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RESEARCH ARTICLE

# Investigation of distribution of radioactivity with effects of heavy metals in toothpastes from Penang markets

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**Abstract** This study was carried out to determine the concentration of $^{222}$Rn, $^{226}$Ra, and $^{238}$U in 25 different toothpastes available in the local market in Penang, Malaysia, using a CR-39 detector. The results showed the maximum concentration of radon/ radium/uranium to be 4197.644 Bq.m$^{-3}$, 54.369 Bq.Kgm$^{-1}$, and 0.044 ppm in Colgate4; the annual effective dose was found (0.402 mSvy$^{-1}$) in S07. The average concentration of radon (42 %, 3.224 KBq.m$^{-3}$) was higher than the concentration of $^{214}$Po, $^{218}$Po in POS (32 %, 2.415 KBq.m$^{-3}$) and POW (26 %, 1.979 KBq.m$^{-3}$). Also the values of pH of samples ranged from 4.21 (highly acidic) in S04 to 9.97 (highly basic) in S07, with an average of 6.33 which tended towards an acidic behavior; a low or high pH for a long period of time can cause harmful side-effects and enamel erosion. Concentrations of heavy metals varied from the maximum value 56.156 ppm in the Ca elements in the Colgate 4 sample to a minimum value of −0.858 ppm in the Cd elements in Colgate 6 (Ca 56.156 ppm > Cd 51.572 ppm > Zn 41.039 ppm > Mg 11.682 ppm > Pb 11.009 ppm]. Monitoring the accumulation of these metals in toothpaste samples is very important: the average annual effective dose (0.3118 mSvy$^{-1}$) was below the range (3–10 mSvy$^{-1}$) reported by ICRP (1993), and therefore there is no evidence of health problems. Significant strong positive correlations were found ($r = 1$, Pearson correlation, $p < 0.000$) in concentration of radon, radium, uranium, annual effective dose, pH, and electrical conductivity.

**Keywords** Radon · Heavy metals · Uranium · Toothpaste · pH · CR-39 detector

## Introduction

Radium, radon, and uranium are grouped together because they are radionuclides, unstable elements that emit ionizing radiation. Uranium, radium, and radon occur naturally in the environment, uranium and radium as solids in rock while radon exists as a gas. The radioactive half-life is the time it takes for a substance to lose one-half of its radioactivity. While the half–life of radon is only about 4 days, the half-lives of the most common isotopes for radium and uranium, radium-226 and uranium-238, are approximately 1,600 years and 4.5 billion years respectively. Uranium is used in the nuclear power industry, for military ammunition, radiation shielding, and weighting balances for aircraft. Naturally occurring uranium has a smaller amount of radioactivity than the uranium used in nuclear reactors, in which uranium-235 isotope has been concentrated or "enriched" (Hezan 2007).

In the early 1900s, when it was newly discovered, no one understood the dangers of radium. People were fascinated with its mysterious properties, especially the luminescence produced when it is mixed with a phosphor. Industries sprang up to manufacture hundreds of consumer products containing radium. Advertisements proclaimed its special powers and unique effects in such products such as hair tonic, toothpaste, ointments, and elixirs. Glow in the dark watch and clock faces were immensely popular (Ali AAbo et al. 2014).

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Ra-226 has been used in numerous applications following its discovery over 100 years ago. At the beginning of the twentieth century, radium was a popular additive in consumer products such as toothpaste, hair creams, and even food items due to its supposed beneficial health properties. Such products soon fell out of vogue, and were prohibited by authorities in many countries because of the potential health effects (FAQs, 2015). This paper presents the investigation of radon, uranium, and radium contents in some toothpaste samples that are used by the Malaysian people and are available in almost all the markets in Penang, Malaysia, by using CR-39 detectors in order to investigate radioactivity of the dose of alpha radiation and investigating these material contents as a radiation health hazard, studying simple materials which may be a direct way to ytransmit these toxins and radioactive substances to humans; the possibility of transmission of radioactive substances to humans is great through saliva or scratches that occur in the gums, thus increasing the proportion of incoming radioactive substances in the human body. It is known that the proportion of uranium in the human body is in the order of 0.0009 ppm, although the entry of one atom can increase the proportion of radiation up to 174 times (Hana 2002).

Although there are a few methods for the determination of heavy metals in toothpaste, methods based on atomic absorption spectrometry (AAS) and inductively coupled plasma mass spectrometry (ICP-MS) are preferred for control of heavy metals in toothpaste. Since toothpastes are considered to be highly complex disperse systems with different organic constituents (Nageswara and Nageswara 2014), therefore, this paper presents the results of an investigation and determination of radioactivity of uranium and radium by using the track detector method, and also the results of investigation and determination of the different elements of heavy metals in toothpastes by using the AAS method.

## Toothpaste

Toothpaste is the most common oral care product for reducing oral becteria flora or for preventing periodontal disease (UMMI 2010). It is a paste or gel dentifrice used with a toothbrush as an accessory to clean and maintain the appearance and health of teeth and to promote oral hygiene. The main purpose of toothpaste is to reduce oral bacterial flora and deliver fluoride to the teeth (Okpalugo et al. 2009). An increasing use of fluoride toothpastes is the most significant factor for improved control of dental caries; the need to strengthen the effective use of fluoride continues to be emphasised as an important public health measure for the prevention of dental caries in the twenty-first century (Poul and Switzerland 2007).

## CR-39 detector

CR-39 is a clear, colorless, rigid plastic, with a density of 1.30 g/ $cm^3$, and chemical formula $C_{12}H_{18}O_7$ (Salih 2008; Enge 1981). made out of polyallyl diglycol carbonate (PADC) resin (Gaillard et al. 2007; Axelson 1995). The rectangular piece of the nuclear track detector (NTD) is $1.5 \times 1.5 \times 0.5$ $mm^3$ in size. The sensibility of CR-39 is such that it is physically able to register low energy alphas. Its high degree of reproducibility from batch to batch ensures the correct determination of the "background signal", giving an accurate estimate of the actual radon concentration (Qiuju et al. 2001).

## Tube-technique (PVC)

PVC tube is a plastic cylinder, made from PVC (polyvinyl chloride) (Salih FN Jaafar 2013) in the form of a cylinder of 2 mm thickness, a diameter of 3 cm and 11 cm long, used in this work to determine the radon concentration and radioactivity in the toothpaste samples (Salih 2008).

