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Variation of essential oil composition of fruits of *Zanthoxylum leprieurii* Guill. et Perr (Rutaceae) from different sample locations and during their maturation period

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ABSTRACT

The fruits of *Zanthoxylum leprieurii* Guill. & Perr. (Rutaceae) are traditionally used in Africa, particularly in Cameroon, as a spice and in the treatment of sickle cell anaemia. The objective of this study was to investigate the fruit essential oils from different sample locations and during their maturation period by using GC and GC-MS. Except for one sample collected in Aniak which consisted mainly of oxygenated monoterpenes, the volatile composition of *Z. leprieurii* fruits exhibited relative high amounts of monoterpene hydrocarbons (61.1–90.9%) such as (*E*)–*β*–ocimene (13.4–75.2%) and *α*-pinene (1.1–40.7%). The influence of the maturity stage on the chemical composition of the essential oils has resulted in an increase of oil yield and variations in the content of volatile compounds (*α*-pinene 13.3–75.6; (*E*)–*β*–ocimene 2.9–64.0%).

Keywords: Senegal, *Zanthoxylum leprieurii* fruits, essential oil, chemical variability, stage of maturity and GC–MS.

INTRODUCTION

Zanthoxylum leprieurii Guil. & Perr., also known as *Fagara leprieurii* Engl., is a tree about 24 m tall from the family Rutaceae, it is widespread throughout tropical Africa. In Cameroon, the dried fruits are traditionally used as spice in soups and its infusion is taken to treat sickle cell anaemia (Guetchueng et al., 2017; Tchinda et al., 2009). Some studies reported that fruit extracts exhibited antiplasmodial (Tchinda et al., 2009), cytotoxic (Choumessi et al., 2012; Ngoumfo et al., 2010) and antioxidant (Tchinda et al., 2009) activities. Previous phytochemical studies on the secondary metabolites of fruit extracts showed the presence of terpenes (Guetchueng et al., 2017), alkaloids (Ngoumfo et al., 2010; Tchinda et al., 2009) and coumarins (Tchinda et al., 2009). Moreover, some papers reported the chemical composition and biological

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activities of fruit essential oils from various geographical origins such as Nigeria and Cameroon. These studies showed chemical variability on the basis of amounts of hydrocarbon monoterpenes ((*E*)-β-ocimene, α-pinene, β-pinene, terpinolene, limonene and δ-3-carene), hydrocarbon sesquiterpene (caryophyllene), oxygenated monoterpenes (β-citronellol, citronellic acid and β-citronellal) (Dongmo et al., 2008; Ekundayo et al., 1986; Fogang et al., 2012; Gardini et al., 2009; Lamaty et al., 1989; Misra et al., 2013; Oyedeji et al., 2008; Reisch et al., 1985; Tatsadjieu et al., 2003).

Generally, the essential oil constituents depend on the internal and external agents influencing the plant such as genetic sequences, biotic, abioticand agricultural factors (Moniodis et al., 2018). In addition, seasonal variations (Matias et al., 2016; Moghaddam et al., 2015; Okoh et al., 2018), the development stage of the collected plant (Mehmood et al., 2019) and the method of preparing samples for analysis (Sefidkon et al., 2006) influence the yield and the different compound contents.

Thus, the aim of the present study was to characterize the chemical compositions of *Z. leprieurii* oil fruits from different sample locations and during their maturation period by using GC and GC-MS.

MATERIALS AND METHODS

Plant material

Twenty-seven samples of *Z. leprieurii* fruit pericarps (Figure 1) were collected between october 27 and 29, 2015 from seven localities of Senegal: Kabrousse (*Kab*, 12°21'N, 16°42'O; eight samples), Emaye (*Em*,12°26'N, 16°33'O; two samples), Essyl (*Es*, 12°31'N, 16°25'O; one sample), Colomba (*Col*, 12°46'N, 16°14'O; four samples), Thionck Essyl (*Th*, 12°46'N, 16°28'O; four samples), Carapan (*Cap*, 12°51'N, 16°19'O; five samples) and Aniak (*An*, 12°34'N, 16°07'O; three samples). A sample corresponds to all the fruits harvested from the same tree. For monitoring the evolution of the chemical composition of essential oil fruits during the maturation, the fruits were taken from a tree located in Colomba (*Col*.2) at different times (from september 1 to october 27, 2015). The plant material was identified by Dr William Diatta from the Department of botanical and pharmacognosy of University Cheikh Anta Diop of Dakar by comparison with the reference specimen deposited in the herbarium of this institution under number 1450.

