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Research Article

Measurement of Uranium Concentrations in Tobacco Cigarettes Consumed by Iraqi Publics using Kinetic Phosphorescence Analyzer (KPA)

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Abstract

Tobacco contains minute quantities of radioactive isotopes of Uranium series, which is radioactive carcinogenic. Smoking of tobacco and its products increases the internal intake and radiation dose due to natural radionuclides. In a number of studies, inhalation of some naturally-occurring radionuclides via smoking has been considered to be one of the most significant causes of lung cancer. In this work, forty seven tobacco samples were collected from the Iraqi markets. Uranium in tobacco was measured using the Kinetic Phosphorescence Analyzer (KPA) techniques. The results demonstrated that Uranium concentrations ranged from 1.371±0.0018 to 3.424±0.0075µg/g, with an overall average about 2.224 µg/g. The annual effective dose due to inhalation for adults (smokers) for Uranium varied from 0.144 to 0.361 mSv/y with an overall average 0.234 mSv/y. The results illustrated that the German tobacco cigarettes have the highest concentration of Uranium compared to the cigarettes produced from other global origins and consumed in the Iraqi markets. Results showed that all tobacco samples are contained Uranium concentrations with varied rates.

Keywords*: Uranium Concentration, Annual Effective Dose, Tobacco Cigarettes, KPA.*

1. Introduction

Uranium is alithophilic element and is present in soils, sediments, plants and aqueous streams. Soils to a depth of 2900 km have a mean concentration of 0.76 µg of Uranium per gram of soil (Mauda, A. 1964). The two principal routes of human intake of Uranium are inhalation and ingestion. Inhalation of Uranium by non-occupationally exposed persons is primarily through re-suspended dust in the air (Singh, N. P., and Wrenn, M. E. 1989). The major fraction of the human total Uranium body burden is believed to occur by ingestion through dietary intake (Singh, N. P., and Wrenn, M. E. 1989;Fisenne, M. *et al* 1983). Uranium solubility depends on its chemical form but soluble Uranium is readily absorbed in the gut after intake and transferred to the blood (Wrenn, M. D.*et al* 1985). The distribution of uranium in the human body depends on the biological half-lives, which vary from tissue to tissue, as well as the rates of Uranium redistribution to specific tissue types, which also vary from tissue to tissue. However Uranium typically redistributes to and stays in the bones (ICRP 1978). In a number of studies inhalation of ^{210}Pb and ^{210}Po from both air and cigarette smoking is given as one of the most significant agents of lung cancer (Martell, E. A. 1987). Along with ^{210}Pb, ^{210}Po is produced as radioactive decay product of ^{226}Ra which is a long-lived member of the ^{238}U chain, as well as, the use of the phosphate fertilizers containing significant concentrations of Uranium. The radon gas, ^{222}Rn (T1/2 = 3.8 days) which has extreme mobility, is the first decay product of ^{226}Ra. Its rapid decay in the atmosphere generates ^{210}Po and ^{210}Pb that are adsorbed by aerosols and returned to earth as surface deposition or rainout (Watson, A. P. 1985). ^{210}Pb and ^{210}Po in tobacco are taken up directly either from the soil or by foliar absorption from the air. They are also generated in the plant after absorption of ^{226}Ra from soil and water (Radford, E. P. and Hunt, V. R. 1964). Tobacco curing and the combustion of trichomes in burning cigarettes produce insoluble particles of high ^{210}Pb activity that are inhaled and deposited in the bronchi of smokers. The subsequent growth of ^{210}Po results in high local-particle irradiation which may account for bronchial cancer among smokers (Martell, E. A. 1974). Tobacco is normally aged for 1-2 yr after being harvested before marketing. In this period, secular equilibrium between ^{210}Pb and ^{210}Po is attained. Nevertheless, Uranium contents have been determined in Indian tobacco samples by thermal neutron activation and lexan polycarbonate track detectors (ChEakarvarti, S. K*et al* 1979). Distribution of α-emitting radionuclides in cigarette filters has been determined using CA-85 plastic detectors (Lal, N. *et al* 1983). The effect of cigarette smoking inside a room on indoor radon daughter's concentration has been studied by thermal neutron activation and lexan polycarbonate detectors (Abu-Jarad, F. 1997). Radon and thoron alpha-activities have been evaluated inside different cigarette smokes of tobacco (Misdaq, M. A.,andFlata, K. 2003)

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using two different types of solid state nuclear track detectors (SSNTDs). In the present work, uranium concentrations have been measured in various tobacco samples for different origins from the world which are available in the Iraqi markets using kinetic phosphorescence analysis (KPA) which represents a very good technique for the determination of uranium concentrations with good resolution.

