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Article

Effect of Flower Development Stages on the Dynamics of Volatile Compounds in Ylang-Ylang (*Cananga odorata*) Essential Oil

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Abstract: Several abiotic factors influence the chemical composition of essential oils. Understanding these factors is an important step in developing quality products that meet market demands. This research work aims to study the chemical composition of the essential oils of ylang-ylang (*Cananga odorata*, forma *genuina*) according to the maturity of flowers. The volatile compounds of ylang-ylang flowers from Reunion Island were extracted by hydrodistillation and the samples were analyzed by high resolution gas chromatography coupled with mass spectrometry (GC-MS) allowing for the identification of 70 volatile compounds. The chemical composition of the essential oils extracted from the flowers at 5 different stages of development varies both qualitatively and quantitatively. The volatile compounds observed belong to 4 chemical groups which are esters, alcohols, terpenes, and ether-oxides. The synthesis of light oxygenated compounds is largely the predominant chemical subgroup in all stages of development. Their relative content is considerably increased during flower ripening with a peak concentration in stage 4 (SD4). The highest concentrations of non-terpene esters and heavy oxygenated compounds are found in stages 1 (SD1) and (SD5), respectively, while no chemical subgroup is dominant in the intermediate stages 2 (SD2) and (SD3). The dynamics of volatile compounds have also been studied. This study established that the stage of development of ylang-ylang flowers significantly influences the dynamics of volatile compounds in the extracted essential oils. Total oxygenated compounds that are highly odoriferous in essential oils increase progressively and significantly with flower maturity, unlike hydrocarbon terpenes, which are less valuable in terms of their contribution to the fragrance and following opposite kinetics; suggesting that odoriferous properties increase with the development of the flower, with a higher intensity at SD4.

Keywords: essential oils; volatile compounds; *Cananga odorata*; stage of development; GC-MS

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1. Introduction

The culture of *Cananga odorata* (Lam.) Hook.f. & Thomson is widespread across the Indian Ocean and a desired essential oil for perfumery, the manufacturing of cosmetics, and soap making. It is obtained by the distillation of its flower [1]. The Union of the Comoros remains the world's largest producer of ylang-ylang essential oil in quantity and quality, and this product is the country's second largest export product, accounting for around 20% of the export goods. Therefore, controlling its quality is crucial.

The chemical variability of essential oils has been known for a very long time and may be due to various factors such as the type of plant organ from which the essential oil is extracted, the extraction time, the stage of flower development, geographical origin, extraction method, and the conditions of analysis [2–9]. The chemical composition of essential oils is largely dependent on the influence of conditions before, during, and/or after

essence extraction, including: climatic stress during growth or maturity, nutrition, harvest time, drying, post-harvest storage, and production process. Furthermore, it depends on the plant organ, ecotype, or chemical variety or chemotypes [10,11]. Because the pickers are paid per kilogram of flower picked, it has been observed in the field that the flowers harvested are always mixed (immature, mature, and overmature). Thus, the study aims to assess the degree of influence of flower development on chemical variability and thus provide scientific knowledge to actors in the ylang-ylang sector in order to be able to improve the quality of the product. It was also reported that the yield of extraction and the proportion of compounds for a given species may vary during its development, as previously observed during the vegetative evolution for three stages for *Coriandrum sativum* L. and *Pimpinella anisum* L. [12]. They may also vary according to the bracts growth in size as observed for monoterpenes in the virens subspecies of oregano (*Origanum vulgare* L. ssp. virens Hoffm. & Link) with the decrease of some hydrocarbon monoterpenes (p-cymene, γ-terpinene) while others increase (linalool, α-terpineol, thymol) [13].

Understanding the factors which influence the chemical composition of essential oils is an important step in the development of products that meet market demands [6,14]. Apart from the standards established by AFNOR (Association Française de Normalisation), very little work related to the quality of ylang-ylang essential oil has been published [15]. The present study aimed to study the dynamism of volatile compounds, chemical groups, and subgroups in relation to the development of the ylang-ylang flowers from flower bud stage to near-wilting. The influence of flower maturity on the yield and density of essential oils produced was also analyzed.

2. Materials and methods

2.1. Plant Material

The ylang-ylang flower (*Cananga odorata*, forma *genuina*) was used in its various stages of maturity as the plant material for the study (Figure 1). The choice of the flower was focused on the *genuina* form because of two forms of *Cananga odorata* (*genuina* and *macrophylla*) and the *fruticosa* variety, *genuina* is the form which gives high quality essential oils and is cultivated in the islands of Indian Ocean [1,4]. Thus, flowers at 5 different stages of development (SD) were concurrently harvested in the town of Saint-Pierre (−21.27724, 55.46689), Reunion Island, France. Because of the difficulty in obtaining flowers in different production areas of the island, and the aleatory characteristic of ylang-ylang flowering, we carried out the 3 repetitions over a sequence of 3 months. Ylang-ylang flowers were selected on the basis of their botanical characteristics to evaluate the dynamic changes and chemical variability of volatile compounds according to different development stages as defined by McGaw [16] and according to our findings in the field: SD1 (green flower buds with closed petals), SD2 (small green flowers with open petals), SD3 (less mature green flowers turning yellow), SD4 (mature yellow flowers with a red-purplish heart), and SD5 which is the last stage of growth of ylang-ylang flowers which tend towards wilting (overly mature flowers of yellow color turning to brown) as shown in Figure 1.

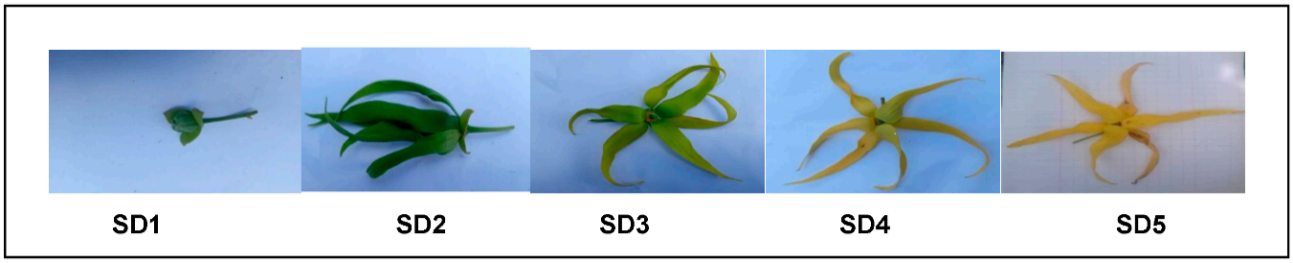

Figure 1. Ylang-ylang flowers at different stages of development.

2.2. Essential Oil Extraction

Essential oils from ylang-ylang flowers were extracted by hydrodistillation using a Clevenger-type device. An amount of 100 g of fresh flowers, 1500 mL of distilled water, and a few pumice stones were used. The cryostat ensuring the cooling of the refrigerating tube, receiving the hot oil and hydrosol vapor, was set at 4 °C.

For each replication, 100 g of flowers from each stage of development were stored at −80 °C after freezing with liquid nitrogen. The cooling of the refrigerating tube, receiving the hot oil and hydrosol vapor, was set at 4 °C. The heating temperature was set at 100 °C then lowered to 60 °C after the recording of the first drops (about 47 min later) and the distillation continued to be stopped 3 h later. Any residual water present in the sample was eliminated with sodium sulfate anhydrous, and then the essential oil obtained was stored in an amber bottle under nitrogen to avoid any possible oxidation. The extracted oils were stored at 4 °C until analysis.

2.3. Density and Yield Measurements

The measured density corresponds to the ratio of the mass of a certain volume of essential oil to the mass of an equal volume of distilled water at 20 °C, measured by an advanced analytical balance, density = m (essential oil)/m (water).

The calculation of the yield (Rdt) was carried out by the ratio of the mass of the essential oil (EO) obtained compared to that of the vegetable matter (VM), both in percentage and yield = m (EO) × 100/m (VM).

2.4. GC-MS Analysis

The samples were analyzed by gas chromatography coupled with mass spectrometry (GC-MS) using Clarus580 SQ8 (PerkinElmer, Villebon-sur-Yvette, Île-de-France, France). The mass spectrometry column was an Elite-5MS Capillary Column in fused silica (PerkinElmer, Wellesley, MA, USA) with a length of 60 m and an internal diameter of 0.25 mm with a 0.25 µmdf film thickness. The column stationary phase was 5% diphenyl/95% dimethyl polysiloxane. The GC-FID column used was an Elite-FFAP column of PerkinElmer (Wellesley, MA, USA) with a length of 30 m and an internal diameter of 0.25 mm with a 0.25 µmdf film thickness. The stationary phase is methyl siloxane, and the film thickness is 0.40 µm. The oven temperature was an initial temperature of 65 °C for 1 min, ramping up to 190 °C at a rate of 4 °C/min, then to 285 °C at a speed of 15 °C/min, and remaining at the final temperature for 2 min. The temperature of injector was 260 °C and the temperature of detector 300 °C. Helium was the carrier gas used at a constant flow rate of 0.6 mL/min. The volume injected was 0.2 mL and the injector was set to 25.0000 pts/s to inject 1 µL. For mass spectrometry conditions, the ionization electron energy was set at 70 eV and mass spectra were acquired in the range of m/z 30–400. The transfer line temperature was set at 250 °C, the ion source temperature at 230 °C and the electron multiplier at 1568. A solvent acquisition delay was set to 6 min and the total run time at 74.5 mn.

Mass spectrometry (MS) has been used for the identification of volatile compounds. Volatile compound peaks were identified using the retention indices of the peaks calculated from their retention times with a series of C7-C30 n-alkanes (49451 U, Sigma-Supelco, Saint-Quentin-Fallavier, France) tested experimentally and analyzed under the same conditions. These retention indices were then compared with those reported in the literature, in particular by the NIST MS database 17 driven by NIST MS search 2.3. Identification was also based on computational matching of mass spectra with NIST Mass Spectrometry Data Center, NIST_ri, Wileyregistry8e, Aro_perkinelmer, PerkinElmer Fragrances & Flavours 2013, mainlib, replib, and terpene libraries. The data processing software used in this study for the characterization was TurboMass V6.1.2 (PerkinElmer, Villebon sur Yvette, France).

