Acetoxy-1,8-cineoles as Aroma Constituents of *Alpinia galanga* Willd.

Kikue Kubota,* Kae Nakamura (Murayama), and Akio Kobayashi

Laboratory of Food Chemistry, Ochanomizu University, 2-1-1 Ohtsuka, Bunkyo-ku, Tokyo 112-8610, Japan

Masayasu Amaike

Technical Research Center, T. Hasegawa Company Ltd., 335 Kariyado, Nakahara-ku, Kawasaki, 211-0022, Japan

Four isomers of acetoxycineoles, (*trans* and *cis*)-2- and 3-acetoxy-1,8-cineoles, were identified as the odorous components of the rhizomes of greater galangal. Their structures were confirmed by comparing the retention indices by GC and the mass spectra with those of synthesized compounds. The concentration of *trans*-3-acetoxy-1,8-cineole was the highest among the isomers. The isomers presented individual odor features: the (*trans* and *cis*)-2 isomers respectively exhibited woody and sweet aromas, while the (*trans* and *cis*)-3 isomers respectively showed sweet floral and camphoraceous aromas. Of these, *trans*-2-acetoxy-1,8-cineole seemed to have the strongest qualitative effect on the characteristic flavor of greater galangal.

Keywords: *Alpinia galanga Willd.; greater galangal; rhizome; flavor component; trans-2-acetoxy-1,8-cineole; cis-2-acetoxy-1,8-cineole; trans-3-acetoxy-1,8-cineole; cis-3-acetoxy-1,8-cineole*

INTRODUCTION

The rhizomes of greater galangal (*Alpinia galanga* Willd.) are widely used as a spice in Asian ethnic dishes, and the plant is also used in traditional medicine. As a characteristic aroma component of *A. galanga*, (*S*)-1′-acetoxychavicol acetate has recently been widely noted because of its biological activities as an antiulcer compound (Mitsui et al., 1976), an antimicrobiological agent (Janssen and Scheffer, 1985), an inhibitor of xanthine oxidase (Noro et al., 1988; Ohnishi et al., 1996) and an antitumor promotor (Itokawa et al., 1987; Kondo et al., 1993). While the essential oil has long been studied, the flavor constituents had not been completely elucidated, leaving much remaining to be investigated (Scheffer et al., 1981; Pooter et al., 1985; Charles et al., 1992). In a previous paper (Mori et al., 1995), we showed 1,8-cineole, linalool, geranyl acetate, eugenol, and chavicol acetate as potent odorants, and acetic acid, bornyl acetate, citronellyl acetate, *trans*-2-acetoxy-1,8-cineole (2-exo-acetoxy-1,8-cineole) and its isomer, methyl eugenol and 1′-acetoxychavicol acetate as other important odor constituents of *A. galanga* by using a modified aroma extract dilution analysis (Ullrich and Grosch, 1987). Our continuing study showed that *A. galanga* contained the isomers of acetoxy-1,8-cineoles in the volatile fraction. Although 2-hydroxy-1,8-cineoles have been found in grape (Bitteur et al., 1990) and ginger lily flowers (Yamada and Ikeda, 1991), acetoxy-1,8-cineoles are not common volatile components in other plants, except for *trans*-2-acetoxy-1,8-cineole which has tentatively been identified in a weed (Vera, 1993). This paper describes the isolation and characterization of 2- and 3-acetoxy-1,8-cineoles in the volatile fraction of the rhizomes of *A. galanga*. In a previous study (Mori et al., 1995), *trans*-2-acetoxy-1,8-cineole was identified from mass spectral and GC−FTIR data. In this present report, we confirm the existence of four isomers of 2- and 3-acetoxy-1,8-cineoles by comparing their spectral data with those of synthesized compounds.

* Author to whom correspondence should be addressed (fax +81-3-5978-5759; e-mail kubota@comfort.eng.ocha.ac.jp).

EXPERIMENTAL PROCEDURES

Reagents. Trioctyl methylammonium chloride, *m*-chloroperbenzoic acid (MCPBA) and α-terpineol (Kanto Chemical Co., Tokyo, Japan), 1,8-cineole (Wako Pure Chemicals, Tokyo, Japan) and piperitenone (Nippon Terpene Chemical Co., Tokyo, Japan) were used.

Materials. Fresh rhizomes of greater galangal (*Alpinia galanga* Willd.) were purchased from a local market in Tokyo, after being grown and transported by air from Thailand.