2. Experimental

Forty seven types of cigarettes have been measured against Uranium concentrations using KPA. Samples collected from Iraqi markets for most consuming kinds of cigarette. KPA provides direct detection of Uranium from the part-per-million to the sub-part-per- billion levels. Optimal ranges for the best precision were determined. These ranges were subsequently used during the statistical evaluation of the method during qualification testing. The main characteristics of the KPA system are given in the Table 1(Chemchek instruments 2006). KPA-11 uses pulsed laser excitation and gated detection for the determination of organic compounds such as poly aromatic hydrocarbons, nitrogen and sulfur heterocyclic, and several lanthanide elements and Uranium. KPA allows for two separate calibrations covering two calibration ranges. Uranium standard solutions were prepared using uranium octoxideU_3O_8. Firstly a stock standard solution of 1000 mg/l (1000 ppm) was prepared by dissolving 117.9 mg of U_3O_8 in 100 ml of 0.82 M nitric acid (HNO_3) in volumetric flask. To construct the calibration curve for KPA two different series of calibration standard were prepared to cover a wide range of Uranium concentrations. Uranium concentrations in the first series of standards were 0.05, 0.1, 0.5, 1, 2, 3, 5, 7, 9 and 10 µg/l, while that of the second series were 0.5, 1, 2, 3, 4, 5, 6 and 7 mg/l. These two sets of standards were used to construct the calibration curves for the low and high ranges of Uranium concentrations respectively.

The calibration curve for the high concentration of Uranium standard range will be used in the present work.
The procedure of samples preparation includes (Chemchek Instruments, INC. 2007):
1-The samples are dried to remove moisture in muffle furnace.
2-Break into smaller pieces by hand and macerate for about 30 seconds by pulsing in a large blender.
3-Remove from blender to a beaker.
4-Put ash in a muffle furnace at 350 °C for about 30 minutes and increase heat by about 40 °C to about 750 °C and hold temperature for about 2.5 hours.
5-Ash should be gray in color. If black color persists, continue muffle furnace treatment to remove residual carbon.
6-Weigh out about 5 grams on an ash-basis in a 250-ml Pyrex beaker.
7-Add 25 ml of concentrated HNO_3 and gradually add 25 ml of concentrated hydrochloric acid (HCl) with agitation.
8-Warm to 400-500 °C on a hot plate.
9-Add 30% deionized water (H_2O_2) in ½ ml, increments from a squeeze bottle. Approximately 5 ml of peroxide may be required to convert the pale yellow color to a clear liquid.

3. Results and Discussion

Tobacco smoking is a risk factor for six out of eight main death causes all over the world; with lung cancer being one of the six causes, tobacco represents the most important one (Zaga, V. *et al* 2011). Each year 1.35 million brand new case is diagnosed, which represents more than 12% of all the modern cancer cases (Zaga, V. *etal* 2011; Greenlee, R. T. *et al* 2000). Furthermore, smoking is responsible for 1.18 million deaths from cancer (17.6% of the world total), of which 21,400 are lung cancers from secondhand smoking. However, this work has investigated the Uranium content of tobacco samples of forty seven different brands (coded T1–T47) used in cigarettes manufactures. Table 2 shows data obtained for Uranium concentrations of tobacco samples of cigarettes origins in different countries from the world and available in Iraqi market. The presence of Uranium in tobacco means that indoor cigarette smoke is a source of Uranium and its decay series to the inhabitants, whether they smoke or not. The data obtained revealed that sample T43 recorded the highest concentration of Uranium, which is made in Germany, whereas the sample T26 contained the lowest concentration of Uranium, which is made in Switzerland. Results in Table 3 were classified by origin country to groups representing the tobacco factory for special kinds of cigarettes, accounting for Egypt (G1), European Union (G2), France (G3), Germany (G4), Jordan (G5), Korea (G6), Switzerland (G7), UAE (G8), and USA (G9). The results show that the set of G4 is the highest concentration of Uranium rate of 2.67±0.0595 µg/g, and a G7 is the least concentration of Uranium at a rate 1.88±0.012 µg/g. Also, from the results, one can be found that samples containing high concentrations of Uranium are brands with good taste and flavor and are heavily used in the Iraqi market. Because of the soil to grow tobacco is usually treated with chemical fertilizers, particularly with phosphate fertilizers to fulfill the requirement of nutrients like nitrogen, phosphorous and potassium. However, to get huge profit in tobacco plantation by the farmers, excessive use of chemical fertilizers has become a common practice causing an elevated level of radioactivity as these fertilizers are rich in Uranium and its decay products. Therefore, various studies have been conducted to estimate the collective and mean individual annual effective doses. UNSCEAR (1982) was evaluated the annual effective dose and found the value 0.16 mSv per year which based on the assumption that a smoker smoking 20 cigarettes per day and that 20% of ^{210}Po and ^{210}Pb are inhaled by the smoker. In this work, and to estimate the annual effective dose, we assumed 0.82 g of tobacco per cigarette and a smoker is smoking 30 cigarettes (one and a half packs) per day or 24.6 g of tobacco per day, and then the annual consumption of tobacco by cigarettes is estimated to be 8.985 kg/y. The fraction of the radionuclide activity concentration that is recovered from cigarette tobacco to cigarette smoke is 0.75, as on the average, about 75 % of the radioisotope in

