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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. xanthioiles T. L. Wu et Senjen (called *Amomum xanthioides* 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 ingredients,[6] 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 (1*R*,2*S*,4*R*,7*S*)-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 component,[17] mainly composed of camphor, lobster acetate, lobster, D-citrin, and camphor,[18,19] 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 strawberries[24] 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,[26] origin[27], and drying methods.[28] The

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research results[26] showed that the composition and content of volatile organic components (VOCs) in seeds, peels, and rhizomes of *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 *Amomum villosum*. The comparative analysis of five drying techniques of *Amomum villosum* fruits performed by Ai *et al.*[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.[28]

The main means of analyzing the volatile aroma of amomums are headspace solid-phase microextraction gas-phase mass spectrometry (HS-SPME-GC-MS),[29] comprehensive two-dimensional gas chromatography-quadrupole time-of-flight mass spectrometry (GC × GC-QTOF-MS),[30] and gas chromatography-mass spectrometry (GC-MS)-electronic nose (E-nose).[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

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.*,[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.*,[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 μ L of 100 mg L^{-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 μ m), 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^{-1}, hold for 5 min, carrier gas helium, injection mode with splitting, total flow rate 34.0 mL min^{-1}, column flow rate 1.00 mL min^{-1}, linear velocity 36.3 cm s^{-1}, septum purge flow 3.0 mL min^{-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^{-1}, and the lowest in sun-dried ones, namely (570 ± 143) μg g^{-1}. Compared with fresh 'Guiyan1', freeze-dried samples had higher contents of

Table 1 The composition and relative contents of VOCs in *Amomum villosum* Lour. 'Guiyan1' samples obtained *via* four drying methods

No.	Retention time	CAS#	Molecular formula	Name	VHPD	HAD	SD	FD	FA	Classification
1	1.464	509-14-8	CN_4O_8	Tetranitro-methane	0.51 ± 0.07	0.39 ± 0.09	0.62 ± 0.2	1.01 ± 0.57	0.41 ± 0.02	Others
2	1.505	75-07-0	C_2H_4O	Acetaldehyde	0	0	0	0	0.04 ± 0.03	Aldehydes
3	1.571	64-17-5	C_2H_6O	Ethanol	0	0	0	0	0.06 ± 0.05	Alcohols
4	2.305	590-86-3	$C_5H_{10}O$	3-Methyl-butanal	0	0.07 ± 0.09	0	0	0	Aldehydes
5	3.036	1066-42-8	$C_2H_8O_2Si$	Dimethyl-silanediol	4.68 ± 0.75	3.72 ± 0.67	4.31 ± 1.4	4.79 ± 1.49	4.54 ± 1.03	Esters