## Methodology

In this study, 25 different samples of different toothpaste types were collected, which are available in the local market in Penang, Malaysia, selected on the basis of their frequency of use.This was done through a questionnaire concerning toothpaste usage which determined the most common and widely used types in the country. Those available in the market were the following; Safi1 (KAYU SUGI), Safi2 (COMPLET CARE), Safi3 (Anti-Kaviti), Colgate1 (Total), Colgate2 (Fresh and White), Colgate3 (Kayu Sugi), Colgate4 (SPIDER-MAN), Colgate5 (Maximum Cavity Protection), Colgate6 (ADVANCED WHITE), Colgate7 (Barlie), Raiya, Pureen, LION, Pro-tech, Vicco, Pepsodent, Sparkle, Halagel, Everyday, Parodontax, DARLIE, PEARL, T-CARE, JUNIOR, BaruTesco.

Samples for testing were prepared by weighing amounts of samples using an electronic balance, drying the toothpaste in an oven for 3 days at 105 °C (Sérgio et al. 2011; Nisar et al. 2014), and then dissolving approximately 100 mg of the dried samples in can preparation for this reason. The samples were then changed to powder in the Medical Physics Laboratory/ USM; they were dried well and then a mill was used to get a good fine powder, homogeneous in terms of distribution of radioactive materials.The 12 gm of each powder sample was placed in cylindrical chambers (long tube-PVC) with a diameter of 3 cm and length of 11 cm; each sample was placed in the bottom of the tube, and the CR-39 detectors were placed on top of the radon dosimeters at 10.5 cm from the surface of the toothpaste samples to register the track of α-particles from the radioactivity during the time of exposure; after installing the detectors in position and sealing the tube properly, the samples were stored and left without moving in an ambient temperature for 90 days. About 98 % of the equilibrium level is reached between radium and radon during this time period (Durrani and Bull 1987). Upon completion of the exposure process during the afore-mentioned period of time, the detectors were taken out from the chambers and prepared for the etching process and track visualization. The process of chemical etching (the simplest and most widely used technique for revealing the latent damage trails of ionizing particles in solids when alpha particles are deposited in the body, thus measuring the radon concentration) (UNSCEAR 1993) was carried out in 6.25 normality of NaOH at 70 ± 1 °C using a water bath for 9 hours (Mohammed 2003) in order to make the tracks large size to appear the alpha emitters emitted from the radon containing the samples on the surface of detectors. Then the detectors were washed in distilled water for 20 minutes to stopping the etching process and to remove the effects of solution etching: a microscope was used to determine the number of alpha particles by measuring the track density formed in the CR-39 reagent.

## Sample analysis

Heavy metals are considered to be one of the main pollutants in the environment. Lead, cadmium, magnesium, manganese, etc. may be chosen as representative trace metals in the environment representing a reliable index of environmental pollution. In this study, the element levels of each toothpaste sample were determined by AAS. Atomic absorption spectroscopy (AAS) is an analytical technique used for the detection of heavy metals. A calibration process was undertaken before using AAS to analyze the samples. Calibration standards of each metal were prepared by using an appropriate dilution of stock solution (concentration usually in ppm) of each element provided by the laboratory (Nageswara and Nageswara 2014).

After the toothpaste samples were dried well, they were kept in separate special tubes; then the experiment were carried out by weighing the samples using an electronic balance; after preparation, the concentration levels of selected heavy metals such as calcium (Ca), cadmium (Cd), magnesium (Mg), manganese (Mn), lead (Pb), and zinc (Zn) in toothpaste samples were analyzed by using AAS (spectrophotometer), the standard addition method: 0.25 g of powdered toothpaste samples were dissolved by the addition of 8 ml concentrated nitric acid (65 % $HNO_3$) and 2 ml concentrated perchloric acid (70 % of HF) to the samples. The sample was heated gently to start the peroxide reaction; it was heated for an additional 15 min without boiling, then the sample was cooled (Muhammad and Stephen 2014). The details of the technique are reported elsewhere (Handbook 2014), and the microwave machine analysis system was used to change the colored toothpaste samples to samples without color using a 35-minute cycle for each process in the Laboratory of Chemistry School.

In addition, the pH and electrical conductivity (EC) of toothpaste samples were measured using the methods described by Hanlon 2009. pH level is the measure of the acidity or alkalinity of a solution. Before carrying out the process of pH, the calibration of equipment of pH with three standard values of pH (pH = 4 acid, pH = 7 normal, pH = 9 basic). The pH of all toothpaste samples was measured by using a pH meter (model 827 pH lab from the company Metrohm) with a normal toothpaste solution in a special tube designed for this study, and electric conductivity was measured with the help of a conductivity meter (Jenway Model 470).

## Statistical analysis

All statistical calculations were performed using SPSS (Statistical Package for Social Sciences) for Windows, standard version 22.0. Description and analysis of the data were carried out by frequency distributions (Pearson correlation, Kruskal–Wallis test and *t*-test (analysis of variance) in order to assess the statistical significance in the data of toothpaste samples.

### Determination of track density and measurement of radon concentration

After the four steps of the track visualization process (etching, washing, drying, and scanning) for CR-39 detectors, the track densities (ρ) were observed according to the following equation (Gil and Kilee (2005):

$$\textbf{Track density } (\rho) = \textbf{average of total pits / area of field view} \quad (1)$$

The measurement of radon concentrations was based on the track densities in the exposure period (90 days) should be determined with the calibration factor of track density, $K(\text{track} \cdot \text{cm}^{-2} \text{ per Bq} \cdot \text{m}^{-3} \cdot \text{day})$, which depends on the average value of critical angle for CR 39 detectors. Equation 2 was used to calculate the calibration factor, as adapted from the studies of (Ismail 2012):.

$$K = \frac{1}{4} r \cdot \left( 2\cos\theta_c - \frac{r}{R_\alpha} \right) \quad (2)$$

Where K = 0.4788 $\text{Track.cm}^{-2}/\text{Bq.m}^{-3}$.d is the calibration factor which was calculated by eq. (2) dependent on the geometrical dimension for detector system (Barillon et al. 1993),

$\theta_C = 35°$ is the average value of critical angle for CR 39 detectors (Durrani and Bull 1987; Barillon et al. 1993), r = 1.5 cm radius of irradiation tube and $R\alpha = 4.15$ cm is the average of range of alpha particle in air to $Rn^{222}$ (Fleischer and Margo 1978).

To calculate and estimate the level of radon concentration in the air inside the chamber, the following relationship was used (Azam et al. 1995; Asumadu-Sakyi et al. 2011; Shashikumar et al. 2011; Alberigi et al. 2004).

$$Ca = C_{Rn}\left(Bq.m^{-3}\right) = \rho/k.\ t \tag{3}$$

The density of the effectiveness of the concentration of radon in the samples can be calculated by using the following relationship (Al-Bataina 2001; Ukla 2004):

$$C = \frac{C_a \lambda_{Rn} h t}{L} \tag{4}$$

Where Ca = concentration of radon in the air space unit $Bq.m^{-3}$, C = concentration of radon in samples unit $Bq.m^{-3}$, ρ = the track density (track. $cm^{-2}$), k = the calibration factor (track.$m^{-3}$)/($cm^{-2}$.Bq.day), t = the exposure period (day), $_{Rn}$ is the radon decay constant (0.1814 $d^{-1}$), h is the distance from the sample surface inside the tube to the face of the detector and is equal to 10.5 cm, t is the irradiation time (90 days), and L is equal to the sample thickness, which is rounded to 0.5 cm.

