 % of the dry matter and oil recoveries (R_C) of 42.9–53.7 %, depending on the operating conditions that were applied during extrusion (Table 1). The mineral content in the press cake was a little higher than in the actual coriander fruits due to the oil pressing, and it varied from 6.0 to 6.3 % of the dry matter

Table 3 Essential oil content in coriander fruits and in press cakes from trials 2, 4, 7, 8 and 10, and antioxidant capacity of press cakes 2, 4, 8 and 10 (before and after hydrodistillation)

	Coriander fruits	Press cake number 2	Press cake number 4	Press cake number 7	Press cake number 8	Press cake number 10
Essential oil content (% of the dry matter)	0.67 ± 0.10^a	0.14 ± 0.00^c	0.14 ± 0.01^c	0.14 ± 0.00^c	0.18 ± 0.01^c	0.31 ± 0.02^b
Antioxidant capacity (μmol of TE per g of press cake)						
Before hydrodistillation	n.d.	3.7 ± 0.2^c	4.2 ± 0.0^b	n.d.	5.1 ± 0.1^a	5.0 ± 0.2^a
After hydrodistillation	n.d.	2.4 ± 0.2^a	2.8 ± 0.3^a	n.d.	n.d.	n.d.

Means in the same line with the same superscript letter (a–c) are not significantly different at $P < 0.05$

n.d. Not determined

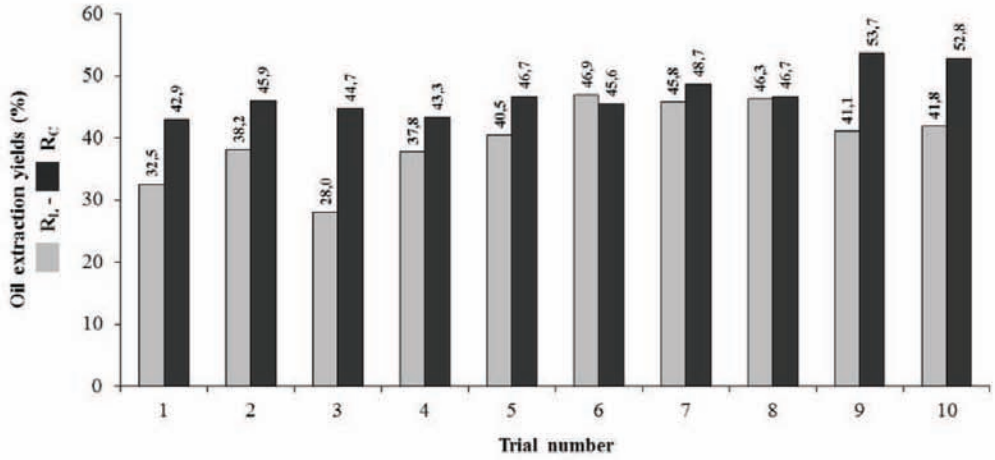


Fig. 3 Variation in oil recoveries for trials 1–10

(Table 1). In addition, for the two screw profiles tested successfully, the lower the oil content in the press cake, the higher its mineral content. The oil recovery (R_L), defined as the ratio of the pressed oil to the total oil contained within the fruit, varied from 28.0 to 46.9 % (Table 1). It was lower than the oil recovery (R_C) due to the oil retained in the filtrate foot (Fig. 3) and the difference between these two oil recoveries was minimal with the lowest filtrate foot contents (trials 6–8).

Moreover, the mass content of the foot in the filtrate was never more than 18 %, instead of 47–66 % for oil pressing from coriander fruits of Tunisian variety [24], meaning that the screw configurations used in this study were much more suited. Further reduction of the filtrate foot content could be accomplished through the application of a filter section with smaller perforations, but this was not possible during the course of this study. Low foot contents are important for the stability and the capacity of the extrusion process, as the centrifugation foot is commonly reintroduced at the inlet of the extruder, thus forming an additional feed flow.

For all screw profiles tested, the trituration zone was composed of a succession of 10 monolobe paddles and 10 bilobe paddles. It was the same as the one in the optimized screw profile used for the extraction of oil from jatropha seeds by mechanical pressing [22]. These elements ensure a profound reduction in the particle size of the material, leading to the rupture of the cell structures. In the third screw profile tested (profile 3), CF2C screws with a −33 mm pitch were still positioned 25 mm from the end of module 6 (Fig. 2). Four experiments were conducted with this profile (trials 1–4). However, when using screw profile 3, there was an accumulation of oil between the end of the filter section and the beginning of the pressing zone which could not drain freely. This presumably led to a decrease in the filtrate's flow rate as well as in the oil recovery (R_L). A fourth screw profile was therefore tested (profile 4), with the CF2C screws with a −33 mm pitch being positioned immediately after the filtration module (Fig. 2). This profile was applied for trials 5–10. Comparing trials 1 and 5, conducted with screw profiles 3 and 4, respectively, while maintaining the same operating conditions, screw profile 4 was found to be significantly more effective in oil extraction. It further resulted in a significantly lower residual oil content in the press cake and a slight reduction in the filtrate's foot content, leading to an increase in both oil recoveries (Table 1). However, the increase in the motor's torque resulted in a modest augmentation of the specific mechanical energy per unit weight of pressed oil (+21 %), which was shown to be significant through statistical analysis. In conclusion, screw profile 4 was considered as the optimized screw configuration for this study.

Next, the influence of the device's filling coefficient on oil extraction efficiency was assessed for screw profiles 3 and 4. When comparing trials 1–3, using the third screw profile, the increase in the inlet flow rate of coriander fruits from 3.12 kg/h (trial 1) to 6.19 kg/h (trial 3) directly affected the device's filling coefficient, increasing it from 31 to 62 g/h rpm (Table 1). Because it was more filled, the crushing ability of the trituration zone certainly diminished and the size reduction of solid particles became less significant, thus resulting in a decrease in the filtrate's foot content. Furthermore, the degree of filling of the CF2C screws increased as the device's filling coefficient increased, resulting in an enhanced pressure buildup inside the extruder and thus to an improved solid/liquid separation. As a result of this, the motor's torque significantly increased, resulting

in a higher specific mechanical energy per unit weight of pressed oil and a lower residual oil content of the obtained press cake, the latter being significantly different from the residual oil content of press cake from trial 1 according to the statistical analysis. Consequently, the oil recovery (R_C) increased with an increase in the device's filling coefficient. In addition, the oil recovery (R_L) showed an even more distinct increase due to the concurrent decrease of the filtrate's foot content. Similar findings were reported for the mechanical pressing of sunflower and jatropha oil [17, 18, 22].

For trial 3, the device's filling coefficient reached 62 g/h rpm. However, even though the motor's torque still increased, this did not result in a decrease in the press cake's residual oil content, the latter exhibiting a slight increase. With such a device's filling coefficient, the solid particles accumulated more upstream from the pressing zone, obstructing part of the filtering screens and thus reducing the filtration surface. This resulted in a decrease of the filtrate's flow rate, causing a significant reduction in the oil recovery (R_L) and an important increase in the specific mechanical energy. With screw profile 3, the machine became, in fact, too highly filled with a 62 g/h rpm device's filling coefficient, and higher filling coefficients were not tested to avoid clogging of the twin-screw extruder.

Comparing trials 2 and 4, the device's filling coefficient was the same (47 g/h rpm), but the screw rotation speed and the inlet flow rate of coriander fruits increased in the same proportions for trial 4: from 100 to 133 rpm and from 4.66 to 6.19 kg/h, respectively (Table 1). The increase in the screw rotation speed led to a more effective size reduction of the solid particles near the trituration zone, resulting in an increase in the foot content in the filtrate. Although the press cake's residual oil content was significantly higher from a statistical point of view, which is due to a less efficient compression of the mixture in the reverse screw elements, there was only a marginal reduction effect on both oil recoveries (R_L and R_C). Furthermore, the increase in the screw rotation speed significantly reduced the motor's torque, further reducing the specific mechanical energy per unit weight of pressed oil. In conclusion, a device's filling coefficient close to 50 g/h rpm was better suited for such a screw configuration, and higher productivity of the extruder resulted in the production of similar oil yields at a lower cost.

The effect of the device's filling coefficient on the oil extraction efficiency was further evaluated for the fourth screw profile through trials 5–7. As previously observed with screw profile 3 (trials 1–3), a decrease in the filtrate's foot content was observed with the increase of the device's filling coefficient, which is due to a lowering of the crushing ability of the trituration zone, thus leading to larger solid particles at the outlet of this zone. A significant increase in the motor's torque was observed at the same time due to the increase in the degree of filling of the CF2C screws and thus resulting in a better pressing action on the matter in this location. Comparing trials 5 and 6, this further led to an improvement in the liquid/solid separation, as illustrated by the relative increase in pressed oil quantity, resulting in an increased oil recovery (R_L) without any significant effect on specific mechanical energy per unit weight of pressed oil, as shown by the statistical analysis. Trial 6 was also the most efficient experiment of the entire study for oil extraction, exhibiting an oil recovery (R_L) of 46.9 % and producing a press cake with good quality (residual oil content of 16.8 % of the dry matter).

For a higher device's filling coefficient (47 g/h rpm, trial 7), the relative quantity of pressed oil slightly decreased compared to trial 6, illustrating the same phenomenon as that observed more significantly for trial 3 with the third screw profile, i.e., the obstruction of part of the filtering screens by an accumulation of solid particles. Despite a slightly excessive filling of the machine, the effect on the oil recovery (R_L) was quite limited and the corresponding specific mechanical energy only increased by 5 % compared to trial 6. At the same time, the press cake was of better quality with a significantly reduced residual oil content, resulting in an increase in the oil recovery (R_C). Therefore, analogous to what was found for screw profile 3, the 47 g/h rpm device's filling coefficient was considered to be a good compromise for screw profile 4 between oil extraction efficiency and the press cake's reduction in lipids.

As a second important operating parameter for twin-screw extrusion, the effect of the pressing temperature on the oil extraction efficiency was assessed for screw profile 4 using the optimized device's filling coefficient through trials 7–10. The applied pressing temperatures were 120, 100, 80 and 65 °C, respectively (Table 1). Contrary to what has been reported for sunflower and jatropha seeds [17, 18, 22], decreasing the pressing temperature from 120 to 65 °C did not substantially improve the oil extraction efficiency. A decrease in the pressing temperature of the extruder leads to a significant increase in the material viscosity, resulting in a profound impact on the residence time and energy consumption [42]. As the increased viscosity of the material flow causes a higher degree of filling of the extruder, the residence time and the energy input increased with decreasing pressing temperature. This led to an increase in both the motor's torque and the specific mechanical energy per unit weight of fruit processed, rendering the pressed oil slightly more expensive to produce. However, the oil extraction efficiency and filtrate's foot content at 100 °C (trial 8) were quite similar to that obtained at 120 °C (trial 7). At the same time, the press cake's oil contents were nearly comparable between trials 7 and 8, further resulting in similar oil recoveries (R_C).

Conversely, when the pressing temperature was only 80 °C (trial 9) or 65 °C (trial 10), the flow of material across the CF2C screws became much more viscous, as illustrated by the high values for the motor's torque (Table 1). This led to a better compression of the matter near the reverse pitch screws, and thus to a better reduction of the lipids in the press cakes. Moreover, press cakes from trials 9 and 10 revealed the two lowest residual oil contents of the entire study: 15.0 and 15.2 % of the dry matter, respectively, resulting in the two highest values for the oil recovery (R_C): 53.7 and 52.8 %, respectively. However, because solid particles accumulated more upstream from the pressing zone for these two lowest pressing temperatures, a significant increase in the foot content in the filtrate was also observed. Therefore, oil recoveries (R_L) slightly decreased compared to trials 7 and 8, further contributing to a significant increase (+30 % on average) in the specific mechanical energy.

In conclusion, the highest oil recovery (R_L) for this study was 46.9 %, obtained under the following operating conditions (trial 6): profile 4 screw configuration, 39.4 g/h rpm device's filling coefficient and 120 °C pressing temperature (Table 1). In addition, the residual oil content in the press cake was 16.8 % of the dry matter. The specific mechanical energy was 103 Wh/kg fruit processed or 884 Wh/kg pressed oil, which is quite low compared to other samplings in this study. It should however be noted that these values of energy consumption need further confirmation when twin-screw extrusion of coriander fruits is executed on a larger scale. The amount of foot in the filtrate was only 10.7 % under these optimal conditions.

The lowest press cake's residual oil content (15.0 % of the dry matter) was obtained under the following operating conditions (trial 9): profile 4 screw configuration, 47.1 g/h rpm device's filling coefficient and 80 °C pressing temperature (Table 1). The oil recovery (R_C) was then maximal (53.7 %). However, the amount of foot in the filtrate was much more important for these conditions (17.9 %). The oil recovery (R_L) was slightly lower than for the optimal conditions (41.1 % instead of 46.9 %). Moreover, the corresponding pressed oil was more expensive to produce (126 Wh/kg fruit processed or 1229 Wh/kg pressed oil). Reducing the diameter of perforations in the filter section should diminish the filtrate's foot content.

The abrupt shutdown of the extruder when all experiments were finished allowed the observation of material along the screw profile. From this, it could be seen that near modules 1–5, the trituration zone was significantly more filled than the conveying zones. Furthermore, the particle size of the material was found to be reduced substantially after passing through the monolobe and bilobe paddles, even if no accurate measurement of the particle size was carried out. Additionally, the pressing zone containing the reverse screw elements showed the highest filling due to its inherent pressing action on the material. And, as it was previously observed in the case of jatropha oil pressing [22], this led to an additional size reduction of the material due to the strong shearing force imposed by the reverse screws.

In summary, the extraction of vegetable oil from coriander fruits originating from France by mechanical pressing in a twin-screw extruder was improved compared to results obtained in the previous study, i.e., with fruits of Tunisian origin [24]. Indeed, even if the French variety was richer in vegetable oil than the Tunisian one (27.7 % of the dry matter instead of 21.9 %), the lowest residual oil content in the press cake (15.0 % of the dry matter) was lower than for the Tunisian variety (16.6 %). This means that the reduction in lipids was much more effective, leading to a better oil recovery (R_C): 54 % instead of only 45 % for the Tunisian variety. Furthermore, the filtrate's foot content was never more than 17.9 % due to the use of an optimized screw configuration, while it was at least 47.5 % in the previous study [24]. This considerably facilitates the pressed oil isolation, i.e., elimination of the foot from the filtrate by centrifugation.

