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Carotenoid composition, distribution and degradation to flavour volatiles during black tea manufacture and the effect of carotenoid supplementation on tea quality and aroma

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Abstract

Carotenoid content of tea leaves ranges from 36 to 73 mg/100 g dry weight, and is dominated mainly by β-carotene, lutein and zeaxanthin. Among the cultivars, China contained the maximum and Assam clone the least. Carotenoid fractions were found to degrade to different extents at different stages of tea processing. The carotenoid content was as low as 25 mg/100 g in the made tea. Only a small quantity was leached into the brew, the remaining being retained in the infused leaf/tea residue. The high stability of carotenoid in tea is mainly due to the presence of antioxidants, such as polyphenols and catechins. Carotenoid degradation was found to be greater in the CTC (Crush, Tear, Curl) process than the orthodox process, greater in withered than unwithered, and in the order β-carotene > zeaxanthin > lutin. Vitamin A value was greater in orthodox tea than CTC tea and it varied with clones. The carotenoid degradation was found to yield large quantities of desirable aroma volatiles in made tea, giving a high grown flavour status. An increase in endogenous carotene content enhanced all the quality parameters of tea, the VFC (volatile flavour compounds) index, almost being doubled. The tasters' evaluation also revealed the same trend. It was found that a 1:1 NK application at the rate of 300 kg/ha/year enhanced the carotenoid content of green leaves in the second week after application, with subsequent decline.

Keywords: Carotenoid; Black tea; Degradation; Volatile flavour compounds

1. Introduction

Tea is a perennial evergreen shrub belonging to the Camellia genus of the Theaceae family. It is manufactured by a variety of processes, producing a range of beverages from green, non-fermented tea through to black, fermented tea (Takeo & Hampton, 1992). The consumer acceptability of these beverages is largely dependent on the flavour of the finished product (Ravichandran & Parthiban, 1998c). Tea aroma, which is composed of the volatile flavour compounds (VFC) generated during tea processing, is an important quality parameter, determining the price of made tea. These VFC can be divided into two groups. The Group I compounds are mainly the products of lipid breakdown, which imparts an undesirable grassy odour. However, the Group II compounds, which impart a sweet flavoury aroma to black tea, are mainly derived from terpenoids, carotenoids and amino acids. The flavour of made tea depends on the ratio of the sum of VFC Group II to that of VFC Group I, which is the flavour index or VFC index.

Many volatile compounds, collectively known as the aroma complex, have been detected in tea (Ravichandran & Parthiban, 1998b ; Skobeleva, Petrova, & Bokuchava, 1987). Most of the aroma compounds are formed during tea processing and are derived from carotenes, amino acids, unsaturated fatty acids (Ravichandran & Parthiban, 2000; Yamanishi, 1981) and terpene glycosides (Takeo, 1981). Carotenoids are yellow pigments present in the fresh leaf and their degradation during manufacture leads to the formation of terpenoid flavour compounds in black tea (Hazakira & Mahanta, 1983). The carotenes are therefore important precursors in green leaf for the manufacture of high quality black teas. Owuor and Orchard (1989) have recently reviewed this subject. While the literature on non-carotenoid

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precursors is extensive (Ravichandran & Parthiban, 1998a, 1998b), that of the carotenoids is sporadic (Robinson & Owuor, 1992). The aim of this study was to determine the variation in carotenoid composition in genetically different clones and to study the course of their degradation during each stage of manufacture. The effect of endogenous addition of carotene, to cut dhool, on the made tea aroma has also been studied, along with its enhancement by cultural practices.

2. Materials and methods

Tea leaves of UPASI—3, 9 and 17 clones, representing the genetically diverse Assam, China and Combod cultivars, were harvested from the experimental plot of United Planters Association of Southern India—Tea Research Institute (UPASI-TRI). They consisted mainly of three leaves and a bud. Tea was manufactured (Ravichandran & Parthiban, 2000) in a mini manufacturing unit giving 68% withering, 5 CTC (Crush, Tear, Curl) cuts, optimum fermentation and drying for 30 min at 120 °C. Carotenoids (extracted from Rhodotorula yeast) were a gift from the Central Food Technological Research Institute, Mysore, India, and were added at 1% (to withered leaf) ratio in the third CTC cut by uniform mixing with the cut dhool.