^{1}H- and ^{13}C NMR Spectral Measurements. ^{1}H- and ^{13}C NMR spectra of the isolated compounds and synthesized compounds were measured with a JEOL JNM-JSX 270 FT-NMR spectrometer. Chemical shift data are expressed as δ values in relation to TMS as an internal standard.

GC and GC/MS. The analytical and sniffing GC conditions were as follows: Hewlett-Packard 5890 series II GC instrument equipped with a 0.25 mm × 60 m (0.25 μm film thickness) DB-WAX (J&W Scientific, Folsom, CA) column and a flame ionization detector; He as the carrier gas; oven temperature held at 60 °C for 4 min and then increased at 2 °C/min to 200 °C; injection port and detector temperature at 220 °C. The end of the column divided to feed the FID and ODO-1 sniffing adapter (GL Sciences, Tokyo, Japan) at 1:2. EI-MS data were collected by a Hewlett-Packard 5972 GC-MS instrument with the same type of column operated under the same conditions as those just described.

Preparation of the Oxygenated Volatile Fraction. The fresh rhizomes (200 g) of *A. galanga* were mechanically ground in 1 L of purified water, and the volatiles were collected by steam distillation under reduced pressure (10 Torr) with a rotary evaporator, before being salted-out and extracted with refined diethyl ether. This procedure was conducted three

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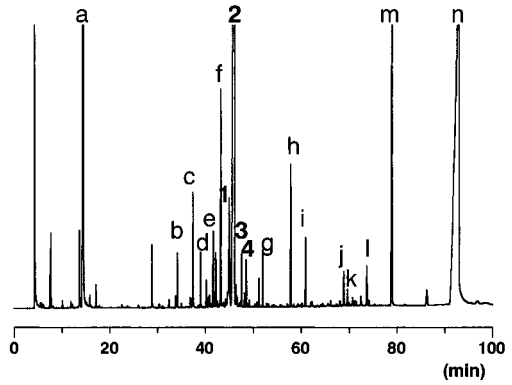

Figure 1. Gas chromatogram of the oxygenated compound fraction eluted with pentane−ether (1:1) by SiO_2 column chromatography. Refer to the Experimental Procedures for details of the conditions. Peaks: a, 1,8-cineole; b, linalool; c, terpinen-4-ol; d, citronellyl acetate; e, δ-terpineol; f, α-terpineol; g, thymol acetate; h, chavicol acetate; i, methyleugenol; g, eugenol; k, thymol; l, eugenyl acetate; m, chavicol; and n, 1′-acetoxychavicol acetate.

times, the combined ethereal fraction was dried over anhydrous sodium sulfate, and the ether was distilled off at 39.5 °C to obtain a volatile concentrate (0.6 g, 0.1%/wet weight of rhizomes). The concentrate was subsequently submitted to silica gel column chromatography to separate the hydrocarbon and oxygenated compounds by eluting with pentane and pentane−diethyl ether (1:1), respectively. The latter fraction (0.08%/wet weight) was submitted to gas chromatographic−mass spectrometric (GC−MS) analysis and preparative GC.

Isolation and Identification of the Unknown Compounds. The biggest unknown peak (peak 2 in Figure 1) was trapped by preparative GC. A GL Sciences 380 GC instrument was used and was equipped with a 2.0 mm i.d. × 1.5 m glass column packed with 5% PEG 20M on gas chrom Q operated under the following conditions: oven temperature, 60 °C with 4-min hold and then increased at 2 °C/min to 200 °C; injection and detection port temperature, 200 °C; carrier gas, He at 40 mL/min. The structure was elucidated from the following MS, IR, and NMR spectral data. IR (film, cm^{-1}): 1739 and 1236 ($COCH_3$). ^1H NMR ($CDCl_3$, TMS): δ 1.08 (3H, s), 1.27 (3H, s), 1.28 (3H, s), 2.07 (3H, s, $COCH_3$), 1.46−1.93 (6H, m), 2.15−2.25 (1H, m), 5.30−5.35 (1H, m). ^{13}C NMR ($CDCl_3$, TMS): δ 14.9, 21.4, 27.0, 28.5, 28.8, 30.9, 37.2, 40.1, 69.4, 70.6, 73.3, 170.8 (C=O).

The structures of compounds **1**−**4** were evaluated by comparing Kovats indices of the GC and mass spectra with those of synthetic compounds.

