Green Pesticides Handbook

Essential Oils for Pest Control

Edited by

Leo M.L. Nollet and Hamir Singh Rathore

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Preface

This book explores one series of biopesticides, essential oils. The number of essential oils with pesticidal possibilities or activities is large—too large to comment on all in this book. The authors made a selection of essential oils. These are discussed in depth. Other interesting properties of essential oils are also considered (in some chapters of Section III). Both editors would like to cordially thank all of the authors for their excellent work. They spent a lot of time and hard work, and the result is superb. We would also like to thank the editorial team of CRC Press/Taylor & Francis for their patience. Finally, Dr. Nollet gives a word of thanks to and expresses his appreciation for his coeditor, Professor Rathore. The idea of this book originated in his head.

Two heads are better than one.

Leo M.L. Nollet
Hamir Singh Rathore

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Editors

Leo M.L. Nollet, PhD, received an MS (1973) and PhD (1978) in biology from the Katholieke Universiteit Leuven, Belgium. He is an editor and associate editor of numerous books. He edited for M. Dekker, New York—now CRC Press/Taylor & Francis—the first, second, and third editions of the books entitled *Food Analysis by HPLC* and the *Handbook of Food Analysis*. The last edition is a two-volume book. He also edited the *Handbook of Water Analysis* (first, second, and third editions) and *Chromatographic Analysis of the Environment*, Third Edition (CRC Press). With F. Toldrá, he coedited two books published in 2006 and 2007: *Advanced Technologies for Meat Processing* (CRC Press) and *Advances in Food Diagnostics* (Blackwell Publishing—now Wiley). With M. Poschl, he coedited the book *Radionuclide Concentrations in Foods and the Environment*, also published in 2006 (CRC Press).

Dr. Nollet has coedited several books with Y.H. Hui and other colleagues: the *Handbook of Food Product Manufacturing* (Wiley, 2007); *Handbook of Food Science, Technology and Engineering* (CRC Press, 2005); *Food Biochemistry and Food Processing* (first and second editions; Blackwell Publishing [Wiley], 2006, 2012); and the *Handbook of Fruits and Vegetable Flavors* (Wiley, 2010). In addition, he edited the *Handbook of Meat, Poultry and Seafood Quality* (first and second editions; Blackwell Publishing [Wiley], 2007, 2012). From 2008 to 2011, he published with F. Toldrá five volumes on animal product–related books, namely, the *Handbook of Muscle Foods Analysis*, the *Handbook of Processed Meats and Poultry Analysis*, the *Handbook of Seafood and Seafood Products Analysis*, the *Handbook of Dairy Foods Analysis*, and the *Handbook of Analysis of Edible Animal By-Products*. Also in 2011 with F. Toldrá, he coedited for CRC Press two volumes: *Safety Analysis of Foods of Animal Origin* and *Sensory Analysis of Foods of Animal Origin*. In 2012, they both published the *Handbook of Analysis of Active Compounds in Functional Foods*. In a coedition with Hamir Rathore, the *Handbook of Pesticides: Methods of Pesticides Residues Analysis* was marketed in 2009, *Pesticides: Evaluation of Environmental Pollution* in 2012, and the *Biopesticides Handbook* in 2015.

Other finished book projects include *Food Allergens: Analysis, Instrumentation, and Methods* (with A. van Hengel; CRC Press, 2011) and *Analysis of Endocrine Compounds in Food* (Wiley-Blackwell, 2011). Dr. Nollet's recent projects include *Proteomics in Foods* with F. Toldrá (Springer, 2013) and *Transformation Products of Emerging Contaminants in the Environment: Analysis, Processes, Occurrence, Effects and Risks* with D. Lambropoulou (Wiley, 2014). In the series, CRC Food Analysis & Properties, Dr. Nollet edited with C. Ruiz-Capillas *Flow Injection Analysis of Food Additives* (CRC Press, 2015) and *Marine Microorganisms: Extraction and Analysis of Bioactive Compounds* (CRC Press, 2016).

Hamir Singh Rathore, PhD, is a retired professor and chairman, Department of Applied Chemistry, Zakir Husain College of Engineering and Technology, Aligarh Muslim University, Aligarh, India. His research work is in the area of applied/industrial chemistry, with more emphasis on synthetic inorganic ion exchangers, pesticides, and development of analytical techniques. He received MSc, MPhil, and PhD degrees from Aligarh Muslim University in 1967, 1970, and 1971, respectively. He worked as a postdoctoral fellow on biosensors with Professor Marco Mascini in the Second University of Rome, Italy, on a fellowship awarded to him in 1987 by the Third World Academy of Sciences, Trieste, Italy.

Professor Rathore taught several courses of applied chemistry to the students of BTech and MTech. After retirement, he became actively engaged in teaching modern

instrumental techniques to MSc students (industrial chemistry), writing a book, and conducting research work in the university.

His research work has been published in the form of 125 papers in journals of international repute. Twenty students have been awarded MPhil and an equal number of PhD degrees under his supervision. Professor Rathore has presented his research work and delivered lectures and invited talks in several national and international conferences held in India and abroad (United States, Italy, Spain, Finland, Hungary, Brazil, etc.). He has attended a dozen summer schools and short-term courses at different institutes, universities, and research laboratories. He is a member of several national and international scientific bodies. He is the editor or referee of some national and international journals.

Professor Rathore has contributed to the following books: *Basic Practical Chemistry* (1982) and *Experiments in Applied Chemistry* (1990), both of which were edited by Ishtiaq Ali and H.S. Rathore and published by AMU, Aligarh; *Handbook of Chromatography: Liquid Chromatography of Polycyclic Aromatic Hydrocarbons* (CRC Press, 1993), edited by Joseph Sherma and Hamir S. Rathore; and *Handbook of Pesticides: Methods of Pesticide Residues Analysis* (2009), *Pesticides: Evaluation of Environmental Pollution* (2012), and *Handbook of Biopesticides* (2014), all edited by Leo M.L. Nollet and H.S. Rathore and published by CRC Press/Taylor & Francis.

He has contributed six chapters in the following handbooks: the *Handbook of Food Analysis* (1996), *Handbook of Water Analysis* (2000), and the revised and enlarged edition of the *Handbook of Food Analysis* (2004). All three books were edited by Leo M.L. Nollet and published by Marcel Dekker.

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7

Cinnamon Oil

Khalid Haddi, Lêda R.A. Faroni, and Eugênio E. Oliveira

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7.1 Introduction

Cinnamon is a common spice that has been used for several centuries by different cultures around the world. It is obtained from different parts of a tropical evergreen tree belonging to the genus *Cinnamomum*. Various reports have dealt with the numerous properties of cinnamon and its major components not only for human health but also for agriculture applications. In this chapter, important aspects of trees from the *Cinnamomum* genus, and their products, such as botany, pharmacology, toxicology, and some end uses, with a special focus on the pesticidal potential for agriculture and indoor uses, are covered.

7.2 Botany of the Plant

The genus *Cinnamomum* (Lauraceae) includes more than 250 aromatic evergreen trees and shrubs of up to 10–20 m, primarily distributed in Southeast Asia, China, and Australia (Barceloux 2009). Investigations conducted at the beginning of the 1980s have shown that this genus has a center of diversity in south India (Ravindran et al. 2003). However, although formerly thought to be a purely Asiatic genus, *Cinnamomum* has been enriched with species such as *Phoebe*, transferred from neotropical genera based on studies and investigations carried out by taxonomists such as Kostermans. Kostermans has also defined the key characteristics for the *Cinnamomum* species identification (Kostermans 1980, 1983), leading the genus to include not only the Asiatic species but also New World ones. A very detailed botanical characterization of different species of the genus *Cinnamomum* can be found in the monograph on cinnamon and cassia written by Ravindran et al. (2003).

There are mainly four types of cinnamon:

1. True cinnamon, *Cinnamomum verum* J. Presl, also called Ceylon cinnamon, *Cinnamomum zeylanicum*, or Mexican cinnamon, *Laurus cinnamomum* L. Moderately sized (10–15 m) evergreen trees with smooth and brown branches when young. The leaves are opposite or subopposite, leathery, ovate or elliptic to broadly ovate, triplinerved with the three main nerves prominent on both surfaces. Young leaves are reddish and later turn dark green. The bark is smooth, light pinkish brown, and up to 10 mm thick. Small, pale yellow, campanulate flowers, arranged in cymes, are borne in axillary or terminal panicles. The flowering time is from October to February. The fruit is a fleshy, ellipsoid to oblong-ovoid drupe, which contains one seed and turns dark purple or black when ripe between May and June.
2. Cassia cinnamon, *Cinnamomum aromaticum* Nees, or Chinese cinnamon, *Cinnamomum cassia* J. Presl. This evergreen tree grows up to 18–20 m high. Young branches are smooth and brown, and the bark is gray to brown colored and is 13–15 mm thick when mature. The leaves are simple, opposite to subopposite, oblong lanceolate or oblanceolate, with three prominent veins. The leaves are glabrous above and with microscopic hairs below. These leaves are reddish when young and dark green when mature. The small, white flowers are borne in axillary or terminal panicles with characteristics similar to those of *C. verum*. The flowering takes place from

October to December. The fruit is a green, one-seeded, fleshy, globose drupe and turns pink-violet when mature. This fruit is similar in size to a small olive.

