

# Comparison of Gas Chromatographic Volatile Profiling Methods for Assessing the Flavour Quality of Kenyan Black Teas

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**Abstract:** A comparison is made of the various ratios used to quantify black tea aroma such as: ratio of sum of gas chromatographic peak areas of compounds eluting before linalool (3,7-dimethyl-1,6-Octadien-3-ol) to those eluting after linalool (Wickremasinghe–Yamanishi ratio); gas chromatographic peak areas of compounds imparting sweet flowery aroma (Group II volatile flavour compounds) to those imparting green grassy aroma (Group I volatile flavour compounds), Owuor's flavour index; ratio of the sum of gas chromatographic peak area of the terpenoids to non-terpenoids, i.e. Mahanta ratio; and gas chromatographic peak area of linalool to E-2-hexenal, i.e. Yamanishi–Botheju ratio.

The Wickremasinghe–Yamanishi ratio and Owuor's flavour index showed a significant relationship, the Mahanta ratio showed a significant but poor relationship, and the Yamanishi–Botheju ratio had no relationship with the tasters' evaluation of Kenyan clonal CTC black teas. Similar results were obtained from orthodox black teas manufactured from different varieties.

**Key words:** black tea, *Camellia sinensis*, aroma, flavour indices.

## INTRODUCTION

The volatile flavour compounds (VFC) of black tea made from the young tender shoots of *Camellia sinensis* (L) O Kuntze are known to contribute towards the quality of the beverage. Several studies have reported various compositions of the VFC or aroma complex, and over 600 individual components have been reported (Robinson and Owuor 1991). Although many of these studies have reported the composition of the VFC in black tea without indicating how the varying levels of the compounds affect its quality (Yamanishi *et al* 1966, 1972; Bondarovich *et al* 1967; Renold *et al* 1974; Aisaka *et al* 1978; Cloughley *et al* 1982; Takeo 1983a, 1984; Takeo and Mahanta 1983; Mick *et al* 1984; Horita and Owuor 1987) it has been recognised that some of these compounds contribute negatively whereas others contribute positively towards black tea flavour. Thus the VFC composition reported in these studies has not helped to quantify how quality of the black teas changed with the varying levels of the VFC. Indeed no re-

lationship has been established between the total VFC and quality *per se*.

In an attempt to overcome this problem, several quantitative methods have been examined (Wickremasinghe *et al* 1973; Yamanishi *et al* 1978, 1989; Baruah *et al* 1986; Owuor *et al* 1986, 1988; Mahanta *et al* 1988). These studies have used ratios of gas chromatographic peak areas of compounds perceived to be beneficial to black tea quality to those which are thought to lower the quality of black tea or vice versa. Wickremasinghe *et al* (1973) and Yamanishi *et al* (1978) used the ratio of the sum of the gas chromatographic peak areas of compounds eluting before linalool to the sum of areas of the linalool plus all compounds with gas chromatographic retention times longer than linalool. In this ratio, henceforth referred to as the Wickremasinghe–Yamanishi ratio, it was assumed that compounds with retention times shorter than linalool were deleterious to quality whereas linalool and the VFC, with longer retention times than linalool, were desirable for black tea quality. Thus, the smaller the ratio, the better the quality.

Later, the Wickremasinghe–Yamanishi ratio was improved by first classifying the compounds using the odour characteristics of the individual VFC reported in the literature (Owuor *et al* 1986 and references therein). Where smell characteristics of the compounds were not available in the literature, the Wickremasinghe *et al* (1973) and Yamanishi *et al* (1978) classification above was used. The compounds imparting green grassy smell to black tea were classified as Group I VFC while those with desirable sweet flowery aroma were classified as Group II VFC. A ratio of the desirable to undesirable VFC, worked as Group II/Group I VFC (henceforth referred to as Owuor's Flavour Index (FI)), was then developed (Owuor *et al* 1986, 1988). FI measures directly and linearly the aroma quality, thus the larger the ratio the better the aroma quality. Again linalool oxides (*cis* (*cis*-2-methyl-2-vinyl-5-hydroxy isopropyl tetrahydrofuran) and *trans* (*trans*-2-methyl-2-vinyl-5-hydroxy isopropyl tetrahydrofuran) furanoid) and benzaldehyde, which have desirable aroma (Yamanishi *et al* 1968), were classified into the correct group. Apart from these compounds, the rest of the classification of the compounds remained the same as those of Wickremasinghe *et al* (1973) and Yamanishi *et al* (1978).

At the same time a group of flavour chemists working at Tocklai in India also developed a ratio based on the sum of gas chromatographic peak areas of terpenoids to non-terpenoids (Baruah *et al* 1986; Mahanta *et al* 1988). In this ratio, henceforth called the Mahanta ratio, the terpenoids were assumed to be desirable while the non-terpenoids were classified as undesirable to tea quality.