About 50 g of tea leaves (or 10 g of tea dust) were extracted with 100 ml methanol, using a homogeniser, and filtered. The methanol extract was pooled. The residue was extracted successively with acetone (100 ml) and peroxide-free diethylether (100 ml). The total extract was washed with water and the organic layer was evaporated in vacuo. In order to remove chlorophylls and fats, the residue was refluxed with 10% methanolic potassium hydroxide (50 ml) for 20 min at 60 °C under nitrogen atmosphere and left overnight at room temperature in the absence of light. Carotenoids were extracted after saponification with peroxide-free diethyl ether, washed free of alkali, concentrated and stored in light petrol (40–60 °C).

The extracted carotenoids were subjected to alumina column chromatography, by gradient elution, employing peroxide–free diethyl ether and petrol (40–60 °C) and acetone-petrol mixtures. Tentative identification was carried out using a UV–visible spectrophotometer, by monitoring absorption maximum and visible absorption spectra (Davis, 1965; Harborne, 1973) and comparing with literature (Foppen, 1971). The estimation of carotenoids was carried out by using the $E^{1\%}_{1cm}$ values of the authentic compounds (Goodwin, 1955). The authentic samples were gifts from CFTRI, Mysore, where they had been isolated from natural sources/synthesised chemically and purified to 99.8% purity. The methods described by Owuor and Odhiambo (1993) and AOAC (1996) methods were followed throughout. The volatiles were extracted with dichloromethane, using a Likens–Nickerson apparatus, by the simultaneous distillation and extraction method and analysed by GC–MS (QP-5000 Mass Spectrometer GC-17 AGC Shimadzu),

Table 1
Carotenoid and chlorophyll composition of tea leaves and dust (mg/100g dry wt.)[a]

Carotenoids	Assam (UPASI-3)		China (UPASI-9)		Cambod (UPASI-17)	
	Leaf	Dust	Leaf	Dust	Leaf	Dust
Pytoene	1.07	0.39	2.18	0.62	1.70	0.46
Antheraxanthin	1.65	0.33	1.77	0.41	1.70	0.37
Lycopene	3.25	0.56	2.81	0.53	2.83	0.53
Pytofluene	0.09	0.04	0.08	0.06	0.05	0.03
α-Carotene	0.31	0.21	2.31	0.81	1.87	0.44
β-Carotene	8.14	3.96	23.1	11.67	16.0	6.11
β-Zeacarotene	2.69	0.22	0.97	0.04	1.37	0.09
Aurochrome	0.16	Trace	0.23	Trace	0.19	0.03
Mutatochrome	0.17	0.21	0.31	0.59	0.23	0.58
β- Cryptoxanthin	0.89	0.03	3.25	0.07	2.67	0.05
Cryptoflavin	0.48	0.50	0.08	0.36	0.22	0.41
Cryptoxanthin-5,8-diepoxide	Trace	0.05	0.46	0.07	0.46	0.05
Lutein	10.0	6.13	211	7.09	16.7	6.56
Zeaxanthin	11.2	5.91	17.71	7.01	15.91	6.88
Lutein-5, 6-epoxide	0.46	3.17	0.06	1.67	0.12	2.27
Violaxanthin	1.49	Trace	0.25	0.11	0.71	0.09
Neoxanthin	0.33	0.23	1.97	0.06	1.58	0.12
Chlorophyll ' a '	782	183	1034	271	719	281
Chlorophyll ' b '	314	69.5	437	101	255	70.9
Total chlorophyll	1095	255	1471	376	1002	249
(a:b)	(2.49:1)	(2.63:1)	(2.46:1)	(2.69:1)	(2.82:1)	(3.96:1)
Total carotenoid	36.7	6.10	73.1	24.7	49.3	11.5

[a] Data are average of three trials in triplicate as mean with standard deviation less than 0.9%. Trace = less than 0.01

Table 2
Pigment degradation during tea manufacture[a]

UPASI-9	Total carotenoid	Total chlorophyll (a:b)	Chlorophyll 'a'	Chlorophyll 'b'	Polyphenols	Catechins
Green leaf	73.1	1471 (2.36:1)	1034	437	31.7	24.6
Withered leaf	70.3	1441 (2.46:1)	1014	412	29.1	22.6
Fermented dhool	61.6	1202 (2.24:1)	830	372	13.4	11.7
Dried tea	24.7	376 (2.69:1)	271	101	10.5	9.6
Brew	9.9	247 (3.19:1)	186	58.5	5.8	4.2
Tea residue	16.4	129 (2.01:1)	84.8	42.3	7.0	6.3