Essential oil isolation

Plant material were air-dried for 14 days at room temperature. Samples were hydrodistilled (6 h) using a Clevenger-type apparatus according to the method recommended in the European Pharmacopoeia (Council of Europe, 1997). The yields of essential oils (w/w, calculated on dry weight basis) are given in Table 1 and 2. The essential oils were stored in amber glass bottles at a temperature of 4 °C.

Gas chromatography (GC) and gas chromatography-mass spectrometry (GC/MS) analysis

Analyses were carried out using a *Perkin-Elmer Autosystem XL GC* apparatus (*Walthon*, MA, USA) equipped with dual flame ionisation detection (FID) system and fused-silica capillarycolumns, namely, Rtx-1 (polydimethylsiloxane) and Rtx-wax (poly-ethyleneglycol) (60 m × 0.22 mm i.d; film thickness 0.25 µm). The oven temperature was programmed from 60 to 230 °C at 2 °C/min and then held isothermally at 230 °C for 35 min: hydrogen was employed as carrier gas (1 mL/min). The injector and detector temperatures were maintained at 280 °C, and samples were injected (0.2 µL of pure oil) in the split mode (1:50). Retention indices (RI) of compounds were determined relative to the retention times of a series of n-alkanes (C5–C30) by linear interpolationusing the Van den Dool and Kratz (1963) equation with the aid of software from *Perkin-Elmer* (Total Chrom navigator). The relative percentages of the oil

Figure 1 : Imature (1) and mature Fruits (2) of *Z. leprieurii.*

constituents were calculated from the GC peak areas, without application of correction factors.

Samples were also analysed with a *Perkin-Elmer Turbo mass* detector (quadrupole) coupled to a *Perkin-ElmerAutosystem XL*, equipped with fused-silica capillary columns Rtx-1 and Rtx-Wax. The oven temperature was programmed from 60 to 230 °C at 2 °C/min and then held isothermally at 230 °C (35 min): hydrogen was employed as carrier gas (1 mL/min). The following chromatographic conditions were employed: injection volume, 0.2 µL of pure oil; injector temperature, 280 °C; split, 1:80; ion source temperature, 150 °C; ionisation energy, 70 eV; MS (EI) acquired over the mass range, 35–350 Da; scan rate, 1 s.

Identification of the components was based on: (a) comparison of their GC retention indices (RI) on non-polar and polar columns, determined from the retention times of a series of n-alkanes with linear interpolation, with those of authentic compounds or literature data; (b) on computer matching with commercial mass spectral libraries (Adams and others, 2007; König et al., 2004; NIST, 2008) and comparison of spectra with those of our personal library; and (c) comparison of RI, MS and NMR spectral data of authentic compounds or literature data.

RESULTS

Chemical variability of fruit essential oils according to geographical origins of samples

We had undertaken a comparative study of the chemical composition of the fruits according to the biotopes (harvesting areas) in order to develop a possible production of essential oils from plants growing spontaneously. This study on different Senegalese biotopes aimed to highlight a possible specificity compared to studies in the literature. This work involved 27 samples of *Z. leprieurii* fruits taken from seven different stations (Kabrousse *Kab*, Emaye *Em*, Essyl *Es*, Colomba *Col*, Thionck Essyl *Th*, Caparan *Cap*

and Aniak *An*). A sample corresponded to all the fruits harvested from the same tree.

Table 1 shows the chemical composition of each sample and gives the essential oil yields. These were calculated relatively to the mass of dry vegetable matter and are between 0.60 and 3.49%.

The analysis of the fruit essential oils by GC-FID and GC/MS allowed the identification of 66 compounds accounting for 79.2 to 96.8% of the total oil weight. These oils had qualitatively similar chromatographic profiles, but their compositions were quantitatively very variable. They were all dominated by hydrocarbon monoterpenes (61.1–90.9%) such as (*E*)-β-ocimene ***16*** (13.4–75.2%) and α-pinene ***2*** (1.1–40.7%). However, there was an exception, the sample An.2 was mainly composed of oxygenated monoterpenes. Samples collected at Kabrousse, Emaye, Caparan (except Cap.5), Essyl and Colomba showed almost the same proportions in (*E*)-β-ocimene ***16*** (22.5–57.9%) and *α*-pinene ***2*** (20.4–40.7%), while those of Thionk essyl and Aniak were characterized by high levels of (*E*)-β-ocimene ***16*** (54.4–72.7%) and low levels of α-pinene ***2*** (1.0–11.2%).