Recently, Yamanishi *et al* (1989) developed another ratio based on gas chromatographic peak areas of linalool and E-2-hexenal, which ignored all the other VFC. This ratio (the Yamanishi–Botheju ratio) was shown to have a relationship with prices at the auction of some Sri Lankan orthodox black teas.

The ratios developed seem to have some unique attributes. In this study a comparison is made of the relationship between the ratios and sensory evaluation scores of Kenyan clonal CTC black teas and orthodox black teas from different varieties. It is hoped that such an assessment may lead to a realisation of the need for identification of a more reliable index for use in black tea aroma research and quality classification.

## EXPERIMENTAL

### Plucking and manufacturing

#### (a) CTC manufacture

The clonal tea used in this study (clones 6/8 and TN 14-3) were obtained from the clonal plots of tea-growing companies in Kenya and the clonal field trials (CFT) of the Tea Research Foundation of Kenya (Timbilil Estate).

The materials were derived from different geographical zones at altitudes of between 1969 and 2260 m above mean sea level. All the teas were obtained within a radius of about 300 km.

The plucking standard conformed to normal practice, i.e. about 90% good leaf comprising mostly two leaves and a bud and a small amount of three leaves and a bud. Samples (1200 g) of the teas were withered for 16 to 20 h until they had 28 to 30% of their original weight.

Manufacturing was done at the Research Foundation of Kenya's factory three times within 3 months for every clone and for all locations. The leaves were manufactured in triplicate by a miniature 'crush, tear and curl' (CTC) method (Owuor *et al* 1989). All the teas were manufactured between 07.00 and 11.00 local time, so that, apart from minimal temperature differences within this period, all manufacturing conditions were similar. The marginal differences in fermentation rates due to changes in temperature during this period were overcome by randomisation. The manufactured teas were subjected to analysis and tasting without sorting.