The quantification of compounds was performed using gas chromatography (GC). The relative abundances of the identified compounds correspond to the ratio between the areas of the peaks of each compound compared to the total sum of the areas of all the integrated peaks. To be able to suppress background noise peaks and column bleeding, the

noise/area threshold and the bunching factor have been set so that the minimum area of a peak that must appear on the quantification report is greater than or equal to 100. Data were processed using TotalChrom V6.3.2 software (PerkinElmer, Villebon sur Yvette, France).

2.5. Statistical Analysis

Various statistical analyses were carried out under the R.4.1.0/Rstudio software for the reliability of the results on possible significant differences between the maturation stages studied. The correlations of the dynamism of these compounds have also been studied by Pearson correlation coefficient. The main statistical tests carried out in this study are: Fisher Test (p-value) to confirm the existence of a significant difference (p-value < 0.005) in chemical composition between the stages; analysis of variance (ANOVA) to gain information about the relationship between the dependent and independent stages of development; least significant difference (LSD) to indicate that at least one stage differs from the other stages when the difference between the means of the stages is significant; and principal component analysis (PCA) to visualize correlations between variables, and identify the homogeneity and heterogeneity of flower development stages.

3. Results and discussion

3.1. Density and Ylang-Ylang Essential Oil Extraction Yield

The influence of flower development for the ylang-ylang essential oil production was first studied in relation to the density and the yield of extraction by hydrodistillation of the 5 different stages of flower development (Figures 2 and 3). The density increases gradually and significantly during the maturity of the flower from the bud SD1 to SD4, before decreasing at SD5 (Figure 2). The rate of this gradual increase in density is 2.86% of SD1 (prime fraction with a density of 0.943) to SD4 (extra-high fraction with a density of 0.970). It can be seen that the density of SD5 is slightly higher than that of SD3 but significantly higher than the first two SD1 and SD2.

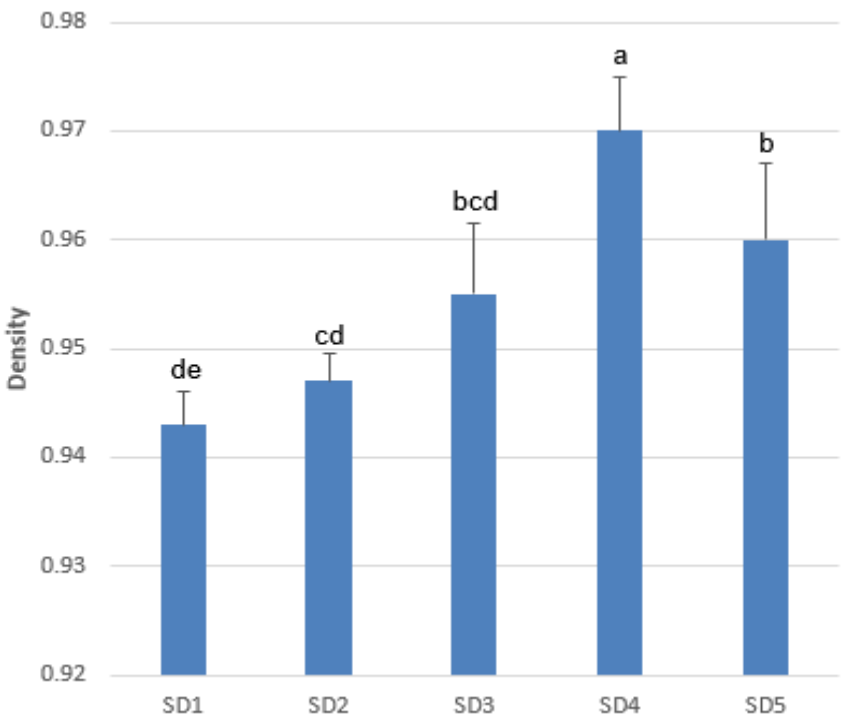

Figure 2. Density (m (essential oil)/m (water)) of oils obtained at different stages of development. Values are mean ± standard error of mean. The different letters (a, b, c, d and e) above the standard deviations of the histograms mean that the densities obtained between the stages of flower development are significantly different ($p \leq 0.05$).

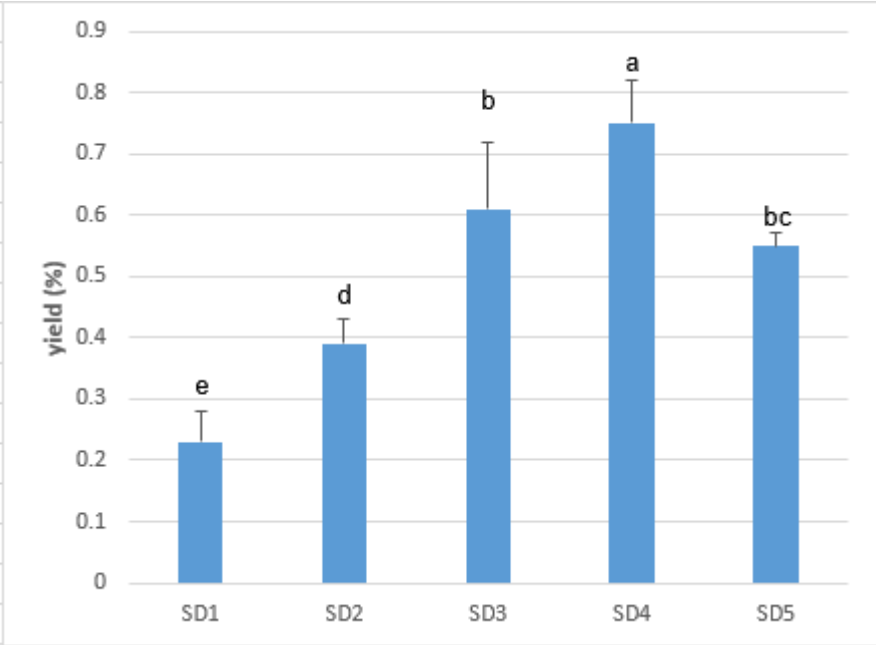

Figure 3. Yield (m (EO) × 100/m (VM)) of oils obtained at different stages of development. Values are mean ± standard error of mean. The different letters (a, b, c, d and e) above the standard deviations of the histograms mean that the yields obtained between the stages of flower development are significantly different ($p \leq 0.05$).

The yield follows the same evolution with a very significant increase of 3.43 times higher from SD1 to SD4 (0.21% yield at SD1 to 0.72% at SD4) (Figure 3). Unlike density, it is the SD3 presents a slightly higher yield than the SD5 but also significantly higher than the first two stages.

Similar to these results, the effects of plant maturity on essential oils yield was also reported on flower heads of *Arnica montana* L. and *Arnica chamissonis* Less. cultivated for industry, showing a highest content of volatile oils in the flower heads from the full flowering phase [17]. The yield of essential oils also ranged from 0.22% to 0.78% at the five leaves appearance stage and 100% flowering stage, respectively for the species *Achillea filipendulina* Lam. studied at four different phenological stages [18].

These results clearly show that the mature flowers of yellow color with a purplish-red heart (SD4) are higher in density and in yield compared to the other stages. This increase in density, one of the qualitative parameters of essential oils, suggests that the leading compounds of ylang-ylang essential oils may gain in proportion during the growth of the flower.

3.2. Diversity of Volatile Compounds in Ylang-Ylang Flowers

Previous research has extensively investigated the chemical compositions of essential oils from the *Cananga odorata* flower using different extraction methods [3,19–23] and some studies have taken into consideration the volatile compounds present at different stages of ylang-ylang flower development. However, so far none has used the hydrodistillation method which is the prevalent method with the stills of the Indian Ocean islands, an area of mass production for ylang-ylang essential oils [14] and this is why we used this processing method for the study. The advantage of this method is due to the temperatures used, which are below 100 °C: most organic compounds have their boiling temperatures approaching 200 °C. This explains why this technique is very widely used in perfumery, where odorous molecules are often fragile and cannot withstand high temperatures.

A total of 70 compounds, representing 99.59–99.87% of the total detected compounds, were identified using gas chromatography-mass spectrometry (GC-MS) and quantified

by GC-flame ionization detector (FID). The results showed clear differences in chemical composition among the essential oils obtained at five different stages of flower development. Most of the compounds were only detected in several, but not all stages of flower development, and some of the compounds were exclusively detected in only one particular stage. Table 1 shows the chemical composition of ylang-ylang essential oils at different stages of development. SD1 and SD2 at the beginning of flowering are dominated by linalool, geranyl acetate, and benzyl benzoate (compounds whose content > 10%). The two last mature stages, SD4 and SD5, were dominated by geranyl acetate, linalool, and benzyl acetate. Regarding intermediate stage SD3, it is only dominated by geranyl acetate and linalool, i.e., with a lesser amount of very abundant benzyl benzoate and benzyl acetate than in the primary and mature stages, respectively. This result might suggest that benzyl benzoate was one of the constituents that contributed significantly during bud development through SD1 to SD2 while benzyl acetate might contribute significantly to the characteristic fragrance of mature flowers because during this time, the aroma of ylang-ylang fragrance was very much intense. It is important to mention that a previous ylang-ylang essential oil characterization by GC×GC-TOFMS conducted by Brokl et al. showed that this essential oil has a diverse chemical composition and can consist of up to 161 individual compounds including those detected as traces [22]. Despite this high number of volatile compounds, only 15 are retained by the French standard ISO-3063 as being characteristic of ylang-ylang from the Comoros archipelago and Madagascar [24]. These observations agree with those found by Afshari et al. [18] on different phenological stages of *Achillea filipendulina*, whose number of volatile compounds identified in the five leaves appearance stage (SD1: 40 compounds) was higher than those identified in the full-flowering stage (SD4: 28 compounds).

Table 1. Proportion of chemical groups in ylang-ylang essential oils in relation to the flower maturity stage.

