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Levels of Aluminium in Green Leaf of Clonal Teas, Black Tea and Black Tea Liquors, and Effects of Rates of Nitrogen Fertilizers on the Aluminium Black Tea Contents

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ABSTRACT

Clonal teas have different aluminium content increasing with age of leaf. Although aluminium and fertilizer nitrogen enhance tea growth and tea yields, respectively, field trials have shown no relationship between total aluminium content in the leaf and clonal tea yields. Higher aluminium concentrations were found in the dust grades of black tea than in the large size grades. However, aluminium levels decreased in the tea liquor from dust grades compared to the large size grades. Only up to 40% of the total aluminium in tea was infused into tea liquors; the amounts infused varied with clones and grading (sorting). Aluminium content of black tea was lowered by increasing rates of nitrogenous fertilizers, more frequent fertilizer application, and application of NPK 20:10:10 as opposed to NPKS 25:5:5:5.

INTRODUCTION

The tea plant *Camellia sinensis* (L.) O. Kuntze is known to accumulate a lot of aluminium (Chennery, 1955). More than 30 000 ppm aluminium have been recorded in the old leaves of the tea plant (Matsumoto *et al.*, 1976). These

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large amounts have not produced any toxic effects in the plant (Matsumoto *et al.*, 1976; Sarishvili & Egorashvili, 1978). In young tea plants, the growth rate is accelerated by addition of aluminium (Chennery, 1955; Chanturiya, 1976; Matsumoto *et al.*, 1976; Ishigaki, 1984; Kinoshi *et al.*, 1985), suggesting that aluminium could be an essential nutrient for tea. In watermelon (Sundstrom *et al.*, 1983) and cocoa (Santana & Cabala-Rosand, 1984), however, no relationship was recorded between yield and leaf aluminium levels. Growth of tea in sand culture only responded up to levels of 50 ppm aluminium in the culture medium (Ishigaki, 1984).

Whether phosphorus is applied as fertilizer or not, excess aluminium in the soil improves the phosphorus uptake by the tea plant (Sivasubramanium & Talibudeen, 1972; Bhattachavya & Dey, 1983; Kinoshi *et al.*, 1985). However, aluminium reduces uptake of potassium (Ishigaki, 1972; Matsumoto & Yamaya, 1986) and calcium (Memon *et al.*, 1981; Korcak, 1984) by tea and cocoa plants (Ng *et al.*, 1970). Calcium also lowers the uptake of aluminium by the tea plant (Memon *et al.*, 1981). Ammonium-nitrogen fertilizer application leads to a higher aluminium content in the tea plant than nitrate-nitrogen (Ishigaki, 1974). Thus, several nutrients needed by the tea plant interact with aluminium in the soil either to increase or reduce the plant uptake of the element. It is therefore important to understand factors which affect the aluminium content in the plant and soil.

Additionally, tea quality has been said to be improved by aluminium contents (Chanturiya, 1976; Edmonds & Gudnason, 1979; Chang & Gudnason, 1982). When aluminium salts were added to tea 'dhool' during manufacture (Edmonds & Gudnason, 1979), or if aluminium fertilizers were applied to tea plants, the resultant teas were normally of superior quality. Recently, Reeves *et al.* (1985) showed that higher quality, as judged by the tea tasters, is the result of theaflavin–aluminium complexation improving the colour of the infusion. It was also demonstrated in this study, that the aluminium–theaflavins complexation could be used profitably as a cheap alternative method of theaflavin (TF) analysis in black tea. However, if a high degree of accuracy is required, this test would be less reliable because tea accumulates high amounts of aluminium, and after aluminium–theaflavin complexation during infusion, TF becomes less soluble in organic solvents (Chang & Gudnason, 1982), which would cause errors in the theaflavins analysis. It is, therefore, important to know how much aluminium is extracted into the tea liquor, as the Flavognost method for theaflavins analysis method (Hilton, 1973) can be affected. This paper reports on the fluctuations in aluminium levels in clonal green tea leaves, total and extractable aluminium content of black tea from different clones and their liquors, and the effects of different rates and frequencies of nitrogenous fertilizers on total black tea aluminium content.

MATERIALS AND METHODS

Aluminium in clonal green leaf

Teas used in this study were obtained from clonal field trials (CFT) of the Botany Department, Tea Research Foundation of Kenya, at latitude 0° 22′ and an altitude of 2178 m above mean sea level. Clonal materials planted in 1983 were used to determine

(a) variations in aluminium content between different clones;
(b) distribution of aluminium in different parts of the shoots from each clone.

