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Assessment of heavy metals in tobacco of cigarettes commonly sold in Ethiopia

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A B S T R A C T

A significant flux of heavy metals, among other toxins, reaches the lungs through smoking. This study reports Cd, Pb, Cu and Zn contents in tobacco of 11 brands of cigarette commonly sold in Ethiopia. The heavy metals were determined by atomic absorption spectrophotometry after wet digestion of cigarette tobacco using HNO_3 and H_2O_2. The concentration of trace metals in the cigarettes ranged (mean) (µg/g), Cd: 1.3–7.6 (2.48±0.32), Pb: 0.50–12.50 (6.24±2.2), Cu: 2.89–25.35 (13.70±4.12) and Zn: 24.40–62.55 (36.22±7.50) while Ni was not detected in all the eleven brands of cigarettes. Comparable levels of trace metals were obtained in the tobacco of both imported and Ethiopian cigarettes. The average trace metal contents of cigarettes available in Ethiopia were Cd 1.82±0.39, Pb 4.23±0.97, Cu 10.2±3.1 and Zn 28.2±7.8 µg/cigarette and a person who smokes 20 cigarettes per day is estimated to increase his/her daily Cd, Pb, Cu and Zn retention by approximately 0.036, 0.085, 0.204, 0.564 mg/day, respectively. The results indicate that smoking and exposure to cigarette smoke is a serious problem to be taken into account when carrying out epidemiological studies on human exposure to trace metals.

Capsule Summary: The heavy metals (Cd, Pb, Cu and Zn) contents in tobacco of eleven brands of cigarette commonly sold in Ethiopia were determined by flame atomic absorption spectrophotometry and results revealed a significant amount of heavy metals in cigarettes and variable among different brands.

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INTRODUCTION

Metals are vital for a huge number of physiological processes in the human body, but can also destroy health when the concentration is not within the physiologically favorable range. Heavy metals are important environmental pollutants and many of them are toxic even at very low concentrations (Memon et al., 2001). Even essential trace elements can cause damage to health or even death at increased concentrations. The form in which an element is ingested also plays a major role in its restorability or toxicity (El Bialy et al., 2005).

Cigarette is made up of tobacco, paper and additives. As much as 600–1400 additives are used in cigarette manufacture, with many of these additives containing environmental contamination and exposure to heavy metals such as mercury, cadmium and lead is a serious growing problem throughout the world (Nnorom et al., 2005). Heavy metals can directly influence behaviour by impairing mental and neurological function, influencing neurotransmitter production and utilization, and altering numerous metabolic body processes (Hu et al., 2003; Dauwe et al., 2004). Systems in which toxic metals can induce impairment and dysfunction include the blood and cardiovascular, eliminative pathways (colon, liver, kidneys, skin), endocrine (hormonal), energy production pathways, enzymatic, gastrointestinal, immune, nervous (central and peripheral). This was attributed to the higher transfer rates of the added ingredients to smoke (Mumtaz et al., 1999). In addition to occupational exposure tobacco smoke is a potential source of some toxic trace elements including inorganic carcinogens (Chang et al., 2005). Only the tobacco companies know exactly what goes into each cigarette, and only some of this information is available.

A wide range of toxic metals is found in tobacco, depending largely on the soil content where the tobacco plant is grown. The use of fertilizers has been blamed for high concentrations of arsenic, mercury, lead, cadmium, chromium, nickel, and selenium in tobacco. Tobacco plant is amenable to absorb and accumulate heavy metal species from the soil into its leaves (Zhang et al., 2005). Cigarette smoke contains particles and gases generated by the combustion of its various components at high temperature. More than 4000 compounds have been identified in environmental tobacco smoke. The cigarette smoke can be inhaled directly by the smoker and non-smokers in cigarette-contaminated environment through passive smoking (DiCarlantonio and Talbot, 1999).