Influence of the Operating Conditions on Oil Quality

The quality of pressed oils was examined through two parameters: the acidity and the petroselinic acid content (Table 2). For screw profile 3, the two pressed oils that were analyzed were those from the two most effective trials in terms of oil recovery (R_L), i.e., trial 2 and trial 4. For screw profile 4, this included the three pressed oils (trials 7, 8 and 10) that were produced with varying pressing temperatures: 120, 100 and 65 °C, respectively.

The oil acidity varied between 1.41 and 1.52 % for all pressed oils that were analyzed, corresponding to acid values between 2.80 and 3.02 mg of KOH/g of oil (Table 2). Thus, these virgin-type vegetable oils exhibit a higher quality than the Soxhlet extracted one (1.80 % acidity). Their acidity was slightly higher than that measured for pressed oils from the Tunisian variety of coriander fruits, which was situated between 1.54 and 2.21 mg of KOH/g of oil [24]. However, they were of excellent quality. Next to this, as revealed by the statistical analysis, it appeared that screw configuration had no significant effect on oil quality. Moreover, a decrease in the pressing temperature in the twin-screw extruder did not induce an oil quality improvement. This could be due to the fact that pressed oils did not properly come into contact with the pressing temperature, but rather with the temperature at the filter section (i.e., module 6). The latter did not exhibit high temperature variation between different trials, fluctuating between 76 and 89 °C for the pressed oils analyzed (Table 1).

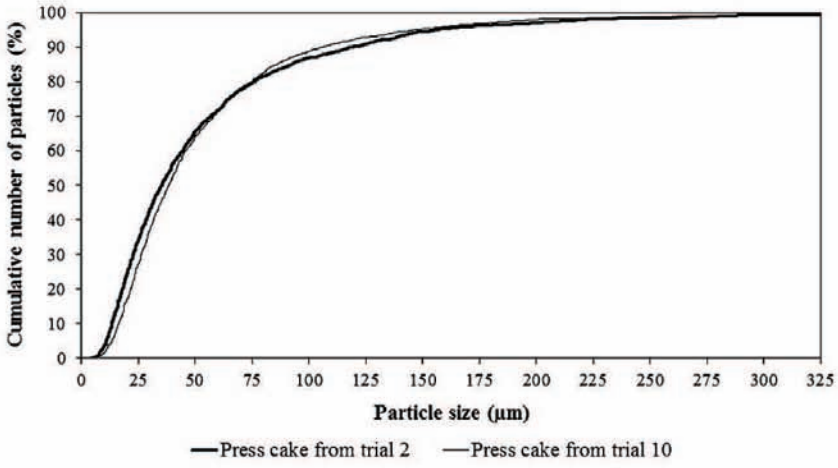


Fig. 4 Cumulative number of particles as a function of particle size for the press cakes from trials 2 and 10

The fatty acid composition of pressed oils was also shown to be nearly independent of the operating conditions used for twin-screw extrusion, i.e., screw configuration and pressing temperature (Table 2). Similar to the oil obtained through solvent extraction, all analyzed pressed oils were very rich in petroselinic acid, exhibiting contents between 72.8 and 73.4 %, while also similar contents of linoleic and oleic fatty acids were found. The minor differences between samples constituted a small variation in the petroselinic/oleic acid ratio. This possibly resulted from a complex integration of GC results, as retention times of both petroselinic and oleic acid were very similar. In conclusion, on the basis of the two quality criteria examined, it was clear that all analyzed pressed oils were high-quality vegetable oils.

Potential Press Cake Applications

In order to establish an economically favorable extrusion process, it is of key importance that extraction by-products such as the press cakes find some industrial applications. The ten press cakes were fine powders composed of almost spherical particles and their particle size distribution was estimated for press cakes from trials 2 and 10, each corresponding to the highest oil recovery (R_C) obtained for the two screw profiles used successfully (i.e., profiles 3 and 4, respectively). It appeared that the particle size distribution of press cakes was independent on the screw profile that was applied (Fig. 4). Indeed, the mean diameter of particles in press cakes from trials 2 and 10 was 35 µm and 38 µm, respectively.

Press cakes still contained part of the essential oil from coriander fruits. However, its content was very low for all operating conditions tested, varying from 0.14 to 0.31 % of the dry matter (Table 3). Further, the residual essential oil content in the press cake mainly depended on the pressing temperature applied in the twin-screw extruder, increasing with its decrease. A maximal essential oil content of 0.31 % was found inside the press cake from trial 10, corresponding to the lowest pressing temperature tested (i.e., 65 °C). Such temperature did not allow strong evaporation of the essential oil contained within the press cake. The residual essential oil content amounted in that case to 40 % of the essential oil in the starting material, leading to the conclusion that the remaining 60 % was co-extracted with the vegetable oil, rendering it pleasantly scented. The simultaneous extraction of both vegetable oil and essential oil through mechanical pressing is an interesting novelty of this research work. The residual essential oil in the press cakes could be extracted by means of hydrodistillation. Its composition was rather similar to that of the essential oil extracted from the fruits (Table 4), linalool being the main component. Linalool is one of the most commonly applied fragrance ingredients in cosmetics, perfumes, shampoos, soaps and even household detergents due to its fresh and flowery scent and it displays a worldwide use of over 1000 tons per year [43]. Further applications include its use as a repellent against mosquitoes [44]. However, because of the low quantities of volatile oil that could be extracted using hydrodistillation and the lack of a strong market for this essential oil, using the press cakes as raw materials to extract the remaining essential oil may seem too ambitious. Its application on an industrial scale would currently not be economical. It might therefore be more interesting to leave the volatile oil within other end products such as agromaterials, providing them with an added value.

Another possible use of the press cakes could be the isolation of some natural antioxidants through methanolic extraction, these being potentially interesting due to their beneficial impact on health. The antioxidant capacity of the

Table 4 Essential oil composition in coriander fruits and in press cakes from trials 2 and 4

	Coriander fruits	Press cake number 2	Press cake number 4
Essential oil composition (%)			
α-Pinene	5.6 ± 0.1^a	1.2 ± 0.2^b	1.2 ± 0.2^b
Camphene	0.8 ± 0.0^a	0.2 ± 0.0^b	0.2 ± 0.0^b
β-Pinene	1.1 ± 0.2^a	0.4 ± 0.0^b	0.4 ± 0.0^b
Myrcene	0.6 ± 0.0^a	0.2 ± 0.0^b	0.2 ± 0.0^b
p-Cymene	1.6 ± 0.2^a	0.8 ± 0.0^b	0.7 ± 0.1^b
Limonene	1.9 ± 0.2^a	0.9 ± 0.1^b	0.8 ± 0.1^b
γ-Terpinene	5.0 ± 0.5^a	3.3 ± 0.2^b	2.8 ± 0.3^b
Linalool	71.7 ± 1.4^b	77.0 ± 1.0^a	77.4 ± 1.1^a
Camphor	4.2 ± 0.1^b	4.8 ± 0.0^a	4.8 ± 0.1^a
Borneol	1.2 ± 0.0^b	1.6 ± 0.0^a	1.5 ± 0.1^a
α-Terpineol	0.4 ± 0.0^b	0.5 ± 0.0^a	0.5 ± 0.0^a
Carvone	n.dc.	0.2 ± 0.0^a	0.2 ± 0.0^a
Linalyl acetate	2.9 ± 0.2^b	4.1 ± 0.2^a	4.2 ± 0.3^a
2-(E)-decenal	n.dc.	n.dc.	0.1 ± 0.0
Geranyl acetate	3.0 ± 0.1^b	4.8 ± 0.3^a	4.9 ± 0.3^a

Means in the same line with the same superscript letter (a–b) are not significantly different at $P < 0.05$

n.d. Not detected

press cakes was determined through DPPH analysis and results varied from 3.7 to 5.1 µmol of TE per g of press cake (Table 3), though still representing rather low antioxidative activity. Press cakes from trials 8 and 10 revealed a better antioxidant capacity due to the decrease in pressing temperature, while for these two press cakes, the antioxidant capacities were shown not to be significantly different. The antioxidative activity of press cakes from trials 2 and 4 was also determined after they had further been subjected to hydrodistillation. This consistently led to a reduction in the antioxidant capacity (Table 3), possibly caused by the high temperatures applied through hydrodistillation for 5 h. However, this may also be a consequence of the migration of some water-soluble antioxidants such as anthocyanins to the hydrodistillation water phase.

The coriander press cakes obtained displayed significant residual oil contents of 15–18 % (Table 1), which may be unfavorable for some applications. However, this does not impose any disadvantages when the cakes are converted into usable energy through combustion, gasification or pyrolysis, possibly rendering the extrusion process self-supporting [45, 46]. In addition, the press cakes could be used as a renewable resource for the production of value-added agromaterials through thermo-pressing [21, 47–51]. Finally, they may be interesting for the biocomposite industry as they could be incorporated into biodegradable polymers such as polycaprolactone and polylactic acid where they could act as reinforcing fillers [52, 53].

Conclusion

The application of twin-screw extrusion for the extraction of vegetable oil from coriander fruits was shown to be effective and resulted in considerable oil recoveries while maintaining low foot contents. The extrusion operating conditions were found to have a significant impact on the oil recovery and energy consumption of the extruder. The highest extraction efficiencies were achieved with a pressing zone situated immediately after the filter and containing -33 mm reverse pitch screws that are 50 mm in length. Further, with a device's filling coefficient of 39.4 g/h rpm and a pressing temperature of 120 °C, an oil recovery of 47 % and a low foot content of 11 % were obtained. The press cake resulting from this trial displayed a residual oil content of 17 %, while a residual oil content of 15 % was reached with an increased device's filling coefficient (47.1 g/h rpm) and a decreased pressing temperature (80 °C). Here, however, the filtrate's foot content was considerable at 18 %, leading to a decrease in the oil recovery based on the filtrate. Because part of the essential oil is co-extracted by mechanical pressing with the vegetable oil, twin-screw extrusion of coriander fruits consistently produced an agreeably scented vegetable oil of good quality with less than 1.5 % free fatty acids and a high petroselinic acid content of 73 %. Potential applications of the press cakes obtained concern the essential oil content, which is rich in linalool, its antioxidant activity and the possible use as a biocomposite.

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RÉSUMÉ

RÉSUMÉ ÉTENDU DE LA THÈSE

I. Introduction

1.1 Contexte scientifique et technique

La coriandre (*Coriandrum sativum* L.) est une plante herbacée annuelle de la famille des Apiacées (Apiaceae), autochtone du bassin méditerranéen. Elle est principalement cultivée dans les régions tempérées du bassin méditerranéen, l'Inde, la Chine, la Thaïlande et l'Europe de l'Est. Elle est utilisée comme condiment à base de plantes pour de nombreuses préparations culinaires asiatiques et méditerranéennes. Cette plante est largement distribuée et principalement cultivée pour ses fruits qui sont utilisées à différentes fins telles que l'aromathérapie, l'alimentation, les médicaments, les cosmétiques et la parfumerie. Les fruits ont aussi des usages médicinaux, pour le traitement des rhumatismes et des douleurs articulaires, les troubles gastro-intestinaux, flatulences et gastralgie, les indigestions, l'insomnie, les convulsions, l'anxiété, la perte d'appétit, la glycémie ainsi que l'hypersomnie. L'intérêt de l'huile de graine de coriandre augmente depuis que l'Union Européenne a autorisé son utilisation comme complément alimentaire.

Les fruits de coriandre contiennent de l'huile végétale (VO) et de l'huile essentielle (EO). Ces deux fractions sont les plus importantes ressources naturelles durables pour la préparation de divers ingrédients alimentaires, additifs alimentaires, composants fonctionnels pour les nutraceutiques, les remèdes naturels, cosmétiques et autres applications. L'huile végétale contient de l'acide pétrosélinique, c'est un acide gras rare qu'on trouve principalement dans les fruits de coriandre. Ces composés représentent environ 70 à 80% des fractions d'huile et il peut être clivé par oxydation pour produire un mélange d'acide laurique, d'un composé utile dans la production de détergents, et d'acide adipique (un acide dicarboxylique en C6 qui est utilisé dans la synthèse de polymère de nylon). En outre, l'acide pétrosélinique est un isomère rare de l'acide oléique qui se trouve dans des niveaux élevés dans une gamme restreinte d'huiles de fruits, principalement de la famille des Apiacées. Il forme un produit oléochimique unique avec un potentiel élevé pour les industries alimentaires, cosmétiques et pharmaceutiques et peut permettre en outre la synthèse d'un grand nombre de molécules à plateforme intéressantes. L'huile essentielle contient du linalol (jusqu'à 60-70%), largement utilisé dans l'industrie des arômes tels que les cosmétiques, les crèmes, les lotions, les shampooings, les parfums, etc.

Aroma Tincto Oleo Crops (ATOC) est un concept de plantes contenant à la fois de l'huile végétale (VO) et de l'huile essentielle (EO). À l'heure actuelle, en fonction du secteur industriel concerné (huile ou aromatique), une seule de ces deux fractions est valorisée, l'autre étant considérés comme du déchet. Le développement d'une valorisation intégrée des ATOC permettant la co-extraction de VO et EO constitue les bases du concept de raffinage ATOC, càd un processus de valorisation séquentielle permettant la co-extraction de VO et EO tout en ne pénalisant pas la valorisation ultérieure des résidus par des produits tels que des molécules biosourcées (antioxydantes ou biocides) ou des substrats pour la conception d'agromatériaux odorants. L'approche technologique a été basée sur le couplage d'une extrudeuse (extrudeuses simples (mono-vis) ou à double vis (bi-vis) à un hydrodistillateur/extracteur pour d'abord la VO vierge à partir de fruit et EO à partir du tourteau extrudé, puis les antioxydants et les cocktails de molécules biocides à partir du tourteau distillé et enfin de valoriser le résidu solide ultime comme substrat soit pour la conception des agromatériaux (par thermopressage ou de moulage par injection) soit pour la production de bio-énergie (par la combustion de pellets formulés). Des expériences ont été menées avec une plante modèle sélectionnée couramment utilisé comme un condiment ou une épice dans les régions méditerranéennes et du Sud-Est asiatique: la coriandre (*Coriandrum sativum* L.). Les résultats obtenus en laboratoire et à l'échelle préindustrielle avec les fruits de coriandre mettent en évidence l'extraction de VO et d'EO avec des concentrations élevées (jusqu'à 75%) de composants de valeur (linalol et acide pétrosélinique). L'approche de la raffinerie a alors été étendue aux autres éléments de la plante, à savoir les feuilles et les racines, afin de valoriser le potentiel chimique global de la coriandre. Basé sur ce concept, la bioraffinerie a été développée dans cette thèse. Les matières végétales de coriandre ont été traitées en quatre parties: huile végétale, huile essentielle (ou extrait volatile), extrait méthanolique et résidu final (biomatériaux). De cette manière, les molécules dans les plantes peuvent être extraites séquentiellement et utilisées, évitant ainsi un grand gaspillage de ressources naturelles.