3. Vietnamese cinnamon, *Cinnamomum loureiroi*. Although considered for a long time as a different species, the Vietnamese cinnamon seems to be a *C. cassia*. The difference seen in the final product is a result of different harvesting processes between Vietnam and China. The confusion is believed to derive from the fact that the original *C. loureiroi*, described by Loureiro and on which he based his study, is a very rare species or even could be mislaid or lost (Dao 2003).
4. Indonesian cinnamon, *Cinnamomum burmannii*. It is a small evergreen tree, up to 15 m tall. Bark smooth, grayish brown, 2–3 mm. Leaves are opposite or subopposite, elliptical-ovate to oblong and triplinerved. They are pale red and finely hairy when young. Older leaves are glabrous, glossy green above and glaucous pruinose below. Inflorescences axillary or subterminal, slender, paniculate-cymose. Flowers are green to dark red. The fruit is ellipsoid or oblanceoloid with a pointed tip.
5. Other species include Indian cassia, *Cinnamomum tamala*, and camphor, *Cinnamomum camphora*. Indian cassia is a moderate-sized (around 8 m) evergreen tree with four morphotypes. Leaves are alternate, subopposite or opposite, glabrous, three-nerved from the base and are pink when young. Morphotypes are differentiated according to the morphology of the leave. The flowering starts from May. The fruit is slender, ellipsoid, acutish cup obconical, and fleshy, and fruits ripen between June and July. *C. camphora* is a small to medium-sized tree with small triplinerved leaves that are glabrous on both surfaces or sparsely puberulent beneath only when young. Flowers are small, yellowish-white, and similar to those of *C. verum*. They appear in April to May. The fruit is a small, purplish-black, ovate or subglobose drupe and ripens in August to November.

7.3 Methods of Extraction of Oil

Traditionally, the main products of the *Cinnamomum* genus are formed by its leaves and the dried, inner bark extracted from shoots, traded as quills, quillings, ships, and powder, and extensively used in flavoring of various dishes and processed food. But recently, interest in the value-added products, such as bark oil and leaf oil, extracted mainly from *C. verum* cinnamon bark, has been consistently growing. These oils are used in food, pharmaceutical, and perfume industries. The high value of these oils is a result of time- and effort-consuming extraction processes. The high price of these oils depends to a large extent not only on the quality of raw material used but also on the extraction process, methods, and final use of the oil.

Like most other essential oils, cinnamon oils can be extracted using a large array of techniques (Ravindran et al. 2003; Wang and Weller 2006; Tongnuanchan and Benjakul 2014; El Asbahani et al. 2015). Such techniques and methods can be classified into two broad classes: conventional and advanced methods (De Castro and Garcıa-Ayuso 1998; Huie 2002; Doughari 2012; El Asbahani et al. 2015).

The conventional methods include distillation and organic solvent extraction, while the advanced methods include a number of innovative techniques, such as supercritical

fluid extraction (SFE), subcritical extraction liquids, ultrasound-assisted extraction, microwave-assisted extraction, solvent-free microwave extraction, microwave hydrodiffusion and gravity, microwave steam distillation (MSD), and microwave steam diffusion (Doughari 2012; Dima and Dima 2015; El Asbahani et al. 2015). A subclass of the innovative methods mostly used at laboratory and microsampling analysis scales includes Clevenger distillation, microdistillation, and headspace solid-phase microextraction (Dima and Dima 2015).

7.3.1 Conventional Methods for Oil Extraction

Distillation is one of the oldest, simplest, and most widespread methods of extracting cinnamon essential oils, especially at commercial levels (Meyer-Warnod 1984; Ravindran et al. 2003; Wong et al. 2014; Dima and Dima 2015). In the cinnamon bark or leaf hydrodistillation process, water vapors are used as solvent driving, at boiling temperature, the cinnamon essential oil molecules (codistillation). The extraction device is simple and includes a heating source surmounted by a copper or steel tank, partially filled with water, where bark or leaves are added. The distillate produced from the tank passes through a precooling system to a condenser, consisting of copper tubing immersed in a large water tank, and a decanter to allow condensation and separation of essential oil and water. In some cases, there are various tanks connected to the same condenser (Ravindran et al. 2003). When the raw material is not immersed in water but maintained at a certain distance above the water surface using a grid or perforated support in a way that allows the vapor circulation from the bottom upward and across the raw material, the process is termed vapor hydrodistillation. The last variant of the distillation process is called steam distillation, and it uses two separate tanks for vapor generation and essential oil extraction. The steam produced in the first tank is introduced into the lower part of the second tank (extractor) and allowed to pass through the raw material.

Although widely used, especially in small-scale extraction units, the distillation method suffers from some drawbacks: prolonged extraction time (3–6 hours); degradation of some temperature-sensitive molecules; simultaneous extraction of other components, such as plant pigments; and environmental negative impacts (El Asbahani et al. 2015).

Together with steam distillation, solvent extraction has been widely used for the extraction of essential oils from various plant parts, including cinnamon bark and leaves. This technique uses either pure organic solvents or mixtures of them. Different solvents, including hexane, petroleum ether, methanol, propanol, methylene chloride, and ethanol, can be used for extraction (Areias et al. 2000; Pizzale et al. 2002; Kosar et al. 2003; Tongnuanchan and Benjakul 2014), but acetone is the most commonly used one. Basically, in this method, the solvent is mixed with the fine grounded plant material, heated to extract the essential oil, and then filtrated. Subsequently, the filtrate is concentrated by solvent evaporation. The resulting concentrate is a resin or a combination of wax, fragrance, and essential oil (concrete), from which the absolute essential oil is obtained using an alcohol-based distillation (Tongnuanchan and Benjakul 2014). At an industrial level, the Soxhlet extraction method is the most used among the solvent-based extraction methods. In conventional Soxhlet, the sample is placed in a "thimble" made of strong filter paper, which is placed in a thimble holder in the chamber of the Soxhlet apparatus, and gradually filled with condensated fresh solvent from a distillation heated flask. When the liquid reaches the overflow level, a siphon aspirates the solute of the thimble holder into the distillation flask in a continuous process until complete extraction is achieved (De Castro and Garcıa-Ayuso 1998; Ravindran et al. 2003).

7.3.2 Advanced Methods for Oil Extraction

Supercritical fluid extraction is one of the innovative techniques used for cinnamon oil and other essential oil extraction (Aghel et al. 2004; Khajeh et al. 2004; Braga et al. 2005; Carvalho et al. 2005; Moura et al. 2005; Fornari et al. 2012). It is using the supercritical state of a solvent fluid, usually CO_2, achieved when the temperature and the pressure of the solvent are raised above its critical value (31°C; 74 bar) (Wang and Weller 2006). Fluids reaching their supercritical state have both gas and liquid characteristics and present a density similar to that of liquids, low viscosity, and a high diffusion coefficient. Moreover, fluids like CO_2 gas are cheap and available at high purity, nontoxic and nonflammable, easily manipulated, and adjusted by varying the pressure and temperature, which is nonaggressive for thermosensitive molecules. The technique has great versatility, and the end product is virtually free from any solvent traces. In this process, the temperature and pressure of CO_2 are adjusted to reach the supercritical state; after that, the solvent is allowed to pass through finely ground raw plant material. The fluid and the dissolved compounds are transported to one or more separators for a depression step, where the CO_2 is gradually decompressed, resulting in lower solubility of the solute and leading to the separation of the solute from the solvent. Once the material is separated, the gas is compressed and recycled back to be reused again in the extraction process (Fornari et al. 2012). When water is used as the fluid, the technique is called superheated water extraction.

Although the supercritical extraction technique is considered a good tool to overcome the disadvantages of the conventional methods, it is not practiced for commercial cinnamon or cassia oil production because this technology is very costly and has little final product quality enhancement compared with solvent-extracted product (Doughari 2012; Ravindran et al. 2003).

Two other relatively newer methods used to extract cinnamon oil mainly for small-scale and laboratory analyses are ultrasound-assisted extraction and microwave-assisted extraction (Gallo et al. 2010; Gursale et al. 2010; Dvorackova et al. 2015; Sowbhagya 2016). The ultrasound-assisted extraction uses ultrasonic waves with frequency higher than 20 kHz to induce mechanical vibration leading to the destruction of cell and storage gland walls of plant material immersed in water or solvent and the release of cell contents, including essential oils (Wang and Weller 2006; El Asbahani et al. 2015). The microwave-assisted extraction uses electromagnetic radiations (frequency higher than 0.3 GHz) that can interact with cellular water and create heat, leading to cell disruption and facilitating the release of cell contents.

7.3.3 Conventional versus Advanced Methods for Oil Extraction

A continuous search for economically and ecologically sound extraction technologies as an alternative to conventional extraction methods has been growing over recent years. These novel techniques aim to overcome the drawbacks of conventional ones with respect to extraction time, solvent consumption, extraction yields and purity of the essential oil, reproducibility, and energy consumption and operating costs.

