NATURAL RADIOACTIVITY AND HEAVY METAL POLLUTANTS IN STAPLE FOODSTUFF AND HUMAN TEETH COLLECTED FROM SELECTED AREAS IN PENINSULAR MALAYSIA

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ABSTRACT

This study is aimed at the evaluation of natural radioactivity (natural decay chains of ^{238}U and ^{232}Th, and non-series long-lived natural radionuclide ^{40}K) and heavy metal (Pb, Cd, As, Hg, Cr, Cu, Sr, Al, Mg, Sb, Ba, Bi, Zn, Rb, Mn and Ni) exposures resulting from the consumption of staple foodstuffs such as rice, vegetables and marine animals. In addition, heavy metal levels in human teeth have been assessed as a bio-indicator of exposure to environmental pollution. The studied samples were collected from different locations across the Peninsular Malaysia. The radioactivity concentrations have been measured by γ-ray spectrometry using high resolution high purity germanium (HPGe) and scintillation NaI(Tl) detector and the heavy metals were determined by inductively coupled plasma-mass spectrometry (ICP-MS). The results showed that the radioactivity in different varieties of vegetable samples varied from 0.5–127Bq kg^{-1} for ^{226}Ra, 0.21–43 Bq kg^{-1} for ^{228}Ra (^{232}Th) and 56–2483 Bq kg^{-1} for ^{40}K. The radioactivity in rice in different areas varied from 1.5±0.4–2.8±0.7 Bq kg^{-1} for ^{226}Ra, 3.6±1.4–7.5±2.7 Bq kg^{-1} for ^{232}Th and 59.9±6.0–92.2±5.4 Bq kg^{-1} for ^{40}K. On the other hand, the activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in marine animals varied from 0.6±0.19–7.83±0.78 Bq kg^{-1}, 0.19±0.17–6.21±0.53 Bq kg^{-1} and 34±13–398.6±20.2 Bq kg^{-1}, respectively. The estimated soil-to-tapioca, -sweet potato and -rice transfer factors for ^{226}Ra and ^{232}Th were found much higher than the International Atomic Energy Agency (IAEA) reported values. The annual effective dose obtained due to ingestion of radionuclides via the consumption of the studied foodstuffs were found below the world average value (290 μSv y^{-1}) and the associated life-time cancer risk were also below the acceptable limit of 10^{-3} for radiological risk, thus discarding any significant radiological risks to the population of Peninsular Malaysia. The concentrations of heavy metals (mg kg^{-1}) in vegetables were found as: Pb (0.001–0.006), Hg (0.0019–0.005), Ni

(0.0004−0.008), Cr (0.0003−0.006), Al (0.06−58.1), Sr (0.01−0.72), Bi (0.05−0.80), Sb (0.1−1.2), Ba (0.001−0.034), Rb (0.02−0.13), Mn (0.012−2.1), Zn (0.01−0.59), Fe (0.05−1.6), Cu (0.001−0.06) and Mg (1.8−47.0), while in marine fishes were found as: As (0.3249−0.7485), Pb (0.02), Hg (0.0247), Al (3.115−4.489), Cr (0.0108−0.0516), Sr (19.56−27.68), Co (0.0007−0.0038), Cu (0.0712−0.1972), Mn (0.4372−1.841), Zn (5.607−15.35), Fe (3.546−4.857), Rb (0.0978−0.2312), Ba (0.2845−0.6597), Bi (1.598−4.143) and Mg (80.43−115.0). The concentrations of all the studied heavy metal in vegetables and fishes were below the daily limit on intake recommended by the international organizations, indicating yet pose no threat to public health. Conversely, trace amount of heavy metals were found in almost all teeth samples. Increased accumulation of Pb, Hg, As, Cr, Mn, Sr, Ba, Sb, Cu, Zn, Mg and Sn in teeth dentin was observed with respect to the teeth age. Some elevated levels of concentrations of heavy metals in the teeth dentin reflect the relation to pollution from industrial emissions and urbanization. Human teeth dentin, therefore, can be used as reliable bio-indicator of environmental pollution by heavy metals. Since higher concentration of radioactive- and heavy metal substances in the environment is undesirable, continuous monitoring should be undertaken to detect the concentration of radioactive and heavy metals in foodstuffs in order to take necessary radiological and dosimetric measures with the aim of minimizing the potential harmful effects of ionizing radiation.

ABSTRAK

Kajian ini bertujuan untuk menilai radioaktif semulajadi (rantaian pereputan semulajadi ^{238}U dan ^{232}Th), dan ^{40}K radionuklid pendedahan pada logam berat (Pb, Cd, As, Hg, Cr, Cu, Sr, Al, mg, Sb, Ba, Bi, Zn, Rb, Mn dan Ni) yang disebabkan oleh pengambilan makanan utama seperti beras, sayur-sayuran dan haiwan marin juga dikaji. Di samping itu, tahap logam berat dalam gigi manusia telah dinilai sebagai penunjuk-bio bagi dedahan kepada pencemaran alam sekitar. Sampel kajian telah dikumpulkan dari lokasi yang berbeza di seluruh Semenanjung Malaysia. Kepekatan bahan radioaktif telah diukur dengan menggunakan spektrometer sinar-γ Germanium huper-tulen (HPGe) beresolusi tinggi dan pengesan NaI(Tl) manakala logam berat dianalisis dengan induktif ditambah spektrometri jisim plasma (ICP-MS). Hasil kajian menunjukkan bahawa kepekatan radioaktif dalam pelbagai jenis sampel sayur-sayuran berbeza dari 0.5–127 Bq kg^{-1} untuk ^{226}Ra, 0.21–43 Bq kg^{-1} untuk ^{228}Ra (^{232}Th) dan 56–2483 Bq kg^{-1} untuk ^{40}K. Radioaktif dalam beras di kawasan yang berbeza berubah dari 1.5±0.4–2.8±0.7 Bq kg^{-1} untuk ^{226}Ra, 3.6±1.4–7.5±2.7 Bq kg^{-1} untuk ^{232}Th dan 59.9±6.0–92.2±5.4 Bq kg^{-1} untuk ^{40}K. Sebaliknya, kepekatan aktiviti ^{226}Ra, ^{228}Ra dan ^{40}K pada haiwan marin berubah dari 0.6±0.19–7.83±0.78 Bq kg^{-1}, 0.19±0.17–6.21±0.53 Bq kg^{-1} dan 34±13–398.6±20.2 Bq kg^{-1}, masing-masing. Faktor permindahan tanah ke ubi kayu, tanah ke keledak dan tanah ke beras bagi ^{226}Ra dan ^{232}Th dianggarkan lebih tinggi daripada nilai-nilai yang dilaporkan oleh Agensi Tenaga Atom Antarabangsa (IAEA). Dos tahunan berkesan disebabkan oleh pengambilan radionuklid dari bahan makanan yang dikaji didapati kurang daripada nilai purata dunia (290 μSv y^{-1}) dan risiko kanser seumur hidup juga di bawah had yang boleh diterima iaitu 10^{-3} untuk risiko radiologi, sekali gus menjadikan risiko radiologi tidak signifikan penduduk di Semenanjung Malaysia. Kepekatan logam berat (mg kg^{-1}) dalam sayur-sayuran didapati

sebagai: Pb (0.001−0.006), Hg (0.0019−0.005), Ni (0.0004−0.008), Cr (0.0003−0.006), Al (0.06−58.1), Sr (0.01−0.72), Bi (0.05−0.80), Sb (0.1−1.2), Ba (0.001−0.034), Rb (0.02−0.13), Mn (0.012−2.1), Zn (0.01−0.59), Fe (0.05−1.6), Cu (0.001−0.06) dan Mg (1.8−47.0), manakala dalam ikan laut didapati sebagai: As (0.3249−0.7485), Pb (0.02), Hg (0.0247), Al (3.115−4.489), Cr (0.0108−0.0516), Sr (19.56−27.68), Co (0.0007−0.0038), Cu (0.0712−0.1972), Mn (0.4372−1.841), Zn (5.607−15.35), Fe (3.546−4.857), Rb (0.0978−0.2312), Ba (0.2845−0.6597), Bi (1.598−4.143) dan Mg (80.43−115.0). Kepekatan semua logam berat yang dikaji dalam sayur-sayuran dan ikan adalah di bawah had pengambilan harian sebagaimanapun disyorkan oleh organisasi antarabangsa tidak menimbulkan ancaman kepada kesihatan orang awam. Sebaliknya, logam berat pada kepekatan surih ditemui pada hampir semua sampel gigi. Peningkatan terkumpul Pb, Hg, As, Cr, Mn, Sr, Ba, Sb, Cu, Zn, Mg dan Sn dalam dentin gigi didapati berkaiten dengan umur gigi. Peningkatan tahap kepekatan logam berat dalam dentin gigi mencerminkan hubungan kepada pencemaran dari pengeluaran perindustrian dan pembandaran. Dentin gigi manusia, boleh menjadi penunjuk-bio pencemaran alam sekitar oleh logam berat. Memandangkan kepekatan yang tinggi, bahan-bahan radioaktif dan logam berat dalam persekitaran tidak diingini, pemantauan berterusan perlu diambil untuk mengesan kepekatan radioaktif dan logam berat dalam makanan bagi memastikan keperluan dan dosimetri diambil bagi memastikan langkah radiologi buruk sinaran mengion.

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DEDICATIONS

This thesis is dedicated to my

Father

Md. Hossen Ali Khandoker

And

Mother

Mrs. Asiya Begum

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LIST OF SYMBOLS AND ABBREVIATIONS

AAS	:	Atomic absorption spectrophotometry
Ag	:	Silver
Al	:	Aluminium
APM	:	Airborne particulate matter
As	:	Arsenic
ATSDR	:	Agency for toxic substances and disease registry
Ba	:	Barium
Bi	:	Bismuth
Ca	:	Calcium
CAC	:	Codex alimentarius commission
Cd	:	Cadmium
CDC	:	Centers for disease control and prevention
CICAD	:	Concise international chemical assessment document
CNS	:	Central Nervous System
Co	:	Cobalt
Cr	:	Chromium
Cu	:	Copper
DNA	:	Deoxyribonucleic acid
DPP	:	Differential Pulse Polarography
EC	:	Electron capture
EC	:	European commission
EFSA	:	European food safety authority
EDI	:	Estimated daily intake
EDXRF	:	Energy dispersive X-ray fluorescence

ELCR	:	Excess lifetime cancer risk
ETAAS	:	Electro-thermal atomic absorption spectrometry
EU	:	European union
FAO	:	Food and agriculture organization
Fe	:	Iron
Ge	:	Germanium
GFAAS	:	Graphite furnace atomic absorption spectrometry
GI	:	Gastro-intestinal
Hg	:	Mercury
HG-AFS	:	Hydride generation atomic fluorescence spectrometry
HPGe	:	High purity germanium
IAEA	:	International atomic energy agency
ICP-AES	:	Inductively coupled plasma atomic emission spectrometry
ICP-MS	:	Inductively coupled plasma mass spectrometry
ICP-OES	:	Inductively coupled plasma optical emission spectrometry
ICRP	:	International commission for radiological protection
IDPH	:	Illinois department of public health
K	:	Potassium
keV	:	Kilo electron volt
LA-ICP-MS	:	Laser ablation inductively coupled plasma mass spectrometry
Li	:	Lithium
MDA	:	Minimum detectable activity
MeV	:	Mega electron volt
Mg	:	Magnesium
Mn	:	Manganese
Na	:	Sodium

NAA	:	Neutron activation analysis
NCRP	:	National council on radiation protection
Ni	:	Nickle
NIH	:	National institute of health
NORM	:	Naturally occurring radioactive material
Pb	:	Lead
PIXE	:	Proton induced X-ray emission
R_fD	:	Reference oral doses
Sb	:	Antimony
Se	:	Selenium
SEPA	:	Scottish environment protection agency
Si	:	Silicon
Sn	:	Tin
Sr	:	Strontium
TENORM	:	Technologically enhanced naturally occurring radioactive material
TDS	:	Total diet study
TF	:	Transfer Factor
UNSCEAR	:	United nations scientific committee on the effects of atomic radiation
USEPA	:	United state energy protection agency
V	:	Vanadium
WHO	:	World health organization
XRFS	:	X-ray fluorescence spectrometry
Zn	:	Zinc
α	:	Alpha
β	:	Beta
γ	:	Gamma

LIST OF APPENDICES

CHAPTER 1: INTRODUCTION

1.1   Research background

NORMs which refers to naturally occurring radioactive materials have always been a part of our daily life and still remain so. Our planet, its atmosphere and practically all living and non-living things even the human body contains NORMs (Hunter-Smith, 2012). Radionuclides found in nature are usually categorized into two distinct forms: Cosmogenic or Extra-terrestrial– originated from the high-energy cosmic ray and/or particles (from Sun, stars and from galactic and intergalactic plasma) incident on the earth's atmosphere, and Terrestrial origin – arising from the earth crusts and earth born materials (Al-Sulaiti, 2011; Canbazoğlu & Doğru, 2013; Faanu, 2011; UNSCEAR, 2000). Terrestrial radioisotopes are also mentioned to as primordial radionuclides because of their source in the solar nebula or primordial dust cloud.

Among the natural radionuclides, uranium, thorium and their progenies are ubiquitous; mostly contribute to the human radiation exposure. The three main decay series exist in nature headed by ^{238}U with a half-life of 4.5 billion years, ^{235}U with a half-life of 700 million years, and ^{232}Th with a half-life of 14.1 billion years. A detailed information on their decay schemes and relevant numerical values are presented in Appendices A and B. In each of the decay chain headed by ^{238}U, ^{235}U and ^{232}Th, the nuclides decay by way of alpha or beta particles followed by gamma radiation until to reach a final and most stable ^{206}Pb, ^{207}Pb and ^{208}Pb nuclides, respectively (Al-Sulaiti, 2011; Hunter-Smith, 2012). Interactive chart of nuclides can be seen in the appendix C (https://nucleus.iaea.org/Pages/nu-dat-2.aspx).