1.2 Objectifs de la thèse

Notre objectif principal était d'évaluer globalement le rendement, la composition de l'huile végétale et de l'huile essentielle, les propriétés antioxydantes de la coriandre. L'objectif était aussi d'évaluer la recherche initiale sur le traitement des déchets de la coriandre pour en faire des sources potentielles de matières premières en service industriel pour améliorer

l'efficacité de la production et du traitement de la coriandre et de ses produits. (i) Déterminer la composition de la VO et EO de différentes cultivars de coriandre (à partir de différentes origines géographiques). Ces cultivars ont été cultivées à l'état organique à Auch. (ii) Ensuite déterminer la composition de la VO et EO des différentes parties de la coriandre (fruits, feuilles, tige et les racines). (iii) Evaluer la bioaccumulation des VO et EO pendant la maturation des fruits de la coriandre. (iv) Déterminer les propriétés antioxydantes des extraits par différentes méthodes (DPPH, ORAC, FRAP) provenant de différentes parties de leurs résidus de distillation de cultivars de coriandre. (v) L'optimisation des conditions de fonctionnement de l'extrudeur mono-vis et bi-vis afin d'évaluer la faisabilité d'un traitement d'huile de fruits d'extraction. (vi) L'étude de faisabilité et l'influence des conditions de fonctionnement de ces machines à évaluer les caractéristiques du matériau de résidus finaux.

II. Bioaccumulation

II.1 Huiles végétales analyse

II.1.1 La teneur en huile et la composition des acides gras dans les différentes parties de la coriandre

Dans ces expériences, le cultivar coriandre français (FR1 - Dourou) a été choisi pour étudier la composition en acides gras et la teneur en huile des différentes parties de la coriandre (fruits, des feuilles et de la tige, racines). Le cultivar coriandre français a été cultivé le 25 Mars 2011 à l'état organique. Les échantillons prélevés sur les différentes parties de la coriandre (*Coriandrum sativum* L.) ont été analysés pour déterminer la teneur en acide gras et le rendement de l'huile. Les résultats des analyses montrent qu'il y a de grandes différences de teneur en huile entre les fruits, des feuilles et de la tige, les racines. La teneur en huile des fruits de coriandre est la plus élevée (jusqu'à 25%), suivie par la teneur en huile de la fleur (5,08%), la teneur en huile des feuilles et la tige (1,48%), et la teneur en huile des racines de coriandre (0,85%). Sur la base sur ce résultat, on distingue que les teneurs en huile de coriandre fleurs, feuilles et racines sont très faibles par rapport aux teneurs en huile des fruits et fruits de coriandre (Figure 1). Par conséquent, l'étude de l'huile végétale et huile essentielle des fleurs, les feuilles et les racines de coriandre n'est pas vraiment attrayant pour les agronomes. C'est la raison pourquoi notre étude est uniquement concentrée sur les fruits de coriandre en raison de leurs promesses d'avenir.

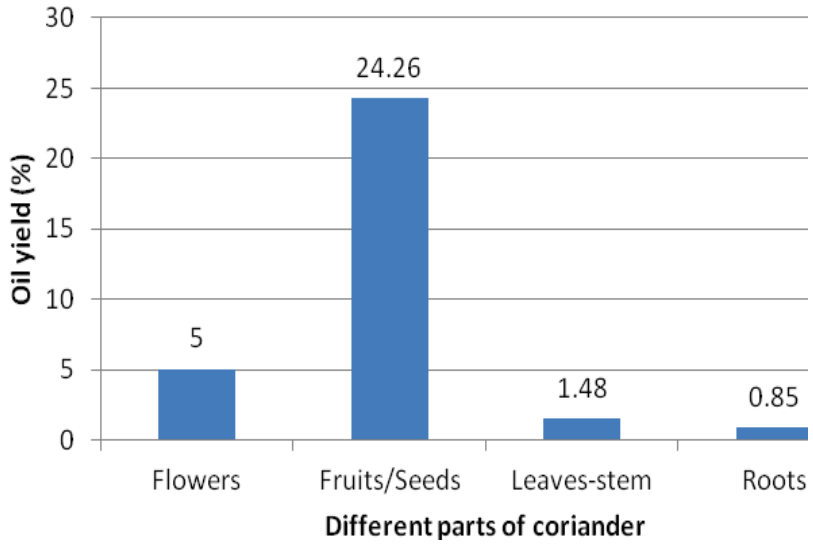


Figure 1: La teneur en huile de coriandre fleurs, feuilles, racines et fruits

Le profil des acides gras et le pourcentage d'acides pétrosélinique sont des déterminants principaux de la qualité globale de l'huile de coriandre. Cependant, les comparaisons entre les différentes parties de coriandre ont été faites, l'acide pétrosélinique apparait faiblement sur les feuilles et les fleurs, et est absent des racines: peut-être que c'est la raison principale pour laquelle il y a très peu de recherches sur la composition en acides gras des fleurs, feuilles et racines de coriandre.

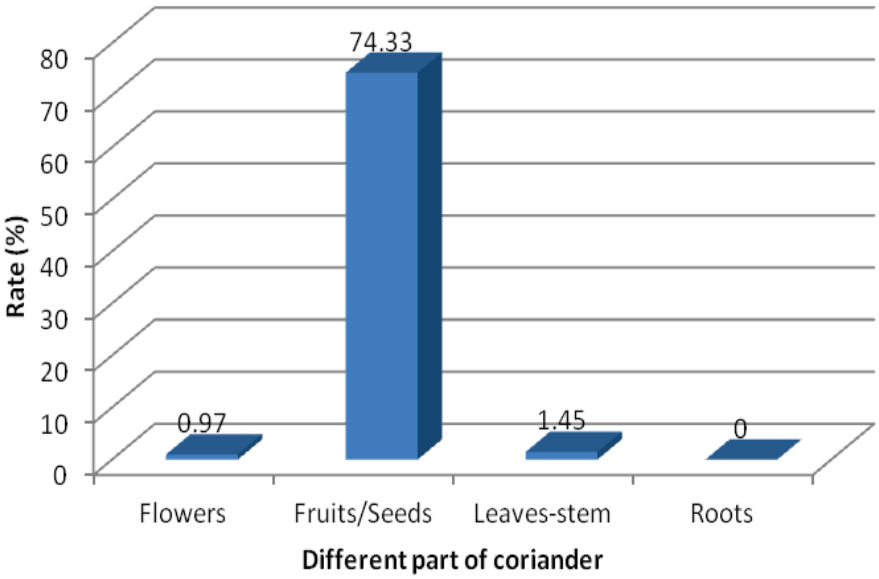


Figure 2: Composition d'acide pétrosélinique dans différentes parties de coriandre

A partir des résultats de la présente enquête, les pourcentages d'acide palmitique et linoléique dans les fleurs, les feuilles-tige et les racines sont plus élevés que dans les fruits de coriandre. Mais leur pourcentage d'acide pétrosélinique est très faible (<1% en fleur et <1,4% dans les feuilles et la tige), même l'acide pétrosélinique est pratiquement absent des racines de coriandre (Figure 2). D'autre part, le rendement en huile des feuilles - tiges (<1,5%) et de racines (<1%) est beaucoup plus faible celui des fruits et des fruits (jusqu'à 25%). Par conséquent, c'est pour nous la principale raison de nous concentrer sur l'étude des fruits de coriandre plutôt que sur les feuilles et les racines de coriandre dans les prochaines études de cette thèse.

II.1.2 Influence de la date de semis sur le rendement et la composition en acides gras de la coriandre

Dans cette étude, le cultivar coriandre français (FR1 - Dourou) a été choisi afin d'étudier les effets de la date de semis sur le rendement en huile et la composition en acide gras des fruits de coriandre. Ces essais ont été cultivés à différentes de semis dans des conditions organiques. Les résultats ont montré que la date de semis est l'un des facteurs importants qui influent sur la composition en acides gras et le rendement de l'huile des fruits de coriandre. Il y avait des différences significatives de teneur en huile entre trois dates de semis, DS1 (24,26%) ont enregistré les plus hautes valeurs de rendement en huile par rapport au rendement de DS2 (20,19%) et DS3 (17,36%). En outre, la diminution de l'acide pétrosélinique a diminué sensiblement : de 74,33% en DS1; 71,30% en DS2 à DS3 67,83%. L'influence de la date de semis sur le rendement et la teneur en acide gras est significatif. La date de semis est déterminante pour fournir aux plantes les conditions pour utiliser au maximum les paramètres environnementaux, en particulier la température et de rayonnement. Si les facteurs environnementaux sont appropriés pour l'écologisation, l'établissement et la survie des semis, alors le rendement maximal sera atteint à la date de semis appropriée. En outre, établir des date de semis appropriées sera un élément fondamental d'un système agricole réussi.

II.1.3 L'accumulation des acides gras durant la maturation de la coriandre

Le cultivar coriandre française (FR1 - Dourou) a été cultivé sous agriculture biologique au cours de la campagne agricole (2009, 2010 et 2011). Les semis ont été faits le 23 Mars, le 9 Avril et le 25 Mars en 2009, 2010 et 2011, respectivement. Les prélèvements de semences ont été effectuées tous les cinq jours (en moyenne) entre le début de la floraison

et la maturité. La période de récoltes a été lancée à partir de 2 jours DAF (après la fleur) à 59 DAF pour évaluer l'accumulation d'acides gras dans la maturité de la coriandre. Les changements dans le rendement de l'huile de coriandre au cours de la floraison à maturité en 2009, 2010 et 2011 ont montré que le rendement de l'huile a augmenté très rapidement entre le 10 au 35 jour après la période de floraison d'échantillonnage et a atteint son niveau maximum à l'échéance (jusqu'à 25%) (Figure 3), le rendement en huile était maximal et sa valeur était semblable à d'autres rapports portant sur les fruits mûrs de coriandre. La tendance de l'évolution de l'accumulation de l'huile dans les fruits de la coriandre était similaire aux résultats rapportés dans des Brassicacées (Chen et al., 2011; Trémolières et al., 1978), Asteraceae (Lardans et Trémolières, 1991;. Roche et al., 2010; Roche et al., 2006). Une importante accumulation d'acides gras est lancée à partir du 2 DAF. Différentes tendances ont été observées pour les acides gras détectés. La teneur en acides gras saturés était haute de 2 DAF jusqu'au 10 DAF puis a chuté rapidement, ce déclin se poursuit jusqu'à la pleine maturité. Les acides gras de cette catégorie dans la coriandre sont les acides palmitique, myristique et stéarique qui ont suivi la même tendance que les acides gras saturés. En revanche, les acides gras monoinsaturés, surtout représentés par l'acide pétrosélinique, étaient présents à faible quantité au début de la formation des fruits (2 DAF) et continuaient à augmenter durant la période de maturation (Figure 4).

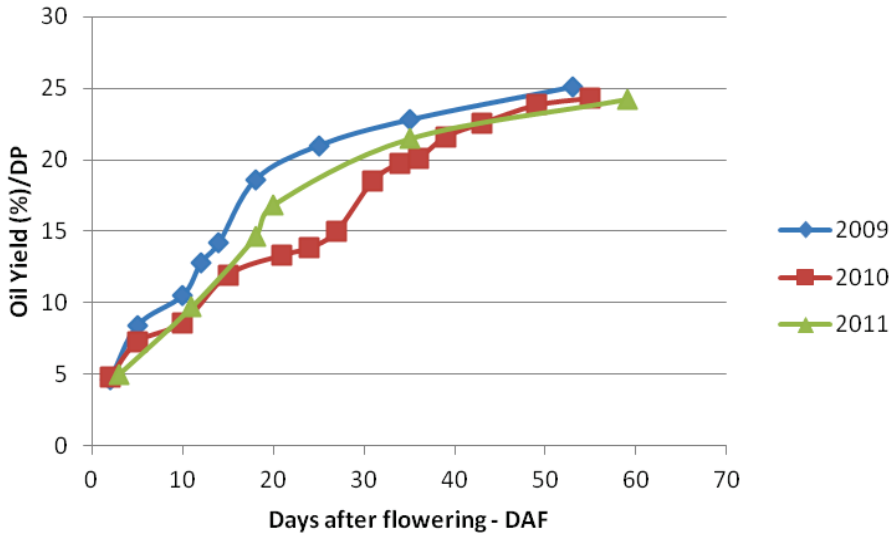


Figure 3: Variations des rendements en huile de coriandre de la floraison à la maturité (2009, 2010, 2011)

En outre, le cultivar coriandre vietnamienne (Vinaseed - VS1), cultivée sous agriculture biologique au cours de la campagne agricole 2012 à Ha Noi (Viet Nam) a été sélectionnée en vue d'étudier l'accumulation d'acide gras durant la maturation de la coriandre. Au cours de l'évolution des fruits, la tendance de rendement en huile et la composition en acides gras est relativement similaires à la coriandre française cultivée à Auch (2009, 2010, 2011). Nos résultats sont en accord avec ceux rapportés dans la littérature. Des niveaux plus élevés d'acides gras polyinsaturés et saturés ont déjà été signalés aux premiers stades de la maturation des fruits (Lakshminarayana et al., 1981; Msaada et al., 2009.). Le rapport des acides gras saturés aux acides gras polyinsaturés a diminué de manière significative au cours de la maturation des fruits. Des résultats similaires ont été signalés dans d'autres espèces d'oléagineux (Chen et al., 2011; Peiretti et al., 2004; Roche et al., 2006; Vuorinen et al., 2014). Le niveau d'acide pétrosélinique était conforme aux résultats déjà rapportés dans la coriandre allant de 51,6 à 90,7% (Angelini et al., 1997; Guidotti et al., 2006; Lakshminarayana et al, 1981; Msaada et al., 2009). Le montant le plus élevé d'acide pétrosélinique a été atteint entre 18 et 35 DAF. Ce résultat était d'accord avec le rapport de Msaada (Msaada et al., 2009), qui a souligné qu'une période de 32 à 35 DAF était suffisante pour l'utilisation de coriandre fruits.

