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# Chromium contamination in black tea and its transfer into tea brew

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#### Abstract

A survey was conducted in south India to find out the amount of chromium (Cr) present in marketable black tea. Before assessing the Cr content in black tea, the Cr content in the green shoots was assessed, in order to know the Cr contamination after completion of the manufacturing process. Samples of black tea manufactured by CTC (Crush, Tear and Curl) and Orthodox processes were collected and analysed for Cr content. The study revealed that the content of Cr was high in CTC-manufactured black tea, which involved the use of stainless steel rollers. A pilot scale experiment was carried out, using newly sharpened rollers, and the black tea samples prepared were analysed for Cr content. A similar study was carried out in a commercial factory and samples analysed for Cr for one month. The study confirmed that Cr content was positively correlated to the sharpening of CTC rollers used for manufacturing. © 2007 Published by Elsevier Ltd.

Keywords: Contamination; Black tea; Chromium; CTC rollers

### 1. Introduction

Tea (*Camellia sinensis*) is one of the world's most popular beverages. Since tea is known to contain several minerals, trace elements and antioxidants, it is considered a healthy beverage. It is known that heavy metal contamination in black tea could be caused by agricultural input, especially by the soil application of synthetic fertilizers, as well as foliar sprays. The toxicity of chromium (Cr) is well established, but in contrast to other toxic metals like cadmium, lead, mercury and aluminium, it has received little attention from agricultural scientists (Becquer, Quantin, Sicot, & Boudot, 2003; Shanker, Cervantes, Tavera, & Avudainayagam, 2005).

During the manufacturing of black tea by the crush, tear and curl (CTC) process rollers made of stainless steel are used. Chromium present (17%) in the stainless steel combines with atmospheric oxygen to form a thin, invisible layer of chrome-containing oxide, called the passive film.

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The size of the chromium atom and its oxides are similar, and therefore they neatly pack together on the surface of the metal, forming a stable layer only a few atoms thick. If the metal is cut or scratched, the passive film is disrupted and more oxide will be formed, which will cover the exposed surface, protecting it from oxidative corrosion (Burgan, 1993). In the present study, tea samples were collected from different parts of south India to determine their chromium contents. In addition, the extent of leaching of Cr from a miniature CTC unit was measured, in order to estimate the amount of chromium transferred to tea, due to the use of newly sharpened rollers in the commercial CTC tea factory. Also the transfer of chromium from black tea to tea infusion was measured. The results obtained are discussed.

### 2. Materials and methods

# 2.1. Survey

Samples of CTC and Orthodox processed teas were collected from different parts of south India to quantify the Cr present.

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### 2.2. Miniature factory trial

A pilot scale study was conducted in the CTC miniature manufacturing unit of UPASI Tea Research Institute, to assess the extent of leaching of Cr from newly sharpened rollers into black tea. For this purpose, CTC rollers were installed after sharpening and black tea was manufactured using tea shoots of the cultivar UPASI-9. Manufacturing was done on seven consecutive days after the installation of sharpened rollers. The drier mouth bulk black tea samples were collected for the analysis of Cr.

# 2.3. Commercial factory trial

Black tea samples were collected everyday continuously for one month from a commercial tea factory located in Valparai (Tamil Nadu, India), situated at an elevation of 1150 m above sea level. About 100 g drier mouth bulk tea sample was collected and packed in an aluminium foil-lined bag. Details of dates of sharpening of CTC rollers were also recorded for correlation with chromium content of tea.

In both the miniature and commercial factory trials the same type of stainless steel rollers were employed; the rollers used in the miniature factory trial were smaller than the rollers used in the commercial factory trials. Due to their continuous usage during the tea manufacturing process the sharpness of the rollers will decrease and the rollers are eventually replaced. During successive batches of manufacturing the rollers are washed with pressurized water.

# 2.4. Analysis of chromium by atomic absorption spectrometry

Black tea samples obtained from the above experiments were subjected to chromium analysis using standard procedure (AOAC, 2000). About 0.5 g of black tea sample were weighed and digested to complete dryness, using a mixture of concentrated nitric acid and concentrated hydrochloric acid (60 ml, 3:1, v/v). The dried residue was dissolved using deionised water and filtered through Whatman (No. 1) filter paper and made up to 25 ml. The blank solution was prepared in similar way without black tea.