Various studies have compared two or more methods for cinnamon oil extraction considering several of the above-cited criteria. Gallo et al. (2010) analyzed the total polyphenols of different extracts of *C. zeylanicum*, and other species, using microwave-assisted and ultrasound extraction methods. Considering factors such as the extraction time and the solvent wastage, the results suggested that the microwave-assisted method was more effective than the ultrasound extraction method, as higher recoveries for *C. zeylanicum*

were obtained. The microwave-assisted method was considered effective in extracting antioxidant components from cinnamon. Using the ethanol extraction technique, Yang et al. (2012) obtained a higher yield than with the supercritical CO_2 extraction when studying the antioxidant activity of various parts of *C. cassia*, while Golmohammad et al. (2012) compared two aqueous solutions of *C. zeylanicum* bark obtained by superheated water extraction and distillation methods, followed by a solid-phase extraction method. Golmohammad et al.'s results showed that the distillation yielded a higher quantity of essential oil, but the superheated water extraction improved the purity of the oil extracted. Another recommendation of Golmohammad et al. is the use of the solid-phase extraction method as an alternative to liquid–liquid extraction for its simplicity and low cost. More recently, Wong et al. (2014) compared steam distillation with Soxhlet methods and concluded that although the quantity of oil extracted was higher with the Soxhlet method, steam distillation was the most suitable method for extracting cinnamaldehyde, as it uses a lower temperature. Dvorackova et al. (2015) concluded that the best option to extract phenolic compounds from cinnamon was the classical solvent-based extraction method, and they considered the extraction methods based on sonication and shaking to be inappropriate when they studied phenolic compounds from *C. cassia* using four different extraction methods: classical solvent, ultrasonication, maceration, and shaking.

7.4 Methods of Analysis of Oil

The important demand for essential oils by the flavor, cosmetic, health, and phytomedicine industries is leading to large quantities of cinnamon extracts, and particularly cinnamon essential oil, being produced and traded worldwide. Thus, analysis of the physical and chemical compositions of such extracts and oils is becoming a pertinent issue to ensure quality, consumer safety, and fair trade (Figueiredo et al. 1997, 2008; Do et al. 2015). It is also well known that the chemical composition of essential oils, including cinnamon oils, depends on many factors, such as growing conditions, harvest periods and techniques, drying processes, and extraction and isolation methods used. Such issues were reported earlier, in the 1970s and 1980s, as problems faced by the food industry to distinguish between commercially available cinnamons and cassia (Lawrence 1967; Archer 1988). Moreover, cases of falsification and fraud have been reported (Kubeczka 2002; Price and Price 2007; Do et al. 2015). Thus, for both consumers and chemical companies, it is necessary to determine a profile (e.g., physical, organoleptic, or chemical characteristics) of the constituents of essential oils (Dima and Dima 2015; Do et al. 2015).

Traditionally, essential oil analysis was performed to investigate their quality aspects, focusing on their purity and identity. However, with the improvements in instrumental analytical chemistry, the characterization of essential oils has allowed the scanning of a greater number of molecular constituents of essential oils. Most of the data available on cinnamon's physical and chemical composition were determined earlier by conventional methods (reviewed in Wijesekera 1977; Senanayake et al. 1978; Senanayake and Wijesekera 2003). These data have been fine-tuned with more recent and innovative methods that include gas chromatography (GC), chiral GC, isotope-ratio mass spectrometry, high-performance liquid chromatography (HPLC), high-performance thin-layer chromatography (HPTLC) analysis, vibrational spectroscopy (infrared [IR], Fourier transform infrared [FTIR], and near infrared [NIR]), and their coupled and multidimensional chromatography

variants (Jayaprakasha et al. 2002, 2003; El-Baroty et al. 2010; Jayawardena and Smith 2010; Jayaprakasha and Rao 2011; Golmohammad et al. 2012; Kamaliroosta et al. 2012; Khoddami et al. 2013; Li et al. 2013a,b; Wong et al. 2014; Dvorackova et al. 2015). Very detailed descriptions of all these methods can be found in Kubeczka (2002) and Zellner et al. (2009).

The early analytical techniques were used for physical measurements, such as relative density, optical activity, and refractive index, or melting, congealing, and boiling point determinations of cinnamon essential oil. But when combined with modern analytical techniques, such as column-based liquid chromatography and mass spectrometry, all these techniques resulted in the identification of essential compounds present in very small quantities (Ravindran et al. 2003). Chromatography is a separation procedure based on the relative affinities of the compounds to be separated toward stationary and mobile phases. The mixture of compounds to be separated is subjected to flow by mobile liquid through the stable stationary phase. Compounds with higher affinity to the stationary phase travel slower and for a shorter distance, while compounds with lower affinity travel faster and longer. The separated compounds are further identified by other techniques, like ultraviolet (UV)–visible, infrared, nuclear magnetic resonance (NMR), and mass spectroscopy. Chromatography can be planar with the stationary phase consisting of a plane surface, like in thin-layer chromatography (TLC) and paper chromatography (PC), or columnar with the stationary phase lying in the walls of a capillary tube, while the mobile phase is flushed through the column like in column chromatography, gas chromatography, and high-pressure liquid chromatography. In general, the volatile fraction of an essential oil is analyzed by GC, while the nonvolatile by liquid chromatography (LC).

Finally, organoleptic properties and nutritive and mineral values of cinnamon essential oil can be assessed with different methods. They include olfactive and sensory analyses by specialized individuals, with high risks of inconsistency deriving from variability between individuals and official methods of analysis based on standard procedures like those of the Association of Official Analytical Chemists (AOAC 2003) used by Gul and Safdar (2009) to determine the nutritive and mineral compositions of cinnamon.

7.5 Composition of Cinnamon Oil

The chemical composition of cinnamon oils varies depending on several factors that include the part of the plant used, age of trees, growing season and location, and extraction methods (Kaul et al. 2003; Rajeswara et al. 2007; Barceloux 2009; Wang et al. 2009; Paranagama et al. 2010; Geng et al. 2011; Li et al. 2013a,b; Pandey et al. 2014; Wong et al. 2014; Chakraborty et al. 2015).

One of the first detailed studies of cinnamon oil composition was carried out by Senanayake et al. (1978). Different parts of the cinnamon plant have different primary constituents: cinnamaldehyde is majorly found in bark oil, eugenol in leaf oil, and camphor in root-bark oil (Wijesekera 1977). In the European Pharmacopoeia (2008), and according to a summary report on the essential oil of cinnamon bark by the Committee for Veterinary Medicinal Products, the cinnamon bark essential oil mainly contains cinnamaldehyde (55%–76%), eugenol (5%–18%), and saffrole (up to 2%). Cinnamon bark contains up to 4% of essential oil, consisting primarily of cinnamaldehyde (60%–75%), eugenol (1%–10%), cinnamyl acetate (1%–5%) (WHO 1999), β-caryophyllene (1%–4%), linalool (1%–3%), and 1.8-cineole (1%–2%) (ESCOP 2003). Wang et al. (2009) reported that the main constituents

found in the leaves of *C. zeylanicum* are eugenol (79.75%), *trans*-cinnamaldehyde (16.25%), and linalool (0.14%).

Leela (2008) summarized the results of several authors and reached 124 and 66 different volatiles that could be found in different parts of *C. verum* and *C. cassia* plants, respectively. At least 94 volatile components have been found in cinnamon bark (Gong et al. 2004). A total of 26 compounds have been characterized from the *C. zeylanicum* flower oil, with (E)-cinnamyl acetate, *trans*-alpha-bergamotene, and caryophyllene oxide being the major compounds (Jayaprakasha et al. 2002, 2003; Jayaprakasha and Rao 2011). The oil of *C. zeylanicum* buds contains 34 compounds consisting of terpene hydrocarbons and oxygenated terpenoids, with alpha-bergamotene and alpha-copaene found to be the major compounds (Jayaprakasha et al. 2002). The volatile oil from *C. zeylanicum* fruit stalks contains more than 27 compounds, including (E)-cinnamyl acetate and (E)-caryophyllene as major compounds (Jayaprakasha et al. 2003). Geng et al. (2011) found that the majority of compounds in different parts of *C. cassia* oil belonged to the sesquiterpene hydrocarbon and oxygenated sesquiterpene fractions, with *trans*-cinnamaldehyde (33.95%–76.4%), cinnamyl alcohol acetate (0.09%–49.63%), 2-methoxycinnamaldehyde (0.09%–6.69%), and copaene (1.09%–14.3%) as major compounds.

The main components of the essential oil obtained from the bark of *C. zeylanicum* are eugenol, cinnamaldehyde, and linalool (Kubeczka 2002; Kubeczka and Formáček 2002), while *C. cassia* bark contains cinnamaldehyde, cinnamic acid, cinnamyl alcohol, and coumarin (Ranasinghe et al. 2013). Other *Cinnamomum* species were found to have lower contents of cinnamaldehyde (He et al. 2005).

Some constituents frequently encountered in cinnamon bark oil include eugenol, eugenol acetate, cinnamyl acetate, cinnamyl alcohol, methyl eugenol, benzaldehyde, cuminaldehyde, benzyl benzoate, linalool, monoterpene hydrocarbons (e.g., pinene, phellandrene, and cymene), carophyllene, and safrole. Cinnamon leaf oil also contains many of the major constituents present in cinnamon bark oil (e.g., cinnamaldehyde, cinnamyl acetate, eugenol acetate, and benzaldehyde), as well as other minor compounds, like humulene, isocaryophyllene, alpha-ylangene, coniferaldehyde, methyl cinnamate, and ethyl cinnamate (Leung and Foster 1996).

