Chapter

Investigation of Toxic Metals in the Tobacco of Pakistani Cigarettes Using Proton-Induced X-Ray Emission

Iram Mahmood, Sadaqat Khan, Waheed Akram, Raphael M. Obodo, Tariq Mehmood, Ishaq Ahmad and Ting-Kai Zhao

Abstract

A particle-induced X-ray emission (PIXE) study has been carried out to find out whether available local and imported cigarette brands in Pakistan have elevated concentration of metals or not. The results are compared within the brands examined in this study and with the results of related studies in literature. A sum of 19 different cigarette brands was purchased randomly from different Pakistani markets which included local and imported brands. The concentration of elements like Cd, Pb, Zn, Fe, Mn, Ni, Cu, and Co was investigated. Results showed that different cigarette brands have different metal contents. The mean concentration of the heavy metals is Cd—4.92 μg/g, Co—0.12 μg/g, Cu—0.97 μg/g, Ni—0.13 μg/g, Pb—1.02 μg/g, and Zn—12.91 μg/g per dry weight. Compared with the reported results of other international studies, Pakistani cigarettes are observed to have lower heavy metal contents except for cadmium which was higher. This study will provide adequate data for all concerned departments. This study will also create awareness among people about the toxicity of metals present in tobacco of cigarettes.

Keywords: toxic metals, PIXE, tobacco, human health, Pakistan

1. Introduction

Smoking of different tobacco products is increasing rapidly throughout the world. Smoking causes many health problems. Cigarette consumption is one of the major reasons for mortality in the world. During the tobacco plantation, various kinds of pesticides, fungicides and herbicide are used to cope with the different diseases and parasites [1]. Galazyn-Sidorczuk et al. [2] also stated that tobacco plant has a high tendency to uptake metals from soils and accumulate them in leaves. Tobacco plants can normally accumulate metals like Pb, Cd, Ni, Zn, and Cu that comes in cigarettes which are variable in the concentration of metals in soil and applied chemicals; tobacco plant have different levels of metals in different countries. Due to these reasons, tobacco is contaminated with different toxic metals and chemical compounds. Many studies reported that tobacco smoke is toxic, carcinogenic, and genotoxic. Cigarette smoke contains 4000 identified toxic chemical compounds that are potentially harmful to human health [1].

Various metallic and nonmetallic elements and heavy metals like lead, cadmium, mercury, antimony, etc. are present in tobacco. Biological samples from the bodies of people who use cigarette have been identified with a higher concentration of these elements than nonsmokers [2–4]. Lead is carcinogenic to humans and probably a major reason for cancer in human belonging to group 1 or group 2 [5]. Tobacco is a key source of lead present in the body of the indirect smokers (children and adolescents) in the United State. Children who live with one or more smokers contain 14–24% higher level of lead in blood than those who live with nonsmokers [6]. Beside it, in addition to other toxic elements, 87 organic carcinogens are produced by tobacco smoke is being inhaled into the lungs [7]. Some of the toxic metals like cadmium, nickel, and lead frequently move along with inhaled smoke to the blood that is usually deposited in various organs like the liver and kidney [8]. Excessive intake of toxic elements or essential element deficiency disturbs the homeostatic control which causes chronic physiological disorders such as hypertension, rheumatoid arthritis, and heart diseases [9]. Lead potentially harms the nervous system, brain, and red blood cells in human. In an estimate, a person inhales 1–5 μg lead per day by the smoking 20 cigarettes [10–13].

Including Pakistan, tobacco smoking is a general practice all over the world, especially in young or adult men and women. Due to the high health risk of tobacco smoking, it is necessary to monitor the elevated level of metals in cigarettes. Thereby, in the current study, we investigate 19 different brands of cigarettes (local and imported) to make fresh data and measure the heavy metals (e.g., lead, nickel, cobalt, copper, zinc, and cadmium). The results are compared to the cigarette brands used in the current study and also with the results of other studies that addressed the elevated level of metals in cigarettes worldwide.

2. Ion beam for material analysis techniques

Recently, analysis using ion beam became an order of modern analytical techniques by the use of MeV ion beams to study the elemental compositions. During bombardment, interactions of the ion with matter results in elastic and inelastic scattering, nuclear reaction, and excitation of the electromagnetic waves. Below is a demonstration of the typical techniques (**Figure 1**).

