Effects of four drying methods on *Amomum villosum* Lour. ‘Guiyan1’ volatile organic compounds analyzed via headspace solid phase microextraction and gas chromatography–mass spectrometry coupled with OPLS-DA

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This paper analyzed the effects of four drying methods (heat pump drying, hot air drying, sun drying, and freeze drying) on the volatile organic compounds (VOCs) in fresh ‘Guiyan1’ *Amomum villosum* Lour. Via separation, component differentiation, and overall variance analysis via HS-SPME-GC/MS coupled with OPLS-DA, 133 kinds of VOCs, mainly composed of hydrocarbons, esters, and alcohols, were identified. The differences in ‘Guiyan1’ processed by freeze-drying and the other three drying methods were the most significant and easily distinguishable. The main VOCs in the dried samples were bornyl acetate and 2-bornanone, with the largest increase in 2-bornanone and the largest decrease in bicyclogermacrene. The obtained data provided guidance for optimizing the processing and storage of ‘Guiyan1’.

1. Introduction

*Amomums* are the dried and ripe fruits of cardamom plants of the ginger family, including *Amomum villosum* Lour., *Amomum longiligulare* T. L. Wu, and *Amomum villosum* Lour. var. xanthioles T. L. Wu et Senjen (called *Amomum xanthioles* Wallich (Zingiberaceae)).”1,2 *Amomums* are mainly distributed in tropical and subtropical regions, China’s southern regions (Guangdong, Guangxi, and Hainan Provinces), and Southeast Asian countries, such as Laos and Vietnam.3 The cultivation climate and soil environment have significant effects on the quality of *amomums*,4 the optimal conditions being reported in Yangchun, Guangdong Province of China.5

*Amomums* belong to the “Four Southern Medicines” in China. They contain polysaccharides, terpenoids, flavonoids, volatile essential oils, and other active ingredients6, which are effective in the treatment of spleen and stomach stagnation, antibacterial and anti-inflammatory properties, and analgesia.7–10 The total sugar content in fresh amomums was estimated as 37.10% in studies,11,12 including 10.69% obtained via ultrasonic-assisted hydro extraction and alcoholic precipitation method, and 3.97% via alkali extraction method.13 It was reported in ref. 14 that polysaccharides in amomums had inhibitory effects on transplanted tumors S180 and H22 in mice and had strong in vitro antioxidant, antibacterial and antitumor properties. Ding et al.15 identified eight terpene components from amomums using HR-ESI-MS and other spectroscopic techniques. They also performed hypoglycemic activity tests in an STC-1-cell model and two enzymatic (GPa and PTP1B) models, proving that (1R,2S,4R,7S)-vicodiol 9-O-β-D-glucopyranoside 6 had significant GPa inhibitory activity with an IC50 of 78.6 μ mol L⁻¹. Li et al.16 determined the total flavonoid content of Changtai amomums to be approximately 2.85 mg g⁻¹. The essential oil of *S. aureus* was found to be another crucial active component17 mainly composed of camphor, lobster acetate, lobster, n-citron, and camphor18,19 which inhibited the growth of *Staphylococcus aureus*,20 *Pseudomonas aeruginosa*, and *Candida albicans*21 and had sound effects in anti-inflammatory and skin wound repair,22,23 preservation of fruits and vegetables such as strawberries24 and improvement of alfalfa silage quality.25

The quality of *amomums* strongly depends on their aroma characteristics, which, in turn, are affected by the variety, fruit/seed/peel/other parts,6 origin27, and drying methods.28 The

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research results\textsuperscript{26} showed that the composition and content of volatile organic components (VOCs) in seeds, peels, and rhizomes of \textit{Amomum villosum} L. Significantly differed, exhibiting bornyl acetate contents of 52.46, 40.35, and 18.34\%, respectively. Various drying methods (including sun drying, hot air drying, heat pump drying, and freeze-drying ones) were found to have different effects on the quality of \textit{Amomum villosum}. The comparative analysis of five drying techniques of \textit{Amomum villosum} fruits performed by Ai et al.\textsuperscript{28} revealed that freeze drying achieved the best color retention, the lowest shell burst ratio, and the best retention of flavor profiles due to the complete glandular trichome structure.\textsuperscript{28}

The main means of analyzing the volatile aroma of amomums are headspace solid-phase microextraction gas-phase mass spectrometry (HS-SPME-GC-MS),\textsuperscript{29} comprehensive two-dimensional gas chromatography-quadrupole time-of-flight mass spectrometry (GC × GC-QTOF-MS),\textsuperscript{30} and gas chromatography-mass spectrometry (GC-MS)-electronic nose (E-nose).\textsuperscript{31} This study adopted the SPME-GC-MS technique combined with OPLS-DA to investigate the effect of four drying methods on VOCs in amomums to provide basic data support for their processing.

2. Materials and methods

2.1 Main materials

\textit{Amomum villosum} Lour. ‘Guiyan1’ was collected from Bu Lian Tun, Shang Meng Village, Ping Shan Township, Long A County, Guangxi Zhuang Autonomous Region of China, and was the primary local amomum cultivar. The samples were collected in clean self-sealing bags and stored at −20 °C in the refrigerator for later use.

2.2 Dry method

2.2.1 Heat pump drying. Referring to the method used by Ai et al.,\textsuperscript{28} some parameters have been modified. A total of 500 g of fresh amomum was laid flat on the mesh tray and dried in a heat pump drying oven (L3. 5TB1, Guangdong Weierxin Industry Co., Ltd., China) at a temperature of 55 °C and a humidity of 5\% for 12 h, packed in a vacuum packing bag, and stored at room temperature until use.

2.2.2 Hot air drying. Referring to the method used by Ai et al.,\textsuperscript{28} some of the parameters have been modified: 500 g of fresh amomum was laid flat on the mesh tray and dried in a hot air drying oven (DHG-9140A type, Shanghai Yiheng Scientific Instruments Co., Ltd., China) at a temperature of 65 °C for 16 h, packed in a vacuum packing bag, and stored at room temperature until use.

2.2.3 Sun drying. Fresh amomum (500 g) was laid flat on the mesh tray, dried under the sun at a temperature of 20–28 °C for 50 h, packed in a vacuum packing bag, and stored at room temperature until use.