6	3.321	108-88-3	C_7H_8	Toluene	0	0.05 ± 0.05	0	0	0	Hydrocarbons
7	3.685	66-25-1	$C_6H_{12}O$	Hexanal	0.1 ± 0.09	0.09 ± 0.03	0.12 ± 0.1	0	0.1 ± 0.01	Aldehydes
8	4.735	123-92-2	$C_7H_{14}O_2$	3-Methyl-1-butanol, acetate	0.15 ± 0.14	0.07 ± 0.09	0	0	0	Esters
9	5.137	111-71-7	$C_7H_{14}O$	Heptanal	0.07 ± 0.06	0.04 ± 0.05	0	0	0.05 ± 0.01	Aldehydes
10	5.246	514-14-7	$C_{10}H_{16}$	2,7,7-Trimethylbicyclo[2.2.1]hept-2-ene	0	0	0	0	0.05 ± 0.04	Hydrocarbons
11	5.529	508-32-7	$C_{10}H_{16}$	Tricyclene	0.28 ± 0.32	1.22 ± 1.06	0.61 ± 0.78	0.1 ± 0.17	0.06 ± 0.06	Hydrocarbons
12	5.604	2867-5-2	$C_{10}H_{16}$	3-Thujene	2.73 ± 0.53	1.14 ± 1.06	1.2 ± 0.61	4.05 ± 2.72	7.94 ± 6.88	Hydrocarbons
13	5.725	80-56-8	$C_{10}H_{16}$	α-Pinene	12.82 ± 2.82	12.75 ± 4.32	11.75 ± 5.46	13.86 ± 7.39	38.5 ± 0.88	Hydrocarbons
14	5.885	36262-09-6	$C_{10}H_{14}$	4-Methylene-1-(1-methylethyl)-bicyclo[3.1.0]hex-2-ene	1.92 ± 0.6	1.3 ± 0.79	1.94 ± 1.1	2.36 ± 1.36	0.71 ± 0.02	Hydrocarbons
15	5.986	79-92-5	$C_{10}H_{16}$	Camphene	14.01 ± 5.54	22.36 ± 12.02	18.47 ± 13.13	4.88 ± 0.63	3.93 ± 0.11	Hydrocarbons
16	6.177	100-52-7	C_7H_6O	Benzaldehyde	0	0.41 ± 0.5	0.44 ± 0.51	0	0.22 ± 0.19	Aldehydes
17	6.384	3387-41-5	$C_{10}H_{16}$	4-Methylene-1-(1-methylethyl)-bicyclo[3.1.0]hexane	11.35 ± 2.06	5.91 ± 1.34	3.64 ± 4.63	16.1 ± 10.47	0	Hydrocarbons
18	6.471	18172-67-3	$C_{10}H_{16}$	6,6-Dimethyl-2-methylene-bicyclo[3.1.1]heptane	24.84 ± 3.49	17.69 ± 2.6	15.55 ± 13.73	32.92 ± 17.51	117.7 ± 0.46	Hydrocarbons
19	6.616	123-35-3	$C_{10}H_{16}$	β-Myrcene	14.64 ± 4.67	18.56 ± 8.99	17.5 ± 9.85	7.4 ± 2.06	16.6 ± 0.2	Hydrocarbons
20	6.736	123-96-6	$C_8H_{18}O$	2-Octanol	10	10	10	10	10	Alcohols
21	6.807	124-13-0	$C_8H_{16}O$	Octanal	0	0	1.07 ± 0.94	2.01 ± 1	0	Aldehydes
22	6.883	99-83-2	$C_{10}H_{16}$	α-Phellandrene	0	0	0	2.46 ± 1.24	0	Hydrocarbons
23	6.89	99-83-2	$C_{10}H_{16}$	L-Phellandrene	2.02 ± 2.03	3.32 ± 1.63	2.06 ± 2.35	0	4.8 ± 0.19	Hydrocarbons
24	6.972	142-92-7	$C_8H_{16}O_2$	Acetic acid, hexyl ester	0	0.41 ± 0.26	0	0	0	Esters
25	7.093	99-86-5	$C_{10}H_{16}$	Terpinene	4.2 ± 0.41	2.66 ± 0.54	2.09 ± 0.86	7.26 ± 4.71	15.53 ± 0.14	Hydrocarbons
26	7.226	99-87-6	$C_{10}H_{14}$	Cymene	5.03 ± 1.78	4.79 ± 1.54	4.35 ± 1.82	7.96 ± 4.7	10.19 ± 0.47	Hydrocarbons
27	7.293	5989-54-8	$C_{10}H_{16}$	L-Limonene	0	0	0	13.16 ± 4.86	0	Hydrocarbons
28	7.31	138-86-3	$C_{10}H_{16}$	Limonene	22.56 ± 6.27	31.8 ± 14.76	29.39 ± 14.11	0	26.4 ± 0.47	Hydrocarbons
29	7.393	3779-61-1	$C_{10}H_{16}$	Ocimene	1.33 ± 0.36	0.38 ± 0.33	1.45 ± 0.01	1.47 ± 0.9	5.76 ± 0.34	Hydrocarbons
30	7.671	10054-09-8	$C_{10}H_{18}$	2-Methyl-6-methylideneoct-2-ene	0.07 ± 0.06	0	0	0	0.09 ± 0.08	Hydrocarbons
31	7.744	2363-89-5	$C_8H_{14}O$	2-Octenal	0	0	0.38 ± 0.45	0	0	Aldehydes