Around 2.7 mg kg^{-1} of uranium is present in the earth's crust (Faanu, 2011). Uranium and Radium is usually more soluble than the Thorium in a naturally uninterrupted environment. In the natural environment, uranium contains 99.28 % of

^{238}U, 0.72 % of ^{235}U and trace amounts of about 0.0058 % of ^{234}U (Faanu, 2011). Upon their decay chain, ^{238}U undergoes fourteen radioactive decay steps resulting in the release of eight α-particles with maximum energy of 7.687 MeV and six β-particles with maximum energy of 1762.6 keV (Faanu, 2011; https://nucleus.iaea.org/Pages/nu-dat-2.aspx). Throughout the decay scheme, gamma photons are also emitted at energy range of 46.53 - 2447.86 keV. In the ^{238}U series, the decay chain segment starting from radium (^{226}Ra) is radiologically the most important because it behaves chemically similar to calcium, being incorporated on bone surfaces and areas of mineral metabolism therefore, reference is often made to ^{226}Ra instead of ^{238}U.

Monazite, a rare-earth material and thorium phosphate mineral are the primary source of thorium. Small quantities of thorium is also found in most rocks and soils, where it is about four times more abundant than uranium (Faanu, 2011). The solubility of thorium is extremely low in natural waters and is totally transported in particulate form. It is adsorbed on the surface of clay minerals. During the decay chain, ^{232}Th follows 11 radioactive decay stage emitting mainly seven α-particles with maximum energy of 8.784 MeV and five β-particles with maximum energy of 834.2 keV (https://nucleus.iaea.org/Pages/nu-dat-2.aspx). During the course of ^{232}Th decay chain, gamma rays are also emitted at energies of 13.51 to 2614.533keV. ^{232}Th isotope undergoes α-decay to form ^{228}Ra which is equally soluble to ^{226}Ra and radiologically important, consequently, reference is often made to ^{228}Ra instead of ^{232}Th.

Potassium is normally distributed in the earth's crust and presents in all environmental media including foodstuffs and even in the human body. Under normal conditions, ^{40}K is the most abundant naturally occurring radioactive constituent within the human body (Asaduzzaman, et al., 2015; Rahman & Faheem, 2008). The isotopic abundance of ^{40}K (the radioactive isotope of terrestrial importance) is 0.0118 % and has

a specific activity of 31.4 Bq g^{-1} of natural potassium (Bakım & Görgün, 2014; Kathren, 1998). Naturally occurring ^{40}K undergoes β$^-$ decay to produce stable ^{40}Ca (88.8%) and the remaining 11.2% of ^{40}K decays by electron capture (EC) and by positron emission to stable ^{40}Ar (James et al., 2013). During the decay process, ^{40}K follows 100 disintegrations, out of them, 89 results in the release of β-particles with maximum energy of 1.33 MeV and 11 follows in the emission of characteristic gamma photons with maximum energy of 1.46 MeV (https://nucleus.iaea.org/Pages/nu-dat-2.aspx ; James et al., 2013). This gamma line is generally use to detect and measure ^{40}K by the way of gamma spectrometry. The decay scheme of ^{40}K is presented in the appendix A. Among the natural radionuclides, ^{40}K is the most significant isotopes contributing maximum portion to the natural radioactivity in the environment (James et al., 2013). ^{40}K is the main contributor to the ingestion dose, with a contribution more than 90% of total ingestion dose from natural radioactivity which is about 0.21mSv y^{-1} (James et al., 2011).

Furthermore, heavy metals are also ubiquitous in nature; however, their presence in the environment can be enhanced as a result of anthropomorphic activities. Environmental pollution by heavy metals is a major concern all over the world. By definition, those metals having specific gravity of ≥5g/cc are referred to as heavy metal (Al-Jubouri & Bashbosh, 2012; Jarup, 2003). However, metals or metalloids that are well-known for their potential toxicity, particularly in environmental perspectives are classified as heavy metal (Srivastava & Goyal, 2010). Heavy metals are also regarded as trace elements because of their existence in trace quantities in numerous environmental media (e.g., food, water and air). The incidence of heavy metal in both the terrestrial and aquatic environment is reasonably important in assessing potential threats for human health once exist in terrestrial as well as aquatic food chain (Arruda-Neto et al.,

2010; Asaduzzaman et al., 2015; Bhuiyan, Dampare, Islam, & Suzuki, 2015; Khandaker, et al., 2015; Zheng et al., 2013). Some metals/elements including Calcium (Ca), Sodium (Na), Magnesium (Mg), Iron (Fe), Potassium (K) etc. are known as macro-minerals, are required in good quantities for proper metabolism and functioning the organs of the body while, some other elements such as Manganese (Mn), Copper (Cu), Zinc (Zn), Chromium (Cr), Selenium (Se), Lithium (Li), Germanium (Ge), Cobalt (Co) etc. are among the essential micronutrients metals that are needed in minuscule quantities in human body of not more than a few mg/day (Li et al., 2015; Lokeshappa, Shivpuri, Tripathi, & Dikshit, 2012; Lu et al., 2015; Santos, Lauria, & Porto da Silveira, 2004). Moreover, metals like Strontium (Sr), Nickle (Ni), Silicon (Si), Vanadium (V), Tin (Sn), Silver (Ag), Aluminum (Al) etc. are possibly beneficial micro-minerals, even though their beneficial role in human body yet to be established (Lokeshappa et al., 2012; Santos et al., 2004; Tchounwou, Yedjou, Patlolla, & Sutton, 2012). Metals that are needed in minute quantities or trace level (micronutrients) in human bodily functions become toxic and can cause adverse health effects, if they exist in greater amounts (Asaduzzaman et al., 2015; Islam, Ahmed, Habibullah-Al-Mamun, & Masunaga, 2015; Khan et al., 2014; Lokeshappa et al., 2012). Even Ca and Na are rather toxic in excess. On the other hand, toxic heavy metal consists of a group of minerals (e.g., Lead (Pb), Cadmium (Cd), Mercury (Hg), Arsenic (As), Barium (Ba), Antimony (Sb), Bismuth (Bi) etc.) that have no known routine biological function in the human body and indeed detrimental even at miniscule quantity (Alina et al., 2012; Asaduzzaman et al., 2015; Hajeb, Sloth, Shakibazadeh, Mahyudin, & Afsah-Hejri, 2014; Lokeshappa et al., 2012; Tchounwou et al., 2012). Overindulgences, deficiencies or imbalances in the supply of minerals from alimentary sources can cause significant malicious impact on the health of human life (Santos et al., 2004; WHO, 1996). Content of metals beyond the maximum permissible levels can lead to a variety of,

cardiovascular, renal, and neurological impairment as well as bone diseases, mutagenic effects, damaged central nervous function, lungs, kidneys, liver, and other vital organs (Asaduzzaman et al., 2015; Jolly, Islam, & Akbar, 2013).

However, environmental heavy metal can be assessed by chemical method, by physical method and by biological indicators (Kamberi, Kocani, & Dragusha, 2012). Presently, there has been a curiosity among the researchers on the use of human bio-indicators such as blood, urine, bone, teeth, nails from finger and toes, hair and saliva to evaluate the environmental pollution by toxic heavy metals (Abdullah et al., 2012; Arruda-Neto et al., 2010; Barton, 2011; Brown et al., 2004; Kamberi et al., 2012; Kantamneni, 2010). Since the metals deposited in teeth during formation and mineralization processes are to a large extent retained, human teeth receive a significant attention as the indicators of the heavy metal exposure. As a bio-indicator of environmental exposure, teeth cover a much longer lifespan as substrates for toxicological analyses, better than bone, blood, nail, saliva, or hair (Arora & Austin, 2013; Barton, 2011; Kang, Amarasiriwardena, & Goodman, 2004). Several studies showed that teeth are superior to blood, nail, saliva, or hair as an indicator of chronological metal exposure from environment because the losses from teeth are much slower as there is no significant turnover of apatite in teeth, as in other biological indicators (Alomary, Al-Momani, & Massadeh, 2006; Arruda-Neto et al., 2009; Kamberi et al., 2012; Kantamneni, 2010; Kumagai, Fujita, Endo, & Itai, 2012).

Present study deals to obtain a comprehensive information of radioactive- and heavy metals, e.g., the determination of the prevailing concentrations of radioactive- and heavy metals in foodstuffs, and their accumulation (specifically heavy metals) in human teeth upon the consumption of foodstuffs. More specifically:

1. Natural radioactivity in staple foodstuffs;

2. Heavy metal pollutants in staple foodstuffs and
3. Heavy metal pollutants in human teeth; a bio-indicator of metal exposure to environmental pollution.

1.1.1   Natural radioactivity in foodstuffs

Uranium and thorium are the most encountered radioactive materials in the earth crust that are associated with igneous and sedimentary rocks from which the soil originates (Faanu, 2011). The concentration of radionuclides in earth surface, specifically in soil, depends on the rock types, the concentrations are lower in sedimentary rock and higher in granitic igneous rocks (UNSCEAR, 2000). Troposphere which is the lower region of the atmosphere adjacent to the earth's surface (spreads to around 15 km in altitude) is the core medium of radioactive releases from NORM industries (Eisenbud & Gesell, 1997). This layer comprises about 75% of the atmospheric mass and nearly all of its moisture and dust wherein the distribution of radionuclides affects mainly on the vertical temperature gradient, pressure, wind speed and direction (Eisenbud & Gesell, 1997). If the wind is insufficient to keep the radionuclides suspended in air, the rainout (droplet formation of the rainfall around aerosols) and washout (scrubbing) action remove aerosols from the troposphere and settle them on the earth's surface and may integrate on soil, water, edible flora and eventually get into the human food chain and finally infiltrate into the human body via ingestion and inhalation (Eisenbud & Gesell, 1997).

In most natural substances, the concentration of NORM is too low to create significant radiological risk, and is usually considered as negligible. Materials that may comprise any of the NORMs for example ^{238}U, ^{235}U, ^{232}Th and their radioactive decay progenies as well as single decay radionuclides, mainly ^{40}K, are disrupted by various human activities. Consequently, their concentration in the environmental media

including land produced and aquatic (marine animals) foodstuffs can be enhanced through anthropomorphic activity, including agricultural inputs (e.g. use of fertilizers, fungicides, insecticides and herbicides), mining, mineral and metal processing, rapid urbanization, industrial activity such as coal-, oil-, and gas-exploration and exploitation, combustion of fossil fuel, energy production, radioactive wastes and industrial effluents disposal, further added to by artificial sources of radioactivity introduced into the environment which beyond the scope of this work (Amin et al., 2013; Asaduzzaman et al., 2015; Awudu et al., 2012; Hunter-Smith, 2012; Khandaker, et al., 2015a, 2015b; UNSCEAR, 2008b; USEPA, 2011). The radionuclides in the decay series are essentially in radiological equilibrium with their parents in the naturally undisturbed environments. But, the aforementioned human activities or natural catastrophe can break this equilibrium, resulting in either an enhancement or reduction of radionuclides concentrations compared to the original matrix (Faanu, 2011; UNSCEAR, 2000).

The presence of radionuclides in terrestrial and aquatic environments may perhaps occur from the emission of industrial solid and liquid effluents into that environment. If the solid and liquid effluents directly discharges into the land, then they may transfer to the food chain through soil-crop plant transfer, but in the case of straight release into the land, radionuclides tend to be more condensed and localized. On the other hand, if solid and liquid effluents discharge into the aquatic environment, distribution of radionuclides into that environments depends geographical and metrological factors for example, place and time of discharge, depth of water at which the effluent/pollutant being released, type of sea/riverbed, tidal and wind factors, temperature and physical state of the pollutant etc. (Eisenbud & Gesell, 1997). The solid state wastes may be attached to the surface of the aquatic plants, be settled on the sea/riverbed or be filtered by the aquatic animals. If the discharge is in liquid form, the adjourned inorganic and organic

matters remediate the contaminant by sorption, or aquatic animals and plants might be accrue it through dermal absorption and/or consumption/uptake.

The radioactive contamination in the terrestrial and aquatic environment might be occur via natural radionuclides from the earth's crusts, soil, rock, oceans, rivers and atmosphere and/or by airborne deposition caused by atmospheric release on the ecosystem. It is then integrated into soil sub-compartment, absorbed by plants through rooting system, transfer it to various parts of the plants eventually edible portion and get into the food chain (Asaduzzaman, Khandaker, Amin, & Mahat, 2015; Pulhani, Dafauti, Hegde, Sharma, & Mishra, 2005; Shanthi, Thanka Kumaran, Gnana Raj, & Maniyan, 2012; Vandenhove et al., 2009). Particularly ^{40}K, ^{238}U and ^{232}Th and their various decay products are the most common radionuclides in foods and water (Asaduzzaman et al., 2015). These radionuclides appear in plants either through direct atmospheric interception onto aerial parts of the plant surfaces or from the re-suspended material and absorbed metabolically by the plant surfaces and indirectly through the uptake of radionuclides from soil via the root system (Asaduzzaman et al., 2014; Chauhan & Kumar, 2015; Jabbar et al., 2010; James et al., 2011; Karunakara et al., 2013; Vandenhove et al., 2009). However, the root uptake path is more effective because soil is the main reservoir of natural radionuclides of ^{40}K, ^{238}U and ^{232}Th. The solubility of radionuclides in water enhance their transmission in soil and hence root uptaking.