2.2.4 Freeze-drying. Freeze-dried fresh amomum (500 g) was laid flat on the mesh tray and dried in a freeze-dryer (Mill Rock ST85B3, MILLROCK, USA) at a temperature of −40 °C and a vacuum of 13.33 Pa for 50 h, packed in a vacuum packing bag, and stored at room temperature until use.

2.3 Analysis method

2.3.1 Sample pretreatment and injection conditions. Approximately 1.0 g of the ‘Guiyan1’ samples was cut into small pieces (approximately 0.5 cm × 0.5 cm) into a 20 mL headspace vial, 10 μL of 100 mg L\textsuperscript{−1} 2-octanol internal standard solution was added, sealed and mixed well, and the DVB/CAR/PDMS (50/30 μm) extraction head was extracted at 55 °C for 30 min with an equilibration time of 5.0 min and thermal desorption at 220 °C for 5.0 min. The data were collected together with the start-up at the same time.

2.3.2 GC-MS conditions. HP-5ms column (30 m × 0.25 mm, 0.25 μm), inlet temperature 220 °C, and the column with programmed ramp-up: start at 50 °C, hold for 1 min, ramp up to 280 °C at 10 °C min\textsuperscript{−1}, hold for 5 min, carrier gas helium, injection mode with splitting, total flow rate 34.0 mL min\textsuperscript{−1}, column flow rate 1.00 mL min\textsuperscript{−1}, linear velocity 36.3 cm s\textsuperscript{−1}, septum purge flow 3.0 mL min\textsuperscript{−1}, splitting ratio 30. The ion source temperature was 220 °C, the interface temperature was 280 °C, the solvent delay time was 1.00 min, and the mass scan range (m/z) was 35–450 amu.

2.4 Data processing and analysis

All samples were measured three times in parallel, and to ensure reproducibility of the data, compounds that appeared in at least two parallel trials were used as the analytes. The data obtained were analyzed qualitatively using the NIST Chemical Structures library (2014) and the Wiley Library (9). OriginPro, SIMCA 14.1, and Photoshop were used for plotting, data processing, and statistical analysis.

3. Results and analysis

3.1 Composition and relative content of VOCs in ‘Guiyan1’ obtained by different drying methods

In total, 133 VOCs in ‘Guiyan1’ samples obtained by different drying methods were identified and listed in Table 1. In particular, 66, 67, 67, 89, and 69 VOCs were identified in heat pump drying, hot air drying, sun drying, freeze-drying, and fresh ‘Guiyan1’, respectively. Fig. 1(A) shows that the number of hydrocarbon compounds in ‘Guiyan1’ was the highest, while the number of hydrocarbon species in the freeze-drying method was the highest (31). Compared with fresh ‘Guiyan1’, the number of VOC species in samples obtained via the freeze-drying method increased, and the total VOC content in samples obtained by the other three methods decreased. The number of alcohols and hydrocarbon compounds increased in freeze-drying and decreased in other methods; the number of ketones, esters, and other compounds grew in all samples obtained via the four drying methods. As shown in Table 1 and Fig. 1(B), the total content of VOCs was the highest in freeze-dried samples, namely (870 ± 120) μg g\textsuperscript{−1}, and the lowest in sun-dried ones, namely (570 ± 143) μg g\textsuperscript{−1}. Compared with fresh ‘Guiyan1’, freeze-dried samples had higher contents of
Table 1  The composition and relative contents of VOCs in Amomum villosum. ‘Guiyan1’ samples obtained via four drying methods

