

## DIFFERENTIATION OF TEAS BY THE VARIATIONS OF LINALOOL AND GERANIOL CONTENTS

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**ABSTRACT.** The linalools and geraniols contents of clonal black teas varied widely due to time from last pruning, altitude, withering and maceration method. The ratio of linalools to linalools plus geraniols (Terpene Index) was specific for the different clones. Similar observations were also made on tea from mixed clones manufactured in one batch due to driers and grading. Thus terpene index can also be used to classify tea made from one batch.

### INTRODUCTION

Many clones of tea (*Camellia sinensis* (L.) O. Kuntze) have been developed in efforts to improve both quality and yield of tea. Although some of the clones look similar, at present only visual characteristics are used to identify the clones. Takeo (1,2) suggested a chemotaxonomic method of classifying the tea clones based on the ratio of the gas chromatographic peak areas due to sum of linalools (i.e. linalool oxides and linalool) to sum of linalools plus geraniols (i.e. geraniol plus (E)-geranic acid). This ratio is called terpene index (1,2).

Many terpenes have been identified in tea, however the terpenes are dominated by linalool and geraniol. Although Selvendran et. al. (3) suggested that linalool is produced from an oxygenated isoprenoid hydrocarbon, Takeo (1,2,4) showed that linalool and geraniol are products of hydrolysis of terpene-glycosides. Indeed, a linalool glycoside was recently isolated from fresh tea shoots (15). Hydrolysis of terpene-glycoside is initiated by glycosidase (4) and occurs mainly during the withering (3,6,7,8), maceration (3,8,10) and fermentation (3,8,9,10) stages of tea manufacture.

Recently, (11) it was shown that the sums of linalools and geraniols varied with the method of withering, rates of nitrogenous fertilizer application; geographical area of production and plucking standard. However, the terpene index (i.e. ratio of peak areas due to linalools to those of linalools and geraniols) was constant and only varied due to different plucking standards. The index has been subjected to further tests and the results obtained are presented in this report.

### EXPERIMENTAL

Tea used to determine the effects of different withering (11) and different maceration methods (12) on the linalools and geraniols composition of tea were obtained from the Tea Research Foundation Clonal Field Trials, while teas to determine the effect of time from last pruning (13) and altitude were obtained from commercial estates of the African Highlands Produce Company, Kericho.

Each treatment used 1200 g fresh leaf. Plucking conformed to commercial standard (11).

Effects of driers and grading on the linalools and geraniols composition were determined in Commercial Kenya Tea Development Authority Kapkoros Factory (14). Details of the manufacture have been published elsewhere (7,12,13,14).

Extraction of terpene mixture was by the simultaneous distillation and extraction (SDE) method (12-16) and gas chromatographic analyses were done as described by Barua et. al. (17).

## RESULTS AND DISCUSSION

Tea quality and tea chemical composition has been shown to vary with time from last pruning (13). In clone S 15/10, the sum of GC peaks due to linalools and geraniols showed wide variations due to time from last prune, with coefficient of variations of 14.8 and 12.9%, respectively (Table 1). However, the ratio of peak areas due to the sum of linalools plus geraniols (terpene index) (TI) showed little variations; 1.9%. Thus, TI of clonal tea does not change due to time from last prune. Earlier TI of clone S 15/10 was found to be 0.32 (11,18) however in the present study a value of 0.26 was determined. This difference is due to improved plucking standard of tea in the study compared to the earlier study.

Table 1. Effect of pruning time on TI of clone S 15/10.

Period since last pruning	Linalools	Geraniols	Terpene Index
37 months	1.86	5.29	0.26
23 months	1.56	4.50	0.26
12 months	1.47	4.28	0.26
3 months	1.31	3.93	0.25
Mean	1.55	4.50	0.26
C.V. % <sup>a</sup>	14.8	12.9	1.9

<sup>a</sup> as standard deviation x mean/100

In the previous study (11), it was shown that although geographical area of tea production caused large variations in sum of linalools and geraniols in teas, TI was not affected. The variations in the chemical composition noted in the early study was difficult to interpret as the teas come from different sources and were of undetermined period since last prune. Using clones 6/8, S 15/10 and TN 14-3 grown at different altitudes in Kericho and in same period since last prune, it has been shown that the sum of linalools and geraniols show large variations (with i.e. coefficient of variations between 9.8 and 17.2%) due to changes in altitude (Table 2). TI however, did not change with altitude for the three clones studied.

Since last study (11) there have been attempts to make withering more efficient and to use the available withering space more economically. To achieve this, two stage withering systems are being introduced in tea manufacture (19). The use of the different withering techniques were recently shown to cause changes in the chemical composition and quality of teas (19). The changes in linalools and geraniols levels due to the different withering methods are presented in Table 3. Indeed there were large changes in the amounts of those chemicals due to different withering methods. However, the TI did not change as a result

Table 2. Effects of Altitude on Terpene Index

Source	Altitude (meters above sea level)	Clone								
		6/8			S.15/10			TN 14/3		
		Linalools	Geraniols	Terpene Index	Linalools	Geraniols	Terpene Index	Linalools	Geraniols	Terpene Index
Tea Research Foundation	2180	1.55	1.89	0.45	1.55	4.67	0.25	1.90	0.21	0.90
Cheppoiben	2120	1.21	1.42	0.46	1.67	5.28	0.24	1.68	0.17	0.91
Cheptabas	1940	1.58	1.84	0.46	1.34	3.72	0.26	1.82	0.16	0.91
Kapropret	1860	-	-	-	1.33	3.79	0.26	1.99	0.23	0.90
Mean		1.45	1.72	0.46	1.47	4.37	0.25	1.80	0.19	0.91
C.V.% <sup>a</sup>		14.2	15.0	1.3	11.3	17.1	3.8	9.8	17.2	0.6

<sup>a</sup> as in Table 1

Table 3. Variations of linalools, geraniols and terpene index with withering of clone 6/8.