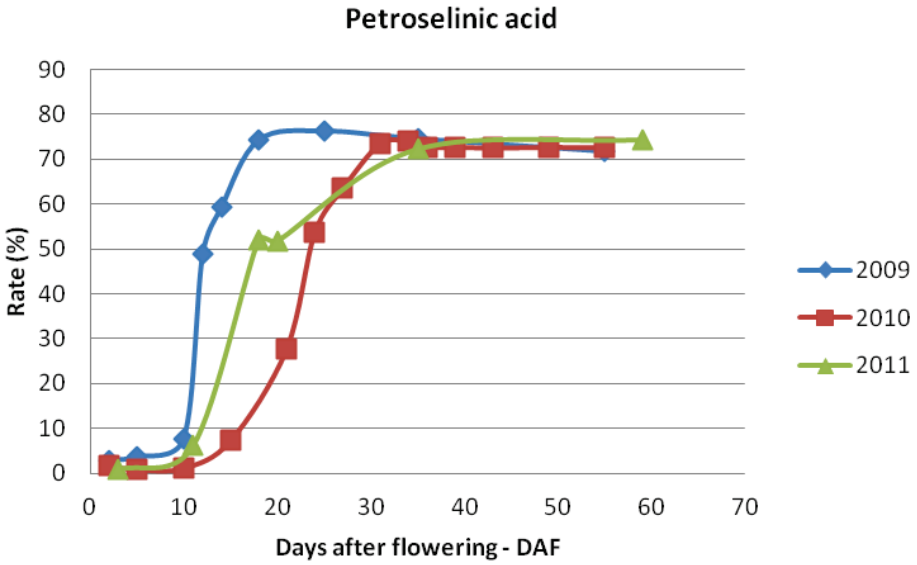


Figure 4: La tendance de l'acide pétrosélinique de coriandre de la floraison à la maturité (2009, 2010, 2011)

II.1.4 Effets des différents emplacements sur le rendement la teneur en acides gras de la coriandre

Le but de cette étude était d'examiner les compositions d'acides gras et la teneur en huile dans les fruits de coriandre cultivés à des endroits différents. Trois sites ont été choisis pour la plantation de coriandre, et les fruits récoltés à partir de trois emplacements ont été analysés pour déterminer les effets des différents lieux sur le rendement de l'huile et de la composition en acides gras de la coriandre vietnamienne. La coriandre vietnamienne (Vinaseed - VS1) a été cultivé à trois endroits: Ha Noi; Thai Binh (nord du Vietnam - delta du fleuve Rouge) et Ha Tinh (côte centrale du Nord du Vietnam). Selon les résultats d'analyse, la teneur en huile de coriandre a été significativement affectée par l'emplacement de la croissance et la teneur en huile a varié entre 17,23% et 19,19% selon la localisation, le taux d'huile le plus élevée a été observé à Ha Noi. La température à Ha Noi était supérieure à celle de Thai Binh et Ha Tinh dans la saison de floraison de la coriandre (de mai à Juillet 2012), cela peut être la cause du rendement d'huile plus élevé de la coriandre à Hanoi. A partir des résultats obtenus, on peut voir que les acides gras saturés et insaturés dans l'huile de coriandre changeait sensiblement en fonction de l'emplacement de croissance. L'acide gras saturé dans cette huile était l'acide palmitique (allant de 3,99 à 4,69%). Les deux principaux acides gras mono-insaturés sont l'acide pétrosélinique (allant de 68,96 à 72,55%) et l'acide oléique (allant de 6,72 à 6,92%). Il y a quatre acides gras essentiels : l'acide pétrosélinique, l'acide linoléique, l'acide oléique et l'acide palmitique sont les acides gras significatifs en termes de qualité et de quantité d'huile de fruits de coriandre, parmi lesquels l'acide gras majoritaire est l'acide pétrosélinique (de 68,96 à 72,55% du total des acides gras). De ce résultat, nous pouvons voir que cette étude a révélé que les différences écologiques, notamment la température, l'humidité et les précipitations impacteront significativement à la fois la teneur en huile de la graine et la composition des acides gras dans la coriandre. Par conséquent, l'emplacement de la croissance est un facteur important pour satisfaire les besoins du marché de la graine de coriandre en termes de qualité de l'huile. En dehors des conditions de croissance, il faut aussi tenir compte d'autres facteurs tels que le cultivar, le moment de la plantation et de l'irrigation, le mode de culture afin d'obtenir un rendement de désir et de la qualité.

II.1.5 Composition en acides gras de différentes cultivars

Cette étude a été réalisée afin d'évaluer le rendement des cultivars de coriandre à Auch. Il est nécessaire de développer des cultivars plus adaptées à la production de semences pour satisfaire la demande croissante de cette culture d'épices. La sélection d'une meilleure cultivar peut être d'une immense utilité au producteur pour la poursuite de l'amélioration et du développement de la culture. Par conséquent, la présente enquête a été menée afin d'évaluer les conditions appropriées aux cultivars de coriandre collectées et pour sélectionner les cultivars prometteurs pour un rendement grainier plus élevé en France. L'expérience a été menée dans des conditions d'agriculture biologique à Auch, de mars à septembre 2011. Les sept cultivars (Canada, Tunisie, Lituanie, Algérie, France (FR1 (Dourou) - DS1), Vietnam (VS2) et Chine) ont été cultivées et les matières premières récoltées à l'étape de pleine maturation (Septembre 2011) ont été étudiées.

Selon les résultats d'analyses, la teneur en huile varie de 15,28 à 24,26% selon les cultivars. Parmi sept cultivars de coriandre, la teneur en huile de trois cultivars de coriandre (Algérie, Canada et Chine) était relativement faible, allant de 15,28% à 16,87%. La coriandre française avait la teneur en huile plus élevée (jusqu'à 24,26%), suivie par celle de Lituanie (19.04%), de Tunisie (18,70%) et du Vietnam (17,64%). Toutes les cultivars sont riches en acides gras insaturés, les teneurs sont : Canada (93,19%), Tunisie (94,28%), Lituanie (94,91%), Algérie (94,05%), Chine (94,14%), Vietnam - VS2 (94.21) et France - DS1 (95,2%). L'acide pétrosélinique est l'acide gras le plus abondant parmi les sept cultivars avec un taux de 69,65% à 74,33%, la teneur de la cultivar de coriandre française (74,33%) a été statistiquement la plus élevée. D'autre part, afin de comparer la teneur en huile et la composition en acides gras entre les cultivars cultivées à Auch (dans des conditions organiques) avec les fruits d'origine, les résultats obtenus à partir de l'analyse préliminaire ont montré qu'il existe des variations considérables entre les profils d'acides gras et les teneurs en huile entre cultivars cultivées à Auch et les cultivars d'origine. La teneur en huile de la plupart des cultivars à Auch était inférieure à celle des semences d'origine, entre 16,5% à 17,9% pour la cultivar canadienne, entre 19% à 21,1% pour la lituanienne, entre 24,3% et 25% pour les françaises et entre 17,6% à 21,1% pour les cultivars vietnamiennes. Les différences entre les teneurs en huile de fruits peuvent probablement être dues à la génétique, la croissance, les facteurs climatiques (tels que la température et l'humidité) pendant les différents stades de croissance, les conditions environnementales,

d'analyse et l'emplacement. Cette étude est la première à fournir des informations comparatives sur la teneur en huile et en acide gras des sept cultivars (Canada, Tunisie, Lituanie, Algérie, Françaises (FR1 (Dourou) - DS1), Vietnam (VS2) et Chine) sous condition d'agriculture biologique. Les résultats de la présente étude peuvent être utiles pour trouver des cultivars de coriandre appropriées qui peuvent cultiver dans des conditions d'agriculture conventionnelles à Auch. En outre, nos résultats contribuent au choix des cultivars adaptées dans des conditions climatiques similaires.

II.2 Composition de l'huile essentielle

II.2.1 Influence de temps de semis sur le rendement et la composition de l'huile essentielle de coriandre

Dans cette étude, le cultivar coriandre française (FR1 - Dourou) a été choisi afin d'étudier les effets de la date d'ensemencement sur le rendement de l'huile et la composition de l'huile essentielle de fruits de coriandre. Ces essais ont été cultivés à différentes dates moment d'ensemencement, en conditions d'agriculture biologique. Le premier essai (DS1) a été planté le 25 Mars, le deuxième (DS2) le 14 avril, le troisième (DS3) le 30 Avril à Auch (2011). Dans l'expérience, la matière première a été récoltée à pleine maturité (Septembre 2011).

Les résultats ont montré une différence significative en teneur en huile entre les différentes dates d'ensemencement (essai DS1, DS2 et DS3). Plus l'ensemencement est tardif, plus le rendement en huile a tendance à diminue. Le taux de rendement moyen de l'huile essentielle se situait entre 0,41 et 0,47%. La différence entre les valeurs moyennes est significative, la valeur la plus élevée a été obtenue avec le premier essai DS1 (0,47%), suivie par DS2 (0,44%) et la plus faible valeur est obtenue avec DS3 (0,41%). Ces variations de rendement en huile pourraient être liées aux changements dans les facteurs environnementaux tels que la température, l'humidité et les précipitations pendant la maturation des fruits et au processus de maturation physiologique des fruits de coriandre (Msaada et al., 2007; Telci et al., 2006).

Dans ce résultat, treize (en essai DS1 et DS3) et quatorze (en essai DS2) composantes de l'huile totale ont été identifiée. Le linalool est le composant le plus caractéristique de l'huile de coriandre et constitue, dans tous les essais étudiés, 74,05 à 76,59% de la quantité totale d'huile. Les autres composantes principales caractéristiques des huiles de fruits de

coriandre sont le α-pinène (03/30 à 04/27%), γ-terpinène (3,15% - 4,13%), l'acétate de linalyle (2,51 à 4,28%), l'acétate de géranyle (2,89 à 3,95%), de p cymène (1.27 à 1.29%). Parmi elles, le linalol et α-pinène ont tendance à diminuer l'impact de la date d'ensemencement. Par contre, le γ-terpinène, l'acétate de linalyle, l'acétate de géranyle, le p-cymène ont tendance à augmenter légèrement l'impact de la date d'ensemencement. Le linalool, un monoterpène oxygéné, était le principal composant de l'huile essentielle de fruits mûrs. Il représente plus de deux tiers des éléments volatiles de l'huile de fruits de coriandre et est considéré comme l'un des composés qui impactent la saveur de l'huile essentielle de fruits (Cadwallader et al., 1999). Il est connu que les facteurs climatiques tels que les jours nuageux, une température plus basse pendant la maturation ou de fortes précipitations peuvent avoir des effets défavorables sur l'accumulation de linalol (Sangwan et al., 2001). Les résultats du rendement en huile et ses composantes selon les dates d'ensemencement ont déjà été mentionnés ci-dessus. Il y avait des différences significatives de teneur en huile dans les trois dates d'ensemencement (DS1, DS2 et DS3). DS1 a montré les valeurs les plus élevées de rendements en huile et en linalool, Par conséquent, DS1 (fin mars) pourrait être proposée comme date d'ensemencement appropriée pour la plantation des culitivars de coriandre en conditions biologiques à Auch.

II.2.2 Composants essentiels de l'huile de différentes cultivars de coriandre

La coriandre (*Coriandrum sativum* L.) ont été obtenus en supermarché, entreprise de semences, magasins de détail de différents pays européens et asiatiques (sept cultivars - Canada, Tunisie, Lituanie, Algérie, Chine, France (FR1 (Dourou) - DS1) et Vietnam (VS2). Après avoir été recueillies, des cultivars de coriandre ont été cultivées en agriculture biologique à Auch, France. Ces cultivars ont été cultivées le 25 Mars 2011.

Dans ce résultat, le rendement en huile essentielle a varié de 0,43 à 0,95%, la teneur en huile maximale (jusqu'à 1,21%) a été observée avec la cultivar lituanienne, suivie par la cultivar vietnamienne (0,65%), chinoise (0,61%), tunisienne (0,59%), algérienne (0,45%), française DS1 (0,44%) et canadienne (0,43%). Sa valeur était semblable à ce qui se trouvait dans d'autres rapports qui indiquent que dans les fruits mûrs, la teneur en huile essentielle est aussi faible (typiquement, moins de 1%) (Telci et al., 2006). Dans les études précédentes, la teneur en huile essentielle des fruits de coriandre fruits varie d'une valeur très faible (0,03%) à une valeur maximum de 2,7% (Purseglove et al. 1981). Les résultats montrent que quatorze composés différents ont été identifiés dans l'huile essentielle de cinq

cultivars (canadienne, tunisienne, lituanienne, algérienne, vietnamienne) et treize composés dans l'huile essentielle de deux autres cultivars (chinoise et française). Le linalol, le γ-terpinène, le Campor, l'acétate de linalyle, l'acétate de géranyle, α- pinène ont été identifiés comme les principaux composants de l'huile de toutes les cultivars. L'huile essentielle de cinq cultivars a une teneur en linalol de plus de 75% : canadienne (77,67%), lituanienne (78,34%), chinoise (75,78%), française DS1 (76,59%) et vietnamienne (78.05%) a montré une teneur en linalol plus de 75 %. Deux culitvars ont montré une teneur en linalol de moins de 75% : cultivars tunisienne et algérienne. Dans ces résultats, toutes les cultivars étudiées ont plus de 74% de linalol, montrant une haute qualité de fruits de coriandre qui peuvent être utilisés dans les aliments, les produits pharmaceutiques et d'autres industries connexes. Les résultats obtenus par plusieurs auteurs comparés aux résultats de ce travail, montrent que ces composés présentent des variations, qui peuvent être liées au climat et à l'état du sol, à la saison de la récolte et de développement de la plante. Le linalool, comme un terpène alifatic, était un composant majeur de sept culitvars de coriandre plantées dans des conditions d'agriculture biologique. Les résultats de la présente étude peuvent être utiles pour recommander les cultivars de coriandre optimales pour la plantation dans des conditions d'agriculture biologique à Auch. De plus, les résultats ont révélé la pertinence de sept cultivars pour une utilisation en tant que matériaux génétiques initiaux pour la sélection de cultivars de coriandre appropriées à la France.