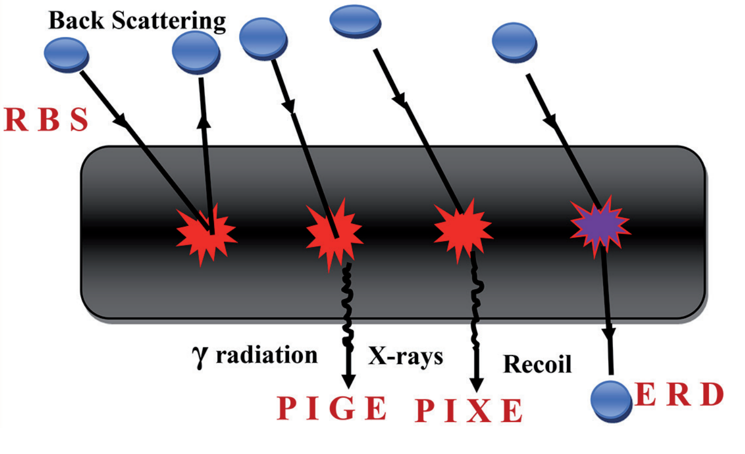

Figure 1.
Ion beam analysis techniques.

2.1 Rutherford backscattering spectrometry (RBS)

Rutherford backscattering spectrometry (RBS) is one of most frequently used ion beam analysis. It is used to examine the elemental composition with depth profiling of samples by measuring the energy of an elastically backscattered ion beam, which depends on the mass of the targeted sample and on the penetrating distance, which the back scattering occurred (**Figure 2**).

2.2 Elastic recoil detection analysis (ERDA)

Elastic recoil detection analysis (ERDA), known as forward recoil scattering can be used to examine the light elemental concentration with depth profile in a thin film. In this technique, light element such as hydrogen could be recoiled by heavy element in the forward direction and detect (**Figure 3**).

2.3 Particle-induced X-ray emission (PIXE)

Particle-induced X-ray emission or proton-induced X-ray emission (PIXE) is a technique used in determining the elemental content of a sample from Na to heavier

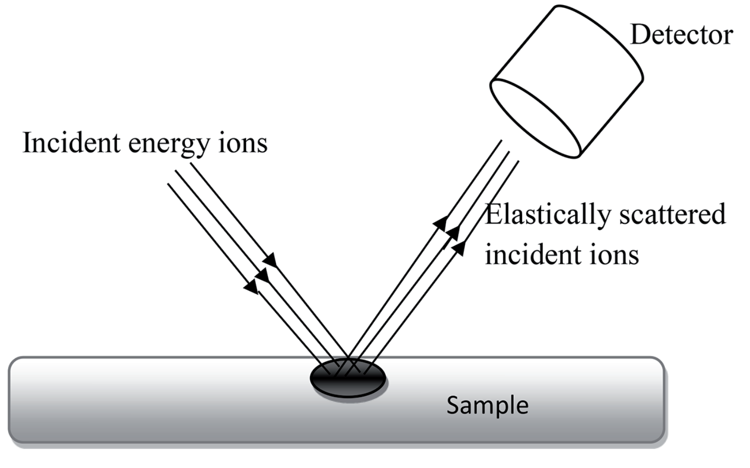

Figure 2.
Schematic of RBS setup.

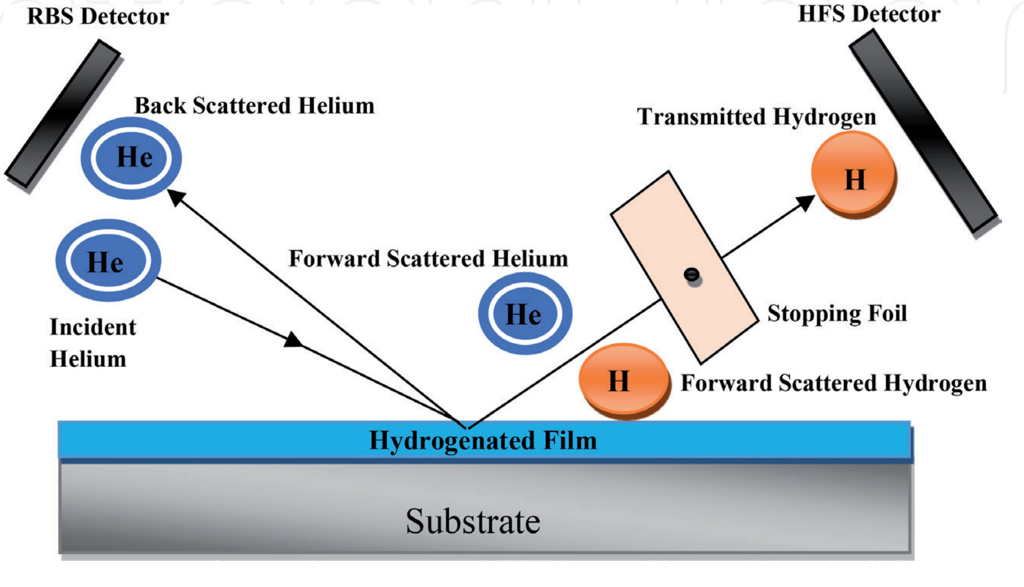

Figure 3.
Typical ERDA setup.