Soil-to-plant and plant-to-human body is one of the foremost corridors for transmission of radionuclides to human being (Aswood, Jaafar, & Bauk, 2013; IAEA, 1982, 2010; Jabbar et al., 2010). After uptake by root, radionuclides are shifted to plant along with other nutrients or minerals necessary for their growth; accumulate in several parts including the edible portions and would lead to endless radiation dose to man once consumed (Carini, 2001; Pulhani et al., 2005). Therefore, it is important to study the

spatial distribution of natural radioactivity in soil and related radiation exposures through specific land produced food-stuffs. Soil-to-plant transfer factor (TF) is one of the key parameters in calculation of radionuclides concentration in agrarian crops, also extensively used for the estimation of internal radiation dose to humans (Asaduzzaman et al., 2014; Tsukada, Hasegawa, Hisamatsu, & Yamasaki, 2002). However, the rate of root uptake (TF) and distribution of radionuclides in plants depend on several factors such as: (i) the form in which the activity enters or present in soil (i. e., as particles, aerosol or solution); (ii) the physicochemical properties of radionuclides and its distribution coefficient; (iii) the type and physicochemical characteristics of the soil environment (such as soil texture, pH, exchangeable Ca and K, the kind and amount of clays, and organic matter contents); (iv) metabolic requirements of the plant; (v) crop management practices (e.g., irrigation, application of fertilizer, ploughing, limiting etc.); (vi) type of crop (crop species and variety, and cultural practices); (vii) Climate condition; (viii) time after contamination etc. (Asaduzzaman et al., 2014; Asaduzzaman et al., 2015; Ewers, Ham, & Wilkins, 2003; IAEA, 2010; Pulhani et al., 2005; Shanthi et al., 2012).

In the marine environment, radionuclides have been released from a multiplicity of sources, both the natural and artificial. Radioactivity in the marine environment is contributed to by the natural processes of weathering and mineral recycling of terrestrial rocks, seabed movement arising from under sea earthquakes and submerged volcanic activity (Khandaker et al., 2015a). Various types of geological minerals as for example igneous rocks and ores which often contain increased levels of natural radionuclides, as a result ^{238}U) and ^{232}Th) are infiltrate to marine water by the process of leaching action (Abbasisiar, Hosseini, & Heravi, 2004). Human activities such as fossil fuel, coal-burn electricity power plants, natural gas and oil production, mining and mineral and ores processing, phosphate processing industries (such as fertilizer, chemicals, detergents),

biofuel , post nuclear disposal of radioactive and industrial waste, underwater nuclear device tests, accident including leakage from nuclear power plants (for example Daiichi nuclear power plant accident-2011, Fukushima, Japan), reprocessing of spent fuel etc. are also known to increase the radioactivity in the marine environment, with bioaccumulation of marine animals, therefore acknowledged growing concern to monitor the content of radionuclides in marine lives particularly in edible fishes, molluscs and crustaceans (Al-Qaradawi et al., 2015; Alam & Mohamed, 2011; Amin et al., 2013; Khandaker et al., 2015a, 2015b; Mayeen Uddin Khandaker, Wahib, Amin, & Bradley, 2013). While radionuclides present in the ocean shows complex behaviour (for example, ^{238}U is fairly soluble in sea water whereas ^{232}Th is almost insoluble but its daughter product ^{228}Ra is soluble in water), they can be transported in the oceanic environment in the following ways: dissolved in the seawater, attached to the plankton suspended in seawater and to the sediment, and radiologically pollutes the marine biota (Carvalho, Oliveira, & Malta, 2011). Marine animals has the propensity to burrow down in the bottom sea sediments and rocks, filtering and/or consuming the organic particles and algae along with tiny fishes and planktons which may cause to boost the uptake of radionuclides from their surroundings (Y.M Amin et al., 2013; Khan, Benjamin, & Godwin, 2011; Khan & Wesley, 2011; Khandaker et al., 2015a).

Food is an essential constituent for life support and none can imagine the life on Earth without food. Malaysia, a rapidly growing industrial country, is susceptible to pollution via large-scale industrial engagements and associated human activities. One particular concern is the potential impact upon the quality of locally resourced rice, vegetables and marine lives that contain important nutrients necessary for good health, forming an essential part of the Malaysian diet. A significant amount of radionuclides may attain human body via the ingestion (food chain, water etc.) and inhalation that may get into the terrestrial and aquatic foodstuffs from natural and human activities.

The presence of natural radioactivity in foodstuff results internal radiation exposures to humans and forming the principal contributor to the ingestion radiation dose received by the general public (Awudu et al., 2012; Korkmaz Gorur, Keser, Akcay, & Dizman, 2012). However, the elevated levels (above the recommended threshold limit) of radioactivity ingested through food supply could be a significant source of human exposure and could cause in radiological hazards. For that reason, it is essential to assess the level of radioactivity in the major human foods and the associated radiation dose for the food safety and minimize the radiological risk.

There are several possible pathways that transfer the radionuclides to the human food chain such as atmospheric, terrestrial (soil sub-compartments) and aquatic paths, each contributing human exposure by externally and/or internally (Al-Sulaiti, 2011; Asaduzzaman et al., 2014). The main pathways of human radiation exposure through the environment can be seen schematically in appendix D. After ingestion to human body via food web (especially rice, fish, vegetables, water etc.), a fraction of the absorbed radionuclides can come out from the body via urinary and facial excretion before intervening their physical half-life. Rest of the radioisotopes may persist in the body organ and became a continuous source of radiation exposure for the rest of individual's lifecycle. When a radionuclide is admitted into the blood along with other body fluids by way of ingestion, inhalation or by any means, it circulates within the systemic circulation in the individual's blood, tailed by transfer to various body organs and distributed uniformly throughout the body tissues (ICRP, 2002). Particularly alpha and beta emitters are of concern because they transfer huge amounts of energy directly to human tissues once ingested or inhaled, creating damage to the molecular structure of the DNA (deoxyribonucleic acid) in the human cell because of long-term exposure, consequently biological effects of radiation exposure such as cancer might be occurred

if this damage is unrepaired and continuous to divides into new mutated cells (ICRP, 2002; UNSCEAR, 2000).

^{226}Ra and ^{228}Ra, the radiologically most important radionuclides (present in soil), is uptake by plants along with their nutrients and finally come into human body through foodstuffs and have a tendency to follow chemically homologous element calcium metabolic process to become deposited in bones, teeth and mineral metabolism areas (Hashim & Najam, 2015; IDPH, 2008; Shakir et al., 2010). Majority (around 80%) of the ingested radium is quickly excreted from the body through faeces, and rest of them is absorbed into the bloodstream from the gastro-intestinal (GI) tract and then concentrated heterogeneously in bone following the behaviour of chemically similar calcium and reside in the bones throughout the entire lifespan causing continuous radiation exposure to the organs (Argonne National Laboratory, 2005; Asaduzzaman et al., 2015; California Environmental Protection Agency, 2006; EPA, 2015). Detectable amounts of ^{226}Ra and ^{228}Ra are found in vegetables, rice, cereals and fish, whereas the concentration is quite high in shellfish. A little amount of environmental radium can lead to accumulate successively in bone tissue can cause damages of bone marrow and be able to mutate bone cells that can produce cancerous cell (Jankovic, Todorovic, Todorovic, & Nikolov, 2012). Long term body exposure to low levels of radium creates severe health effects including depression of the immune system, anemia, cataracts, sores, bone, liver and breast cancer and several other body disorders (Hashim & Najam, 2015; IDPH, 2008; Mahur, Khan, Naqvi, Prasad, & Azam, 2008). Subsequent ingestion by any means, uranium quickly seems in the bloodstream (depending its solubility) and largely attached to the red blood cells and then accumulates in the skeleton (major place of accumulation), kidneys and the lung, while a lesser amount is absorbed by liver (Akhter, Rahman, Orfi, & Ahmad, 2007; Al-Sulaiti, 2011; Hakonson-Hayes, Fresquez, & Whicker, 2002; Hashim & Najam, 2015; La Touche, Willis, & Dawydiak, 1987).

Total body burden of uranium in human beings is estimated as 40 µg, whereby about 40% of this being found in muscles, skeleton contains 20% and 10%, 4%, 1% and 0.3% in the blood, lungs, liver and kidneys, respectively (Igarashi, Yamakawa, & Ikeda, 1987). It is estimated that approximately 15% of the ingested uranium comes from the consumption of foodstuffs and high ingestion of this isotope may cause clinical effects in human organ including damage to the kidneys (Hashim & Najam, 2015). Similarly, following ingestion into the human body, thorium generally deposited in tissues of skeleton, lungs liver and other tissues and have a tendency to retain in skeleton as well as accumulated on the surface of the bone whereby it has a long biological half-life which is about 22 years and create radiation damage to the respective organs via decay progenies (Akhter et al., 2007; NCRP, 1998). Furthermore, potassium is homologous to sodium and uniformly distributes to the whole body has a tendency to accumulate in muscle and cardiac rhythm (Akhter et al., 2007; Al-Sulaiti, 2011). The content of potassium (biological half-life is 300 days) is homeostatically controlled in the body and is little influenced by environmental variations and consequently its level in the body is reasonably constant (Al-Masri et al., 2004; Asaduzzaman et al., 2015; Scheibel & Appoloni, 2007; UNSCEAR, 2000).

1.1.2 Heavy metal pollutants in foodstuffs

In addition to pathways from natural radioactivity in the terrestrial and marine foodstuffs into the human diet, a further concern is heavy metal contaminants through consumption of foods leading to potential health risks via long-term exposure. Therefore, it is important to evaluate the heavy metals in human foodstuffs as well as human populations to assess the degree of body poisoning with toxic metals. Heavy metals can be toxic for humans, in particular those that are not metabolized by the body

and which consequently accumulate in the soft and calcified tissues (Asaduzzaman et al., 2015; McSheehy Ducos, Hamester, & Godula, 2010).

Various industrial, agricultural, domestic, technological and medical applications of heavy metals have led to their extensive distribution in the environment and dramatically increased the human exposure; growing ecological and worldwide public health issue over their potential effects on human health and environment (Tchounwou et al., 2012). Heavy metals are bio-accumulated and bio-transferred together by natural and anthropogenic sources. Although heavy metals occurs naturally in the environment that are originated from the earth's crust and distributed in the environment by natural geogenic processes (e.g., weathering, volcanic eruptions, bacterial activity, erosion etc), most of the environmental pollution and human exposures arise from anthropogenic activities, as for instance mining, smelting procedures, combustion of fossil fuel, steel and iron industry, industrial production and use, domestic and agricultural use of metals, chemical and metallurgical industry, municipal waste, forest fire, coal burning in power plants, petroleum combustion, plastics, textiles, e-waste, paper processing plant, wood preservation, application of metal based pesticides and herbicides, swage sludges in agriculture etc. (Florea & Buesselberg, 2006; Liu, Liu, Wang, & Wang, 2015; Nazir et al., 2015).

Contamination of water, soil and air is the main factor in the transfer of heavy metals to human and animal foodstuffs. The heavy metals presence in foods is frequently determined by its availability in the soil. The occurrence of heavy metals in the environmental matrix such as in foodstuffs may enhance due to rapid industrial growth, advances in agricultural chemicals such as fertilizer, pesticides, fungicides, herbicides, and/or urbanization process (Orisakwe, Blum, Sujak, & Zelikoff, 2014).

Heavy metals can enter into the human body through the consumption of foodstuffs, inhalation of contaminated air and by dermal contact via the skin (Amr, 2011; Zheng et al., 2013). Consumption of vegetables is one of the major roots of human exposure to heavy metals (Liu et al., 2015). Contamination of vegetables and other land produced food crops occurs via the plant uptake of pollutants once grown up in polluted soil, waste water irrigated soil, and/or surface contamination through aerial deposition. Literature revealed that the use of heavy metal contaminated waste water for irrigation over long period of time rises the heavy metal contents in soils which may uptake by plants and distributed by different parts of plants including edible parts, and ultimately enter into the human body following their consumption. Contamination of plants as well as foods and water by heavy metals is one of the main issues to be faced all over the world due to their non-biodegradability and toxic effects. Long term exposure to heavy metals and its compounds could cause in dysregulation of cellular pathways resulting subsequent poisonousness that may interfere with function of the central nervous system, metabolism of cells. Ingestion of heavy metals through the food chain could modify the metabolism of other essential trace elements, as for example Cu, Zn, Fe and Se (Florea & Buesselberg, 2006).

Food is one of the major ingestion source of toxic heavy metals by humans (Hajeb et al., 2014; Millour et al., 2012). Pollution of air, water and soil are contributing to the occurrence of heavy metals, such as lead, cadmium, mercury, arsenic, chromium, manganese, aluminum, tin, copper, iron, antimony, strontium, bismuth, nickel etc, in human foodstuffs (Orisakwe, Nduka, Amadi, Dike, & Bede, 2012). Excessive intake of dietary heavy metals results development of several severe health problems. Moreover, ingestion of heavy metal contaminated food may extremely reduce some essential nutrients in the body resulting a decline in immunological defenses, impaired psycho-

social behaviours, intrauterine growth retardation, malnutrition related disabilities and high prevalence of upper gastrointestinal cancer (Orisakwe et al., 2012).

The presence of toxic and/or heavy metals in the environment is rather significant in assessing possible risks for human health once present in the food chain. The rate of human consumption of metal is directly associated to alimentary habits and their content in foodstuffs. The concentration of heavy metals in foodstuff influenced by the properties of soil, such as pH, organic matter content, clay mineralogy that can affect the bioavailability of metallic elements. Vegetables and other agricultural crops can uptake high levels of metals from polluted soil, water and air. In addition to environmental pollution, a matter of concern is the application of fertilizers, insecticides, herbicides and fungicides to crops, which may contain various metal and increase the content of metal in soil and water as well as food crops (Santos et al., 2004). Moreover, the chemical and physical forms in which metals are distributed may rise the metal availability for plants and increase the concentrations of metal in vegetation.