Other minor constituents also reported to be found in cinnamon essential oil include oligopolymeric procyanidins, cinnamic acid, phenolic acids, pentacyclic diterpenes, cinnzeylanol and its acetyl derivative cinnzeylanine, and the sugars mannitol, L-arabino-D-xylanose, L-arabinose, D-xylose, and α-D-glucose, as well as mucilage polysaccharides (ESCOP 2003). Several nonvolatile compounds (e.g., cinncassiols, cinnzeylanol, cinnzeylanin, anhydrocinnzeylanol, anhydrocinnzeylanin, several benzyl isoquinoline alkaloids, flavanol glucosides, coumarin, b-sitosterol, cinnamic acid, protocatechuic acid, vanillic acid, and syringic acid) have been also reported to be found in cinnamon essential oils (Leela 2008).

7.6 Physical and Chemical Properties of Oil

Essential oils are oily aromatic liquids that are soluble only in organic solvents. They are immiscible with water due to their hydrophobic nature and lower density compared with that of water. In the ISO standards lists (ISO 2003), cinnamon oil is described as the essential oil obtained by steam distillation of the leaves of *Cinnamomum zeylanicum* (Lauraceae),

growing mainly in Sri Lanka. It is a clear, mobile liquid with a light to dark amber color. It is characterized by a spice-like odor reminiscent of eugenol. At 20°C, its relative density is between 1.037 and 1.053; its refractive index is between 1.527 and 1.540; and its optical rotation ranges from –2.5° to + 2°. It shall not be necessary to use more than 2 volumes of ethanol, 70% (volume fraction), to obtain a clear solution with 1 volume of essential oil. The phenol content of cinnamon leaves oil should be between 75% and 85%.

Sri Lanka is the major world exporter of cinnamon essential oil. The Sri Lanka Standards Institution specifies that the cinnamon leaves' oil refractive index has to be between 1.530 and 1.540, its specific gravity has to be between 1.034 and 1.050, its solubility should be 1.5 volumes of 70% (v/v) ethanol at 28°C, and it should contain no less than 75% of total phenols. For the cinnamon bark oil, the values are as follows: refractive index between 1.555 and 1.580, specific gravity between 1.010 and 1.030, solubility of 1.5 volumes of 70% (v/v) ethanol at 28°C, and containing no more than 18% of total phenols for the superior special and average grade. Its content in cinnamic aldehyde was also specified for superior grade (not less than 60% m/m), special grade (55%–60%), average grade (45%–55%), and ordinary grade (30%–45%).

Leela (2008), based on the works of Baslas and Baslas (1970), reported some of the physicochemical properties of *C. verum* leaf's oil. The specific gravity ranges from 1.044 to 1.062, the refractive index is between 1.522 and 1.530, its optical rotation is 3.60, and its eugenol content is estimated at 65%–87.2%. It shall not be necessary to use more than 2 volumes of ethanol, 70% (volume fraction), to obtain a clear solution with 1 volume of essential oil. All these data were obtained at 30°C.

For the Open Chemistry Database (NCBI 2016), the physicochemical characteristics of a product named cinnamon oil are listed under the reference PubChem CID 6850781. This reference lists the synonyms of cinnamon oils as cassia oil, Chinese cinnamon, cassia bark oil, and cinnamon bark oil. The cinnamon oil is described there as a slightly water-soluble liquid that darkens and thickens on exposure to air with a molecular formula of $C_{19}H_{22}O_2$ and a molecular weight of 282.37678 g/mol. When heated to decomposition, it emits acrid smoke and irritating fumes. Its density at 20°C is 1.052–1.070 for cassia oil, 1.037–1.053 for cinnamon leaf oil, and 1.010–1.030 for cinnamon bark oil (at 25°C). The optical rotation at 20°C varies from –2.5 to +2 for cinnamon leaf oil and from –2 to 0 for cinnamon bark oil. The index of refraction at 25°C ranges from 1.6 to 1.5910 for cassia oil, 1.5730 to 1.5910 for cinnamon bark oil, and 1.53 to 1.54 for cinnamon leaf oil (20°C). The solubility at 20°C is 1 volume in 3 volumes of 70% ethanol for cassia oil, 1 volume in 2 volumes of 70% ethanol for cinnamon leaf oil, and 1 volume in at least 3 volumes of 70% ethanol for cinnamon bark oil.

The Open Chemistry Database also gives a detailed physicochemical description of the major constituents of the cinnamon oils. A summary of the physicochemical properties of cinnamaldehyde, eugenol, linalool, coumarin, and camphor are given in Table 7.1.

7.7 General Uses of Oil

7.7.1 Usage in the Ancient Periods

Cinnamon and cassia have been used since ancient times as flavoring and medicinal ingredients. Cinnamon is mentioned in the Bible as a component of the oil used by Moses

TABLE 7.1

Physicochemical Properties of Major Constituents of the Cinnamon Oils

Compound	Molecular Formula	Molecular Weight (g/mol)	Boiling Point (°C at 760 mmHg)	Melting Point (°C)	Density (g/cm³)	Refractive Index (at 20°C)	Description
Cinnamaldehyde	C_9H_8O	132.15922	253	–7.5	1.048–1.052 (25°C)	1.618–1.623	Clear yellow oily liquid with an odor of cinnamon and a sweet taste Solubility in water: 1420 mg/L (25°C) Dissolves in 1:2.5 (v/v) of 70% alcohol
Eugenol	$C_{10}H_{12}O_2$	164.20108	225	–9.2 to –9.1	1.0652 (20°C)	1.5405	Clear colorless pale yellow or amber-colored liquid; odor of cloves; spicy pungent taste; darkens and thickens on exposure to air Solubility in water: 2460 mg/L (25°C) 1 ml dissolves in 2 ml 70% alcohol
Linalool	$C_{10}H_{18}O$	154.24932	198	–	0.858–0.868 (25°C)	1.4627	Colorless liquid; odor similar to that of bergamot oil and French lavender with a good stability Solubility in water: 1600 mg/L (25°C) Soluble in alcohol, ether, fixed oils, propylene glycol; insoluble in glycerin
Coumarin	$C_9H_6O_2$	146.14274	301.71	71	0.935 (20°C)	–	Colorless crystals, flakes, or colorless to white powder with a pleasant fragrant vanilla odor and a bitter aromatic burning taste Solubility in water: 1.900 mg/L (20°C) Soluble in ethanol; very soluble in ether and chloroform
Camphor	$C_{10}H_{16}O$	152.23344	209	174–179	0.992 (25°C)	1.5462	Colorless or white crystals with a penetrating, aromatic odor Solubility in water: 1600 mg/L (25°C) 1 g dissolves in about in 1 ml alcohol, 1 ml ether, 0.5 ml chloroform

for the purpose of anointment (to make a person holy). It has also been used as an ingredient in many ancient Indian medicinal preparations. In Egypt, cassia, along with other exotic herbs, was used not only in daily life, like cooking and bathing, by the privileged classes, but also as the botanical ingredients in mummification rituals. Cinnamon was sometimes exchanged in the barter system with other goods under the Roman Empire extension.

7.7.2 Food Uses

Cinnamon is used as a spice, a condiment, and flavoring material principally in cookery; chocolate preparation, especially in Mexico; many dessert recipes, such as apple pie, doughnuts, and cinnamon buns, as well as spicy candies; coffee; tea; hot cocoa; and liqueurs. In the Middle East, in Turkish and Persian cuisine, cinnamon is often used in chicken and lamb meat dishes and in a variety of thick soups, drinks, and sweets.

Cinnamon is an excellent spice used with meat and poultry in Indian and Moroccan dishes. It is an essential part of the curry pastes used across Asia. It is also used, along with other spices, in pickles, sauces, soups, confectionaries, and canned fruits. Cinnamon is a popular flavoring in numerous alcoholic beverages, such as "cinnamon liqueur," which is popular in Europe (Willard 2013). Krishnamoorthy and Rema (2003) reported that cinnamon bark oil is used frequently in the food, pharmaceutical, and perfume industries. It has largely replaced cinnamon powder in the processing industry, since it can be measured accurately according to well-established replacement ratios for ground spice using oils and oleoresins, such as the ones elaborated by Tainter and Grenis (1993).