Table 1 (*Contd.*)

No.	Retention time	CAS#	Molecular formula	Name	VHPD	HAD	SD	FD	FA	Classification
32	7.796	99-85-4	$C_{10}H_{16}$	1-Methyl-4-(1-methylethyl)-1,4-cyclohexadiene	4.66 ± 4.05	3.37 ± 2.96	3.95 ± 1.39	12.06 ± 7.77	23.17 ± 0.21	Hydrocarbons
33	7.943	15537-55-0	$C_{10}H_{18}O$	Sabinene hydrate	0	0.14 ± 0.25	0.41 ± 0.42	3.02 ± 2.84	2.32 ± 0.15	Alcohols
34	8.03	5989-33-3	$C_{10}H_{18}O_2$	Linalool oxide *cis*	1.16 ± 1.17	0	1.7 ± 2.45	4.78 ± 3.33	0	Alcohols
35	8.07	541-05-9	$C_6H_{18}O_3Si_3$	Hexamethyl-cyclotrisiloxane	1.76 ± 0.35	1.5 ± 0.91	0.64 ± 0.74	1.29 ± 0.84	2.37 ± 0.05	Others
36	8.305	554-61-0	$C_{10}H_{16}$	2-Carene	0	0	4.25 ± 5.16	10.3 ± 6.01	0	Hydrocarbons
37	8.309	586-62-9	$C_{10}H_{16}$	1-Methyl-4-(1-methylethylidene)-cyclohexene	0	2.96 ± 2.82	0	0	12.78 ± 0.3	Hydrocarbons
38	8.507	78-70-6	$C_{10}H_{18}O$	Linalool	61.48 ± 13.73	41.6 ± 7.82	54.6 ± 7.86	107.54 ± 0	93.87 ± 7.23	Alcohols
39	8.626	56114-69-3	$C_{13}H_{22}O_3Si_2$	2,5-Bis[(trimethylsilyl)oxy]-benzaldehyde	0.62 ± 0.29	0.79 ± 0.03	0.59 ± 0.42	0	0	Others
40	8.712	21195-59-5	$C_{10}H_{14}$	*p*-Mentha-1,5,8-triene	0	0	0	0	1.3 ± 0.71	Hydrocarbons
41	8.806	546-80-5	$C_{10}H_{16}O$	α-Thujone	0.58 ± 0.72	0.54 ± 0.59	0.65 ± 0.44	1.22 ± 0.72	0	Ketones
42	8.875	29803-82-5	$C_{10}H_{18}O$	1-Methyl-4-(1-methylethyl)-2-cyclohexen-1-ol	0	0	0	0.41 ± 0	0	Alcohols
43	8.957	91819-58-8	$C_{10}H_{16}O$	Campholenic aldehyde	12.28 ± 3.67	7.41 ± 4.52	12.6 ± 1.32	21.13 ± 0	7.52 ± 0.69	Aldehydes
44	9.222	547-61-5	$C_{10}H_{16}O$	[1*S*-(1α,3α,5α)]-Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-methylene	0	0	0	20.21 ± 0	5.56 ± 0.55	Alcohols
45	9.369	464-49-3	$C_{10}H_{16}O$	2-Bornanone	80.11 ± 19.46	109.78 ± 16.33	40.73 ± 3.46	74.08 ± 0	0	Ketones
46	9.431	24545-81-1	$C_{10}H_{14}O$	4-Methyl-1-(1-methylethyl)-bicyclo[3.1.0]hex-3-en-2-one	0	0	0	2.11 ± 1.33	0	Ketones
47	9.448	564-94-3	$C_{10}H_{14}O$	(−) Myrtenal	0	0	0	4.24 ± 3.75	1.04 ± 0.93	Aldehydes
48	9.485	4916-87-4	$C_9H_{18}O$	1-Methyl-4-(2-hydroxyethyl)-cyclohexane	0	0	0	5.81 ± 0	0	Others
49	9.502	124-76-5	$C_{10}H_{18}O$	Isoborneol	3.19 ± 0.37	2.92 ± 1.66	0	0	0	Alcohols
50	9.592	30460-92-5	$C_{10}H_{14}O$	Pinocarvone	11.18 ± 4.12	7.44 ± 4.74	11.61 ± 7.76	18.77 ± 10.22	7.69 ± 0.21	Ketones
51	9.646	464-45-9	$C_{10}H_{18}O$	1-Borneol	20.49 ± 6.2	11.84 ± 13.61	26.14 ± 15.31	24.11 ± 12.23	0	Alcohols
52	9.711	507-70-0	$C_{10}H_{18}O$	*Endo*-borneol	0	11.78 ± 1.45	7.69 ± 1.39	0	0	Alcohols
53	9.806	562-74-3	$C_{10}H_{18}O$	Terpinen-4-ol	16.5 ± 2.74	13.92 ± 3.69	6.55 ± 2.74	29.95 ± 0	32.94 ± 0.82	Alcohols
54	9.912	6931-54-0	$C_{10}H_{16}O$	6,6-Dimethyl-spiro[bicyclo[3.1.1]heptane-2,2′-oxirane]	0	0	2.41 ± 1.57	2.19 ± 3.78	0	Others
55	9.917	57129-54-1	$C_{10}H_{14}O$	α-Thujenal	0	0	0	0	1.51 ± 1.32	Aldehydes
56	9.919	4764-14-1	$C_{10}H_{16}O$	6,6-Dimethyl-bicyclo[3.1.1]heptane-2-carboxaldehyde	2.03 ± 2.02	0	0	0	0	Aldehydes
57	10.005	98-55-5	$C_{10}H_{18}O$	α,α,4-Trimethyl-3-cyclohexene-1-methanol	0	8.67 ± 5.87	6.07 ± 4.56	10.6 ± 0	6.97 ± 0.2	Alcohols
58	10.131	18486-69-6	$C_{10}H_{14}O$	Myrtenal	15.58 ± 4.97	12.57 ± 6.47	22.41 ± 3.24	19.3 ± 10.35	7.58 ± 0.27	Aldehydes
59	10.247	16721-38-3	$C_{10}H_{18}O$	3-Methyl-6-(1-methylethyl)-2-cyclohexen-1-ol	0	0	0	0	0.86 ± 0.05	Alcohols
60	10.346	1196-01-6	$C_{10}H_{14}O$	4,6,6-Trimethyl-bicyclo[3.1.1]hept-3-en-2-one	1.27 ± 0.37	0.65 ± 0.91	1.14 ± 0.86	3.2 ± 2.28	0	Ketones
61	10.452	13851-11-1	$C_{12}H_{20}O_2$	Fenchel acetate	0.56 ± 0.49	1.44 ± 0.63	0.7 ± 0.09	0	0	Esters
62	10.542	1197-06-4	$C_{10}H_{16}O$	2-Methyl-5-(1-methylethenyl)-2-cyclohexen-1-ol	1.9 ± 1.01	0	2.82 ± 0.05	3.06 ± 1.68	3.05 ± 0.46	Alcohols

Table 1 (*Contd.*)