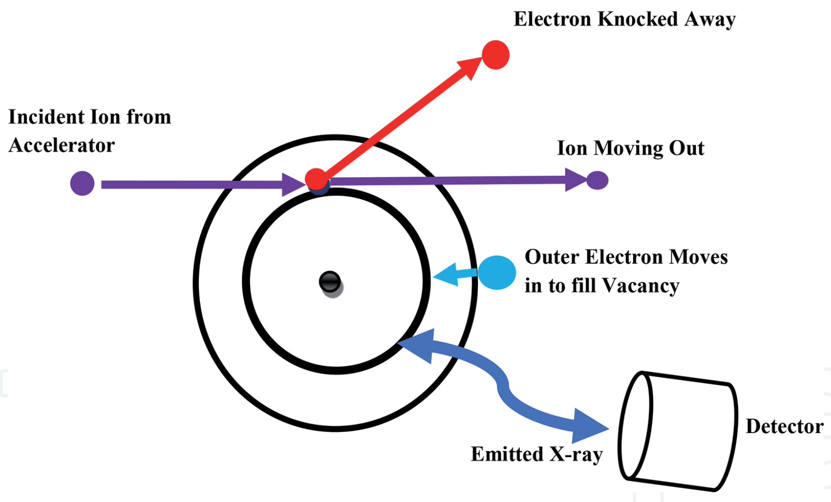

Figure 4.
Schematic of PIXE setup.

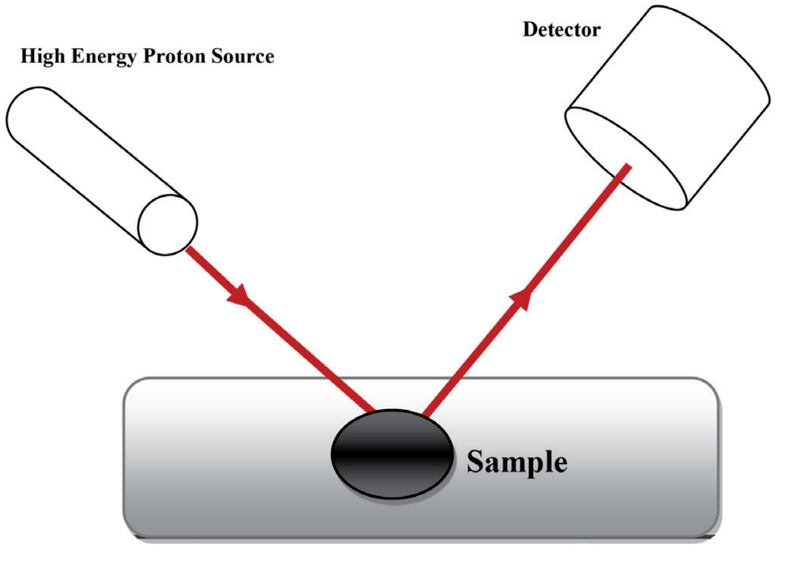

Figure 5.
Typical NRA setup.

elements. These incident beam particles (usually protons) expel internal shell electrons from the target atoms, which results in the emission of characteristic X-rays (**Figure 4**).

2.4 Nuclear reaction analysis (NRA)

Nuclear reaction analysis (NRA) involves the study of sample by irradiating them with select projectile nuclei with very-high-energy ions to induce nuclear reactions in the target nuclei. Targeted samples undergo a nuclear reaction under resonance conditions for a sharply defined resonance energy. The reaction product is usually a nucleus in an excited state, which immediately decays, emitting ionizing radiation that can be analyzed and interpreted (**Figure 5**).

3. Experiment detail

3.1 Sample preparation of tobacco

A sum of 19 different commonly sold cigarette brands (locally manufactured (8) and imported (11)) were randomly purchased from the market of Islamabad (**Table 3**).

Two cigarettes were taken from a pack of each brand, and then tobacco was separated from paper and filter. The samples of tobacco were air dried for the removal of moisture in the covered container. Each sample was placed in polyethylene vials to prevent the contamination. A fine homogeneous powder of dried samples was obtained by grinding in mortar and pestle. Pellets of 7 mm diameter and 2 mm thickness of this fine powder were produced by using a tabletop hydraulic press (pressure 120 kg/cm^2). The pellets were placed in desiccators. Then samples were put into 5 MV Pelletron Tandem Accelerator for analysis. The particle-induced X-ray emission (PIXE) was used for this investigation of toxic metals. The PIXE is a very effective and reliable technique for multi-elemental analysis of materials. The GUPIXWIN and Excel software were used for results and calculations. The standard reference material of 1515 apple leaves from NIST of the USA was used for calibration and analytical quality control.

3.2 PIXE analysis

With the approach of atomic-based analytical strategies in the previous 40 years, proton-induced X-ray emission (PIXE) has set up a part of the advanced elemental investigation of various materials [14]. PIXE is a technique with a diverse array of applications in biology, geology, materials science, and others.