The aquatic especially marine environments contamination via heavy metals is a great global concern due to their toxicity, long term persistence, non-biodegradability and succeeding accumulation in aquatic/marine habitats (Bhuiyan et al., 2015; Hajeb et al., 2014). The marine environments is contaminated by untreated industrial effluents, rapid urbanization (waste stream of urbanization), discharge of municipal wastes, shipping activities, increasing tanker trafficking, oil-gas exploration, operation of power plant and fishing activities (Alina et al., 2012; Hajeb et al., 2014; M. U. Khandaker et al., 2015a). Furthermore, natural disaster, natural processes like weathering and erosion of parent materials can intensify the heavy metal loads in marine environment. The habitats contaminated by heavy metals may perhaps amass in microorganisms and

benthic flora and fauna that may come into the human food chain and pose a significant threat to human health (Bhuiyan et al., 2015). Marine animal has tendency to burrow down in the bottom sea sediments and rocks, filtering on organic particles and algae along with tiny fishes and planktons, which may lead to increase the uptake of heavy metals from their surroundings (Khandaker et al., 2015a). Heavy metal pollutants through consumption of marine products, with bioaccumulation leading to potential risks by way of long term exposure (Elnabris, Muzyed, & El-Ashgar (2013).

1.1.3 Heavy metal pollutants in human teeth; a bio-indicator of metal exposure to environmental pollution

Human civilization is currently exposed to the highest levels of heavy metals in recorded history. Environmental pollution is a major problem in Malaysia, not only due to its rapid industrialization and urbanization but also from wind-borne pollution from bordering countries (e. g., Indonesia) (Afroz, Hassan, & Ibrahim, 2003).

Heavy metals can accrue in the environment such as in the human food chain, entering the human body through the normal ingestion of food and water or by deliberate consumption of soil, through the skin or by inhalation (Amr, 2011; Asaduzzaman et al., 2015; Bhuiyan et al., 2015; Keshavarzi et al., 2015). Once in the body they accumulate in various organs including calcified tissue like bones and teeth and pose a risk to human health due to their toxicity and long-term persistence (Alina et al., 2012; Alomary et al., 2006; Barton, 2011; Lu et al., 2015). The ingestion of heavy metals by human beings is directly connected to dietary habits and lifestyle (Santos et al., 2004). Knowledge of the amount of heavy metals in the body can provide significant information on potential environmental exposure, either at their place of work or elsewhere; their dietary habits and their health (Castro, Hoogewerff, Latkoczy,

& Almirall, 2010; Kern & Mathiason, 2012). Therefore, it is imperative to assess heavy metals in human organs to evaluate the degree of poisoning by toxic metals.

Evaluation of environmental pollution can be performed using physical and chemical methods and with bio-indicators (Kamberi et al., 2012). Bio-monitoring of metal level in human organs gives a sign of an individual's current body burden, which is a function of recent and/or past environmental exposure (Kantamneni, 2010). Thus, the right choice of and development of suitable biomarkers to assess heavy metal exposure is of crucial importance, for primary prevention, health care management and decision making in public health (Arora et al., 2006; Barbosa, Tanus-Santos, Gerlach, & Parsons, 2005). Recently, there has been growing interest among researchers on the use of human bodily fluids and organs such as blood, urine, bone, teeth, nails, hair and saliva as bio-indicators to investigate environmental pollution by means of toxic heavy metals (Abdullah et al., 2012; Arruda-Neto et al., 2009; Arruda-Neto et al., 2010; Barton, 2011; Brown et al., 2004; Kamberi et al., 2012; Kantamneni, 2010).

Each of them is associated with some advantages and limitations. Blood and urine data reflects information on recent exposures (Arruda-Neto et al., 2009; Arruda-Neto et al., 2010). Till now, blood-lead levels are widely used as a marker of Pb exposure. The half-life of Pb in blood is very short (approximately 28–30 days), thus it is not a reliable indicator of chronic exposure. Hair and fingernails are regarded as medium-range bio-monitoring agents, associated with exposure times from a few months to years (Arruda-Neto et al., 2009; Arruda-Neto et al., 2010). Moreover, they are habitually contaminated by external agents, such as dust in air, hair coloring, shampoo, nail polish etc., consequently these samples are often impure and not ideal as bio-indicators (Barbosa et al., 2005; Kern & Mathiason, 2012; Kumagai et al., 2012). Calcified tissues such as bone and teeth have a high affinity to accumulate heavy metals when they are exposed

during development (Arruda-Neto et al., 2009; Arruda-Neto et al., 2010; Gdula-Argasinska, Appleton, Sawicka-Kapusta, & Spence, 2004; Zhang, Wang, Cheng, Xia, & Liang, 2011). Bone is a relatively suitable bio-indicator for long-term exposure, but human bone is usually not readily available for sampling and measurement (Kern & Mathiason, 2012; Kumagai et al., 2012). On the other hand, dental tissues are very hard and similar to the materials that make up the bone (Anjos et al., 2004; Arruda-Neto et al., 2009; Arruda-Neto et al., 2010; Webb et al., 2005). Unlike bone, in which the mineral phase is subject to turnover, the dental hard tissues (e.g., dentin and enamel) are not subject to significant turnover and therefore provide a permanent, cumulative and quite sound record of past and/or recent environmental exposure of heavy metals (Alomary et al., 2006; Appleton, Lee, Sawicka-Kapusta, Damk, & Cooke, 2000; Kolak et al., 2011; Oprea, Szalanski, Gustova, Oprea, & Buzguta, 2013; Prodana, Meghea, Stanciu, Hristu, & Demetrescu, 2010). Teeth (dentin, enamel or whole teeth) thus offer some advantages as suitable bio-indicators of heavy metal exposures (Abdullah et al., 2012; Alomary et al., 2006; Mohamed Amr & Helal, 2010; Arora et al., 2006; Arruda-Neto et al., 2010; Barton, 2011; Gdula-Argasinska et al., 2004; Kamberi et al., 2012; Kantamneni, 2010; Kolak et al., 2011; Kumagai et al., 2012; Zhang et al., 2011).

Teeth are readily accessible biopsy tissues and are physically stable for analysis. These biopsy tissues recently receive substantial attention for research in biological modeling, because of their easy extraction and very low rate of pollutant clearance relative to other organs (Kumagai et al., 2012). Consequently, a precise chronological record of exposure to a number of elements is retained in the hard calcified tissues of the teeth (Gdula-Argasinska et al., 2004). Furthermore, teeth of different ages of people can be easily accessed to compare the metal concentrations of multiple generations at one time (Kern & Mathiason, 2012).

Many studies have been devoted to analyze metal concentrations in whole teeth to make correlations between samples and environmental pollution by heavy metals (Adams, Romdalvik, Ramanujam, & Legator, 2007; Alomary et al., 2006; Amr, 2011; Mohamed Amr & Helal, 2010; Appleton, Lee, Sawicka-Kapusta, Damk, & Cooke, 2000; Arruda-Neto et al., 2009; Arruda-Neto et al., 2010; Báez, Belmont, García, & Hernández, 2004; Barton, 2011; Brown et al., 2004; Castro et al., 2010; Chew, Bradley, Amin, & Jamil, 2000; Karahalil, Aykanat, & Ertas, 2007; Kern & Mathiason, 2012; Tvinnereim, Eide, & Riise, 2000; Zhang et al., 2011). But data on the heavy metal concentration in tooth dentin is scarce. To increase knowledge of the spatial distribution of elements in each tissue of human teeth, (such as dentin, enamel, pulp and cementum) and their affinity for environmental pollution, it is important to study the elemental concentrations in the dentin and enamel separately.

During the sixth week in utero, dental hard tissues, specifically enamel and dentin, begin to grow and then teeth in each mandible become the deciduous teeth that are later replaced by the permanent teeth (Kohn, Morris, & Olin, 2013; Webb et al., 2005). Dentin, richer in organic content than enamel is biologically more active than enamel. It is a typical composite material, containing inorganic hydroxyapatite crystals and organic collagen matrix proteins (Arnold & Gaengler, 2007; Webb et al., 2005). Odontoblasts, situated in the pulp adjacent to the dentin, continuously produce dentin throughout the whole lifespan of a tooth, until it's shed (Arnold & Gaengler, 2007). Protoplasmic protuberances of odontoblasts persist after completion of dentin development and some metabolism is mediated through these growths. Dentin is not affected by the oral environment, since it is surrounded by enamel and cementum (Kumagai et al., 2012). There is no active metabolism of elements occurring after the completion of dental dentin (Kumagai et al., 2012). Tooth dentin can be a superior bio-

indicator of recent health and mineral status; and of environmental pollution by heavy metal since it is deposited during the course of life (Brown et al., 2004).

1.2   Motivation of this study

Most of the human foodstuffs primarily come either from land (such as land produced agricultural crops) or from aquatic system (as for example, marine and fresh water fishes, crustacean, molluscs and so forth). In the terrestrial ecosystem, soil is the main reservoir for radioactivity contamination and the ^{238}U, ^{232}Th, their progenies and also ^{40}K are the most common radionuclides in all land produced and aquatic food-stuffs. Soil-to-plant-to-human body is the main pathway of internal radiation dose experienced by individuals (Asaduzzaman et al., 2015; Tsukada et al., 2002). Diet is the leading root of human exposure to radioactive elements which guides to internal radiation doses (Chen, Zhua, & Hu, 2005; Gaso, Segovia, Cervantes, Herrera, & Perez-Silva, 2000; M.A Saeed et al., 2012). It is estimated that natural decay chain radionuclides (^{238}U, ^{232}Th, ^{235}U) represent the most significant source of ionizing radiation to earth contributing about 83% of the total effective dose experienced by individuals whereas 16% is contributed by ^{40}K, and the remaining 1% is due to the anthropogenic radionuclides (UNSCEAR, 2008a). Since a large fraction of at least one-eighth of the average annual effective dose has been credited to human body via the consumption of food, assessment of radiation level in foodstuffs finds great significance in addressing the associated health concerns (Jibiri, Farai, & Alausa, 2007; UNSCEAR, 2000). Consequently, it is important to study the spatial distribution of natural radioactivity and associated radiation exposures resulting from specific agrarian and marine food-stuffs.

Among the foodstuffs, rice, fishes and vegetables (root, fruit and leafy) are the main staple food that are widely produced and consumed as the daily diet in humid tropical

and sub-tropical regions, Malaysia as an exemplar. In order to determine the radiological food safety and minimize the radiation health risk of population, different kinds of foods (which constitute the major food nutritive requirement and large percentage of total diet) that are produced and commonly consumed by the Malaysian communities were collected directly from the farm land (rice and vegetable) and marine fish landing jetties (fishes, Crustesian and molluscs) in several sites of Peninsular Malaysia (Malaysian map showing the sampling location can be seen in the appendix E) in natural background conditions. However, one of the former Tin (Sn) mining area is also included in the present study for the investigation of radioactivity levels and their transfers to root vegetables. In the course of the milling of the Tin-ore to extract Tin, other minerals such as monazite, zircon, columbite, ilmenite etc. are also produced and extensively disseminated into the surrounding environment whereby they can be localized, consequently levels of natural radioactivity is intensified significantly (Jibiri et al., 2007). The waste resulting from the mining activities can modify the level of natural radioactivity in the soil, and hence the levels in crop grown on such soil may also boost via root uptake mechanisms from soil-plant pathways (Jibiri, Alausa, & Farai, 2009).

Seafood (e.g., fishes, crustaceans and molluscs) and their products can typically be one of the major sources of protein to populations in coastline regions all over the world as well as in Malaysia. It is noted that Malaysia is considered as one of the highest marine fish consumption country in the world, to extent that information on radionuclide balance in marine fish assumes greater importance (Alam & Mohamed, 2011; Mayeen Uddin Khandaker et al., 2013). Straits of Malacca is the main fishing zone of Malaysia which is one of the most important shipping lanes in the world, transporting around one-third of the world's targeted goods (Amin et al., 2013; Freeman, 2004; Khandaker et al., 2015a). About three million barrels of crude oil are

daily shipped via the Malacca Straits, subjecting the sensitive marine environment to the risk of accidental oil spillage; over the 10 years period 1981−1999, an average of two−three oil spill occurrences per year were recorded in these waters (Freeman, 2004). There is distinct possibility of enhanced levels of radio-contaminants from various sources including association of transportation, with increasing tanker traffic adding to the concern, offshore oil-gas exploration and exploitation and also the waste stream of urbanization may pose a substantial radiological threat to marine animals and ultimately humans following their consumption (Korkmaz Gorur et al., 2012).

The above circumstances indicate that the radioactivity monitoring in agrarian and marine foodstuffs are essential to control and avoid unwanted radiation exposure to the general public. This is to assertion that food safety is not compromised and the committed doses caused by the consumption of foodstuffs remain within the safety limits recommended by national and various international authorities such as UNSCEAR, IAEA, ICRP, EC, NCRP, WHO etc. After the Chernobyl nuclear plant accident on April 1986 and recent Fukushima Daiichi, Japan nuclear power plant accident on 11 March 2011, Nuclear Malaysia in collaboration with Ministry of Health had performed radiological food monitoring program to verify the levels of radioactivity of artificial radionuclides ^{134}Cs, ^{137}Cs and ^{131}I in imported foods through all of the points in Malaysia to ensure the public safety (Jaffary et al., 2011). This food monitoring program reported that imported food products do not show any traces of the said artificial radionuclides. Locally produced food monitoring measures for natural radioactivity are yet to be implemented in Malaysia (Yusof, Wo, & Ishak, 2011). The safety of food is one of the vital matters which always receive the civic attention. For the enhancement and facilitate human economic and social development, safe and nutritious food supply is of prime importance.

