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Elements of Natural Radioactive Decay Series In Iranian Mineral Water and Cigarettes

S Mohammadi

Physics Department, Payam Noor University, Mashad 91735, Iran
E-mail: Mohammadi@pnu.ac.ir

Abstract

The Uranium (^{238}U) decay series provides the most important isotopes of elements Radium (^{226}Ra), Radon (^{222}Rn) and Polonium (^{210}Po) with half-lives of 1600 years, 3.8 days and 140 days, respectively. Although the chemical structure of radium is very similar to calcium, the fact that it produces a radioactive gas (Radon) complicates its handling in the laboratory and natural environment.

The average concentrations of naturally occurring radio nuclide ^{226}Ra in drinking water at different parts of Iran were used to estimate the annual effective dose equivalent. In another research, concentrations of ^{210}Po in Iranian cigarettes were measured for the internal intake of this radionuclide and its concentration in the lung tissues. The results indicate that the average concentration of ^{226}Ra in drinking water is low compared to 100 mBq/l that is recommended by the World Health Organization while the average concentration of ^{210}Po and ^{210}Pb in Iranian cigarette is relatively high in comparison with other cigarettes found in the market.

Keywords: Uranium decay series – Mineral water – Cigarette - Radon

Introduction

Uranium is a naturally occurring primordial radionuclide; that is it was one of the elements created after the Big Bang. It is the last naturally occurring element in the periodic table. The decay products of uranium pass over 10 elements, all with very different chemical properties. These elements are transported by groundwater (migrate), the solute composition of which varies with the surrounding rock/soil minerals. The different elements migrate at different rates due to their different chemistries, dissolving in some areas and precipitating in others. In the ^{238}U decay

series 8 α-particles are emitted from ^{238}U to ^{206}Pb. Alpha particles cause extensive ionization in matter.

The uranium decay series provide the most important isotopes of elements radium, radon and polonium, which are all radioactive and alpha emitters. Although the chemistry of radium is relatively simple, the fact that it produces a radioactive gas (Radon) complicates its handling in the laboratory and natural environment. The decay of radon produces radioactive atoms of At, Po, Bi, and Pb.

The radioactive noble gas ^{222}Rn is produced continuously from the decay of radium in the ground. It dissolves in groundwater, which often carries it in high concentrations and releases it to areas inhabited by humans. It diffuses readily through soil and into the atmosphere. Thus it is always present in the air and water at levels, which are determined by local geology and meteorology. It imparts the highest organ dose to the lungs of any radioactive environmental contaminant.

The relatively high activity concentrations of ^{210}Po that are found in cigarette increase internal intakes of this radionuclide and its concentration in the lung tissues. That contributes to an increase in the internal radiation dose among smokers.

Natural Radioactivity from Drinking Water

All waters have impurities in them and there is no pure water. Successfully solving problems of monitoring, protection and rational use of water from rivers and mountains demands complex systematic study of laws of space-time distribution and migration of heavy elements such as ^{226}Ra. Radium (half life=1600 years) is a decay product of uranium and is generally in radioactive equilibrium with it on a global basis. It is much more soluble in water than uranium is, and thus is leached out of the soil by groundwater and makes its way to drinking water and food, and this route of exposure is important if high concentrations are found in drinking water.

The actual concentration of ^{226}Ra varies greatly in different locations. Radium in the body becomes incorporated in bone, where it remains virtually indefinitely. The protection standard for radium was originally based on a maximum level in the body (maximum body burden).

The average concentrations of naturally occurring radio nuclide ^{226}Ra in drinking water was used to estimate the annual effective dose equivalent. In this work, during June 2003 to June 2004, samples of mineral water from Northwest, Northeast, West, Center and South of Iran were gathered in winter and summer, and ^{226}Ra activity in each sample were determined.

Radium decays to radon, ^{222}Rn that is a gaseous intermediate. ^{222}Rn (half life= 3.8 days) is an inert noble gas and the immediate daughter product of ^{226}Ra. Natural radiation accounts for the majority of human exposure to radiation, and ^{222}Rn and its short-lived daughter products are the largest contributions to this radiation dose. Because of the radon health concern, several methods have been developed to monitor for radon and its daughters in air (Ziegelheim, et al . 1982).