Syntheses of 2- and 3-Acetoxy-1,8-cineoles. *trans*-2-Hydroxy-1,8-cineole was synthesized according to the method of Bitteur et al. (1990) by oxidizing α-terpineol with *m*-chloroperbenzoic acid (MCPBA). *cis*-2-Hydroxy-1,8-cineole was prepared from 1,8-cineole and MCPBA according to the literature (Asakawa et al., 1988), the desired compound being separated by SiO_2 column chromatography (benzene/ethyl acetate). *cis*-3-Hydroxy-1,8-cineole was synthesized from piperitenone and trioctylmethylammonium chloride in a 35% sulfuric acid solution, being reduced with $LiALH_4$ in the way described in the patent (Kobayashi et al., 1997). *trans*-3-Hydroxy-1,8-cineole was prepared by inverting the *cis* isomer with 3,5-dinitrobenzoic acid, triphenylphosphine and diethyl azodicarbonate, and then hydrolyzing with a base (Kobayashi et al., 1997). The structures of the synthesized hydroxy-1,8-cineoles were confirmed by comparing their spectral data with those in the literature. The desired acetoxy-1,8-cineoles were prepared by acetylating the corresponding hydroxy-1,8-cineole with acetic anhydride in pyridine in the usual way, the structure then being confirmed by spectral analyses. The mass spectra of the synthesized acetoxy cineoles were almost the same as shown in Table 1, and the IR absorbance in the neighborhood of 1740 and 1235 cm^{-1} shows the existence of an acetoxy group in each molecule. NMR analytical data ($CDCl_3$, TMS, numbering of carbon atoms refers to Figure 2) are as follows.

*trans-2-Acetoxy-1,8-cineole (**1**)*: ^1H NMR (270 MHz): δ 1.04 (3H, s, Me-7), 1.21 (3H, s, Me-9), 1.28 (3H, s, Me-10), 1.25−1.33 (1H, m, H-3), 1.48−1.55 (2H, m, H-4, 5), 1.57−1.65 (1H, m, H-6), 1.81−1.88 (1H, m, H-6), 2.0−2.05 (1H, m, H-5), 2.06 (3H, s, H-12), 2.56−2.68 (1H, m, H-3), 4.66−4.71 (1H, m, H-2); ^{13}C NMR (67.8 MHz): δ 21.3 (C-12), 22.0 (C-5), 24.1 (C-7), 25.9 (C-6), 28.8 (C-10), 28.9 (C-9), 32.7 (C-3), 33.8 (C-4), 70.8 (C-1), 72.9 (C-2), 73.7 (C-8), 170.5 (C-11).

*cis-2-Acetoxy-1,8-cineole (**4**)*: ^1H NMR (270 MHz): δ 1.05 (3H, s, Me-7), 1.27 (3H, s, H-9), 1.29 (3H, s, Me-10), 1.36−1.56 and 1.68−2.17 (7H, m, H-3, 4, 5, 6), 2.10 (3H, s, H-12), 4.68 (1H, dd, J = 3.3, 9.9, H-2); ^{13}C NMR (67.8 MHz): δ 21.2 (C-12), 21.8 (C-5), 22.9 (C-7), 28.0 (C-6), 28.8 (C-10), 29.7 (C-9), 32.3 (C-3), 33.3 (C-4), 70.8 (C-1), 72.8 (C-2), 73.7 (C-8), 171.1 (C-11).

*trans-3-Acetoxy-1,8-cineole (**2**)*: ^1H NMR (270 MHz): δ 1.08 (3H, s, Me-7), 1.27 (3H, s, Me-10), 1.29 (3H, s, Me-9), 1.35−1.48 (1H, m, H-2), 1.47−1.54 (1H, m, H-6), 1.63−1.92 (4H, m, H-4, 5, 6), 2.07 (3H, s, H-12), 2.16−2.23 (1H, m, H-2), 5.30−5.33 (1H, m, H-3); ^{13}C NMR (67.8 MHz): δ 14.9 (C-5), 21.4 (C-12), 27.0 (C-7), 28.4 (C-10), 28.7 (C-9), 30.9 (C-6), 37.1 (C-4), 40.1 (C-2), 69.4 (C-3), 70.5 (C-1), 73.2 (C-8), 170.7 (C-11).

*cis-3-Acetoxy-1,8-cineole (**3**)*: ^1H NMR (270 MHz): δ 1.12 (3H, s, Me-7), 1.24 (3H, s, Me-10), 1.35 (3H, s, Me-9), 1.39−1.48 (1H, m, H-5), 1.39−1.54 (2H, m, H-6), 1.58−1.60 (1H, m, H-4), 1.70−1.76 (1H, m, H-2), 2.06 (3H, s, H-12), 2.03−2.13 (2H, m, H-2, 5), 4.95−5.0 (1H, m, H-3); ^{13}C NMR (67.8 MHz): δ 21.1 (C-5), 21.5 (C-12), 26.8 (C-7), 30.2 (C-6, 10), 30.4 (C-9), 37.4 (C-4), 40.3 (C-2), 70.0 (C-1), 72.8 (C-3), 73.2 (C-8), 170.7 (C-11).