Leaves to determine effects of age of pluckable shoots on aluminium content were plucked at three leaves and a bud plucking standard. Subsequently, the leaves were separated into a bud, first leaf, second leaf, third leaf, stem, and two leaves and a bud. Clones were the main treatments with shoot parts as split treatments. The experiment was replicated three times. Yields from this experiment were obtained during 1986 using a seven-day plucking interval.

Aluminium in clonal black tea

Leaf from different CFT planted in 1982 was manufactured and the aluminium measured in both black tea and black tea liquors for each clone. A commercial plucking standard was adopted, i.e. 90% good leaf of two leaves and a bud plus a small amount of three leaves and a bud, and the teas were manufactured by CTC maceration method (Owuor *et al.*, 1987*a,b,c*, 1988). The infusions (liquors) were made using 375 g boiling water and 9 g dry black tea (Reeves *et al.*, 1985).

Variations of aluminium due to drier type and sorting

The effect of the type of drier and sorting (grading) on the aluminium content of black tea was investigated in commercial teas from Kapkoros Tea Factory of the Kenya Tea Development Authority (see Owuor *et al.*, 1987*b*, for manufacturing procedure). The tea liquors were made as in the previous section.

Effects of compound nitrogen fertilizers

The changes in the black tea aluminium content due to different rates of nitrogen were determined from two experiments. In the first experiment, seedling tea planted in 1960 was used. The tea was receiving NPKS 25:5:5:5

fertilizer at 50, 100, 200, 400 and 800 kg N/ha/year. Each level was applied at different frequencies, i.e. once a year, 1/2 rate at six month intervals, and 1/4 rate at three month intervals. The experiment was replicated three times. First fertilizer applications were done in September 1983, and subsequently, in December, March, June and September every year, i.e. depending on when each plot was due to receive fertilizers. For miniature manufacture (Owuor *et al.*, 1987*a, b, c*, 1988), teas were sampled in April 1988.

In the second experiment, different rates of 0, 100, 150, 300, 450 and 600 kg N/ha/year, split for NPKS 25:5:5:5 and NPK 25:10:10, were applied to high yielding clone S 15/10. The first experimental treatments were done in September 1984. Subsequently, the teas received fertilizer in September every year. Manufacture was done in April 1988, as outlined earlier (Owuor *et al.*, 1987*a,b,c*, 1988). Tea liquors were made as outlined previously (Hilton, 1973; Reeves *et al.*, 1985).

Aluminium determination

Aluminium was determined using atomic absorption spectrophotometry with a nitrous oxide-acetylene flame at 309 nm (Puchyr & Shapiro, 1986).

RESULTS AND DISCUSSION

The distribution of aluminium in the different parts of the pluckable shoots of clonal teas is presented in Table 1. In all the clones studied, the level of aluminium significantly increased with age of the leaf. Thus, the bud had the lowest aluminium content, while third leaf had the highest. Similar observations had been made in earlier studies (Chennery, 1955; Matsumoto *et al.*, 1976; Memon *et al.*, 1981). Thus, one of the possible methods of reducing black tea aluminium content is practising fine plucking, i.e. plucking younger and more tender shoots only. Different clones exhibited varying ability to accumulate aluminium in their young shoots, and aluminium contents were not linearly correlated with yields (Table 1). In earlier studies (Ishigaki, 1984; Kinoshi *et al.*, 1985; Matsumoto & Yamaya, 1986), it was observed that aluminium stimulated growth of young tea plants. Such a relationship was not observed in the clonal teas studied. Ishigaki (1984) had demonstrated that the growth of tea responded only up to levels of 50 ppm in the soil. All the tea soils in these studies have aluminium levels above 100 ppm (Owuor, unpublished) which may explain the lack of correlation between aluminium levels and growth.