A few black tea samples collected from the CTC factory were brewed. Brewing was performed with 2 g of black tea sample in 100 ml boiled water for 6 min (ISO 3103, 1990). The infused solution was made up to 100 ml in volumetric flask using hot water and analysed by atomic absorption spectrometry for chromium content. A standard addition method was carried out to enhance the accuracy. To the tea brew a standard Cr solution was added and analysed.

The Perkin–Elmer AAnalyst 800 Flame AAS instrument was used for Cr determination. The operating parameters for chromium was set as recommended by the manufacturer (Perkin–Elmer, 2000). Quantification of Cr was carried out in air/acetylene flame without background correction against a NIST-traceable standard reference material of Cr. The relative standard deviations were less than 3% for all observations.

# 3. Results and discussion

The Cr content in the collected green tea shoots was below the detection limit of 0.7 mg/kg. The chromium content in leaf grade of CTC-processed black tea samples from the Nilgiris region was found to be between 3.0 and 4.9 mg/ kg while in the dust grade, the content of chromium was higher (3.9–21.2 mg/kg). However in 90% of the samples Cr content was below 10 mg/kg. In Orthodox processed black tea samples, the chromium content was lower and ranged between 1 and 1.6 mg/kg (Table 1). Ferrara, Montesanoa, and Senatore (2001) reported that chromium in black tea samples from different countries were in the range of 17.9-115 mg/kg. Since the rollers used in CTC tea manufacturing are made of stainless steel, containing chromium (17% w/w), CTC-processed black teas contained higher level of Cr, whereas in Orthodox processed tea the Cr content was lower, as the machinery employed is made up of gun metal having only trace levels of chromium.

The pilot scale factory study on Cr showed that on the 1st and 2nd day after installation of newly sharpened rollers, the CTC black tea contained 18.2 mg/kg of Cr and on the two subsequent days the leaching of Cr from rollers came down to 7.5 and 7.8 mg/kg, respectively. On the 5th day, Cr content was found to be 11.4 mg/kg (Table 2). The variations in Cr content could be due to the formation of various thicknesses as of stable layers of Cr atoms exposed to atmospheric oxygen. The study confirmed that the higher content of Cr was caused by sharpening of the rollers.

The results from the commercial tea factory indicated an increase in the Cr content of black tea whenever newly sharpened rollers were installed. The present study revealed higher contents of Cr *viz.*, 6.27, 7.59, 7.36, 7.58 and 6.91 mg/kg, on the days of fixing the newly sharpened rollers. On other days the content of Cr was below 6.66 mg/kg (Table 3). This study also confirmed that the source of Cr was the newly sharpened CTC rollers.

Since the CTC rollers are made of stainless steel containing chromium, which requires periodical sharpening for proper cutting, damage to the invisible layer of chromecontaining passive film occurs. The damaged passive film

Table 1
Survey on chromium in black tea in south India

Region	Grade/type of tea	Chromium (mg/kg)	
Anamallais	CTC fanning and CTC dust	5.8-8.4	
High range	CTC dust	21.2	
High range	Orthodox broken	1.6	
Nilgiris	Orthodox broken	1.2	
Nilgiris	CTC leaf	3.0-4.9	
Nilgiris	Orthodox (organic)	1.0	
Central Travancore	CTC dust	6.5–7.9	
Wayanad	CTC dust	3.9	

Table 2

Chromium content in pilot plant CTC-manufactured black tea after installation of newly sharpened rollers

Day of manufacturing after sharpening rollers	Chromium (mg/kg) <sup>a</sup>	
lst	$18.2\pm0.59$	
2nd	$18.3\pm0.44$	
3rd	$7.5\pm0.23$	
4th	$7.8\pm0.16$	
5th	$11.4\pm0.25$	
6th	$8.6\pm0.39$	
7th	$2.7\pm0.06$	

<sup>a</sup> Values (mean  $\pm$  SD); mean of three replicate analyses.