As far as our concern, no comprehensive data of natural radioactivity in soil, their transfers to foodstuffs, and radiological hazard parameters upon the consumption of staple foodstuffs are exist in the studied region. However, some efforts have been discretely made to study the radioactivity in marine animals in the Straits of Malacca. But, information on bioaccumulation and distribution of natural radioactivity in marine life within the available literature is still lacking. This data scarcity motivate us to investigate the levels of natural radioactivity (^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K) in the staple foodstuffs such as rice, vegetables and marine fishes which constitute the major dietary fraction of Malaysian people. The obtained data were used for a preliminary estimate of intake doses of radionuclides for Malaysian populace, and the associated health risks. The present study would therefore make a valuable contribution to the establishment of a standard database of the natural radioactivity in foodstuff sources in the country with a view to launch a radionuclide monitoring program through food chain. It can also be serve as a reference for the assessment of radiological impact through food consumption by the inhabitants of Malaysia and bordering regions. The outcomes of this work in turn can be treated as benchmark data to evaluate transfer factor into the food chain. The data generated herein can be used as baseline by the legislative bodies to setup and/or periodic evaluation and amendment of the existing guideline for the radiological protection of the population.

Similar to natural radioactivity, a further concern is the heavy metal pollutants through the ingestion of foods leading to potential health risks. Vegetables and marine fishes are the common foods in human diet in the tropic including Malaysia. Food constitutes the major source of ingestion of heavy metals in humans (Matos-Reyes, Cervera, Campos, & de la Guardia, 2010; Millour et al., 2012). A small amount of toxic/heavy metals are naturally originated in the terrestrial and aquatic environment, and trace amounts of them are always present in foodstuffs, but their levels can be high

in foodstuffs grown in contaminated soil/areas and/or polluted aquatic environment. Human food contaminated with toxic metal becomes a matter of worldwide concern because of their non-biodegradability and long term persistence; contaminated food is capable of transfer heavy metal into human body by the way of ingestion and potentially cause serious health risks, for instance stomach ache, renal dysfunction, pulmonary emphysema, cardiovascular and neurological destruction, bone disease, mutagenic effect, central nervous system, kidneys, lungs, liver etc, impairment (Asaduzzaman et al., 2015; Keshavarzi et al., 2015).

Heavy metals are natural trace elements in the aquatic media, however their concentrations have augmented owing to various industrial, agricultural and mining activities. As such, heavy metal analysis in human foodstuffs receives great attention in addressing the concomitant health concerns. Human foodstuffs can be contaminated by heavy metals via the introduction of mechanized farming, cumulative use of chemicals, such as various fertilizer, pesticides, herbicides, preservatives, processing and packaging of food (Matos-Reyes et al., 2010). Thus, the knowledge of heavy metal concentrations in both the terrestrial and marine foodstuffs can provide necessary information on the impact of the use of chemical products in crops and on levels of environmental contamination in agricultural lands. Data of heavy metal via food composition are essential to both consumers and health professionals and for food labelling legislation. Dietary ingestions of heavy metals that are of great global concern for the health of general public, need to be checked on a regular basis and current data on dietary intakes of metals should be updated frequently and publicly.

Food safety in light of heavy metal is one of the important matters that always find the public concern. Supply of safe and nutritious foods is the human rights for the improvement and expedite socio-economic development. As far as author's interests are

concerned, no representative data exist for the toxic/heavy metals in vegetables grown in the studied region. However, some effort has been discretely made to study some heavy metals in marine animals in the Straits of Malacca. Information on bioaccumulation and distribution of heavy metals in marine organisms within the available literature is scarce. This data scarcity motivate us to investigate the heavy metal pollutants (e.g., Pb, Cd, Hg, As, Cr, Mn, Al, Sb, Ba, Sr, Sn, Zn, Cu, Bi, Fe and Mg,) in the staples foods such as vegetables and marine fishes which constitute a major dietary fraction of Malaysian population. This study can make a significant input to create a standard database of the said heavy metals in foodstuff sources in the country with a view to promote a heavy metals monitoring program through food chain. It can also be serve as a reference for the evaluation of heavy metal threat through food consumption by the inhabitants of Malaysia. The results of this effort can be treated as benchmark data to assess the daily intake of metals for food consumption. The data generated herein can be used as baseline by the legislative bodies to setup and/or periodic evaluation and amendment of the existing guideline for the toxicological protection of the population.

In addition, bio-monitoring of heavy metal exposures to human is a sign of an individual's body burden, which is a function of recent and/or past record of exposure from environment (Kantamneni, 2010). Thus, the right choice and development of suitable biomarkers of heavy metals (e.g., Pb, Cd, Hg etc.) exposure is of crucial importance for the action of primary prevention, health care management purpose and decision making in public health sector (Arora et al., 2006; Barbosa et al., 2005).

Recently, there has been a growing curiosity among the researchers on the use of human bio-indicators such as blood, urine, bone, teeth, nails from finger and toes, hair and saliva to investigate the environmental pollution by means of toxic heavy metals

(Abdullah et al., 2012; Arruda-Neto et al., 2010; Kantamneni, 2010). However, due to short half-life (blood), impure sample (hair, saliva and fingernails) and not readily available for sampling (bone), these samples are not ideal as bio-indicators of chronological exposure and body burden of heavy metal (Arruda-Neto et al., 2010; Jones, 2014; Kern & Mathiason, 2012; Kumagai et al., 2012). For this reason, there exists a demand to develop alternative biological indicators to evaluate the level of exposure and better understand the health effects of environmental toxic metal exposure.

Literature revealed that teeth are superior to blood, nail, saliva, or hair as an indicator of chronological metal exposure from environment because the losses from teeth are much slower as there is no significant turnover of apatite in teeth, as in other biological indicators, therefore, teeth are to be the greatest convenient biological markers of exposure to environmental pollution (M. M. Abdullah et al., 2012; Alomary et al., 2006; Kumagai et al., 2012).

Many studies have been devoted to analyze the metal concentrations in whole teeth to make correlation between samples and environmental pollution by heavy metals. Individually, the data on the heavy metals concentration of tooth dentin and enamel is scarce. To increase the existing knowledge of spatial distribution of elements in each tissue of human teeth (such as dentin, enamel, pulp and cementum) and their influences by environmental pollution, it is important to study the elemental concentrations separately. More depth information may be gained on teeth as bio-marker of environmental pollution by investigating the dentin and enamel compartment independently.

1.3 Objectives of this study

This research work has been carried out to meetup the following objectives:

1. To investigate the levels of radiologically important natural radionuclides ((^{226}Ra (^{238}U), ^{232}Th (^{228}Ra) and ^{40}K)) in the staple foodstuffs produce in natural terrestrial (such as rice and vegetables) and marine (e.g., fishes) ecosystems, and estimate the levels of human exposures (e.g., daily and/or annual intake of radionuclides, annual effective dose etc.) from the consumption of these foods and lifetime cancer risk.

2. To evaluate the soil-to-plant (vegetation) transfer factor (migration or uptake) of these radionuclides.

3. To evaluate the heavy metal pollutants scenario in staple foodstuffs.

4. To investigate the heavy metal levels in human tooth dentin. Goals are to understand environmental pollution by heavy metals, to evaluate the correlation with a number of parameters; including the ethnicity of the tooth donor, age, sex, tooth condition and tooth type.

1.4 Account of research progress

After making a general introduction in Chapter 1, the rest of this thesis is designed in the following ways: Chapter 2 reviews and discusses the existing literature on the levels of NORMs and heavy metals in foodstuffs, and heavy metal pollutants in human teeth in relation to environmental pollution; Chapter 3 compiles the publications and the findings of this study. The objectives of this thesis and the findings of each published paper is linked and briefly described as follows:

Publication 1 is based on the study of natural radioactivity (^{226}Ra, ^{232}Th and ^{40}K) levels in rice which constitute the essential and main part of the daily diet of Malaysian populace. The rice and their associated soil samples were collected from the major rice

growing areas of Malaysia. Soil-to-rice transfer factor of radionuclides, and levels of human exposure such as daily intake of radionuclides, annual effective dose and lifetime cancer risk from the consumption of the rice has been evaluated.

Publication 2 is the comprehensive study of natural radioactivity (^{226}Ra, ^{232}Th and ^{40}K) levels in vegetables which also form the essential and main part of the Malaysian daily diet. The vegetables and their associated soil samples were collected from the various vegetable growing regions of Malaysia. Soil-to-vegetable transfer factor of radionuclides, and various radiological hazard indicators such as daily intake of radionuclides, committed effective dose and lifetime cancer risk due to the consumption of different types of vegetables have been assessed.

Publication 3 is based on the study of natural radioactivity (^{226}Ra, ^{232}Th and ^{40}K) levels in root vegetables which supply the food starch that the important part of the Malaysian daily diet. The root vegetables (tapioca and sweet potato) and their associated soil samples were collected from the various tapioca and sweet potato growing areas including a former tin (Sn) mining area of Malaysia. Soil-to-tapioca and soil-to-sweet potato transfer factor of radionuclides, daily intake of radionuclides, annual committed effective dose due to the consumption of these root vegetables have been appraised.

Publication 4 is the comprehensive study of natural radioactivity (^{226}Ra, ^{228}Ra and ^{40}K) and heavy metal (Pb, Hg, Cd, As, Cr, Ni, Mn, Cu, Rb, Sb, Sr, Al, Fe, Ba, Bi, Zn and Mg) levels in vegetables (practically all categories of vegetable such as root, fruit and leafy vegetables) that are produced and commonly consumed by the Kuala Selangor communities of Malaysia. Various radiological hazard indicators such as annual intake of radionuclides, committed effective dose and lifetime cancer risk due to the

consumption of different types of vegetables have been evaluated. The concentration and daily intake of heavy metals in these vegetables have also been assessed.

Publication 5 is based on the study of natural radioactivity (^{226}Ra, ^{232}Th and ^{40}K) and heavy metal (Pb, Hg, Cr, Cd, As, Mn, Cu, Rb, Se, Sr, Al, Co, Fe, Ba, Bi, Zn and Mg) levels in marine fish which also constitute the essential and main part of the Malaysian daily diet. The marine Fish (*Rastrelliger kanagurta*) were collected from different fish landing jetties of Straits of Malacca, Malaysia. Various radiological hazard indicators such as daily intake of radionuclides, committed effective dose and lifetime cancer risk due to the consumption of this popular and hugely caught fish species have been assessed. The concentration and daily intake of heavy metals in this marine fish species have also been evaluated.

Publication 6 is based on the study of natural radioactivity (^{226}Ra, ^{228}Ra and ^{40}K) levels in marine animals such as fishes, crustaceans and molluscs which also constitute the essential and main part of the Malaysian daily diet. The marine animal samples were collected from different fish landing jetties of East and West coast of Peninsular Malaysia. Radiological hazard indicators such as daily intake of radionuclides and committed effective dose to the consumption of these marine animal species have been evaluated.

Publication 7 deals with the study of heavy metal levels in human teeth used as a bio-indicator of metal exposure to environmental pollution. The teeth samples were collected from heavily polluted Klang valley, Malaysia to evaluate the presence and levels of heavy metal in human teeth in relation to environmental pollution, and to evaluate the correlation with a number of parameters including the ethnicity of the tooth donor, age, sex, tooth condition and tooth type.

Finally Chapter 4 ends this research with a brief conclusions from the present study and suggestions for future work.

CHAPTER 2: LITERATURE REVIEW

2.1   Introduction

The discovery of X-ray by German Mechanical Engineer and Physicist Wilhelm Conrad Roentgen in 1895 and after the discovery of radioactivity in 1896 by French Physicist Antoine Henri Becquerel, the science of the radioactivity has been widely studied and established the basic phenomenon of radioactivity (Allisy, 1996). Radionuclides that emit nuclear radiations are the sources of radioactivity. In the following decades, intensive investigations carried out by Marie and Pierre Curie, Rutherford and many other Scientists discovered that these emissions are ionizing radiation, comprises positively and negatively charged and neutral particles, first categorized by Ernest Rutherford (according to their penetrating and ionizing competences) as alpha particles, beta particles and gamma rays (Lilley, 2001, Rutherford, 1911).

While by definition natural levels of radioactivity and heavy metals are ubiquitous in nature, their presence in the environment can be enhanced through anthropomorphic activity, including agricultural inputs (e.g. use of fertilizers, fungicides, insecticides and herbicides), rapid urbanization, industrial activity such as coal-, oil- and gas-exploration and exploitation and radioactive wastes and industrial effluents disposal (Awudu et al., 2012; Santos et al., 2002; Korkmaz Gorur et al., 2012; Santos et al., 2004). Food is the major source of ingestion of radioactivity and heavy metals in humans. The presence of radioactivity and heavy metals in vegetation can be enhanced by such activities, posing an additional loading with consequences for human health.

This chapter deals with the reviews of the early literature on the subject of radioactivity (e.g., basic concept of radioactivity and radioactive decays together with radioactive equilibrium, nuclear decay processes, gamma-rays interaction with matters

etc.). The biological effect of low dose radiation are briefly discussed. Environmental sources of radioactivity and its detection techniques are also focussed. Moreover, reviews on environmental sources of heavy metal and its impact on human health are highlighted. Finally, the recent literature on natural radioactivity in rice, vegetables and marine animals (e.g., fish, crustacean and molluscs) and heavy metal pollutants in vegetables and marine fishes are reviewed. Emphasis has also been placed on the literature review on heavy metal in human/animal teeth- a bio-monitor of environmental pollution by heavy metal.

2.2   Radioactivity and radioactive decay

Radioactivity is a statistical progress defining the spontaneous transformation of unstable atomic nuclei (parent nuclei) into a relatively more stable nuclei (formation of daughter nuclei) without any influence of chemical and physical state (Lilley, 2001; L'Annunziata, 2007; Choppin, Liljenzin, & Rydberg, 2002). If the progenies of parent nuclei are also unstable, the process of decay will continue until a progeny nucleus attain its stable state (Lilley, 2001). During the decay process, the energy of transformation may be released by the way of the emission of nuclear particles and/or in the form of electromagnetic radiations (Gilmore, 2008; L'Annunziata, 2007).