The pelletized samples were irradiated with a 3 MeV proton beam from the 5 MV Pelletron Tandem accelerator installed at Experimental Physics Lab, National Centre for Physics, Islamabad. A standard reference material, NIST 1515 (apple leaves, National Institute of Standards and Technology, USA), was taken as the analytical quality control. The analytical outcomes concurred well with the standard qualities (**Table 1**), confirming the steady quality of the analytical outcomes in this work. The collimated proton beam was of 2 mm diameter. Mylar, "funny" filter having 100 μm thick, was used during the measurements, and this reduced the count

Elements	Determined values	Certified values
S	1557.8 ± 194	1800
Cl	545.9 ± 62	579 ± 23
K	13468.6 ± 2254	16,100 ± 1200
Ca	12,013.1 ± 1689	15,260 ± 1150
Cr	0.32 ± 0.057	0.3
Mn	52.9 ± 6.6	54 ± 3
Fe	78.5 ± 16.4	83 ± 5
Co	0.07 ± 0.053	0.09
Ni	0.76 ± 0.23	0.91 ± 0.12
Cu	6.2 ± 2.02	5.6 ± 0.24
Zn	15 ± 2.3	12.5 ± 0.3
Sr	22 ± 5.1	25 ± 2
Cd	0.04 ± 0.007	0.013 ± 0.002
Sb	0.019 ± 0.008	0.013
Hg	0.14 ± 0.15	0.044 ± 0.004
Pb	0.61 ± 0.19	0.57 ± 0.024

Table 1.
Analytical results for NIST 1515 (μg/g).

rate ensuring a less than 10% dead time at beam currents of 2–5 nA. No apparent damage in samples was observed after irradiation. A 30 mm^2 Si(Li) detector with an energy resolution of 138 eV (FWHM) at 5:9 keV of Mn was used to detect the emitted X-rays. The PIXE data was analyzed using the computer code GUPIXWIN.

4. Results and discussion

The quantitative analysis of tobacco of various imported and local cigarettes, purchased in Pakistan, was performed with the help of PIXE. The consequences of standard reference material for tobacco were in great concurrence with the certified values for compound components given in **Table 1**. In this study concentration of chemical elements including copper, lead, cadmium, ferric, manganese, zinc, nickel, sulfur, etc. was analyzed. **Figures 6–8** show the typical PIXE spectrum of NIST 1515 apple leaf and different tobacco samples obtained by using 5 MV Pelletron Tandem accelerator of the National Centre for Physics, Islamabad.

The toxic elements obtained from tobacco of different brands exhibited a large elemental concentration fluctuation. The order of the concentration of metals in local brands is Fe>Mn >Zn >Cd >Pb >Cu >Ni >Co. And the order of concentration of metals in imported brands is as Fe >Mn>Zn>Cd>Co>Cu >Pb>Ni. The outcomes of toxic elements in the tobacco of cigarettes were denoted as mean ± standard deviation as shown in **Tables 2** and **3.**

Lead is a very toxic metal even in very small concentration. Smoking is a key source of lead inhaled by human; thereby, it is more important to eradicate its contribution to overall lead load in humans. The past implementation of various policies has a successfully reduced lead level in the environment especially in the case of reduction of lead emissions origination from petrol, which has been reduced currently by utilization of unleaded petrol. It was estimated a sum of 50% total lead taken up by humans comes from petrol-originated emissions besides its ingestion of lead through the food chain is also important to be investigated [15]. It is widely admitted that cigarette contains about 1.2 μg lead, and approximately 6% passes over to mainstream smoke [7]. The current study showed the average concentration of lead in the tobacco of local cigarettes was 1.02 μg/g. The minimum mean concentration of lead was observed in the tobacco of Thrill which was 0.6 μg/g, and maximum concentration was 1.55 μg/g in the tobacco of Gold Leaf Special (**Table 2**). In the imported

