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Investigation of Toxic Metals in the Tobacco of Pakistani Cigarettes and Related Health Issues Using PIXE

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Abstract:. This study was carried out for the investigation of toxic metals in tobacco cigarettes available in Pakistan using Particle Induced X-ray emission (PIXE). A 5 MV Pelletron Tandem Accelerator at National Centre for Physics, Islamabad was used for this analysis. PIXE is well suited for rapid and quantitative analysis of large number of samples. Nineteen various cigarettes brands were randomly purchased from different Pakistani markets which included local and imported brands. The concentrations of elements like Cd, Pb, Zn, Fe, Mn, Ni, Cu and Co were investigated. Higher concentrations of these elements are toxic to human health, for example, cadmium can damage kidney and lungs and lead causes damage to the nervous system. It was found that different cigarettes brands have variation in concentrations of these elements. The results of this study were compared with other international studies.

Key words: Toxic Metals · PIXE · Tobacco · Human Health

INTRODUCTION

The smoking of different tobacco products is increasing rapidly throughout the world. Cigarette smoking is a health hazard and is one of the major causes of human mortality in the world. In tobacco growing different pesticides, herbicides and fungicides are used to control the various parasites and plant diseases. Due to these reasons tobacco is contaminated with different toxic metals and chemical compounds. The tobacco smoke has toxic, genotoxic and carcinogenic properties. Cigarette smoke contains 4000 identified chemical compounds which are toxic and harmful for human health [1].

The tobacco contains heavy metals like lead, cadmium, mercury, antimony etc and other metallic and non-metallic elements. It is observed that contents of these toxic elements in biological samples of human bodies of smokers are much more than nonsmokers [2-4]. Lead is classified as carcinogenic to humans or possibly carcinogenic to humans (group 1 or 2) [5]. Tobacco is considered to be an important source of lead in second hand smokers (children and adolescents) in the United States. It was observed that blood lead level in children was 14% and 24% higher who are living with 1 or more smokers than children living with nonsmokers [6]. In addition to other toxic elements, tobacco smoke transfers 87 organic carcinogens to lungs [7]. Plants can easily take up metals from soil and concentrated them in leaves. This contamination is different in every country in which tobacco plant is harvested and processed. Heavy metals like cadmium, nickel can pass to the blood via inhale smoke and even can be deposited in specific organs such as kidney and liver [8]. Lack of essential elements can result in loss of homeostatic control, or an excessive intake of toxic elements causes different disorder like hypertension, cardio-vascular diseases and rheumatoid arthritis [9]. Lead has the tendency of causing serious effects on brain, nervous system and red blood cells. It has been estimated that smoking of 20 cigarettes a day resulted in the inhalation of 1-5 µg lead [10-13].

In Pakistan, many young men and women enjoy smoking and this ratio is increasing day by day. It is necessary to know the levels of toxic metals in tobacco of cigarettes. In this investigation, 19 samples of different cigarettes brands both local and imported were randomly purchased from market to make fresh data and measure heavy metals (e.g. lead, nickel, cobalt, copper, zinc and cadmium). The acquired data of imported and local cigarettes are discussed and compared with each other and with the existing ones in different countries.

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MATERIALS AND METHODS

Preparation of Tobacco Samples: Nineteen different brands of commonly sold imported and locally manufactured cigarettes in Islamabad market were randomly purchased (Table 2 and 3). Two cigarettes were selected from each brand and then the tobacco was separated from paper and filter. The samples of tobacco were air-dried for the removal of moisture in a covered container. Each sample was placed in a polyethylene vial to prevent contamination. The dried samples were grinded into a fine powder using a mortar pestle. These Powdered samples were pressed into pellets of 7 mm diameter and 2 mm thickness using a table-top hydraulic press (pressure120kg/cm^2). The pellets were placed in desiccators. Then the samples were analysed using Particle Induced X-ray emission (PIXE) on a 5 MV Tandem pelletron Accelerator. PIXE is a very effective and reliable technique for multi-elemental analysis of materials. GUPIXWIN [Ref] and MS Excel software were used for quantitative analysis. The standard reference material 1515 apple leaves from NIST of USA were used for calibration and analytical quality control.

PIXE Analysis: With the advent of nuclear-based analytical methods in the last 40 years, Proton Induced X-ray emission (PIXE) has established a role in the elemental analysis of different 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. The diameter of the collimated proton beam was 2 mm. A 100 ìm thick Mylar, "funny" filter was used during the measurements and this reduced the count-rate to ensure a dead time of less than 10% at beam currents of 2–5 nA. Observation of the samples after irradiation showed no apparent damage. The emitted X-rays were detected using a 30 mm^2 Si(Li) detector with an energy resolution of 138 eV (FWHM) at 5:9 keV of Mn. The PIXE data was analyzed using the computer code GUPIXWIN. A standard reference material, NIST 1515 (Apple Leaves, National Institute of Standards and Technology, USA), was used for analytical quality control. The analytical results agreed well with the standard values (Table 1), confirming the reliability of the analytical results in this work.