The intensity or strength of radioactivity is termed as activity and the activity of a radioactive substance is defined as the rate of nuclei number decaying (i.e., the number of disintegration per unit time) (Knoll, 2000; L'Annunziata, 2007). The radioactive disintegration rate is a first order process and depend on the radioactive atoms present in particular radionuclide in the source. The decay rare of radioactive material decreased exponentially with time. The rate of radioactive decay is related to activity and is directly proportional to the number of atoms of radionuclide present in the radioactive

source, which can be stated by the fundamental law of radioactive decay (Gilmore, 2008; Knoll, 2000):

$$A = -\frac{dN}{dt} = \lambda N \qquad (2.1)$$

where, A is the activity of radioactive source is equal to the number, dN of radioactive nuclei that disintegrate in a time, dt, which is proportional to the original number of radioactive nuclei, N present at time, t and λ is the decay or disintegration constant which defines the probability per unit time for the decay of a nucleus (Gilmore, 2008; Knoll, 2000).

The exponential law of radioactive decay can be found by integrating the differential equation (2.1); i.e. (Gilmore, 2008),

$$N_t = N_0 e^{-\lambda t} \qquad (2.2)$$

where N_0 represent the original number of nuclei present at time t = 0 and N_t is the number of nuclei present at later time t.

The activity, A_t is the decay rate which occurs in a given source and can be achieved by differentiating equation (2.2) and taking into account equation (2.1) (Gilmore, 2008):

$$A_t = A_0 e^{-\lambda t} \qquad (2.3)$$

where A_0 and A_t are the initial and present activities of a radioactive substance at time t = 0 and later time t, respectively.

The original (historical) unit of activity is curie (Ci), which is numerically equal to 3.7×10^{10} disintegration per second, which is based on the estimation of the activity of

one gram of pure ^{226}Ra (Cember & Johnson, 2009; Curie et al., 1931; Knoll, 2000). Later, the Becquerel (Bq) has become the standard unit of activity, which is defined as one disintegration per second (dps) (Knoll, 2000; Lilley, 2001). Therefore, 1 Ci = 3.7×10^{10} dps = 3.7×10^{10} Bq (Cember & Johnson, 2009; Gilmore, 2008; L'Annunziata, 2007).

Even though the activity of a given sample is used as a measure of the quantity of radioactivity present, it does not bear any information on the volume or mass of the sample/material in which the radioactive decay is occurred. In practice, it is more useful to use the term 'specific activity' which is defined as the activity per unit mass (W) of the radioactive sample, i.e.,

$$A_s = \frac{A}{W} = \lambda \frac{N}{W} \qquad (2.4)$$

It is measured in Bq/kg).

2.2.1 Types of radioactive decay

Radioactive nuclei are usually unstable and has a tendency to reach more stable state by discharging energetic particles. Although various modes of radioactive decay exists, principally, the most common types are the alpha (α), beta (β) and gamma (γ) decays whereby radioactive nuclei can transform to create other nuclei (Krane, 1988; Lilley, 2001; Tipler & Llewellyn, 2007). These nuclear transformation processes are usually statistical in nature. These emitted particles can undesirably affect the cells of the body thereby causing in death or unrestrained multiplication of cells in the human body.

2.2.1.1 Alpha (α) decays

Decay by the alpha particles occurs naturally in heavy nuclei in the radioactive decay chains, which involves the emission of an electron that is positively charged and

transform the mass number of the nucleus while decaying (Gilmore, 2008; Krane, 1988; L'Annunziata, 2007). Alpha-particles are in nucleus of helium-4 isotope composed of two proton and two neutrons and has a very low penetrating power, which cannot penetrate the dead layers of human skin (Gilmore, 2008; Cember & Johnson, 2009; Knoll, 2000). Because of their large mass and electric charge, this particles cannot travel very far away in the environment. The heavy unstable nucleus, both the naturally occurring radionuclides of which atomic number is greater than 82 and anthropogenic produced nuclides of which atomic number is bigger than 92 emit alpha-particle as a decay product (Lilley, 2001). The emission of alpha-particle from the initial nuclei leads to the decrease in both the mass and charge on the final nuclei can be represented in the following decay process.

$$_Z^A X_N \rightarrow _{Z-2}^{A-4} Y_{N-2} + \alpha \qquad (2.5)$$

where A and Z represent the mass and atomic number of the nuclear species, respectively, X is the parent decaying nucleus and Y is the final recoil (final) nucleus (Das & Ferbel, 2003; Knoll, 2000). Alpha radioactivity is usually a stream of positively charged particles with very high ionization and low penetration range. Generally, the energy ranges of α-particles emitted from the radioactive decay are between 1 and 10.5 MeV (L'Annunziata, 2007).

2.2.1.2 Beta (β) decays

Beta (β) particle is an electronically charged particle originates from the neutron-rich or proton-rich unstable nuclei (Lilley, 2001). There are three decay processes by which a nuclei may undergo radioactive beta (β) decay: positron emission, referred as beta plus decay (β+ decay); electron emission, termed as beta minus decay (β− decay) and electron capture (Harvey, 1969; Lilley, 2001). In this decay process, the atomic number

and the neutron number of a nucleus change by one unit, however the atomic mass number remains the same (Krane, 1988).

Negative beta decay (β− decay) occur when the daughter nucleus is energetically more stable than the parent nucleus, where a neutron directly transforms to a proton, electron and anti-neutrino (Lilley, 2001). The proton that formed in this process remains in the nucleus and the electron is ejected as a β− particle. The following expression represents an example of β− decay process (Krane, 1988):

$$^{210}_{82}Pb_{128} \rightarrow {}^{210}_{83}Bi_{127} + e^- \ (\beta-) + v \qquad (2.6)$$
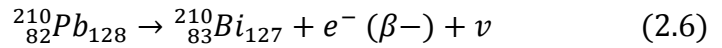
While, the positive beta (β+) decay occurs if the ratio of protons to neutrons of a radioactive nucleus is greater than the most stable isobar in the particular isobaric chain (Cember & Johnson, 2009; L'Annunziata, 2007). In process of β+, the weak nuclear force converts a proton into a neutron with the release of β+ particle and a neutrino. This kind of emission occurs only if the transformation energy is bigger than 1.02 MeV. The following expression represents an example of β+ decay process (Krane, 1988):

$$^{25}_{13}Al_{12} \rightarrow {}^{25}_{12}Mg_{13} + e^+ \ (\beta+) + v \qquad (2.7)$$
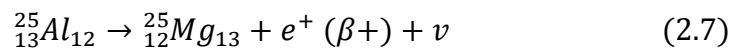
Conversely, electron capture (EC) is an alternative process of positron decay (β+), which occurs when the energy of transformation is insufficient to form an electron pair (Cember & Johnson, 2009; L'Annunziata, 2007). In this process, an atomic electron whose orbits is close to the nucleus is captured by a proton-rich unstable nucleus (Das & Ferbel, 2003; Gilmore, 2008; Lilley, 2001). The electron combines with a proton and produce a neutron with the emission of a neutrino of fixed energy. In the process of electron capture (EC), the mass of an electron is transformed into energy. An example of EC process is as follows:

$$^{208}_{83}Bi_{12}5 \rightarrow ^{208}_{82}Pb_{126} + v \qquad\qquad (2.8)$$

β-particle is much lighter than α-particle which means that for a given energy, β-particle is more penetrating than α-particle. Beta radiation is usually composed of a stream of electrons with a continuous energy sweep associated with the β-particles (L'Annunziata, 2007; Lilley, 2001). The level of highest energy of β-particle is a function of particular radionuclide.

2.2.1.3   Gamma (γ) decays

Gamma radiation is a form of electromagnetic radiation which has no mass and electric charge and originate from the nucleus of a radioactive nuclei (Gilmore, 2008; L'Annunziata, 2007). Unlike alpha and beta particles, gamma radiation is a process by which some radionuclides release their energy in the form of electromagnetic radiation and which travels at the speed of light. The origin of gamma radiations can be correlated with decays from nuclear excited states that may have been populated to subsequent alpha and beta decay processes (Krane, 1988). The excited nuclei may release some of their energy through a transition to lesser and more stable energy state by way of gamma radiation (Krane, 1988). As for example, ^{222}Ra is produced when the natural radionuclide ^{226}Ra undergoes alpha decay, which is accompanied by a gamma decay with an energy of 186.21 keV. Characteristically, the energies of gamma rays cover a range from 0.1 to 10 MeV (Krane, 1988; Lilley, 2001).

2.3   Radioactive equilibrium

The term radioactive equilibrium is generally used to elucidate the state when the members of the radioactive series decay at the same rate as they are produced (Prince, 1979). There are three predominant limiting situations concerning the term equilibrium,

these are, secular equilibrium, transient equilibrium and the state of no equilibrium (L'Annunziata, 2007).

2.3.1 Secular equilibrium

Secular equilibrium is the most common case of radioactive equilibrium, is a steady-state condition in which the half-life of the parent nuclide is infinitely longer than that of its progeny nuclide (Cember & Johnson, 2009; Krane, 1988). Under secular equilibrium condition, the parent nuclide undergoes a very slow rate of decay with no noticeable change its activity for the period of several half-lives of its decay products, whereas its progenies grow-in and continue to decay. Once the secular equilibrium is established, the activity of the progenies becomes equal to that of its parent with time (Cember & Johnson, 2009; L'Annunziata, 2007). The progeny nuclide can reach their parent activity in a closed system if the system elapsed for a sufficiently longer period than the daughter half-life. For example, assuming secular equilibrium, the activity concentrations of the several daughter radionuclides that attend the parent can be estimated for natural radionuclide of ^{238}U with its first 6 daughters to ^{226}Ra (Denagbe, 2000). Generally, secular equilibrium is put in practice in the indirect measurement of activity of parent radionuclide via its daughter using gamma-ray spectrometry in which the activity of any of the daughters is a representative activity of the parent. To achieve secular equilibrium of ^{238}U and ^{232}Th series with their daughters and to measure the radioactivity of unknown samples, the samples must be sealed in a container for a length of period to prevent their escape prior to gamma-ray spectrometric analysis.

2.3.2 Transient equilibrium

Transient equilibrium occurs when the half-life of the parent nuclide is not significantly longer or nearly the same as the daughter (Cember & Johnson, 2009; Choppin et al., 2002; L'Annunziata, 2007). When the transient equilibrium exists, the

ratio of the number of radionuclide tends to constant value and the rate of decay of the parent and progeny radionuclides will the same which is the main characteristic of the transient equilibrium (Cember & Johnson, 2009; Harvey, 1969; L'Annunziata, 2007). The decay of ^{212}Pb with half-life 10.64 hours to ^{212}Bi with half-life 60.55 minutes is an example of transient equilibrium. The establishment of transient equilibrium between the parent and its daughter radionuclides depends on the degrees of their half-lives, i.e., the smaller the half-life of the daughter compared to the parent, the faster the state of transient equilibrium will be attained (Cember & Johnson, 2009).

2.3.3   No equilibrium

In the situation where the half-life of the daughter products are much longer than that of its parent nuclide, the parent decays relatively quickly leaving behind the daughter and hence the state of equilibrium will not be reached (Cember & Johnson, 2009; Gilmore, 2008; Krane, 1988). In these conditions, the product activity growths to a maximum and decays with its own characteristic half-life and equilibrium does not exist (Cember & Johnson, 2009; Choppin et al., 2002). The decay of ^{218}Po with half-life 3.1 minutes to ^{214}Pb with half-life 26.8 minutes is an example of the state of no equilibrium.

2.3.4   Radioactive disequilibrium

Radioactive disequilibrium occur if the daughter radionuclides in a decay chain partially or totally separated from the series system (IAEA, 2003). Disequilibrium occur mainly due to some geological processes, such as erosion and leaching which are common in uranium (U) decay chains and takes place at different points within the decay chains/series (because U-decay chains are composed of different elements) (Condomines, Jean-Claude, & Vale´rie, 1995). Radioactive disequilibrium in the U-decay series affects the state of the activity concentration and therefore appear as a source of error in the gamma-ray spectrum.

2.4   Interaction of radiation (basically gamma-rays) with matter

Principally, radiation is a stream of elementary particles and quanta of energy. Their energy of radiation and physical characteristics governs their categorization and interaction with matter (Gilmore, 2008). When a beam of radiation passes through the tissue or other absorbing medium, the beam can be transmitted to or scattered from the media; whereas in some situations, the photon energy is imparted to the medium that either ionizes or excites the medium. The instrumental detection of any radiation or particle depends on the production of the secondary charged particles which can be collected to produce an electrical signal. The charged particles alpha and beta can produce a signal within the detector by ionization and excitation of the detector material directly, while the gamma photons are uncharged and thus cannot do this (Gilmore, 2008; Knoll, 2000). Whereas, the detection of gamma-ray depends on other types of interaction which transform the gamma ray energy to electrons within the detector material (Gilmore, 2008). The excited electrons have charge and lose their energy by excitation and ionization of atoms of the detector media, giving rise to several electron-hole pairs and produce electrical signal.

Although the interaction of gamma-ray photons in the matter can occur by several distinct mechanisms, only three mechanisms play the most important roles in the radiation measurement (Debertin & Helmer, 1988; Knoll, 2000). These mechanisms are: (i) photoelectric interactions or effect or absorption which occur at low photon energies, (ii) compton scattering which is leading in mid-energy range and (iii) pair production, visible at high energy, all contribute to the experimental response in gamma-ray spectrometry (Gilmore, 2008; Knoll, 2000). In each of these cases, gamma-rays transfer their energy either partially or completely to the electrons of the detection medium (Gilmore, 2008).

