

Chemical Composition of Essential Oil of Coriander Seeds (*Coriandrum sativum*) Cultivated in Amazon Savannah, Brazil

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Coriandrum sativum (Apiaceae), popularly known as coriander, is one of the most consumed and cultivated vegetables in tropical and subtropical regions of the world, mainly in the North and Northeast of Brazil. This species has functional properties, nutraceutical and bioactive compounds of biotechnological interest. For those reasons, coriander has been widely studied in different lines of research, especially the oil of its seeds. In this sense, the objective of this work was to evaluate the phytochemical composition of the essential oil of coriander seeds cultivated in the Savannah tropical region Brazil, in the state of Roraima, by gas chromatography coupled to mass spectrometry (GC-MS) and high resolution gas chromatography (GC-FID). To obtain the oil, the seeds were processed and subjected to hydrodistillation process in Clevenger-type apparatus. The identification and quantification of the volatile constituents were provided by gas chromatography coupled by mass spectrometry (GC-MS) and high-resolution gas chromatography (GC-FID). The chromatographic analysis presented ten components, with the four main constituents corresponding to 80.3%: linalool (64.4%); 2-dodecanal (5.5%); palmitic acid (5.3%); and geraniol (5.1%). Among the chemical constituents of the profile, mainly monoterpenes were found, with linalol being the main one. This botanical genotype also has significant concentrations of palmitic and myristic acids.

Keywords: Chromatography, Linalool, Chemotypes, Phytochemistry

1. Introduction

Coriandrum sativum, popularly known as coriander, belongs to the family Apiaceae or Umbelliferae, and is originated in the Eastern Mediterranean, being widely produced and consumed in North Africa, Central Europe and Asia (Abdelkader et al., 2018). In Brazil, its production area is mainly concentrated in the North and Northeast (Brazil, 2018) and is generally managed by family farmers. This cultivated plant is part of the food culture and composes the nutritional habits of the population of Northern Brazilian region, playing an important socioeconomic role in these territories.

Moreover, the products derived from processing this vegetable, mainly leaves and seeds, are also widely used in the cosmetics, pharmaceutical and food industries, as well as popularly used for culinary and therapeutic purposes. Due to its physiological and adaptive characteristics and the bioactive potential of the chemical constituents, it can be exploited throughout the year, provided that the necessary agronomic requirements are met (Da Silva et al., 2018). Biological activities of coriander are also widely investigated in several areas of knowledge (Begnami et al, 2018; Abdella et al., 2018), including prospecting studies of their oils (Aelenei et al., 2018). Studies have demonstrated the potential of coriander's oil as antimicrobial (Salamon et al., 2018);

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antibacterial (Mansouri et al., 2018), and cytotoxic (Caputo et al., 2016) agents, among others. These biological activities can be related to the synergic effect of secondary metabolites (Elhidara et al., 2018; Hu et al., 2019) or related to the isolated activity of some constituents, like linalool, identified as the major secondary metabolites in the essential oil of coriander seeds in the previously cited studies. Despite variations in the concentration of the monoterpene alcohol linalool in the phytochemical profiles analyzed, independently: i) of the chemotypes evaluated; ii) stage of its phenological cycle; or, iii) the condition edafoclimatic and ecophysiological that the seeds were submitted, this monoterpene, in general, is the metabolite present in the highest concentration in the essential oil of coriander seed (Ben et al., 2018a).

It is worth highlighting that the literature shows expressive differences between the compositions of the essential oil of coriander, according to the part of the plant, showing that the biosynthesis of secondary metabolites varies in this botanical species (de Almeida Freires et al., 2014). For instance, the essential oil of the leaves have four aldehydes and a fatty acids as the major components (decanal, 2-decenal, 2-dodecanal, tetradecenal, and palmitic acid) but linalool was not identified in the phytochemical analysis of coriander leaves. Mandal and Manda (2015) reported that the concentration of linalool in the seeds and also the phytochemical differences are due changes occurred during the ontogenesis process, which gives a unpleasant odor to the immature fruits of coriander, known as the "stink bug" due to *trans*-tridecene constituent contained in its oil.

