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Note

Effects of Storage Conditions on the Composition of *Citrus tamurana* Hort. ex Tanaka (*Hyuganatsu*) Essential Oil

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Compositional changes of *Citrus tamurana* Hort. ex Tanaka (*hyuganatsu*) essential oil were analyzed after storage for 1, 3, 6, and 9 weeks at −21, 5, 20, and 30°C. The total amount of oxides increased greatly. The contents of monoterpene alcohols and ketones also increased. The total amount of monoterpene hydrocarbons was unchanged up to 30°C. However, the contents of myrcene, γ-terpinene, and terpinolene decreased, while there was a considerable increase in *p*-cymene. Among the sesquiterpene hydrocarbons, considerable increases in the (−)-cedrene, γ-elemene, and α-humullene contents were noted in samples stored at 30°C, and the *trans*-β-farnesene content decreased during storage. The total content of monoterpene alcohols was increased slightly. The content of *trans*-carveol increased during storage. *p*-Cymene, *trans*-carveol, isopiperitone, and limonene oxide contents increased, while γ-terpinene, terpinolene, and citronellal contents decreased significantly during storage. The changes of these compounds during storage can serve as a quality index for *hyuganatsu* essential oil.

Key words: *Citrus tamurana* Hort. ex Tanaka; cold-pressed oil; storage period; storage temperature; compositional change

Quantitative analyses of cold-pressed citrus oils are becoming increasingly important, particularly in relation to quality control. The flavor quality of cold-pressed peel oils of citrus fruits depends on a number of factors including degree of maturity,[1] postharvest treatment,[2] storage conditions,[3,4] and hybridization.[5] The economic importance of citrus oils and their usefulness in the flavor and fragrance industries necessitate the acquisition of accurate compositional data. Citrus oils are subject to oxidative changes in the presence of air and light, and during storage.[6] These changes usually have adverse effects on flavor quality. Most flavoring materials are to some extent thermolabile or heat-sensitive. The degree of change is usually a function of both temperature and time. Less stable compounds may also change due to chemical interactions with other constituents.[6]

The composition of the essential oil of *Citrus tamurana* Hort. ex Tanaka (*hyuganatsu*) has been quantitatively and qualitatively measured.[7] In this paper, we report the effects of storage conditions on the composition of *hyuganatsu* essential oil. *Hyuganatsu* samples in maturity were obtained from the Kochi Prefectural Fruit Tree Experimental Station, Kochi, Japan. The peel oil was extracted within 24 h after harvest by the cold-pressing method described by Sawamura and Kuriyama.[8] *Hyuganatsu* peel oil samples (0.5 g) were weighed and placed in 5-ml screw-capped amber glass bottles. Four different conditions were adopted for the storage test of *hyuganatsu* peel oils, where each test included duplicate samples. The results were expressed as peak area percentage and as the average of duplicate runs. The samples were kept at −21, 5, 20, and 30°C, respectively. The qualitative composition of the samples was analyzed upon storage for 1, 3, 6, and 9 weeks at each temperature compared to that of fresh oil. GC, GC/MS, and identification methods of components in this study were the same as described in a previous report.[7]

In our previous work[7] we reported the composition of *hyuganatsu* essential oil. Limonene, γ-terpinene, myrcene, linalol, and α-pinene were the most abundant components in *hyuganatsu* oil. The contents of *trans*-β-farnesene and *l*-carvone in *hyuganatsu* oil were higher than in oils of other citrus fruits. Chemical changes in volatile compounds are known to occur due to light, oxygen, moisture, heat, and storage period, resulting in the formation of artifacts or polymerization.[9] In this study, the relative peak area percentages of 91 components in *hyuganatsu* oil were analyzed after storage at four temperature levels. The results are shown in Table 1.