2.4.1 Photoelectric effect

Photoelectric absorption occurs by the interaction of gamma-ray photon with a bound electron in an atom of the absorber material in which the photon is completely absorbed and an energetic electron termed as photoelectron is ejected from its shell with the kinetic energy, E_c, expressed as (Gilmore, 2008; Knoll, 2000):

$$E_c = hv - E_b \qquad (2.9)$$

Photoelectric interaction is only possible if the incoming photon energy (hv) of the gamma-ray exceeds the ionization (binding) energy (E_b) of the ejected electron. The binding energy of these electron varied from a few keV (for material with lower atomic number) to tens of keV for the materials which have higher atomic number (Knoll, 2000). This interaction also creates an ionized absorber atom with the vacancy in the bound electron shells causing in an excited state. Then the excited free electron can be captured by the medium or a de-excitation occur by the rearrangement of electrons from the outer shells to fill in a vacancy leading to the production of characteristic X-ray photons. Photoelectric process is dominant at low energy state, however less significant at greater energies.

2.4.2 Compton scattering

In the Compton scattering mechanisms, the incident gamma-ray photon directly collides with weakly bound electron in the absorbing medium and a portion of its energy is transferred to the recoil electron (Knoll, 2000). The gamma-ray photon is then deflected or scattered through an angle θ relative to its original direction.

The energy, hv′ of the scattered gamma ray photon is a function of its scattering angle θ and can be expressed as (Das & Ferbel, 2003; L'Annunziata, 2007):

$$h\acute{v} = \frac{hv}{1 + \left(\frac{hv}{m_0 c^2}\right)(1 - cos\theta)} \tag{2.10}$$

where $m_o c^2$ is the rest mass energy of the ejected electron is equal to 511 keV.

The probability of Compton scattering strongly depends on the number of electron per nit mass of the interacting medium and thus increases linearly with atomic number of the medium. This scattering is the leading interaction process for gamma ray energies ranging from 0.1 MeV to 10 MeV. At greater energies, another interaction mechanism called pair production is more significant.

2.4.3   Pair production

Pair production process occurs close to the nuclei of the absorbing medium, because this point possesses high electric field. The pair production process is energetically possible if the incident gamma-ray energy exceeds twice the rest mass energy of an electron (i.e., $2m_o c^2 \geq 1.022$ MeV) (Knoll, 2000). This type of interaction occurs mainly within the nuclear Coulomb field of a nucleus in which the high energy gamma ray photon is absorbed into the vacuum and is transformed into an electron-positron pair.

Since a gamma photon energy of 2 $m_o c^2$ (1.022 MeV) is essential for the formation of electron-positron pair, thus any excess energy (beyond 1.022 MeV) carried by the gamma ray photon is imparted to and shared by the electron-positron pair as kinetic energy, which can be expressed by (Turner, 2007)

$$E_{e^-} + E_{e^+} = hv - 2m_0 c^2 \tag{2.11}$$

The probability of pair production process depends on the gamma ray energy beyond threshold (1.o22 MeV) and becomes the significant interaction process for gamma energies greater than 10 MeV (Das & Ferbel, 2003).

2.5   Biological effects of radiation

When radionuclides are entered into the human body by any means (ingestion and inhalation), some of them may be cleared by the body relatively quickly, even their physical half-life has elapsed and rest of them remain in the body. The alpha and beta emitters radionuclide transfer their huge amounts of energy to the body organs/tissues for the rest of individual's span of life, causing potentially direct damage to the DNA in the human cell (ICRP, 2002). If the human body is exposed by any types of radiation, either from internal (ingestion or inhalation) or external (gamma radiation) or both sources, ionization and excitation of atoms and molecules in the living cell can be occurred. Therefore, the interaction of radiation with biological tissues can cause in the damage and death of the living cells and/or mutation of the genetic material (1997; Lilley, 2001; Noz & Maguire, 2007; UNSCEAR, 2000). The amount of ionization and the energy absorbed by the specific cells related to biological efficacy which can be measured in terms of radiation dosimetry.

The biological effect of ionizing radiation begin if the living cell molecules interact with radiation energy via exposure and/or deposition. When a massive radiation dose (>20 Gy) is delivered in shorter period of time, the symptoms of acute radiation damage will be predicted in the first few hours and/or days. Conversely, when the delivered dose is considerably smaller and recurrent over lengthier periods of time prolonging to several years, the biological effect of this low dose exposure may not manifest for several decades (Eisenbud & Gesell, 1997).

Biological effects from exposure to ionizing radiation on living cells can take place either by direct or indirect actions. Direct actions occur if the radiation causes excitation in the same molecule of the cell, in which the initial radiation is deposited and engrossed. Whereas, indirect actions/effects take place if the ionizing radiation is

absorbed in the water molecules of the human cell/body and generates short lived chemically reactive products that can react with other molecules in the body (Henriksen & Maillie, 2003). Thus, if the human body/organism is irradiated by ionizing radiation, the chemically reactive products can be formed either directly by ionization and produce hydrated electron (e⁻) or due to excitation of water molecules that quickly lose their energy by cleaving the bond and produce a hydroxyl radical (OH⁻) and a hydrogen atom (H⁺) that are often called free radicals which are highly toxic, being the cause behind several biological effects in human body (Cember & Johnson, 2009).

These chemically highly reactive products act as a radiation product within the body or body cells and can directly react with biomolecules, i.e., DNA (deoxyribonucleic acid) and causing critical damage to the chemical structure of DNA, consequently, lead to potential for development to cancer (Comatic mutation) or genetic mutation (hereditary disease) (Cember & Johnson, 2009; Henriksen & Maillie, 2003; UNSCEAR; 2000). In reality, when the damage of DNA is misrepaired and carry on to divides into new mutated cells, a cancerous polyp may developed as direct result of exposure to ionizing radiation.

2.6 Environmental sources of radioactivity/radioactivity in nature

The world is naturally radioactive since its creation due to naturally occurring radioactive materials (NORMs) in its environment, thus life on earth has settled under the ubiquitous presence of environmental radiation to which all living beings are continuously exposed (Ahmad, Jaafar, Bakhash, & Rahim, 2015; Eisenbud & Gesell, 1997). Natural radioactivity is widely distributed in the environment and can be found in various environmental media, for instance, soil, rock, plants and foods, air, water and building materials in different quantities (Abojassim, Al-Gazaly, & Kadhim, 2014; Kant, Gupta, Kumari, Gupta, & Garg, 2015). Several types of biochemical mechanisms

and actions of some entities transfer these radionuclides from their distinct geologic medium into the biosphere in which they get bio-accumulated in the chain of food, and serve as a major exposure route to human beings.

Human beings are continually exposed to the environmental radioactivity that can be categorized into two main sources: namely, natural and anthropogenic (man-made) sources. Natural sources of radioactivity mostly arise from the terrestrial radionuclides having long half-lives relative to earths and are extensively distributed in the earth's crust and extra-terrestrial sources arise from the bombardment of cosmic rays (Lilley, 2001; NCRP, 1975; UNSCEAR, 2000). Along with the natural sources, human activities concerned with the utilization of radiation and radioactive materials (e.g., industrial purposes and medical applications) from which radionuclides may release into the environment and enhance the radioactivity in nature (Eisenbud & Gesell, 1997; UNSCEAR, 2010).

2.6.1   Natural sources of radioactivity

According to their origin, natural radionuclides can be classified into two types, the terrestrial and cosmogenic. Since some of the terrestrial radionuclides have very long half-lives (hundred million years or more), large fraction of these radionuclides are still exists on earth today. These radionuclides are common in soil, rocks, food and water, ocean etc (UNSCEAR, 2010). Terrestrial radionuclides are again sub-divided into two categories, namely, primordial (non-series) and decay chains (series) radionuclides (Eisenbud & Gesell, 1997; UNSCEAR, 2010).

2.6.1.1 Primordial radionuclides

Several single (non-series) primordial radionuclides have been identified that occurs naturally and decays directly to stable nuclides. Most of them are radioactive isotopes with very long half-lives (10^{10}–10^{15} years) comparable to the age of the earth and

extensively low isotopic abundance, causing in their negligibly less activities and usually not regarded as important in terms of human radiation exposure (Lilley, 2001). However, the radioactive isotopes ^{40}K and ^{87}Rb of them are significant sources of natural radioactivity. Most of them decay by beta and alpha emission, whereas some of them follows electron capture (Eisenbud & Gesell, 1997; NCRP, 1975). The main primordial singly radionuclides are listed in appendix B. ^{40}K is widely distributed in the earth's crust and a dominant radionuclide in normal foodstuffs and human tissues. ^{40}K is a major source of internal radiation dose due its high energetic beta emission, but homeostatically controlled by the human body and thus makes the dose from ^{40}K to be constant within the human body (Eisenbud & Gesell, 1997; UNSCEAR, 2000).

2.6.1.2 Decay chains (series) radionuclides

Natural decay series radionuclides (^{238}U, ^{232}Th and ^{235}U) and their succeeding radioactive decay progenies and non-series radionuclides ^{40}K are the greatest encountered radionuclides in virtually all environmental media including soil, rocks and foodstuffs, exposure dose resulting predominantly from these radionuclides in drinking water and foodstuff that internally irradiate the various organs with alpha and beta particles (Awudu et al., 2011; Canbazoğlu & Doğru, 2013; IAEA, 2003; Rahman & Faheem, 2008; UNSCEAR, 2000)

The three decay series radionuclides (^{238}U, ^{232}Th and ^{235}U) which have a half-life compared to the age of the earth can still be present observed on earth environment that form the main portion of our natural radiation (Krane, 1988; Lilley, 2001; NCRP, 1975). These chains radionuclides decay through a sequence of radioactive progeny nuclides and finally attain a stable state of lead. The decay schemes of each radioactive series and details of each radionuclides of these series are presented in appendixes A and B, respectively.

2.6.2   Cosmic radiation/Cosmogenic radionuclides

Natural radioactivity in the form of cosmic radiation arises from both the primary highly energetic protons (positively charged particles) and high energy photons of extra-terrestrial origin which strike the atmosphere of earth and the secondary particles known as cosmogenic radionuclides are constantly produced by the bombardment of stable nuclides in the upper atmosphere (NCRP, 1975; Silberberg & Tsao, 1990; UNSCEAR, 2000). A huge number of radioactive isotopes with a half-life of several minutes to millions years that are produced by cosmic-ray are found everywhere in the living earth. However, only four of the main cosmogenic radionuclides ^{3}H, ^{7}Be, ^{14}C and ^{22}Na contribute significantly measurable dose of radioactivity in humans (Eisenbud & Gesell, 1997; NCRP, 1975; UNSCEAR, 2000). The list of cosmogenic radionuclides of natural origin can be seen in appendix B.

2.6.3   Anthropogenic (man-made) sources of radioactivity

In addition to natural radioactivity, technological progression and increased growth in the use of nuclear energy introduce the concept of anthropogenic radioactivity in the environment. Human activities that mentioned in section 2.1 and the waste of nuclear medicine can boost the levels of natural radioactivity in every component of the environment which is termed as TENORM (technologically enhanced naturally occurring radioactive material).

The radionuclides in the decay series are essentially in radiological equilibrium with their parents in the naturally undisturbed environments. But, the aforementioned human activities (mentioned in section 2.1) or natural catastrophe can break this equilibrium, resulting in either an enhancement or reduction of radionuclides concentrations compared to the original matrix (Faanu, 2011; UNSCEAR, 2000).

Radioactivity in soil as well as human foodstuffs can be boosted by the soil amendment, such as application of phosphate and other fertilizers, pesticides, insecticides, fungicides, irrigation by contaminated water, radioactive wastes and industrial effluents disposal etc (Amin et al., 2013; Awudu et al., 2012; UNSCEAR, 2008b).

Detectable amounts of artificial radionuclides (mainly ^{137}Cs) are widely spread in the atmoshphere, particularly those are pruduced by the testing of nuclear weapon since 1945, accidental failures of nuclear power stations in 1986 (Chernobyl, Ukrain) and in 2011 (Fukushima, Japan) and from nuclear power fuel cycle (Lilley, 2001; NCRP, 1977). This radionuclide has been globally disseminated and deposited via rain and dry deposition on the terrestrial surfaces, which can be uptake by plants and subsequently accumulated into foodstuffs (Watson, Jones, Oatway, & Hughes, 2005). Annual average dose of ionizing radiation to the world population from various sources can be seen in appendix B.

2.7 Radiation detection

There are several types of radiation detectors/instruments available for the measurement of ionizing radiation in the samples such as gas filled detectors (ionization chamber counters, proportional counters and Geiger-Muller counters), scintillation and semiconductor detectors (solid-state detector) (Cember & Johnson, 2009; Knoll, 2000; L'Annunziata, 2007). All kinds of radiation detection methods/instruments fundamentally involve the generation of electrical signals resulting from the interaction of radiation/gamma-ray photons with the detector medium (i.e., gas, liquid or solid materials). Each types of gamma ray interactions (photoelectric effect, Compton scattering and pair production) that discussed in section 2.4 can occur within the active volume of the detector, however, photoelectric effect and Compton scattering plays the

most significant roles in the detection of gamma ray photons (Cember & Johnson, 2009; Knoll, 2000).

The basic requirement of the radiation measuring equipment is that, the radiation/gamma-ray photon interacts with the detector in such a way that the degree of the equipment's response is proportional to the effect or property of radiation being measured (IAEA, 1989). Due to their interactions, the primary gamma ray photons / and or scattered secondary photons undergoes interaction with the detector atoms and produce fast electrons (by ionization) within the sensitive volume of the detector. As these fast electrons propagate through the volume of the detector, can able to generate secondary electrons which can be collected to produce electrical signals or output pulses. These pulses (charges) can be transformed by a charge sensitive preamplifier to a voltage in which the amplitude of the voltage pulse is proportional to the initial energy of the gamma ray that deposited/absorbed in the detector (Knoll, 2000).