Chemical Groups	Stage of Development				
	SD1	**SD2**	**SD3**	**SD4**	**SD5**
Esters	43.80 ± 6.87 ^{abcd}	51.89 ± 8.53 ^{bcde}	48.74± 7.62 ^{cde}	52.81 ± 7.81 ^{de}	59.14 ± 8.39 ^{e}
Alcohols	33.98 ± 4.52 ^{abcd}	28.22 ± 4.93 ^{bcde}	32.52± 6.17 ^{cde}	30.11 ± 6.25 ^{de}	25.50 ± 5.03 ^{e}
Terpenes	18.44 ± 1.58 ^{a}	10.43 ± 1.18 ^{bc}	8.43 ± 0.80 ^{cde}	6.06 ± 0.67 ^{de}	5.38 ± 0.66 ^{e}
Ether oxides	2.33 ± 1.42 ^{ab}	5.24 ± 2.74 ^{bcde}	6.80 ± 3.39 ^{cde}	8.57± 4.54 ^{de}	6.56± 3.75 ^{e}
Phenols	1.74 ± 2.94 ^{a}	2.64 ± 0.55 ^{bcde}	2.4 ± 1.0 ^{cde}	2.09 ± 0.94 ^{de}	2.14 ± 0.72 ^{e}
Oxides	0.83 ± 0.37 ^{a}	1.25 ± 0.23 ^{a}	0.38 ± 0.62 ^{a}	0.81 ± 0.44 ^{a}	0.54 ± 0.42 ^{a}
Nitrogeneted compounds	-	-	-	0.06 ± 0.06 ^{a}	0.14 ± 0.09 ^{a}
Alcenes	-	-	0.02 ± 0.03 ^{a}	0.32 ± 0.39 ^{d}	0.03 ± 0.05 ^{a}
Aldehydes	0.10 ± 0.02 ^{a}	0.82 ± 0.37 ^{a}	0.13 ± 0.07 ^{a}	0.09 ± 0.16 ^{a}	-

Values are mean ± standard error of means; means followed by a different letters (a, b, c, d and e) within the column mean that the proportions of chemical groups obtained between the stages of flower development are significantly different ($p \leq 0.05$).

Figure 4 shows that SD1, SD2, SD3, SD4, and SD5 consisted of 54, 51, 47, 43, and 43 identified compounds, accounting for 99.59%, 99.74%, 99.68%, 99.87%, and 99.82% of the total essential oil composition, respectively. SD1 is the richest in number of volatile compounds and this number decreases along flower development. For all the stages of flower development, 31 compounds are common, while five are specific for SD1, six for SD3, and one for SD4. No specific compounds were characterized for flowers with open petals or intermediate SD2 and at the very mature stage SD5.

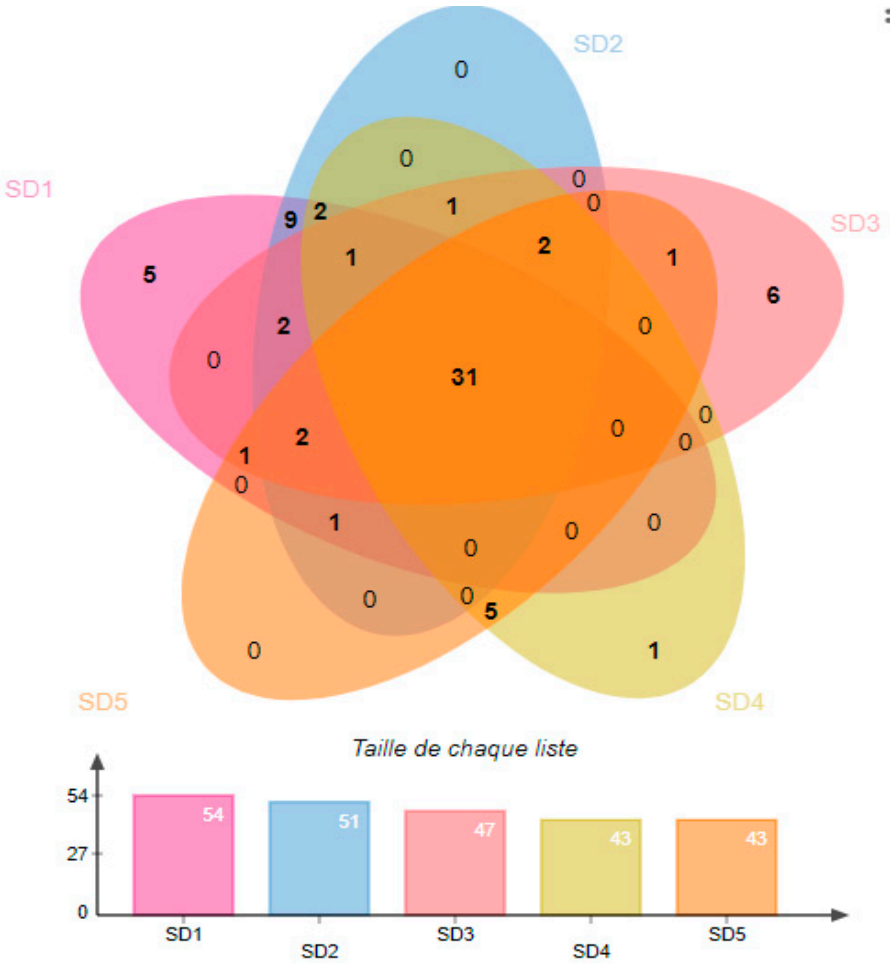

Figure 4. Jvenn diagram showing the number of specific or shared compounds by stage of development.

This decrease in the number of compounds with the development of the flower can be explained by the fact that certain compounds of the embryonic stage are precursors for the synthesis of others. Actually, Schade et al. [25] reported on fragrance volatiles of developing and senescing carnation flowers that flower buds do not have a scent, and that the characteristic fragrance of a flower appears during anthesis as the petals open. For example, flower fragrance compounds are formed from precursor compounds in the flower buds of *Jasminum polyanthum* upon treatment with a crude enzyme preparation from opening flowers [26]. This suggests that the final biosynthetic reactions are developmentally regulated and only occur as the flowers begin to open [25]. It has also been reported in the literature that the volatile components of *Nicotiana suaveolens* flowers increase dramatically post-anthesis [27]. The increase in concentration of certain volatile compounds with flower maturity may also be due to precursors of other plant organs. Indeed, it has also been proposed that precursors of floral scent are formed in leaves and transported through the phloem to flowers [28].

These results differ from those described by Stashenko et al. [3] on essential oil resulting from three stages of development of ylang-ylang flowers from Colombia, obtained by combined steam distillation-solvent extraction. In this study, only 40 compounds were detected in the extract of small green flowers (SD2), whereas the extracts obtained from flowers of intermediate maturity (SD3) and from completely matured yellow flowers (SD4 and SD5) presented 51 and 65 volatile compounds, respectively, at concentrations > 0.1 ppm [3]. The difference can be explained by the detection threshold considered. On the other hand, such variations in the number of volatile compounds might be attributed in part to the harvest time, geographic origin, agro-climatic conditions, and extraction methods [29–32].

Among essential oils, flower essential oils are particularly known to present a variation of composition depending on environmental conditions [6,21,31,33–36].

3.3. Evolution of Groups, Subgroups, and Oxygenated Compounds between Stages of Development

Table 1 lists the chemical groups and subgroups impacted or not by the maturity of the flower in this study. The multiple comparison tests show that out of seven chemical groups studied, the four most abundant groups (esters, alcohols, hydrocarbon terpenes, and ether-oxides), representing more than 95% of the sample, are significantly influenced by the stages of development of the flower (p-value < 0.05). A general upward trend is observed for ester and ether oxide groups and to a lesser extent for phenols and nitrogenated compounds. Data indicates that the ester content of the least abundant stage SD1 (43.80%) is significantly lower than that of the most abundant SD5 (59.14%) with no notable difference in the three middle stages. Ether-oxides also gradually increase from SD1 (2.33%), which is significantly different from the last three stages (6.80%, 8.57% and 6.56%). Conversely, a decreasing tendency is observed in the proportion of alcohol and hydrocarbon terpene groups along flower maturation. The alcohols decrease slightly during the growth of the flower with a significant difference between the first stage (33.98%) and the last (25.50%). The greatest variability of the chemical groups is observed for the hydrocarbon terpenes whose level gradually and significantly decreases from SD1 (18.44%) to SD5 (5.38%). The multiple comparison test shows major differences between SD1 and the other four higher stages (SD2, SD3, SD4 and SD5). SD2 is also significantly different from mature SD4 and SD5 (Table 1).

These data are consistent with a previous work reporting that some terpenes that are very present at the beginning of flowering in *Michelia alba* species have completely disappeared during the maturation stage such as α-cubebene, β-farnesene, and Chamigrene, and others have disappeared at the end of maturity like Germacrene D and α-caryophyllene [37].

Among chemical subgroups (Table 2 below), monoterpene and non-terpene esters show an increasing tendency with flower development but this is not significant according to the statistical tests carried out. The sum of the two subgroups thus contributed to the significant increase in esters during flower maturity. The level of sesquiterpene alcohols decreased slightly and progressively from SD1 (33.78%) to SD5 (24.52%) with a significant difference between these two stages only. A slight unexpected drop in the content of these compounds should be noted in SD2. The significant decrease of the total alcohol content from SD1 to SD5 was due to the sesquiterpene alcohols according to the ANOVA and LSD tests processed. The non-overlapping decrease of hydrocarbon sesquiterpenes is significantly observed between SD1 with all the other later stages. The content of these compounds in the last two mature stages (SD4 and SD5) is practically four times lower (4.42 and 4.44%, respectively) than that of the primitive SD1 (16.65%). The gradual drop in hydrocarbon monoterpenes content during floral development from SD1 to SD5 is only 0.76% when this rate is 13.06% for hydrocarbon sesquiterpenes (Table 2).

Table 2. Proportion of chemical subgroups of ylang-ylang essential oils in relation to the flower maturity stage.