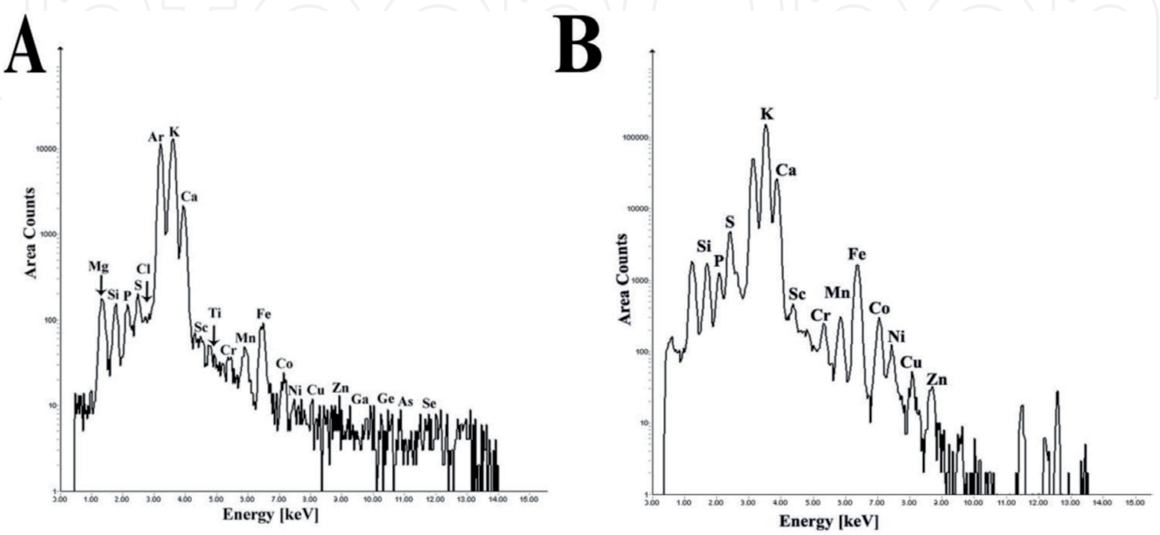

Figure 6.
PIXE spectrum of (A) NIST 1515 apple leaf and (B) Benson & hedges tobacco samples obtained by using 5 MV Pelletron Tandem accelerator of the National Centre for Physics, Islamabad.

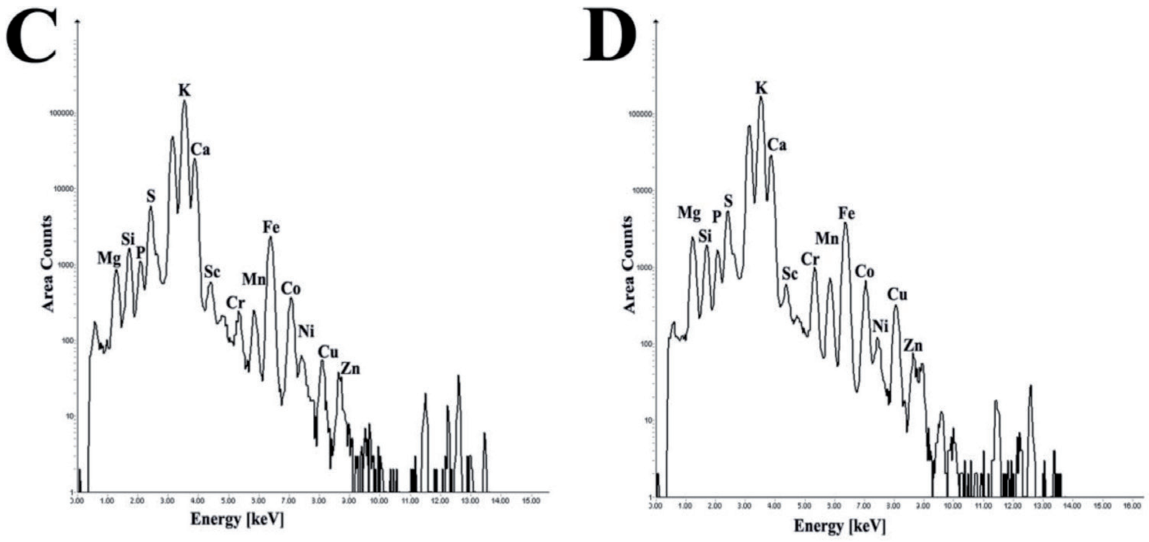

Figure 7.
PIXE spectrum of (C) Dunhill and (D) Marlboro tobacco samples obtained by using 5 MV Pelletron Tandem accelerator.