the cigarette tobacco was contained in the cigarette smoke, which is partially inhaled and deposited in the lung tissues and about 25 % was retained in the cigarette filter and ash (Khater, A. E. 2004).

Table 1 The main characteristics of the KPA

Parameter	Laser KPA
Selectivity	Tunable for U, Eu, Sm. analysis
Laser source	Nitrogen laser of 337nm
Pulse duration	3 nsec.
Repetition rate	20 pulses/sec
Emission wavelength	515nm for U
Pulse power	120 µJ
Buffer	Uraplex
Sample volume	1 ml
MDL	0.01ng/ml (0.13mBq/l)
Precision (RSD)	1-3% at U> 0.01 ng/ml
	7-10% at U< 0.01 ng/ml
Analysis range	0.01 500,000 ng/ml
Data processing	Computer software

Then the data of Table 2 are derived for the annual effective dose, HE (Sv/ y), due to inhalation for adults (smokers), according to the equation:

$$H_E = 0.75 \times M_T \times Ci \times F \quad (1)$$

where M_T (kg/y) refers to the annual amount (in mass) of tobacco consumed, Ci (µg/g) refers to the concentration of the Uranium, and F (Sv/Bq) refers to the dose conversion factor (ICRP 1978). From the data of Table 2, it is shown that the annual effective dose due to Uranium varied from 0.144 to 0.361 mSv/y with an overall average about 0.234 mSv/y. The German tobacco models is the highest concentration gave the highest annual effective dose, while the Swiss tobacco models is the smallest annual effective dose. This dose must be compared with the average worldwide exposure to natural radiation sources 2.4 mSv/y and especially the part due to inhalation which is 1.26 mSv/ y (UNSCEAR 2000).

Conclusions

Tobacco is a source of natural radioactivity of Uranium. Tobacco smoking is fatal in many ways and has severe health, economic, and social consequences. Although natural radioactivity in tobacco could be one of the main reasons for the health impacts of tobacco smoking. This work aimed to shed more light on the concentrations of Uranium in cigarette tobacco that is consumed in Iraq. Based on the results obtained from this work, the concentrations of Uranium in forty seven tobacco samples showed that the highest concentration was observed in the sample T43 with 3.424±0.0075 µg/g, the lowest concentration was observed in the sample T26 with1.371±0.0018 µg/g, and the overall average about 2.224 µg/g. In the estimating the radiation effective dose induced from smoking, it was concluded that the annual effective dose to lungs due to inhalation for adults (smokers) averaged to 0.234 mSv/y for Uranium, which is

Table 2 Uranium Concentration and annual effective doses of smokers who smoke 30 cigarettes per day