There are several studies reported in the literature on heavy metal contents in the cigarettes sold and smoked in different parts of the world. Vincent et al. (2011) have reported a comparative evaluation and toxicity assessment of heavy metals (Pb, Cd, Ni, Cr and Zn) in commonly smoked cigarette brands and local tobacco snuff purchased and consumed in Nigeria. Pourkhabbaz and Pourkhabbaz (2012) have investigated the levels of toxic metals (Cd, Cu, Co Ni, Zn and Pb) in the tobacco of different Iranian cigarette brands and related health issues. Ashraf (2012) has reported levels of Cd and Pb in popular cigarette brands sold and/or produced in Saudi Arabia. Level of heavy metals (Cr, Cd and Pb) in selected cigarettes and tobacco leaves in Benue State, Nigeria has been reported by Eneji et al. (2013). Sebiawu et al. (2014) have reported the analysis of heavy metals (As, Pb, Cu, Fe, Zn, Mn, Cd, Ni, Cr and Se) content of tobacco and cigarettes sold in Wa Municipality of Upper West Region, Ghana. Caruso et al. (2014) have determined the toxic metal concentrations (As, Cd, Cr, Ni and Pb) in cigarettes obtained from U.S. smokers in 2009 under the umbrella of International Tobacco Control United States Survey Cohort. Regassa and Chandravanshi (2016) have reported the levels of heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) in the raw and processed Ethiopian tobacco leaves.

In Ethiopia, there are imported and locally manufactured cigarettes with unknown metal contents. The different brands of cigarettes sold in Ethiopia are Nyala, Gisela, Marlboro Light (local), and Rothmans, Marlboro Red, Benson, Winston, Kents, London, Gold seal, Ronson and Ghamdan (imported). However, there is no study reported in the literature on the levels of heavy metals in tobacco of different cigarettes sold in Ethiopia. Hence it is worth to determine the contents of toxic heavy metals in cigarettes sold in Ethiopia.

The objectives of this study were (i) to determine the levels of heavy metals in different brands of cigarettes sold in Ethiopia, (ii) to compare the levels of heavy metals in the Ethiopian cigarettes with that of imported cigarettes sold in Ethiopia, and (iii) to compare the levels of heavy metals in the cigarettes sold in Ethiopia with literature reports on cigarettes sold or smoked around the world.

MATERIAL AND METHODS

Instruments

An atomic absorption spectrometer (Buck model 210VGP, East Norwalk, USA) equipped with deuterium ark background correctors and air-C_2H_2 flame was used for the determination of heavy metals (Pb, Cd, Cu and Zn).

Standards and reagents

Chemicals and reagents used for the analysis were of analytical reagent grade. Nitric acid (69-72%, Loba Chemie, India), hydrochloric acid (35-38%, Loba Chemie, India), and hydrogen peroxide (30%, ADWIC, Egypt) were used for the digestion of cigarettes tobacco samples. Stock standard solutions of the metals (Pb, Cd, Cu and Zn), 1000 mg/L calibration standards (Buck Scientific, USA), prepared as nitrates for each element in 2% HNO_3, were used for the preparation of calibration curves for the determination of metals in the samples. Distilled-deionized water was used for preparation of standard solutions, and dilution.

Sample collection

Eleven brands of cigarette samples, which are sold in Ethiopia, were bought from the retailers' shops from different places in Addis Ababa, Bahir Dar and Shewa Robit, Ethiopia. Three packs of a particular cigarette were bought from three different places for the purpose of random sampling. The cigarette brands were: Nyala, Gisela and Marlboro Light (local), and Rothmans, Marlboro Red, Benson, Winston, Kents, Gold seal, Ronson and Ghamdan (imported). The description of cigarette samples is given in Table 1.

Sample preparation

The average weight of each cigarette tobacco was determined by weighing 5 sticks of each brand after removing the filter and paper. The mean weight of each cigarette tobacco was calculated. Composites of each brand were prepared by removing the papers and filters from 60 cigarettes of 3 packs. The samples were dried in an oven at a temperature of 80 °C for 6 hours and allowed to cool in a desiccator. The dried tobacco was ground in a mortar with a pestle until powdered finely as much as possible for homogenization, to simplify weighing and to facilitate organic matter destruction (digestion).