Recently, there have been implications of aluminium in Alzheimer's disease in the Western world. Tea was thought of as one of the beverages

TABLE 1

Clonal Variations of Aluminium (ppm) with Age of Pluckable Tea Leaves and Clonal Tea Yield Variations

Clone	*Bud*	*First leaf*	*Second leaf*	*Third leaf*	*Stem*	*2 + Bud*	*Mean*	*Yield 1985/86 (kg/made tea/ha/year)*
6/8	147	208	397	581	166	277	296	1 040
54/40	193	230	337	513	227	299	300	1 400
56/89	199	227	335	544	263	290	310	1 296
7/9	119	231	418	819	189	301	346	1 042
7/14	160	237	470	573	278	314	339	1 255
31/8	134	198	301	659	219	291	300	1 308
31/11	149	279	365	833	219	327	354	852
100/5	138	189	350	559	193	287	386	1 179
108/82	126	195	477	925	219	355	383	1 120
12/12	193	259	498	609	250	284	349	1 124
Mean	156	220	395	662	222	302	—	—
r[a]	0·12	−0·42	−0·31	−0·64	0·43	−0·27	−0·42	—

CV%	Clone	2·96			10·86		
	Leaf	18·08					
LSD	*P* =	0·05	0·01	0·001	0·05	0·01	0·001
Clone		41	56	NS	216	296	NS
Leaf		30	40	31			
Clone and leaf		95	126	162			

[a] Correlation coefficient of linear regression analysis between aluminium levels and yield. CV, Coefficient of variation; LSD, least significant difference; *P*, probability level; NS, not significant.

containing excessive amounts of aluminium. In black clonal tea, there were also variations of aluminium with clones (Table 2). The results presented here (Tables 1 and 2) suggest that aluminium in tea can be drastically reduced by proper clonal selection.

Although tea accumulates a lot of aluminium, not all the aluminium in black tea is extracted during tea making (Tables 2 and 3). Only between 30 and 45% of the aluminium is extracted. Also the amounts extracted depend on the clone (Table 2) and the grades of tea (Table 3). It was noted that the smaller grades of teas, Pekoe Dust (PD) and Dust 1 (D1) had higher aluminium content than the larger grades of tea, Broken Pekoe (BP 1) and Pekoe Fannings (PF 1). However, the aluminium extracted (%) was lower with the smaller grades than with the larger grades (Table 3). These results suggest that, despite the high accumulation of aluminium in tea leaf, the amount that is infused into the liquors is not as high. Thus, the contribution of aluminium in human diet by tea is not as high as might be expected from

TABLE 2
Clonal Variations of Aluminium in Black Tea, Black Tea Liquors and Tasters' Evaluation

Clone or seedling stock	*Total aluminium (ppm)*	*Total infused (ppm)*	*Infusion (%)*	*Tasters' evaluation*[a]
6/8	472	144	30·5	36·08
St. 18	372	117	31·5	35·08
311/235	498	195	39·2	35·08
311/238	482	171	25·5	36·83
311/246	443	152	34·3	36·67
311/254	451	136	30·2	37·33
311/282	440	162	36·8	36·83
337/40	378	120	31·7	37·00
337/100	498	168	33·7	36·42
337/105	414	155	37·4	37·42
337/113	451	200	44·3	36·75
337/138	425	139	32·2	36·17
CV%	12·96	18·43	—	2·54
LSD $P = 0{\cdot}05$	97	48	—	0·53
r[b]	0·08	0·03	—	—

[a] Based on brightness, briskness, colour, thickness, flavour and infusion on a scale of 0 to 10 for each component.
[b] Correlation coefficient of linear regression analysis between aluminium levels and tasters' evaluation.

the total aluminium in tea. Additionally, tea is drunk as an infusion with a dilution factor of about 40 (41·7 in this study). Thus, the actual aluminium concentration in tea infusion is below 5 ppm.

Increasing rates of nitrogenous fertilizers generally increase soil acidity (Bhavanandan & Sunderlingham, 1971), which is known to increase soil-available aluminium. In Russia, Sarishvili and Egorashvili (1978) noted that raising rates of nitrogen from 100 to 500 kg N/ha/year caused an increase in soil-available aluminium. However, in this study, increasing rates of nitrogenous fertilizers (Tables 4 and 5) reduced the total aluminium in black teas. This result is the reverse of what is expected from the soil aluminium availability data (Bhavanandan & Sunderlingham, 1971; Sarishvili & Egorashvili, 1978). This aspect needs further critical study. It is postulated that the decrease in tea plant uptake of aluminium with increasing rates of nitrogen may be a reflection of the competition between the uptake of aluminium and ammonium ions. As the nitrogenous fertilizer rates increase, more ammonium ions are available to the tea plant and are probably absorbed in preference to aluminium. Tea has been shown to take up