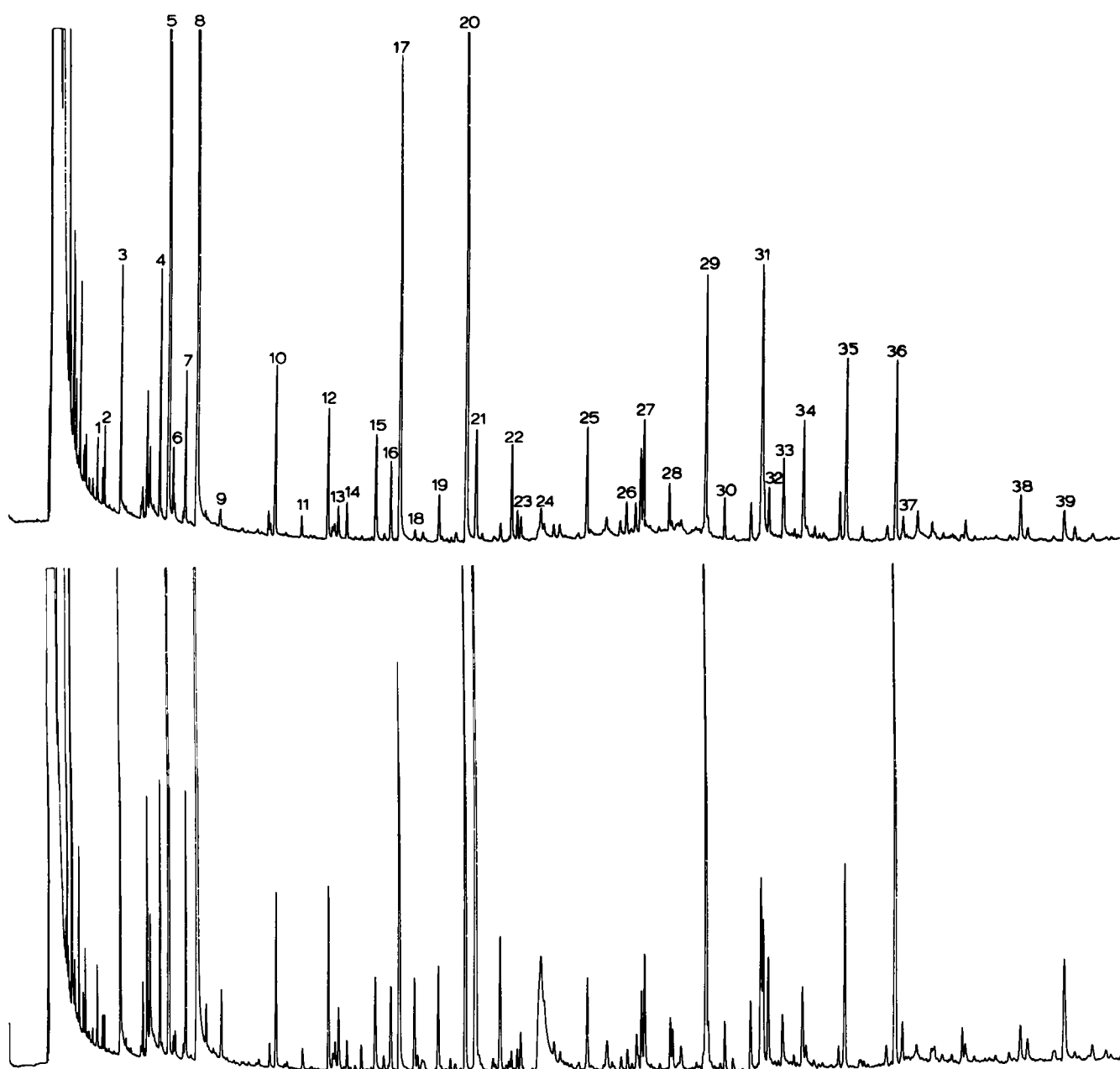
#### (b) Orthodox manufacture

Clones 31/8, 31/11, S15/10 and 6/8 and seedling stock St 18 from *Camellia sinensis* var *assamica*, clone 14/1 and seedling China tea from variety *sinensis* and clones 301/4, 301/5 and 301/6 from variety *assamica lasiocalyx* (Shan tea) were manufactured by miniature orthodox method as outlined earlier (Owuor *et al* 1989). The young tender shoots of the plants were obtained from different CFTs at Timbilil Estate. Plucking standard was similar to that used in the CTC manufacture, but withering was done until the shoots had lost 35–40% of the original weight.

### Analysis of the volatile flavour compounds (VFC)

Simultaneous steam distillation extraction (SDE, water/diethylether) was used to extract the VFC (Likens and Nickerson 1964) as outlined earlier (Owuor *et al* 1986) using cumene or ethyl decanoate as an internal standard.

The dried (anhydrous Na<sub>2</sub>SO<sub>4</sub>) ether mixture was concentrated to about 100 µl and the concentrate was subjected to gas chromatography (GC) and gas chromatography–mass spectrometry (GC–MS) analysis under the conditions of Baruah *et al* (1986). GC was done on a Shimadzu model GC-7A gas chromatograph or Varian 3300 GC equipped with flame ionisation detector, while GC–MS was done on a Hitachi model 663 GC coupled with a Hitachi M.80A and M.003 mass spectrometer. Peak identifications were done by comparing retention times with those of authentic samples and confirmed by GC–MS. The quantities of the compounds are expressed as the ratio of area of peak to that of the internal standard.



**Fig 1.** Gas chromatographic traces of black tea aroma concentrates. 1 = 2-methylbutanal, 2 = pentanal, 3 = hexanal, 4 = 1-penten-3-ol, 5 = cumene (internal standard), 6 = heptanal, 7 = Z-3-hexenal, 8 = E-2-hexenal, 9 = pentanol, 10 = Z-3-Penten-1-ol, 11 = hexanol, 12 = Z-3-hexenol, 13 = *n*-nonanal, 14 = E-2-hexenol, 15 = linalool oxide (*cis* furanoid), 16 = E,Z-2,4-heptadienal, 17 = linalool oxide (*trans* furanoid), 18 = E,E-2,4-heptadienal, 19 = benzaldehyde, 20 = linalool, 21 =  $\alpha$ -cedrene, 22 = 3,7-dimethyl-1,5,7-octatrien-3-ol, 23 =  $\beta$ -cyclocitral, 24 = phenylacetaldehyde, 25 =  $\alpha$ -terpineol, 26 = linalool oxide (*cis* pyranoid), 27 = methyl salicylate, 28 = nerol, 29 = geraniol, 30 = benzyl alcohol, 31 = 2-phenylethanol, 32 = butylated hydroxy toluene (an artifact), 33- $\beta$ -ionone, 34 = E-2-hexenoic acid, 35 = nerolidol, 36 = cedrol, 37 = bovolide, 38 = 6,10,14-trimethyl pentadecan-2-one, 39 = E-geranic acid.

### Tea tasting

Sensory evaluation for the clonal CTC black teas was by a panel of three regular tea tasters, and scores were based on briskness, brightness, infusion, thickness and flavour on a scale of 0 to 10 for each component. For orthodox manufacture, the black teas were tasted by professional tasters from Africa Tea Brokers in Mombasa, and scores were on a scale of 0 to 20 each for briskness, brightness,

infusion, thickness and flavour. The best quality teas were given the highest score.