Chemical Subgroups	**Stage of Development**				
	SD1	**SD2**	**SD3**	**SD4**	**SD5**
Sesquiterpenes alcohol	33.08 ± 5.08 ^{abcd}	27.51 ± 5.6 ^{bcde}	31.53 ± 7.04 ^{cde}	29.38 ± 7.13 ^{de}	24.52 ± 5.74 ^{e}
Sesquiterpenes	16.65 ± 1.76 ^{a}	9.04 ± 1.34 ^{bcde}	7.06 ± 0.91 ^{cde}	4.42 ± 0.76 ^{de}	4.44 ± 0.75 ^{e}
phenylpropanoïde	-	2.34 ± 0.51 ^{bcde}	2.18 ± 0.72 ^{cde}	1.44 ± 0.20 ^{de}	1.82 ± 0.54 ^{e}
Non-terpene ester	25.23 ± 4.85 ^{a}	24.81 ± 4.07 ^{a}	22.89 ± 2.63 ^{a}	27.39 ± 3.68 ^{a}	32.64 ± 4.52 ^{a}
Monoterpenic Ester	18.57 ± 5.71 ^{a}	27.08 ± 1.87 ^{a}	25.85 ± 6.29 ^{a}	25.42 ± 4.86 ^{a}	26.50 ± 2 ^{a}
Monoterpenes	1.79 ± 0.34 ^{a}	1.40 ± 0.18 ^{a}	1.37 ± 0.13 ^{a}	1.10 ± 0.17 ^{a}	1.12 ± 0.12 ^{a}
Monoterpenic alcohols	0.90 ± 0.44 ^{a}	0.65 ± 0.38 ^{a}	0.88 ± 0.39 ^{a}	0.73 ± 0.38 ^{a}	0.98 ± 0.50 ^{a}

Values are mean ± standard error of means; means followed by a different letter within the column are significantly different ($p \leq 0.05$).

Hydrocarbon sesquiterpenes are therefore responsible for the significant drop in hydrocarbon terpenes throughout flower development. These results are confirmed by correlation values and *p*-values between terpenes and sesquiterpenes (R^2 = +0.996; *p*-value = 6.44×10^{-15}), while the decrease in hydrocarbon terpenes is not correlated with that of monoterpenes (R^2 = +0.4086167; *p*-value = +0.1305). Hydrocarbon sesquiterpene results are similar to those found by Li et al. [38] from *Luculia pinceana* flower whose sesquiterpene content was much higher in the bud stage (44.41%) and significantly dropped during flower development: at 13.37%, 16.79%, and 13.91%, respectively, for SD2, SD3 and SD4.

The oxygenated compounds (light and heavy) are a large proportion of the chemical compounds identified in the samples (Table 3) and represent at least 80% of the volatile content, contributing strongly to the organoleptic properties of the essential oil [39].

Table 3. Sum of light and heavy oxygenated compounds of ylang-ylang essential oils in relation to the flower maturity stage.

Oxygenated Compounds	Stage of Development				
	SD1	SD2	SD3	SD4	SD5
Light	54.47 ± 5.31 [a]	70.15 ±6.76 [b]	80.39 ± 7.3 [cde]	86.83 ± 7.52 [d]	84.76 ±7.19 [e]
Heavy	26.60 ± 4.24 [a]	19.02 ±3.66 [b]	10.70 ± 1.77 [cde]	7.46 ± 1.28 [d]	9.53 ± 1.60 [e]

Values are mean ± standard error of means; means followed by a different letter within the column are significantly different ($p \leq 0.05$).

They are significantly influenced by the maturity of the flower, representing approximately 81.07% of the sample at the start of flowering, increasing gradually to reach 94.29% at the two mature stages combined (SD4 and SD5), i.e., an increase of +13.22%. The dynamism of light oxygenated compounds, representing at least 66% of this family, is observed with a significant increase of +31.96% from stage SD1 (54.47%) to SD4 (86.43%). Even if a slight decrease is later observed at SD5 (84.76%), the jump is still important compared to the two intermediate stages SD2 (70.15%) and SD3 (80.39%). The heavy oxygenated elements present in large quantities at the embryonic stage follow opposite kinetics (26.60% from SD1 to 9.53% for SD5). Overall, total oxygenated compounds increase progressively with a percentage of evolution of +12.59% during floral development (81.7%, 89.35%, 91.09%, 94.29%, and 94.29%). This gradual and significant increase is due to volatile compounds from various chemical families, in particular certain esters, alcohols, ether-oxides, and phenols.

3.4. Evolution of Individual Volatile Compounds

The dynamics of the 15 characteristic compounds of ylang-ylang (forma *genuina*) according to the AFNOR standard was studied. Figure 5 shows that the dynamism of the 5/15 of these compounds are significantly influenced by the stage of development.

Thereby, benzyl acetate becomes 10 to 13 times more concentrated in SD4 and SD5 relative to its initial content in SD1, following a progressive increase from SD1 to SD5 (1.12%, 2.44%, 6.49%, 11.69%, and 14.84%). This result is similar to that reported for the flower of *Jasminum auriculatum* on which the emission of benzyl acetate showed much higher levels at fully bloomed SD5 [40]. P-cresyl methyl ether which barely quadruples its concentration from SD1 (2.27%) to SD4 (8.29%) and methyl benzoate which evolve from 0.87% (SD1) to 5.5% (SD4) follow this same increasing kinetics but with a slight decrease in SD5 at 6.27% and 4.94%, respectively. E-cinnamyl acetate, unidentified at the primitive stage of flower development (SD1), increases its content gradually from SD2 (1.73%) until reaching 4.06% at SD5. Beta-caryophyllene was exclusively present at SD1 (1.04%). These results agree with the observations of Verdoonk et al. [37] who reported that in few stages of *Petunia* flower development, the peak of methyl benzoate became more obvious during the expansion of the corolla limbs and opening of the flower, and then increased when the flower developed further.

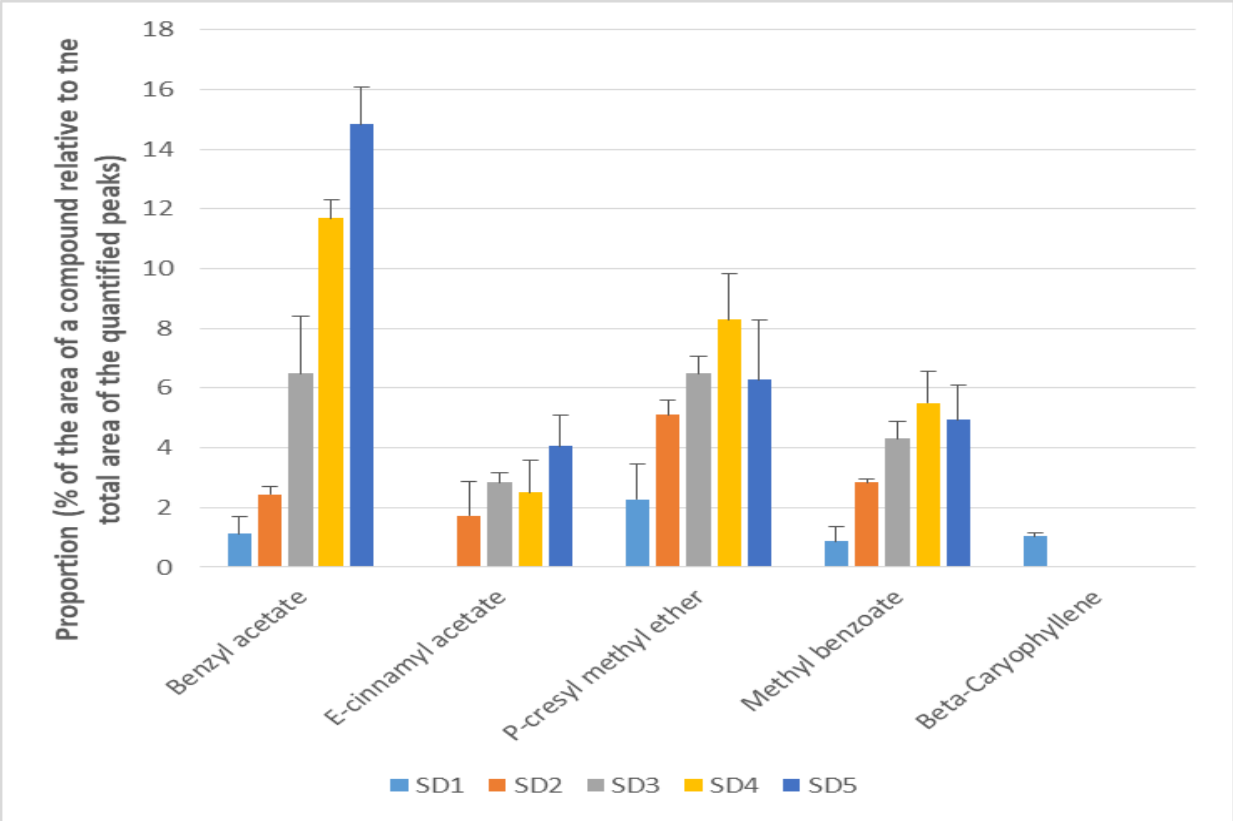

Figure 5. Kinetics of five compounds among the 15 characteristic of ylang-ylang oils according French Association for Standardization.

The volatile compounds (identified by GC-MS) of ylang-ylang essential oils in relation to the flower maturity stage are reported in Table 4 below.