No.	Retention time	CAS#	Molecular formula	Name	VHPD	HAD	SD	FD	FA	Classification
63	10.606	7492-41-3	$C_{11}H_{18}O_2$	Bornyl formate	0	0.8 ± 1.16	0.9 ± 0.49	0.45 ± 0.21	0	Esters
64	10.727	106-26-3	$C_{10}H_{16}O$	3,7-Dimethyl-2,6-octadienal	0	0	0	0.77 ± 0.51	0	Aldehydes
65	10.73	556-67-2	$C_8H_{24}O_4Si_4$	Octamethyl-cyclotetrasiloxane	1.08 ± 0.25	1.78 ± 1.55	0.74 ± 0.73	1.57 ± 0	0	Others
66	10.793	34246-57-6	$C_{10}H_{12}O$	3-Isopropylbenzaldehyde	0	0	0	2.69 ± 4.65	0	Aldehydes
67	10.848	122-03-2	$C_{10}H_{12}O$	Cuminaldehyde	2.61 ± 0.9	0	0	10.56 ± 0	6.08 ± 0.53	Aldehydes
68	10.895	106-25-2	$C_{10}H_{18}O$	3,7-Dimethyl-2,6-octadien-1-ol	0	0	0	0	1.87 ± 0.26	Alcohols
69	10.965	35907-10-9	$C_{10}H_{16}O$	2-Methylene-5-(1-methylethenyl)-cyclohexanol	0	0	0	0	0.92 ± 0.06	Alcohols
70	11.036	55050-40-3	$C_{10}H_{16}O$	7-Methyl-3-methylene-6-octenal	0	0	0	0	0.8 ± 0.04	Aldehydes
71	11.066	106-24-1	$C_{10}H_{18}O$	*trans*-Geraniol	0.46 ± 0.43	0	0	0	0	Alcohols
72	11.223	141-27-5	$C_{10}H_{16}O$	*E*-Citral	0	0.43 ± 0.21	0	0.74 ± 0.42	1.06 ± 0.09	Aldehydes
73	11.342	2111-75-3	$C_{10}H_{14}O$	4-(1-Methylethenyl)-1-cyclohexene-1-carboxaldehyde	0	0	0	3.97 ± 2.27	3.65 ± 0.25	Aldehydes
74	11.48	92618-89-8	$C_{12}H_{20}O_2$	Acetic acid, 1,7,7-trimethyl-bicyclo[2.2.1]hept-2-yl ester	0	0	0	0	19.11 ± 1.48	Acids
75	11.619	76-49-3	$C_{12}H_{20}O_2$	Bornyl acetate	170.73 ± 46.39	180.92 ± 35.07	197.38 ± 114.08	202.12 ± 11.32	0	Esters
76	11.679	30649-97-9	$C_{11}H_{16}O_2$	Methyl 6,6-dimethylbicyclo[3.1.1]hept-2-ene-2-carboxylate	0	0	8.76 ± 3.88	7.5 ± 3.75	13.32 ± 1.01	Esters
77	11.868	1686-15-3	$C_{12}H_{18}O_2$	Pinocarvyl acetate	0	0	0	0	2.56 ± 0.29	Esters
78	12.057	1079-01-2	$C_{12}H_{18}O_2$	Myrtenyl acetate	1.06 ± 1.01	0.65 ± 0.01	1.81 ± 1.41	2.59 ± 1.61	0	Esters
79	12.094	673-84-7	$C_{10}H_{16}$	Alloocimene	0	0	0	0	0.32 ± 0.28	Hydrocarbons
80	12.408	80-26-2	$C_{12}H_{20}O_2$	Terpinyl acetate	0	3.01 ± 1.69	0	0	0	Esters
81	12.599	62635-56-7	$C_9H_{20}O_3$	1,2,6-Trimethoxy-hexane	1.59 ± 1.93	4.35 ± 4.78	1.12 ± 0.67	24.34 ± 15.13	6.65 ± 1.09	Others
82	12.679	541-02-6	$C_{10}H_{30}O_5Si_5$	Decamethyl-cyclopentasiloxane	0.48 ± 0.03	0	1.09 ± 1.25	0	0	Others
83	12.758	141-12-8	$C_{12}H_{20}O_2$	2,6-Octadien-1-ol, 3,7-dimethyl-, acetate	1.54 ± 0.08	3.6 ± 2.49	0	2.54 ± 0	0	Esters
84	12.871	138874-68-7	$C_{15}H_{24}$	Copaene	2.36 ± 1.3	5.39 ± 6.75	2.94 ± 3.68	1.13 ± 0.67	5.71 ± 0.14	Hydrocarbons
85	12.915	14010-23-2	$C_{19}H_{38}O_2$	Heptadecanoic acid, ethyl ester	0	0	0	6.25 ± 0	0	Esters
86	12.991	72439-85-1	$C_{20}H_{54}O_7Si_6$	3,5-Dibutoxy-1,1,1,7,7,7-hexamethyl-3,5-bis(trimethylsiloxy)tetrasiloxane	0.8 ± 0.24	0	1.02 ± 0.2	2.14 ± 1.23	1.96 ± 0.12	Others
87	13.13	18479-51-1	$C_{10}H_{20}O$	Dihydro linalool	0	0	0	0.39 ± 0.34	0	Alcohols
88	13.147	112-54-9	$C_{12}H_{24}O$	Dodecanal	0	0	0	0	1.77 ± 0.1	Aldehydes
89	13.372	151283-74-8	$C_{15}H_{24}$	Gurjunene	0.25 ± 0.22	0.27 ± 0.1	0.5 ± 0.73	0.34 ± 0.16	0	Hydrocarbons
90	13.456	512-61-8	$C_{15}H_{24}$	1,7-Dimethyl-7-(4-methyl-3-pentenyl)-tricyclo[2.2.1.0(2,6)]heptane	0.47 ± 0.53	0.81 ± 1.05	0.74 ± 1.07	0	0	Hydrocarbons
91	13.536	13877-93-5	$C_{15}H_{24}$	Caryophyllene	1.91 ± 0.9	2.71 ± 2.33	1.85 ± 1.65	3.51 ± 2.62	12.21 ± 0.63	Hydrocarbons
92	13.629	80923-88-2	$C_{15}H_{24}$	Himachalene	0	0	0	0.12 ± 0.21	0	Hydrocarbons
93	13.64		$C_{15}H_{24}$	α-Bergamotene				0		Hydrocarbons

Table 1 (*Contd.*)