### RESULTS AND DISCUSSION

A typical gas chromatogram of a tea extract is presented in Fig 1, and it is noted that there are larger amounts of geraniol (2-*trans*-3,7-dimethyl-2,6-octadien-1-ol) and

TABLE 1

Changes in the composition\* of the essential oil and tasters' evaluation of clone 6/8 black CTC due to geographical area of production

Volatile compounds	Location of production							
	Karirana (Limuru)	Timbilil (Kericho)	Sitoi (Nd H) <sup>a</sup>	Kangaita (Mt K) <sup>b</sup>	Kagochi (Mt K)	Changoi (Kericho)	Kaimosi (Kaimosi)	Sotik (Sotik)
Hexanal	0.28	0.40	0.24	0.32	0.16	0.37	0.31	0.40
1-Penten-3-ol	0.10	0.12	0.13	0.29	0.07	0.10	0.11	0.16
Heptanal	0.03	0.02	0.02	0.03	0.02	0.02	0.02	0.03
Z-3-Hexenal	0.08	0.20	0.19	0.22	0.11	0.23	0.18	0.17
E-2-Hexenal	2.17	3.12	2.67	4.15	1.40	2.41	2.42	2.80
Pentanol	0.03	0.03	0.02	0.04	0.02	0.04	0.02	0.05
Z-2-Penten-1-ol	0.09	0.06	0.08	0.24	0.09	0.06	0.09	0.10
Hexanol	0.08	0.06	0.10	0.12	0.09	0.08	0.06	0.14
Z-3-Hexen-1-ol	0.11	0.16	0.18	0.28	0.12	0.15	0.11	0.10
Nonanal	0.06	0.06	0.06	0.07	0.05	0.04	0.07	0.06
E-2-Hexen-1-ol	0.14	0.08	0.14	0.24	0.21	0.08	0.12	0.07
Linalool oxide ( <i>cis</i> furanoid)	0.13	0.08	0.05	0.19	0.08	0.06	0.08	0.07
E,Z-2,4-Heptadienal	0.04	0.03	0.03	0.05	0.03	0.04	0.05	0.07
Linalool oxide ( <i>trans</i> furanoid)	0.40	0.25	0.16	0.53	0.20	0.23	0.23	0.23
E,E-2,4-Heptadienal	0.05	0.03	0.03	0.03	0.03	0.05	0.06	0.11
Benzaldehyde	0.04	0.05	0.06	0.07	0.04	0.04	0.05	0.05
Linalool	1.31	0.91	1.00	1.33	0.67	0.44	0.80	0.88
$\alpha$ -Cedrene	0.41	1.65	0.24	0.06	0.06	1.22	0.41	1.12
3,7-Dimethyl-1,5,7-octatrien-3-ol	0.01	0.24	0.03	0.02	0.03	0.17	0.05	0.15
$\beta$ -Cyclocitral <sup>c</sup>	0.08	0.08	0.04	0.10	0.06	0.03	0.06	0.07
Phenylacetaldehyde	0.91	0.61	0.81	1.16	0.74	0.39	0.73	0.79
$\alpha$ -Terpineol <sup>d</sup>	0.09	0.06	0.06	0.13	0.08	0.04	0.06	0.04
Linalool oxide ( <i>cis</i> pyranoid) <sup>e</sup>	0.08	0.02	0.01	0.03	0.02	0.01	0.01	0.01
Methyl salicylate	0.70	0.69	0.42	0.77	0.61	0.54	0.82	0.75
Nerol <sup>f</sup>	0.09	0.06	0.06	0.12	0.04	0.02	0.06	0.03
Geraniol	2.34	1.15	1.08	2.05	0.66	0.54	0.88	1.06
Geranyl acetone <sup>g</sup>	0.02	0.02	0.01	0.02	Tr	Tr	0.02	0.03
Benzyl alcohol	0.02	0.01	0.02	0.06	0.03	0.01	0.04	0.01
2-Phenylethanol	0.02	Tr	Tr	Tr	Tr	Tr	Tr	Tr
$\beta$ -Ionone <sup>h</sup>	0.05	0.07	0.02	0.06	0.02	0.05	0.04	0.06
Nerolidol	0.11	0.12	0.02	0.08	0.05	0.08	0.15	0.13
Cedrol	0.24	1.23	0.15	0.05	0.03	1.02	0.21	0.66
Bovolide <sup>i</sup>	0.03	0.04	0.02	0.02	0.01	0.03	0.03	0.03
E-Geranic acid	0.58	0.41	0.50	0.64	0.36	0.28	0.22	0.31
Indole	0.01	0.01	Tr	Tr	0.01	0.01	Tr	0.01
Total VFC	10.93	12.13	8.65	13.57	6.2	8.88	8.57	10.75
Tasters' evaluation	39.00	38.63	36.03	33.63	38.63	38.25	37.13	37.88

\* As ratio of the gas chromatographic peak area to that of internal standard.