We also reported two atypical chemical compositions for two samples Cap.5 and An.2 The main components of Cap.5 are *δ*-3-carene ***10*** (17.1%), α-pinene ***2*** (14.9%), terpinolene ***20*** (14.5%) and (*E*)-β-Ocimene ***16*** (13.4%) while those of An.2 were geranyl acetate ***53*** (51.6%), geraniol ***47*** (19.4%), cironellol ***45*** (4.7%) and (*E*)-β-ocimene ***16*** (4.7%). In addition, among these compounds, five had very low or no abundance in the other 25 samples: *δ*-3-carene ***10*** (0.0–0.1%), terpinolene ***20*** (0.0–0.6%), geranyl acetate ***53*** (0.0–3.2%), geraniol ***47*** (0.0–3.6%) and cironellol ***45*** (0.0–2.5%)

Evolution of the chemical composition of essential oils of fruits during their maturation period

It should be emphasized that the valuation of an essential oil requires a judicious determination of the harvest period. In order to determine the impact of the fruit ripening process, we followed the evolution of the chemical composition of essential oil over a period of two months (September - October) for fruits always harvested on same tree Col.2 (Figure 1). First, we found that the yield of essential oil, expressed as a percentage (%) relative to the dry matter, increases during fruit ripening. Indeed, the best yields (0.97 to 1.64%) were obtained in October when the harvested fruits were yellow-orange in color. For immature fruits harvested in September; the yields are from 0.51 to 0.81% (Table 2).

The chemical composition of the essential oils was qualitatively identical throughout the ripening of the fruits. Essential oils were mainly characterized by hydrocarbon monoterpenes (78.5 to 86.2%), with a high abundance of *α*-pinene and (*E*)-*β*-ocimene. The results and discussion of this study focused only on these two major constituents. However, we noted quantitative variations between the oil compositions of fruits from the first two harvests (September 1 and 8, 2015) characterized by *α*-pinene rising to 75.6 and 57.9%, respectively and those of the fruits of the following harvests (from September 22 to October 27, 2015) characterized by the combination of α-pinene (13.3 to 32.2%) with (E) β-ocimene (40.5 to 64.0%) which was always the major compounds (Figure 2).

Table 1: Chemical variability of essential oils from *Z. leprieurii* fruits according to sample locations in Senegal.

No[a]	Compounds	IRIa[b]	RIa[c]	RIp[d]	Chemical variability of fruit essential oils[e]																									