No.	Retention time	CAS#	Molecular formula	Name	VHPD	HAD	SD	FD	FA	Classification
		17699-05-7			0.76 ± 0.34	0.98 ± 1.24	1 ± 1.22		0.86 ± 0.74	
94	13.81	109119-91-7	$C_{15}H_{24}$	Aromadendrene	0	0	4.54 ± 6.55	6.64 ± 5.75	8.83 ± 0.67	Hydrocarbons
95	14.006	6753-98-6	$C_{15}H_{24}$	α-Humulene	2.32 ± 1.09	2.66 ± 1.47	1.55 ± 0.97	16.41 ± 13.5	5.63 ± 0.31	Hydrocarbons
96	14.118	25246-27-9	$C_{15}H_{24}$	[1ar-(1aα,4aβ,7α,7aβ,7bα)]-1*H*-Cycloprop[*e*]azulene, decahydro-1,1,7-trimethyl-4-methylene	2.98 ± 1.72	1.46 ± 1.12	1.8 ± 1.23	3.58 ± 2.07	2.81 ± 0.11	Hydrocarbons
97	14.23	22567-17-5	$C_{15}H_{24}$	γ-Gurjuene	0	0	0	0.21 ± 0.36	0.75 ± 0.65	Hydrocarbons
98	14.26	3856-25-5	$C_{15}H_{24}$	α-Copaene	0.55 ± 0.66	0	1.21 ± 1.94	0.34 ± 0.19	0	Hydrocarbons
99	14.405	26620-71-3	$C_{15}H_{24}$	Aristolochene	0	0	0.35 ± 0.42	0.31 ± 0.54	5.39 ± 0.17	Hydrocarbons
100	14.448	17066-67-0	$C_{15}H_{24}$	Decahydro-4a-methyl-1-methylene-7-(1-methylethenyl)-, [4ar-(4aα,7α,8aβ)]-naphthalene	0	0	0	17.83 ± 15.44	0	Hydrocarbons
101	14.456	20071-49-2	$C_{15}H_{24}$	γ-Maaliene	0.61 ± 0.71	0	0	0.72 ± 0.63	0	Hydrocarbons
102	14.458	97408-24-7	$C_{15}H_{24}$	Maaliene	1.53 ± 1.32	0	0	0	0	Hydrocarbons
103	14.545	21747-46-6	$C_{15}H_{24}$	Ledene	0	0	1.33 ± 1.31	0	0	Hydrocarbons
104	14.553	74409-93-1	$C_{15}H_{24}$	Viridiflorene	0	0	0	10.6 ± 0.18	0	Hydrocarbons
105	14.566	100762-46-7	$C_{15}H_{24}$	Bicyclogermacrene	6 ± 2.69	7.64 ± 7.14	0	1.58 ± 0.73	9.2 ± 0.77	Hydrocarbons
106	14.692	508-77-0	$C_{30}H_{44}O_9$	Cymarin	0	0	0	1.69 ± 0.56	0	Others
107	14.71	90131-02-5	$C_{15}H_{26}O$	Sesquicineole	0.54 ± 0.2	0.91 ± 0.62	0	0	0	Others
108	14.847	16729-01-4	$C_{15}H_{24}$	δ-Cadinene	0	0	0	1.74 ± 1.51	0	Hydrocarbons
109	14.86	483-76-1	$C_{15}H_{24}$	δ-Cadinene	2.4 ± 2.08	4.86 ± 6.18	2.65 ± 3.4	0.79 ± 1.37	4.01 ± 0.43	Hydrocarbons
110	15.007	17699-14-8	$C_{15}H_{24}$	α-Cubebene	0	0	0	0	0.61 ± 0.54	Hydrocarbons
111	15.011	5986-49-2	$C_{15}H_{26}O$	Palustrol	0	0	0	1.91 ± 0	0	Alcohols
112	15.061	95841-70-6	$C_{27}H_{45}BrO_2$	5β,6β-Epoxy-7α-bromocholestan-3β-ol	0.97 ± 0.08	0	0	0	0	Alcohols
113	15.54	577-27-5	$C_{15}H_{26}O$	Ledol	0	0	0	1.7 ± 0	0	Alcohols
114	15.661	72203-24-8	$C_{15}H_{24}O$	Spathulenol	3.48 ± 1.81	2.06 ± 1.37	3.26 ± 1.19	4.36 ± 2.88	9.46 ± 0.92	Alcohols
115	15.741	81968-62-9	$C_{15}H_{24}O$	(1*R*,7*S*,*E*)-7-Isopropyl-4,10-dimethylenecyclodec-5-enol	0	0	0	6.08 ± 0	0	Alcohols
116	15.758	17627-43-9	$C_{15}H_{24}O$	Caryophyllene oxide	1.03 ± 0.89	0	0	0.57 ± 0	2.15 ± 0.12	Ethers
117	15.868	19078-39-8	$C_{15}H_{26}O$	Viridiflorol	0	0.3 ± 0.14	0	1.15 ± 0	0	Alcohols
118	15.943	62376-14-1	$C_{12}H_{20}$	1,2,3,6-Tetramethyl-bicyclo[2.2.2]oct-2-ene	0	0	0	2.68 ± 0	0	Others
119	16.089	19888-34-7	$C_{15}H_{24}O$	Humulene epoxide II	0.22 ± 0.26	0.38 ± 0.36	0.29 ± 0.33	2.85 ± 2.34	0.91 ± 0.1	Others
120	16.397	79580-01-1	$C_{15}H_{24}O$	11,11-Dimethyl-, 4,8-bis(methylene)-bicyclo[7.2.0]undecan-3-ol	0	0	0	0	4.17 ± 0.29	Alcohols
121	16.457	556-68-3	$C_{16}H_{48}O_8Si_8$	Hexadecamethyl-cyclooctasiloxane	0.68 ± 0.67	0.81 ± 0.83	1.48 ± 1.53	1.51 ± 0.86	0	Others
122	16.61	6750-60-3	$C_{15}H_{24}O$	[1ar-(1aα,4aα,7β,7aβ,7bα)]-1*H*-Cycloprop[e]azulen-7-ol, decahydro-1,1,7-trimethyl-4-methylene	0	0	0	3.41 ± 0	0	Alcohols
123	16.642	1139-30-6	$C_{15}H_{24}O$	Caryophyllene oxide	1.18 ± 0.93	1.39 ± 0.68	3.46 ± 2.91	2.85 ± 2.15	3.22 ± 1.08	Others