Tobacco Samples	Uranium Concentration	Annual Effective Dose
	(µg/g)	H_E(mSv/y)
T1	1.62±0.011	0.171
T2	2.34±0.0028	0.247
T3	2.63±0.009	0.277
T4	2.71±0.023	0.286
T5	2.35±0.0042	0.248
T6	2.28±0.011	0.24
T7	2.69±0.026	0.283
T8	2.66±0.004	0.28
T9	1.83±0.0012	0.193
T10	1.99±0.015	0.21
T11	1.87±0.0043	0.197
T12	1.65±0.0013	0.174
T13	2.289±0.022	0.241
T14	2.19±0.015	0.231
T15	1.63±0.0014	0.172
T16	2.52±0.0014	0.266
T17	1.732±0.0034	0.183
T18	1.54±0.025	0.162
T19	1.85±0.031	0.195
T20	2.44±0.0021	0.257
T21	1.93±0.0045	0.203
T22	2.304±0.0042	0.243
T23	1.724±0.008	0.182
T24	2.44±0.009	0.257
T25	2.608±0.017	0.275
T26	1.371±0.0018	0.144
T27	2.608±0.006	0.275
T28	1.772±0.028	0.187
T29	2.68±0.0016	0.282
T30	2.45±0.007	0.258
T31	1.42±0.0045	0.15
T32	2.464±0.0039	0.26
T33	1.92±0.0054	0.202
T34	2.608±0.018	0.275
T35	1.86±0.012	0.196
T36	2.84±0.012	0.299
T37	1.78±0.0054	0.188
T38	1.432±0.008	0.151
T39	1.88±0.031	0.198
T40	2.456±0.0054	0.259
T41	2.44±0.018	0.257
T42	2.704±0.008	0.285
T43	3.424±0.0075	0.361
T44	2.12±0.0009	0.223
T45	3.172±0.027	0.334
T46	2.74±0.0054	0.289
T47	2.58±0.0054	0.272
Average	2.22±0.0102	0.234

representing a high level comparing to the effective doses happened from another reasons. The cigarette's flavored and good taste characteristic is that contain high concentrations of Uranium, and therefore, the annual effective dose will be more. The results showed that cigarettes produced in Germany, the highest concentration of Uranium from other countries producing cigarettes and

Table 3 Uranium concentrations in different tobacco samples classified according to the original country

Tobacco samples	Groups	Origin	Uranium Concentration (µg/g)	Average
T9	G1	Egypt	1.83±0.0012	2.06±0.0027
T22		Egypt	2.304±0.0042	
T28	G2	European Union	1.772±0.028	
T32		European Union	2.464±0.0039	
T37		European Union	1.78±0.0054	2.118±0.01
T40		European Union	2.456±0.0054	
T17	G3	France	1.732±0.0034	
T47		France	2.58±0.0054	2.156±0.0044
T3	G4	Germany	2.63±0.009	
T4		Germany	2.71±0.023	
T24		Germany	2.44±0.009	
T29		Germany	2.68±0.0016	
T30		Germany	2.45±0.007	2.67±0.0595
T33		Germany	1.92±0.0054	
T36		Germany	2.84±0.012	
T41		Germany	2.44±0.018	
T43		Germany	3.424±0.0075	
T45		Germany	3.172±0.027	
T2	G5	HKJ	2.34±0.0028	
T8		HKJ	2.66±0.004	2.33±0.0072
T10		HKJ	1.99±0.015	
T34	G6	Korea	2.608±0.018	
T38		Korea	1.432±0.008	2.26±0.0104
T46		Korea	2.74±0.0054	
T15	G7	Switzerland	1.63±0.0014	
T19		Switzerland	1.85±0.031	
T20		Switzerland	2.44±0.0021	
T25		Switzerland	2.608±0.017	1.88±0.012
T26		Switzerland	1.371±0.0018	
T31		Switzerland	1.42±0.0045	
T39		Switzerland	1.88±0.031	
T44	G8	UAE	2.12±0.0009	
T5		UAE	2.35±0.0042	
T18		UAE	1.54±0.025	2.068±0.0088
T23		UAE	1.724±0.008	
T27		UAE	2.608±0.006	
T1	G9	USA	1.62±0.011	
T6		USA	2.28±0.011	
T11		USA	1.87±0.0043	2.19±0.0084
T14		USA	2.19±0.015	
T16		USA	2.52±0.0014	
T42		USA	2.704±0.008	

consumed in the Iraqi market. Finally, we wish to emphasize the urgent needs for more research on the activity concentration of natural radionuclides in tobacco and tobacco products, their behavior during smoking, and on their concentration in smoke and smoker's intake.

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