7.7.3 Medicinal Uses

Cinnamon and cassia are believed to have a broad spectrum of medicinal and pharmacological applications. In folk medicine, cinnamon is used for the treatment of impotence, frigidity, dyspnea, eye inflammations, leukorrhea, vaginitis, rheumatism, and neuralgia, as well as wounds and toothaches (WHO 1999). In African and Chinese pharmacopoeias and traditional systems of medicine, cinnamon is indicated for the treatment of dyspeptic conditions, including mild spastic conditions of the gastrointestinal tract, fullness and flatulence, and loss of appetite. Cinnamon is also known to be a carminative, expectorant, and antidiarrheal, and to be useful for bronchitis, itching, and urinary disease (Leela 2008). Cassia is traditionally used for digestive problems, such as flatulence, colic, dyspepsia, diarrhea, and nausea, as well as colds, influenza, fevers, arthritis, and rheumatism (Barceloux 2009). Recent pharmacological studies have shown that besides its role as a spice, cinnamon can be used as a hypoglycemic and cholesterol-lowering (Khan et al. 2003), wound pro-healing (Kamath et al. 2003), and anti-inflammatory compound (Chao et al. 2005). It is a risk-reducing agent for colon cancer (Wondrak et al. 2010) and can prevent bleeding due to its anticoagulant properties (Husain and Ali 2013). Several studies have reported the anti-inflammatory activity of cinnamon and its essential oils (Sosa et al. 2002; Li et al. 2003; Matu and Staden 2003; Chao et al. 2005; Tung et al. 2008, 2010). Cinnamaldehyde, the major compound of cinnamon, exhibited anti-inflammatory activity by inhibiting the activation of the nuclear factor kappa-light-chain enhancer of activated B cells (Reddy et al. 2004; Lee and Balick 2005). Other constituents of cinnamon oil belonging to the flavonoid group have been demonstrated to possess anti-inflammatory activities (Kim et al. 2004; Stoner and Wang 2013).

7.7.3.1 Antioxidant Properties

Cinnamon bark has been shown to contain very high concentrations of antioxidants (Dragland et al. 2003). The higher antioxidant activities of cinnamon, compared with those of other spices, have been previously described (Murcia et al. 2004; Shan et al. 2005). Considerable antioxidant activities of various extracts of cinnamon have been reported by Mancini-Filho et al. (1998), and their inhibition of fatty acid oxidation and lipid peroxidation was demonstrated *in vitro* (Shobana and Naidu 2000). Singh et al. (2007) evaluated the antioxidant potential of cinnamon volatile oils and oleoresins of leaf and bark and their major components by comparing their lipid inhibitory activities with selected antioxidant activities and concluded that the volatile oils and oleoresins of cinnamon leaf and bark have good antioxidant properties. Yang et al. (2012) compared the antioxidant activities of extracts of various parts of *C. cassia* (barks, buds, and leaves) obtained by supercritical carbon dioxide extraction and ethanol extraction and showed that the extracts of *Cinnamon* barks exhibited higher antioxidant activity than other parts of cinnamon. Moreover, Yang et al. (2012) also demonstrated that ethanol is the best solvent to obtain the main antioxidant constituents.

Etheric, methanolic, and aqueous cinnamon extracts also inhibited oxidative processes *in vitro* (Dhuley 1999; Mathew and Abraham 2006; Lin et al. 2007). Several *in vitro* studies have demonstrated the antioxidant effects of the essential oil obtained from the bark of *C. zeylanicum* and its main components (Lee et al. 2002; Jayaprakasha et al. 2003; Chericoni et al. 2005; Lee and Balick 2005). Studying the free radical–scavenging activities of various medicinal plants, Okawa et al. (2001) concluded that different flavonoids extracted from cinnamon have good antioxidant properties. The study of Lee et al. (2002) showed that cinnamaldehyde and other compounds of cinnamon have inhibitory activities against the production of nitric oxide. *In vivo* study carried out by Lin et al. (2003) showed that the ethanolic extract of *C. cassia* exhibited significant antioxidant activity compared with the natural antioxidant α-tocopherol. El-Baroty et al. (2010) found that cinnamon essential oil exhibited appreciable *in vitro* antioxidant activity. Volatile oils of *C. zeylanicum* showed significant antioxidant activities, as reported by Jayaprakasha and Rao (2011).

7.7.3.2 Hypoglycemic Properties

Pharmacological studies, on human and animals both *in vitro* and *in vivo*, have recently been trying to show that cinnamon may play a possible role in improving glucose and insulin metabolism (Imparl-Radosevich et al. 1998; Onderoglu et al. 1999; Broadhurst et al. 2000; Kar et al. 2003; Khan et al. 2003), and that cinnamaldehyde may have a potential role as an antidiabetic agent with contrasting results.

In his analysis of randomized controlled trials including more than 500 patients, Allen et al. (2013) established that when taken in a dose ranging from 0.12 to 6.0 g/day for approximately 4 months, cinnamon contributed to a statistically significant decrease in the levels of fasting plasma glucose, coupled with an improvement in the lipid profile. The same conclusions were reached by Alanazi and Khan (2015) in their meta-analysis of 16 randomized control trials with 638 patients, where they concluded that the consumption of cinnamon is associated with a statistically significant decrease in the levels of fasting plasma glucose, total cholesterol, and triglyceride. However, the high degree of heterogeneity in the studies analyzed may limit the ability to apply these results to patient care.

Regarding the cinnamon compound mechanism of action in diabetes, Sheng et al. (2008) explained that its role in insulin resistance derived from the increased expression

of peroxisome proliferator-activated receptors (PPARs) α and γ. Moreover, the effect of cinnamon on PPAR γ was found to be analogous to that of the thiazolidinediones in type 2 diabetes (Rafehi et al. 2012). A different role played by *C. cassia* in mitigation of insulin resistance was advanced by Jitomir and Willoughby (2009) and consisted of the enhancement of expression of insulin-sensitive glucose transporters by acting on the phosphorylation of signaling proteins. Finally, cinnamon has also been reported to have an insulin mimetic and insulin-sensitizing action (Howard and White 2013). A good review of potential mechanisms of action may be found in Medagama (2015).

7.7.3.3 Other Medicinal Uses

Cinnamon is also frequently used as flavor in chewing gums due to its effects and ability to remove bad breath, and it has been traditionally used as tooth powder and for dental problems such as toothaches and bad breath (Aneja et al. 2009; Jakhetia et al. 2010; Gupta et al. 2012). The active component cinnamaldehyde is said to be cardioprotective (Song et al. 2013) and has a vasorelaxative effect (Alvarez-Collazo et al. 2014). A systemic review of previous studies has suggested that cinnamon-supplemented diets can result in a significant fall in blood pressure (Wainstein et al. 2011; Akilen et al. 2013).

Much research has been done to see the effect of cinnamon on melanoma cells, and the results of a study suggested that *C. cassia* can inhibit the survival, viability, and proliferation of tumor cells *in vitro* without having a significant effect on the normal cells (Han et al. 2004). *C. cassia* bark extracts also effectively inhibited the virus-induced cytopathogenicity in MT-4 cells infected with HIV (Premanathan et al. 2000). Cinnamon, cinnamon extracts and essential oils, and constituents of cinnamon, such as monoterpenoids and cinnamaldehyde, have all been reported to exhibit anticancer, antitumor, antiproliferative, and antimutagenic effects (Shaughnessy et al. 2006; Bhattacharjee et al. 2007; King et al. 2007; Wu and Ng 2007; Duessel et al. 2008; Dong et al. 2009; Lin et al. 2009; Sharififar et al. 2009).

7.8 Pesticidal Uses of Cinnamon Essential Oil

7.8.1 Antibacterial Properties

Cinnamon oils have been widely studied for their antimicrobial effects on various bacteria (Hili et al. 1997; Chao et al. 2000; Matan et al. 2006; Shan et al. 2007; Singh et al. 2007; Abdollahzadeh et al. 2014). Cinnamon oils and extracts, as well as their major components cinnamaldehyde and eugenol, have been found to exhibit antimicrobial effects on both gram-positive and gram-negative bacteria such as *Salmonella enterica*, *Escherichia coli* (Friedman et al. 2004), and *Listeria monocytogenes* (Yuste and Fung 2002). El-Baroty et al. (2010) found that cinnamon essential oil exhibited a strong antibacterial activity against *Bacillus subtilis*, *Bacillus cereus*, *Staphylococcus aureus*, *Streptococcus faecalis*, and *Micrococcus luteus* and gram-negative bacteria *Alcaligenes faecalis*, *Enterobacter cloacae*, *Pseudomonas aeruginosa*, *Klebsiella pneumoniae*, and *Serratia marcescens* (Chao et al. 2000). Cinnamon bark oil and its major components showed antibacterial effects on the major respiratory and gastrointestinal tract pathogens *Haemophilus influenzae*, *Streptococcus pneumoniae*, *Streptococcus pyogenes*, and *S. aureus* (Inouye et al. 2001a,b). Furthermore, when incorporated in biofilms, cinnamaldehyde has also been reported to have negative effects on *E. coli* and *Pseudomonas*

spp. (Niu and Gilbert 2004), *Burkholderia* spp. (Brackman et al. 2009), uropathogenic *E. coli* (Amalaradjou et al. 2010), *Vibrio* spp. (Brackman et al. 2011), methicillin-resistant *S. aureus* and *Staphylococcus epidermidis* (Jia et al. 2011; Kavanaugh and Ribbeck 2012), *Candida* spp. (Khan and Ahmad 2012), *Listeria* spp. (Upadhyay et al. 2013), *Salmonella* spp. (Zhang et al. 2014), *S. pyogenes* (Shafreen et al. 2014), and *P. aeruginosa* (Kim et al. 2015). Ouattara et al. (2000) reported that incorporating cinnamaldehyde into chitosan film reduced the growth of *Lactobacillus sakei*, *Serratia liquefaciens*, and Enterobacteriaceae on the surface of meat products.

Antibacterial activities of cinnamon bark oil and cinnamaldehyde have been attributed to considerable alterations in the structure of cell envelopes (Di Pasqua et al. 2007). The membrane permeability may be affected by an inhibition of energy generation, probably due to the inhibition of glucose uptake or utilization of glucose (Gill and Holley 2004). The cinnamaldehyde of the biofilm is partially caused by the downregulation of quorum-sensing systems (Kim et al. 2015).