## Estimation of specific activity of radon ($A_{Rn}$)

$$A_{Rn} = CV \tag{5}$$

$$V = \pi r^2 L \tag{6}$$

V is the volume of the chamber
L is the distance between the sample and the detector

## Estimation of radium and uranium concentration

The concentration of uranium and radium were determined by calculating the number of atoms $N_{Rn}$ in the relationship:

$$A_{Rn} = \lambda_{Rn} N_{Rn} \tag{7}$$

By using the low of ideal radioactivity balance, the number of atoms of uranium in the samples from the equation was determined:

$$N_U \ \lambda_U \ = \ N_{Rn} \ \lambda_{Rn} \tag{8}$$

Where $\lambda_U$ is the decay constant of uranium (4.8833 x$10^{-18}$ $s^{-1}$), and the mass of uranium $W_U$ (g) in the samples was found by Eq. 9 (Ukla 2004).

$$W_U = \frac{N_U A_U}{N_{av}} \tag{9}$$

$A_U$ is mass number of $^{238}U$, Nav is the number of Avogadro (6.022 x $10^{23}$ $mol^{-1}$). The concentration of uranium (ppm) was found by equation 10 (Ukla 2004). Since the mass samples $W_S$ was used equal to 12 g:

$$C_U(ppm) = \frac{W_U}{W_S} \tag{10}$$

## Estimation of the radon effective radium content

The effective radium $C_{Ra}$ content in the sample is the amount of the contain virtually the sample of radium which slimming to radon that was calculated in units of component Bq.kg-1 of the following relationship 11 (Azam et al. 1995). Effective exposure time of radon was calculated by using the following relationship because the resulted in the exposure (90 days) of the radon to variable levels of radon concentration starting from zero concentration level to equilibrium level (Durrani and Bull 1987) and effective exposure time was calculated by Eq.12. When $T_{eff} = 87.349$ day and $_{Rn} = 2.1$x 10-6 $S^{-1} = 0.007567\ h^{-1}$

$$C_{Ra}\text{-} = \left(\frac{\rho}{k \cdot T_e}\right)\left(\frac{h \cdot a}{M}\right) \tag{11}$$

$$T_{eff}\left[T_{tot}\text{-}\lambda_{R_n}^{-1}\left(1\text{-}e^{-\lambda_{Rn}\,T_{tot}}\right)\right] \tag{12}$$

To calculate the mass exhalation rate of radon using the following relationship (Azam et al. 1995):

$$C_{Rn}\left(B_q.kg^{-1}.d^{-1}\right) = C_{\rho(Ra)} \cdot \frac{\lambda_{Ra}}{\lambda_{Rn}} \cdot \frac{1}{T_{eff}} \tag{13}$$

For estimating the effective dose from $^{222}Rn$ progeny in the toothpaste, the relationship between the effective dose and radon concentration is given by Najeeb et al. 2005.

## Estimation of the radon progeny concentration

The contribution and concentrations of radon progeny (polonium) were emitting alpha particles ($^{214}Po$, $^{218}Po$) which were deposited on the walls (POW) of the irradiation chamber and on the face (POS) of the detector (Barillon et al. 1993) were calculated using the relationships explained by Salih 2008; Sérgio et al. 2011.

$$D_{Po^{218}} = D_{Po^{214}} = \frac{C}{4} r \left(\frac{h}{r+h}\right) \cos\theta_c \tag{14}$$

$$D_{Po^{218}} = D_{Po^{214}} = \frac{C}{4} r \left(\frac{h}{r+h}\right) \left(\cos\theta_c \frac{r}{R_\alpha}\right) \tag{15}$$

## Results and discussion

This research aimed to study the contents of the toothpastes in common dental use in the country, and determine the concentrations of radioactive materials, on the principle that some imported materials should be monitored for radiation, as well as other industrial controls, to minimize radioactive contamination problems, especially since the mouth is a susceptible quick source of entry of radioactive materials to the human body and ionizing radiation can cause toxicity; therefore, these substances are a health risk only if taken into the body by ingestion or inhalation (Hezan 2007). This study was carried out to determine the levels of radon in the different types of toothpaste which are available in Malaysia markets were measured, in order to be able to take action to avoid exposure of consumers to radiation. We used the CR-39 solid-state nuclear track detection technique because is widely used as a tool to measure α-track activity (Akram et al. 2008). CR-39 NTDs possess the stability to withstand various environmental factors sensitive to alpha particles within the energy range emitted by radon. The sensibility of CR-39 is such that it is physically able to register low energy alphas. Its high degree of reproducibility from batch to batch ensures the correct determination of the “background signal”, giving an accurate estimate of the actual radon concentration (Qiuju et al. 2001). About 98 % of equilibrium level is reached between radium and radon during this time period (Asumadu-Sakyi et al. 2011).

During the time of storage, alpha particles from the decay of radon and their daughters bombarded the CR-39 NTDs, which recorded the α-emitters from toothpaste samples. When a particle interacts with a CR-39 detector, the polymeric chains along its trail are affected, which results in the formation of latent track. The etching produces a ‘hole’ in the detector along the path of the particle, which can be easily obvious and visible under an optical microscope after being subjected to chemical etching. Track visualization using etching generally consists of four steps: etching, washing, drying, and observation under an optical microscope (Salih FN Jaafar 2013). $^{222}$Rn is the most significant among these isotopes because of its longer half-life Therefore, this study focuses