II.3 Propriétés antioxydantes

Dans cette étude, la quantité de composés phénoliques totaux dans les extraits de sept cultivars de coriandre variait de 12,71 à 42,88 mg GAE/g. La cultivar tunisienne avait la plus grande valeur de PTC de 42,88 mg GAE/g, suivie par la cultivar canadienne (39.29 mg GAE/g d'extrait). La quantité de composés phénoliques de la cultivar française de coriandre est la plus basse. L'analyse des antioxydants montre que tout extrait méthanolique des cultivars de coriandre a une action antioxydante. Cependant, leur capacité de piégeage des radicaux (RSC) varie dans une large mesure. Dans ces résultats préliminaires, les activités antioxydantes de l'extrait méthanolique de sept culitvars de coriandre a été étudiée. L'action antioxydante de la coriandre était suffisamment élevée pour que la plante devienne une nouvelle source naturelle de substances antioxydantes pour son utilisation comme additifs dans les aliments naturels. Les cultivars tunisiennes et canadiennes ont montré la plus grande quantité sur les polyphénols totaux, la tunisienne a

montré la plus forte activité DPPH de balayage, FRAP et le dosage ORAC. En outre, les capacités antioxydantes variaient considérablement selon les cultivars de coriandre et dépendent de la composition phénolique des cultivars de plantation dans des conditions organique à Auch.

III. Bioraffinerie de fruits de coriandre, une première enquête

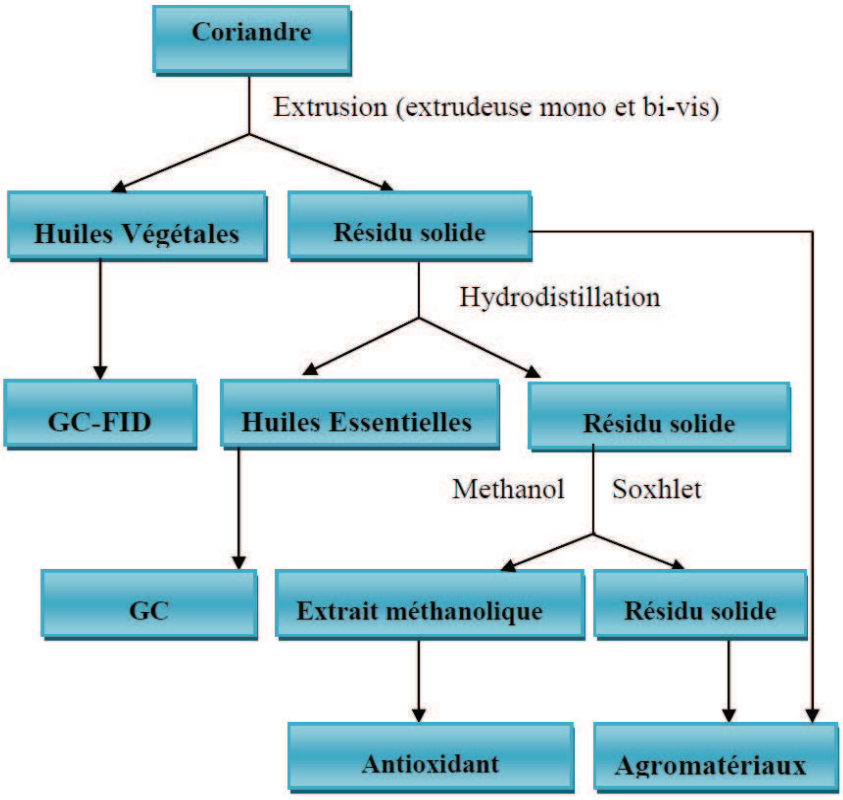


Figure 5: Schéma de préparation et l'analyse d'extraits de coriandre

Basé sur le concept de bioraffinerie, l'objectif de ce travail est de traiter une plante pour atteindre la valeur la plus élevée possible sur les matières premières de biomasse, à savoir la valorisation de la plante entière par des extractions séquentielles de molécules d'intérêt.

On peut voir à partir de la Figure 5 que l'extrait d'huile végétale et le résidu solide (tourteau) de coriandre sera produit par extrusion en utilisant les technologies d'extrudeur mono-vis ou bi-vis. Ensuite, l'extrait d'huile essentielle est obtenu à partir du tourteau (résidu) par hydrodistillation. Pour le résidu solide, il peut être traité avec du methanol pour obtenir des extraits méthanoliques. Le résidu solide final peut être transformé en agromatériaux et biocomposites, utilisé comme engrais, pour la production d'énergie, à

l'alimentation animale ou directement comme carburant. Des extraits méthanoliques seront évalués pour leur action antioxydante. Les plantes ayant une action antioxydante élevée seront les sources potentielles d'antioxydants naturels. En modélisant des études dans cette étude, les molécules dans la matière végétale peuvent être pleinement utilisées, évitant ainsi un grand gaspillage de ressources naturelles.

Actuellement, deux techniques d'extraction différentes sont généralement utilisées pour obtenir de l'huile végétale à partir de fruits, à savoir l'extraction de solvant organique et le pressage mécanique. Actuellement, le pressage mécanique devient la méthode la plus couramment utilisée pour l'extraction commerciale d'huile. Bien que le rendement d'extraction correspondant est légèrement inférieur, la vis de pression est la méthode d'extraction de l'huile la plus populaire. En effet, ce processus est simple, continu, flexible et sûr (Singh et Bargale, 2000; Zheng et al., 2005). Pour l'extraction commerciale de l'huile, cette technologie présente de nombreux avantages, y compris sa polyvalence, une productivité élevée, le faible coût et la capacité de produire des formes de produits uniques et de haute qualité (Köksel et al., 2004). Par conséquent, l'application du processus d'extraction de l'huile en utilisant la technologie d'extrusion sera étudiée dans cette étude en utilisant deux extrudeuses mono- et bi-vis). Les objectifs de ce travail étaient d'évaluer la faisabilité de la pression mécanique d'extraction d'huile végétale à partir de fruits de coriandre, et d'étudier l'influence des conditions d'exploitation, soit le diamètre de la buse et la buse/vis distance (cas d'extrusion monovis) ou la configuration de vis, dispositif de coefficient de remplissage et de la température de pressage (de cas d'extrusion bi-vis), sur le rendement de l'extraction de l'huile.

III.1 L'extraction de l'huile de coriandre en utilisant la technologie d'extrudeur mono-vis

III.1.1 Effet des conditions d'exploitation sur le rendement de l'huile dans l'extrudeur mono-vis

Afin d'évaluer les effets du diamètre de la buse et la buse/distance de vis dans l'extraction d'huile à partir des fruits de coriandre, tous les essais ont été réalisés avec un seul lot de fruits de coriandre (coriandre Français - GSN). Un poids de 500 g de fruits est utilisé pour chaque expérience. Le taux d'humidité était de 9,77 ± 0,10% (norme française NF V 03-903). La teneur en huile déterminée par extraction au Soxhlet en utilisant du n-hexane comme solvant d'extraction était de 27,67 ± 0,57% de matière sèche (NF V 03-908). Dix-

huit essais ont été réalisés. Les résultats obtenus à partir des différentes conditions de fonctionnement testées (par exemple le diamètre de la buse et la distance entre la buse et la vis) sont présentés dans le Tableau 1, et les rendements en huile obtenus (R_S) sont présentés sur la Figure 6. Les données dans le Tableau 1 montrent que les différentes conditions opératoires mises en oeuvre ont une influence sur l'extraction de l'huile végétale à partir de fruits de coriandre. Le rendement en huile R_S varie de 12,8% à 16,0%, et le rendement de l'extraction du pétrole tend à augmenter de l'essai 1 à l'essai 18.

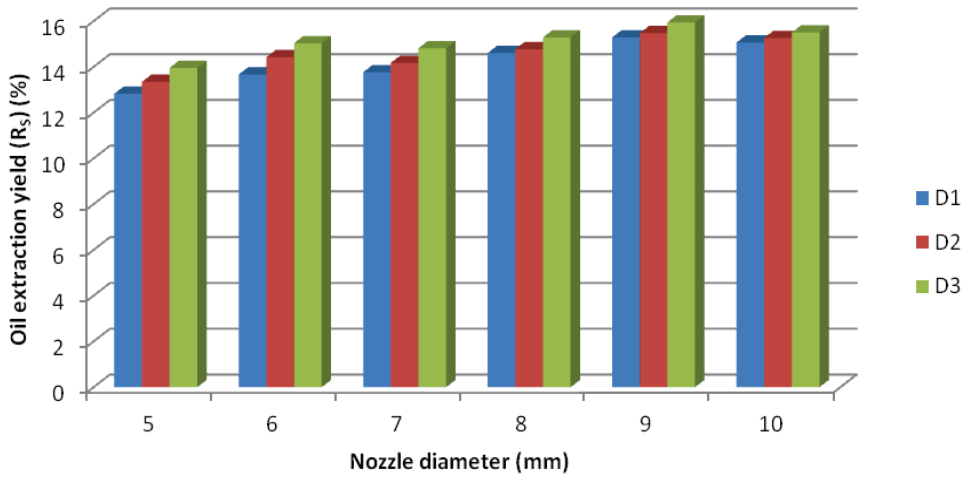


Figure 6: Variation du rendement d'extraction d'huile (R_S) de la configuration de vis différent

En général, le diamètre de la buse (allant de 5 à 10 mm) affecte l'efficacité de l'extraction de l'huile. Les rendements en huile obtenus pour des diamètres de buse de 8, 9 et 10 mm sont plus élevés que les rendements d'huile obtenus pour des diamètres de buse de seulement 5, 6 et 7 mm. Le rendement le plus élevé d'extraction d'huile (16,0%) a été obtenu pour un diamètre de buse de 9 mm et la buse / la distance de vis de 3 mm, tandis que le plus bas (par exemple 12,8%) a été obtenu pour un diamètre de buse de seulement 5 mm et la buse / la distance de la vis de seulement 1 mm (Figure 6). En outre, à partir de ces résultats, le rendement d'extraction de l'huile augmente lorsque la distance entre la buse et la vis augmente, pour tous les diamètres de buses testés et en particulier pour des diamètres de buse de 8, 9 et 10 mm. Par exemple, pour le diamètre de la buse de 9 mm, le rendement en huile de R_S varie de 15,3% à la distance de 1 mm à 16,0% pour la distance de 3 mm. Ainsi, l'augmentation de la distance entre la buse et la vis (de 1 à 3 mm) contribue à l'augmentation de la pression sur le matériau. En outre, la distance entre la buse supérieure

et la vis entraîne l'augmentation du temps de séjour de la matière première dans la zone de compression, ce qui augmente à la fois la pression et la matière rendement d'extraction de l'huile. D'autre part, plus la distance buse / vis est grande, plus la teneur en pied dans le filtrat (c.à.d particules solides entraînée moins à travers le filtre au cours de la séparation liquide/solide) est faible. La qualité des huiles pressées a également été examinée (Tableau 1). L'acidité de l'huile varie de 1,3 à 1,8%. A partir de ces résultats, les conditions de fonctionnement (par exemple de diamètre de buse et la distance entre la buse et la vis) n'ont aucune influence sur la qualité de l'huile, cette dernière étant toujours satisfaisante.

Dans cette étude, les différentes configurations ont été testées afin d'évaluer l'impact des conditions de fonctionnement de l'extrusion sur l'efficacité de l'extraction de pétrole. L'effet sur le rendement en huile a été évalué grâce à la détermination de trois rendements différents: R_S (abordé précédemment), R_L et R_C. Le rendement en huile (R_L) est défini comme le rapport de l'huile pressée à l'huile totale contenue dans le fruit. Dans le Tableau 1, le rendement en huile (R_L) varie de 41,8 à 52,0%. Le rendement le plus élevé de l'huile (R_L) pour cette étude (c.à.d 52,0%) a été obtenu dans les conditions d'exploitation suivantes: diamètre de la buse 9 mm et 3 mm de distance buse / vis. Cette valeur est similaire à d'autres résultats rapportés dans la littérature pour l'extraction de l'huile de coriandre dans une extrudeuse à vis unique (Sriti et al., 2011a). En outre, la teneur en huile résiduelle dans le tourteau varie de 14,2 à 16,8% de la matière sèche. Cela conduit à un rendement en huile (R_C), sur la base de la teneur en huile résiduelle dans le tourteau, compris entre 49,8 et 57,1% (53,8% pour les conditions optimales) (Tableau 1).

Pour les teneurs en acides gras des huiles pressées, les résultats ne montrent aucun changement réel dans les proportions relatives des acides gras selon les conditions d'extrusion utilisées. Dans tous les essais, neuf acides gras ont été identifiés. Toutes les huiles pressées sont très riches en acide pétrosélinique (72,6 à 74,2% selon les conditions utilisées, à savoir la buse/la distance de la vis et le diamètre de la buse), suivie par les acides linoléique, oléique et palmitique, représentant respectivement de 13,2 à 14,1%, 7.1 3,3 à 4,0% et 7,8% des acides gras totaux. Dans tous les essais, les acides gras saturés (AGS) représentaient 4,3 à 5,0% des acides gras totaux, tandis que les acides gras monoinsaturés (AGMI) représentaient 80,6 à 81,9% et les acides gras polyinsaturés (AGPI) représentaient 13,5 à 14,3%. En résumé, les conditions opératoires ont joué un rôle important pour influencer le rendement d'extraction de l'huile dans l’extrudeuse à simple

vis (mono-vis), et des rendements en huile plus élevés ont été obtenus à partir de la configuration permettant une plus grande pression des matériaux, à savoir un diamètre de buse de 8 à 10 mm. En général, la modification des conditions d'extrusion n'a pas influencé la qualité de l'huile. En outre, le meilleur état de fonctionnement (3 mm buse/vis distance et du diamètre de la buse 9 mm) conduit à un rendement en huile de 16,0% (R_S) et à un rendement en huile de 52,0% (R_L).