The antibacterial actions of natural extracts of cinnamon have been suggested to be a relevant tool in the control of pathogens of aquatic animals. In their investigations, Yeh et al. (2009) demonstrated that shrimp treated with natural extracts of cinnamon exhibited an enhanced disease resistance to *Vibrio alginolyticus.*

7.8.2 Antifungal Properties

Based on several *in vivo* and *in vitro* studies, cinnamon essential oils and its major components have been found to exhibit significant inhibitory effects against several fungi, including *Coriolus versicolor*, *Laetiporus sulphureus*, *Eurotium* spp., *Aspergillus* spp., and *Penicillium* (Chipley and Uraih 1980; Cao 1993; Mastura et al. 1999; Guynot et al. 2003; Simić et al. 2004; Cheng et al. 2006). *trans*-Cinnamaldehyde, a component in the oil of *C. zeylanicum*, was the most active compound against 17 micromycetes (Simić et al. 2004). The essential oils of several *Cinnamomum* species showed anticandidal and antidermatophytic activity *in vitro* (Lima et al. 1993; Mastura et al. 1999). Quale et al. (1996) reported that the use of *C. zeylanicum* allowed overcoming the resistance to fluconazole in *Candida* isolates. Singh et al. (2007), using several methods to study the antifungal efficacy of cinnamon essential oil and its oleoresin, reported that the volatiles elicited from the essential oils extracted from cinnamon leaves were found to be 100% antifungal against *Aspergillus niger*, *Aspergillus terreus*, *Fusarium moniliforme*, *Fusarium graminearum*, *Penicillium citrinum*, and *Penicillium viridicatum*, but not against *A. ochraceus* and *A. terreus*. The leaf oleoresin showed complete mycelial zone inhibition for *P. citrinum*, and volatiles elicited from the essential oils extracted from cinnamon barks showed complete inhibition against fungi such as *F. graminearum*, *F. moniliforme*, *P. citrinum*, *P. viridicatum*, and *A. terreus* (Singh et al. 2007). Moreover, Singh et al. (2007) also suggested that among cinnamon oil constituents, cinnamaldehyde possessed the best antifungal activity. El-Baroty et al. (2010) found that cinnamon essential oil has a strong antifungal activity against four fungal strains: *A. niger*, *Penicillium notatum*, *Mucora heimalis*, and *Fusarim oxysporum*. Cinnamon oils and extracts showed good antifungal activities against important plant diseases. Wilson et al. (1997) found that among 49 essential oils tested, *C. zeylanicum* demonstrated a great antifungal activity against *Botrytis cinerea*, while Montes-Belmont and Carvajal (1998) reported that *A. flavus* was totally inhibited with *C. zeylanicum*. In other studies, *C. zeylanicum* was fungicidal against pathogens isolated from banana, including *Colletotrichum musae*, *Lasiodiplodia thebromae*, and *Fusarium proliferatum* (Ranasinghe et al. 2002); exerted antifungal activity toward *Oidium murrayae* (Chu et al. 2006); and inhibited conidial germination

of *Colletotrichum gloesporioides* (Barrera-Necha et al. 2008). In *in vitro* experiments, it was found to have a good mycelial inhibition of the corn rot *F. oxysporum* f.sp. *gladioli* (Barrera-Necha et al. 2009), to be highly effective against the growth of *Rhizoctonia solani* (Nguyen et al. 2009), and to have an excellent antifungal activity against early blight of tomato *Alternaria solani* (Yeole et al. 2014). The investigations of Wang et al. (2014) showed that cinnamon microemulsions had high *in vivo* control activity against gray mold of pears *Botritys cinerea.*

7.8.3 Insecticidal Properties

7.8.3.1 Against Vector of Human Diseases

As many other essential oils, cinnamon essential oils offer great potential in medical entomology, especially against mosquitoes, which represent one of the most relevant vectors of human diseases. They have been shown to be effective larvicides against mosquitoes (Cheng et al. 2004, 2009; Chang et al. 2006). Larvicidal tests demonstrated that the components of leaf essential oils, such as cinnamaldehyde–cinnamyl acetate and cinnamyl alcohol, had an excellent inhibitory effect against the fourth-instar larvae of the yellow fever mosquito *Aedes aegypti* (Cheng et al. 2004; Chang et al. 2006). Results of mosquito larvicidal assays also showed that the most effective constituents in leaf essential oils were cinnamaldehyde, eugenol, anethole, and cinnamyl acetate. Cinnamon has also shown excellent repellency in tests conducted on blood-starved females of *Ae. aegypti* mosquitoes (Chang et al. 2006). Reviewing the literature of mosquito larvae control using botanical larvicides, Pavela (2015) concluded that from 122 initially studied plant species, 3 *Cinnamomum* species were among the 7 most significant botanical larvicides that may be considered suitable sources for substances to control mosquito larvae.

7.8.3.2 Against Agricultural Insect Pests

Cinnamon oils and its components, such as cinnamaldehyde, are well-known insecticidal compounds that have been studied against a variety of other insects (Huang and Ho 1998; Lee et al. 2001; Chang and Cheng 2002; Lee et al. 2008). The antitermitic activities of the essential oils from the leaves of *C. osmophloeum* and its chemical ingredients against the Formosan subterranean termite *Coptotermes formosanus* were investigated by direct contact application (Chang and Cheng 2002). Results have demonstrated that the indigenous cinnamon leaf essential oil has a good effective antitermitic activity, and that cinnamaldehyde, eugenol, and α-terpineol extracted from indigenous cinnamon leaf essential oil are responsible for the high antitermitic effectiveness. Cheng et al. (2008) described that the leaf essential oil of *C. osmophloeum* exhibits effective toxicity in both open and closed exposure against red imported fire ants, with *trans*-cinnamaldehyde as the major component in the essential oil playing the key role in controlling the red imported fire ant *Solenopsis invicta*. Park et al. (2000), using a fumigation test, found that the *Cinnamomum* bark–derived compounds were much more effective against larvae of the oak nut weevil *Mechoris ursulus* in closed cups than in open ones, indicating that the insecticidal activity of tested compounds was attributable to fumigant action. Park et al. (2000) concluded that the *Cinnamomum* bark–derived materials could be useful as a preventive agent against damage caused by *M. ursulus*. Ovicidal activity of *C. zeylanicum* oil was reported for the rice moth *Corcyra cephalonica* (Bhargava and Meena 2001). Passino et al. (1999) reported insecticidal activity of *C. zeylanicum* oil against the Mediterranean fruit fly *Ceratitis capitata*.

7.8.3.3 Against Stored Product Pests

In stored product pests, the susceptibility of the rice weevil *Sitophilus oryzae* to fumigant actions of cinnamon oil was investigated, along with other essential oils, and resulted in 100% mortality within 1 day of treatment in closed containers (Kim et al. 2003). In a different study, Paranagama et al. (2003) concluded that *C. zeylanicum* leaf essential oils can be used as stored paddy rice protectant, as it kept the samples studied free of *S. oryzae* and the angoumois grain moth *Sitotroga cerealella*, two major pests of stored grains, without altering the quality of the stored rice.

C. cassia and its major constituent, cinnamaldehyde, exhibited fumigant toxicity and residual effects against *S. oryzae* (Lee et al. 2008), and *C. cassia* vapor caused the highest mortality of various life stages of the red flour beetle *Tribolium castaneum* (Mondal and Khalequzzaman 2009). The insecticidal effects of cinnamon oil and other essential oils were evaluated by Karahroudi et al. (2010) for three of the most important stored product pests of the Indian mealmoth *Plodia interpunctella*, the confused flour beetle *Tribolium confusum*, and the pulse beetle *Callosobruchus chinensis*, and the results indicated that cinnamon essential oil has good fumigant activity against the three tested species.

In their study, Jumbo et al. (2014) evaluated the insecticidal (e.g., lethal toxicities, disturbances on reproductive traits, and persistence of action) and repellent activities of cinnamon, *C. zeylanicum*, and clove, *Syzygium aromaticum*, essential oils on the bean weevil *Acanthoscelides obtectus* in a nonfumigant manner. Jumbo et al. (2014) concluded that cinnamon not only has a good insecticidal activity but also significantly reduced the bean weight losses caused by *A. obtectus*. Similar results were reported by a study on *C. maculatus* and *S. oryzae* (Brari and Thakur 2015), where the essential oil of *C. zeylanicum* and its two components (cinnamaldehyde and linalool) were found to exhibit contact and fumigant toxicity against the adults of both insect species tested.

7.8.3.4 Against Medical–Veterinary Insect Pests

In veterinary area, the application of camphor and cinnamon oils to water buffalo, at concentrations similar to the ones used in the laboratory studies, resulted in a large decline in numbers of the ungulate lice *Haematopinus tuberculatus* up to 6 days after application (Khater et al. 2009). The same study reported a decrease in a number of three fly species (*Stomoxys calcitrans, M. domestica*, and *Hippobosca equina*) on treated cattle. Results also indicated that the essential oils from cinnamon and its most predominant compound had high ovicidal activity against various harmful flies (Shen et al. 2007).