| Retention No. | CAS# | Molecular formula | Name                                      | VHPD ± | HAD | SD | FD | FA ± | Classification       |
|---------------|------|-------------------|------------------------------------------|--------|-----|----|----|------|----------------------|
| 1             | 509-14-8 | CN₄O₈     | Tetranitro-methane                        | 0.51 ± 0.39 | 0.62 ± 1.01 | 0.41 | Others |
| 2             | 75-07-0  | C₆H₆O    | Acetaldehyde                              | 0.07 | 0.09 | 0.2 | 0.57 | ± 0.02 | Aldehydes ± 0.03 |
| 3             | 64-17-5  | C₆H₄O    | Ethanol                                   | 0     | 0    | 0  | 0   | 0.04 | Aldehydes ± 0.05 |
| 4             | 590-86-3 | C₁₀H₁₈O    | 3-Methyl-butanal                          | 0     | 0.07 | 0  | 0   | 0.06 | Aldehydes ± 0.05 |
| 5             | 1066-42-8 | C₆H₆O₂Si  | Dimethyl-silanediol                       | 4.68 | 3.72 | 4.31 | 4.79 | 4.54 | Esters |
| 6             | 108-88-3 | C₆H₆     | Toluene                                   | 0.75 | 0.67 | 1.4 | 1.49 | ± 1.03 | Hydrocarbons |
| 7             | 66-25-1  | C₆H₁₂O    | Hexanal                                   | 0.1  | 0.09 | 0.12 | 0.1  | ± 0.01 | Aldehydes |
| 8             | 123-92-2 | C₆H₁₄O₂   | 3-Methyl-1-butanol, acetate               | 0.15 | 0.07 | 0  | 0   | 0.05 | Esters |
| 9             | 111-71-7 | C₆H₆O    | Heptanal                                  | 0.07 | 0.04 | 0  | 0   | 0.05 | Aldehydes ± 0.01 |
| 10            | 514-14-7 | C₁₃H₁₈    | 2,7,7-Trimethylbicyclo[2.2.1]hept-2-ene   | 0     | 0    | 0  | 0   | 0.05 | Hydrocarbons ± 0.04 |
| 11            | 508-32-7 | C₁₃H₁₈    | Tricyclene                                | 0.28 | 1.22 | 0.61 | 0.1  | ± 0.06 | Hydrocarbons |
| 12            | 2867-5-2 | C₁₀H₁₈    | 3-Thujene                                 | 2.73 | 1.14 | 1.2 | 4.05 | ± 7.94 | Hydrocarbons |
| 13            | 80-56-8 | C₁₃H₁₈    | α-Pinene                                  | 12.82 | 12.75 | 11.75 | 13.86 | 38.5 | Hydrocarbons ± 0.88 |
| 14            | 36262-09-6 | C₁₀H₁₈    | 4-Methylene-1-[1-methylthyl]-bicyclo[3.1.0]hex-2-ene | 1.92 | 1.3 | 1.94 | 2.36 | ± 0.71 | Hydrocarbons |
| 15            | 79-92-5 | C₁₃H₁₈    | Camphene                                  | 14.01 | 22.36 | 18.47 | 4.88 | ± 3.93 | Hydrocarbons ± 0.11 |
| 16            | 100-52-7 | C₆H₆O     | Benzaldehyde                              | 0.41 | 0.44 | 0  | 0   | 0.22 | Aldehydes ± 0.19 |
| 17            | 3387-41-5 | C₁₃H₁₈    | 4-Methylene-1-[1-methylthyl]-bicyclo[3.1.0]hexane | 11.35 | 5.91 | 3.64 | 16.1 | ± 0.6 | Hydrocarbons |
| 18            | 18172-67-3 | C₁₃H₁₈    | 6,6-Dimethyl-2-methylenecyclo[3.1.1]heptane | 24.84 | 17.69 | 15.55 | 32.92 | 117.7 | Hydrocarbons |
| 19            | 123-35-3 | C₁₃H₁₈    | β-Myrcone                                 | 14.64 | 18.56 | 17.5 | 7.4  | ± 16.6 | Hydrocarbons ± 0.2 |
| 20            | 123-96-6 | C₆H₆O     | 2-Octanol                                 | 0.79 | 0.79 | 1.1 | 1.36 | ± 0.02 | Alcohol |
| 21            | 124-13-0 | C₁₃H₁₈    | Octanal                                   | 0     | 0    | 1.07 | 2.01 | ± 0.94 | Aldehydes ± 1.24 |
| 22            | 99-83-2 | C₁₃H₁₈    | α-Phellandrene                            | 0     | 0    | 0  | 2.46 | ± 0.46 | Hydrocarbons ± 1.24 |
| 23            | 99-83-2 | C₁₃H₁₈    | l-Phellandrene                            | 2.02 | 3.32 | 2.06 | 0    | ± 0.8 | Hydrocarbons ± 0.19 |
| 24            | 142-92-7 | C₆H₆O₂    | Acetic acid, hexyl ester                  | 2.03 | 1.63 | 2.35 | 0    | ± 0.26 | Esters |
| 25            | 99-86-5 | C₁₃H₁₈    | Terpinen                                  | 4.2  | 2.66 | 2.09 | 7.26 | ± 15.3 | Hydrocarbons ± 0.14 |
| 26            | 99-87-6 | C₁₃H₁₈    | Cymene                                    | 0.41 | 0.54 | 0.86 | 4.71 | ± 10.14 | Hydrocarbons ± 0.47 |
| 27            | 5989-54-8 | C₁₃H₁₈    | l-Limonene                                | 1.78 | 1.54 | 1.82 | 4.7  | ± 10.14 | Hydrocarbons ± 0.47 |
| 28            | 138-86-3 | C₁₃H₁₈    | Limonene                                  | 22.56 | 31.8 | 29.39 | 0    | ± 26.4 | Hydrocarbons ± 0.47 |
| 29            | 3779-61-1 | C₁₃H₁₈    | Ocimene                                   | 1.33 | 0.38 | 1.45 | 1.47 | ± 5.76 | Hydrocarbons ± 0.34 |
| 30            | 10054-09-8 | C₁₃H₁₈    | 2-Methyl-6-methylideneoct-2-ene           | 0.36 | 0.33 | 0.01 | 0.9  | ± 0.09 | Hydrocarbons ± 0.08 |
| 31            | 2363-89-5 | C₆H₆O     | 2-Octenal                                 | 0.07 | 0    | 0  | 0.07 | ± 0.09 | Aldehydes ± 0.08 |

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| Retention time | CAS# | Molecular formula | Name | VHPD ± 1SD | HAD ± 1SD | SD ± 1SD | FD ± 1SD | FA ± 1SD | Classi  
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| No. | Retention time | CAS#     | Molecular formula | Name                                      | VHPD  | HAD   | SD   | FD   | FA  | Classification  |
|-----|----------------|----------|-------------------|-----------------------------------------|-------|-------|------|------|-----|-----------------|
| 63  | 10.606         | 7492-41-3| C₁₁H₁₈O₂          | Bornyl formate                          | 0     | 0.8±  | 1.16 | 0.49 | 0.21| Esters         |
| 64  | 10.727         | 106-26-3 | C₁₀H₁₆O           | 3,7-Dimethyl-2,6-octadienal             | 0     | 0     | 0.77 | 0.40 | 0.51| Aldehydes      |
| 65  | 10.73          | 556-67-2 | C₄H₉O₂Si₄         | Octamethyl-cyclosiloxane                | 1.08± | 0.25  | 0.74 | 0.55 | 0   | Others         |
| 66  | 10.793         | 34246-37-6| C₁₀H₁₈O₂          | 3-Isopropylbenzaldehyde                 | 0     | 0     | 2.69 | 0.68 | 4.65| Aldehydes      |
| 67  | 10.848         | 122-03-2 | C₁₀H₁₆O           | Cuminaldehyde                           | 2.61± | 0.9   | 0    | 0.45 | 0   | Aldehydes      |
| 68  | 10.895         | 106-25-2 | C₁₀H₁₈O₂          | 3,7-Dimethyl-2,6-octadien-1-ol          | 0     | 0     | 0    | 0.36 | 0   | Alcohol        |
| 69  | 10.965         | 35907-10-9| C₁₀H₁₆O           | 2-Methylene-5-(1-methylene)-cyclohexanol| 0     | 0     | 0    | 0.92 | 0.76| Alcohol        |
| 70  | 11.036         | 55050-40-3| C₁₀H₁₆O           | 7-Methyl-3-methylene-6-octenal          | 0     | 0     | 0    | 0.8  | 0.44| Aldehydes      |
| 71  | 11.066         | 106-24-1 | C₁₀H₁₈O₂          | trans-Geranol                           | 0.46± | 0     | 0    | 0   | 0   | Alcohol        |
| 72  | 11.223         | 141-27-5 | C₁₀H₁₆O           | E-Citral                                | 0.43  | 0     | 0.74 | 0.42 | 0.09| Alcohol        |
| 73  | 11.342         | 2111-75-3| C₁₀H₁₈O₂          | 4-(1-Methylene)-1-cyclohexene-1-carboxylic| 0     | 0     | 3.97 | 0.42 | 0.09| Alcohol        |
| 74  | 11.48          | 92618-89-8| C₁₂H₂₀₂O₂         | Acetic acid, 1,7,7-trimethyl-bicyclo[2.2.1]hept-2-yl ester | 0     | 0.8   | 0    | 0   | 0   | Alcohol        |
| 75  | 11.619         | 76-49-3 | C₁₂H₂₀₂O₂         | Bornyl acetate                          | 170.73| 180.92| 197.38| 202.12| 11.32| Esters         |