					Kabrousse								Emaye		Caparan					Colomba				Essyl	Aniak			Thionck Essyl			
					Kab 1	Kab 2	Kab 3	Kab 4	Kab 5	Kab 6	Kab 7	Kab 8	Em 1	Em 2	Cap 1	Cap 2	Cap 3	Cap 4	Cap 5	Col 1	Col 2	Col 3	Col 4	Es	An 1	An 2	An 3	Th 1	Th 2	Th 3	Th 4
1	*a*-Thujene	932	922	1023	0.1	0.1	0.3	-	0.1	0.1	0.1	-	0.3	0.2	-	-	0.1	0.1	0.1	-	0.3	0.1	-	0.1	-	-	tr	-	-	0.3	0.1
2	*α*-Pinene	936	931	1015	35.5	20.4	33.3	40.7	35.2	29.0	38.7	28.4	23.7	30.7	34.4	25.9	30.1	30.2	14.9	29.1	30.4	16.1	26.1	35.3	1.1	2.0	2.5	11.2	9.7	6.4	6.0
3	Camphene	950	943	1066	0.5	0.2	0.6	0.8	0.4	0.4	0.5	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.1	0.3	0.4	0.2	0.3	0.4	-	-	tr	0.1	0.1	0.1	0.1
4	Thuja-2,4-(10)-diene	946	946	1123	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.1	-	0.1	0.1	-	0.1	0.2	-	0.1	-	0.1	0.1	0.3	-	-	-	-	-	-	-
5	Sabinene	973	964	1120	0.5	0.4	3.2	0.1	1.1	0.1	0.8	0.1	4.0	2.2	0.1	0.1	0.1	0.1	0.1	0.1	2.7	0.3	0.1	0.5	-	-	1.1	0.1	0.1	6.0	3.5
6	*β*-Pinene	978	970	1108	0.8	0.4	1.2	1.0	0.9	0.5	0.9	0.6	0.8	0.7	0.6	0.4	0.5	0.5	0.2	0.5	0.9	0.3	0.4	0.7	-	tr	0.1	0.3	0.2	0.4	0.4
7	Myrcene	987	979	1159	1.0	0.6	1.5	1.6	1.1	1.0	1.1	0.8	1.2	1.0	0.7	1.0	1.0	0.7	4.5	0.9	1.3	0.6	0.8	0.7	0.3	2.9	0.4	0.6	0.6	0.9	0.7
8	Octanal	981	980	1290	0.1	0.1	tr	Tr	0.1	tr	0.1	0.1	0.2	tr	-	-	-	-	tr	0.1	0.2	0.1	tr	tr	0.1	0.2	0.1	0.1	-	-	-
9	⊩2-Carene	1000	996	1131	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.5	-	-	-	-	-	-	-	-	-	-	-	-
10	⊩3-Carene	1010	1005	1147	-	-	-	-	-	-	-	-	-	-	-	-	-	-	17.1	-	-	-	-	-	-	-	-	-	-	0.1	-
11	⊩Terpinene	1013	1008	1178	0.1	0.6	0.5	-	0.1	0.3	0.2	0.2	0.5	0.2	-	-	-	-	0.6	tr	0.4	-	-	0.2	0.2	0.2	0.1	-	-	0.5	0.5
12	*p*-Cymene	1015	1011	1268	0.3	0.1	0.4	0.2	0.5	tr	0.3	0.1	0.6	0.3	0.1	-	0.2	0.3	2.0	0.2	0.5	-	-	0.3	0.1	0.2	0.3	-	-	0.6	0.7
13	Limonene	1025	1020	1201	0.6	0.3	0.7	0.9	0.7	0.5	0.6	0.6	0.6	0.4	0.4	0.4	0.5	0.4	0.2	0.5	0.6	0.2	0.6	0.5	0.1	0.2	0.2	0.3	0.2	0.2	0.3
14	1,8-Cineole	1024	1020	1209	1.5	1.6	7.6	0.4	1.9	0.6	1.2	0.4	0.6	0.3	0.2	0.2	0.5	0.1	2.5	0.3	0.5	0.4	0.3	0.7	0.1	0.1	0.2	0.2	0.2	0.6	0.3
15	(*Z*)-*β*-Ocimene	1029	1024	1230	0.6	0.6	0.5	0.5	0.4	0.7	0.6	0.5	0.6	-	0.5	0.8	0.5	0.6	0.3	0.7	0.7	1.0	0.8	0.4	1.0	0.1	0.8	0.9	1.1	1.0	0.7
16	(*E*)-*β*-Ocimene	1041	1034	1247	41.0	51.5	33.1	32.6	24.2	45.8	36.4	29.1	41.4	49.7	36.4	52.6	32.5	39.6	13.4	48.7	50.9	62.6	57.9	22.5	72.7	4.7	65.5	70.4	75.2	64.1	54.4
17	⊩Terpinene	1051	1047	1243	0.2	0.2	0.8	0.1	0.3	0.1	0.2	0.1	1.1	0.4	0.1	-	0.2	tr	0.1	tr	0.7	0.1	0.1	0.3	-	-	0.4	-	-	1.0	1.2