<sup>a</sup> Nandi Hills area.

<sup>b</sup> Mt Kenya area.

<sup>c</sup> 2,2,6-trimethyl-6-cyclohexen-1-carboxaldehyde.

<sup>d</sup> 1-methyl-4-isopropenyl cyclohexan-1-ol.

<sup>e</sup> Tetrahydro-2,2,6-trimethyl-6-vinyl propan-3-ol.

<sup>f</sup> 2-*cis*-3,7-dimethyl-2,6-octadien-1-ol.

<sup>g</sup> 2,6-dimethyl undeca-2,6-dien-10-one.

<sup>h</sup> 4(2,6,6-trimethyl-1-cyclohexenyl)-3-buten-2-one.

<sup>i</sup> 2,3-dimethyl-4-keto-2-nonenoic acid, enol-lactone.

E-geranic (3,7-dimethyl, 2*E*, 6-octadienoic) acid in the *sinensis* variety black tea (Fig 1b) than in the *assamica* variety black tea (Fig 1a).

Clonal black teas prepared from green teas grown in different locations of Kenya (Owuor *et al* 1988) were used in this study. The VFC data are presented in Tables

TABLE 2

Changes in the composition\* of the essential oil and tasters' evaluation of clone TN 14-3 black CTC due to geographical area of production

Volatile compounds	Location of production							
	Karirana (Limuru)	Timbilil (Kericho)	Sitoi (Nd H) <sup>a</sup>	Kangaita (Mt K) <sup>b</sup>	Kagochi (Mt K)	Changoi (Kericho)	Kaimosi (Kaimosi)	Sotik (Sotik)
n-Hexanal	0.51	0.40	0.40	0.45	0.56	0.41	0.44	0.41
1-Penten-3-ol	0.22	0.19	0.29	0.25	0.25	0.09	0.17	0.24
Heptanal	0.08	0.04	0.05	0.09	0.06	0.02	0.04	0.03
Z-3-Hexenal	0.30	0.19	0.25	0.22	0.38	0.19	0.17	0.22
E-2-Hexenal	3.16	2.98	4.92	3.37	3.66	2.15	2.22	3.24
Pentanol	0.04	0.04	0.02	0.02	0.04	0.05	0.04	0.06
Z-2-Pentenol	0.17	0.13	0.23	0.20	0.21	0.08	0.14	0.19
n-Hexanol	0.07	0.08	0.07	0.06	0.06	0.02	0.05	0.06
Z-3-Hexenol	0.17	0.29	0.30	0.14	0.15	0.17	0.12	0.24
Nonanal	0.13	0.10	0.15	0.18	0.14	0.08	0.17	0.10
E-2-Hexenol	0.07	0.07	0.07	0.09	0.07	0.03	0.06	0.06
Linalool oxide (cis furanoid)	0.17	0.09	0.11	0.11	0.12	0.08	0.11	0.12
E,Z-2,4-Heptadienal	0.05	0.03	0.05	0.05	0.07	0.04	0.05	0.08
Linalool oxide (trans furanoid)	0.68	0.41	0.44	0.43	0.44	0.39	0.43	0.57
E,E-2,4-Heptadienal	0.07	0.03	0.06	0.05	0.05	0.04	0.03	0.09
Benzaldehyde	0.09	0.07	0.09	0.07	0.11	0.07	0.11	0.09
Linalool	1.48	1.03	1.61	1.60	1.10	0.69	0.86	1.13
$\alpha$ -Cedrene	0.43	1.10	0.26	0.03	0.08	1.46	0.40	1.13
3,7-Dimethyl-1,5,7-octatrien-3-ol	0.05	0.16	0.03	Tr	0.07	0.21	0.05	0.16
$\beta$ -Cyclocitral	0.10	0.07	0.10	0.08	0.13	0.11	0.18	0.16
Phenylacetaldehyde	1.02	0.64	1.12	1.32	1.41	0.78	1.18	0.97
$\alpha$ -Terpineol	0.11	0.06	0.10	0.11	0.02	0.07	0.08	0.08
Linalool oxide (cis pyranoid)	0.04	0.01	0.02	0.02	0.09	0.02	0.03	0.02
Methyl salicylate	0.58	0.41	0.37	0.59	0.57	0.40	0.44	0.54
Nerol	0.06	0.04	0.05	0.06	0.04	0.01	0.05	0.02
Geraniol	0.18	0.13	0.21	0.22	0.13	0.08	0.13	0.16
Geranylacetone	0.02	Tr	0.02	0.02	0.01	Tr	0.02	0.02
Benzyl alcohol	0.03	0.02	0.02	0.04	0.04	0.02	0.02	0.02
2-Phenyl ethanol	0.02	0.01	0.01	0.02	0.02	0.02	0.02	0.01
$\beta$ -Ionone	0.08	0.12	0.11	0.09	0.07	0.08	0.07	0.09
Nerolidol	0.07	0.09	0.03	0.07	0.04	0.04	0.05	0.07
Cedrol	0.11	0.47	0.10	Tr	0.04	1.78	0.14	0.37
Bovolide	0.03	0.03	0.02	0.02	0.01	0.04	0.02	0.03
E-Geranic acid	0.13	Nil	0.04	0.08	0.13	0.02	0.14	0.08
Indole	0.02	0.05	0.01	0.01	0.03	0.07	0.02	0.04
Total VFC	10.54	9.58	11.73	10.16	10.40	9.81	8.25	10.90
Tasters' evaluation	37.25	36.38	33.75	36.38	35.63	39.00	36.00	37.88