III.1.2 Influence de la cultivar de coriandre sur l'efficacité de l'extraction de l'huile

Des expériences ont été menées sur l'extraction de l'huile végétale dans l'extrudeur mono-vis de cinq cultivars différentes de fruits de coriandre. Les cultivars de coriandre tunisienne, lituanienne et vietnamienne ont été obtenues en supermarché. La coriandre française a été fournie par la société GSN Semences (France) et la cultivar française Dourous DS1 a été cultivée à Auch (France).

Lorsque l'on étudie l'effet des conditions de fonctionnement sur l'extraction d'huile à partir de fruits de coriandre française (GSN) dans l'extrudeuse à vis unique, le rendement le plus élevé de l'huile a été obtenu à partir de la configuration suivante: 3 mm buse/distance de vis et diamètre de la buse 9 mm. Cette configuration est la meilleure optimisation à appliquer pour l'extraction d'huile végétale, ce qui conduit à la meilleure efficacité de l'extraction de l'huile. Par conséquent, cette configuration de vis sera appliquée à partir de différentes cultivars de coriandre. Quinze essais ont été réalisés, à savoir trois répétitions pour chaque variété.

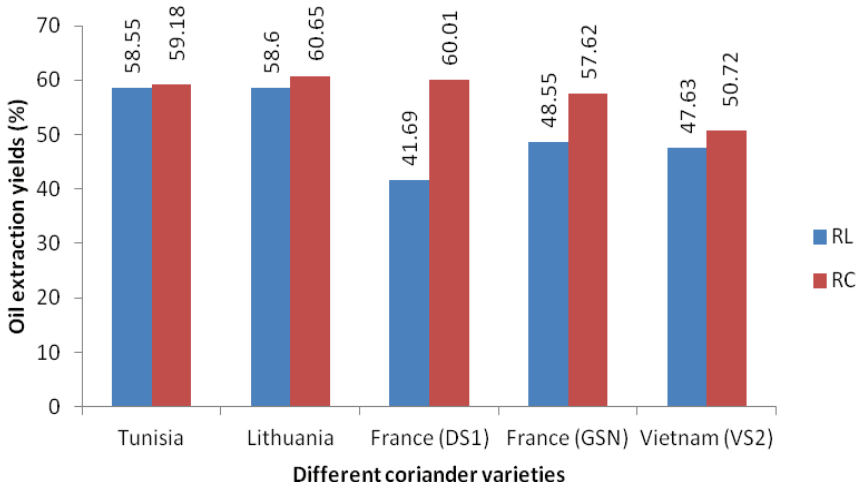


Figure 7: Variation des rendements d'extraction d'huile (%) pour les différentes cultivars de coriandre testé

Pour les cinq cultivars de coriandre testées, l'effet sur l'extraction de l'huile a été évalué grâce à la détermination des trois rendements en huile différents: R_S, R_L et R_C. Les résultats obtenus sont présentés dans le Tableau 2, et les rendements en huile associés (R_L et R_C) sont présentés sur la Figure 7. En fonction de la cultivar de coriandre, le rendement en huile (R_S) varie de 11,1 à 14,9%. Le rendement en huile plus élevée (c.à.d 14. 9%) est obtenu à partir de la coriandre française (GSN), suivie par la cultivar française (DS1), vietnamienne, tunisiennes, lituaniennes, comptant respectivement 14,4%, 13,8%, 11,6% et 11,1%. Sur la Figure 7, le rendement en huile de R_C est plus élevé que celui de R_L dans tous les essais, car il comprend tout l'huile végétale dans le filtrat, à savoir non seulement l'huile pressée, mais aussi de l'huile contenue à l'intérieur du pied. De la Figure 8, on peut voir que le rendement en huile extraite par pressage mécanique à l'aide de l'extrudeuse mono-vis est toujours inférieur à l'extraction par solvant (méthode Soxhlet). Ceci est en accord avec l'opinion de Shahidi (2005) que la vis de pression est utilisée pour la récupération de l'huile jusqu'à 90%, tandis que l'extraction par solvant est capable d'extraire 99% (Shahidi, 2005). Bien que le rendement de l'huile obtenue en utilisant l'extrudeuse à vis unique est inférieure par rapport à une recherche précédente (Sriti et al., 2011), cette méthode est encore une méthode d'extraction d'huile populaire car le processus est simple, continu, flexible et sûr.

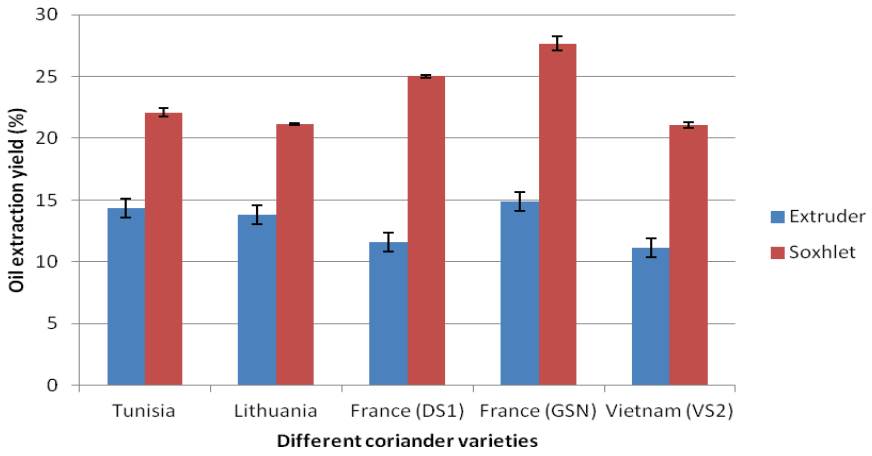


Figure 8: Comparaison entre les rendements d'extraction de l'huile dans la extrudeuse mono-vis et en utilisant le procédé de Soxhlet avec du n-hexane comme solvant d'extraction.

Les résultats de la composition en acides gras dans les huiles pressées ont été identifiés. La composition en acides gras de tous les huiles de coriandre pressées a été caractérisée par

des quantités importantes d'acide pétrosélinique, l'acide linoléique, l'acide oléique et l'acide palmitique, la variation en acides gras en fonction de la cultivar de coriandre utilisé. L'acide gras insaturé (AGMI, plus AGPI) contenus étaient toujours élevé (>94%). Généralement, les composés d'acides gras n'ont pas changé de façon significative entre les deux méthodes d'extraction (d'extrudeur mono-vis et d'extraction de Soxhlet).

III.1.3 Les utilisations potentielles des tourteaux produits dans l'extrudeur mono-vis

Basé sur le concept de bioraffinerie qui a été développé dans ce travail, des matières végétales de coriandre ont été traitées en différentes parties: huile végétale, huile essentielle (ou extrait volatile), extrait méthanolique et résidu final (biocomposites). De cette manière, les molécules dans les plantes peuvent être extraites séquentiellement et utilisées, évitant ainsi un grand gaspillage de ressources naturelles. Après avoir recueilli l'huile et le résidu solide (tourteau) de la méthode de pressage, ce tourteau contenait encore une partie de l'huile essentielle, sa teneur variant 0,26 à 0,35% de la matière sèche. La teneur maximale en huile essentielle (0,35%) a été trouvée à l'intérieur du tourteau de la coriandre lituanienne, et la plus faible (0,26%) était dans le tourteau de la coriandre vietnamienne. L'huile essentielle résiduelle dans les tourteaux pourrait être extraite au moyen d'hydrodistillation. Quatorze composés ont été identifiés et les plus importants sont les suivants: le linalol, le γ-terpinène, le camphre, l'acétate de géranyle, l'α-pinène, le limonène, le p-cymène. Parmi eux le linalol étant encore le principal composant pour tous les tourteaux: 77,8% pour la coriandre tunisienne, 78,8% pour la coriandre lituanienne, 75,4% pour la cultivar française DS1, 76,6% pour la cultivar française fournie par GSN entreprise et 78,1% pour la coriandre vietnamienne.

Une autre utilisation possible des tourteaux peut être l'isolement de certaines molécules naturelles afin de tester leur action antioxydante. Dans cette étude, les antioxydants ont été extraits des résidus (tourteaux) de fruits de coriandre obtenus par extrusion mono-vis, après extraction de l'huile essentielle par hydrodistillation. Une extraction au methanol utilisé comme solvant d'extraction a été utilisé pour obtenir les antioxydants (par exemple des extraits méthanoliques). Ensuite, une comparaison des résultats a été faite afin de savoir qui fournit une cultivar de coriandre résiduelle avec une concentration d'antioxydants plus élevée. L'extrait a été mesuré par différentes méthodes de dosage (FRAP, ORAC, dosage de DPPH), et les composés phénoliques totaux ont aussi été déterminés. Tous les extraits méthanoliques de tourteaux des différentes cultivars de coriandre avaient une action

antioxydant. Les extraits de tourteaux de cultivars tunisiennes et lituaniens ont une meilleure action antioxydante dans le dosage de FRAP, DPPH dosage et le dosage ORAC. Cela est directement lié à leur teneur en composés phénoliques totaux plus élevés, ce qui conduit à une action anti-oxydante supérieure.

III.2 L'extraction de l'huile de coriandre en utilisant la technologie d'extrudeur bi-vis

III.2.1 Influence des conditions de fonctionnement sur l'efficacité de l'extraction de l'huile

Les expériences ont été effectuées en utilisant un Clextral BC 21 (France) extrudeuse à double vis à co-rotation et co-pénétrante. Dans cette étude, les quatre profils de vis testés ont été basés sur ceux utilisés dans Evon et al. (Evon et al., 2010b) pour l'extraction de l'huile de jatropha. Les différences entre elles concernent les zones de pressage, toutes situées dans le module 7. Les vis à pas inverse utilisées dans les profils 1 et 2 étaient de 50 mm de long, avec un pas de -25 mm. Ils ont été positionnés immédiatement après le module de filtration pour le profil 1, à 25 mm de l'extrémité du module 6 pour le profil 2. Les vis à pas inverse utilisés dans les profils 3 et 4 ont la même longueur (50 mm), mais leur pas est plus grand (-33 mm au lieu de -25 mm). Ils ont été placés à 25 mm de l'extrémité du module 6 pour le profil 3 et immédiatement après le module de filtration pour le profil 4.

Afin d'évaluer l'impact des conditions de fonctionnement d'extrusion sur l'extraction de l'huile, des profils de vis différents ont été testés; le coefficient de remplissage de l'appareil et la température de pressage ont varié. L'effet sur l'efficacité d'extraction a été évalué grâce à la détermination de deux rendements en huile (R_L et R_C), le contenu du pied du filtrat et la consommation d'énergie de l'extrudeuse.

Dans le premier profil de vis testé (profil 1), les vis à pas CF2C inverse avaient 50 mm de long, avec un pas de -25 mm, et ils ont été placés immédiatement après le module de filtration (Figure 9). Avec une telle configuration de la vis et une température de pressage de 120 °C, un grand nombre de particules solides ont été rapidement poussés à travers le filtre, ce qui empêche l'huile d'être drainée librement et donc sa séparation à partir d'un tourteau. Ainsi, elle a simplement abouti à l'obstruction de l'extrudeuse à double vis. Ce phénomène a persisté malgré la réduction du coefficient de remplissage de l'appareil à moins de 10 g/h tours par minute.

Profile 1	1		2			3		4		5		6			7				
	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -25	C2F 25	C2F 33		
Profile 2	1		2			3		4		5		6			7				
	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -25	C2F 33			
Profile 3	1		2			3		4		5		6			7				
	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -33	C2F 33			
Profile 4	1		2			3		4		5		6			7				
	T2F 50	T2F 50	C2F 33	C2F 25	DM 10×10 (45°)	C2F 33	C2F 25	BB 10×10 (90°)	C2F 33	C2F 33	C2F 25	C2F 25	C2F 16	C2F 16	CF2C -33	C2F 25	C2F 33		

Figure 9: Configurations à vis pour l'extraction d'huile végétale à partir de coriandre fruits

T2F (*vis trapézoidal et à double filet*); C2F (*vis conjuguée et à double filet*); DM (*disque malaxeur mobolobe*); BB (*disque malaxeur bilobe*); CF2C (*contre-filet conjjugée et à double filet*).

Par conséquent un deuxième profil de vis (profil 2) a été testé, les mêmes vis de CF2C avec un pas de -25 mm étant alors positionnés à 25 mm de l'extrémité du module 6 (Figure 9). Cependant, le même phénomène, à savoir le colmatage de la machine, a été observé après seulement quelques dizaines de secondes. Comme les vis de CF2C avec un pas -25 mm utilisés pour les profils de vis 1 et 2 étaient trop restrictives, ces vis à pas inverse ont été remplacés dans les profils de vis 3 et 4 par des vis CF2C avec un pas supérieur, moins restrictif (-33 mm au lieu de -25 mm).