Withering method	Linalools	Geraniols	Terpene Index
1. Immediate maceration without moisture loss (no wither)	2.10	2.49	0.46
2. Immediate warm air moisture loss then maceration	2.29	2.71	0.46
3. Immediate cold air moisture loss then maceration	2.37	2.73	0.46
4. Normal withering	2.01	2.23	0.47
5. Storage with no moisture loss then maceration	1.98	2.27	0.47
6. Storage with moisture loss - warm air moisture loss - maceration	2.42	2.85	0.46
7. Storage with no moisture loss - cold air moisture loss - maceration	2.07	2.57	0.45
8. Immediate warm air moisture loss - storage - maceration	2.34	2.80	0.46
9. Immediate cold air moisture loss - storage - maceration	2.49	3.08	0.45
Mean of withering methods	2.23	2.64	0.46
C.V.% <sup>a</sup>	8.6	10.5	1.6

<sup>a</sup> as in Table 1

of withering.

Methods of maceration normally cause changes in the chemical composition of teas (10,12). Comparing the effects of ("Crush, Tear and Curl") CTC and Orthodox maceration, it was noted that generally the Orthodox maceration

Table 4. Manufacturing effects on Terpene Index

Manufacture	Terpenes	Clone 6/8	Clone 31/8	Clone S 15/10
CTC	Linalools	1.73	2.73	2.47
	Geraniols	2.00	0.23	7.09
Terpene	Linalools	0.46	0.92	0.26
	Geraniols	4.20	4.43	2.77
Orthodox	Geraniols	4.69	0.38	7.88
Terpene Index		0.47	0.92	0.26
Mean Terpene Index		0.47	0.92	0.26

Table 5. Drier and grading effect on terpene Index

Drier	Grade	Linalools	Geraniols	Terpene Index
Conventional Drier	BP 1	4.44	2.01	0.69
	PF 1	3.92	1.68	0.70
	PD	2.93	1.30	0.69
	D 1	2.56	1.14	0.69
Mean CD		3.46	1.53	0.69
C.V.%		25.1	25.5	50.7
Fluid Bed Drier (FBD)	BP 1	4.43	1.99	0.69
	PF 1	4.37	1.81	0.71
	PD	3.64	1.55	0.70
	D 1	2.66	1.17	0.69
Mean FBD		3.78	1.63	0.70
C.V.%		21.9	21.8	1.4
Mean Grades	BP 1	4.44	2.00	0.69
	PF 1	4.15	1.75	0.7
	PD	3.29	1.43	0.70
	D 1	2.61	1.16	0.69
C.V.%		23.0	23.2	0.8

produced teas with higher amounts of the volatile flavour compounds than CTC maceration (10,12). However, CTC maceration produced teas with higher amounts of theaflavins and thearubigins than Orthodox maceration (12). In the present study, using clones 6/8, 31/8 and S 15/10 CTC manufacture produced less amounts of linalools and geraniols than Orthodox manufacture. However, TI for the clones were specific and did not change due to the mode of maceration (Table 4).

Driers and grading of tea were recently shown to cause variation in the chemical composition of teas (14). Using mixed teas, it has been demonstrated (Table 5) that both driers and grading of teas cause changes in the amounts of linalools and geraniols in tea. However, TI does not change due to either driers or grading. Thus apart from using TI to classify clones (11), it can additionally be used to identify teas from the same batch.

Although *Assamica* and *sinensis* varieties of tea from North-East India and China, respectively were introduced in Kenya in the 1920s, most of the tea planted at present is the *Assamica* variety. This is due to the relatively larger leaf size of *Assamica* variety compared to *sinensis* variety thus making harvesting (plucking) easier. However, several inter-breedings have been done in Kenyan

clonal teas and although some clones may have morphological features of *Assamica* variety their chemical constitution resemble the *sinensis* variety. Takeo (1,2,20) showed that the *Assamica* varieties of tea have higher amounts of linalools than geraniols while the *sinensis* varieties show the reverse effect. Clone 6/8 and S 15/10 used in this study (Table 1 to 3) were therefore noted to have TI closer to that of the *sinensis* varieties than *Assamica* varieties. Their morphological features are however similar to other *Assamica* varieties. Thus although TI is unique to a particular clonal tea, where several inter-breedings between *Assamica* and *sinensis* varieties of tea have been done, the index cannot be used to classify teas into varieties.

Additionally, use of TI as a chemotaxonomic criterion however, has some problems. Earlier it was demonstrated that plucking standards affect TI (11). Although this can be profitably used to measure plucking standards, it implies that to use it to identify made teas as being from a particular clone, the plucking standard must be known or be constant. Secondly, TI can only have values between 0 and 1; but there are many clones. This implies that some clones have same TI and therefore other additional parameters must be used to identify them.

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