Table 1 (*Contd.*)

No.	Retention time	CAS#	Molecular formula	Name	VHPD	HAD	SD	FD	FA	Classification
124	16.683	515-20-8	$C_{15}H_{24}O$	Costol	0	0	0	1.2 ± 0	0	Alcohols
125	16.753	53820-13-6	$C_{15}H_{24}O_2$	Chrysantenyl 2-methuylbutanoate	0	0	0	0.55 ± 0	0	Esters
126	16.793	552-02-3	$C_{15}H_{26}O$	Epiglobulol	0	0	0.38 ± 0.42	0	0	Alcohols
127	17.03	71579-69-6	$C_{18}H_{52}O_7Si_7$	3-Isopropoxy-1,1,1,7,7,7-hexamethyl-3,5,5-tris(trimethylsiloxy)tetrasiloxane	0	0	0.62 ± 0.12	1.17 ± 1.02	0	Others
128	17.133	145344-72-5	$C_{16}H_{48}O_{10}Si_9$	2-(2′,4′,4′,6′,6′,8′,8′-heptamethyltetrasiloxan-2′-yloxy)-2,4,4,6,6,8,8,10,10-nonamethylcyclopentasiloxane	0.32 ± 0.09	0.31 ± 0.27	0.2 ± 0.09	0.34 ± 0.59	0.34 ± 0.03	Others
129	17.14	19095-24-0	$C_{16}H_{50}O_7Si_8$	1,15-Dihydrogenhexadecamethylo	0.11 ± 0.1	0.09 ± 0.07	0	0.13 ± 0.23	0.27 ± 0.18	Ethers
130	17.76	3155-71-3	$C_{14}H_{22}O$	2-Methyl-, 4-(2,6,6-trimethyl-1-cyclohexen-1-yl)-but-2-enal	0	0.09 ± 0.08	0	0	0	Aldehydes
131	18.142	638-36-8	$C_{20}H_{42}$	2,6,10,14-Tetramethyl-hexadecane	0	0	0	0	0.05 ± 0.04	Hydrocarbons
132	18.309	540-97-6	$C_{12}H_{36}O_6Si_6$	Dodecamethyl-cyclohexasiloxane	0.99 ± 0.91	0.98 ± 0.66	2.35 ± 0.42	3.27 ± 1.71	0.13 ± 0.12	Others
133	18.438	3243-36-5	$C_{16}H_{26}O$	Ambrial	0.55 ± 0.05	0.49 ± 0.21	1.03 ± 0.45	0.69 ± 0.33	3.57 ± 0.27	Ketones

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-

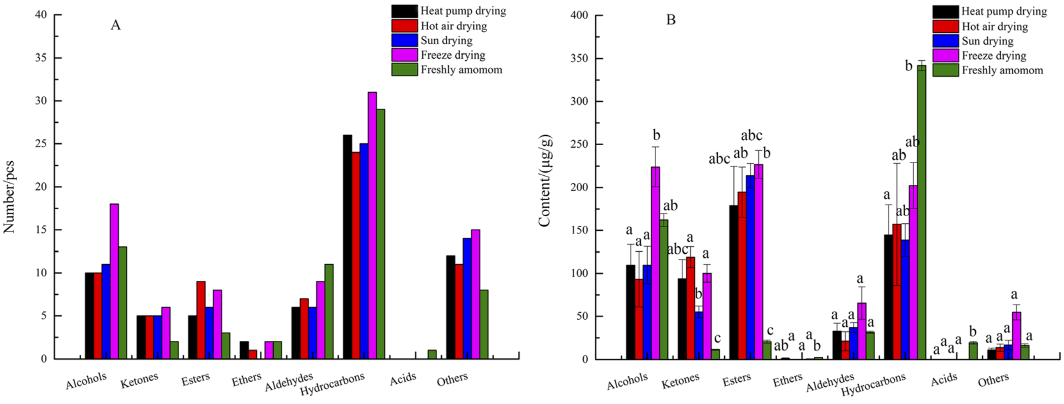
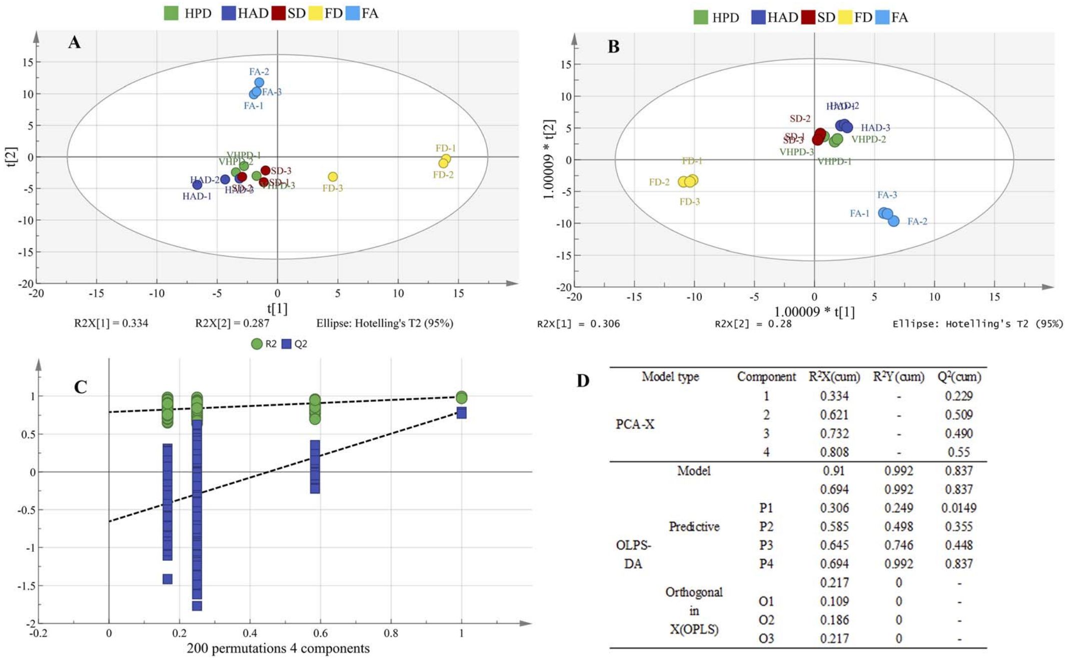