Investigations with chemotypes of this botanical species cultivated under Amazon Savannah conditions were not identified in the literature in this line of research. Knowing this, the present study had the objective to evaluate the phytochemical composition of the essential oil of seeds of *Coriandrum sativum* cultivated in Roraima's Savannah, Brazil, by Gas Chromatography Coupled to Mass Spectrometry (GC-MS) and High Resolution Gas Chromatography (GC-FID).

2. Materials and methods

2.1 Collection, processing and obtaining of essential oil

The seeds were collected in the area of Savannah (Lavrado) in the locality of Murupú (3°04'15.6" N - 60°48'53.3" W), during the dry season, in the state of Roraima, in Brazilian Amazon. The climate of this region is characterized as Tropical with rain (Aw), according to the types of climatic classification of Köppen. The access was duly registered in the National System of Management of Genetic Heritage and Associated Traditional Knowledge - SisGen linked to the Ministry of the Environment - MMA, according to specific Brazilian legislation, under number A5FD8B7.

After the collection, the following procedures were performed in the Laboratory of Environmental Chemistry of the Federal University of Roraima - UFRR, Brazil: Seeds were selected, washed, dried (50 °C) until reaching a constant mass, and then duly weighed and grinded in a household blender. Later, the samples were separated - obtaining homogeneous material with granulometry between 20-40 Mesh, and, again weighed and submitted to hydrodistillation in a Clevenger type device, double condenser of the brand Spell, to obtain the essential oil. The extraction was carried out for two hours without interruption, in triplicate. Finally, the sample was stored in an amber bottle and preserved until further analysis.

2.2 Phytochemical Screening

To determine the constituents of the essential oil of *Coriandrum sativum* (EOS) seeds, part of the work sample was sent to the Laboratory of Chemistry of the Federal University of Minas Gerais - UFMG, Brazil, and supplied by GC-MS Spectrometric Gas Chromatography) and High Resolution Gas Chromatography (GC-FID), as described by Melo Filho et al. (2018).

For identification of the phytochemical constituents present in the essential oil, a GCMS-QP2010 ULTRA (Shimadzu) equipment, equipped with a column Rxi-1MS 30 m x 0.25 mm x 0.25 µm (Restek) was used. Column temperature was 50 °C (3 min), 3 °C.min⁻¹, to 220 °C. The injector worked with initial temperature of 220 °C, with split mode ratio (1: 50), GC-MS interface at 220 °C. Detector MS (Electronic Impact at 70 eV) at 220 °C. Flow of entrainment gas (Helium) was 3.0 mL / min. The mass spectrum of each essential oil component of *C. sativum* seeds was carefully analyzed in comparison to spectra of the library spectra (NIST11), using GC-MS Solution (Shimadzu) software and also by comparison with the indices of Kovats.

The quantification of the essential oil components of *C. sativum* seeds was performed using gas chromatography on a HP 7820A Gas Chromatograph (Agilent) equipped with a capillary column with dimensions 30 m x 0.32 mm x 0.25 µm (Agilent), with a temperature of 50 °C (0 min), 3 °C / min, up to 220 °C. Samples containing 1 µL of essential oil diluted (1%) in chloroform with an initial temperature of 200 °C in the split ratio (1: 50) was injected. The CG-FID worked with a temperature of 220 °C. Drag gas, Helium at 3 mL / min. Data acquisition software was Ezchrom Elite Compact (Agilent).

3. Results and discussion

The chemical constituents of *C. sativum* seeds, identified by GC-MS and quantified by GC-FID, are presented in Table 1. It was possible to quantify and identify 96.9% of the fatty acids present in the oil composition.

Table 1: Phytochemical composition of the essential oil of Coriandrum sativum seeds grown in the Savannah area of the state of Roraima, Brazil, in the dry period, by Gas Chromatography Coupled to Mass Spectrometry (GC-MS) and by Chromatography High Gas Chromatography Resolution (GC-FID).

Constituents	%	KI*
Linalool	64.4	1099
Camphor	2.9	1131
Decanal	3.4	1198
2-decenal	3.6	1256
Geraniol	5.1	1363
Geranyl Acetate	2.1	1388
2-dodecanal	5.5	1470
Tetradecenal	2.7	1671
Myristic acid	1.9	1764
Palmitic acid	5.3	1936
Others	3.1	-
Total	100	-

*Kovats Index

Linalool was identified as the major constituent of the essential oil of seeds (EOS) of *Coriandrum sativum* under conditions in which the present study was conducted. This result is in agreement and corroborates with other investigations that have been developed with the essential oil of coriander in different phases of its phenological stage and in several regions of the world. Linalool is a monoterpene alcohol of biotechnological interest, mainly because of its widely researched bioactive potential (Shaaban et al., 2018; Jabir et al., 2019).