Continued...
| Retention No. | CAS#  | Molecular formula | Name                                      | VHPD      | HAD      | SD      | FD      | FA      | Classification  |
|--------------|-------|-------------------|------------------------------------------|-----------|----------|--------|--------|--------|-----------------|
| 94           | 13.81 | C15H24            | Aromadendrene                             | 0.76 ± 0.98 | 1 ±     | 0.86   |        |        | Hydrocarbons    |
| 95           | 14.06 | C15H24            | z-Humulene                                | 0.34 ± 1.24 | 1.22    | ± 0.74 | 6.64 ± 8.83 | 5.75 | ± 0.67         |
| 96           | 25246-1| C15H24 | [1ar-(1ax,4ax,7a,7aββ,7βz)]-1H-Cycloprop[ez]azulene, decahydro-1,1,7-trimethyl-1H-methylene | 2.32 ± 2.66 | 1.55 ± | 16.41 | 5.63 | | Hydrocarbons |
| 97           | 14.23 | C15H24            | γ-Gurjene                                 | 1.09 ± 1.47 | 0.97    | ± 13.5 | 3.58 ± 2.81 | 2.07 | ± 0.11         |
| 98           | 14.26 | C15H24            | α-Copaene                                 | 1.72 ± 1.12 | 1.23    | 0.21 ± 0.75 | Hydrocarbons | 0.36 | ± 0.65         |
| 99           | 14.405| C15H24           | Aristolochene                             | 0.55 ± 0.66 | 1.21    | 0.34 ± 0 | Hydrocarbons | 0.19 |               |
| 100          | 14.448| C15H24           | Decahydro-4a-methyl-1-methylene-7-(1-methylethynyl), [4ar-(4ax,7a,8aβ)]-naphthalene | 0.0 ± 0.42 | 0.35 ± 0.21 | 17.83 | 0.54 ± 0.17 | Hydrocarbons |
| 101          | 14.456| C15H24           | γ-Maileine                                | 0.61 ± 0.71 | 0.72 ± 0 | Hydrocarbons | 0.63 |               |
| 102          | 14.458| C15H24           | Maileine                                  | 1.53 ± 0.74 | 0.0      | Hydrocarbons | 0.0   |               |
| 103          | 14.545| C15H24           | Ledene                                    | 1.3 ± 1.32  | 1.33 ± 0 | Hydrocarbons | 0.0   |               |
| 104          | 14.553| C15H24           | Viridiflorene                             | 0.0 ± 0.0  | 10.6 ± 0 | Hydrocarbons | 0.18 |               |
| 105          | 14.566| C15H24           | Bicyclogermacrene                         | 6 ± 7.64 ± 0.269 | 7.14 | 1.58 ± 9.2 | Hydrocarbons | 0.73 | 0.77          |
| 106          | 14.692| C15H24           | Cymarin                                   | 0.0 ± 0.0  | 1.69 ± 0 | Others    | 0.56 |               |
| 107          | 14.71 | C15H26O          0.0                  | Sesquicineole                             | 0.54 ± 0.91 | 0.0      | Others   | 0.0   |               |
| 108          | 14.847| C15H24           | δ-Cadinene                                | 0.2 ± 0.62  | 0       | Hydrocarbons | 1.74 | 0.33          |
| 109          | 14.86 | C15H24           | δ-Cadinene                                | 2.4 ± 4.86 ± 2.08 | 2.65 | 0.79 ± 4.01 | Hydrocarbons | 1.37 | ± 0.43         |
| 110          | 15.007| C15H24           | α-Cubebeane                               | 0.0 ± 0.0  | 0.0      | Hydrocarbons | 0.61 | 0.54          |
| 111          | 15.011| C15H26O          0.0                  | Palustrol                                 | 0.0 ± 0.0  | 1.91 ± 0 | Alcohols | 0.0   |               |
| 112          | 15.061| C15H24           | 5β,6β-Epoxy-7x-bromocholestan-3β-ol       | 0.97 ± 0.08 | 0       | Alcohols | 0.0   |               |
| 113          | 15.54 | C15H26O          0.0                  | Ledol                                     | 0.0 ± 0.0  | 1.7 ± 0.0 | Alcohols | 0.0   |               |
| 114          | 15.661| C15H24           | Spathulenol                               | 3.48 ± 2.06 ± 1.81 | 3.26 | ± 4.36 | Alcohols | ± 9.46 | 2.88 | ± 0.92         |
| 115          | 15.741| C15H26O         0.0                   | (1R,7S,E)-7-Isopropyl-4,10-dimethylenecyclodec-5-enol | 0.0 ± 0.0  | 0.0      | Alcohols | 0.0   |               |
| 116          | 15.758| C15H24           | Caryophyllene oxide                       | 1.03 ± 0.0  | 0.57 ± 2.15 | Others   | 0.0   | ± 0.12        |
| 117          | 15.868| C15H26O          0.0                  | Viridiflorol                              | 0.0 ± 0.0  | 1.15 ± 0 | Alcohols | 0.0   |               |
| 118          | 15.943| C15H26O          0.0                  | 1,2,3,6-Tetramethyl-bicyclo[2.2.2]oct-2-ene | 0.0 ± 0.0  | 0.14     | Alcohols | 0.0   |               |
| 119          | 16.089| C15H26O          0.0                  | Humulene epoxide II                       | 0.22 ± 0.38 | 0.29 ± 2.85 | Others   | 0.91 | ± 0.43         |
| 120          | 16.397| C15H26O          0.0                  | 11,11-Dimethyl-, 4,8-bis(methylene)-bicyclo[7.2.0]undecan-3-ol | 0.26 ± 0.36 | 0.33    | 2.34 ± 0.1 | ± 0.61 | 4.17 | ± 0.29         |
| 121          | 16.457| C15H26O          0.0                  | Hexadecamethylene-cyclooctasiloxane       | 0.68 ± 0.81 | 1.48 ± 1.51 | Others   | 0.0   |               |
| 122          | 16.61 | C15H26O          0.0                  | [1ar-(1ax,4ax,7β,7aβ]-1H-Cycloprop[ez]azulene-7-ol, decahydro-1,1,7-trimethyl-4-methylene | 0.0 ± 0.0  | 0.0      | Alcohols | 0.0   |               |
| 123          | 16.642| C15H26O          0.0                  | Caryophyllene oxide                       | 1.18 ± 1.39 | 3.46 ± 2.85 | Others   | 0.32 | ± 0.68         |