[a] Chlorophyll expressed as mg/g dry weight. Carotenoid expressed as mg/100 g dry weight. Polyphenols and catechins expressed as g/100 g. Data are average of three trials in triplicate as mean with standard deviation less than 0.8%.

using a carbowax 20 M (50 m × 0.25 mm) fused capillary column with FID detector. MS ion source temperature was 200 °C and electron energy was 70 eV. The data obtained were compared to those of the authentic compounds (Ravichandran & Parthiban, 1998d). Sensory evaluation was carried out by professional tea tasters.

3. Results and discussion

Table 1 shows the carotenoid composition of Assam, Cambod and China clonal materials which are genetically diverse cultivars. It is very clear from the Table that the three varieties differ very much in their carotenoid composition. While the China clone dominates in the carotenoid distribution, the Assam clone contains the least with the Cambod variety falling in between these two. Of the different carotenes, β-carotene, lutein and zeaxanthin constitute the bulk, in all three varieties. This suggests that, much of the α-carotene formed may be hydroxylated to lutein and that little of the β-carotene formed might be converted to corresponding xanthophylls (Robinson & Owuor, 1992).

Analysis of carotenoid composition of dust reveals a close resemblance to that of leaves. The trend observed in green leaf is also found in dust. It is surprising to find, especially in view of the extreme lability of carotenoids, that most of the carotenoids detected in tea leaf could also be detected in dust. As in leaves, β-carotene and lutein-zeaxanthin constitute the bulk of hydrocarbon and xanthophylls, respectively. The vast difference in carotenoid composition of leaf and dust clearly indicates that the loss during manufacturing was significant, being as high as 20–30%. The individual carotenoids seem to get decomposed to different degrees. Also, a careful observation of the data, reveals that the hydrocarbon carotenoids decompose to a greater extent than the xanthophylls during manufacture. Chlorophyll composition and degradation were also studied, as it is the principal pigment present in tea.

In order to elucidate the extent of carotenoid degradation at different stages of manufacture, a systematic study was undertaken to monitor the pigment changes during manufacture. Table 2 shows the extents of carotenoid and chlorophyll degradation at different stages of manufacturing. The extent of degradation is quite different at different stages of manufacturing. The carotenoid degradation was much less during the withering stage and increased a little during the fermentation stage. However, the major loss was observed during the firing stage and in brewing. The carotenoid decomposition occurs mainly by an oxidative enzymatic reaction which takes place during withering and fermentation and a pyrolytic reaction during firing (Sanderson & Graham, 1973). Apart from these, carotenes also undergo photo- and auto-oxidative reactions (Sanderson & Graham, 1973). It has been reported that the carotene loss increases with degree of wither (Hazakira & Mahantha, 1983). However, the extent of degradation in tea, compared to isolated carotenoid, is very much less and this may be due to the presence of antioxidants such as polyphenols and catechins (Howard, 1978). The high retainment of carotenoid in the tea residue supports this suggestion. Little of it is leached into the brew. The above study indicates that around 70–80% of the total carotenoid is decomposed during processing.

The changes in carotenoids during CTC and orthodox methods of manufacture are given in Table 3. The degradation of carotenoids is greater in CTC teas than orthodox teas. Again, the rates of degradation of the various fractions were in the order: β-carotene > zeaxanthin > lutein. Also, the carotenoid degradation in withered CTC tea is slightly greater than in unwithered CTC teas. Minor degradation of carotenoids during withering may be attributed to the cell permeability and photoisomerisation which gains momentum during rolling and fermentation. Significantly higher degradation of cartenoids in CTC teas than in orthodox teas may be due to the greater exposure of the polyphenols caused by severe cell damage in the CTC machine. It is noteworthy that the random and greater degradation of carotenoids in CTC leaves may not be commensurate with the expected flavour of tea. On the contrary, the orthodox teas with less carotenoid degradation are in fact more flavoursome. This is probably due to greater retention of essential flavour components, such as ionones and linalool and its oxides, in orthodox teas

Table 3
Degradation of carotenoids (mg/100 g) in orthodox and CTC teas manufactured from UPASI 9[a]

Sample	β-carotene	Lutein	Zeaxanthin
Fresh leaf	23.1±0.09	21.1±0.12	17.7±0.10
Orthodox tea (withered)	16.6±0.11	16.8±0.08	12.6±0.06
CTC tea (withered)	17.7±0.07	14.2±0.03	8.8±0.02
CTC tea (unwithered)	13.8±0.06	15.1±0.06	10.0±0.06

[a] Data are average of three trials in triplicate as means with standard deviation.