**Table 1** Radon and radium concentrations in the toothpaste samples

| Code of sample | Toothpaste | Co. of radon (Ca) Bq.m$^{-3}$ | Co. of radon (Cs) Bq.m$^{-3}$ | Radon activity (A) Bq* 10$^{-3}$ | Co. of radium Bq.Kg$^{-1}$ |
|---|---|---|---|---|---|
| S01 | Safi1 | 29.521 | 3534.372 | 549.347 | 45.779 |
| S02 | Safi2 | 30.891 | 3698.481 | 574.855 | 47.904 |
| S03 | Safi3 | 27.553 | 3298.775 | 512.728 | 42.727 |
| S04 | Colgate1 | 19.451 | 2328.747 | 361.957 | 31.163 |
| S05 | Colgate2 | 27.657 | 3311.242 | 514.666 | 42.888 |
| S06 | Colgate3 | 20.241 | 2423.397 | 376.668 | 30.989 |
| S07 | Colgate4 | 35.061 | 4197.644 | 652.439 | 54.369 |
| S08 | Colgate5 | 31.063 | 3718.993 | 578.043 | 48.170 |
| S09 | Colgate6 | 28.459 | 3407.330 | 529.601 | 44.133 |
| S10 | Colgate7 | 21.753 | 2604.422 | 404.805 | 33.734 |
| S11 | Raiya | 23.358 | 2796.602 | 434.676 | 36.223 |
| S12 | Pureen | 20.972 | 2510.856 | 390.262 | 32.522 |
| S13 | LION1 | 24.398 | 2921.122 | 454.029 | 37.836 |
| S14 | Pro-tech | 21.323 | 2552.959 | 396.806 | 33.067 |
| S15 | LION2 | 27.615 | 3306.204 | 513.883 | 42.823 |
| S16 | Pepsodent | 28.157 | 3370.984 | 523.952 | 43.662 |
| S17 | Sparkle | 34.369 | 4114.872 | 639.574 | 53.298 |
| S18 | Halagel | 33.188 | 3973.435 | 617.591 | 51.466 |
| S19 | Everyday | 30.444 | 3644.858 | 566.520 | 47.210 |
| S20 | Parodontax | 29.211 | 3497.304 | 543.586 | 45.298 |
| S21 | DARLIE | 21.114 | 2527.926 | 392.915 | 32.743 |
| S22 | PEARL | 29.971 | 3588.339 | 557.735 | 46.478 |
| S23 | T-CARE | 24.883 | 2979.156 | 463.050 | 38.587 |
| S24 | JUNIOR | 31.433 | 3763.235 | 584.919 | 48.743 |
| S25 | BaruTesco | 28.074 | 3361.124 | 522.419 | 43.535 |
| Mean | | 27.206 | 3257.295 | 506.281 | 42.190 |

on $^{222}Rn$, in order to assess the radioactivity in the samples of different types of toothpaste marketed in Malaysia. Analysis was also carried out of the concentrations of heavy metals in toothpaste samples, and pH and electrical conductivity of the samples.

## Concentration of radon, radiom, uranium in the toothpaste samples

Table 1 shows the measurements obtained of radon and radium concentrations for different types of toothpaste available in Malaysia markets: it can be seen that radon concentrations in 12 gm of toothpaste changed from a minimum value of 2328.747 Bq.m$^{-3}$ in the Colgate1(Total) sample to a maximum value of 4197.644 Bq.m$^{-3}$ in the Colgate4 sample; the small variation in radon values inside samples are due mainly to the difference in the natural uranium, which when it decays, changes into different elements that are also radioactive, including radon. Also, from Table 1, it can be seen that radium concentrations changed from a minimum value of 30.989 Bq.Kgm$^{-1}$ in the Colgate3 sample to a maximum value 54.369 Bq.Kgm$^{-1}$ in the Colgate4 sample (the variation from one toothpaste sample to another depends on the component formation: therefore, the observed variations of radon concentrations among various types of toothpaste can be attributed to quantity of fluoride that is added to toothpaste. The aim of this research is to know whether the concentration levels of the radiation elements which are found in toothpastes exceeds permitted world limits in order to avoid importing them. In Table 2, uranium concentrations vary from a minimum value of 0.024 ppm in the Colgate1 (Total) sample to a maximum value of 0.044 ppm in the Colgate4 sample. This also compared the effectiveness of radon with radon concentration in samples but noted that there was a decrease or an increase in radon concentration of the sample to the other because, depends on the sample mass used and therefore the radon concentration in the samples will depend heavily on the mass of those samples and for this

**Table 2** Radon activity and uranium concentration in the toothpaste samples

| Code of sample | Toothpaste | Radon activity (A) Bq X $10^{-3}$ | *N* of atoms uranium x$10^{21}$ | Wu gm x $10^{-2}$ | Co. of uranium ppm |
|---|---|---|---|---|---|
| S01 | Safi1 | 549.347 | 112.495 | 4445.245 | 0.0370 |
| S02 | Safi2 | 574.855 | 117.718 | 4651.647 | 0.0387 |
| S03 | Safi3 | 512.728 | 104.996 | 4148.930 | 0.0345 |
| S04 | Colgate1 | 361.957 | 74.121 | 2928.908 | 0.0244 |
| S05 | Colgate2 | 514.666 | 105.393 | 4164.61 | 0.0347 |
| S06 | Colgate3 | 376.668 | 77.134 | 3047.951 | 0.0254 |
| S07 | Colgate4 | 652.439 | 133.606 | 5279.454 | 0.0439 |
| S08 | Colgate5 | 578.043 | 118.371 | 4677.445 | 0.0389 |
| S09 | Colgate6 | 529.601 | 108.451 | 4285.462 | 0.0357 |
| S10 | Colgate7 | 404.805 | 82.896 | 3275.629 | 0.0273 |
| S11 | Raiya | 434.676 | 89.012 | 3517.338 | 0.0293 |
| S12 | Pureen | 390.262 | 79.917 | 3157.95 | 0.0263 |
| S13 | LION | 454.029 | 92.976 | 3673.948 | 0.0306 |
| S14 | Pro-tech | 396.806 | 81.258 | 3210.903 | 0.0267 |
| S15 | Vicco | 513.883 | 105.233 | 4158.275 | 0.0346 |
| S16 | Pepsodent | 523.952 | 107.294 | 4239.749 | 0.0353 |
| S17 | Sparkle | 639.574 | 130.972 | 5175.351 | 0.0431 |
| S18 | Halagel | 617.591 | 126.470 | 4997.463 | 0.0416 |
| S19 | Everyday | 566.520 | 116.012 | 4584.205 | 0.0382 |
| S20 | Parodontax | 543.586 | 111.315 | 4398.623 | 0.0366 |
| S21 | DARLIE | 392.915 | 80.461 | 3179.42 | 0.0265 |
| S22 | PEARL | 557.735 | 114.213 | 4513.12 | 0.0376 |
| S23 | T-CARE | 463.050 | 94.823 | 3746.938 | 0.0312 |
| S24 | JUNIOR | 584.919 | 119.779 | 4733.091 | 0.0394 |
| S25 | BaruTesco | 522.419 | 106.981 | 4227.348 | 0.0352 |
| Mean | | 506.281 | 103.676 | 4096.760 | 0.0341 |

reason does not get consensus between the increase or decrease in the effectiveness of radon and its concentration in the samples used for different block those samples.