^{226}Ra Determination Method

In this experiment, the activity of ^{226}Ra was measured by *radon emanation method* with a minimum detection limit of 2 mBq/l (Ziegelheim, et al . 1982).

Briefly, ^{226}Ra in drinking water sample is concentrated and separated by co precipitation on Barium Sulfate. The precipitate is dissolved in EDTA reagent, place in sealed bubbler and stored for ingrowths of ^{222}Rn. After ingrowths, the gas is purged into a scintillation cell. When the short-lived ^{222}Rn daughters are in equilibrium with the parent (4h), the scintillation cell is counted for alpha activity.

We used the following formula to calculate the annual effective dose of ^{226}Ra in drinking water:

$D = C_{Ra} \times U_a \times D_f$

Where C_{Ra} is concentration of ^{226}Ra in Becquerel per letter (Bq/l), U_a is annual intake of drinking water by adults (l/a), and D_f is conversion factor of radioactivity to absorbed dose (or dose equivalent per Bq) in μSv / Bq. The United Nations Scientific Committee on the Effects of Ionizing Radiation (UNSCEAR) recommendation of D_f for intake of ^{226}Ra is 0.28 μSv / Bq (UNSCEAR 2000).

Table 1 shows ^{226}Ra measurements in water for different parts of Iran. The results show that the ^{226}Ra concentration in investigated waters did not exceed the level of 100 mBq/l that is recommended by the World Health Organization (WHO). In the southern part of Iran, the concentration of ^{226}Ra is relatively high in comparison to other parts.

Table 1: Radioactivity of water for different parts of Iran.

Location	**Average ^{226}Ra concentration (mBq/l)**
North-West	2.1 ±0.1
North-East	2.6±0.2
Center	2.3±0.2
West	2.1 ±0.1
South[a]	6.2±0.7

(a) Water from wells.

Natural Radioactivity from Cigarette Smoke

The relatively high activity concentrations of ^{210}Po that are found in cigarettes increase internal intakes of this radionuclide and its concentration in the lung tissues.

Polonium is a reactive, silvery-gray metal that dissolves in dilute acids. It is fairly volatile and about half will evaporate within two days if kept at 55^{o} C. A gram capsule of Polonium will reach 500^{o} C because of the intense alpha-radiation (Emsley 2000). That might contribute significantly to an increase in the internal radiation dose among smokers.

In addition to ^{210}Po, ^{210}Pb with a half-life of 22 years is found in cigarettes as well. Since the burning temperature of a cigarette is around 700^{o} C and ^{210}Pb is supposed to volatilize at temperature over 500^{o} C, the percentage of ^{210}Pb effectively inhaled will be an additional source of ^{210}Po for the lungs. The annual effective doses were calculated on the basis of ^{210}Po and ^{210}Pb intake with the cigarette smoke.

All soils contain radium, a radioactive element that decays into ^{210}Pb and ^{210}Po. In addition, phosphate ore used to make fertilizers used on tobacco fields contains these isotopes in relatively high concentrations. While tobacco plants can absorb ^{210}Pb and ^{210}Po through their roots, relatively little enters this way. When ^{226}Ra, a component of soil and fertilizer decays, it gives to ^{222}Rn, a gas that escapes to the air. This radon decays into its daughters, which have high electric charges that make them, attach to dust particles. These dust particles sick to the tips or heads of the hairs of Tobacco leaves. Almost all the radioactivity of tobacco comes from this air deposition process. During processing of tobacco to cigarettes, this radioactivity is not removed from the tobacco leaves.

Equivalent doses resulting from single disintegration of ^{210}Po (alpha particle decay) are a thousand times greater than in the case of ^{210}Pb decay (beta particle decay) (Parfenov 1974). The concentration of ^{210}Po in cigarettes are in the range of 3-37 Bq/Kg and vary with cigarette brand, due to different varieties of tobacco and also manufacturing procedures (Skwarzec, et al. 2001).