**Table 1 (Contd.)**
alcohols (223 ± 23) µg g⁻¹, esters (226 ± 16) µg g⁻¹, aldehydes (65 ± 18) µg g⁻¹, and other compounds. The content of ketones increased in hot air-dried samples to (118 ± 12) µg g⁻¹ and decreased in samples obtained by other methods. The content of ethers and hydrocarbons was reduced by all drying methods.

The significance of the VOC results was assessed that compared with fresh ‘Guiyan1’. The differences in alcohols obtained by the freeze-drying method and the other three methods were significant. The differences in ketones and ethers in ‘Guiyan1’ obtained by hot air drying, sun drying, and freeze-drying were significant, in contrast to hydrocarbons. The differences between the four drying methods were significant for esters and acids and not significant for aldehydes and other compounds. In addition, as shown in Fig. 1(A) and (B), there was no significant correlation between the number of species and the content of VOCs in ‘Guiyan1’ samples obtained by the four drying methods.

### 3.2 Modeling and model evaluation of VOCs in ‘Guiyan1’ obtained by different drying methods

As shown in Fig. 2(A) and (B), ‘Guiyan1’ samples obtained by the four drying methods and fresh ones were within the 95% confidence interval. In the PCA-X model, five groups of samples were relatively dispersed, the hot air-dried, heat pump-dried, and sun-dried ones had some overlap and could not be effectively distinguished, while the clustering of freeze-dried samples was poor. In the OPLS-DA model, all five groups of samples were well clustered; except for the overlapping of some areas of the heat pump- and sun-dried samples, others were easily distinguished. To avoid a possible overfitting of the OPLS-DA model, a cross-validation was performed for all groups of samples, and the minimum XCV err r was chosen to be the model with the best stability.

### Table 1

| No. | Retention time | CAS# | Molecular formula | Name                                      | VHPD | HAD | SD | FD | FA | Classification  |
|-----|----------------|------|-------------------|-------------------------------------------|------|-----|----|----|----|----------------|
| 124 | 16.683         | 515-20-8 | C₁₅H₂₄O       | Costol                                    | 0    | 0   | 0  | 1.2±0 | 0  | Alcohols       |
| 125 | 16.753         | 53820-13-6 | C₁₅H₂₄O₂     | Chrysantenyl 2-methylbutanoate            | 0    | 0   | 0  | 0.55±0 | 0  | Esters         |
| 126 | 16.793         | 552-02-3 | C₁₅H₂₆O       | Epiglobulol                               | 0    | 0   | 0.38±0 | 0.28 | 0  | Alcohols       |
| 127 | 17.03          | 71579-69-6 | C₁₆H₂₆O₂Si₆   | 3-Isopropoxy-1,1,1,7,7,7-hexamethyl-3,5,5-tris(trimethylsiloxy)tetrasiloxane | 0    | 0   | 0.62±0 | 1.17±0 | 0  | Others         |
| 128 | 17.133         | 145344-72-5 | C₁₈H₄₈O₇Si₉   | 2-(2',4',6',8',8'-heptamethyltetrasiloxan-2-yloxy)-2,4,6,6,8,8,10,10- nonamethyleclopentsiloxane | 0.32±0 | 0.31±0 | 0.2±0 | 0.34±0 | 0.34±0 | Others |
| 129 | 17.14          | 19095-24-0 | C₁₅H₅₀O₂Si₆  | 1,15-Dihydrogenhexadamethyl          | 0.11±0 | 0.09±0 | 0.13±0 | 0.27±0 | 0  | Ethers         |
| 130 | 17.76          | 3135-71-3 | C₁₄H₂₄O       | 2-Methyl-, 4-(2,6,6-trimethyl-1-cyclohexen-1-yl)-but-2-enal | 0    | 0   | 0.09±0 | 0.08 | 0  | Aldehydes      |
| 131 | 18.142         | 638-36-8 | C₂₀H₄₂         | 2,6,10,14-Tetramethyl-hexadecane       | 0    | 0   | 0   | 0.05±0 | 0.18±0 | Hydrocarbons  |
| 132 | 18.309         | 540-97-6 | C₁₂H₃₆O₇Si₉   | Dodecamethyl-cyclohexasiloxane        | 0.99±0 | 0.98±0 | 2.35±0 | 3.27±0 | 0.13±0 | Others         |
| 133 | 18.438         | 3243-36-5 | C₁₅H₂₆O₆Si₈  | Ambrial                                 | 0.91±0 | 0.66±0 | 0.42±0 | 1.71±0 | 0.12±0 | Ketones        |

Fig. 1  Analysis of species (A) and content of VOCs (B) of Amomum villosum Lour. ‘Guiyan1’ samples obtained via four drying methods.
DA model, which could deteriorate its ability to assess the new sample dataset effectively, the model reliability was validated via the permutation test and cross-validation analysis (CV-ANOVA). As shown in Fig. 2(C), the OPLS-DA model had no overfitting, being stable and reliable. The intercept of $R^2$ and $Q^2$ curves with vertical coordinates was less than 1, and the intercept of $Q^2$ in vertical coordinates was less than 0. Besides, the significant probability $P$ value was below 0.05 in the CV-ANOVA analysis. It can be seen in Fig. 2(D) that the OPLS-DA model featured values $R^2X = 0.910$ and $Q^2 = 0.837$ and, thus, presented more data variation than the PCA-X model with $R^2X = 0.808$ and $Q^2 = 0.550$. Thus, the OPLS-DA model could better differentiate the ‘Guiyan1’ samples obtained by the four drying methods under study than the PCA-X model.