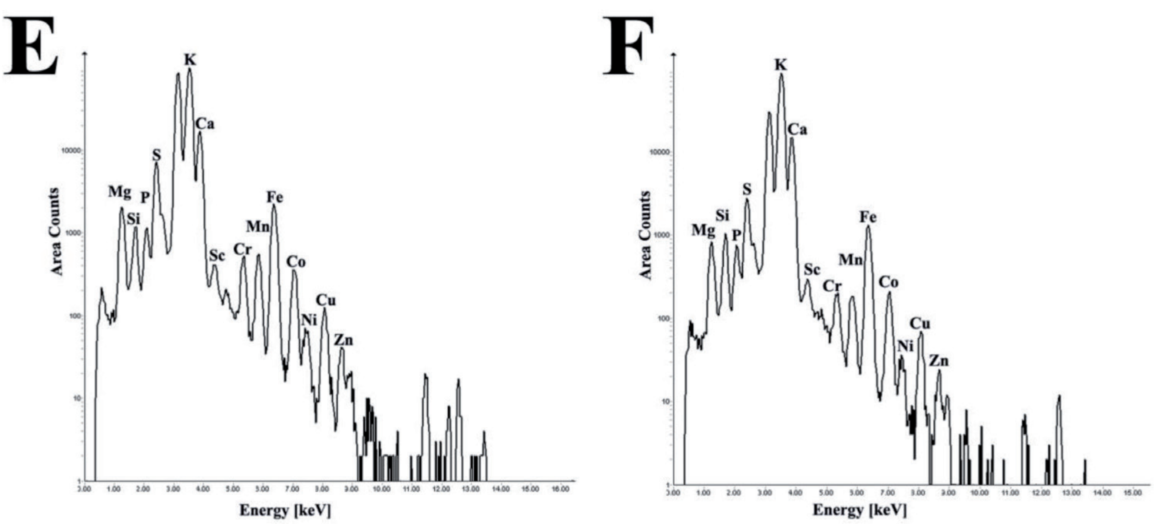

Figure 8.
PIXE spectrum of (E) gold leaf and (F) Capstan tobacco samples obtained by using 5 MV Pelletron Tandem accelerator.

cigarettes, the average concentration of lead present in the tobacco was 1.08 μg/g. The minimum concentration of lead that was observed in tobacco of imported brands was 0.67 μg/g, and maximum concentration was 1.3 μg/g (**Table 3**). Regarding the health issues, Mortada et al. [16] reported 101.6 + 30.9 μg/l and 143.7 + 33.8 blood lead levels in non-smoker and smokers, respectively. On the other hand, lead serum levels reported by Satarug et al. [17] in nonsmokers and smokers are 4.2 + 5.4 μg/l and 9.0 + 12, respectively. The variation of lead concentration between serum and blood is due to the fact that lead in the circulation is chiefly associated with erythrocytes [15]. The elimination of lead is a slow process carried out by urine resultantly; it accumulates in the skeleton. Lead is considered to be impermeable for blood and brain barriers, but the children are highly affected by neurotoxicity of lead as it accumulates in the brain and central nervous system that cause neurological disorder and mental retardation [18]. Besides it, lead accumulates in the blood of children by passive smoking due to the smoking habit of their parents. Hence protection of children from both type of smoking (i.e., active and passive) is a matter of great concern. Furthermore, peripheral arterial diseases, hypertension [19], and cataract

Brand name	Mean ± SD									
	Cd	Pb	Cu	Ni	Mn	Zn	Fe	Cr	Cl	Ca
Gold Leaf Special	3.55±5.02	1.55±0.35	0.33±0.04	1.03±0.24	47.3±0.99	14.4±1.13	175.1±4.7	0.28±0.1	3357.85±207.39	24217±1483.37
Marlboro	4.85±0.07	1.05±0.08	1.1±0.0	0.5±0.06	85.75±12.5	16.1±3.96	92.95±3.3	0.19±0.0	3473. 9±414.22	11743.2±238.01
Benson & Hedges	5.55±1.91	1.3±0.14	0.36±0.06	0.46±0.06	46.45±6.72	13.85±0.21	73.35±1.8	0.07±0.0	2932.15±566.75	19102.1±30.41
Press	6.1±5.09	0.93±0.52	0.41±0.07	0.15±0.04	18.95±2.05	9.25±1.2	52.55±6.7	0.05±0.0	4201.1±3762.23	14812.7±3042.54
Red & White	6.45±.0.35	1.15±0.07	0.61±0.07	0.2±0.01	36.7±1.84	11.45±0.78	100.52±1.3	0.08±0.0	3514.9±151.6	18578±361.33
Tender	9.4±0.28	0.68±0.12	0.22±0.03	0.08±0.02	19.1±1.41	7.3±0.57	50.85±2.3	0.04±0.0	6641.4±86.13	18565.95±844.36
Bond Street	1.86±2.61	0.65±0.92	5.71±7.48	0.3±0.23	77.65±24.7	24.35±22.3	47.55±67	29.03±41	3046.95±130.32	15270.7±819.96
Thrill	3.7±0.14	0.6±0.09	0.26±0.07	0.1±0.01	16.55±1.34	9.3±2.55	50.4±1.3	0.04±0.0	2193.95±118.44	11987.4±288.92
Gold Flake	3±0.14	1.05±0.07	0.38±0.05	0.18±0.0	25.05±1.34	9.75±0.07	78.3±6.8	0.08±0.0	2125.35±159.88	13843.85±102.88
Gold Flake Style	3.95±0.92	0.89±0.04	0.17±0.06	0.13±0.04	23.4±1.84	8.1±0.85	70.35±0.6	0.06±0.0	2006.15±118.16	13235.65±133.29
Capstan	6.05±1.06	1.35±0.35	0.58±0.09	0.18±0.02	31.95±2.33	10.6±3.82	101.9±1	0.07±0.0	3033.7±231.37	17944.05±485.15
Diplomat	6.4±0	0.95±0.07	0.25±0.09	0.18±0.01	36.2±2.55	11.4±1.13	92.7±3.5	0.06±0.0	4303.15±380.07	17061±866.35
Dunhill	5.3±0.14	1.35±0.07	2.9±0.14	0.76±0.07	86.8±8.34	28.55±10.7	162.45±7.6	0.36±0.0	2903±194.6	21253.8±625.51
Gold Leaf	3.45±0.07	0.82±0.02	0.81±0.26	0.23±0.05	25.1±0.0	8.2±0.57	60.9±5.7	0.08±0.0	1528.65±181.66	10615.05±260
Morven Gold	4.2±0.14	1±0.29	0.39±0.05	0.21±0.06	34.05±1.63	11.1±0.14	106.9±8.3	0.04±0.0	3838.9±34.51	16580.6±813.03
Mean values	4.9206667	1.02134	0.96534	0.31267	40.73334	12.91334	87.78467	2.03533	3251.488	16320.74