### Mass exhalation rate of radon and annual effectivee dose of $^{222}$Rn

Mass exhalation rate is another point that this study focused on. Mass radon exhalation rate was measured by continuous radon dosimetry (CR-39). Table 3 shows that the maximum values and minimum values of mass exhalation rate annual effective dose of $^{222}$Rn of toothpaste samples were found to be 12.165 Bq.Kg$^{-1}$ d$^{-1}$, 0.402 mSvy$^{-1}$ in S7 and 6.749 Bq. Kg$^{-1}$ d$^{-1}$, 0.223 mSvy$^{-1}$ in S4 respectively. The annual effective dose depends on the exposure period (Abu-Jarad 1982), therefore when the time exposure increases the annual effective dose also increase. Also, the value of mass exhalation rate depends on the track density of the radon.

Positive correlation has been observed between radon concentration and radium concentration and other radioactivity factors in toothpaste samples; this measurement is important from the health protection point of view, so the passive method is a simple and reliable analytical method. This correlation of radon exhalation rate with track production rate indicates the track recording efficiency of CR-39, so it is the most suitable track detector for radon measurements.

Estimated values of radon exhalation rates for all samples were found to be lower than the international accepted value 57.60 Bq.Kg$^{-1}$ h$^{-1}$ (UNSCEAR 2000): hence, it is concluded that the toothpaste samples analyzed may be used for cleaning teeth as they have no health hazards.

### Radon progeny concentration

The maximum values and minimum values of concentrations of radon progeny ($^{214}$Po, $^{218}$Po) deposited on the walls of the irradiation chamber (POW) and on the surface of the detector (POS) were found to be 2.577 KBq.m$^{-3}$, 3.144 KBq.m$^{-3}$, and 1.429 KBq.m$^{-3}$, 1.744 KBq.m$^{-3}$ respectivily, depending on the average value of critical angle of alpha particles indicated on the CR 39 detectors (Barillon et al. 1993; Durrani and Bull 1987) and radius of irradiation PVC tube, as shown in Table 4.

**Table 3** Mass exhalation and annual effective dose of $^{222}$Rn

| Code of sample | Toothpaste | Track density track/cm$^2$ | Radium effective $10^{-3}$ Bq.Kg$^{-1}$ | $C_{Rn}$ x$10^{-6}$ Bq.Kg$^{-1}$ d$^{-1}$ | Annual effective dose- mSvy$^{-1}$ |
|---|---|---|---|---|---|
| S01 | Safi1 | 1272.119 | 393.975 | 10.243 | 0.338 |
| S02 | Safi2 | 1331.186 | 412.268 | 10.718 | 0.354 |
| S03 | Safi3 | 1187.321 | 367.713 | 9.560 | 0.315 |
| S04 | Colgate1 | 838.181 | 259.584 | 6.749 | 0.223 |
| S05 | Colgate2 | 1191.808 | 369.103 | 9.596 | 0.317 |
| S06 | Colgate3 | 872.248 | 270.135 | 7.023 | 0.232 |
| S07 | Colgate4 | 1510.849 | 467.909 | 12.165 | 0.402 |
| S08 | Colgate5 | 1338.569 | 414.555 | 10.778 | 0.356 |
| S09 | Colgate6 | 1226.393 | 379.814 | 9.875 | 0.326 |
| S10 | Colgate7 | 937.404 | 290.314 | 7.548 | 0.249 |
| S11 | Raiya | 1006.575 | 311.736 | 8.105 | 0.267 |
| S12 | Pureen | 903.727 | 279.884 | 7.277 | 0.240 |
| S13 | LION | 1051.393 | 325.616 | 8.466 | 0.279 |
| S14 | Pro-tech | 918.881 | 284.577 | 7.399 | 0.244 |
| S15 | Vicco | 1189.995 | 368.541 | 9.582 | 0.316 |
| S16 | Pepsodent | 1213.311 | 375.762 | 9.769 | 0.322 |
| S17 | Sparkle | 1481.057 | 458.683 | 11.925 | 0.394 |
| S18 | Halagel | 1430.15 | 442.917 | 11.515 | 0.380 |
| S19 | Everyday | 1311.886 | 406.291 | 10.563 | 0.348 |
| S20 | Parodontax | 1258.777 | 389.843 | 10.136 | 0.334 |
| S21 | DARLIE | 909.871 | 281.787 | 7.326 | 0.242 |
| S22 | PEARL | 1291.543 | 399.991 | 10.399 | 0.343 |
| S23 | T-CARE | 1072.281 | 332.085 | 8.634 | 0.285 |
| S24 | JUNIOR | 1354.493 | 419.486 | 10.906 | 0.360 |
| S25 | BaruTesco | 1209.762 | 374.663 | 9.741 | 0.321 |
| Mean | | 1172.391 | 363.089 | 9.440 | 0.312 |

**Table 4** Concentrations of radon progeny ($^{214}Po$, $^{218}Po$) in the irradiation chamber

| Code of sample | Toothpaste | Radon con. $KBq.m^{-3}$ | Concentration of (POW) $KBq.m^{-3}$ | Concentration of (POS) $KBq.m^{-3}$ |
|---|---|---|---|---|
| S01 | Safi1 | 3.534 | 2.170 | 2.647 |
| S02 | Safi2 | 3.698 | 2.271 | 2.770 |
| S03 | Safi3 | 3.298 | 2.025 | 2.470 |
| S04 | Colgate1 | 2.328 | 1.429 | 1.744 |
| S05 | Colgate2 | 3.311 | 2.033 | 2.480 |
| S06 | Colgate3 | 2.423 | 1.488 | 1.815 |
| S07 | Colgate4 | 4.197 | 2.577 | 3.144 |
| S08 | Colgate5 | 3.718 | 2.283 | 2.785 |
| S09 | Colgate6 | 3.407 | 2.092 | 2.552 |
| S10 | Colgate7 | 2.604 | 1.599 | 1.951 |
| S11 | Raiya | 2.796 | 1.717 | 2.094 |
| S12 | Pureen | 2.512 | 1.541 | 1.880 |
| S13 | LION | 2.921 | 1.793 | 2.188 |
| S14 | Pro-tech | 2.553 | 1.567 | 1.912 |
| S15 | Vicco | 3.306 | 2.031 | 2.476 |
| S16 | Pepsodent | 3.371 | 2.069 | 2.525 |
| S17 | Sparkle | 4.115 | 2.526 | 3.082 |
| S18 | Halagel | 3.973 | 2.439 | 2.976 |
| S19 | Everyday | 3.645 | 2.238 | 2.729 |
| S20 | Parodontax | 3.497 | 2.147 | 2.619 |
| S21 | DARLIE | 2.528 | 1.552 | 1.893 |
| S22 | PEARL | 3.588 | 2.203 | 2.687 |
| S23 | T-CARE | 2.979 | 1.829 | 2.232 |
| S24 | JUNIOR | 3.763 | 2.310 | 2.818 |
| S25 | BaruTesco | 3.361 | 2.063 | 2.517 |
| Mean | | 3.224 | 1.979 | 2.415 |

The average value of concentration of radon (3.224 $KBq.m^{-3}$) at 42 % was higher than the concentration of polonium (POW) (1.979 $KBq.m^{-3}$) at 26 %, and POS (2.415 $KBq.m^{-3}$) at 32 %. The sensitivity of the detector to alpha particles emitted by radon depends on the dimensions of the chamber that is used (Nikzic et al. 1996), and the distribution of the tracks on the face of the detector at 2.415 $KBq.m^{-3}$ was higher than the distribution of the tracks on the walls of the detector at 1.979 $KBq.m^{-3}$; therefore, CO. Rn > CO. POS > CO. POW, rspectively. It should be noted that that the concentration of $^{214}Po$ and $^{218}Po$ is low and within the limits of error in the measurement of concentrations of radon, and this can be neglected due to the effects of this progeny.