RESULTS AND DISCUSSION

The gas chromatogram of the oxygenated compound fraction eluted with pentane−diethyl ether (1:1) by silica gel column chromatography of the essential oil from the rhizomes is shown in Figure 1. Although most of the components, peaks a−n, and *trans*-2-acetoxy-1,8-cineole (**1**) were identified in our previous work (Mori et al., 1995), peaks 2−4 (compounds **2**−**4**) remained unidentified. Compound **2** occupied 17.3% of the total peak area in GC and it was one of the main volatile constituents, as well as 1,8-cineole (peak a) and 1′-acetoxychavicol acetate (peak n), in the essential oil, but its structure could not be determined by GC−MS analysis, because the detailed MS data and retention indices by GC could not be found in the literature, except for *trans*-2-acetoxy-1,8-cineole (**1**). Consequently, since compounds **1**−**4** were thought to be structurally related from their mass spectral data and presented individually interesting odors, more detailed investigations were carried out.

Identification of 2- and 3-Acetoxy-1,8-cineoles. The retention indices by GC, respective concentrations in the rhizomes, and mass spectra of compounds **1**−**4** are shown in Table 1. The results of the GC−MS analysis showed that they had the same molecular ion at *m*/*z* 212, which indicated that they were isomers of each other. In addition, compounds **1** and **4**, and compounds **2** and **3** respectively presented almost the same MS fragmentation profiles; they are considered to have been stereoisomers of each other in each group. In our previous study (Mori et al., 1995), the structure of compound **1** had been determined as *trans*-2-acetoxy-1,8-cineole (*trans*-1,3,3-trimethyl-2-oxabicyclo[2.2.2]oct-6-yl acetate) from its MS and FT-IR data. Consequently, compound **4** is proposed to have been *cis*-2-acetoxy-1,8-cineole because of the analogous MS data. The MS fragment profiles of **2** and **3** were slightly different from

Table 1. Concentration and Mass Spectral Data of Acetoxy-1,8-cineoles Isolated from the Rhizomes of Greater Galangal

compound	RI[a]	ppm[b]	MS data, *m/z* (relative intensity)
trans-2-acetoxy-1,8-cineole (**1**)	1737	13	212 (M^+, 16), 155 (3), 137 (4), 126 (40), 109 (32), 108 (69), 93 (32), 82 (28), 71 (33), 43 (100)
trans-3-acetoxy-1,8-cineole (**2**)	1749	128	212 (M^+, 6), 197 (10), 152 (28), 137 (40), 109 (43), 108 (16), 93 (34), 83 (19), 82 (17), 67 (13), 43 (100)
cis-3-acetoxy-1,8-cineole (**3**)	1780	3	212 (M^+, 1), 197 (20), 152 (13), 137 (19), 109 (36), 93 (31), 83 (20), 67 (10), 43 (100)
cis-2-acetoxy-1,8-cineole (**4**)	1797	2	212 (M^+, 15), 155 (5), 137 (4), 126 (35), 109 (23), 108 (65), 93 (30), 82 (28), 71 (31), 43 (100)

[a] Kovats index (DB-WAX column). [b] Concentration for the wet weight of the rhizomes calculated from the yield of the aroma concentrate and peak area % by GC.

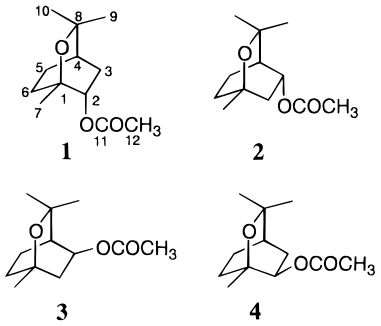

Figure 2. Structures of the identified acetoxycineoles: **1**, *trans*-2-acetoxy-1,8-cineole; **2**, *trans*-3-acetoxy-1,8-cineole; **3**, *cis*-3-acetoxy-1,8-cineole; and **4**, *cis*-2-acetoxy-1,8-cineole.