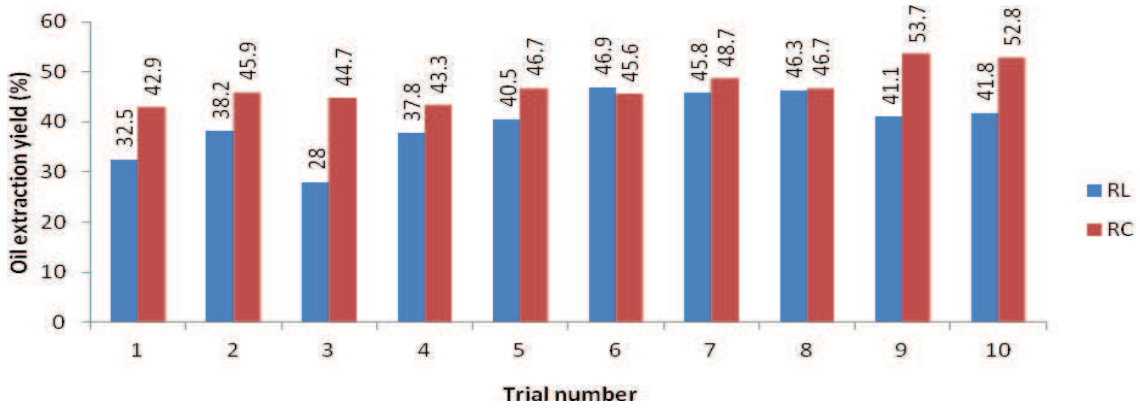


Figure 10: Variation in oil extraction yields for trials 1 to 10

Pour toutes les expériences menées avec ces deux derniers profils de vis, les échantillons de filtrat et les échantillons de tourteaux ont toujours été collectés séparément. La teneur en huile dans les tourteaux était plus faible que dans les fruits de coriandre et elle variait de 18,1 à 15,0% de la matière sèche, selon les conditions opératoires appliquées. Cela a conduit à un rendement en huile (R_C), sur la base de la teneur en huile résiduelle dans les tourteaux, comprise entre 42,9 et 53,7%. Comme prévu, plus fabiel était la teneur en huile dans le tourteau, plus le rendement en huile (R_C) était élevée, et cette relation est linéaire. Le rendement en huile (R_L), défini comme le rapport de l'huile pressée par rapport à l'huile totale contenue dans le fruit, variait de 28,0 à 46,9% (Tableau 3). Il a été plus faible que le rendement en huile (R_C) en raison de l'huile retenue dans le pied du filtrat (Figure 10) et la différence entre ces deux rendements en huile a été minimale avec la présence la plus basse de pied dans le filtrat (essais 6 à 8). En résumé, l'extraction de l'huile végétale à partir de fruits de coriandre provenant de France par pressage mécanique dans une extrudeuse à double vis (bi-vis) a été améliorée par rapport aux résultats obtenus dans l'étude précédente, qui utilisait des fruits de la cultivar tunisienne (Sriti et al., 2012). En effet, même si la cultivar française était plus riche en huile végétale que celle tunisienne (27,7% de la matière sèche au lieu de 21,9%), la plus faible teneur en huile résiduelle dans le tourteau (15,0% de la matière sèche) était inférieur à celui la cultivar tunisienne (16,6%). Cela signifie que l'appauvrissement en lipides était beaucoup plus efficace, conduisant à un meilleur rendement de l'huile (R_C): 54% au lieu de 45% seulement pour la cultivar tunisienne. En outre, la proportion du pied dans le filtrat ne dépassait jamais 17,9% en raison de l'utilisation d'une configuration de vis optimisée, alors qu'il était au moins à 47,5% dans la précédente étude (Sriti et al., 2012). Ceci facilite considérablement l'isolement de l'huile pressée, à savoir l'élimination du pied du filtrat par centrifugation.

III.2.2 Les utilisations potentielles des tourteaux produits dans l'extrudeur bi-vis

Les tourteaux contenaient encore une partie d'huile essentielle de fruits de coriandre, son contenu variant de 0,14 à 0,31% de la matière sèche. En outre, la teneur en huile essentielle résiduelle dans le tourteau dépend principalement de la température de pression appliquée dans l'extrudeuse à double vis : la teneur augmente lorsque la température diminue. Une teneur en huile essentielle maximale de 0,31% a été trouvée à l'intérieur du tourteau de l'essai 10, correspondant à la plus basse température de pressage testée (soit 65°C). Le linalool en étant la principale composante, il est l'un des ingrédients d'arôme les plus

couramment appliqués dans les cosmétiques, les parfums, shampooings, savons et même des détergents ménagers en raison de son parfum frais et fleuri, et il affiche une utilisation mondiale de plus de 1000 tonnes par an (Lapczynski et al., 2008). Une autre utilisation possible des tourteaux pourrait être l'isolation de certains antioxydants naturels par extraction méthanolique, ceux-ci étant potentiellement intéressants en raison de leur impact bénéfique sur la santé. La capacité antioxydante des tourteaux a été déterminée par l'analyse de DPPH, et les résultats variait de 11,8 à 77,0 μmol de TE/g de tourteau. Les tourteaux provenant des essais 8 et 10 ont révélé une meilleure capacité antioxydante due à la diminution de la température de pressage utilisée dans l'extrudeur bi-vis.

III.3 Nouveaux panneaux de fibres renouvelables et biodégradables issus de tourteaux de coriandre

Comme mélanges de protéines et de fibres ligno-cellulosiques, les tourteaux de coriandre obtenus après pressage mécanique des fruits à l'aide des extrudeuses mono- ou bi-vis peuvent être considérés comme des composites naturels. Par conséquent, ils sont transformables en agromatériaux biodégradables par thermo-compression (Evon et al., 2014; Evon et al., 2010a; Evon et al., 2012; Evon et al., 2015). Cette étude avait pour but d'évaluer l'influence des conditions de thermo-pressage (température du moule, pression appliquée et le temps de moulage) sur les propriétés mécaniques, l'épaisseur de gonflement et l'absorption d'eau de panneaux de fibres fabriqués à partir d'un tourteau de coriandre produit dans une extrudeuse mono-vis.

Les onze panneaux de fibres résultant de ces conditions de thermo-pressage étaient tous solides, avec des protéines et des fibres agissant respectivement comme liant naturel et charges de renfort. Le meilleur compromis entre les propriétés mécaniques, le gonflement en épaisseur et l'absorption d'eau est représentée par le panneau de l'essai 7. Il a été produit à partir des conditions de thermo-pression suivantes: température du moule 200 °C, pression 36,8 MPa et temps de moulage appliqué de 180 secondes. En outre, la pression appliquée au cours du moulage a donné lieu à la mise en évidence d'une partie de l'huile résiduelle dans le tourteau. Cela a conduit à une diminution de la teneur en huile résiduelle à l'intérieur des panneaux de fibres (7,7% de la matière sèche pour l'essai 7, et jusqu'à 7,0% pour l'essai 10) et à une augmentation du rendement total en huile (de 47,6% après l'extrusion à 79,7% après thermo-pressage pour l'essai 7, et jusqu'à 81,2% pour l'essai 10). En conclusion, en ce qui concerne les propriétés mécaniques de l'essai 7 et sa sensibilité à

l'eau, un tel panneau de fibres serait utilisable en tant que feuille inter-couches pour les palettes, pour la fabrication de récipients ou de meubles, ou dans le secteur du bâtiment (plancher sous-couche, cloison intérieure ou dalle de plafond). En outre, le thermo-pressage ne produit pas seulement des panneaux de fibres de cohésion, mais aussi augmente considérablement l'efficacité de l'extraction de l'huile.

IV. Conclusion

- ✓ Concernant le rendement et le profil d'acides gras de l'huile végétale et de l'huile essentielle, nous pouvons réaliser que la coriandre (*Coriandrum sativum* L.) est une ressource potentielle de ressources ATOC. Selon les résultats décrits, différentes parties de coriandre (fleurs, feuilles, tiges, fruits et racines) permettent d'obtenir à la fois de l'huile essentielle et de l'huile végétale Parmi elles, le fruit sera la partie optimale pour une exploitation efficace. Cette étude indique que ces organes sont d'excellents sites pour la bioaccumulation de métabolites fonctionnels secondaires.
- ✓ Les changements de composition en acides gras de la coriandre mûre avaient également défini l'influence des stades de maturité, ce est le rapport préliminaire pour étudier l'accumulation d'huile et d'acide gras de coriandre pendant la maturation des fruits cultivés dans des conditions organiques en trois saisons (2009, 2010 et 2011). Nos résultats ont démontré que le rendement le plus élevé de l'huile a été atteint à la pleine maturité. Les profils d'acides gras varient considérablement au cours de la maturation des fruits. À des stades antérieurs, les acides gras saturés et polyinsaturés sont plus élevés et diminuent avec la maturité des fruits. L'acide pétrosélinique était le principal acide gras après le DAF 12. Ce dernier a montré une évolution inverseme de celle de l'acide palmitique qui peut soutenir une corrélation fonctionnelle entre les deux acides gras. Cette étude a fourni des données pour l'utilisation de l'huile de coriandre et concernant sa composition en acides gras pour différentes applications industrielles en raison de sa teneur en acide pétrosélinique.
- ✓ La variation de la composition en acides gras de coriandre a également défini l'influence de la région de croissance. Sur la base de ces résultats, nous pouvons voir que les différentes zones écologiques notamment la température, l'humidité et les précipitations ont un impact significatif à la fois sur la teneur en huile de la

graine et sur la composition en acides gras dans la coriandre. Par conséquent, l'emplacement de la croissance est un facteur important pour satisfaire les besoins du marché de la graine de coriandre en termes de qualité de l'huile. En dehors des conditions de croissance, il faut aussi tenir compte d'autres facteurs tels que la cultivar de coriandre, le moment de la plantation et de l'irrigation afin d'obtenir le rendement et la qualité désirés.

- ✓ La date de semis est un des facteurs importants qui influent sur la composition en acides gras et sur le rendement en huile de graine de coriandre. Les résultats sur le rendement et la composition en acides gras des différents moments d'ensemencement ont déjà mentionné ci-dessus. Il y avait des différences significatives de teneur en huile entre les trois dates, DS1 (25 mars) a montré les valeurs les plus élevées de rendement en huile avec des différences significatives par rapport au rendement de DS2 (14 avril) et DS3 (30 avril). Donc, on pourrait recommander que pour de bonnes récoltes, fin mars est la date appropriée pour la plantation de coriandre. L'effet de la date de semis sur le rendement et la présence d'acide gras est significatif. La date de semis permet de fournir aux plantes les conditions pour utiliser au maximum les paramètres environnementaux, en particulier la température et de rayonnement. Si les facteurs environnementaux sont appropriés pour l'écologisation, l'établissement et la survie des semis, le rendement maximal peut être réalisé à la date de semis appropriée. En outre, l'établissement de la date de semis appropriée sera utile pour la santé des plantes et de la surface agricole; elle est un élément fondamental d'un système agricole réussi.
- ✓ Les recherches sur l'huile végétale et la composition en huile essentielle de différentes cultivars de coriandre (sept cultivars - Canada, Tunisie, Lituanie, Algérie, France (FR1 (Dourou) - DS1), Vietnam (VS2) et Chine) ont été réalisées pour évaluer le rendement de chaque cultivar à Auch. D'après les résultats, les cultivars tunisienne et lituanienne pourraient être sélectionnées pour être plantées dans des conditions d'agriculture biologique (à Auch) en raison de leur potentiel d'exploitation. Il est nécessaire de développer des cultivars plus adaptées à la production de semences pour satisfaire la demande croissante de cette culture d'épices. La sélection d'une meilleure cultivar peut être d'un immense apport à l'éleveur pour la poursuite de l'amélioration et le développement de la culture. Par conséquent, la présente enquête a été menée afin d'évaluer les conditions

appropriées des cultivars de coriandre collectées et pour sélectionner les cultivars prometteurs pour un rendement grainier plus élevé en France.

- ✓ Pour les recherches sur la capacité d'action antioxydante, l'action antioxydante des extraits méthanoliques de sept cultivars de coriandre a été identifiée. L'action antioxydante de la coriandre était relativement élevée pour en faire une nouvelle source naturelle de substances antioxydantes, pour être utilisée comme additif dans les aliments naturels. Les cultivars de coriandre tunisienne et canadienne ont montré la plus grande quantité de polyphénols totaux et la cultivar tunisienne a montré la plus forte activité dans les essais DPPH, FRAP et ORAC. Les techniques FRAP, DPPH, ORAC étaient simples, effectués rapidement, ils seraient donc une technique appropriée pour déterminer l'action antioxydante dans les extraits de coriandre.
- ✓ Pour l'extraction de l'huile de coriandre végétale en utilisant la technologie d'extrusion mono-vis, le rendement de l'extraction d'huile dépend des conditions de fonctionnement suivantes: le diamètre de la buse et la distance buse/vis. L'augmentation du diamètre de la buse (jusqu'à 8 à 10 mm) conduit à une augmentation du rendement en huile et le rendement d'extraction d'huile maximal (soit 52%) a été obtenu à partir des conditions d'extrusion permettant la plus forte pression de la matière et la plus forte apparition d'huile, soit environ 3 mm de distance buse/vis et 9 mm de diamètre de la buse. L'acidité de l'huile pressée obtenue est faible (moins de 2%). Par ailleurs, la composition en acides gras des huiles pressées n'est pas affectée par la modification des paramètres de la machine. Elle est caractérisée par l'abondance des acides gras monoinsaturés (80%), dans laquelle l'acide pétrosélinique est le principal composé (variant de 73,5 à 75,8% en fonction de la cultivar de coriandre utilisé comme matière de départ). Enfin, les tourteaux peuvent être utilisés comme sources à la fois d'huile essentielle à haute teneur en linalol (75%) et d'antioxydants naturels. En effet, tous les tourteaux produits ont révélé une action antioxydante prometteuse, en particulier les tourteaux des cultivars tunisiennes et lituaniens. Ceci est lié à des teneurs en phénoliques totaux plus élevés dans leurs extraits méthanoliques.
- ✓ L'extraction de l'huile végétale à partir de fruits de coriandre par pressage mécanique a également été réalisée avec succès en utilisant la technologie d'extrusion bi-vis. Les conditions opératoires examinées dans cette étude (à savoir

la configuration de la vis, le coefficient de remplissage de l'appareil et la température de pressage) ont eu une influence sur le rendement en huile et de l'énergie mécanique spécifique (par unité de poids de l'huile pressée). Le contenu de pied dans le filtrat ont toujours été faible (8-18%), et les meilleurs rendements d'extraction d'huile ont été obtenus avec des à pas inverse de -33 mm, de 50 mm de longueur et positionnés dans la zone de pressage immédiatement après le module de filtration. Le rendement en extraction d'huile était au moins de 41% avec un tel profil de vis, et il a atteint 47% dans des conditions de fonctionnement du coefficient de remplissage de 39,4 g / h rpm et une température de pressage de 120 °C. La teneur en masse du pied dans le filtrat est de seulement 11% dans ces conditions et la qualité du tourteau correspondant était raisonnable, avec une teneur en huile résiduelle de moins de 17% de la matière sèche. Un meilleur appauvrissement en lipides du tourteau (15%) a été obtenu avec l'augmentation du coefficient de remplissage de l'appareil (47,1 g / h tours par minute) et la diminution de la température de pressage (80 °C), ce qui conduit à un rendement en huile de 54% basé sur la teneur résiduelle en huile du tourteau. Cependant, la teneur en pied du filtrat a été beaucoup plus importante (18%) dans ces conditions. En outre, le processus d'extrusion a systématiquement produit une huile agréablement parfumée et de bonne qualité (<1,5% d'acidité), avec une teneur en acide pétrosélinique élevée (73%). Enfin, les tourteaux peuvent être utilisés comme sources d'huile essentielle à haute teneur en linalol (77%) et/ou d'antioxydants naturels.