## Toothpaste characteristics — pH, electrical conductivity

### *pH*

pH is the measure of the acidity or alkalinity of a solution. The pH of toothpaste should be neutral or close to neutral. The pH of all toothpaste samples was measured by using a pH meter with a normal toothpaste solution in a special tube designed for this study. A lower pH indicates acidity while a higher pH indicates alkalinity. A pH value of 7 is considered neutral. This study measured the pH of 25 commercially available toothpaste products: Table 5 shows the pH and conductivity of the toothpaste samples collected from different markets in Penang: the results indicate that the pH of the different toothpaste samples ranged from 4.21 (highly acidic) in S04 to 9.97 (highly basic) in S07, with an average of (6.33) at 21.5 °C, which agrees with the study Richard et al. 2000, which tended toward an acidic behavior. pH has effects on enamel; low pH and high acid concentrations lead to adverse side-effects such as enamel erosion. Using ANOVA statistical analysis showed that there was a significant difference ($p$ <0.05); exposure of the teeth and oral tissues to a low or high pH for an a long period of time may cause harmful side-effects, cause enamel erosion 28,29 (Richard et al. 2000). Also the pH of toothpaste ranges from 4.21 to 9.97 depending on the additives it contains (Price et al. 2000).

**Table 5** pH and conductivity in toothpaste samples

| Code of sample | Toothpaste | pH | EC (mS/cm) |
|---|---|---|---|
| S01 | Safi1 | 7.71 | 0.28 |
| S02 | Safi2 | 5.00 | 0.26 |
| S03 | Safi3 | 6.82 | 0.23 |
| S04 | Colgate1 | 4.21 | 0.18 |
| S05 | Colgate2 | 7.49 | 0.25 |
| S06 | Colgate3 | 6.38 | 0.29 |
| S07 | Colgate4 | 9.97 | 0.32 |
| S08 | Colgate5 | 7.22 | 0.22 |
| S09 | Colgate6 | 5.54 | 0.37 |
| S10 | Colgate7 | 7.18 | 0.20 |
| S11 | Raiya | 4.65 | 0.17 |
| S12 | Pureen | 5.72 | 0.19 |
| S13 | LION | 5.22 | 0.27 |
| S14 | Pro-tech | 5.78 | 0.14 |
| S15 | Vicco | 6.72 | 0.16 |
| S16 | Pepsodent | 7.51 | 0.25 |
| S17 | Sparkle | 5.49 | 0.19 |
| S18 | Halagel | 7.18 | 0.16 |
| S19 | Everyday | 5.74 | 0.27 |
| S20 | Parodontax | 4.88 | 0.34 |
| S21 | DARLIE | 5.33 | 0.34 |
| S22 | PEARL | 7.04 | 0.13 |
| S23 | T-CARE | 5.91 | 0.29 |
| S24 | JUNIOR | 7.38 | 0.17 |
| S25 | BaruTesco | 6.82 | 0.24 |
| Mean | | 6.33 | 0.23 |

*Electrical conductivity*

Conductivity is a solution's ability to transmit an electrical charge. Electrical conductivity was measured with the help of a conductivity meter (Jenway Model 470) in the same samples and at the same time as pH. The results indicate that the conductivity varied from 0.13 mS/cm in S22 to 0.37 mS/cm in S09, with an average of conductivity of the samples of 0.23 at 21.5 °C, as shown in Table 5 and Fig. 1.

## Heavy metals

Heavy metals are potentially toxic and/or clinically undesirable (Hardman 2006). Most heavy metals are toxic and their accumulation over time in the bodies can cause severe diseases if there is long-term exposure. In the present investigation we used the AAS method because it is highly sensitive and specific and has been developed and validated (Hardman 2006). The concentration of uranium, zinc, cadmium, lead, calcium, magnesium, and cobalt in toothpaste samples from markets in Penang in Malaysia were analysed, in order to understand the correlation which exists between the concentration of uranium and these heavy metals. A positive correlation was observed between the concentration of uranium and heavy metals, as shown in Table 6.

There are many techniques used for heavy metal digestion procedures such as dry, wet, and microwave. Both dry and wet digestions are slow and time-consuming, whereas microwave digestion is much more rapid and involve low consumption of time and reagents (Esen and Balci 2008) A microwave reaction system (MWRS) was utilized in this study to digest the samples. The main advantages of microwave digestion are that it requires small amounts of sample and oxidizing materials, short digestion times, and provides ease of sample handling (Ingrid et al. 1998). Although there are a number of methods for determinating the concentration of heavy metals in toothpaste, methods based on AAS are preferred (Nageswara and Nageswara 2014). To ensure precision and accuracy, each sample was analyzed in triplicate and the instrument was continuously calibrated during analysis.

Table 6 shows the concentrations of Ca varied from 56.156 ppm in the Colgate4 sample to −2.384 ppm in the LION sample; the concentrations of Cd varied from 51.572 ppm in the Everyday sample to −0.858 ppm in the Colgate6 sample, the concentrations of Mg varied from 11.682 ppm in the Sparkle sample to −2.642 ppm in the Pepsodent sample, the concentrations of Pb varied from 11.009 ppm in the Parodontax sample to −2.168 ppm in the Everyday sample, and the concentrations of Zn varied from 41.039 ppm in the Pro-tech sample to −2.342 ppm in the Colgate3 sample. The concentrations of heavy metals overall varied from a maximum value of 56.156 ppm in the Ca elements in the Colgate4 sample to a minimum value of −0.858 ppm in the Cd elements in the Colgate6 sample; therefore, Ca 56.156 ppm > Cd 51.572 ppm > Zn 41.039 ppm > Mg 11.682 ppm > Pb 11.009 ppm respectively. The average concentration of metals determined in the toothpaste samples included: Zn (32 %, 6.457 ppm) > Cd (29 %, 5.529 ppm) > Ca (24 %, 5.101 ppm) > Pb (8 %,1.521 ppm) > Mg (7 %, 1.449 ppm). Presence of heavy metals above the maximum permissible limit could be harmful, as shown in Fig. 1.