Although the embryonic stage (SD1) with 54 chemical compounds is quantitatively richer in chemical composition than the mature stages, being about 13 compounds more at SD1 compared to SD4 and SD5; the relative percentage of numerous main compounds are lower at SD1. This is much illustrated by the five compounds listed above. We also note that geranyl acetate, linalool, and anethole gradually evolve respectively from 18.57% (SD1) to 26.5% (SD5), from 18.88% (SD1) to 25.92% (SD4), and 0.07% to 0.29% (SD5). The observation made by Barman et al. [41] on Jasminum species agree with these results as the emission of linalool was also higher at flower blooming in SD4. The two phenolic compounds (trans-isoeugenol and p-cresol) characterized in this work are absent at the bud stage (SD1). P-cresol undergoes increasing kinetics from SD2 to SD5. Some compounds follow a progressive decrease until mature SD4 such as benzyl benzoate (16.36%—13.92%—6.72%—4.71%), phenyl ethyl acetate (1.27%—1.07%—0.34%—0.09%), germacrene D (3.22%—1.65%—0.9%—0.07%), and delta-cadinene (1.38%—0.36%—0.25%—0.14%). The decrease in germacrene-D content during flower maturity agrees with the results of Verdoonk et al. [37]. Some volatile compounds overlap, including benzyl salicylate, geraniol, alpha-terpineol, junenol, and trans-isoeugenol. Schade et al. [25] and Sanimah et al. [42] reported a similar pattern in carnation flowers and Michelia alba flowers. It was found that the steady state level of 10 volatiles including benzaldehyde, benzyl benzoate, and caryophyllene changes independently as the flowers develop and senesce, suggesting that the synthesis of these volatile compounds was physio-biochemically regulated. Some compounds are specific to the embryonic stage only, such as phenol-2-methoxy-4-(2-propenyl)–acetate (2.83%), beta-caryophyllene (1.04%), and alpha-cubebene (0.12%), with others characterized under form of traces (geranyl benzoate; trans-nerolidol . . .). Cubenol (0.09%—0.06%), Cubenene (0.15%—0.02%), and beta-elemene (0.16%—0.03%) are only present in the first two SD1

and SD2. On the other hand, α-Muurolene (0.78%—1.17%—0.25%), Zonarene (0.06%—0.18%—0.1%), and cinnamyl alcohol (0.02%—0.03%—0.11%) are specific to the first three stages of growth of the flower. The only nitrogenous compound identified in this study, 2-Phenylnitroethane is exclusively presently on the two mature stages, SD4 and SD5 (0.04%—0.14%), in agreement with previous study [3]. On the other hand, relative content of δ-cadinene, cubebol, and cubenol at the bud stage were higher than that at the other stages of flower development, which is suitable with the results obtained by Li et al. [38] on Luculia pinceana flower.

Figure 6 below shows the signature of total volatile compounds characterized by GC-MS. More than 10 compounds showed a clear regular increasing dynamic with flower development, this includes some of the main components of the ylang-ylang essential oil (benzyl acetate, cinnamyl acetate, P-cresyl methyl ether, linalool, methyl benzoate, alpha-terpineol, p-Cresol, methyl-anisoate, alloaromadendrene, 2-phenylnitroethane, 4-Isopropenyl-1-Methyl-1-Cyclohexene, citral and Benzoic acid, and 2-methoxy methyl ester). The increase of compounds such as Trans-Trans-Farnesal and Cinnamyl Alcohol stops at the intermediate stages SD2 and SD3, which follows a similar pattern to flower maturity. An opposite specific kinetic is observed for two esters (benzyl benzoate and acetate phenylethyle), 2 alcohols farnesol, and tau-muurolol) and five hydrocarbon terpenes (alpha-farnesene, beta-carryophyllene, D-germacrene, delta-elemene, and delta-cadinene) which sign a decreasing dynamic. Finally, a last group can be identified for compounds which have an important signature exclusively on one particular stage (tau-muurolol at SD1), on two stages (anethole at SD3 and SD5) or on three stages (nerol at SD1, SD2 and SD4) and seem to be synthesized independently of flower development.

3.5. Principal Component Analysis of Maturity of Ylang-Ylang Flowers

A principal component analysis by covariance matrix (PCA) was carried out on the 70 volatile components characterized by GC-MS to compare the profile of volatile compounds between maturation stages. The variance contribution rate of PC1 (principal component 1) and PC2 were 43.4% and 12.4%, respectively (Figure 7).

The PCA results indicated that the samples from the five stages of flower maturation occupied relatively independent spaces in the distribution map. It suggests that the aromatic noses are considerably different between the stages. Across all the flower-life stages, the bud stage is distinctly dissimilar to the four full-flowering stages. Variations of the volatile compositions were also apparently involved in the maturity stages of flower from *Luculia pinceana* [38]. The same phenomena are observed in other plants, such as the flowers of *Penstemon digitalis* [43].

The distribution of the different stages of development (individuals), according to the two dimensions, clearly shows the existence of a difference in the chemical composition of the volatile compounds between the stages. Indeed, SD1 which is positioned to the right of the axes (dimension 1), differs from the four other upper stages almost all projected on the same dimension. The intermediate stages of flower development (SD2 and SD3), being at the intersections of the axes 1 and 2, are also distant from two mature stages (SD4 and SD5) placed clearly to the right (PC1). There is a slight interference of a test (repeatability) from SD3 to the last two (SD4 and SD5).

Figure 8 below shows that methyl benzoate, E-cinnamyl acetate, P-cresyl methyl ether, benzyl acetate, geranyl acetate, linalool, anethole, and p-cresol are projected to axis 1 and are highly positively correlated with mature stages SD4 and SD5. While (E.E)-alpha-farnesene, beta-caryophyllene, germacreneD, benzyl benzoate, (E.E)-farnesyl acetate, geraniol, tau-cadinol, and cubenene also correspond to axis 1 but correlate positively to initial bud stage (SD1). Volatile compounds that had high positive scores on PC 2 included D-limonene, phenylethyl acetate, beta-pinene, alloaromadendrene, citral, cinnamyl alcohol, and phenyl methyl ester.

Table 4. Composition of ylang-ylang essential oils in relation to the flower maturity stage, analyzed by GC-MS.

Group	Subgroup	Compound	Stage of Development				
			SD1	SD2	SD3	SD4	SD5
Ester	Monoterpenic ester	Geranyl acetate *	18.57 ± 5.71 [a]	27.08 ± 1.87 [a]	25.85 ± 6.29 [a]	25.42 ± 4.86 [a]	26.5 ± 2 [a]
Ester	Non-terpene ester	Benzyl acetate *	1.12 ± 0.56 [ab]	2.44 ± 0.28 [b]	6.49 ± 1.92 [c]	11.69 ± 0.61 [d]	14.84 ± 1.26 [e]
Ester	Non-terpene ester	Benzyl benzoate *	16.36 ± 1.75 [ab]	13.92 ± 1.47 [b]	6.72 ± 3.93 [cde]	4.71 ± 2.45 [de]	5.83 ± 3.14 [e]
Ester	Non-terpene ester	Methyl benzoate *	0.87 ± 0.49 [a]	2.85 ± 0.12 [bc]	4.31 ± 0.59 [cde]	5.5 ± 1.07 [de]	4.94 ± 1.17 [e]
Ester	Non-terpene ester	E-cinnamyl acetate *	-	1.73 ± 1.43 [bcd]	2.85 ± 0.33 [cde]	2.51 ± 1.07 [de]	4.06 ± 1.03 [e]
Ester	Non-terpene ester	Benzyl salicylate *	1.51 ± 0.42 [a]	2.58 ± 0.81 [a]	1.66 ± 0.95 [a]	1.79 ± 1.22 [a]	2.38 ± 1.75 [a]
Ester	Non-terpene ester	E,E, Farnezyl acetate *	0.83 ± 0.46 [a]	0.44 ± 0.12 [a]	0.46 ± 0.31 [a]	0.22 ± 0.1 [a]	0.31 ± 0.23 [a]
Ester	Non-terpene ester	Benzoic acid, 2-methoxy-, methyl ester	-	-	-	0.04 ± 0.06 [de]	0.03 ± 0.05 [e]
Ester	Non-terpene ester	Phenol, 2-methoxy-4-(2-propenyl)-, acetate	4.53 ± 1.23 [a]	-	-	-	-
Ester	Non-terpene ester	Acetate phenyl ethyle	1.27 ± 2.19 [a]	1.07 ± 1.85 [a]	0.34 ± 0.47 [a]	0.09 ± 0.09 [a]	0.18 ± 0.07 [a]
Ester	Non-terpene ester	Acetic acid, hexyle ester	-	-	-	0.11 ± 0.19 [a]	0.11 ± 0.2 [a]
Ester	Non-terpene ester	Acetate Neryle	-	-	0.02 ± 0.03 [a]	-	0.03 ± 0.05 [a]
Ester	Non-terpene ester	Methyl-Anisoate	-	-	-	0.04 ± 0.06 [a]	0.05 ± 0.05 [a]
Ester	Non-terpene ester	3-Hexen-1-ol, benzoate, (Z)-	0.02 ± 0.03 [a]	0.02 ± 0.01 [a]	-	-	0.02 ± 0.03 [a]
Ester	Non-terpene ester	9-Octadecenoic acid (Z)-, phenylmethyl ester	-	-	0.06 ± 0.1	-	-
Ester	Non-terpene ester	Geranyl benzoate	0.03 ± 0.05 [a]	0.02 ± 0.01 [a]	-	-	-
Alcohol	Sequiterpene alcohol	Geraniol *	2.24 ± 1.84 [abc]	2.07 ± 1.76 [bc]	2.97 ± 0.15 [c]	1.46 ± 1.19 [de]	1.93 ± 0.8 [e]
Alcohol	Sequiterpene alcohol	Farnesol *	2.84 ± 0.63 [ab]	1.16 ± 0.97 [bce]	1.09 ± 0.66 [cde]	0.32 ± 0.29 [d]	0.83 ± 0.5 [ed]
Alcohol	Sequiterpene alcohol	Linalool *	18.88 ± 4.12 [a]	20.5 ± 3.04 [a]	25.69 ± 1.18 [a]	25.92 ± 2.11 [a]	20.9 ± 5.64 [a]
Alcohol	Sequiterpene alcohol	Tau-Cadinol	2.96 ± 1.64 [a]	0.59 ± 0.32 [bcde]	0.96 ± 0.61 [cde]	0.34 ± 0.07 [de]	0.42 ± 0.19 [e]
Alcohol	Sequiterpene alcohol	Tau-Muurolol	2.76 ± 1.35 [a]	0.84 ± 0.06 [bcde]	0.74 ± 0.61 [cde]	0.19 ± 0.17 [de]	0.43 ± 0.2 [e]
Alcohol	Sequiterpene alcohol	Guaiol	0.84 ± 0.21 [a]	0.07 ± 0.02 [bcde]	0.03 ± 0.03 [cde]	0.02 ± 0.0 [de]	0.03 ± 0.01 [e]
Alcohol	Sequiterpene alcohol	Alpha-terpineol	0.53 ± 0.47 [a]	0.45 ± 0.41 [a]	0.74 ± 0.13 [a]	0.51 ± 0.48 [a]	0.91 ± 0.25 [a]
Alcohol	Sequiterpene alcohol	Junenol	0.58 ± 0.33 [a]	0.05 ± 0.09 [bcde]	0.05 ± 0.09 [cde]	0.12 ± 0.21 [de]	0.04 ± 0.04 [e]
Alcohol	Monoterpenic alcohol	Nerol	0.33 ± 0.43 [a]	0.21 ± 0.36 [a]	0.07 ± 0.06 [a]	0.71 ± 1.1 [a]	0.06 ± 0.09 [a]
Alcohol	Sequiterpene alcohol	Alpha-Elemol	0.08 ± 0.03 [a]	0.01 ± 0.01 [bcde]	-	-	-
Alcohol	Sequiterpene alcohol	10-Epi-γ-eudesmol	0.09 ± 0.1 [a]	0.03 ± 0.02 [a]	0.02 ± 0.01 [a]	-	0.02 ± 0.03 [a]
Alcohol	Sequiterpene alcohol	Cubenol	0.17 ± 0.17 [a]	0.06 ± 0.1 [a]	-	-	-
Alcohol	Sequiterpene alcohol	Epi-Cubenol	0.09 ± 0.11 [a]	0.04 ± 0.06 [a]	0.02 ± 0.01 [a]	0.01 ± 0.01 [a]	0.01 ± 0.02 [a]
Alcohol	Sequiterpene alcohol	Caryophyllenyl alcohol	0.06 ± 0.1 [a]	0.04 ± 0.07 [a]	-	-	-
Alcohol	Sequiterpene alcohol	β-Acorenol	0.25 ± 0.22 [a]	0.31 ± 0.54 [a]	-	0.51 ± 0.59 [a]	-
Alcohol	Sequiterpene alcohol	Cubebol	-	-	0.03 ± 0.05	-	-
Alcohol	Sequiterpene alcohol	Epi-Cubebol	0.03 ± 0.05	-	-	-	-
Alcohol	Sequiterpene alcohol	Trans-Nerolidol	0.02 ± 0.03	-	-	-	-
Alcohol	Non-terpene alcohol	Cinnamyl Alcohol	0.02 ± 0.03 [a]	0.03 ± 0.06 [a]	0.11 ± 0.2 [a]	-	-
Alcohol	Non-terpene alcohol	2-Furanmethanol, 5-Ethenyltetrahydro-α,α,5-Trimethyl-, Trans-	-	-	0.07 ± 0.13	-	-
hydrocarbon Terpene	Sesquiterpene	E,E, alpha-farnesene *	6.05 ± 2.9 [ab]	2.32 ± 1.85 [bcd]	2.41 ± 0.82 [cd]	0.94 ± 0.71 [de]	1.16 ± 0.89 [e]
hydrocarbon Terpene	Sesquiterpene	Beta-Caryophyllene *	1.04 ± 0.11 [a]	-	-	-	-
hydrocarbon Terpene	Sesquiterpene	GermacreneD *	3.22 ± 1.83 [abc]	1.65 ± 1.62 [bcde]	0.9 ± 0.74 [cde]	0.07 ± 0.12 [de]	0.26 ± 0.16 [e]
hydrocarbon Terpene	Sesquiterpene	Alpha-Humulene	3.08 ± 4.81 [a]	3.65 ± 1.15 [a]	2.71 ± 2.71 [a]	2.28 ± 0.94 [a]	2.69 ± 1.39 [a]
hydrocarbon Terpene	Sesquiterpene	Delta-Elemene	0.07 ± 0.01 [a]	0.06 ± 0.08 [b]	-	-	-
hydrocarbon Terpene	Sesquiterpene	Delta-cadinene	1.38 ± 0.63 [a]	0.36 ± 0.32 [bcde]	0.25 ± 0.21 [cde]	0.14 ± 0.12 [de]	0.26 ± 0.11 [e]