In a study on the chemical composition of essential oil from *Coriandrum sativum* seeds in Poland, a central European chemotype was analyzed by GC-MS. The main components identified were linalool (78.45%), α -pinene (5.03%), camphor (3.90%), γ -terpinene (3.80%), D-limonene (2.58%) and geranyl acetate (2.13%) (Huzar et al., 2018). The same authors reported that the replacement of conventional (hydrodistillation) by microwave heating for 15 minutes had a positive influence, increasing the content of some less volatile components, among them, linalool. This may explain the higher percentage of linalool reported by them in comparison with the present study.

When investigating the phytochemical profile and bioactive effects of EOS from a cultivar of *C. sativum* from Turkey, Ildiz et al. (2018) observed that among the ten compounds identified and quantified, linalool was the major constituent in the sample (82.2%), being the second and third most abundant component in the essential oil the compounds camphor (3.21%) and γ -terpinene (2.6%), respectively. The Turkish genotype also showed a lower concentration of palmitic and myristic fatty acids in comparison with the levels determined for the coriander seed from Tropical Roraima's Savannah in Brazil. Nguyen et al. (2015) when evaluating the accumulation of lipids and fatty acids during the development of organic seeds of *C. sativum* in an experiment carried out in southwest France also found that palmitic and myristic acid concentrations were lower than those found in the present study. This difference can be justified by the climatic and management conditions that the plants were subjected to during their growth and development. Species that develop in temperate climates generally accumulate fewer fatty acids in their seeds (Taiz et al., 2017).

From the results of the analysis of essential oil of coriander seeds by GC-MS, Zanusso-Junior et al. (2011) also found linalool as the main component (82.2%). In addition, the other compounds identified were α -pinene (4.0%), camphor (2.6%), α -terpinene (2.6%), linalin acetate (2.4%) showing some coincidences with the values herein reported. The before mentioned work also detected that *C. sativum* L. exhibits anti-edematogenic activity.

When observing the effect of the distillation time in the composition and bioactivity of coriander oil, Zheljzkov et al. (2014) detected eleven constituents and found that the concentration of the main constituent of EOS (linalool) was 50.7% when distillation time was 115 min and gradually increased to 67.9% with the increase of distillation time to 240 min. It was evidenced that distillation time can be used to obtain higher yield of the essential oil of *C. sativum* with differentiated composition. The concentrations of the other constituents present in the oil also varied in nine distillation times evaluated. However, for a period inferior than 40 minutes, the

coriander EOS herein studied reached already a concentration of 64.4% linalool, being very close to that described for the American genotype (USA).

The chromatogram showing the secondary metabolites quantified in the present study is presented in Figure 1. The Brazilian Savannah's chemotype investigated showed a profile containing four main constituents: linalool (64.4%), 2-dodecanal (5.5%), palmitic acid (5.3%), and geraniol (5.1%) corresponding to 80.3% of the compounds identified.

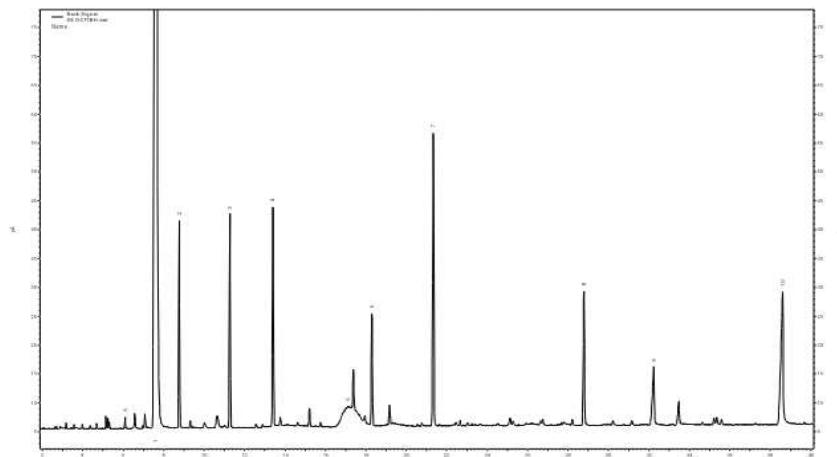


Figure 1: Chromatogram of the phytochemical profile of the major constituents of the essential oil of *C. sativum* seeds grown in Savannah area in the state of Roraima, Brazil, in the dry period, quantified by High Resolution Gas Chromatography (GC-FID).