Sample digestion

The digestion procedure of Campbell and Plank (1998) was used. 0.50 g of dried, ground, and homogenized cigarette tobacco was transferred to a 100 mL flat bottom flask. 5.0 mL of concentrated HNO_3 acid was added and the flask was covered with watch glass and allowed to stand overnight. The covered flask was placed on a hot plate with temperature controller and heated at 200 ^oC for 30 min. The flask was removed and allowed to cool and 2 mL of 30% H_2O_2 was added and digested at the same temperature and time in the same way. This step was repeated to ensure complete digestion. The digest was allowed to dry to 1 to 2 mL at 150 ^oC. Then 5.0 mL of 1% HNO_3 was added to digest residue and filtered quantitatively through Whatman filter paper into a 25 mL volumetric flask and made up to volume with deionized water. This was subsequently analyzed for

Table 1: Description of cigarette samples

No.	Cigarette brand	Place of sample collection	Country of origin (manufacturer)
1	Nyala	Addis Ababa and Bahir Dar	Ethiopia
2	Gisela	Shewa Robit	Ethiopia
3	Ghamdane	Addis Ababa	Yemen
4	Benson	Addis Ababa and Bahir Dar	U.K.
5	Rothmans	Addis Ababa and Bahir Dar	U.K.
6	Kent	Addis Ababa and Bahir Dar	U.S.A.
7	Gold Seal	Addis Ababa	U.K.
8	Marlboro Light	Addis Ababa and Bahir Dar	U.S.A.
9	Marlboro Red	Addis Ababa and Bahir Dar	U.S.A.
10	Ronson	Addis Ababa	U.K.
11	Winston	Addis Ababa	U.S.A.

Table 2: Analytical wavelength, linear range, correlation coefficient and detection limit for the determination of heavy metals (Cd, Pb, Cu and Zn) by flame atomic absorption spectrometry

No.	Heavy metals	Wavelength (nm)	Linear range (μg/mL)	Correlation coefficient (r)	Detection limit in the digest (μg/mL)	Detection limit in the tobacco (μg/g)
1	Cd	228.80	0.01 – 2	0.9998	0.02	1.0
2	Pb	217.00	0.08 – 20	0.9997	0.03	1.5
3	Cu	217.89	0.005 – 50	0.9995	0.03	1.5
4	Zn	213.86	0.005 – 250	0.9996	0.04	2.0

Cd, Pb, Zn and Cu using flame atomic absorption spectrophotometer. Internal quality control with retests of cadmium, lead, nickel, copper and zinc standards prepared in 2% HNO_3 was undertaken. Digestion blanks were also prepared and results reported were average of triplicates.

RESULTS AND DISCUSSION

Calibration and limit of detection

Five points calibration curves was constructed for each metal. Analytical wavelength, linear range, correlation coefficient and detection limit for the determination of heavy metals (Cd, Pb, Cu and Zn) by flame atomic absorption spectrometry are given in Table 2. Good linearity with correlation coefficients r > 0.999 was obtained. Method detection limits (MDLs) of each metal were also determined. Limits of detection of the analysed metals were determined as thrice the standard deviation (3σ) of the blank determinations by FAAS from the mean of six replicate analyses. The detection limits (≤ 2 μg/g) were low enough to detect the heavy metals at trace level in the cigarette tobacco.

Method validation

The validation of the method was evaluated by spiking the standard stock solution of different volume of each metal to the cigarette tobacco samples in triplicate. The spiked samples were digested and analyzed by the same procedure as that of unspiked samples. The results of determinations showed good recoveries (91–106%).

Levels of heavy metals

The weight of the cigarette tobacco (after removing the papers and filters) varies depending on the length of the cigarette and other factors. The average weight of the cigarettes without the filter and paper were in the range 0.66

Table 3: Trace metal contents (mean ± SD) (μg/g, dry weight) of Ethiopian brands of cigarette tobacco

No.	Cigarette brand	Cd	Pb	Zn	Cu
1	Nyala	1.55 ± 0.15	5.52 ± 0.50	26.30 ± 1.75	19.75 ± 1.04
2	Gisela	1.30 ± 0.00	5.10 ± 0.50	62.55 ± 4.50	2.80 ± 0.55
	Mean	1.43 ± 0.15	5.31 ± 0.71	44.43 ± 4.83	11.28 ± 1.8