\* As ratio of the gas chromatographic peak area to that of internal standard.

<sup>a, b</sup> As in Table 1.

1 and 2. Only compounds whose ratios of peak areas to that of internal standard were equal to or more than 0.01 are reported. The variations noted in the VFC associated with differences in the place of cultivation, or with the clones, have been reported elsewhere (Owuor *et al* 1988). There was  $\pm 5\%$  variation in the peak areas of replicated sample analysis.

The various ratios were calculated based on the data in Tables 1 and 2, as explained in the introductory section. The two clones used were chosen on the basis of their

difference in the VFC composition. Whereas clone 6/8 exhibits *sinensis* character, clone TN 14-3 exhibits *assamica* character (Takeo 1981, 1983b; Owuor *et al* 1987; Owuor 1989). Thus clone 6/8 had larger amounts of geraniol than linalool whereas TN 14-3 showed the reverse effect.

The changes in the Mahanta and Yamanishi-Botheju ratios and their correlation coefficients obtained by linear regression against the tasters' evaluation are presented in Table 3. The correlations between the

TABLE 3

Changes in Mahanta and Yamanishi–Botheju ratios with locations and correlation coefficient of linear regression between the index and tasters' evaluation

Location	Clone 6/8			Clone TN 14-3			Clone 6/8			Clone TN 14/3		
	Total non-terpenoids (1)	Total terpenoids (2)	Mahanta ratio (2/1)	Total non-terpenoids	Total terpenoids	Mahanta ratio	E-2-Hexenal (3)	Linalool (4)	Yamanishi–Botheju ratio (4/3)	E-2-Hexenal	Linalool	Yamanishi–Botheju ratio
Karirana	4.96	5.97	1.20	6.80	3.74	0.55	2.17	1.31	0.60	3.16	1.48	0.47
Timbilil	5.74	6.39	1.11	5.77	3.81	0.66	3.12	0.91	0.29	2.98	1.03	0.35
Sitoi	5.20	3.45	0.66	8.48	3.25	0.38	2.67	1.0	0.37	4.92	1.61	0.33
Kangaita	8.14	5.43	0.67	7.22	2.94	0.41	4.15	1.33	0.32	3.37	1.60	0.47
Kagochi	3.83	2.37	0.62	7.88	2.52	0.32	1.40	0.67	0.48	3.66	1.10	0.30
Changoi	4.66	4.22	0.91	4.73	5.08	1.07	2.41	0.44	0.18	2.15	0.69	0.32
Kaimosi	5.26	3.31	0.63	5.49	2.76	0.50	2.42	0.80	0.33	2.22	0.86	0.39
Sotik	5.87	4.88	0.83	6.69	4.21	0.63	2.80	0.88	0.31	3.24	1.13	0.35
<i>r</i>			0.57			0.81**			0.26			0.10
Pooled <i>r</i>			0.70**						0.18			

\*\* Significant at  $P \leq 0.01$ .

Yamanishi–Botheju ratio and tasters' evaluation were insignificant for Kenyan clonal black teas. Thus the Yamanishi–Botheju ratio, which is significantly related to prices of Sri Lankan orthodox black teas (Yamanishi *et al* 1989), was not appropriate for the Kenyan clonal black CTC teas used in this study. The lack of a significant relationship between the Yamanishi–Botheju ratio and tasters' evaluation noted here is not strange. The ratio ignores too many volatile flavour compounds (see Tables 1 and 2), most of which must be contributing towards the aroma quality of tea. The rationale for the use of the Yamanishi–Botheju ratio requires that linalool and E-2-hexenal occur in large amounts in all teas and therefore have a dominant effect. However, the data presented in Fig 1 and Tables 1 and 2 indicate this not to be true for all black teas. Although the aroma values of the individual aroma compounds have not been quantified, it is noted that in some teas  $\alpha$ -cedrene, geraniol, E-geranic acid and/or cedrol sometimes occur at levels similar to or greater than linalool (Owuor *et al* 1986, 1987, 1988, 1989). Again many black teas contain large amounts of hexanal, linalool oxides, phenylacetaldehyde and methyl salicylate (Owuor *et al* 1986, 1987, 1988, 1989). The Yamanishi–Botheju ratio (Yamanishi *et al* 1989) is therefore unlikely to be a useful index in aroma classification, especially for Kenyan black teas and/or their equivalent.