Monoterpene hydrocarbons were predominant in

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Table 1. Changes of Relative Concentrations of Volatile Compounds in the Cold-pressed Oils of *Hyuganatsu* During Storage

Compounds	Peak area percent																
	Fresh oil	−21°C				5°C				20°C				30°C			
		1*	3	6	9	1	3	6	9	1	3	6	9	1	3	6	9
hydrocarbons																	
aliphatic hydrocarbons																	
undecane	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—	tr	tr	tr	—
tridecane	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	tr	—
pentadecane	0.05	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.03	0.03	0.03	0.04	0.03	0.03	0.02
octadecane	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.02	0.01	0.01	0.03	0.04
total	0.06	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.04	0.05	0.06	0.06	0.04	0.06	0.06
monoterpene hydrocarbons																	
α-pinene	1.04	1.04	1.04	1.02	1.00	1.05	1.03	1.01	1.00	1.07	1.04	1.02	1.02	1.07	1.06	1.11	1.14
α-fenchene	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—
camphene	tr	0.01	tr	tr	tr	0.01	tr	tr	tr	0.01	0.01	tr	0.01	0.01	0.01	tr	0.01
β-pinene	0.50	0.50	0.50	0.49	0.48	0.50	0.50	0.48	0.48	0.51	0.50	0.49	0.49	0.50	0.49	0.49	0.48
(+)-sabinene	0.14	0.14	0.14	0.14	0.13	0.15	0.14	0.13	0.13	0.14	0.14	0.13	0.13	0.14	0.14	0.13	0.12
δ-3-carene	0.31	0.29	0.26	0.28	0.34	0.21	0.31	0.31	0.37	0.37	0.34	0.34	0.36	0.39	0.38	0.37	0.33
myrcene	1.84	1.87	1.92	1.86	1.74	2.08	1.84	1.79	1.73	1.79	1.75	1.62	1.43	1.53	1.32	1.05	0.71
α-phellandrene	0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.09	0.04	0.01	tr	0.01	tr	—	—
α-terpinene	0.11	0.10	0.11	0.11	0.11	0.09	0.11	0.11	0.11	0.11	0.03	tr	—	0.01	—	—	—
limonene	84.78	85.32	85.34	84.28	84.94	85.72	85.24	83.30	85.20	85.06	86.02	85.99	85.16	85.63	85.71	85.45	84.94
γ-terpinene	6.48	6.46	6.47	6.36	6.28	6.49	6.43	6.27	6.27	6.57	4.32	2.16	0.01	4.36	2.67	1.00	0.01
p-cymene	0.03	0.08	0.10	0.11	0.09	0.09	0.10	0.12	0.14	0.16	2.15	4.05	5.46	2.09	3.53	5.21	5.93
terpinolene	0.33	0.33	0.34	0.32	0.32	0.34	0.33	0.31	0.31	0.33	0.23	0.11	0.05	0.18	0.11	0.04	0.01
Total	95.64	96.22	96.30	95.05	95.51	96.81	96.11	93.91	95.82	96.21	96.57	95.92	94.12	95.92	95.42	94.85	93.68
sesquiterpene hydrocarbons																	
β-ylangene	0.01	0.01	tr	tr	tr	0.01	0.01	tr	0.01	0.01	0.01	0.01	—	0.01	0.01	tr	—
(−)-α-copaene	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr	tr	tr
β-cubebene	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.01	0.02	0.01	0.01	0.02	0.01	0.01	0.01
(−)-α-cedrene	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.04	0.06	0.02	0.04	0.06	0.08
β-elemene	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	tr	0.01	0.01	0.01	tr