### 3.3 Excavation of potential differences in VOCs in ‘Guiyan1’ obtained by different drying methods

S-plots were used to identify chemical composition differences between two samples and helped to identify metabolites of statistical and potential biochemical significance. The points at the ends of the S-plot indicated variables with the highest contributions to the model, while those with smaller contributions were clustered near the origin. The OPLS-DA model predicted that the ‘Guiyan1’ samples obtained by the four drying methods were better separated from the fresh ones, the freeze-dried samples were better separated from the samples obtained by the other three drying methods, and hot air-dried samples were better separated from sun-dried ones. Given this, this study focused on analyzing the differences in VOCs under the above conditions, yielding the S-plot of VOCs presented in Fig. 3.

The red dots in Fig. 3 indicate metabolites with VIP > 1. As can be seen from Fig. 3, the four components that differed most significantly in fresh and dried samples were bornyl acetate (75), 6,6-dimethyl-2-methylene-bicyclo[3.1.1]heptane (18), 2-borneanone (45), and 1-borneol (51). In the HPD and FD samples, the components with the most significant differences in VOCs (VIP > 2) were linalool (38), bornyl acetate (75), limonene (28), and Terpinen-4-ol (53). In the HAD and FD samples, these were linalool (38), limonene (28), bornyl acetate (75), and camphene (15). In the SD and FD samples, these were linalool (38), bornyl acetate (75), limonene (28), terpinen-4-ol (53), and 6,6-dimethyl-2-methylene-bicyclo[3.1.1]heptane (18). Finally, in the HAD and SD samples, these were bornyl acetate (75), 1-borneol (51), linalool (38), myrtenal (58), and methyl 6,6-dimethylbicyclo[3.1.1]hept-2-ene-2-carboxylate (76).

### 3.4 Differential analysis of the content of VOCs in ‘Guiyan1’ obtained by different drying methods

The VIP analysis results on VOCs in ‘Guiyan1’ obtained by four drying methods are depicted in Fig. 4(A), featuring 33 metabolites with VIP > 1. These included acetate (80), acetic acid, hexyl ester (24), isoborneol (49), 2, 2,4-trimethyl-3-cyclohexene-1-methanol (57), endo-borneol (52), 2,6-octadien-1-ol, 3,7-dimethyl-1, acetate (83), 2-borneanone (45), methyl 6,6-dimethylbicyclo[3.1.1]hept-2-ene-2-carboxylate (76), and sesquicineneole (107).
The variability of VOC contents in ‘Guiyan1’ obtained by four drying methods was assessed using heatmaps with the contents of 33 metabolites with VIP > 1, as shown in Fig. 4(B).

In can be inferred from Fig. 4(B) that the content of 2-borinanone (45) in ‘Guiyan1’ obtained by the four drying methods increased the most, followed by 4-methylene-1-(1-methylethyl)bicyclo[3.1.0]hexane (17). Taking 2-ornanone(45) as an example, hot air drying method yielded the highest content of $(109.78 \pm 16.33) \mu g \ g^{-1}$, followed by heat pump drying, freeze-drying, and sun drying methods with $(80.11 \pm 19.46) \mu g \ g^{-1}$, $(74.08 \pm 0.00) \mu g \ g^{-1}$, and $(40.73 \pm 3.46) \mu g \ g^{-1}$, respectively.

After drying, the contents of terpinene-4-ol (53), 1-methyl-4-(1-methylethylidene)-cyclohexene (37), and bicyclogermacrene (105) in fresh ‘Guiyan1’ decreased the most, namely by $2.99 \pm 0.82 \mu g \ g^{-1}$, $12.78 \pm 0.30 \mu g \ g^{-1}$, and $7.62 \pm 0.04 \mu g \ g^{-1}$ for the freeze-dried samples, respectively. The unique characteristic fraction endo-borneol (52) was present in sun-dried and hot air-dried samples with values of $7.69 \pm 1.39$ and $11.78 \pm 1.45 \mu g \ g^{-1}$, respectively. Terpinyl acetate (80) and acetic acid hexyl ester (24) were present only in hot air-dried samples; 5,6b-epoxy-7α-bromocholestan-3β-ol (112) was present only with the heat.
pump-dried samples, while hexamethyl-cyclotrisiloxane (35) was retained after drying.

4. Discussion

In total, 133 VOCs were identified in ‘Guiyan1’ samples obtained by four drying methods under study. These were mainly hydrocarbons, esters, alcohols, and ketones. Noteworthy is that hydrocarbons had the highest content and species content, in contrast to the earlier results of Ai et al.28 This can be attributed to different combinations of compounds, esters, etc. Compared with fresh ‘Guiyan1’, the contents of alcohols, esters, aldehydes, and other kinds of compounds increased in freeze-dried samples and decreased in samples obtained by the other drying methods. The content of ketones in grew in hot air-dried samples and dropped in those dried by other methods. The content of ethers and hydrocarbons in all dried samples decreased, in contrast with findings of Ai et al.28 Besides, this study revealed that the content of alcohols increased in freeze-dried samples and dropped in samples dried by the other three methods, instead of increasing as in ref. 28. These discrepancies can be attributed to the testing and processing differences.

Drying methods significantly influenced the species and content of VOCs in samples. The main VOCs of fresh ‘Guiyan1’ were 6,6-dimethyl-2-methylene-bicyclo[3.1.1]heptane, (1S)-linalool, α-pinene, terpinene-4-ol, limonene, 1-methyl-4-(1-methylethyl)-1,4-cyclohexadiene, acetic acid, 1,7,7-trimethylbicyclo[2.2.1]hept-2-yl ester, β-myrcene, α-terpinene, etc. The inconsistency of this finding with the results of Chen et al.19 can be attributed to different sample pretreatment processes. After grinding during the assay process, the tissue structure of ‘Guiyan1’ was destroyed more substantially. The essential oil of ‘Guiyan1’ was released, resulting in the VOCs in their amomums being mainly camphor, bornyl acetatecamphor, bornyl acetate, caryophyllene, β-bisabolene, (E)-nerolidol, and cubenol being the predominant compounds. Besides, the extraction method of the aroma during the assay process affected the composition variability.44 After drying, the VOCs in ‘Guiyan1’ were dominated by bornyl acetate (75), 2-boranone (45), limonene (28), and linalool (38). The drying process could promote the release of volatile oils from amomum plant tissues to the kernels’ surface, presenting the characteristic components of lobsteryl acetate and camphor in amomum essential oil.6,35

In this study, a mass spectrometry detection technology combined with OPLS-DA was used to study the effect of drying methods on the VOCs of ‘Guiyan1’, and the drying methods were differentiated according to the specific components. Thus, the origin of amomums according to VOC variability can be identified as in ref. 30 and 36.