In veterinary use, although anthelmintic effects of Ceylon cinnamon were reported as early as in the 1950s by Cavier (1950), it is only recently that Williams et al. (2015) showed for the first time that cinnamon bark has anthelmintic potential *in vitro* using swine nematode *Ascaris suum*, and this derives both from its proanthocyanidin tannins and most notably from *trans*-cinnamaldehyde. However, their *in vivo* experiments with pigs and poultry made them reach a conclusion that for the potential of *trans*-cinnamaldehyde to be used as an anthelmintic against intestinal helminthes, appropriate formulations to stabilize and protect the compound will likely be necessary.

7.8.4 Acaricidal Effects

Cinnamon displayed acaricidal activity against the poultry red mite *Dermanyssus gallinae* (Kim et al. 2004). In investigations conducted by Bahadon and Azarhoosh (2013), it was

demonstrated that plant preparations from *C. cinnamon* and other aromatic plants can be used for controlling *D. gallinae*. On the basis of LC_{50} values, essential oil extracted from cinnamon leaves was one of the most active oils among 24 Tai herbal oils tested against house dust mites *Dermatophagoides pteronyssinus* (Veeraphant et al. 2011), which was attributed to be due to its eugenol content (Veeraphant et al. 2011). By evaluating the acaricidal and repellent effects of cinnamon essential oil on the house dust mite, *Dermatophagoides farina* and *D. pteronyssinus*, Oh (2011) demonstrated that cinnamon bark essential oil was a very effective acaricide at the concentration of 0.125 μl; it had good repellent effect when used at the concentration of 0.094 μl.

The results of dose–mortality experiments, carried out by Shen et al. (2012) to test the effects of *trans*-cinnamaldehyde (a component of cinnamon essential oil) on the common worldwide parasite of rabbits, the rabbit ear mite *Psoroptes cuniculi*, indicated that this compound had a good killing activity against *P. cuniculi* adults, and that *trans*-cinnamaldehyde can be considered as a promising agent for mite control. Similar results were already reported by Fichi et al. (2007), as he showed cinnamon leaf to have high levels of acaricidal efficacy against *P. cuniculi* in rabbits at concentrations of 2.5%.

However, the mechanism of cinnamon oils' and extracts' acaricidal activity is not yet well understood. Ellse and Wall (2014) suggest that the acaricidal efficacy of essential oils may be linked to the vapor pressure to which the mites are exposed, as it affects the concentration of volatile. Vapor assays conducted by Na et al. (2011) using 34 compounds extracted from *Cassia* spp. and *Cinnamomum* spp. showed that α-methyl-E-cinnamaldehyde and E-cinnamaldehyde had acaricidal efficacy comparable to that of the chemical acaricide dichlorvos.

7.8.5 Nematicidal Effects

Park et al. (2005) reported nematicidal activity of plant essential oils, including *C. verum* and its components, against the pine wood nematode *Bursaphelenchus xylophilus*. Analyzing the activity of 88 commercial essential oils against mixed-stage of *B. xylophilus*, Kong et al. (2006) identified highly active *C. zeylanicum* bark essential oils showing nematicidal activity that proved to be higher than those obtained for some commercial synthetic nematicides, such as fenitrothion.

7.8.6 Repellency Effects

In investigations conducted by Prajapati et al. (2005), the essential oil of *C. zeylanicum* proved to be oviposition deterrent and repellent against three mosquito species tested (*Anopheles stephensi*, *Ae. aegypti*, and *Culex quinquefasciatus*). Yang et al. (2004) investigated the repellent activity of methanol extracts and steam distillate from 23 aromatic medicinal plant species against female blood-starved *Ae. aegypti*, and they found that at a dose of 0.1 mg/cm^2, the repellency of extracts of *C. cassia* bark and *C. camphora* steam distillate was comparable to that of deet. The duration of the effectiveness for extracts from *C. cassia* bark was comparable to that of deet.

Laboratory studies suggested that cinnamon may be useful as an insect repellant (Cloyd et al. 2009). Hori (2003) described *C. cassia* as having repellent activity against the cigarette beetle *Lasioderma serricorne*, while in the study of Jumbo et al. (2014), cinnamon oil exhibited repellent actions against *A. obtectus*, in accordance with other investigations, such as the one carried out by Liu et al. (2006), and where they described a good repellent activity exhibited by essential oil from the seeds of *C. camphora* against storage pests *S. oryzae* and

Bruchus rugimanus. In addition, cinnamon essential oils displayed repellant action against the red bud borer *Resseliella oculiperda* (Van tol et al. 2007).

Hanifah et al. (2012) reported that *C. zeylanicum* showed the highest repellency rate compared with the other plants extracts in a study carried out to evaluate the repellency of six plant extracts against the larval stage of *Leptotrombidium deliense*, the mite vector of scrub typhus.

7.8.7 Herbicide Effects

In a laboratory and greenhouse experiments with essential oils from different plants (including cinnamon), Tworkoski (2002) tried to determine the herbicidal effect of plant-derived oils and identify the active ingredient with herbicide activity. Essential oils in aqueous concentrations from 5% to 10% (v/v) added of two adjuvants (nonionic surfactant and paraffinic oil blend at 0.2% [v/v]) were applied to shoots of the common lambsquarters *Chenopodium album*, the common ragweed *Ambrosia artemisiifolia*, and johnsongrass *Sorghum halepense* in the greenhouse; shoot death occurred within 1 hour to 1 day after application. Essential oil (1%, v/v) from cinnamon was one of the most phytotoxic and caused electrolyte leakages, resulting in cell death. Eugenol (one of the major components of cinnamon) was confirmed to be the active ingredient in the essential oil of cinnamon.

Campiglia et al. (2007) evaluated and compared the inhibition effect exerted by the essential oils of cinnamon, peppermint *Mentha* × *piperita* L., and lavender *Lavandula* spp. on seed germination of some of the most common weed species of the Mediterranean environment, like pigweed *Amarantus retroflexus* L., wild mustard *Sinapis arvensis* L., and ryegrass *Lolium* spp. Their results highlighted a control in the weed germination, with cinnamon oil exhibiting the highest inhibition effect compared with lavender and peppermint ones. The dicotyledonous species have been more susceptible to the cinnamon oil inhibition of seed germination than the monocotyledonous ones.

Under controlled and semicontrolled conditions (laboratory and greenhouse), Cavalieri and Caporali (2010) studied the allelopathic effects of essential oil extracted from *C. zeylanicum* on the seed germination of seven Mediterranean weed species (i.e., redroot pigweed *A. retroflexus* L., black nightshade *Solanum nigrum* L., common purslane *Portulaca oleracea* L., common lambsquarters *C. album* L., wild mustard *S. arvensis* L., ryegrass *Lolium* spp. and common vetch *Vicia sativa* L.). Cinnamon oil showed drastic inhibitory effects, and in a semicontrolled condition, the 345.6 mg/L concentration of cinnamon essential oil totally inhibited the seed germination of *A. retroflexus* L. (Cavalieri and Caporali 2010).

7.9 Advantages as a Pesticide

The use of essential oils as pesticides has drawn a large and continuous interest, exemplified by the very high number of studies dealing with extraction, chemistry, toxicology, and uses of essential oils (Ravindran et al. 2003; Bakkali et al. 2008; Barceloux 2009; El-Baroty et al. 2010; Doughari 2012; Rao and Gan 2014; Tongnuanchan and Benjakul 2014; Chakraborty et al. 2015; Dima and Dima 2015; El Asbahani et al. 2015; Medagama 2015). Cinnamon essential oils are no exception to this rule. This wide interest found its basis in the problems faced with chemical pesticides, such as risk to human health and environment, pest resurgence, secondary pest problems, and the development of resistance, as well as the

concerns expressed by consumers and pressure groups about the safety of pesticide residues in food. The essential oils, including cinnamon oils, are considered natural, and their use as spices and in food industry is often regarded as sufficient evidence of their safety. Pesticide products containing certain of these essential oils are exempt from toxicity data requirements by the U.S. Environmental Protection Agency (USEPA).

Essential oils and their main constituents are generally regarded as safe products owing to this positive perception to their very low mammalian toxicity. The estimated oral intake LD_{50} for rat is 1.160 mg/kg (body weight) for cinnamaldehyde, and it is 500 mg/kg for eugenol (Shivanandappa and Rajashekar 2014) and 2790 mg/kg for linalool (OECD 2002), representing very low mammalian toxicity values compared with other insecticides. In the fact sheet of cinnamaldehyde issued by the USEPA, it is written, "Cinnamaldehyde is Generally Recognized As Safe (GRAS) by the Flavoring Extract Manufacturers' Association and is approved for food use by the Food and Drug Administration. Cinnamon oil, which contains 70% to 90% cinnamaldehyde, is also classified as GRAS, and, like cinnamaldehyde, is used in the food and flavoring industry" (USEPA 2015).

This mammalian selectivity was partially attributed to the mode of action of various components of essential oils like eugenol (Enan et al. 1998). In fact, Enan et al. (1998) demonstrated that a number of essential oil compounds act on the octopaminergic system of insects. For instance, eugenol was found to mimic octopamine in increasing intracellular calcium levels in cloned cells from the brain of *Periplaneta americana* and *Drosophila melanogaster* (Enan 2005). Altering the functioning of octopamine by a compound like eugenol results in total interruption of nervous system functioning in insects, which makes the octopaminergic system of insects a sound and rational target for insect control, as these receptors are not found in vertebrates.