β-caryophyllene	0.07	0.06	0.06	0.07	0.06	0.07	0.06	0.06	0.06	0.06	0.06	0.04	0.03	0.06	0.04	0.03	0.02
γ-elemene	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.04	0.10	0.03	0.07	0.16	0.29
trans-β-farnesene	0.58	0.56	0.55	0.55	0.58	0.53	0.58	0.58	0.56	0.52	0.53	0.43	0.45	0.51	0.39	0.36	0.23
α-humullene	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.05	0.10	0.04	0.05	0.12	0.23
germacrene D	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.06	0.06	0.04	0.01	tr	0.02	0.01	tr	—
valencene	tr	tr	0.01	tr	tr	tr	tr	tr	tr	tr	tr	0.01	—	tr	tr	0.01	0.01
sesquiphellendrene	0.03	0.04	0.03	0.03	0.03	0.04	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.03	0.02	0.02	0.01
total	0.82	0.80	0.78	0.78	0.80	0.78	0.81	0.80	0.78	0.73	0.75	0.66	0.77	0.76	0.65	0.78	0.88
aldehydes																	
aliphatic aldehydes																	
decanal	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr
trans-2-decenal	0.02	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr	0.01	0.02	—
dodecanal	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	0.01	tr	0.01	tr
trans-2-undecenal	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.03	0.02	0.02	0.04
trans-2-dodecenal	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
tetradecanal	tr	0.01	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr	tr	tr
tetradecenal	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr	0.02	tr	tr	0.03	0.03
total	0.07	0.06	0.06	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.06	0.05	0.07	0.07	0.05	0.10	0.09
terpene aldehydes																	
citronellal	0.22	0.21	0.22	0.21	0.21	0.21	0.21	0.21	0.21	0.19	0.15	0.10	0.08	0.15	0.10	0.07	0.05
neral	tr	0.01	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.01	0.04	0.01	0.02	0.04	0.04
perillaldehyde	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.02	0.03
total	0.23	0.23	0.23	0.22	0.22	0.22	0.22	0.22	0.22	0.20	0.17	0.12	0.14	0.17	0.13	0.13	0.12
alcohols																	
aliphatic																	
octanol	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.03	0.02	0.04	0.03	0.02	0.02
trans-dodec-2-enol	tr	tr	tr	tr	tr	tr	0.01	tr	tr	tr	tr	0.01	0.01	0.01	0.01	0.01	0.01
total	0.04	0.04	0.04	0.04	0.04	0.04	0.05	0.04	0.04	0.04	0.04	0.04	0.03	0.05	0.04	0.03	0.03
monoterpene alcohols																	
linalol	0.80	0.79	0.78	0.78	0.78	0.81	0.79	0.78	0.78	0.77	0.78	0.72	0.71	0.78	0.71	0.70	0.65
terpinen-4-ol	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.02	0.01	0.01	0.02	0.01
l-menthol	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.01	0.01	tr	0.01
α-terpineol	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.09	0.10	0.09	0.09	0.10	0.09	0.10	0.10