Table 3

Chromium in black tea collected from a commercial CTC factory

Sampling days/day of sharpening of rollers	Chromium content (mg/kg) <sup>a</sup>
Roller sharpened on	
6th day	$6.27\pm0.12$
13th day	$7.59\pm0.19$
19th day	$7.36\pm0.22$
24th day	$7.58\pm0.12$
29th day	$6.91\pm0.36$
Subsequent sampling days	
lst	$3.25\pm0.09$
2nd	$3.48 \pm 0.11$
3rd	$3.77\pm0.19$
4th	$2.75\pm0.39$
5th	$2.95\pm0.09$
7th	$3.40\pm0.15$
8th	$3.56\pm0.26$
9th	$4.27\pm0.51$
10th	$4.61\pm0.37$
11th	$4.85\pm0.24$
12th	$4.36\pm0.17$
14th	$5.97 \pm 0.39$
15th	$5.77\pm0.09$
16th	$5.22 \pm 0.22$
17th	$5.45\pm0.05$
18th	$4.90\pm0.11$
20th	$5.19 \pm 0.25$
21st	$5.34 \pm 0.06$
22nd	$5.23 \pm 0.07$
23rd	$4.76\pm0.16$
25th	$5.73\pm0.37$
26th	$5.29\pm0.16$
27th	$5.63 \pm 0.24$
28th	$5.70\pm0.05$
30th	$6.66\pm0.19$
21 of	$6.17 \pm 0.22$

 $^{\rm a}$  Values (mean  $\pm$  SD); mean of three replicate analyses.

releases chromium into tea while rolling, due to the contact of tea leaves with the metal surface of the roller. Although a considerable portion of moisture is removed during this stage of processing, tea leaves remain wet and the extruded viscous juice increases the chances of various contaminants adhering to the leaf surfaces. Since the mechanical damage and the resulting oxide formation is not uniform, neither is the transfer of Cr into black tea. Immediately after sharpening the Cr content in the black tea was obviously more (Table 3), due to Cr contamination from the metal surfaces

Table 4			
Transfer of chromi	um from b	lack tea to b	orew (infusion)

Cr in black tea <sup>a</sup> (mg/kg)	Cr in tea brew <sup>a</sup> (mg/kg) after 6 min brewing	% of transfer
$2.95 \pm 0.09$	$0.04\pm0.005$	1.4
$3.48\pm0.11$	$0.08\pm0.006$	2.3
$3.56\pm0.26$	$0.09\pm0.01$	2.5
$4.36\pm0.17$	$0.13\pm0.03$	3.0
$4.61\pm0.37$	$0.15\pm0.009$	3.3
$5.19\pm0.25$	$0.17\pm0.01$	3.3
$5.22\pm0.22$	$0.23\pm0.04$	4.4
$5.23\pm0.07$	$0.20\pm0.07$	3.8
$6.91 \pm 0.36$	$0.35\pm0.05$	5.1
$7.36\pm0.22$	$0.40\pm0.11$	5.4
$7.58\pm0.12$	$0.42\pm0.09$	5.5
$7.59\pm0.19$	$0.42\pm0.10$	5.5

<sup>a</sup> Values (mean  $\pm$  SD) mean of three replicate analyses.

of the machine. Potentially, the most contaminating parts of the machinery were the CTC rollers.

### 3.1. Chromium in tea brew

The results of the studies on transfer of Cr from black tea to brew are summarised in Table 4. The content of Cr in the black tea samples, varied between 2.95 and 7.59 mg/kg. Tea infusion contained very low amounts of Cr, between 0.04 and 0.42 mg/kg when brewed for 6 min. Natesan and Ranganathan (1990) reported that 16.5% of Cr present in black tea was transferred into tea brew when the brewing time was 1 min and when the brewing time was increased to 5 min, the percentage transfer of Cr was 42.2. Tascloglu and Kok (1998) reported that the transfer of Cr into tea brew from black tea was higher than from green tea. In the present study the transfer of Cr to the brew was positively correlated with the level of Cr in the black tea and the brewing time. The chemical compositions of the infusions of black tea and green tea are different from one another. In the leaf material (black or green tea), most of the metals are present in complexes and are coordinated by ligands. Brewing may cause partial dissolution of such complexes. The nature of the metal complexes determines the rate of transfer of metals into infusions. Accordingly, transfer of metals from leaves to infusion is variable and depends upon several parameters including brewing time, temperature, pH, etc. (Michie & Dixon, 1977).

### 4. Conclusions

In the present investigation, it was found that CTC-processed black tea contained higher levels of Cr than Orthodox processed black tea. Since tea leaves come into contact with the CTC rollers during the crushing, tearing and curling process, Cr leached into the "dhool" during manufacturing. Factory level study also confirmed that Cr contamination could happen due to sharpening of CTC rollers. Since there is no limit prescribed for chromium in tea, the data generated could be used for fixing a tolerance limit. Tea factories adopting CTC-type manufacturing are advised to periodically check the chromium content in black tea.

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