Table 4. *Cont.*

Group	Subgroup	Compound	Stage of Development				
			SD1	SD2	SD3	SD4	SD5
hydrocarbon Terpene	Sesquiterpene	Copaene	0.33 ± 0.31 [ab]	0.13 ± 0.14 [bcde]	0.04 ± 0.04 [cde]	0.02 ± 0.01 [de]	0.04 ± 0.08 [e]
hydrocarbon Terpene	Sesquiterpene	Alpha-Cubebene	0.12 ± 0.06 [a]	-	-	-	-
hydrocarbon Terpene	Sesquiterpene	Ylangene	0.02 ± 0.02 [a]	0.02 ± 0.02 [a]	0.02 ± 0.04 [a]	-	-
hydrocarbon Terpene	Sesquiterpene	Beta-Elemene	0.16 ± 0.14 [a]	0.03 ± 0.05 [a]	-	-	-
hydrocarbon Terpene	Sesquiterpene	Alloaromadendrene	-	0.04 ± 0.07 [a]	0.38 ± 0.66 [a]	1.3 ± 2.25 [a]	-
hydrocarbon Terpene	Sesquiterpene	α-Muurolene	0.78 ± 0.7 [a]	1.17 ± 2.03 [a]	0.25 ± 0.44 [a]	-	-
hydrocarbon Terpene	Sesquiterpene	Gamma-Cadinene	0.16 ± 0.28 [a]	0.26 ± 0.46 [a]	-	0.08 ± 0.14 [a]	-
hydrocarbon Terpene	Sesquiterpene	Zonarene	0.06 ± 0.07 [a]	0.18 ± 0.31 [a]	0.1 ± 0.17 [a]	0.01 ± 0.01 [a]	0.01 ± 0.01 [a]
hydrocarbon Terpene	Sesquiterpene	Cubenene	0.15 ± 0.21 [a]	0.02 ± 0.03 [a]	-	-	-
hydrocarbon Terpene	Sesquiterpene	Alpha-Cadinene	0.03 ± 0.06 [a]	0.02 ± 0.04 [a]	-	-	-
hydrocarbon Terpene	Monoterpene	Alpha-Pinene	0.94 ± 0.58 [a]	0.6 ± 0.03 [a]	0.47 ± 0.09 [a]	0 ± 0.1 [a]	0.43 ± 0.04 [a]
hydrocarbon Terpene	Monoterpene	Beta-Myrcene	0.27 ± 0.12 [a]	0.3 ± 0.02 [a]	0.23 ± 0.07 [a]	0.17 ± 0.15 [a]	0.17 ± 0.15 [a]
hydrocarbon Terpene	Monoterpene	Beta-Pinene	0.4 ± 0.25 [a]	0.35 ± 0.01 [a]	0.23 ± 0.19 [a]	0.33 ± 0.1 [a]	0.25 ± 0.08 [a]
hydrocarbon Terpene	Monoterpene	D-Limonene	0.18 ± 0.04 [a]	0.15 ± 0.02 [a]	0.36 ± 0.49 [a]	0.05 ± 0.09 [a]	0.15 ± 0.1 [a]
hydrocarbon Terpene	Monoterpene	Gamma-terpinene	0.02 ± 0.03 [a]	-	0.07 ± 0.12 [a]	-	-
Ether-oxyde	-	P-cresyl methyl ether *	2.27 ± 1.18 [ab]	5.11 ± 0.5 [bcde]	6.48 ± 0.58 [cde]	8.29 ± 1.93 [de]	6.27 ± 2.9 [e]
Ether-oxyde	-	Anethole	0.07 ± 0.06 [ab]	0.1 ± 0.09 [bcde]	0.32 ± 0.17 [cde]	0.19 ± 0.17 [de]	0.29 ± 0.09 [e]
Oxide	Monoterpenic oxide	Eucalyptol	0.83 ± 0.37 [a]	1.25 ± 0.23 [a]	0.38 ± 0.62 [a]	0.63 ± 0.59 [a]	0.54 ± 0.42 [a]
Phenol	-	Trans-Isoeugenol	1.7 ± 2.94 [a]	2.42 ± 0.55 [bcde]	2.18 ± 0.72 [cde]	1.89 ± 0.73 [de]	1.82 ± 0.64 [e]
Phenol	-	p-Cresol	-	0.04 ± 0.03 [a]	0.22 ± 0.28 [a]	0.2 ± 0.21 [a]	0.32 ± 0.08 [a]
Aldehyde	Monoterpenic aldehyde	Citral	0.07 ± 0.07 [a]	0.06 ± 0.06 [a]	0.11 ± 0.2 [a]	0.01 ± 0.01 [a]	-
Aldehyde	-	Trans-Trans-Farnesal	0.03 ± 0.05 [a]	0.76 ± 1.31 [a]	-	-	-
Aldehyde	-	Benzene bytanal	-	-	0.02 ± 0.03	-	-
Alcane	-	2-Phenylnitroethane	-	-	-	0.04 ± 0.07 [a]	0.14 ± 0.09 [a]
Alcane	-	Heptacosane	-	-	0.04 ± 0.07	-	-
Alcene	-	Bicyclo [4.1.0] Hept-3-Ene, 3,7,7-Trimethyl	-	-	-	0.3 ± 0.36	-
Alcene	-	4-Isopropenyl-1-Methyl-1-Cyclohexene	-	-	-	0.02 ± 0.03 [a]	0.03 ± 0.05 [a]
Alcene	-	Cyclohexene, 3-Methyl-6-(1-Methylethylidene) -	-	-	0.02 ± 0.03	-	-

Relative percentage (peak area relative to the total peak area, %). Values are mean± standard error of means; means followed by a different letter within the column are significantly different ($p \leq 0.05$). SD1-stage 1 (green flower buds with closed petals), SD2-stage 2 (small green flowers with open petals), SD-stage 3 (less mature green flowers turning yellow), SD4-stage 4 (mature yellow flowers with a red-purplish heart), SD5-stage 5 which is the last stage of growth of ylang-ylang flowers (overly mature flowers of yellow color turning to brown which tend towards wilting). * compounds characteristics of ylang-ylang oils according to the French Association for Standardization.