Essential oils such as cinnamon oils are a mixture of many biosynthetically diverse compounds and analogs. This characteristic diversity is related not only to the age and developmental stage of the part source of the oil, but also to the growing conditions (Regnault-Roger et al. 2012). In the case of cinnamon oil, the cinnamaldehyde is acting on the energy production system, possibly interfering with glucose uptake or utilization, and besides eugenol acting on the octopaminergic system, linalool, a frequently reported monoterpenoid in cinnamon oil, has been demonstrated to act on the nervous system, affecting ion transport and the release of acetylcholine esterase in insects (Re et al. 2000). Finally, synergistic effects between the components of essential oils have been reported by previous studies (Berenbaum 1985; Miresmailli et al. 2006; Joffe et al. 2012; Koul et al. 2013; Faraone et al. 2015; Omolo et al. 2005). This indicates that the effect of the major components needs synergism from secondary constituents in the essential oils. This mixture of compounds with various sites of action and synergized effects between the essential oil constituents may be behind the improved efficacy of essential oils as insecticides and is surely playing a crucial role as a barrier for resistance development.

The essential oils and their components are generally considered safe for the environment, as they are majorly nonpersistent (Isman 2000). The major components of cinnamon oil are also nonpersistent, and hence have little impact on the environment when used as pesticides. The USEPA reports that because cinnamaldehyde is not soluble in water and rapidly degraded in the soil, it is not expected to pose any hazard to nontarget organisms (USEPA 2015). Eugenol is anticipated to be short-lived in the environment, and is rapidly dissipated and degraded via volatilization and atmospheric decomposition (Marin Municipal Water District 2008). Most linalool, both natural and synthetic, is released to the atmosphere, where it is rapidly degraded abiotically with a typical half-life below 30 minutes (OECD 2002). In water, linalool is readily biodegraded under both aerobic and

anaerobic conditions; the same is predicted for soil and sediment. This nonpersistent characteristic is linked to the susceptibility of the essential oil and its compounds to temperature and UV light degradation, resulting in short residual activity with shorter restriction intervals for the treated areas (Miresmailli and Isman 2006).

Historically, aromatic plants and plant extracts were widely used for insect control in traditional agricultural systems in many developing countries. Using techniques as easy and affordable as steam distillation, plants products such as essential oils may be affordable for small farmers under the conditions of improving safety use and knowledge about both the accurate compositions and the pesticidal activities.

7.10 Limitations as a Pesticide

Essential oils such as cinnamon oil have been intensively studied for their pesticidal activities and have been described as a sustainable, effective, and affordable alternative to chemical insecticides. However, such oils still face various challenges regarding their use as pesticides. Various reports have described human toxicity cases of cinnamon and cinnamon oil involving local irritation and allergic reactions (Barceloux 2009). Because of its skin-sensitizing property, the use of bark oil in perfume and cosmetic industry is very limited (Ravindran et al. 2003), and occupational allergic contact dermatites, although rare (Kanerva et al. 1996), have been reported among workers with cinnamon (Kanerva et al. 1996).

The cinnamon essential oil biological activities are linked mainly to their major components: cinnamaldehyde (for bark oil) and eugenol (for leaf oil). Although synergism among major and minor components is well known, each of the dominant constituents of the oils is acting with a single mode of action, making them in this regard similar to conventional synthetic insecticides (Copping and Menn 2000). In fact, Correa et al. (2015) evaluated the toxicity (including the effects on the population growth rates) of cinnamon essential oils, as well as its effects on the behavioral (locomotory) and respiratory rates of four Brazilian populations of *Sitophilus zeamais* with distinct susceptibilities to traditional insecticides (phosphine and pyrethroids) and concluded that although cinnamon essential oil has the potential to control *S. zeamais* populations, insects from the studied populations that are resistant to traditional insecticides (e.g., pyrethroids and phosphine) might share some physiological and behavioral mechanisms to mitigate the actions of such essential oils. Moreover, Haddi et al. (2015), in a recent study, reported that the sublethal exposure to cinnamon essential oil of a population of *S. zeamais* susceptible to conventional insecticides resulted in stimulatory responses in the median survival time and the number of larvae per grain. The sublethal exposure elicited behavioral and physiological mechanisms that these insects normally use to overcome the actions of insecticides. Such findings showed that replacing synthetic insecticides with botanical insecticides like cinnamon essential oils to control insect pests still needs further investigation and scrutiny. More studies are also needed on the potential effects of the use of cinnamon oils on the nontarget and beneficial insects (Isman 2000), especially because a study demonstrated negative effects of the alcohol extract of *C. camphora* on two aphid parasitoids, *Aphidius gifuensis* and *Diaeretiella rapae* (Zhou and Liang 2003).

The nonpersistence of essential oils, first looked at as an environmental advantage, may turn out to be an issue, as higher quantities will be needed to reach the same levels of

control achieved with conventional insecticides, which may lead to higher residues in the environment (Copping and Menn 2000). Pest control in foodstuffs may face a problem of odor acceptability by consumers if cinnamon essential oils are applied as contact and fumigant, due to its strong flavored smell.

The availability of the raw material is another concern for the use of cinnamon oil as a pesticide to a large extent (Isman 2000). Furthermore, the wide use of these oils will depend deeply on the possibilities of growers to provide the essential oil industry with standardized material. Variability and inconsistencies in composition will influence the extraction efficiency and may jeopardize the investors' interest in such a risky sector.

TABLE 7.2

List of Commercially Available Cinnamon-Based Pesticides

Products (Trade Name)	Uses and Activity Description	Composition
Snail & Slug Away	Kills snails and slugs and their eggs	Active ingredients: Cinnamon oil Other ingredients: Soap bark, water, soybean oil, sunflower oil
Weed Zap	Contact, nonselective, broad-spectrum, foliar applied herbicide Controls both annual and perennial broadleaf and grassy weeds; does not translocate	Clove oil: 45% Cinnamon oil: 45% Other ingredients (lactose and water): 10%
Weed-A-Tak	Kills broadleaf weeds, grasses, vines, and brush	Citric acid: 4.0% Cinnamon oil: 1.0% Clove leaf oil: 1.0% Other ingredients: 94% (lecithin, water)
Cinnacure	Fungicide, insecticide, miticide	30% cinnamaldehyde
Cinnamite	Fungicide, insecticide, miticide	30% cinnamaldehyde
Valoram II	Kills and repels numerous insect pests	Clove/cinnamon/mint oils
Armorex II	Kills and repels numerous insect pests	Clove/cinnamon/mint oils
Kinnamon 70	Fungicide, insecticide, accaricide	Cinnamon extract 50% w/w
Citrokinnamon 50–30	Fungicide, insecticide, accaricide	Cinnamon extract 50% w/w, citrus extract 30% w/w
Cinnamon extract JBQ	Combats powdery mildew and spider mite pests Preventive and curative	Cinnamon extract (70%) and conditioners
Flower farm pesticide	Natural insecticide/miticide/fungicide	Cinnamon oil, cottonseed oil, rosemary oil
Final Stop® Pest Control Killer Spray	Controls: Ants, cockroaches, spiders, fleas, wasps, stink bugs, moths, silverfish, mosquitoes, centipedes, earwigs, gnats, chiggers, ticks, pillbugs, crickets, and other nasty creepy-crawly insects	Active ingredients: Cinnamon oil, rosemary oil, sesame oil, peppermint oil, thyme oil, garlic extract Inert ingredients: Beeswax, calcium carbonate, carrageenan, cellulose, citric acid, glycerin, kaolin, lecithin, mustard powder, sodium bicarbonate, sodium chloride, soybean oil, wintergreen oil, distilled water
Spider Killer	Against spider mites, thrips, nematodes, cochineal, aphids, and other insect pathogens	Rich in cinnamon (*Cinnamomum zeylanicum*)

7.11 Essential Oil–Based Pesticides

The review of the available experimental data (Section 7.8) supports the hypothesis that cinnamon oil is an effective natural pesticide and repellant against insects. Cinnamon essential oils and/or their constituents have shown a broad spectrum of insecticidal, miticidal, nematicidal, fungicidal, and bactericidal activity, as well as having a good repellency potential. Moreover, registration specifications indicate that they are user and environment safe. Nevertheless, few cinnamon-based pesticides are available in the market for wide use (see Table 7.2). The majority of the pesticides with composition containing cinnamon or its constituents are targeting mainly small-scale uses, such as home garden and indoor applications. The only major exception is Cinnacure, which is recommended for large-scale uses, as both an insecticide and a fungicide.

7.12 Conclusions

Although cinnamon tree products have played key roles in nutrition, medicine, and religion for centuries, their use as green pesticides is still limited. The chemistry of the genus *Cinnamomum* is interesting, but efforts in the research and development of these tree spices and their products have been restricted mainly to the volatile oil and its constituents. Various studies have dealt with the potential benefits of *Cinnamomum* for human health with controversial results. The pesticidal activities of cinnamon species have received little attention, and more research and scientific investigations are needed to unleash the huge potential of a tree qualified often as the "spice of life."

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