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

Elements	Determined values	Certified Values
S	1557.8±194	1800
Cl	545.9±62	579±23
K	13468.6±2254	16100±1200
Ca	12013.1±1689	15260±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

RESULTS AND DISCUSSION

Toxic Elements in Tobacco of Cigarettes: The quantitative analysis of different local and imported tobacco cigarettes available in Pakistan was performed using PIXE. The results from standard reference material for tobacco were in good agreement with the certified values for chemical elements given in Table 1. In this study concentration of chemical elements including copper, lead, cadmium, ferric, manganese, zinc, nickel, sulphur, etc, were analyzed. The toxic elements obtained from different tobacco brands showed a wide variation with regard to concentration levels of these elements. The order of the concentration of metals in local brands is: Fe>Mn >Zn >Cd >Pb >Cu >Ni. And the order of concentration of metals in imported brands is: Fe >Mn>Zn>Cd>Cu >Pb>Ni. The concentrations of toxic elements in the tobacco of cigarettes were expressed as mean ±standard deviation as shown in Table 2 and Table 3.

Some metals are toxic in very small amount like lead. Smoking is not the only source of Pb uptake by humans, but the contribution of smoking to the total Pb load in humans has become important. The reason for this is the reduction of Pb emissions originating from petrol, which have been reduced in recent decades by the introduction of unleaded petrol. Approximately 50% of total Pb taken up by humans originates from petrol, but Pb ingestion via food is also important [15]. It is generally accepted that Pb content in cigarettes is around 1.2 µg and about 6% passes over to mainstream smoke, which is inhaled by smokers [7]. In this study, the average concentration of lead in the tobacco of local cigarettes was 1.02 µg/g.

Table 2: Concentration of elements in local cigarettes (µg/g)

Brands Name	Cd Mean± S.D	Pb Mean±S.D	Cu Mean±S.D	Ni Mean±S.D	Mn Mean±S.D	Zn Mean±S.D	Fe Mean±S.D
Gold leaf special	3.55±0.52	1.55±0.35	0.33±0.04	1.03±0.24	47.3±6.99	14.4±3.13	175.1±14.7
Marlboro	4.85±0.47	1.05±0.08	1.1±0.08	0.5±0.06	85.75±12.5	16.1±3.96	92.95±13.3
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±11.8
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±10.7
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±11.3
Tander	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±12.3
Bond street	1.86±2.61	0.65±0.92	5.71±7.48	0.3±0.03	77.65±24.7	24.35±22.3	47.55±6.7
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±10.3
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
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
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
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
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
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
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
Mean values	4.9206667	1.02134	0.96534	0.31267	40.73334	12.91334	87.78467

Table 3: Concentration of elements in imported cigarettes (µg/g)

Elements	Benson & Hedges	Dunhill	Marlbro	More	Mean Values
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
Cd	5.8±0.64	4.8±0.28	4.65±0.78	4.2±1.27	4.87
Pb	1.3±0.07	1.1±0.14	0.67±0.17	1.3±0.0	1.08

Table 4: Comparison of the result of studies of elements contents of cigarettes in various countries and results of the present study

Cd	Pb	Cu	Co	Ni	Zn	References
4.9	1.05	1.06	0.84	0.37	14.46	This 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 [31]
0.18	0.64	4.13		2.23		China [30]
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	12.7			36.22	Ethiopia [37]

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 cigarettes, the average concentration of lead present in the tobacco was 1.08µg/g. The minimum concentration of lead observed in tobacco of imported brands was 0.67µg/g and maximum concentration was 1.3µg/g (Table 3). Regarding health issues, Mortada *et al*. [16] reported that blood Pb levels in non-smokers is 101.6+30.9 µg/l and in smokers is 143.7+33.8, whereas Pb serum levels reported by Satarug *et al*. [17] in non-smokers and smokers are 4.2+5.4 µg/l and 9.0+12, respectively. The difference between blood and serum Pb concentrations is due to the fact that Pb in circulation is mainly bound to erythrocytes [15]. Pb is eliminated from the body via urine, but this occurs slowly and accumulation in the skeleton is observed. Although the blood brain barrier is relatively impermeable for Pb, children are at a high risk to accumulate the neurotoxic Pb in the brain and central nervous system, resulting in mental retardation and other neurological disorders [18]. In addition, children from parents who smoke accumulate high levels of blood lead via passive smoking. These facts are just one further example that stresses the urgent need to protect children from active as well as passive smoking. Other possible health consequences of lead accumulation are hypertension and peripheral arterial diseases [19], as well as cataract [20]. The concentrations of lead in tobacco of cigarette measured at various places around the world are shown in Table 4. Among the twelve places shown in Table 4, there are 9 places where the concentration of lead in tobacco of cigarette is higher as compare to this study.