18	Octanol	1063	1065	1531	0.2	-	-	0.1	-	0.1	0.3	-	0.1	-	tr	-	0.1	0.2	-	-	0.1	0.1	-	-	0.2	-	-	-	-	0.1	0.2
19	*p*-Cymenene	1075	1072	1432	-	-	-	-	-	-	-	-	-	-	-	-	-	-	1.1	0.1	-	-	-	-	-	-	-	-	-	0.1	-
20	Terpinolene	1082	1078	1280	-	-	-	-	-	-	-	-	-	-	-	-	-	-	14.5	-	0.4	-	-	-	-	-	-	-	-	0.6	-
21	6-Methyl-3,5-heptadien-2-one		1080	1581	0.2	0.6	0.2	0.1	0.2	0.3	0.3	0.3	-	0.2	0.1	-	0.2	0.2	-	0.3	-	0.5	0.3	0.2	0.2	0.1	0.2	0.3	0.2	0.1	0.2
22	Linalool	1086	1081	1544	0.3	0.4	0.2	0.3	0.3	0.3	0.2	0.7	-	0.3	-	0.1	0.1	0.3	-	0.2	tr	0.2	0.1	0.4	0.4	0.9	0.3	-	0.2	0.3	0.2
23	Rosefurane	1091	1083	1398	0.1	1.9	1.0	0.9	1.2	1.4	1.9	1.7	0.9	1.2	1.2	0.4	2.3	2.0	-	2.0	-	1.5	1.1	3.5	1	0.6	1	1.1	0.8	0.3	1.6
24	Nonanal	1076	1083	1394	1.1	0.2	tr	0.2	0.1	0.2	0.1	0.1	-	0.1	-	-	0.2	0.2	-	0.1	0.1	0.1	0.1	0.2	0.3	0.1	0.1	0.2	0.1	0.1	-
25	Perrilene	1090	1092	1416	-	-	-	0.1	-	-	-	-	-	0.2	-	-	-	-	0.1	0.1	-	-	-	0.1	-	-	-	-	-	-	-
26	-Campholenal	1105	1105	141	0.2	0.1	0.1	0.2	0.2	0.3	0.2	0.3	0.2	0.2	0.1	0.1	0.3	0.2	-	0.3	-	-	0.1	-	0.1	-	-	-	-	-	0.1
27	*p*-Mentha-1,3,8-triene	1123	1122	1433	0.2	0.2	0.2	0.4	0.2	0.1	0.1	0.2	0.2	0.2	0.2	0.1	0.2	0.3	-	0.3	0.7	0.3	0.2	0.3	0.3	-	0.3	0.3	0.4	0.4	0.2
28	(*E*)-Ocimenoxide	1125	1125	1482	0.2	0.3	0.2	-	0.4	0.3	0.3	0.3	0.3	0.4	0.4	0.3	0.6	0.3	-	0.5	0.1	0.3	0.4	0.2	1.1	-	-	0.5	0.7	0.4	0.3
29	*Cis*-Verbenol	1132	1127	1655	0.3	0.3	0.2	0.3	-	0.7	0.7	0.6	-	-	-	-	0.7	0.3	-	0.5	-	-	-	0.4	-	-	-	-	-	-	-
30	Citronellal	1129	1131	1479	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	3.0	-	-	-	-	-
31	Pinocarvone	1137	1136	1558	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.1	0.1	-	-	0.1	0.1	-	-	-	-	-	0.2	-	-	-	-	-	-	0.1
32	*p*-Mentha-1,5-dien-8-ol	1127	1146	1714	0.4	0.4	0.4	0.2	0.3	0.2	0.3	0.4	0.2	0.2	-	-	0.1	0.4	-	0.2	-	0.2	0.2	0.4	0.1	-	0.1	0.2	0.1	tr	0.2
33	Borneol	1150	1148	1698	0.2	0.2	0.2	0.2	0.2	0.1	0.2	0.2	-	0.2	-	-	-	-	-	-	-	-	0.1	0.2	0.1	-	0.2	-	-	-	0.1
34	Nonanol	1149	1156	1635	0.2	0.7	-	0.6	-	0.2	0.2	0.4	-	0.3	-	-	0.4	0.6	-	0.1	-	0.4	0.2	0.1	0.7	0.3	0.5	0.3	0.3	0.3	0.3
35	Cryptone	1160	1157	1667	0.1	-	0.1	-	0.2	0.1	-	-	-	-	-	-	-	-	-	-	-	-	0.1	-	-	-	-	-	-	-	-
36	Terpinen-4-ol	1164	1161	1600	0.6	0.6	2.2	0.3	1.5	0.1	0.7	0.3	3.6	1.0	-	-	-	-	-	-	1.8	0.3	0.2	0.6	0.2	-	3.3	-	-	3.0	6.5
37	*p*-Cymen-8-ol	1169	1170	1833	-	-	-	-	-	tr	-	-	-	-	-	-	0.1	0.2	7.0	0.1	-	-	-	-	-	-	-	-	-	-	-
38	Myrtenal	1172	1172	1628	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	-	-	0.2	-	-	0.1	-	-	0.1	0.2	0.1	-	-	0.1	-	-	0.1
39	Myrtenol	1178	1177	1789	0.1	-	-	0.1	0.1	0.2	0.1	0.1	-	-	-	-	-	-	-	-	-	-	-	0.1	2.4	-	1.8	0.1	-	-	-
40	-Terpineol	1176	1179	1700	0.8	0.9	3.1	0.6	1.9	0.2	0.7	0.7	0.5	0.2	-	-	0.3	0.2	0.9	0.1	0.4	0.4	0.2	0.9	0.3	-	0.6	0.2	0.1	0.4	0.9
41	Verbenone	1183	1184	1707	0.2	-	0.3	-	0.2	0.3	0.3	-	-	0.1	-	-	-	-	-	-	-	-	-	-	-	-	-	0.3	-	-	-
42	Decanal	1180	1185	1498	0.2	0.7	-	0.8	0.7	0.2	0.1	0.4	0.1	-	-	-	0.6	0.6	0.2	0.2	0.1	0.5	0.4	0.4	0.6	0.9	0.4	0.3	1.2	0.3	0.4
43	Octyl acetate	1194	1198	1478	0.6	-	0.2	0.3	0.4	0.4	0.8	0.1	-	-	-	-	-	0.7	-	0.3	-	0.6	-	-	0.6	0.2	-	-	0.3	0.2	0.7
44	*Cis*-Carveol	1210	1208	163	0.1	0.2	0.1	0.2	0.1	0.2	-	0.2	-	-	-	-	-	0.1	-	-	-	-	-	0.1	-	-	-	-	-	-	-