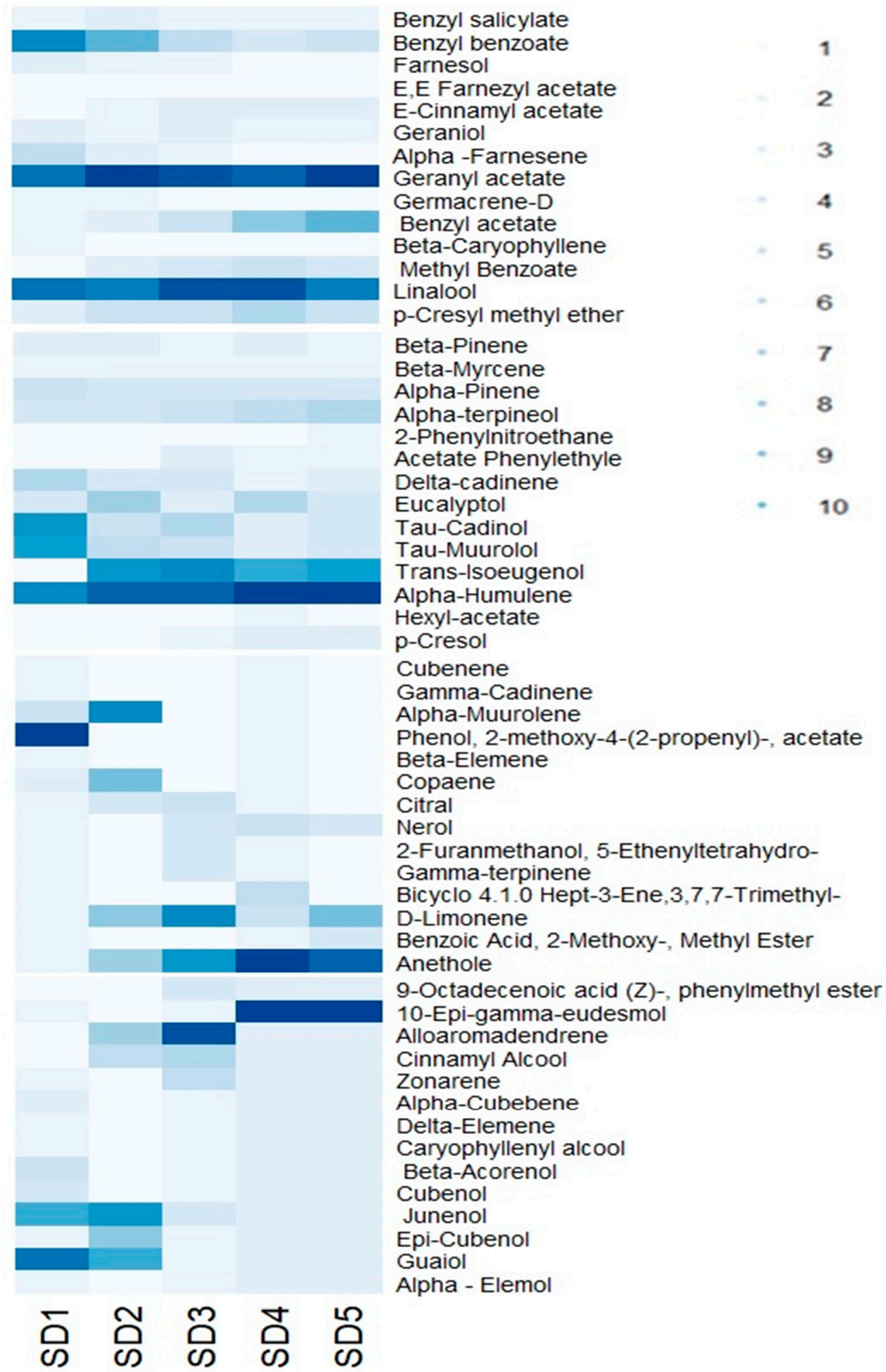

Figure 6. Heatmap of total volatile compounds dynamics characterized by GC-MS in five different stages of flower development. The proportion of each compound in SD1 was considered as reference.

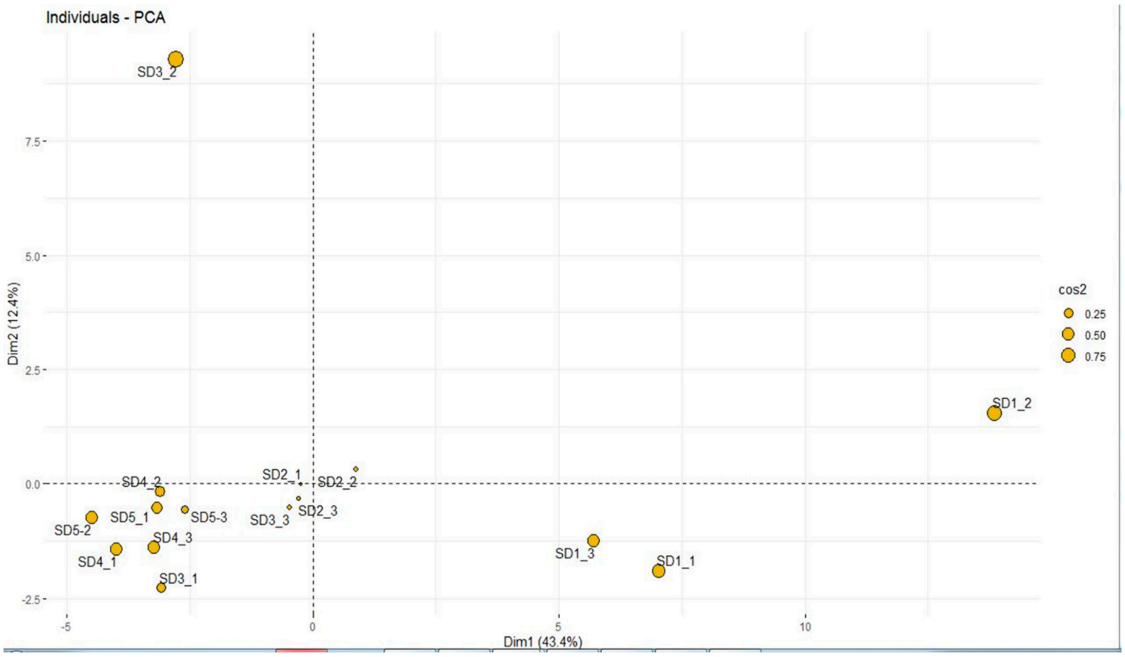

Figure 7. Principal component score chart of the five different stages of development of ylang-ylang flowers.

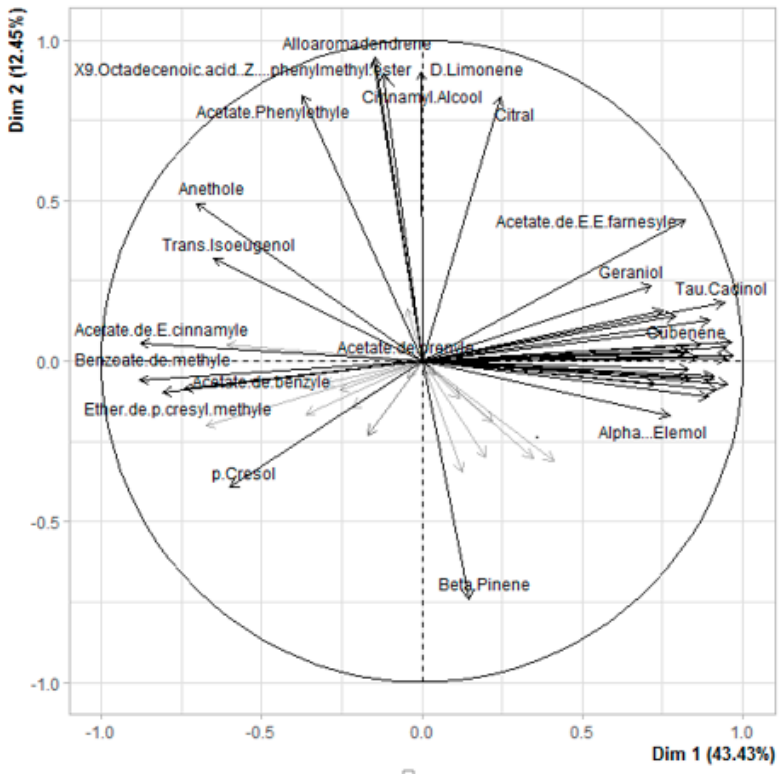

Figure 8. Principal component analysis of the 70 identified compounds on essential oils extracted at five different stage of development of ylang-ylang flowers.

The calculation of correlation coefficients by the Pearson method showed that methyl benzoate is negatively correlated with Benzyl benzoate ($R^2 = -0.950$) and (E,E)-farnesol ($R^2 = -0.946$). That suggests, in accordance with the ACP, that the progressive reduction of benzyl benzoate and (E,E)-farnesol necessarily favors the increase of methyl benzoate with the development of the flower. The first is also correlated with p-cresyl methyl ether

(R^2 = +0.922) and (E,E)-farnesol is positively correlated with benzyl benzoate, R^2 = +0.955 (Table 1).

Following the above, it can be deduced that the volatile compounds having contributed to a significant increase in esters and ether-oxides during flower maturity are mainly benzyl acetate, methyl benzoate, E-cinnamyl acetate, P-cresyl methyl ether, and anethole. The development of the flower has also led to a significant decrease in certain volatile compounds, namely, tau-cadinol, tau-muurolol, farnesol, guaiol, junenol, and geraniol for alcohols and (E,E)-alpha-farnesene, Beta-Caryophyllene, D-Germacrene, Copaene, Delta-cadinene, Alpha-Cubebene, and Delta-Elemene for hydrocarbon terpenes (sesquiterpenes). We also note a very significant increase in the light oxygenated compounds which are the majority for all the stages, at the time when the heavy oxygenated compounds were decreasing significantly. Finally, the two phenolic compounds and the only nitrogen compound identified in this work are absent at the embryonic stage; and other volatile compounds have overlapping kinetics.