The concentrations of uranium varied from a maximum value of 0.049 ppm in the Colgate4 sample to a minimum value of 0.023 ppm in the Colgate7 sample. Because the amount of flouride in toothpaste varies and there is a relationship between the radon and flouride element difference, the average concentration of uranium of 0.034 ppm and the concentration of heavy metals in the range of uranium concentration were centered between 0.02 and 0.05 ppm, as shown in Figure 1. The

**Table 6** The concentration of uranium and heavy metals in toothpaste samples

| Code of sample | Toothpaste | $^{238}$U ppm | Ca ppm | Cd ppm | Mg ppm | Pb ppm | Zn ppm |
|---|---|---|---|---|---|---|---|
| S01 | Safi1 | 0.037 | −0.222 | −0.027 | 0.342 | −0.139 | −2.342 |
| S02 | Safi2 | 0.038 | −0.366 | −0.024 | 0.277 | −0.109 | 3.066 |
| S03 | Safi3 | 0.034 | 1.851 | −0.014 | 1.7538 | −0.136 | 4.521 |
| S04 | Colgate1 | 0.024 | 10.951 | −0.016 | 1.618 | −0.114 | 0.544 |
| S05 | Colgate2 | 0.035 | −0.421 | −0.035 | 0.295 | −0.221 | 0.462 |
| S06 | Colgate3 | 0.025 | 48.539 | −0.034 | 1.453 | 0.165 | −2.342 |
| S07 | Colgate4 | 0.049 | 56.156 | −0.022 | 1.280 | 0.019 | 16.312 |
| S08 | Colgate5 | 0.039 | −1.372 | −0.121 | −2.164 | 3.185 | 6.992 |
| S09 | Colgate6 | 0.037 | 3.023 | −0.858 | −0.283 | 4.651 | 7.556 |
| S10 | Colgate7 | 0.023 | 2.521 | −0.163 | 0.137 | 1.584 | 4.521 |
| S11 | Raiya | 0.029 | 0.814 | 12.291 | −2.552 | 5.422 | 1.832 |
| S12 | Pureen | 0.026 | 0.481 | 0.158 | 3.195 | 3.087 | 13.957 |
| S13 | LION | 0.036 | −2.384 | 0.819 | 6.821 | 5.214 | 0.425 |
| S14 | Pro-tech | 0.026 | 1.972 | 8.432 | 0.583 | 4.413 | 41.039 |
| S15 | Vicco | 0.034 | 0.317 | −0.756 | 0.412 | 1.169 | 26.038 |
| S16 | Pepsodent | 0.035 | 1.917 | −0.764 | −2.642 | 3.693 | 2.382 |
| S17 | Sparkle | 0.041 | 2.154 | −0.810 | 11.682 | 1.184 | 3.066 |
| S18 | Halagel | 0.046 | 0.151 | 41.753 | −2.352 | 0.269 | 4.521 |
| S19 | Everyday | 0.038 | 1.463 | 51.572 | 3.011 | −2.168 | 2.856 |
| S20 | Parodontax | 0.036 | 1.261 | 12.082 | 4.821 | 11.009 | 10.932 |
| S21 | DARLIE | 0.025 | 0.175 | 1.279 | 1.764 | −0.189 | −0.429 |
| S22 | PEARL | 0.037 | −0.136 | 1.584 | 0.742 | −2.168 | 8.632 |
| S23 | T-CARE | 0.031 | −0.334 | 9.653 | −2.352 | −0.742 | 6.137 |
| S24 | JUNIOR | 0.039 | −1.289 | 0.995 | 3.816 | −0.137 | −2.342 |
| S25 | BaruTesco | 0.035 | 0.285 | 1.273 | 4.571 | −0.915 | 3.086 |
| Average | | 0.034 | 5.101 | 5.529 | 1.449 | 1.521 | 6.457 |
| Maximum | | 0.049 | 56.156 | 51.572 | 11.682 | 11.009 | 41.039 |
| Minimum | | 0.023 | −2.384 | −0.858 | −2.642 | −2.168 | −2.342 |

result (0.12 ppm) in Asumadu-Sakyi et al. 2011 is higher than this, and this agrees with the result reported by the researcher Nada 2004.

It is an established fact that high concentration of non-radioactive heavy metals such as Pb, Cd, and Zn in the samples may also lead to teeth damage. The highest concentration was found in Ca (56.156 ppm), while the lowest was observed in Mg (−2.642 ppm). The result showed the maximum value of element concentration was found in the three elements Ca, Zn, Cd in the five toothpaste samples Tesco Choice, Raiya, Tesco Everyday, Colgate Total, and Colgate Fresh and White. The analysis of the heavy metal concentrations indicated that there is presence of cadmium, zinc and magnesium in high proportions. These metals are toxic and may cause severe problems with prolonged exposure (Khan et al. 1988). Monitoring the accumulation of these metals in toothpaste samples is very important, and works to prevent the transportation of these toxic metals into the human system.

## Statistical results

### Statistical results of concentrations

As shown in Table 7, using descriptive statistics and *t*-test, the maximum average values and minimum average values for concentrations of $^{222}$Rn in 12 gm of toothpaste were found

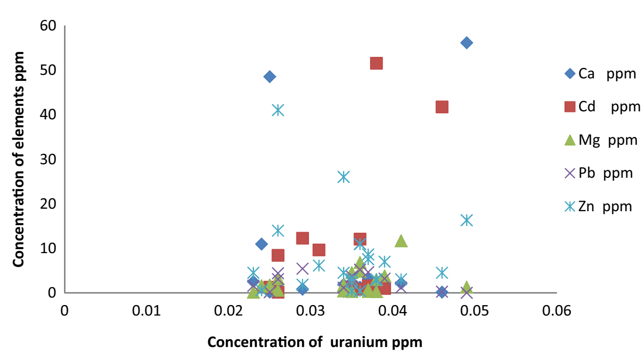


**Fig. 1** The relationship between the concentration of uranium and concentration of heavy metals in the toothpaste under study

**Table 7** The descriptive statistics of concentrations

| Variables | $N$ | Minimum | Maximum | Mean | Std. error | Std. deviation |
|---|---|---|---|---|---|---|
| Radon — Bq.m$^{-3}$ | 25 | 19.45 | 35.06 | 27.206 | 0.915 | 4.579 |
| Uranium —ppm | 25 | 0.02 | 0.04 | 0.034 | 0.001 | 0.005 |
| Radium — Bq.kg$^{-1}$ | 25 | 30.16 | 54.37 | 42.191 | 1.420 | 7.102 |
| Dose mSvy$^{-1}$ | 25 | 0.22 | 0.40 | 0.312 | 0.011 | 0.052 |

to be 35.06 Bq.m$^{-3}$ and 19.45 Bq.m$^{-3}$ with a mean of 27.206 ±4.579, those of $^{238}$U were 0.04 ppm and 0.02 ppm with a mean of 0.034 ± 0.005, those of $^{226}$Ra were 54.37 Bq.kg$^{-1}$ and 30.16 Bq.kg$^{-1}$ with a mean of 42.191 ± 7.102; the maximum average values and minimum average values of the annual effective dose of $^{222}$Rn of toothpaste were found to be 0.40 mSv y$^{-1}$ and 0.22 mSv y$^{-1}$with a mean of 0.312 ± 0.052. The results also showed that the concentration of radon gas varied from one sample to another, depending on the type of toothpaste.