The ratio of terpenoids to non-terpenoid compounds (Mahanta ratio) showed a significant relationship with tasters' evaluation of clone TN 14-3 ( $P \leq 0.05$ ) and for data pooled from clones 6/8 and TN 14/3 ( $P \leq 0.01$ ) (Table 3). However, the relationship was insignificant for clone 6/8 alone (Table 3). The naturally occurring

terpenes or terpenoid compounds in tea have been classified to give black tea superior flavour (Owuor *et al* 1986 and references therein). However, some non-terpenoids in black tea, eg benzaldehyde (Yamanishi *et al* 1968), phenylacetaldehyde (Motoda 1979), methyl salicylate (Aisaka *et al* 1978; Howard 1978) and benzyl alcohol (Aisaka *et al* 1978) have been demonstrated to have a desirable aroma. Some of these compounds occur in large amounts in some black teas (Tables 1 and 2) and thus their inclusion in the group of compounds imparting inferior flavour to black tea (Baruah *et al* 1986; Mahanta *et al* 1988) may be erroneous. This may explain in part the lack of a significant relationship between tasters' evaluation of clone 6/8 and the Mahanta ratio of clone 6/8 black CTC teas.

The changes in the ratio of the sum of gas chromatographic peak areas eluting on a GC column before linalool to those of linalool plus compounds with retention times higher than linalool (Wickremasinghe–Yamanishi ratio, Wickremasinghe *et al* 1973; Yamanishi *et al* 1978) and the correlation coefficient of the ratios and tasters' evaluation are presented in Table 4. There were significant inverse relationships between the ratios and tasters' evaluation for each clone and pooled data for both clones. Although the classification was based on an arbitrary division based on elution time, the ratio seems to have worked very well. Indeed it has been shown that apart from linalool oxide (*cis* and *trans* furanoid) and benzaldehyde, which have been confirmed by sniffing to have sweet flowery aroma (Yamanishi *et al* 1968), the classification of the rest of the volatiles was similar to that used in Owuor's FI (Owuor *et al* 1986 and references therein). The correlation between the

TABLE 4

Changes in Wickremasinghe–Yamanishi ratio and correlation coefficient of linear regression analysis between the ratio and tasters' evaluation

Location	Clone 6/8			TN 14-3		
	Compounds before linalool (1)	Linalool + compounds after it (2)	Wickremasinghe–Yamanishi ratio (1/2)	Compounds before linalool	Linalool + compounds after it	Wickremasinghe–Yamanishi ratio
Karirana	3.83	7.10	0.54	5.98	4.56	1.31
Timbilil	4.75	7.38	0.64	5.14	4.44	1.16
Sitoi	4.16	4.49	0.93	7.50	4.23	1.77
Kangaita	6.87	6.70	1.03	5.78	4.38	1.32
Kagochi	2.72	3.48	0.78	6.37	4.03	1.58
Changoi	4.00	4.88	0.82	3.91	5.90	0.66
Kaimosi	3.98	4.59	0.87	4.35	3.90	1.11
Sotik	4.61	6.14	0.75	5.80	5.10	1.14
<i>r</i>			–0.87***			–0.88***
Pooled <i>r</i>			–0.74***			

\*\*\* Significant at  $P \leq 0.001$ .

TABLE 5

Changes in Owuor's flavour index and correlation coefficient of linear regression analysis between the indices with tasters' evaluation

Location	Clone 6/8			TN 14-3		
	Group I VFC (1)	Group II VFC (2)	Owuor's FI (Group II/Group I)	Group I VFC	Group II VFC	Owuor's FI
Karirana	3.26	7.67	2.35	5.04	5.50	1.09
Timbilil	4.37	7.76	1.78	4.57	5.01	1.10
Sitoi	3.89	4.76	1.22	6.86	4.87	0.71
Kangaita	6.08	7.49	1.23	5.17	4.99	0.97
Kagochi	2.40	3.80	1.58	5.70	4.70	0.82
Changoi	3.67	5.21	1.42	3.37	6.44	1.91
Kaimosi	3.62	4.95	1.37	3.70	4.55	1.23
Sotik	4.26	6.49	1.52	5.02	5.88	1.17
<i>r</i>			0.69*			0.85**
Pooled <i>r</i>				0.76**		

\*, \*\* Significant at  $P \leq 0.05, 0.01$ , respectively.