5. Conclusions

In this study, 133 volatile organic compounds, mainly hydrocarbons, esters, alcohols, and ketones, were isolated and identified in ‘Guiyan1’ samples via HS-GC-MS. Significant differences in the types and contents of VOCs in ‘Guiyan1’ samples dried by four different methods were observed. Among them, freeze-drying and sun-drying yielded the most considerable content of VOCs in ‘Guiyan1’; only freeze-drying helped to promote the content of alcohols, esters, and aldehydes in ‘Guiyan1’; only hot air drying promoted the content of ketones in ‘Guiyan1’, and all four methods decreased the content of ethers and hydrocarbons. The VIP distribution, S-plot, and heatmap diagrams were used to identify 33 significantly different metabolites in ‘Guiyan1’ obtained by different drying methods, among which 2-boranone content increased the most. The results obtained are considered instrumental in optimizing the processing of Amomum villosum Lour.
amomums. Exploring the effects of various drying methods on other varieties of amomum is envisaged in the follow-up study.

Conflicts of interest

The authors declare no conflicts of interest. The sponsors had no role in the study’s design, data collection, analyses, or interpretation, in the writing of the manuscript, or in the decision to publish the results.

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References

1. X. Fangfang, C. Weiying, C. Wanna, M. Ting, W. Yunshan and L. Bo, Research progress of chemical constituents and quality control method of Villous Amomum fruit, World. Chin. Med., 2020, 15, 3881–3886.
2. H. Junjun, Y. Yanli, X. Yinquan, L. Wenxiu, Z. Hualin, T. Xinghao, Z. Qingqian, H. Summing, D. Huabo and L. Ping, Analysis on morphological characteristics and Quality of Amomum villosum Lour. ‘Zhansha 11’, Chin. J. Trop. Crop, 2022, 4(3), 294–302.
3. H. Lv, Research on comprehensive development and utilization of Yangchun Amomum Villous. Resources, Yunnan University of Traditional Chinese Medicine, 2020.
4. X. Hongning, H. Liufang, L. Xile, C. Qingxi and J. Shengguo, Study on quality variation of Amomum villosum Lour from different producing areas, J. Guangdong Pharm. Univ., 2016, 32, 176–180.
5. X. Kaining, X. Xuefeng, X. Wenxue and L. Xueying, New advances in the study of the chemical composition and related pharmacological effects of Amomum villosum Lour, Chinese Medicine Modern Distance Education of China, 2014, 12, 100–101.
6. H. Guozhong, W. Qin, M. Lukai and L. Weifang, Trends in research on the primary active compounds in Amomum villosum, Food Res. Dev., 2021, 42, 219–224.
7. C. JungHyo, L. JongSuk, K. HyeongGeug, L. H. Won, F. Zhigang, K. HyeokHee, K. D. Woon, L. ChangMin and J. JinWoo, Ethyl Acetate Fraction of Amomum villosum var. xanthoiodes Attenuates Hepatic Endoplasmic Reticulum Stress-Induced Non-Alcoholic Steatohepatitis via Improvement of Antioxidant Capacities, Antioxidants, 2021, 10, 998.
8. L. Shanhong, Z. Ting, G. Wen, Y. Xingxin, L. Jianmei, Z. Ronghua and Y. Jie, Volatile Oil of Amomum villosum Inhibits Nonalcoholic Fatty Liver Disease via the Gut-Liver Axis, BioMed Res. Int., 2018, 3589874.
9. S. Ekaruth, P. Mullika, M. Sakulrat and S. Klaokwan, Anti-inflammatory Effect of Ettlingera pavieana (Pierre ex Gagnep.) R.M.Sm. Rhizomal Extract and Its Phenolic Compounds in Lipopolysaccharide-Stimulated Macrophages, Pharmacogn. Mag., 2017, 13, S230–S235.
10. Z. Yang, Q. ChunGuo, Y. Depo, T. Cailin, X. Xinjun, L. EHu, Z. Jingtang, Z. Longping and Z. Zhimin, Purification, Structural Characterization and Immunomodulatory Effects of Polysaccharides from Amomum villosum Lour on RAW 264.7 Macrophages, Molecules, 2021, 26, 2672.
11. W. Zhi, R. Xinmei, D. Taotao, F. Min, Y. Jiancheng, L. Jiao and C. Jun, Structure Characterization and Antioxidant Activity of Polysaccharides from Amomum villosum Extracted with Alkaline Solution, Sci. Technol. Food Ind., 2021, 42, 87–93.
12. W. Zhi, Research and Product Development of Active Ingredients of Hainan Amomum villosum, Nanchang University, 2021.
13. F. Yuijiang, J. Xilin, Z. Shuai, W. Qixiang, Q. Min, Q. Huijuan and W. Na, Optimization of extraction process of Amomum longiligulare T. L. Wu polysaccharides by response surface methodology, J. Hainan Med. Univ., 2021, 27, 467–471.
14. G. Linlin, W. Qing, Z. Jingwen, H. RuqiZang and Z. Xiaowen, Study on purification and antibacterial and antitumor activity of flavonoids from Amomum villosum Lour and Alpinia Oxyphylla Miq, J. Food Saf. Food Qual., 2019, 10, 4659–4666.
15. D. Min, W. Shengli, H. Xiaofeng, Z. Xuemei and G. Changan, Terpene constituents in Amomum villosum Lour and their hypoglycemic activity, Chin. J. Inf. Tradit. Chin. Med., 2022, 1–6.
16. L. QinLi, X. WeiRong, G. MengYao, Z. RuiYan, Z. LiQin and Z. Qingyi, Optimization of the extraction technology of total flavonoids from Changtai Amomum villosum by response surface methodology, J. Jiangxi Univ. Tradit. Chin. Med., 2021, 33, 68–72.