Cadmium is the best studied metal from cigarette smoke and smoking is the main source of cadmium intake by humans. The content of cadmium in cigarettes and cigarette smoke was analysed in a number of studies. Although the Cd amounts varied, the average Cd content per cigarette lies between 0.5 and 1.5 mg per cigarette [15, 7]. Cadmium can be found in large quantities in the earth's crust. It can always occur in combination with zinc. Cadmium is also found in industries as an inevitable by-product of zinc, copper and lead extraction. After being applied it enters the environment mainly through the ground, because it is found in pesticides and manures [21].

The local cigarettes had average concentration of cadmium in tobacco of 4.92µg/g with standard deviation 1.2. The minimum mean concentration of cadmium in a sample was 1.86µg/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 4.88µg/g with standard deviation 0.74 (Table 2). The minimum concentration of cadmium observed in tobacco of imported brands was 4.2µg/g and maximum concentration was 5.85µg/g (Table 3). The concentration of cadmium in this study is at highest level as compared to the 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).

When the cigarette is smoked, Cd is transformed to cadmium oxide, which is then inhaled. Approximately 10% of the Cd is deposited in the lungs and 20–50% is transferred to the circulation [7,15]. Cadmium not only accumulates in the circulation but also in the kidneys, mainly in the kidney cortex, where metal-lothioneins chelate Cd and immobilize it. A number of studies have shown that this accumulation of Cd in kidneys causes tubular dysfunction and renal end stage failure, but there is also evidence that the amount of cadmium delivered by smoking is too small to cause kidney failure [17, 15, 18]. Also cadmium is suspected to cause cataract, emphysema, hypertension and cardiovascular disease [19, 20].

Copper enters the air through the combustion of fossil fuels. It will remain there for eminent period of time and it settles when it starts to rain. Then it will end up in the soils. So soils may also contain large quantity of copper. Copper can be released into the environment by natural sources such as decaying vegetation, forest fires, wind-blown dust and sea spray. Human activities also contribute to copper release such as mining, wood production, metal production and phosphate fertilizer. Soluble copper compounds form the major threat to the human health and occur in the environment after use in agriculture [21].

The presence of copper in the tobacco of local brands was observed, ranging from 0.17 µg/g to 5.71 µg/g with an average 0.97 µg/g (Table 2) and in the imported brands it ranged from 0.41 µg/g to 2.3 µg/g with average 1.14 µg/g (Table 3). In this study, copper is at the lowest level with respect to other studies done at different places of the world (Table 4). There is a link between long-term exposure to high concentrations of copper and a decline in intelligence in young adolescents. In addition, industrial exposure to copper fumes, dusts or mists may result in metal fumes fever with atrophic changes in the nasal mucous membranes. Chronic copper poisoning results in Wilson's disease, characterized by hepatic cirrhosis, brain damage, renal disease, demyelization and copper deposition in the cornea [22]

In the environment, nickel occurs at very low levels. Humans use nickel for many different application. The most common application is the use of Ni as an ingredient of steel and other metal products. It can also be found in jewelry [21]. The concentration of nickel in the tobacco of local brands ranged from 0.08 µg/g to 1.03 µg/g with an average 0.31 µg/g and the concentration of nickel in the tobacco of imported brands ranged from 0.33 µg/g to 0.5 µg/g with an average 0.43 µg/g. Nickel has been shown to cause a number of di?erent forms of cancer, especially of the respiratory tract. The main mechanism responsible for this activity is that Ni is mutagenic, [23] and has been reported to induce sister chromatide exchanges. In addition, there is evidence that Ni affects heart development in unborn mice [24]. Nickel is essential but when the uptake is too high it can be dangerous for health. Humans may be exposed to nickel through breathing air, drinking water, smoking cigarettes or eating food. An uptake of a large quantity of nickel has many effects such as higher chances of lung cancer, nose cancer, larynx cancer and prostate cancer and sickness [25]. The concentration of nickel measured in this study was at second lowest position after concentration measured in tobacco of Turkey (0.22 µg/g) given in Table 4.

Naturally, zinc occurs in air, water and soil. Its concentration is rising unnaturally, due to addition of zinc through human activities. Most zinc is added due to industrial activities, such as mining, coal and waste combustion and steel processing. Soils from some areas are heavily contaminated with zinc, these areas are included where zinc has to be mined or refined, or where sewage sludge from industrial areas has been used as fertilizer [21]. The local cigarettes had average concentration of zinc in tobacco of 12.91 µg/g, the minimum mean concentration of zinc in a sample was 7.3 µg/g and maximum concentration in a sample was 28.55 µg/g. In the imported cigarettes, the average concentration of zinc present in the tobacco was 16.0 µg/g and 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. Large quantities of zinc disturb the protein metabolism and cause arteriosclerosis, or respiratory disorders and can damage the pancreas [26]. The concentration of zinc measured in this study was lower as compared to that measured in other studies referred in Table 4 except previous study on tobacco of Pakistani cigarettes (8.57 µg/g).

CONCLUSION

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

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