Table 4: Trace metal contents (mean ± SD) (μg/g, dry weight) of some imported brands of cigarette tobacco

No.	Cigarette brand	Cd	Pb	Zn	Cu
1	Marlboro Light	2.43 ± 0.10	10.78 ± 0.98	39.50 ± 3.35	16.75 ± 1.25
2	Benson	2.40 ± 0.01	6.10 ± 0.51	27.75 ± 2.70	13.30 ± 0.20
3	Rothmans	7.60 ± 0.05	5.96 ± 0.91	38.70 ± 0.60	6.45 ± 0.01
4	Kent	2.00 ± 0.20	Trace	31.15 ± 0.55	10.35 ± 2.50
5	Gold Seal	2.30 ± 0.00	12.50 ± 0.60	26.60 ± 1.70	25.35 ± 0.55
6	Ghamdane	1.65 ± 0.05	0.50 ± 0.00	24.40 ± 2.50	12.55 ± 2.10
7	Marlboro Red	1.54 ± 0.05	6.00 ± 1.00	34.95 ± 3.20	21.15 ± 1.75
8	Ronson	2.38 ± 0.15	7.50 ± 1.00	37.90 ± 0.95	7.50 ± 0.05
9	Winston	2.28 ± 0.05	2.40 ± 0.50	55.40 ± 3.55	14.70 ± 0.01
	Mean	2.73 ± 0.29	5.75 ± 2.16	35.15 ± 7.22	14.23 ± 3.95

Table 5: Comparison between the results of present study and available reported levels of Cd, Pb, Zn and Cu in cigarette tobaccos in the literature

Heavy metals	This study Mean ± SD (μg/g) Range (μg/g)	Literature value (mean or range) (μg/g)	Country of origin	References
Cd	2.49 ± 0.32 1.30–7.60	1.27 2.3	Nigerian France	(Nnorom et al., 2005)
		0.5–0.8 (0.6 ± 0.1) 1.1–6.1 (3 ± 1.6)	Real (U.K.) Counterfeit/Forged (U.K.)	(Edrydstephens and Calder, 2005)
		0.02-3.6	Nigeria	(Vincent et al., 2011)
		2.71 1.76–3.20	Iran	(Pourkhabbaz and Pourkhabbaz, 2012)
		1.81	Saudi Arabia	(Ashraf, 2012)
		0.012	Nigeria	(Eneji et al. 2013)
		0.86	U.S.A.	(Caruso et al., 2014)
		1.8 1.0-4.1	Ghana	(Sebiawu et al., 2014)
Pb	6.07 ± 2.2 0.50–12.50	0.5–10	Indian	(Shaikh et al., 2002)
		0.3–0.5 (0.4 ± 0.1) 1.3–10.3 (4.1 ± 2.6)	Genuine/Real (U.K.) Counterfeit/Forged (U.K.)	(Edrydstephens and Calder, 2005)
		0.12–3.1	Nigeria	(Vincent et al., 2011)
		2.07 1.05–3.10	Iran	(Pourkhabbaz and Pourkhabbaz, 2012)
		2.46	Saudi Arabia	(Ashraf, 2012)
		0.038	Nigeria	(Eneji et al. 2013)
		0.44	U.S.A.	(Caruso et al., 2014)
		5.82 1.53–8.3	Ghana	(Sebiawu et al., 2014)
Cu	12.70 ± 4.12 2.80–25.35	20–50	Indian	(Shaikh et al., 2002)
		11.7–16.2 (13.0±1.7) 11.8 – 42.2 (16.7± 5.2)	Genuine/Real (U.K.) Counterfeit/Forged (U.K.)	(Edrydstephens and Calder, 2005)
		6.02–15.9	Nigeria	(Vincent et al., 2011)
		9.7 5.18–17.6	Iran	(Pourkhabbaz and Pourkhabbaz, 2012)
		14.53 10.2–21.8	Ghana	(Sebiawu et al., 2014)
Zn	36.22 ± 7.50 24.40–62.55	5.0–100	Indian	(Shaikh et al., 2002)
		26–40 (32 ± 4) 33–100 (49 ± 15)	Genuine (U.K.) Counterfeit/Forged (U.K.)	(Edrydstephens and Calder, 2005)
		7.30-24.0	Nigeria	(Vincent et al., 2011)
		27 18.1–42.2	Iran	(Pourkhabbaz and Pourkhabbaz, 2012)
		127 49.2–277	Ghana	(Sebiawu et al., 2014)