When studying the constituents of the EOS of four coriander genotypes of Turkey, Beyzi et al. (2017) also quantified ten secondary metabolites, with linalool being the major compound, ranging from 89.44% to 91.77 % among cultivars, and camphor was the second main secondary metabolite quantified, with variation between 2.01 % and 2.83% among the investigated genotypes. It is believed that the difference in the concentration of these constituents is directly related to the abiotic and biotic conditions that these specimens were subjected during their phenological cycle. Camargo and Vasconcelos (2015) corroborate that the essential oils of *C. sativum* presented two components in common: camphor and linalool. These same authors added that the differences between components and concentrations of essential oil of seeds from this species may vary due to climatic conditions, soil, period of collection, nutrition and availability of water, among other influences, such as their stage of development.

In this context, when evaluating the composition of essential oils of coriander fruits at three maturation stages by GC-FID and GC-MS, Msaada et al. (2007) found that geranyl acetate (46.27%), linalool 10.96%), nerol (1.53%) and neral (1.42%) were the main compounds in the first stage of maturation (immature fruits). In the intermediate stage, linalool (76.33%), cis-dihydrocarvone (3.21%) and geranyl acetate (2.85%). And in the final stage of maturity (ripe fruits) consisted mainly of linalool (87.54%) and cis-dihydrocarvone (2.36%). Therefore, literature consistently demonstrated that the concentration of chemical constituents of coriander's EOS vary as the plant develops. A study of the composition of *C. sativum* EOS carried out in Iran revealed thirty-four different compounds, the main components being linalool (40.9 - 79.9%), neryl acetate (2.3 - 14.2%), γ -terpinene (0.1 - 13.6%) and α -pinene (1.2 - 7.1%) (Nejad Ebrahimi et al., 2010). Sourmaghi et al. (2015) by means of GC-MS found that EOS of *C. sativum* presented 32 components, where the main constituent was linalool in the range of 50.7 - 67.9%. Wang et al. (2018) observed that essential oil of coriander seeds contained 37 compounds and the main components were: linalool (37.12%), geranyl acetate (35.72%) and menthol (5.07%). Sriti et al. (2014) when analyzing the phytochemical profile of OES of *C. sativum* identified and quantified 39 constituents, so that linalool was the main compound of essential oils with 76.11%. These results show that coriander varieties grown under different ecological and edaphoclimatic conditions have a direct influence on their composition. However, despite the concentration variations, linalool is the major constituent independent of the region of origin and the current study demonstrates the prominent presence of this monoterpene alcohol in the chemotype herein studied.

Studies in Brazil (De Figueiredo et al., 2004) and Pakistan (Ben et al., 2018b) evaluating the phytochemical composition and bioactivity from EOS of coriander genotypes revealed again linalool as the major constituent (78.86% and 77.48%, respectively). In agreement with all the results presented in previously cited investigations, Iqbal et al. (2018) corroborate and meta-analyzes asserted that volatile essential oil of

coriander seeds possess alcoholic monoterpenes as main constituent with bioactive potential that can be exploited for different purposes.

4. Conclusions

The chemical composition of the essential oil of seeds (EOS) of *Coriandrum sativum* of this cultivar, in the experimental conditions of this study, presented a phytochemical profile without major differences when compared to the cultivar studied in other regions of the world. However, an important communication is that the EOS of coriander presented a concentration of linalool (64.4%) lower than that reported for samples from temperate climate specimens discussed in this investigation. Another relevant aspect is that this genotype produces palmitic and myristic acids with have nutraceutical, functional and therapeutic properties. Therefore, systematic and applied studies are recommended to evaluate its activities and biological functions.

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