Wickremasinghe–Yamanishi ratio and sensory evaluation can be improved by re-classifying these compounds. The rest of the compounds with GC retention time less than that of linalool are products of lipid degradation during tea manufacture, and all these compounds impart a green grassy flavour to black tea (Yamanishi 1981).

The results on flavour index (FI) as determined by Owuor *et al* (1986, 1988) (Owuor's FI) are presented in Table 5. As with the Wickremasinghe–Yamanishi ratio there were significant regressions between FI and tasters' evaluations, except that the regressions were linear and

positive. This index was developed as an improvement of the Wickremasinghe–Yamanishi ratio in that the compounds were assigned to the different groups based on their aroma characteristics as perceived by sniffing and reported in the literature (see Owuor *et al* 1986, 1988 and references therein). The only difference between the Wickremasinghe–Yamanishi ratio and Owuor's FI is the classification of linalool oxides (*cis* and *trans* furanoid) and benzaldehyde in the group of VFC imparting sweet flowery aroma to black tea (Yamanishi *et al* 1968). Both the Owuor and Wickremasinghe–Yamanishi indices of

**TABLE 6**  
Comparison of different indices used to qualify black tea aroma using orthodox tea from different varieties

Variety	Clone/seedling stock	Mahanta ratio	Yamanishi-Botheju ratio	Wickremasinghe-Yamanishi ratio	Owuor's FI	Sensory evaluations
<i>Assamica</i>	31/8	0.89	1.35	0.75	2.13	127
	31/11	1.15	1.41	0.82	2.32	125
	S15/10	1.40	0.65	0.52	2.28	120
	6/8	1.17	1.07	0.55	2.21	138
	St 18	1.01	1.37	0.95	1.65	120
<i>Sinensis</i>	14/1	0.58	0.31	1.18	1.02	72
	Seedling China	0.56	0.25	1.44	0.85	60
<i>Lasiocalyx</i> (Shan tea)	301/4	0.55	0.73	0.91	1.33	90
	301/5	0.69	0.85	1.32	1.22	89
	301/6	1.03	1.37	0.95	1.75	120
<i>r</i>		0.83*	0.75*	-0.87***	0.95***	

*r* = Linear regression coefficient between the different indices and sensory evaluation.

\*\*, \*\*\* Significant at  $P \leq 0.05$  and  $0.001$ , respectively.

aroma quantification worked better in this study than the Yamanishi-Botheju ratio or the Mahanta ratio.

In the above studies the ratios were used on CTC clonal black teas. However, the Yamanishi-Botheju ratio was shown to have worked best on Sri Lankan orthodox black teas (Yamanishi *et al* 1989). A comparison of the relationship between sensory evaluation and the different ratios for Kenyan orthodox black teas manufactured from different varieties is presented in Table 6. Best relationships with sensory evaluations were shown by the Wickremasinghe-Yamanishi ratio and Owuor's FI. This was followed by the Mahanta ratio while the Yamanishi-Botheju ratio demonstrated the worst relationship. Similar results are expected from equivalent teas from different parts of the world. Although Owuor's FI is more sensitive to the methods of tea manufacture and discriminates tasters' scores well at FI below 2.0, the method plateaus off above an FI of 2 (Table 6). Thus the method is less sensitive for discriminating black teas with FI above 2. Again it is recognised that the relative concentrations of volatiles as detected by GC following steam distillation may not necessarily reflect the relative concentration above a cup of black tea.

Although these ratios (or indices) may seem to work reasonably well, the data must be used with caution. The indices should at best be treated as qualitative since the olfactory perception limits of different VFC are variable. Some VFC may exist at low levels and affect aroma more than those occurring at high levels (Howard 1978; Kobayashi *et al* 1988). Again the contribution of each VFC to flavour is not proportional to gas chromatographic area. It is known that the relationship between stimulant concentration (represented by GC peak area) and neural response (perceived flavour intensity) is not

linear. Progressive increases in stimulus give progressively smaller increases in neural response. Since for some VFC the range between minimum and maximum concentrations encountered may be considerable, better correlation with sensory evaluation might be obtained using logarithmic or power transformations of the peak areas. This aspect will be addressed in future studies.

The relative concentrations of volatiles as measured by GC after steam distillation do not necessarily represent the concentrations in the headspace of a tea brew. Thus the VFC composition presented in this study could be different from that in a tea brew. It is therefore necessary to develop an aroma extraction procedure which could more closely replicate the composition in the tea brew.

This study employed a relatively small number of black tea samples. It is desirable in future studies to extend the range in order to test whether the ratios are of more general application. However, despite using only two clones for CTC and orthodox manufactured from different varieties in this study, the data presented have demonstrated the need to improve the aroma quantification methods of black tea so that data from different laboratories can be easily compared.

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