Fig. 1 Analysis of species (A) and content of VOCs (B) of *Amomum villosum* Lour. 'Guiyan1' samples obtained *via* four drying methods.



Model type		Component	R²X(cum)	R²Y(cum)	Q²(cum)
PCA-X		1	0.334	-	0.229
		2	0.621	-	0.509
		3	0.732	-	0.490
		4	0.808	-	0.55
OLPS-DA	Model		0.91	0.992	0.837
	Predictive		0.694	0.992	0.837
		P1	0.306	0.249	0.0149
		P2	0.585	0.498	0.355
		P3	0.645	0.746	0.448
		P4	0.694	0.992	0.837
	Orthogonal in X(OPLS)		0.217	0	-
		O1	0.109	0	-
		O2	0.186	0	-
		O3	0.217	0	-

Fig. 2 Score plot of the PCA-X model (A), OPLS-DA model (B), cross-validation of the OPLS-DA model (C), and model parameters of VOCs among *Amomum villosum* Lour. 'Guiyan1' samples obtained *via* four drying methods (D). Hereinafter, abbreviations HPD, HAD, SD, FD, and FA correspond to heat pump-dried, hot air-dried, sun-dried, freeze-dried, and fresh 'Guiyan1' samples, respectively.

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.[32] It can be seen in Fig. 2(D) that the OPLS-DA model featured values $R^2\text{X} = 0.910$ and $Q^2 = 0.837$ and, thus, presented more data variation than the PCA-X model with $R^2\text{X} = 0.808$ 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[33]. 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-bornanone (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), α, α,4-trimethyl-3-cyclohexene-1-methanol (57), *endo*-borneol (52), 2,6-octadien-1-ol, 3,7-dimethyl-, acetate (83), 2-bornanone (45), methyl 6,6-dimethylbicyclo [3.1.1] hept-2-ene-2-carboxylate (76), and sesquicineole (107).