Table 2.
Content (μg/g) of metals in tobacco of local cigarette brands.

are including in expected consequences of lead accumulation [20]. The concentration of lead in tobacco of cigarette measured at various places around the world is shown in **Table 4**. Among the 12 places shown in **Table 4**, there are nine places where the concentration of lead in tobacco of cigarette is higher as compared to this study.

Cigarette smoke has been widely studied for cadmium which is claimed as a key source of cadmium inhaled by a human. Many studies addressed the elevated level of cadmium in cigarette and cigarette smoke with the mean cadmium concentration lies 0.5–1.5 mg per cigarette [15, 7]. In earth's crust, cadmium can be found in higher concentration usually combined with zinc. Usually cadmium is found as a by-product in copper, zinc, and lead extraction industries. Cadmium is also present

Brand names	Benson & Hedges	Dunhill	Marlboro	More	Mean values
S	2522.05±17.18	1700.6±108.97	1931.6±44.26	1948.7±10.82	2025.76
Cl	6413.2±837.07	3434.9±171.05	2429.5±188.44	2145±53.74	3605.67
K	12301.5±474.68	10217.6±71.56	14042.1±553.88	14911.05±979.84	12868.09
Ca	12167±666.8	11179.05±299.32	10629.05±556.14	14197.2±1339.97	12043.08
Mn	39.8±2.62	57.1±1.27	79.9±7.85	77.3±1.77	63.56
Fe	67.1±7.78	68.4±1.77	95.8±1.48	91±4.67	80.6
Ni	0.33±0.07	0.45±0.04	0.45±0.01	0.5±0.08	0.4325
Cu	2.3±0.57	1.1±0.0	0.75±0.13	0.41±0.04	1.14
Zn	15±0.14	14.4±0.99	15.8±0.57	18.8±2.12	16
Sr	13.3±0.64	10.9±0.71	12.35±1.77	8.8±12.52	11.36
Ru	0.33±0.05	0.13±0.03	0.05±0.08	0.18±0.0	0.17
Cd	5.8±0.64	4.8±0.28	4.65±0.78	4.2±1.27	4.87
Sb	0.19±0.0	0.12±0.05	0.15±0.02	0.1±0.14	0.14
Pb	1.3±0.07	1.1±0.14	0.67±0.17	1.3±0.0	1.08

Table 3.
Metal contents (μg/g) in the tobacco samples of imported cigarette brands.

Copper	Pb	Cu	Co	Ni	Zn	
4.92	1.02	0.97	0.12	0.13	12.91	Present study
2.71	2.07	9.7	4.42	17.93	27.02	Iran [28]
0.9	0.74	13	–	–	31.9	UK [29]
1.02	1.35	7.73	–	–	38.5	Korea [29]
0.4	1.6	18	0.91	3.6	29	India [30]
0.18	0.64	4.13	–	2.23	–	China [31]
1.7	1.02	2.45	–	0.22	–	Turkey [32]
1.95	1.2	9.7	–	2.4	49.8	Germany [33]
2.64	2.67	12.9	–	–	55.62	Jordan [34]
0.5	14.53	7.89	–	–	8.57	Pakistan [27]
0.9	4.3	39	–	3	39.5	India [35]
0.45	1.94	14	–	8.79	27	India [36]
2.48	6.07	12.70	–	–	36.22	Ethiopia [37]

Table 4.
Comparison of metal content (μg/g) in cigarettes of present study with international studies.

in manures and many pesticides, so it becomes easily a part of the environment after their application [21].

The local cigarettes had an average concentration of cadmium in tobacco which was 4.92 μg/g with standard deviation of 1.2. The minimum mean concentration of cadmium in a sample was 1.86 μg/g, and maximum concentration in a sample was 9.4 μg/g. In the imported cigarettes, the mean concentration of cadmium present in the tobacco was 4.88 μg/g with standard deviation of 0.74 (**Table 2**). The minimum concentration of cadmium observed in tobacco of imported brands was 4.2 μg/g, and maximum concentration was 5.85 μg/g (**Table 3**). The concentration of cadmium in this study is at the highest level as compared to other studies around the world (**Table 4**). The lowest cadmium concentration was observed in tobacco of Indian cigarettes which was 0.4 μg/g (**Table 4**).