Esters (benzyl acetate, geranyl acetate, salicylate methyl, etc.) are one of the major groups contributing to the smell of ylang-ylang, whose acetate benzyl is the most significant. Benzyl acetate and geranyl acetate provide the fruity-flowery body of ylang-ylang. Ethers, like p-cresyl methyl ether, is characteristic of medicinal smell diffuse and penetrating ylang-ylang. Linalool, a monoterpene alcohol which increases gradually but not significantly, is responsible for the fresh and flowery notes. Hydrocarbon sesquiterpenes (caryophyllene, cadinene, farnesene, etc.) create the base of the warm notes on which the other molecules cling. Phenols (p-cresol, iso-eugenol, etc.) are responsible for the spicy, balsamic warm notes characteristic of the smell of ylang-ylang [1,3,14,44–46]. Benzyl acetate, geranyl acetate, and linalool represent 38.57%, 50.74%, 58.03%, 63.03% and 62.24% of the volatile compounds from SD1 to SD5. We therefore assume that the fresh, fruity, and flowery notes of ylang-ylang essential oils connected to these molecules increase with the development of the flower from SD1 to SD4 then decrease slightly at SD5. Also, the p-cresyl methyl ether content is 2.27%, 5.11%, 6.48%, 8.29% and 6.27 from SD1 to SD5, suggesting that the medicinal diffuse and penetrating smell of ylang-ylang increases with flower maturity from SD1 to SD4 then decreases slightly at SD5. The content of two identified phenolic compounds (p-cresol and trans-isoeugenol) increased from SD1 to SD2 to remain almost identical until stage SD5 (1.74%, 2.64%, 2.4%, 2.09% and 2.14%), which should provide intense spicy and balsamic warm notes from SD2 to SD5 compared to the embryonic stage. This is unlike hydrocarbon sesquiterpenes, which are also linked to the base warm notes and which decrease during flower maturity. This decrease should not have a strong influence on the organoleptic properties since hydrocarbon terpenes do not contribute significantly to the flavor or smell of the essential oils because of hydrolysis reactions occurring when exposed to heat and light, and thus, is occasionally associated with a loss of aromatic and flavor qualities of the essential oil [47,48]. The oxygenated compounds are usually appreciated by the industry for their aroma and quality [49,50]. Hydrocarbon monoterpenes are also less valuable than oxygenated compounds in terms of their contribution to the fragrance of the essential oil. Conversely, the oxygenated compounds are highly odoriferous and, hence, the most valuable [39]. All of these observations lead to the hypothesis that the organoleptic properties increase with the development of the flower, with a higher intensity at SD4.

The volatile organic compounds are produced by the plants mainly to deter herbivores, act as repellents, and help in pollination [40]. All of these compounds belong to two distinct groups: terpenes and terpenoids and aromatic and aliphatic compounds. Terpenes consist of a combination of five carbon units called isoprene. The biosynthesis of terpenes consists of the synthesis of isopentenyl diphosphate (IPP) and the repetitive addition of IPP to form prenyldiphosphate, a precursor of the different classes of terpenes. The prenyldiphosphate is then modified by the intervention of specific enzymes in order to form the terpene skeleton. Finally, a secondary enzymatic activity modifies the terpene skeletons and attributes their different functional properties. The great diversity of terpenes therefore comes from the large number of enzymes involved in their synthesis process

but also due to the fact that a terpene synthase has the ability to form multiple products from a single substrate [51–53]. The abundance of these compounds at the beginning of flowering suggests that the biosynthesis of these hydrocarbon terpenes is necessary for the development of the flower at the bud stage. Then, the steady-state levels of these hydrocarbon terpenes decrease as the flowers develop and senesce, suggesting that their synthesis is developmentally regulated and the final biosynthetic reactions of these volatile compounds occurs as the flowers begin to open. Wanatebe et al. [26] have proposed that the enzymes mediating these terminal reactions are either newly induced or activated during flower opening. Aromatic compounds on the other hand, are derived from phenylpropane (shikimate pathway). The aromatics biosynthetic pathway is usually separated from the biosynthetic pathway terpenes in plants, but both pathways can coexist in some plants [51]. The shikimate pathway leads to the formation of three aromatic amino acids—the phenylalanine, tyrosine, and tryptophan—which, in addition to intermediates of this pathway, are at the base of many aromatic compounds [54]. Regarding aliphatic compounds, they are, for the most part, volatile derivatives of fatty acids mainly resulting from the degradation of linolenic acid (lipoxygenase pathway) [53,55].

Volatile compounds may not always be released from the floral tissue immediately after biosynthesis, but rather are retained quite often in floral tissues in soluble phase and latter emitted as scent upon vaporization [56]. Further, several volatile compounds are also stored in vacuoles as water soluble glycosides [26,41], and upon physiological necessity, cleaved out by hydrolytic enzymes and subsequently released as fragrant molecules from floral tissue [57].

4. Conclusions

The results of this study confirm the presence of a chemical variability of certain terpenes and oxygenated compounds, predominant molecules in the organoleptic qualities of essential oils extracted at five different stages of flower development. It also indicates that the optimum stage for obtaining essential oils of higher yield and high density is the yellow flowers with a purplish red heart (SD4), which is also richer in light oxygenated compounds, representing up to 80% of the samples studied. Thereby, to improve practices and produce high quality essential oils, it would be best for producers/distillers in the ylang-ylang sector to harvest only the flowers of the SD4 stage and/or rigorously sort the flowers picked to take only this SD4 into account for distillation. However, the differences observed for these two parameters are low with the SD3 (in yield) and SD5 (in density), unlike in primary stages SD1 and SD2. The presence of one or more characteristic compounds of a specific stage, as well as the knowledge and control of the aromatic profiles of these stages, would constitute a considerable asset for the development of products that meet the market requirements of this product which has become emblematic in the Indian Ocean. A deep study of olfactometry on the essential oils of ylang-ylang related to the stage of the development of flowers will allow us to highlight the predominant compounds of a good quality essential oil.

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Data Availability Statement: The authors declare that the data of the study are stored on the analytical instruments of the laboratories of the UMR-Qualisud of CIRAD-Réunion. They confirm the availability of providing this data to the Journal if necessary.

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Appendix A

Table 1. Pearson coefficients depicting linear correlations between 14 major volatile compounds according to the stage of development. Numbers **1** to **14** mean the 14 compounds identified by GC-MS among the 15 characteristics of ylang-ylang oils according to the French Association for Standardization. * If *p*-value of 0.05, ** if 0.01 and *** if 0.001.

	Compounds	1	2	3	4	5	6	7	8	9	10	11	12	13	14
1	Methyl benzoate	-	0.81 ***	−0.75 ***	−0.95 ***	−0.21	0.52 *	−0.77 ***.	−0.71 **	−0.83 ***	−0.94 ***	0.67 **	−0.71 **	0.92 ***	0.77 ***
2	Benzyl acetate	0.81 ***	-	−0.56	−0.82 ***	0.06	0.37	−0.57 *	−0.66 **	−0.75 ***	−0.78 ***	0.30	−0.89	0.62 *	0.81 ***
3	Farnesyl acetate	−0.75 ***	−0.56	-	0.72 **	0.30	−0.71 **	0.59 **	0.63 **	0.83 ***	0.85 ***	−0.68 **	0.59 *	−0.78 ***	−0.56 *
4	Benzyl benzoate	−0.95 ***	−0.82 ***	0.72 **	-	0.34	−0.53 *	0.64 **	0.63 **	0.79 ***	0.95 ***	−0.70 **	0.65 **	−0.83 ***	−0.68 **
5	Benzyl salicylate	−0.21	0.06	0.30	0.34	-	−0.24	−0.01	−0.08	0.04	0.24	−0.60 **	0.18	−0.34	0.34
6	Geranyl acetate	0.52 *	0.37	−0.71 **	−0.53 *	−0.01	-	−0.59 *	−0.27	−0.61 *	−0.68 **	0.27	−0.34	0.38	0.57 *
7	Beta-Caryophyllene	−0.77 ***	−0.57 *	0.59 *	0.64 **	−0.24	−0.59 *	-	0.69 **	0.73 ***	0.73 ***	−0.41	0.54*	−0.69 **	−0.87 ***
8	D-Germacrene	−0.71 **	−0.66 **	0.63 **	0.63 **	−0.08	−0.27	0.69 **	-	0.82 ***	0.71 **	−0.55 *	0.64 **	−0.66 **	−0.69 **
9	Apha-Farnesene	−0.83 ***	−0.75 ***	0.83 ***	0.79 ***	0.04	−0.61 **	0.73 ***	0.82 ***	-	0.86 ***	−0.63 **	0.72 **	−0.77 ***	−0.78 ***
10	Farnesol	−0.94 ***	−0.78 ***	0.85 ***	0.95 ***	0.24	−0.68 **	0.73 ***	0.71 **	0.86 ***	-	−0.70 **	0.66 **	−0.83 ***	−0.73 ***
11	Linalool	0.67 **	0.30	−0.68 **	−0.70 **	−0.60 *	0.27	−0.41	−0.55 *	−0.63 **	−0.70 **	-	−0.21	0.81 ***	0.23
12	Geraniol	−0.71 **	−0.89 ***	0.59 *	0.65 **	−0.18	−0.34	0.54 *	0.64 **	0.72 **	0.66 **	−0.21	-	−0.62 **	−0.82 ***
13	P-cresyl methyl ether	0.92 ***	0.62 ***	−0.78 ***	−0.83 ***	−0.34	0.38	−0.69 **	−0.66 **	−0.77 ***	0.83 ***	−0.81 ***	−0.62 **	-	0.62 **
14	E-cinnamyl acetate	0.77 ***	0.81 ***	−0.56 *	−0.68 **	0.34	0.57 *	−0.87 ***	−0.69 **	−0.78 ***	−0.73 ***	0.23	−0.82 ***	0.62 **	-

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