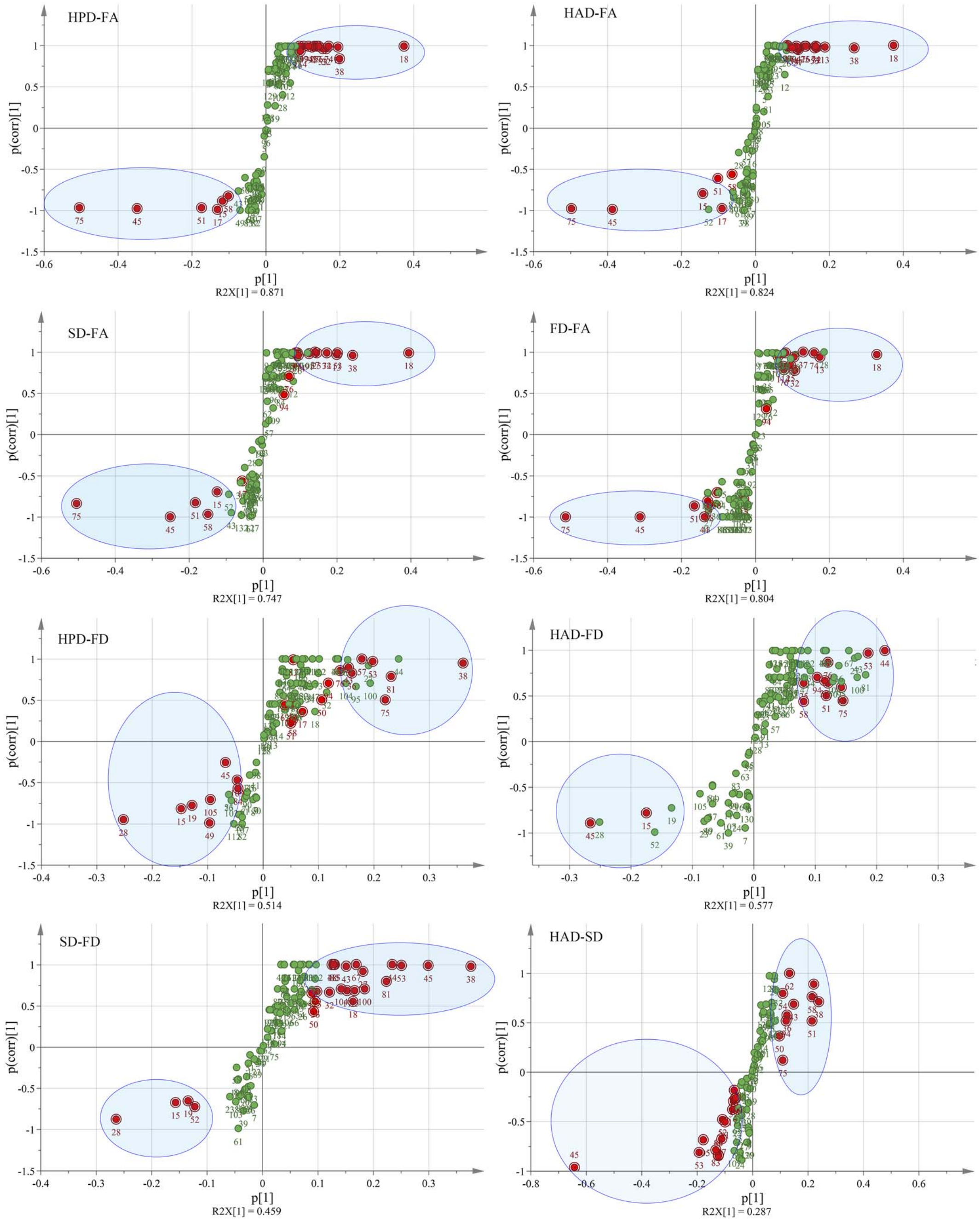


Fig. 3 S-plot of VOC-based OPLS-DA model analysis of *Amomum villosum* Lour. 'Guiyan1' samples obtained *via* four drying methods.

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-bornanone (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 ± 16.33) μg g^{-1}, followed by heat pump drying, freeze-drying, and sun drying methods with (80.11 ± 19.46) μg g^{-1}, (74.08 ± 0.00) μg g^{-1}, and (40.73 ± 3.46) μg 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 ± 0.82 μg g^{-1}, 12.78 ± 0.30 μg g^{-1}, and 7.62 ± 0.04 μg 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 ± 1.39 and 11.78 ± 1.45 μg g^{-1}, respectively. Terpinyl acetate (80) and acetic acid hexyl ester (24) were present only in hot air-dried samples; 5β,6β-epoxy-7α-bromocholestan-3β-ol (112) was present only with the heat

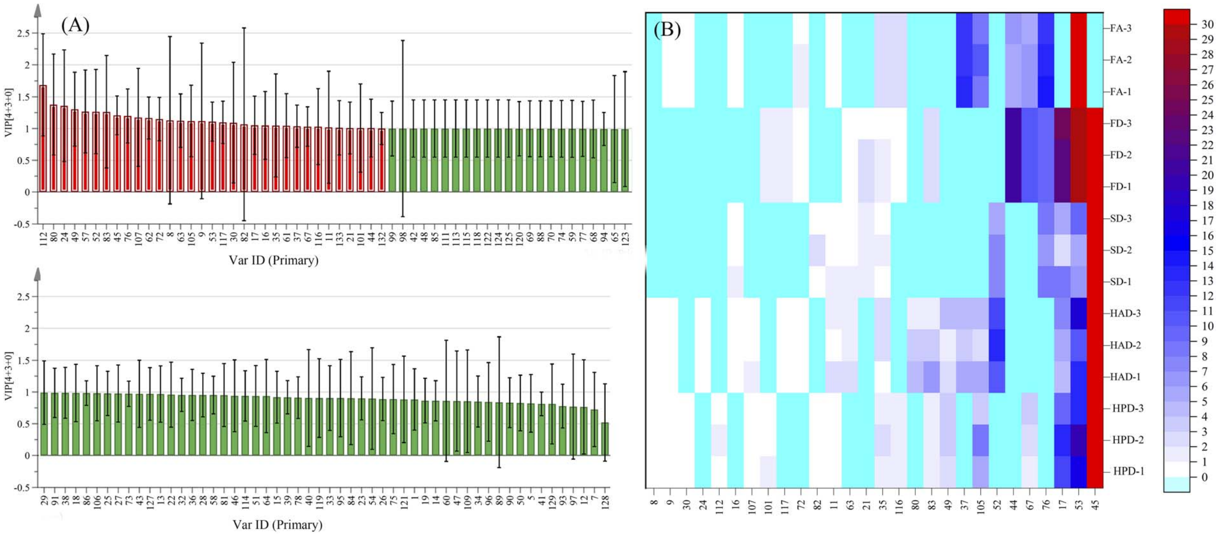


Fig. 4 Distribution of VIP values predicted *via* OPLS-DA (A) and the heatmap of different metabolites in *Amomum villosum* Lour. 'Guiyan1' samples obtained *via* four drying methods (B).

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, (1*S*)-linalool, α-pinene, terpinene-4-ol, limonene, 1-methyl-4-(1-methylethyl)-1,4-cyclohexadiene, acetic acid, 1,7,7-trimethyl-bicyclo[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 acetate camphor, 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.[34] 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.[26,35]

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-bornanone 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 amomums 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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