Table 1. Continued

Compounds	Fresh oil	Peak area percent															
		−21°C				5°C				20°C				30°C			
		1*	3	6	9	1	3	6	9	1	3	6	9	1	3	6	9
citronellol	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.03	0.03	0.02	0.03	0.03
nerol	0.03	0.03	0.04	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.04	0.04	0.04	0.03	0.04	0.06
p-mentha-1-en-9-ol	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.01	0.01	0.01	0.01	0.01	0.01
cinnamyl alcohol	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.02	tr	tr	tr	tr
trans-carveol	0.01	tr	tr	tr	tr	tr	0.01	tr	tr	tr	0.02	0.06	0.13	0.02	0.05	0.07	0.22
isoeugenol	tr	tr	tr	0.01	tr	tr	tr	0.01	tr	0.01	0.01	0.01	0.02	0.01	0.01	0.02	0.03
p-mentha-1,8-dien-10-ol	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—	0.01	tr	tr	—
limonene-diol	0.01	0.01	tr	0.01	0.01	tr	0.01	tr	tr	0.01	tr	0.01	tr	0.01	tr	0.01	tr
total	0.98	0.96	095	0.96	0.95	0.97	0.97	0.95	0.94	0.94	0.98	0.99	1.08	1.03	0.94	1.00	1.12
sesquiterpene alcohols																	
cis-nerolidol	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.04	0.03	0.03	0.04	0.05
trans-nerolidol	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.03	0.03	0.04	0.03	0.03	0.04	0.03	0.04	0.03
globulol	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.04	0.03	tr	0.01	0.02	0.01	0.01	tr
elemol	tr	tr	0.01	tr	tr	tr	tr	tr	0.01	tr	0.01	tr	tr	tr	tr	tr	—
cedrol	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	tr
spathulenol	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
(−)-α-bisabolol	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr	0.01	0.01
β-eudesmol	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	0.01	0.02	tr	tr	0.02	0.03
cis, *trans*-farnesol	tr	tr	tr	tr	tr	0.01	tr	tr	0.01	tr	tr	0.01	tr	tr	tr	0.01	tr
trans, *trans*-farnesol	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	0.01	0.01	tr	0.01	tr
total	0.16	0.16	0.17	0.16	0.16	0.17	0.16	0.16	0.17	0.14	0.14	0.10	0.13	0.13	0.09	0.15	0.13
ketones																	
α-thujone	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr	0.01	tr	0.01
β-thujone	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—	tr	tr	tr	—
menthone	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.01	0.02	0.02	0.01	0.01
δ-camphor	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.01	0.01	0.01
l-carvone	0.34	0.34	0.33	0.33	0.33	0.34	0.33	0.33	0.32	0.30	0.19	0.13	0.22	0.16	0.14	0.25	0.44
isopiperitone	tr	tr	0.01	tr	0.01	tr	tr	tr	0.01	tr	0.02	0.06	0.16	0.03	0.06	0.18	0.32
total	0.36	0.36	0.36	0.35	0.36	0.36	0.35	0.35	0.35	0.32	0.23	0.21	0.40	0.22	0.24	0.45	0.79
esters																	
ethyl acetate	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.01	0.01
2-methylbutyl butyrate	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	tr	0.02	0.02	tr	—
linalyl acetate	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.03	0.06	0.09	0.05	0.07	0.10	0.12
nonyl acetate	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
bornyl acetate	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.06	0.07	0.06	0.07	0.08	0.06	0.07	0.06
citronellyl formate	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.01
decyl acetate	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	tr	0.02
terpinyl acetate	0.01	0.01	0.01	tr	0.01	0.01	tr	0.01	0.01	0.01	0.01	tr	tr	tr	tr	0.03	—
neryl acetate	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03
geranyl acetate	0.04	0.04	0.03	0.04	0.04	0.04	0.03	0.03	0.03	0.03	0.03	0.04	0.07	0.04	0.04	0.07	0.10
geranyl propionate	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—
trans, *trans*-farnesyl acetate	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.01	0.01	0.02	0.01	0.01	tr
total	0.23	0.22	0.21	0.21	0.22	0.22	0.20	0.21	0.21	0.21	0.23	0.23	0.30	0.27	0.27	0.35	0.36
oxides																	
cis-linalol furanoxide	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.02	0.01	0.01	0.02	0.02
(+)-*cis*-limonene oxide	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.07	0.08	0.08	0.10	0.09	0.16	0.17
(+)-*trans*-limonene oxide	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.03	0.15	0.24	0.06	0.11	0.16	0.18
trans-linalol furanoxide	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.01	0.02	0.01	0.01	0.01	0.02
cis-linalol pyranoxide	tr	tr	0.01	tr	tr	tr	tr	0.01	tr	tr	0.01	tr	tr	tr	tr	tr	tr
caryophyllene oxide	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	0.02	0.03	0.01	0.01	0.03	0.03
nerol oxide	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—
total	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.13	0.27	0.39	0.19	0.23	0.38	0.42
acids																	
octanoic acid	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—
nonanoic acid	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr
undecanoic acid	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	—
total	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	tr	0.01	tr
total volatiles (%)	98.60	99.11	99.17	97.88	98.37	99.68	98.98	96.76	98.64	98.90	99.34	98.64	97.49	98.87	98.10	98.29	98.68

* Week.