- ✓ Comme composites naturels, les tourteaux peuvent être transformés en panneaux de fibres biodégradables et renouvelables par thermopressage, des protéines et des fibres lignocellulosiques agissant respectivement comme liants naturels et charges de renforcement. Le meilleur compromis entre les propriétés mécaniques (c.à.d propriétés de flexion, la dureté de surface et résistance aux chocs), le gonflement en épaisseur et l'absorption de l'eau est le panneau moulé aux conditions suivantes: température du moule à 200 °C, 36,8 MPa de pression et 180 seoncdes de temps de moulage. En ce qui concerne ses propriétés mécaniques (11,3 MPa de résistance à la flexion à la rupture, 2,6 GPa module d'élasticité et 71 ° Shore D dureté de surface) et sa sensibilité à l'eau (épaisseur de 51% le gonflement et l'absorption de l'eau de 33%), un tel panneau de fibres serait utilisable comme feuilles inter-

couches de palettes, pour la fabrication de récipients ou des meubles, ou dans l'industrie du bâtiment (sol, sous-couches, les cloisons intérieures ou les carreaux de plafond). En outre, le thermo-pressage ne produit pas seulement des panneaux de fibres de cohésion, mais aussi augmente considérablement l'efficacité de l'extraction de l’huile (jusqu'à 81% pour le rendement total de l'extraction de l’huile, après l'extrusion et le thermo-pressage).

Tableau 1: Effet des conditions de configuration de vis sur l'extraction de l'huile avec extrudeur

N° de manipulation	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**
Conditions opératoires													
Diamètre de buse (mm)	5	5	5	6	6	6	7	7	7	8	8	8	9
distance buse/vis (mm)	D_1	D_2	D_3	D_1	D_2	D_3	D_1	D_2	D_3	D_1	D_2	D_3	D_1
T^oC initiale	26	27	25	27	28	26	25	25	28	24	26	25	27
T^oC finale	57	62	65	63	60	65	56	58	57	63	65	62	62
Filtrat													
Masse (g)	65.35	67.31	70.72	69.89	72.21	74.35	69.78	71.37	73.65	74.35	74.31	75.89	77.0
T_L (%)	88.54	89.45	89.69	88.25	90.17	91.28	89.01	89.62	90.77	88.66	89.75	90.89	89.5
T_F (%)	11.46	10.55	10.31	11.75	9.83	8.72	10.99	10.38	9.23	11.34	10.25	9.11	10.4
(% IA)	1.61	1.65	1.65	1.74	1.53	1.56	1.56	1.63	1.63	1.66	1.66	1.62	1.70
Tourteau													
Masse (g)	418.89	412.54	411.19	412.08	408.32	405.1	416.88	410.26	409.89	413.98	409.54	410.45	405.0
L_C (%)	15.6	15.2	14.2	16.2	15.8	14.8	15.6	15.2	15.0	16.6	16.2	15.6	16.8
H_C (%)	7.81	8.01	8.32	8.3	8.46	8.41	8.0	8.21	8.25	8.67	8.55	8.14	8.1
Rendement en huile (%)													
R_S	12.83	13.35	13.96	13.67	14.43	15.04	13.77	14.18	14.82	14.61	14.78	15.29	15.2
R_L	41.82	43.52	45.52	44.58	47.06	49.06	44.98	46.23	48.32	47.65	48.20	49.86	49.8
Rc	51.75	53.80	57.12	50.97	52.69	56.02	52.15	54.16	54.82	49.79	51.40	52.91	49.9

T_L: teneur en huile exprimée du filtrat, T_F: teneur en pied du filtrat, L_C: teneur résiduelle en huile de tourteau, H_C: rendement en huile calculé par rapport à la graine, R_L: rendement en huile calculé par rapport à l'huile que la graine exprimée, calculé par rapport à l'huile résiduelle du tourteau. IA: indice d'acide, D_1: distance buse/vis = 1 mm, D_2: buse/vis = 3 mm.

Tableau 2: Effet de la cultivar de coriandre sur l'efficacité de l'extraction de l'huile avec extrudeu

Cultivar de coriandre	**Tunisie**			**Lituanie**			**France (DS1)**			**France**	
N°de manipulation	1	2	3	4	5	6	7	8	9	10	1
Conditions opératoires											
Diamètre de buse	Diamètre de buse: 9 mm			Diamètre de buse: 9 mm			Diamètre de buse: 9 mm			Diamètre de	
distance buse/vis (mm)	D_3	D_3	D_3	D_3	D_3	D_3	D_3	D_3	D_3	D_3	D
T^oC initiale	27	27	30	31	27	28	25	25	30	26	2
T^oC finale	60	62	64	60	58	65	57	58	55	63	5
Filtrat											
Masse (g)	68.21	75.98	76.35	66.89	72.11	73.35	59.05	64.42	61.07	71.76	77.
T_L (%)	89.40	86.69	87.73	88.17	87.19	87.23	83.22	85.78	85.30	89.01	88.
T_F (%)	10.60	13.31	12.27	11.83	12.81	12.77	16.78	14.22	14.70	10.99	11.
(% IA)	1.47	1.42	1.53	1.32	1.47	1.39	2.20	1.97	1.91	1.42	1.4
Tourteau											
Masse (g)	415.08	406.91	402.56	416.08	415.33	409.56	427.68	407.68	416.2	414.33	403
L_C (%)	14.2	13.4	13.2	12.8	13.3	12.8	11.5	12.2	11.6	14.6	14
H_C (%)	8.36	8.17	8.18	8.19	8.14	8.21	7.99	8.20	8.11	8.00	8.
Rendement en huile (%)											
R_S	13.56	14.64	14.89	13.13	14.00	14.25	10.90	12.26	11.56	14.16	15.
$\overline{R_S}$	14.36 ± 0.70^a			13.79 ± 0.58^a			11.57 ± 0.68^b			14.89 ±	
R_L	55.26	59.69	60.70	55.80	59.48	60.53	39.28	44.17	41.64	46.17	49.
$\overline{R_L}$	58.55 ± 2.98^a			58.60 ± 2.48^a			41.69 ± 2.44^b			48.55 ±	
Rc	56.74	59.89	60.92	60.84	59.66	61.46	59.87	59.87	60.66	55.42	57.
$\overline{R_C}$	59.18 ± 2.17^a			60.65 ± 0.91^a			60.01 ± 0.58^a			57.62 ±	

T_L: teneur en huile exprimée du filtrat, T_F: teneur en pied du filtrat, L_C: teneur résiduelle en huile de tourteau, H_C: rendement en huile calculé par rapport à la graine, R_L: rendement en huile calculé par rapport à l'huile que la graine exprimée, calculé par rapport à l'huile résiduelle du tourteau. IA: indice d'acide, D_3: distance buse/vis = 3mm.

Tableau 3: Les résultats des expériences d'expression effectuées avec la Clextral BC 21

N° de manipulation	**1**	**2**	**3**	**4**	**5**	**6**	**7**
Conditions opératoires							
Profil de vis	3	3	3	3	4	4	4
S_S (rpm)	100	100	100	133	100	100	100
Q_S (kg/h)	3.12	4.66	6.19	6.19	3.15	3.94	4.71
C_F (g/h.rpm)	31.2	46.6	61.9	46.5	31.5	39.4	47.1
θ_{c6} (°C)	83.0 ± 0.0	79.4 ± 0.8	77.7 ± 0.6	77.9 ± 0.3	95.2 ± 0.8	90.6 ± 0.6	89.1 ± 0.
θ_{c7} (°C)	122.0 ± 4.6	119.0 ± 0.6	118.7 ± 0.5	120.4 ± 0.8	120.1 ± 0.7	118.1 ± 2.1	119.5 ± 0
Filtrat							
Q_F (kg/h)	0.31	0.51	0.49	0.70	0.38	0.52	0.59
T_L (%)	82.8	87.3	88.3	83.5	83.6	89.3	91.5
T_F (%)	17.2	12.7	11.7	16.5	16.4	10.7	8.5
H_F (%)	-	-	-	-	3.32 ± 0.05	3.60 ± 0.01	3.74 ± 0.0
L_F (% masse de sèches)	-	-	-	-	59.33 ± 1.71	63.34 ± 2.31	73.48 ± 0.
Tourteau							
Q_C (kg/h)	2.58	3.84	5.08	5.07	2.61	3.31	3.92
H_C (%)	2.16 ± 0.02	3.85 ± 0.07	4.60 ± 0.04	4.46 ± 0.06	2.32 ± 0.14	3.73 ± 0.08	4.05 ± 0.0
L_C (% masse de sèches)	17.63 ± 0.07	17.06 ± 0.05	17.62 ± 0.03	18.10 ± 0.08	16.48 ± 0.21	16.82 ± 0.59	16.06 ± 0.
Rendement en huile (%)							
R_L	32.5	38.2	28.0	37.8	40.5	46.9	45.8
R_C	42.9	45.9	44.7	43.3	46.7	45.6	48.7
L'énergie consommée							
CP (bars)	2.1 ± 0.2	3.6 ± 0.4	5.4 ± 0.5	3.3 ± 0.3	1.4 ± 0.3	1.6 ± 0.2	1.8 ± 0.4
T (%)	16.4	42.7	57.2	32.5	25.0	37.2	45.6
P (W)	180.0	467.9	626.7	473.1	274.2	407.8	499.0
SME (W h/kg)	57.7	100.4	101.3	76.5	86.9	103.5	105.9
SME' (W h/kg)	710.2	1053.8	1447.2	810.4	859.1	884.1	925.6

S_S est la vitesse de rotation des vis, Q_S le débit d'alimentation de solide, Q_F est le débit du filtrat, T_L est la teneur en huile du filtra teneur en lipides du pied, Q_C est le débit du tourteau, H_C est l'humidité du tourteau, R_L: rendement en huile calculé par rappor rendement en huile exprimée, calculé par rapport à l'huile résiduelle du tourteau. (-) Non déterminé.

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ABSTRACT

Apiaceae could be defined as Aroma Tincto Oleo Crops (ATOC), e.g. plants containing both vegetable oil and essential oil. Applying agroreffinery concept to ATOC led to propose a sequential fractionation process coupling co-extraction of vegetal oil and essential oil to a valorization of by-product residues as biosourced active molecules and substrates for designing agromaterials. The aim of this thesis is to determine the biological and technological feasability of application of the ATOC-refinery concept to coriander (*Coriandrum sativum* L.). Chapter I reports a bibliographic state of the art study on extraction and characterisation of coriander vegetal oil and essential oil while chapter II describes materials and methods setting up during the thesis for sampling, extraction, analysis and data processing. Chapter III focus on the study of major various biological parameters influencing bioaccumulation of vegetal oil and essential oil in coriander (different plant cultivars, different plant organs, different biological stages) and their impact on anti-oxidant activity of extracts obtained from extraction residues. In chapter IV, coriander fruits are processed by extrusion technology (mono screw and twin-screw extruder) in order to evaluate the feasability of mechanical pressing for extracting flavored vegetal oil. Influence of operating parameters on vegetal oil extraction yields (nozzle diameter and nozzle/screw distance (single-screw extruder) or screw configuration, device's filling coefficient and pressing temperature (twin-screw extruder)) is studied while the feasability of valorization of extraction cake as agromaterial (thermopressing) was stated.

Keywords: ATOC-refinery, coriander, vegetable oil, essential oil, single and twin-screw extruder, antioxidant, agromaterials.

RÉSUMÉ

Les apiaceae peuvent être définéies en tant qu' Aroma-Tincto-Oleo-Crop (ATOC), plantes qui contiennent à la fois une huile végétale et une huile essentielle. Appliquer le concept d'agroraffinage aux ATOC revient à proposer un procédé séquentiel alliant une co-extraction huile végétale et huile essentielle à une valorisation des résidus en tant que source de molécules biosourcées et de susbtrat pour la formulation d'agromatériaux. Les objectifs de cette thèse seront donc d'étudier la faisabilité biologique et technologique d'application du concept d'ATOC-raffinage à la coriandre (*Coriandrum sativum* L.). Le chapitre I présente l'état de l'art bibliographique sur l'extraction et l'analyse des huiles végétales et huiles essentielles de coriandre tandis que dans le chapitre II sont décrits les matériels et méthodes mis en oeuvre au cours de la thèse tant au niveau échantillonnage, extraction, analyse que traitement des données. Le chapitre III est centré sur l'étude des différents paramètres biologiques pouvant influencer la bioaccumulation des huiles végétales et huiles essentielles dans la coriandre (différentes cultivars, différents organes de la plante, différents stades de développement biologique) et leur impact sur l'activité anti-oxydante des extraits obtenus à partir des résidus d'extraction. Dans le chapitre IV, la technologie d'extrusion (mono-vis et bi-vis) a été appliquée aux fruits de coriandre dans le but d'évaluer la faisabilité du pressage mécanique du fruit de la coriandre pour l'extraction d'une huile végétale aromatisée. L'influence des conditions expérimentales sur le rendement d'extraction en huile végétale (diamètre de buse et distance buse-vis (extrudeur mono-vis) ou configuration de vis, coefficient de remplissage et température de pressage (extrudeur bi-vis)) a été étudiée tandis que la faisabilité de la valorisation du résidu solide d'extraction en agromateriaux (thermo-pressage) a été montrée.

Mots clés: ATOC-raffinage, coriandre, huile végétale, huile essentielle, extrusion mono-vis et bi-vis, antioxydants, agromateriaux.