45	Citronellol	1213	1208	1764	-	-	-	1.1	0.4	-	-	2.5	0.5	-	-	-	-	-	0.6	-	-	-	-	-	-	4.7	-	-	-	0.3	-
46	Nerol	1210	1211	1799	0.3	0.2	tr	-	-	0.2	-	-	-	0.1	-	-	-	0.1	-	-	-	-	-	-	-	0.3	-	-	-	-	-
47	Geraniol	1235	1232	1844	-	-	-	0.5	0.3	-	-	0.7	0.9	-	-	-	-	-	3.6	-	-	-	-	-	0.4	19.4	-	-	-	-	-
48	Geranial	1244	1244	1731	-	-	-	-	-	-	-	0.1	-	-	-	-	-	-	-	-	-	-	-	0.1	0.3	0.4	0.2	-	-	-	-
49	Decanol	1259	1261	1765	-	-	-	0.5	-	-	-	0.1	-	-	-	-	-	-	-	-	-	-	-	-	0.4	-	-	-	-	-	-
50	Bornyl acetate	1270	1269	1575	0.1	-	0.1	-	0.1	-	-	0.1	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
51	Citronellyl acetate	1337	1331	1657	0.1	-	-	0.3	0.1	tr	-	0.7	0.1	-	-	-	-	-	0.1	-	-	-	-	-	-	1.1	-	-	-	-	-
52	Neryl acetate	1342	1342	1725	0.2	0.1	tr	0.1	0.3	0.1	0.1	0.3	0.6	0.1	-	0.1	-	0.1	0.1	-	0.2	0.2	-	-	0.1	-	-	-	-	0.5	0.2
53	Geranyl acetate	1362	1361	1752	0.9	0.3	-	1.1	0.5	0.2	tr	1.1	3.0	0.2	-	0.1	-	0.2	3.2	-	0.4	0.1	-	-	0.5	51.6	-	-	-	0.5	0.1
54	3,5-heptdienal, 2-ethylidene-6-methyl		1387	2039	0.2	-	0.2	0.3	0.3	0.4	-	0.6	0.2	-	-	-	-	0.3	-	-	-	-	0.3	-	-	0.1	-	-	0.5	0.2	0.4
55	-Caryophyllene	1421	1424	1591	0.6	0.7	0.8	0.8	1.6	1.2	0.5	1.3	1.1	0.4	4.5	5.0	1.1	1.5	0.6	0.8	0.5	0.7	0.7	1.2	0.3	0.2	2.2	0.6	0.4	2.3	1.6
56	*α*-Humulene	1455	1450	1660	0.2	0.2	0.2	0.2	0.4	0.3	0.2	0.4	0.3	0.1	1.0	1.2	-	0.4	0.1	0.2	0.1	0.2	0.2	0.4	-	-	0.5	0.2	0.1	0.5	0.4
57	*β*-Selinene	1486	1483	1712	-	-	-	-	-	-	-	0.3	-	-	-	0.2	-	-	-	-	-	0.1	-	0.2	-	-	-	-	-	-	0.1
58	-Cadinene	1520	1516	1752	-	0.1	-	-	-	-	-	0.2	-	-	0.1	-	-	-	-	-	-	-	-	-	-	-	0.1	-	0.1	-	-
59	(*E*)-Nerolidol	1553	1546	2037	0.1	0.3	0.2	0.1	0.6	0.3	-	0.5	0.7	0.5	0.5	0.2	-	0.2	-	0.1	0.1	0.3	0.1	0.1	-	0.2	0.4	-	0.5	-	0.6
60	Germacrene B	1552	1552	1828	-	-	-	0.2	0.5	-	-	0.2	-	-	-	-	-	-	-	-	-	0.2	-	0.2	0.5	-	-	0.3	-	-	0.4
61	Caryophyllene oxide	1578	1576	1980	2.8	4.5	2.2	2.1	5.0	5.9	2.8	5.0	3.0	1.9	9.5	5.2	4.7	9.8	1.3	6.5	0.7	2.9	3.1	10.7	0.9	1.6	6.8	1.6	1.7	2.7	5.6
62	Humulene epoxide II	1602	1598	2044	0.6	1.0	0.8	0.4	1.0	0.9	0.6	1.0	0.6	0.3	1.2	0.7	-	1.4	0.2	1.0	-	0.5	0.5	2.2	0.2	0.3	1.1	0.4	0.3	0.4	0.8
63	Caryophylla-4(14),8(15)-dien-5-		1626	2285	0.1	0.3	-	0.1	0.2	0.2	-	0.4	0.1	-	-	0.1	-	0.3	-	-	-	-	-	-	0.1	-	0.2	-	-	-	0.1
64	14-Hydroxy-9-epi-E-caryophyllene		1656	2316	0.3	0.4	0.1	0.1	0.4	0.6	0.2	0.4	-	-	-	0.5	-	0.9	tr	0.4	-	-	0.3	1.1	-	-	0.6	-	-	0.1	0.3
65	(*2E,6E*)-Farnesol		1706	2336	0.4	-	-	0.5	0.4	0.1	0.1	0.4	1.0	0.3	-	0.3	0.3	0.1	0.2	-	-	-	-	-	0.7	0.2	0.2	-	-	0.2	-
66	(*E-E*)-Farnesyl acetate	1822	1822	2260	0.2	-	-	0.1	0.1	Tr	0.1	0.4	0.2	-	-	tr	-	-	tr	-	-	-	-	0.1	0.1	-	tr	-	-	0.1	-
	Hydrocarbon Compounds				82.3	76.7	77.4	80.3	67.8	80.2	81.4	63.5	76.7	87.1	79.5	88.0	67.4	75.2	70.5	82.6	91.5	83.1	88.3	64.6	76.6	10.5	74.5	85.3	88.2	85.5	71.3
	Oxygenated Compounds				14.2	16.5	19.9	13.3	19.9	15.5	12.9	21.9	17.8	8.3	13.2	8.3	11.8	20.1	19.9	13.4	4.7	9.6	8.2	23.1	12.3	86.3	18.3	5.9	7.2	11.1	20.3
	Hydrocarbon Monoterpenes				81.5	75.7	76.4	79.1	65.3	78.7	80.7	61.1	75.3	86.6	73.9	81.6	66.3	73.3	69.8	81.6	90.9	81.9	87.4	62.6	75.8	10.3	71.7	84.2	87.6	82.7	68.8