hyuganatsu essential oil and accounted for 95.64% of the fresh oil. The total amount of monoterpene was not influenced by storage temperatures up to 30°C. The contents of myrcene, γ-terpinene, and terpinolene decreased with storage duration and temperature increase; the reverse was true for *p*-cymene. Njoroge *et al.*[3] has suggested that the increase of *p*-cymene during storage could occur by rearrangement, hydrogenation, or dehydrogenation of α- and γ-terpinenes and limonene. When the essential oil of *hyuganatsu* was stored at 5°C, the effect of temperature on the terpinolene content was minimal, but at 20 and 30°C conditions the amount of terpinolene decreased significantly as a function of the storage period, as shown in Table 1. The limonene content was not much changed during storage. The acyclic monoterpene myrcene can be formed from the chemical modification of either geranyl pyrophosphate or neryl pyrophosphate.[10] Dieckmann and Palamand[11] suggested the possibility of myrcene cyclizing to α- and γ-terpinenes and limonene, however, this was not observed in this study.

The content of sesquiterpene hydrocarbons did not change much during storage at -21, 5, 20, and 30°C. However, the contents of $(-)$-α-cedrene, γ-elemene, and α-humullene increased considerably at 30°C. The content of *trans*-β-farnesene in *hyuganatsu* oil was higher than in oils of other citrus fruits. A large reduction in the *trans*-β-farnesene content was noted in samples stored at 30°C. Changes also took place in the aldehyde contents, but these were not prominent. Citronellal was the major terpene aldehyde and underwent considerable change during storage. The content of this aldehyde decreased to about one-fifth after 9 weeks storage at 30°C.

The changes in the monoterpene alcohols could be related to degradations of some monoterpene hydrocarbons. As shown in Table 1, the total content of monoterpene alcohols increased slightly. The content of linalol decreased during storage, while an increase occurred in *trans*-carveol. It is interesting that the content of octanol, which was investigated as an odor-active compound of *hyuganatsu* flavor,[12] was nearly constant. Linalol was also investigated as an odor-active volatile of *hyuganatsu*.[12] The content of linalol was considerable in fresh cold-pressed oil (CPO) of *hyuganatsu* but gradually declined during storage. The measurable amounts of *cis*- and *trans*-linalol oxides in the *hyuganatsu* CPO upon storage might account for the decreased linalol content observed in this study. The content of globulol decreased and it may have been decomposed by oxidative reactions and polymerization. Tressl *et al.*[13] suggested that globulol may arise via the biogenetic transformation of germacrene D, the content of which also decreased during storage.

Isopiperitone, a rarely reported component of *Citrus* oil, was detected in the CPO of *hyuganatsu* at a level of less than 0.01%. The content of isopiperitone increased considerably during storage. The content of esters remained nearly constant. Only the contents of linalyl and geranyl acetates increased significantly. Most oxides, except *cis*-linalol pyranoxide and nerol oxide, increased considerably during storage of *hyuganatsu* peel oil. The trace amounts of *cis*- and *trans*-limonene oxides, and *cis*- and *trans*-linalol furanoxides were initially present in *hyuganatsu* oil but gradually increased during storage. When the oxide amounts of limonene and linalol were compared, *cis*- and *trans*-limonene oxides showed significantly higher concentration than those of *cis*- and *trans*-linalol oxides in *hyuganatsu* oil.

Quantitative analyses of cold-pressed citrus oils are becoming increasingly important, particularly in relation to quality control. A temperature rise may have caused the polymerization. The flavor of *hyuganatsu* was easily changed by storage conditions. Compositional change in the volatile profile, resulting from storage conditions, caused alteration in *hyuganatsu* flavor. *p*-Cymene, *trans*-carveol, isopiperitone, and limonene oxide contents increased siginificantly, and γ-terpinene, terpinolene, and citronellal contents decreased significantly during storage. The changes of these compounds during storage may be used as a quality index for *hyuganatsu* essential oil.

Some high molecular weight nonvolatile polymers or artifacts could also be generated. However, this study was focused only on the compositional change of volatile compounds of *hyuganatsu* oil during storage. Further research is necessary to determine the formation of new compounds during the storage of *hyuganatsu* oil.

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