TABLE 3
Variations of Aluminium Contents (ppm) in Black Tea and Black Tea Liquors, due to Driers and Sorting (grading)

Drier	*Grade*	*Aluminium in black tea*	*Aluminium in tea liquor*	*Extraction (%)*
Conventional drier	BP 1	504	183	36·3
	PF 1	502	183	36·5
	PD	544	161	29·6
	D1	526	172	32·7
Mean		519	175	33·7
Fluid bed drier	BP 1	522	184	35·2
	PF 1	504	179	35·5
	PD	537	175	32·6
	D 1	517	164	31·7
Mean		519	176	33·9
Grade means	BP 1	513	184	35·9
	PF 1	503	181	36·0
	PD	539	168	31·2
	D 1	522	168	32·2
CV%	Grade	1·25	1·68	—
	Drier	4·43	3·41	—
(LSD $P = 0{\cdot}05$)	Grade	18	8	—
	Drier	NS	NS	—
	Grade × Drier	NS	NS	—

TABLE 4
Effects of Rates and Frequency of NPKS 25:5:5:5 Fertilizers on the Aluminium Content of Black Tea (ppm)

Yearly frequency of application	*Rate of Nitrogen (kg N/ha/year)*					*Mean frequency*
	50	*100*	*200*	*400*	*800*	
Once	655	508	383	374	412	467
Twice	483	417	324	402	425	410
Four times	478	318	359	411	328	379
Mean rate	539	414	355	396	388	—
C.V. %		4·80				
LSD *P*		0·05	0·01	0·001		
Rate		25	34	45		
Frequency		19	26	35		
Rate × Frequency		43	58	78		

TABLE 5
Effects of Rates and Sources of NPKS Fertilizers on Aluminium Content of Tea from Clone S 15/10

Type of nitrogen	*Rate of Nitrogen (kg N/ha/year)*						*Mean type of nitrogen*
	0	*100*	*150*	*300*	*450*	*600*	
NPKS 25:5:5:5	893	761	763	717	692	655	739
NPK 20:10:10	873	551	574	416	480	415	552
Mean rate of nitrogen	883	634	669	567	586	534	—
CV%							
Nitrogen Rate			1·08				
Nitrogen Source			2·78				
LSD *P*			0·05	0·01	0·001		
Nitrogen Rate			18	26	37		
Nitrogen Source			13	18	25		
Nitrogen Rate × Source 31			45	63	NS		

ammonium ions in preference to nitrate ions (Ishigaki, 1974; Watson & Wettasinghe, 1982). Splitting the fertilizer application caused more reduction of aluminium uptake by the tea plant as reflected in black tea aluminium content. Thus non-split (single) application of nitrogenous fertilizer produced black teas with higher aluminium content. Significant interactions were also noted between rates and frequency of fertilizer application in black tea aluminium content. The NPKS 25:5:5:5 fertilizer caused the tea to have more aluminium than NPK 20:10:10 (Table 5). This was noted at all rates of application.

Earlier studies have indicated that aluminium enhances tea quality (Chanturiya, 1976; Edmonds & Gudnason, 1979; Chang & Gudnason, 1982). For the clonal teas used in this study (Table 2), no significant relationship was noted between tasters' evaluation with total aluminium in tea ($r = 0{\cdot}08$) or aluminium in infused liquors ($r = 0{\cdot}04$). Thus, there are factors causing quality differences in clonal teas other than aluminium (Owuor *et al.*, 1987*a*, *c*, 1988).

The presence of variable amounts of aluminium in different teas (Tables 2 to 4) causes a problem in the TF analysis by the Flavognost method (Hilton, 1973). In this method of TF analysis, tea that has been infused is cooled and the TF is partitioned into an isobutylmethylketone (IBMK) (4-methyl pentan-2-one) layer prior to complexing with Flavognost reagent. The total TF is measured as the amount partitioned into the IBMK layer. Chang and Gudnason (1982) demonstrated that TF, which has complexed with aluminium during infusion, dissolves in the water during infusion and is not extracted by organic solvents such as ethylacetate or IBMK. The complexation of TF and aluminium therefore causes inaccuracy in TF determination by the Flavognost method. The results of the study suggest that this inaccuracy will vary from clone to clone (Table 2) and with the grading of teas (Table 3) as the amount of aluminium extracted varies.

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