Cadmium inhaled in its oxidized form as cadmium oxide while smoking. It is roughly estimated that 10% of cadmium deposit in lungs, and about 20–50% become a part of circulation [15, 7]. Cadmium accumulates in the circulation as well as deposits in kidney mainly in the cortex of the kidney by the late reaction of cadmium and metallothioneins. Although smoking generates small amount of copper which is unable to cause kidney failure, many studies stated that copper accumulates in kidney and is the main cause of renal end-stage failure and tubular dysfunction [17, 15, 18]. Many of other health disorders included emphysema, cataract, hypertension, as well as cardiovascular disease are also under investigation to know possible consequences of copper accumulation in these diseases [19, 20].

Copper mainly comes in air from fossil fuel burning and remains in the air for a long time. Usually, copper settles down in soil due to rain where it becomes bioavailable to plants. Naturally, copper comes from the soil through weathering of parent material, decaying of natural vegetation, forest fire, dust, windblown, and sea spray. Copper is also released into the environment by anthropogenic activities mainly by mining, metal production, wood production, and phosphate fertilizer. Human health is potentially affected by the soluble copper compounds which enter into the food chain through agricultural practices [21].

The concentration of copper in the tobacco of local and imported brands was observed, ranging from 0.17 to 5.71 μg/g with an average of 0.97 μg/g (**Table 2**) and from 0.41 to 2.3 μg/g with an average of 1.14 μg/g (**Table 3**), respectively. In this study, copper is at the lowest level with respect to other studies was done in different places of the world (given in **Table 4**). Long-term exposure to a higher level of copper causes decline in intelligence in young adolescents. Industrial exposure to copper fumes, dust, or mists generated by industries cause metal fumes fever and atopic retardation in nasal mucous membranes. Copper deposit in cornea and chronic copper toxicity causes various diseases like Wilson's disease, characterized by hepatic cirrhosis, renal disease, brain damage, and demyelination [22].

Nickel is present in low concentration in the environment and use in many things made by a human. Commonly nickel is used in steel and metal products as well as in jewelry [21]. The concentration of nickel in the tobacco of local and imported brands was observed, ranging from 0.08 to 1.03 μg/g with an average of 0.31 μg/g and from 0.33 to 0.5 μg/g with an average of 0.43 μg/g, respectively.

Nickel is a mutagen and carcinogen that causes many types of cancer in human especially related to the respiratory track. It induces sister chromatid exchanges by mutation [23]. Experiments showed the affected heart development in the unborn mice due to Nickel toxicity [24]. Although nickel is essential, but in excessive amount it is dangerous to health which causes sickness and enhance chances of various types of cancer like lung cancer, larynx cancer, prostate cancer, and nose cancer. Human exposure to nickel is usually through drinking water, breathing air, the food chain, and smoking of cigarettes [25]. The concentration of nickel measured in this study was at the second lowest position after concentration measured in tobacco of Turkey (0.22 μg/g), given in **Table 4**.

Zinc is present ubiquitously in nature with a variable concentration that mostly adds up to human activities. There are many anthropogenic sources of zinc are present in our surrounding especially in steel production, smelting, mining, and coal and waste combustion. In many countries, zinc present in soil with very high concentration is due to mining and refining of metals and use of sewage sludge as fertilizer [21]. The mean, minimum, and maximum concentrations of zinc in tobacco of local cigarette were 12.91, 7.3, and 28.55 μg/g, respectively. In the imported cigarettes, the average concentration of zinc present in the tobacco was 16.0 μg/g, the minimum concentration of zinc observed in the tobacco of imported brands was 14.4 μg/g and maximum concentration was 18.8 μg/g.

Too much zinc can cause a number of health problems, such as vomiting, nausea, anemia, skin irritations, and stomach cramps. A large quantity of zinc disturb the protein metabolism and cause arteriosclerosis or respiratory disorders that can damage the pancreas [26].

The concentration of zinc measured in this study was lower as compare to measure in other studies, referred in **Table 4** except the previous study on tobacco of Pakistani cigarettes (8.57 μg/g).

5. Conclusion

The available data on toxic metals in tobacco of Pakistani cigarettes was insufficient; this study will provide adequate data to all concerned departments. This study will also create awareness among people about the toxicity of metals present in tobacco of cigarettes.

Acknowledgements

The authors are thankful to the deanship of Scientific Research at King Khalid University and Abha Kingdom of Saudi Arabia for their technical and administrative support. The Higher Education Commission, Pakistan, is gratefully acknowledged. Plagiarism has been carried out via ID 1066213984 (similarity index 13%) in Turnitin software.

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