^{210}Po and ^{210}Pb Determination Method

For this experiment we used the method suggested by Saito (Saito 1996). Several Iranian brands of cigarettes were selected from market. For each analysis accomplished in triplicate, the content of three packages was homogenized. From the mixture, 15 grams were reserved for ^{210}Pb determination and another 15 grams for ^{210}Po determination.

To a dry sample of 5 gram of tobacco, 1ml of ^{208}Po tracer was added. The sample was leached with 40 mL of concentrated nitric acid, under heating at temperature not exceeding 70^{o} C in order to avoid losses of polonium by volatilization. At close to the dryness, it was added more 30 mL of nitric acid. This procedure was repeated once more.

Following, 10 mL of hydrogen peroxide was added for the destruction of the organic matter and 10 mL of nitric acid 8M was added under heating. At close to the dryness, it was added more 10 mL of nitric acid 8M. The sample was filtered directly into a separation funnel containing 5 mL of TBP (tributilphosphate). The mixture was shaken by 5 minutes. After 10 minutes in rest, the polonium is found in the aqueous phase. The extraction was repeated twice with 10 mL of nitric acid 8M.

Then, the aqueous phase was taken close to the dryness by heating, in order to eliminate the nitric acid. After, 20 mL of concentrated chloridric acid was added under heating. At close to the dryness, more 20 mL of chloridric acid 2M was added.

The iron was complexed by adding L (+) ascorbic acid until the change of the coloration from yellow to colorless was observed.

The solution was transferred to a plating cell, where the ^{208}Po and ^{210}Po were spontaneously deposited onto a copper disc, after 4 hour under continuous agitation and heating at temperature bellow 70^{o} C. The disc was washed with deionized water and let to dry at room temperature. The alpha spectrum was obtained by counting in a surface barrier detector.

The counting efficiency was determined using an electrodeposited source of ^{241}Am. The value obtained was 0.125 ± 0.001. The yield of the process ranged from 23 to 67% and the typical lower limit of detection of this method was 5x10^{-5} mBq/gr for 1000 minutes time counting.

The average activity concentration of ^{210}Po in Iranian cigarettes was 26 mBq per cigarette. Assuming that 50% of ^{210}Po activity contained in cigarette is inhaled, the average committed effective dose due to ^{210}Po and ^{210}Pb intake by smoking one pack (20 cigarettes) per day during one year is 0.4-0.5 mSv. Table 2 shows an activity comparison between Iranian cigarettes and those from other countries that are mostly found in Iranian markets and reported in different literature (Colangelo, et al. 1992, Peres, et al. 2002). As it can be seen, the average concentration of ^{210}Po in Iranian cigarette is relatively high in comparison with other cigarettes.

Table 2: The average activity concentrations of ^{210}Po in Iranian cigarettes and in those from other countries[7,8].

Country	**Average ^{210}Po Concentration (mBq/cigarette)**
Iran	26
France	23
Brazil	19
Russia	14
Egypt	14
Turkey	14

Conclusions

Concentration levels of ^{226}Ra were measured in different samples of bottled natural mineral water commercially available in Iran's supermarkets. ^{226}Ra was analyzed using radon emanation method with a minimum detection limit of 2 mBq/l. Concentration of ^{226}Ra ranged from 2.10 to 6.24 mBq/l in different parts of Iran. As the results show, the ^{226}Ra concentration in investigated mineral water did not exceed the level of 100 mBq/l that recommended by the WHO for drinking water. The isotope ^{226}Ra present in the under round water in the Northwest and Northeast of Iran

which most people use for medical treatments have been measured and the results indicate that the concentration of ^{226}Ra is very small.

Cigarette smoking increases the internal intake of ^{210}Po and ^{210}Pb, which are contained in cigarette tobacco. ^{210}Po and ^{210}Pb that are inhaled and deposited in the lung will contribute to an increase in the internal radiation dose. The result of this study indicates that the average activity concentration of ^{210}Po in Iranian cigarettes is 26 mBq/cigarette, which is relatively high in comparison with other cigarettes found in the market.

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