Table 4
Vitamin A value of orthodox and CTC teas (mg/100 g)[a]

Clone	Orthodox tea	CTC tea
UPASI-3	6.3±0.08	1.98±0.06
UPASI-9	8.3±0.09	5.84±0.07
UPASI-17	7.1±0.06	3.06±0.07

[a] Data are average of three trials in triplicate as means with standard deviation.

than in CTC teas. Moreover, it may also be expected that higher amounts of oxidised polyphenols in CTC leaves would influence the formation of low boiling secondary oxidation products of carotenoids, such as hydrocarbons and aldehydic compounds, such as phenylacetaldehyde.

The vitamin A values of orthodox and CTC teas were calculated, based on residual β-carotene (Ceechi & Rodriguez, 1981). Table 4 shows the vitamin A values of orthodox and CTC teas. The lower carotenoid degradation accounts for the higher vitamin A value in orthodox teas. The study of carotenoids in tea with good vitamin A value may prove to be significant from therapeutic and nutritional points of view. Irrespective of the method of tea manufacture, clonal variation in vitamin A content is also observed.

In order to account for the loss in carotenoids, further studies were undertaken on the aroma profile of the made tea. It is established that carotenoids undergo degradation, giving rise to volatile substances of flavour

Table 5
Volatile flavour components profile of made tea with added carotenoid[a]

VFC Compounds	Control	Carotenoid added
n-Hexanal	0.24	0.25
Cis-3-Hexenal	1.12	1.10
Trans-2-Hexenal	3.27	3.28
Cis-3-Hexenol	0.21	0.19
Trans-2-Hexenol	0.49	0.48
n-Hexanol	0.12	0.13
1-Penten- 3- ol	0.13	0.13
2-Penten- 1- ol	0.10	0.11
1-Octen- 3- ol	0.11	0.09
Linalool	0.99	1.21
Linalool oxides	0.13	0.26
Phenyl acetaldehydes	1.32	1.36
Methyl salicylate	0.47	0.51
Geraniol	0.93	0.92
2-Phenyl ethanol	0.36	0.33
Benzaldehyde	0.06	0.06
Benzyl alcohol	0.09	0.08
α-ionone	0.32	0.59
β-ionone	0.26	0.66
3-Hydroxy-β-ionone	0.02	0.05
3-Hydroxy-5,6-epoxy ionone	0.01	0.04
3,5-Dihydroxy-4,5-dihydro-6,7-didehydro-α-ionone	Trace	0.02
Dihydro actinidiolide	Trace	0.02
2, 2, 6-Tri methyl cyclohexanone	Trace	0.04
5, 6-epoxy ionone	0.01	0.05
2, 2, 6-Trimethyl- 6- hydroxy cyclo hexanone	Trace	0.02
Theaspirone	0.01	0.04
β-Damascenone	Trace	0.02
α-Damascone	Trace	0.03
β-Damascone	Trace	0.04
3-Oxo-β-ionone	Trace	0.05
1, 2-epoxy-1′, 2′-dihydro-β-ionone	0.01	0.05
Loliolide	Trace	0.03
Dehydrovomifoliol	Trace	0.01
3, 7-Dimethyl-1, 5-octadien-3, 7-diol	Trace	0.01

[a] As ratio of peak area to that of internal standard. Data are averages of three trials in triplicate as mean with standard deviation less than 0.7%. Trace = less than 0.01.