17. Y. Jianjun, Z. Shulei, Z. Bo, R. Faisah, L. Zuhani, L. Xiaohua, Z. Yongyu, N. Qu and Q. Mingfeng, Efficacy and mechanism of active fractions in fruit of Amomum villosum Lour for Gastric Cancer, J. Cancer, 2021, 12.
18. S.-K. Lee, C. H. Eum and C.-G. Son, Analysis of volatile compounds and metals in essential oil and solvent extracts of Amomi Fructus, Anal. Sci. Technol., 2015, 28, 436–445.
19. L. X. Chen, Y. F. Lai, W. X. Zhang, J. Cai, H. Hu, Y. Wang, J. Zhao and S. P. Li, Comparison of volatile compounds in different parts of fresh Amomum villosum Lour. from different geographical areas using cryogenic grinding combined HS-SPME-GC-MS, Chin. Med., 2020, 15, 97.
20. C. Tang, J. Chen, Y. Zhou, P. Ding, G. He, L. Zhang, Z. Zhao and D. Yang, Exploring antimicrobial mechanism of essential oil of Amomum villosum Lour through metabolomics based on gas chromatography-mass spectrometry in methicillin-resistant Staphylococcus aureus, Microbiol. Res., 2020, 242, 126608.
21. T. Huong Le, T. Viet Nguyen, N. Sam Ly, N. Giang Cao, H. Hung Nguyen, N. Nai Do and A. Ogunwande Isiaka,
Antimicrobial activity of the Essential Oils from the Leaves and Stems of Amomum rubidium Lamxay & N. S. Ly, *Bol. Latinoam. Caribe Plant. Med. Aromat.*, 2021, 20, 81–89.
22 H. Fengting, W. Mianjie and Z. Danyan, Chemical constituents from Amomum villosum Leaf oil and its activity of promoting wound healing, *Journal of Guangdong Pharmaceutical University*, 2017, 33, 466–470.
23 W. Mianjie, Analysis of the chemical composition and preliminary pharmacological study of Amomum villosum leaf oil, *J. Guangzhou Univ. Tradit. Chin. Med.*, 2017, 1.
24 H. Lin, H. Yanzhang, X. Guosheng, W. Kaituo, H. Qianfang and Z. Yuanyuan, Analysis of the composition of the essential oil of saxifrage and its effect on the freshness of strawberries, *Food Ferment. Ind.*, 2012, 38, 199–203.
25 C. Dekui, G. Xiang, Z. Minyang, C. Xiaoyang, Z. Wei and Z. Qing, The Effects of Amomum Villosum Lour. Essential Oil on the Fermentation Quality of Alfalfa, *Acta Agrestia Sin.*, 2021, 29, 855–860.
26 H. Lv, F. Xingqing, X. Jia, W. Ru, Z. Ronghua and B. Niman, GC-MS analysis of volatile components in different parts of Amomum villosum L., *J. Chengdu Univ. Tradit. Chin. Med.*, 2020, 43, 42–46.
27 X. Weishan, H. Xikun, X. Yanping, W. Wanwen, L. Zhixiao and C. Zhixiong, Determination and analysis of trace elements in Amomum villosum from different habitats, grades and its confusions, *Chin. Manipulation Rehabil. Med.*, 2022, 13(9), 77–80.
28 Z. P. Ai, S. Mowafi and Y. H. Liu, Comparative analyses of five drying techniques on drying attributes, physicochemical aspects, and flavor components of Amomum villosum fruits, *LWT*, 2022, 154, 112879.
29 H. Jianing, G. Chen, D. Xingming, L. Guang and L. Guoqing, Analysis of the volatile fractions of Amomum villosum from different origins based on headspace solid-phase microextraction coupled with gas-phase mass spectrometry, *J. Guangzhou Univ. Chin. Med.*, 2020, 37, 1366–1371.
30 C. Xiaotian, X. Xue, Q. Chenyu, S. Juyi and X. Zhangmin, Analysis of volatile oil components in Amomum villosum from different producing areas by full two-dimensional gas chromatography-quadrupole time-of-flight mass spectrometry, *Chin. Condiment*, 2021, 46, 142–148.
31 H. Zhou, D. Luo, H. Gholamhosseini, Z. Li, B. Han, J. He and S. Wang, Aroma characteristic analysis of Amomi Fructus from different habitats using machine olfactory and gas chromatography-mass spectrometry, *Pharmacogn. Mag.*, 2019, 15, 392–401.
32 Y. J. Liu, Y. Y. Qian, B. Shu, Y. Y. Liu, X. H. Tu, H. J. Ouyang, Y. Li, G. Tan, Z. W. Yu, F. Chen and L. J. Lin, Effects of four drying methods on Ganoderma lucidum volatile organic compounds analyzed via headspace solid-phase microextraction and comprehensive two-dimensional chromatography-time-of-flight mass spectrometry, *Microchem. J.*, 2021, 166, 1–13.
33 C. P. Jianghao, Differentiation of Panax Quinquefolius grown in the USA and China using LC/MS-based chromatographic fingerprinting and chemometric approaches, *Anal. Bioanal. Chem.*, 2011, 399, 1877–1889.
34 S. Juyi, C. Xiaotian, L. Shuqin, B. Rui and X. Zhangmin, Comparison of extraction effect of headspace-solid phase microextraction and simultaneous distillation on volatile components in Amomum Villosum based on comprehensive two-dimensional gas chromatography-quadrupole time-of-flight mass spectrometry, *Phys. Test. Chem. Anal., Part B*, 2022, 58, 181–192.
35 L. Jinlin, W. Mei, T. Jia, Q. Fei, Z. Junbo, Z. Xiaohei, C. Chundao, Z. Qin and S. Yajun, Effect of environmental factors on main components of Amomi Fructus by headspace-gas chromatography-mass, *Mod. Chin. Med.*, 2018, 20, 1504–1508.
36 T. C. Lin, Y. D. Po, C. J. Li, Z. L. Xia, D. Ping, X. X. Jun, L. W. Jian, X. J. Chun and Z. Z. Min, Geographical origin discrimination of Amomi Fructus using an ultra-performance liquid chromatography-quadrupole time-of-flight mass spectrometry-based metabolomics approach combined with antioxidant activity analysis, *Pharmacogn. Mag.*, 2021, 17, 492–498.