The concentrations and annual effective dose for each toothpaste samples were non-normally distributed, the non-parametric Friedman test was used and a significant difference ($p > 0.000$) was found in all the data presented, as shown in Table 8.

## Correlation of the differences in laboratory data analysis

As shown in Table 9, significant strong positive correlations were found ($r = 1$, Pearson correlation, $p < 0.000$) in concentration of radon, concentration of radium, concentration of uranium, annual effective dose, pH, electrical conductivity, and heavy metals for 25 samples of toothpaste: correlation is significant at the $p < 0.01$ level.

The concentrations of these metals were not normally distributed, thus non-parametric tests were used. The Kruskal–Wallis test was the appropriate statistical test in order to find out the difference among results of these experiments. Significant difference ($p > 0.00$) was found in the toothpaste concentrations of these metals, but significant correlations were found between different heavy metals; this correlation provides valuable information on heavy metal sources. The data showed strong correlations between Ca and Cd ($r = 0.659$), Ca and Pb ($r = -0.735$), Mg and Pb ($r = -0.698$), and Mg and Mn ($r = 0.762$).

**Table 8** The statistical summary of data analysis

| Variables | Mean rank | $N$ | Chi-square | df | $P$ value |
|---|---|---|---|---|---|
| Radon | 3.00 | 25 | 24 | 3 | 0.000 |
| Uranium | 1.00 | 25 | 24 | 3 | 0.000 |
| Radium | 4.00 | 25 | 24 | 3 | 0.000 |
| Dose | 2.00 | 25 | 24 | 3 | 0.000 |

Friedman test

This study showed that the resulting concentrations from radon, radium and uranium in the selected samples were significantly lower than the limits in previous studies. Comparing these results with those of some studies:

> Khan et al. conducted a study to determine the radon content in the target samples of tobacco, tea, and toothpastes using the CR-39 detector and a technique slander: the average radon concentration in toothpastes was reported to be about 79.505 Bq.m$^{-3}$ in the range 26.38–413 Bq.m$^{-3}$ (Khan et al. 1988).
> In 1994, Salmon and others presented a study on the level and distribution of $^{210}$Pb and $^{210}$Po accompanying a low level of $^{226}$Ra concentration: the results show the distribution of these radioactive nuclei to be irregular, with a link between $^{210}$Po and abrasion of teeth by foreign objects, and a link between $^{226}$Ra and tooth pulp (Salmon et al. 1994).
> Hana submitted a study to determine the concentration of radium ($^{226}$Ra) and then extended it by calculating the number of effects of radon recorded by mediated CR-39 detection. The results showed that the radon content in the air space of the irradiation chamber in the samples was around 20.627–56.151 Bq.m$^{-3}$, while the values of effectiveness (A) were up to 0.678 x10$^{-3}$–1.846 x 10$^{-3}$

**Table 9** Correlations between laboratory data and demographic characteristics

| Variables | Laboratory data analysis | |
|---|---|---|
| Concentration of radon | Pearson correlation | 1.000 |
| | $p$ value | <0.000 |
| Concentration of radium | Pearson correlation | 1.000 |
| | $p$ value | <0.000 |
| Concentration of uranium | Pearson correlation | 1.000 |
| | $p$ value | <0.000 |
| Annual effective dose | Pearson correlation | 1.000 |
| | $p$ value | <0.000 |
| pH | Pearson correlation | 1.000 |
| | $p$ value | <0.000 |
| Electrical conductivity | Pearson correlation | 1.000 |
| | $p$ value | <0.000 |
| Heavy metals | Pearson correlation | 1.000 |
| | $p$ value | <0.000 |

Correlation is significant at the 0.01 level (2-tailed)

Bq, and effective radium content ranged between 0.103 and 0.471 Bq.kg$^{-1}$ and that of uranium was 7.5 to 34.5 ppb (Hana 2002).

Ali AAbo et al. conducted a study to determine the concentrations of radium and uranium by counting the number of radon tracks using the CR-39 plastic nuclear track detector. The levels/ranges of radon, radium, and uranium concentration were found to be 6.03 ± 0.70–101.08 ± 7.44) B.m$^{-3}$, 0.08 ± 0.009–1.416 ± 0.104 Bq.Kgm$^{-1}$ and 2.962 ± 0.345–49.620 ± 3.653 ppb respectively (Ali AAbo et al. 2014).

The results showed that the average dose of absorption in the studied samples was in a range about 0.3118 mSvy-1, which was below the range 3–10 mSvy-1 reported by ICRP (1993). The annual effective dose values represent a very small component of the total effective dose from natural sources (2.4 mSvy$^{-1}$) (Hana 2002), therefore, there is no evidence of health problems.

## Conclusions

Radon and heavy metal concentrations are of interest since this gives very important information in the monitoring of environmental contamination. This study showed that the maximum concentration of radon in toothpaste samples was lower than the global permissible limit of exposure to radon of 200 Bq.m$^{-3}$. From the results, we observed that variations of radon concentrations among various types of toothpaste can be attributed to differences in the nature and nuclei content of these samples, such as distribution of uranium and radium. Significant strong positive correlations were found ($r = 1$, Pearson correlation, $p < 0.000$) in concentration of radon, concentration of radium, concentration of uranium, annual effective dose, pH, electrical conductivity, and heavy metals. Monitoring the accumulation of these metals in toothpaste samples is very important, and helps to prevent the transportation of these toxic metals into the human system. The information gained from this study could help dentists and consumers in Malaysia to choose the best type of toothpaste for reduction of oral bacteria, thus improving dental health; it is important also from the viewpoint of general health and environmental problems. This research helped us identify whether the concentration rate of radiated elements found in toothpastes exceeded permitted global limits in order to avoid importing toothpaste types which do so.

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