value (Robinson & Owuor, 1992). Table 5 details the aroma profile of made tea. To determine the contribution of carotenoids to the aroma and made tea quality, experiments were undertaken with the endogenous addition of natural carotenoids to cut dhool before fermentation. It is seen that the addition of carotenoid enhances the VFC index, which indicates an increase in VFC group II compounds. The formation of aroma compounds from carotenoids occcurs by the various pathways of degradation mentioned above. Evidently β-ionone is a major degradation product of β-carotene, while β-ionone, α-ionone, 3-hydroxy-5,6-epoxyionone, 3, 5-dihydroxy-4,5-dihydro-6,7-didehydro- α-ionone, linalool and other terpenoid aldehydes and ketones are degradation products of other carotenes present in tea. Dihydro-actinidiolide, 2,2,6-trimethylcyclohexanone, 5,6-epoxyionone, 2,2,6-trimethyl-6-hydroxy cyclohexanone and theaspirone are possibly formed from the primary oxidation product of carotene ie. β-ionone. Also, the addition of carotenoids influenced the formation of β-damascenone, α-damascone, β-damascone, 3-oxo-β-ionone, 1,2-epoxy-1′,2′-dihydro-β-ionone, loliolide, dehydrovomifoliol and 3,7-dimethyl-1,5-octadien-3.7-diol, suggesting their formation from carotenoids. Thus the compounds produced from carotenoids have a major effect on the aroma of tea. Flavoursome teas are normally produced from green leaf with high carotene contents (Wickremasinghe, 1974). The quality parameters also showed a positive trend for acceptability. The endogenous addition of carotenoids (Table 6) increased the desirable parameters, such as theaflavin, thearubigin, highly polymerised substances and total liquor colour. A rise in total liquor colour indicates a corresponding increase in cuppage and colour of the liquor. The increase in VFC index is mainly due to the contribution of flavoursome note. All these chemical parameters support tasters' evaluation which also revealed the above findings.

Considering the restrictions in the use of chemicals (natural/synthetic) in black tea manufacture, a field experiment was conducted to study the effects of agronomic practices on the carotenoid content of tea. Several experiments conducted at UPASI Tea Research Institute on the use of NK (nitrogen/potassium), have revealed highly positive effects on quality (Upasi, 1995). Considering the contribution of carotenoids to the overall aroma, a study was undertaken to elucidate the effect of NK application on carotenoid content and hence quality. Results are in Table 7. It is very clear that there is a linear response of carotenoid content with N levels up to 300 kg but it declines sharply above this level. Also, carotenoids build up and reach a maximum during the second week of fertiliser application and tail off thereafter. The ratio of N:K also seems to affect carotenoid content. An equal amount of N and K is shown to yield more carotenoids. Thus, the present study clearly demonstrates that the carotenoid content contributing to tea quality, particularly the volatiles, can also be enhanced by proper application of NK fertiliser.

The above investigation reveals that tea leaves contain appreciable amounts of carotenes, which decrease during tea manufacture with the resultant production of various aroma compounds. By controlling the reactions in black tea manufacture, it may be possible to enhance the formation of essential and characteristics flavoursome volatile products. The endogenous addition of natural carotenoids significantly and economically enhances the aroma and quality of made tea. The carotenoid content of green leaf can be improved through proper NK application with suitable intervals and plucking rounds.

Table 6
Changes in quality parameters upon addition of carotenoids[a]

Parameters	Control	Carotenoid added
Theaflavin (%)	0.87	0.95
Thearubigin (%)	8.50	10.34
Highly polymerised substances (%)	6.00	8.14
Total liquor colour	2.75	2.90
VFC index	1.43	2.61
Water extract (%)	36.22	36.01
Tasters' evaluation		
Appearance	Grey/dull	Grey/fair
Infusion	Bright/coppery	Bright/coppery
Aroma	Clonal flavour	High grown flavour
Liquor	Bright/moderate Strength	Bright/high Strength
Comments	Preferred	More preferred

[a] Data are average of three trials in triplicate as mean with standard deviation less than 0.9%.

Table 7
Effect of NK application on the carotenoid content in tea [a]

N:K	Level (kg/ha/year)	Carotenoids (μg/g fresh weight)			
		7th day	14th day	21st day	28th day
2:1	50	118	165	155	148
2:1	100	135	190	168	163
2:1	200	146	200	178	173
2:1	300	160	215	188	175
2:1	400	140	160	140	133
2:1	500	138	155	140	130
1:1	50	120	169	158	150
1:1	100	138	195	183	173
1:1	200	158	200	186	183
1:1	300	175	230	198	190
1:1	400	140	173	168	163
1:1	500	138	166	160	155

[a] Data are average of three trials in triplicate as mean with standard deviation less than 1.19% and CV less than 2.1%.

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