Oxygenated Monoterpenes	7.3	8.3	16.4	7.4	10.9	6.4	7.5	12.6	11.8	4.9	2.0	1.3	5.5	5.1	18.0	4.6	0.6	4.1	3.5	8.2	7.4	82.3	7.9	3.0	2.8	6.6	11.3
Hydrocarbon Sesquiterpenes	0.8	1.0	1.0	1.2	2.5	1.5	0.7	2.4	1.4	0.5	5.6	6.4	1.1	1.9	0.7	1.0	3.4	1.2	0.9	2.0	0.8	0.2	2.8	1.1	0.6	2.8	2.5
Oxygenated Sesquiterpenes	4.5	6.5	3.3	3.4	7.7	8.0	3.8	8.1	5.6	3.0	11.2	7.0	5.0	12.7	1.7	8.0	0.8	3.7	4.0	14.2	2.0	2.3	9.3	2.0	2.5	3.5	7.4
Oxygenated non-terpenic compounds	2.4	1.7	0.2	2.5	1.3	1.1	1.6	1.3	0.4	0.4	0.0	0.0	1.3	2.3	0.2	0.8	0.5	1.8	0.7	0.7	2.9	1.7	1.1	0.9	1.9	1.0	1.6
Total identified	96.5	93.2	97.3	93.6	87.7	95.7	94.3	85.4	94.5	95.4	92.7	96.3	79.2	95.3	90.4	96.0	96.2	92.7	96.5	87.7	88.9	96.8	92.8	91.2	95.4	96.6	91.6
Yields (w/w vs dry material)	1.42	1.69	2.19	0.74	1.21	1.35	1.73	1.45	1.34	0.84	1.32	1.76	0.60	1.16	2.72	3.49	1.64	1.95	0.98	0.98	0.85	2.05	0.88	2.89	1.77	1.69	1.06