those of **1** and **4**. Since **2** presented a large peak area by GC of the oxygenated compound fraction as shown Figure 1, it was trapped by preparative GC and submitted to the spectral analyses that are summarized in the Experimental Procedures. The absorbances at 1739 and 1236 cm^{-1} in the IR spectrum, a singlet signal at δ 2.07 in the ^1H NMR spectrum and the signal of a carbonyl carbon at δ 170.8 in the ^{13}C NMR spectrum indicated the presence of an acetoxy group. The signals of three methyl groups at δ 1.08, 1.27, and 1.28, and the methylene protons at δ 1.46−2.25 in the ^1H NMR spectrum supported a bicyclic structure, and the chemical shift profiles by ^1H and ^{13}C NMR are similar to those of *trans*- or *cis*-3-hydroxy-1,8-cineole in the literature (Miyazawa et al., 1989). In addition, compared with the data of synthesized *cis*-3-acetoxy-1,8-cineole in the literature, the chemical shift of a methyne proton (δ 5.3−5.33, H-5) attached to the carbon-bearing acetoxy group was observed in a field lower by 0.35 ppm than in the literature for ^1H NMR (De Boggiatto et al., 1987). On the basis of these facts, the structure of **2** was determined to be *trans*-3-acetoxy-1,8-cineole (*trans*-1,3,3-trimethyl-2-oxabicyclo[2.2.2]oct-5-yl acetate), not the *cis*-isomer. Consequently, **3** was determined to be *cis*-3-acetoxy-1,8-cineole in analogy with **4**.

To establish the structures of **1**−**4**, all of them were synthesized as described in the Experimental Procedures. 2- and 3-hydroxy-1,8-cineoles (*trans* and *cis*) were first synthesized and isolated. After their structures had been established by comparing their spectral data with those in the literature (Miyazawa et al., 1989), each corresponding acetate was prepared in the usual way. Their NMR spectra were assigned by using C−H COSY and by reference to the data for *cis*-3-acetoxycineole (De Boggiatto et al., 1987) and for the glucosides of hydroxycineoles (Orihara and Furuta, 1994), as summarized in the Experimental Procedures. These results indicated that four isomers of acetoxy-1,8-cineoles had been synthesized, and the structures of compounds **1**−**4** in the rhizomes were established to be those shown in Figure 2 from the identical KI values by GC and MS analyses to those of the synthesized compounds.

Although 2- and 3-hydroxy-1,8-cineoles have been found in grape (Bitteur et al., 1990) and ginger lily flowers (Yamada and Ikeda, 1991) as volatile components, these acetyl derivatives are not naturally common. 3-Acetoxy-1,8-cineoles (*trans* and *cis*) and *cis*-2-acetoxy-1,8-cineole were identified for the first time as natural flavor constituents in this study.

Concentration and Odor Characteristics. The concentration of *trans* isomers was much higher than that of *cis* isomers in both 2- and 3-acetoxy-1,8-cineoles (Table 1). The proportion of the *trans*-3 isomer was also about 9-fold higher than that of the *trans*-2 isomer. The formation mechanism for these acetoxy derivatives of 1,8-cineole in the rhizomes is a subject worthy of further investigation. In addition, each acetoxy-1,8-cineole exhibited some interesting odor features: the *trans*-2 isomer exhibited a woody and galangal-like odor; the *cis*-2 isomer had a sweet aroma; the *trans*-3 isomer presented a sweet floral aroma; and the *cis*-3 isomer presented a camphorous odor which distinguished it from the other isomers. Although Mariani et al. (1995) have described the odorous note of synthesized *trans*-2-acetoxy-1,8-cineole to be woody, pine oil-like, and violet-like, the olfactory characters of the *cis*-2- and (*trans* and *cis*)-3-acetoxy-1,8-cineoles were also clarified for the first time in this study. In the previous work, the authors reported that galangal soup had a more woody, minty, and floral odorous note than ginger, and both *trans*-2- and *trans*-3-acetoxy-1,8-cineoles were selected as potent odorants of greater galangal by using an aroma extract dilution analysis (Mori et al., 1995). The results of this study allow us to conclude that the *trans*-2 and *trans*-3 isomers respectively contributed to the woody note and floral note of the rhizomes of greater galangal, while both the *cis*-2 and *cis*-3 isomers seem not to have had individual effects on the odor of the rhizomes, but in combination because of their low concentration.

Since each acetoxy-1,8-cineole identified here has three asymmetric carbons in the molecule, they have optical isomers. Their enantiomeric purity in *A. galanga* and the different odor characteristics of the optical isomers will be described elsewhere.

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