± 0.01 (Ronson and Marlboro Light) to 0.98 ± 0.05 g (Gissla). The levels of heavy metals in the Ethiopian and imported cigarettes are given in Table 3 and 4, respectively.

A comparison of metals (Cd, Pb, Cu and Zn) contents of cigarettes studied in various countries and the results of the present study are given in Table 5. Most results of this study are in good agreement with the literature results. The concentration of trace metals in the cigarettes ranged, Cd: 1.3 to 7.6 µg/g with mean 2.49 ± 0.32 µg/g, Pb: 0.50 to 12.50 µg/g with mean 6.24 ± 2.20 µg/g, Cu: 2.80 to 25.35 µg/g with mean 13.70 ± 4.12 µg/g, and Zn: 24.40 to 62.55 µg/g with mean 36.22 ± 7.50 µg/g. There is no significant difference between the determined values and literature values as it is seen in Table 5. However, there is a significant variation of Cd in Rothmans and mean values of Pb from literature values. The average trace metal contents of cigarettes sold in Ethiopia are Cd: 1.82 ± 0.39 µg/cigarette (range 1.27 – 5.4 µg/cigarette, Pb: 4.23 ± 2.34 µg/cigarette (range trace – 10.21 µg/cigarette), Cu.: 10.22 ± 3.15 µg/cigarette (range 2.74 – 20.50 µg/cigarette) Zn: 28.18 ± 7.81 µg/cigarette (range 18.79 - 61.30 µg/cigarette). The mean Pb content of Ethiopian cigarettes is comparable with imported brands. Ethiopian cigarettes show higher Zn content than imported cigarettes.

Imported cigarettes show higher Cd and Cu contents than Ethiopian cigarettes. It is likely that the major source of trace metals in tobacco leaves and cigarettes probably result from the widespread use of chemical fertilizers (Stephen and Calder, 2003). According to Nnorom et al. (2005), the mean metal contents of cigarettes varied markedly depending on the area of production.

However it has not been possible to obtain any evidence to suggest that the differences are related to the area of production or the extent of industrial development of the area. Nnorom et al. (2005) has reported that some species of plant have been observed to accumulate high concentrations of some metals, especially Cd, in leaf tissue rather than in roots. The processing, packaging and other technological processes (including the use of additives) used to bring raw items to the consumer can significantly increase heavy metal contents in cigarette tobacco (Stephen and Calder, 2003). An International (WHO/UNEP) program for assessment of human exposure to heavy metals has reported higher levels of Cd in kidney cortex samples of smokers compared to non-smokers (Stephen and Calder, 2003). According to this study the recommended cigarette with low mean trace metal contents are Ghamdane and Kent.

In this study the level of As and Hg, which are commonly found in tobacco, were not determined. Because to determine these elements, another procedure is required, i.e. hydride generation for As and cold vapor generation for Hg which requires much more sophisticated instruments than the direct determination of elements in single aliquots using FAAS.

CONCLUSIONS

This study showed that the levels of Cd, Pb, Cu and Zn in cigarettes sold in Ethiopia compares well with levels in cigarettes from other parts of the world. However, the level of Pb and Zn are slightly higher in cigarettes sold in Ethiopia than in cigarettes from other parts of the world. Nickel was not detected in any of the eleven brands of the cigarette consumed in Ethiopia. The results also showed the presence of significant amounts of the two toxic heavy metals Cd and Pb in the cigarette sold in Ethiopia. Thus the smokers in Ethiopia have higher intake of Cd and Pb than the non-smokers. Hence efforts should be made by the concerned organization at discouraging consumption of cigarettes.

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