[a] Order of elution is given on apolar column (Rtx-1).
[b] Retention indices of literature on the apolar column (IRIa) (König et al., 2004).
[c] Retention indices on the apolar Rtx-1 column (RIa).
[d] Retention indices on the polar Rtx-Wax column (RIp).
[e] Localities of sampling.
tr: trace (%<0.05%)

Table 2: Evolution of chemical composition of fruit essential oils of *Z. leprieurii* from Colomba Station (Col 2) during their maturation

Compounds (%)	**01-sept**	**08-sept**	**22-sept**	**29-sept**	**06-oct**	**13-oct**	**20-oct**	**27-oct**
α-pinene	75.6	57.9	26.8	17.1	13.3	18.0	32.2	30.5
(E)-β-ocimene	2.9	8.9	40.5	58.2	64.0	64.0	53.1	55.7
Yields (%)	0.57	0.66	0.62	0.81	1.03	1.13	0.97	1.64

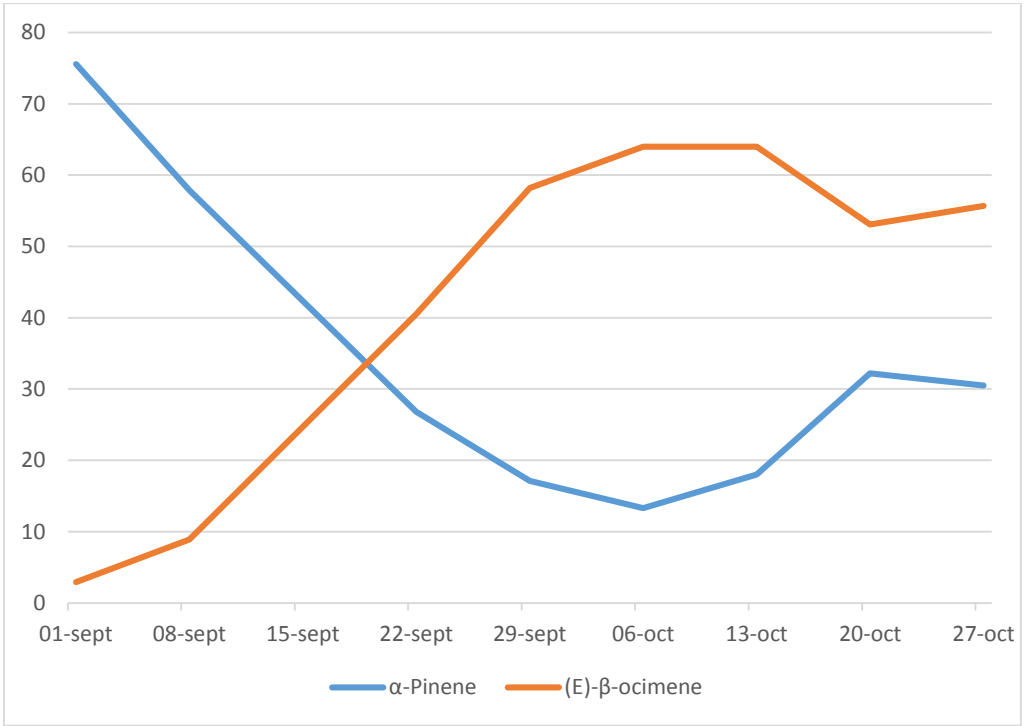

Figure 2: Evolution of the chemical composition of fruit essential oils of *Z. leprieurii* from Colomba Station (sample Col 2) during their maturation period.

DISCUSSION

Chemical variability of fruit essential oils according to geographical origins of samples

The richness of fruit essential oils in (*E*)-β-ocimene has also been described in the literature, with relative percentages between 29.4% and 80.7% (Dongmo et al., 2008; Fogang et al., 2012; Lamaty et al., 1989; Misra et al., 2013; Tatsadjieu et al., 2003). On the other hand, an interesting content of terpinolene and δ-3-carene in the fruit essential oils (26.0% and 14.5%, respectively) has also been reported by Lamaty et al. (1989) from Cameroon (Lamaty et al., 1989). However, the chemotype, geranyl acetate/geraniol/cironellol/(*E*)-β-ocimene of the sample An.2, was well as the combination of (*E*)-β-ocimene and α-pinene in significant proportions (13.4–62.6%, 14.9–40.7% respectively) has never been described in the literature to our knowledge.

Evolution of the chemical composition of essential oils of fruits during their maturation period

Despite the available reports on fennel essential oil composition, there are no previous references to the evaluation of the chemical composition of sweet fennel oil during fruit maturation. It is in this case not possible to compare our data with those of previous studies. Nevertheless, such studies were undertaken in other medicinal plants. In plants with different essential oil compositions in various ontogenetic periods such as coriander, essential oil composition is different. Schimitberger et al. (2018) reported δ-3-carene (46.67%) and α-pinene (14.98%) as the main compounds in immature fruit essential oil of *Schinus terebinthifolia* Raddi, while limonene (37.49%) and δ-3-carene (19.98%) were the main compounds of mature fruit

(Schimitberger et al., 2018). Telci et al. (2006) reported similar findings in coriander (*Coriandrum sativum* L.) with limonene contents varying from immature fruits (30%) to full mature fruits (77%) (Telci et al., 2006).

Conclusion

This study reported chemical variabilities of essentials oils of *Z. leprieurii* fruits from Senegal. According to the geographic origin of the populations, the composition of the fruit oils exhibited quantitative variations in the contents of hydrocarbon monoterpenes ((*E*)-β-ocimene and α-pinene) except for a sample from Aniak which consists mainly of oxygenated monoterpenes. The influence of the maturity stage on the chemical composition of the essential oils has resulted in an increase of oil yield and variations in the content of volatile compounds. Therefore, this study could be useful for the valuation of the essential oil of the fruits of *Z. leprieurii* which requires a judicious determination of the harvest period.

COMPETING INTERESTS

The authors state that they have no conflict of interest.

AUTHORS' CONTRIBUTIONS

YT, JP, AW and JC conceived and coordinated the study. YT and JP designed, performed, and analyzed the experiments shown in the text. All authors reviewed the results and approved the final version of the manuscript.

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