The assessment of environmental radioactivity is achieved predominantly by the gamma-ray spectrometry technique. In this technique gamma ray detector must act as an energy conversion medium for the incident gamma-ray photons to have a high interaction probability for the production of fast electrons and also used as a conventional detector for the secondary electrons (Knoll, 2000). In this method, pulse amplitudes of the gamma ray photons are analysed after the amplification and digitization. Therefore, the output of the gamma spectrometer represents the energy spectrum of the detected radionuclide which provides complete information that is useful for the identification of the unknown radionuclides.

Gas-filled detection systems are certainly suitable only for the counting of low energy electrons, ions and photons because of their poor stopping capacity of gas as a detection medium for the gamma-rays. To improve the absorption capability of

detecting medium, greater atomic number and/or high density liquid or solid materials are commonly used for the measurement of extremely penetrating (i.e., gamma radiation) radiations (Gilmore, 2008; Krane, 1988).

High resolution gamma-ray spectrometry system is the most frequently used radioactivity measurement methods which give relatively low detection limits for radioactive nuclides. Basically, thallium doped sodium iodide (NaI (Tl) scintillation and high purity germanium (HPGe) are the two commonly used detectors in gamma ray spectrometry system, which is characterized by low impurity concentration, high atomic number and low ionizing energy required for the production of electron-hole pair. The detectors that are based on the mechanism of scintillation (NaI(Tl) is one of the oldest and most useful methods for the detection and measurement of a broad range of radiations, especially in-situ measurement system in order to their room temperature operation and simplicity (Knoll, 2000). The NaI crystal suffers numerous excitations by the incident gamma radiation, which is then followed by a series of de-excitations within the same crystal and produce a quanta of light. These light pulses which are captured/strike on the photosensitive cathode of the photomultiplier tube (optically coupled to the crystal) and ultimately released as electrical charge (electrons are ejected from the cathode). This charge is amplified by the dynode chain to produce detectable electrical signal which is finally processed. The scintillation detectors have a high efficiency for the detection of gamma radiation but the energy resolution is poor. Due to their poor energy resolution, this type of detectors do not provide the energy selectivity for dealing with a complex gamma ray spectrum (Cember & Johnson, 2009; Knoll, 2000). To attain a good energy resolution, a semiconductor detectors (such as HPGe) are employed.

Detectors which is made from semiconductor materials offer alternatives to scintillation detectors that result in much number of charge carriers than any other kinds of radiation detector. Unlike NaI(Tl) detectors, HPGe detectors directly convert the gamma photon into charge carriers. For this reason, semiconductor detectors, especially HPGe detectors achieve the best energy resolution and become the major, appropriate extensively used equipment for radiation spectrometry measurements for gamma photon detection (in 0.1 MeV to 10 MeV) (Knoll, 2000). HPGe detectors are consists of semiconductor diodes which are configured as reverse biased condition. By the incidence of radiation, electron-hole pairs are produced in the semiconductor crystal, in which they drifts under an electric field towards the electrodes where they are collected as electrical signals/pulses. The amplitude of the resultant electrical signal is proportional to the amount of energy that is deposited in the detector. The electrical signals from the detector are amplified by instrumental amplifier and are finally processed by multichannel analyser (MCA).

The efficiency of the NaI(Tl) detectors is higher but the energy resolution is much lower than that of HPGe detectors. For gamma energy of 1 MeV, the energy resolution of NaI(Tl) detectors are normally falls between 7−9%, whereas the energy resolution is of the order of 0.1% for HPGe detectors. The lesser the energy resolution, the better the detector's capability to distinguish between two discrete energies lie close to each other.

For NaI (Tl) detector, the 609.320 keV gamma line (^{214}Bi) is used for ^{226}Ra, and 583.187 KeV (^{208}Tl) and 911.204 keV (^{228}Ac) gamma lines are used for ^{232}Th in this work. In reality, the resolution of NaI detector is comperatively poorer than HPGe but NaI (Tl) offers higher detection efficiency. Hoewver, the line 352 keV for ^{226}Ra and 232 keV for ^{232}Th have a high gamma ray emission probability (intensity) of 35.6% and

43.6%, respectively to produce sufficiently intense peak. These lines may be used by adjisting the ADC gain and proper detector shielding to reduce background rariation.

A cylindrical multi-nuclide standard source (initial radioactivity: 5.046 μCi; source number: 89-90; reference date: 1 September 2013; nature of source: solid; source package: 500 ml plastic Marinelli beaker) from Isotope Products Laboratories, Valencia, California, USA, was used to calibrate the system (detector) for the required gamma line used in this study including 1836 keV gamma line of [88]Y.

2.7.1   Natural radioactivity in foodstuffs

Naturally occurring radionuclides particularly [238]U and [232]Th decay series and the primordial [40]K are the most common radionuclides in all environmental media including foodstuffs which are the main sources of natural radiation dose to human beings (Canbazoğlu & Doğru, 2013).

These decay series radionuclides represent the most significant source of ionizing radiation on Earth contributing about 83% of the annual effective dose experienced by the global inhabitants, while about 16% of this dose is contributed to by the primordial radioisotope [40]K and the remaining only 1% comes from the artificial sources of radionuclides (Asaduzzaman, et al., 2015; Chau et al., 2011; UNSCEAR, 2008b). Natural radionuclides are present in aquatic and terrestrial food chains that can subsequently transfer to the human body following their consumption (Alrefae & Nageswaran, 2013). It has been assessed that about one-eighth of the average annual effective doses from the natural sources have been credited via the consumption of foodstuffs (Awudu et al., 2012). Consequently, radiation exposure in humans owing to the intake of radionuclides from the food consumption is of global concern (Ababneh, Alyassin, Aljarrah, & Ababneh, 2009; Alrefae & Nageswaran, 2013; IAEA, 2010).

In the recent years, several studies have been performed in different geological locations across the globe to evaluate the radioactivity concentration in various foodstuffs and associated radiation dose received by the population due to consumption of foods. Saeed et al. (2011) estimated the activity concentrations in six varieties of marketed rice in Malaysia by gamma-ray spectrometry coupled with HPGe detector and documented the higher levels of radioactivity for radiologically important nuclides ^{238}U and ^{232}Th (25.10±1.35 Bq kg^{-1} and 64.97±0.36 Bq kg^{-1}, respectively) in white glutinous rice than the other plane brand of rice. The total effective dose received through the consumption of these rice varied from 0.02–0.03 μSv y^{-1}, which were extremely lower than the tolerable limit of 1mSv y^{-1}. But this study did not mentioned the source of origin of the studied rice. In 2012, the same research groups performed the similar study (using HPGe detector) together with evaluation of soil to rice transfer factor (TF) of these (^{238}U, ^{232}Th and ^{40}K) radionuclides in some rural and urban granary areas of Kedah, Malaysia. The finding of this study showed that both the activity concentrations of rice and soil, and soil to rice TFs of radionuclides were comparatively higher in rural areas than urban one. The soil to rice TF of ^{238}U, ^{232}Th and ^{40}K in urban areas were estimated as 0.09, 0.11 and 4.12, respectively, whereas in rural areas, this TF values were found as 0.20, 0.15 and 1.43, respectively. The study concluded that the physico-chemical characteristics of the soil and crops and soil amendment such as application of fertilized and other chemicals in the soil affects the activity concentrations and TFs. Moreover, the estimated effective dose was below the permissible limit of 1 mSv y^{-1} set by the IAEA. In the Penang island of Malaysia, Alsaffar et al. (2015) assessed the radioactivity (^{226}Ra, ^{232}Th and ^{40}K) concentrations in soil and their distribution (TF) to different parts of rice plant including grain using HPGe spectroscopy. The reported concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K in rice grain were ranged from 0.53–2.82, 0.51–1.57 and 43.51–108.47, respectively and the corresponding soils

activity were ranged from 49.4–208.51, 68.22–194.13 and 138.31–943.11, respectively. This study showed that the radionuclides are more concentrated in roots that grains. Soil to rice grain TFs were 0.0018, 0.0009 and 0.235 for ^{226}Ra, ^{232}Th and ^{40}K, respectively which were within the range of IAEA (2010) reported values of 2.2×10^{-4}–2.8×10^{-2} for ^{226}Ra, 2.2×10^{-5}–3.0×10^{-2} for ^{232}Th and 1.8×10^{-2}–7.8×10^{-1} for ^{40}K. Their findings identified that the uptake of radionuclides by plants is not only depends on their levels in soil but also influenced by physical and chemical properties of soil such as soil texture, organic matter content, cation exchange capacity, pH and electrical conductivity.

Natural radioactivity of ^{238}U, ^{232}Th and ^{40}K and fallout ^{137}Cs in marketed rice of Kuwait has been performed by Alrefae and Nageswaran (2013) employing HPGe spectrometry technique with a view to evaluate the population dose. The average activity levels of ^{238}U, ^{232}Th and ^{40}K and ^{137}Cs were documented as 0.62±0.19, 0.48±0.10, 48.60±18.34 and 0.10±0.012 Bq kg^{-1} (^{137}Cs was found only one brand imported from Germany), respectively. This study informed that the rice imported from India contains comparatively higher activity than rice imported from other countries. They also estimated effective doses 33 μSv y^{-1} for adult population and 60 μSv y^{-1} for children, which were much lower than the world average value of 0.29 mSv y^{-1}. The authors concluded that rice marketed in Kuwait is radiologically safe for human consumption without the estimation of fatal cancer risk, however, suggests to investigate the radioactivity of gamma emitters together with alpha and beta emitters in all types of foodstuffs to establish more robust baseline data. Similar study has been carried out in Iran by Pourimani and Anoosheh (2015) using the same methods which reported the activity levels in rice samples ranged of <1.27–2.89±0.86, <0.42–15.24±1.68, 84.66±3.38–122.66±4.90 and <0.27–1.00±0.26 Bq kg^{-1} for ^{226}Ra,

^{232}Th, ^{40}K and ^{137}Cs, respectively. They also measured the activity of the corresponding soil to evaluate the soil to rice TFS which varied from 0.02–0.07 for ^{226}Ra and 0.09–0.13 for ^{40}K, being greater than the IAEA (2010) reported values and find the TFs for ^{232}Th and ^{137}Cs in most of the varieties of rice lower than the minimum detectable activity of the spectrometry system. However, the TF of ^{137}Cs was found only one rice sample which was 0.23. The annual effective dose due to rice for Iranian individuals were ranged as 20.50 ± 0.74–$68.40\pm11.71\mu$Sv y^{-1}, which was lower than the global average of 0.29 mSv y^{-1}.

On the other hand, Aswood et al. (2013) investigated the natural radioactivity of ^{238}U and ^{232}Th in vegetables and corresponding soils in Cameron Highlands and Penang, Malaysia by neutron activation analysis (NAA) technique and found that activity concentrations of soils as well as vegetables were higher in highlands that the low lands. The activity levels of ^{238}U and ^{232}Th in vegetables in Cameron Highlands varied from 1.3 ± 0.14 to 6.25 ± 1.58 and 0.41 ± 0.12 to 2.5 ± 1.55 Bq kg^{-1}, respectively. However, in Penang, the activity ranged from $<1.3\pm0.41$ to 3.10 ± 1.10 and $<0.41\pm0.12$ to 0.41 ± 0.12 Bq kg^{-1}, respectively. The TFs of radionuclides from soil-to-vegetables were ranged from 0.006 to 0.031 for ^{238}U and 0.002 to 0.013 for ^{232}Th in Cameron Highlands, while in Penang, this factor varied from 0.012 to 0.028 for ^{238}U and 0.003 to 0.003 for ^{232}Th. The highest TF was estimated in the case of cucumber and eggplant. The authors mentioned that the radionuclides uptake from soil-to-vegetables depends on physico-chemical properties of soil such as cation exchange capacity, organic matter contents, pH, texture, clay content etc, and also depends on plant species, stage of growth, physical and chemical characteristics of radionuclides etc.

In south west India, Shanthi et al. (2010) evaluated the radioactivity levels of ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K in some foodstuffs by NaI(Tl) scintillation spectrometry. Among

the foodstuffs, the activity levels in rice (3.07±0.41, 4.54±0.8, 34.3±2.7, and 120.2±15.8 Bq kg^{-1}, respectively) and tapioca (3.57±1.2, 5.42±0.3, 27.4±4.8 and 181.1±12.4 Bq kg^{-1}, respectively) showed comparatively higher than the vegetables and fruits. The dose received by the population were estimated as 0.201, 0.659, 0.475 and 0.460 mSv y^{-1} for ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K with a total of 1.796 mSv y^{-1}, which was indeed higher than the world average of 0.29 mSv y^{-1} and hence this study remarked that special care should be taken to consume the studied foodstuffs. In 2012, the same research groups studied the radioactivity in food crops and soil in South India using the same methods with a view to assessed the soil to rice, vegetables and fruits TFs. The result exhibited that the soil-to-rice TF of ^{226}Ra, ^{232}Th (^{228}Th), ^{238}U and ^{40}K were found to be 8.8×10^{-2}, 14.2×10^{-2}, 5.8×10^{-2} and 6.3×10^{-2}, respectively. The corresponding values of vegetables varied from (0.3–4.4) ×10^{-2}, (0.7–2.7) ×10^{-2}, (BDL–0.425) ×10^{-2} and (2.4–8.0) ×10^{-2}. Soil-to-tapioca TFs were 6.2×10^{-2}, 11.0×10^{-2}, 1.9×10^{-2} and 8.9×10^{-2}, respectively. In the case of fruits, TF varied as (0.39–4.6) ×10^{-2}, (0.08–2.5) ×10^{-2}, (BDL–0.89) ×10^{-2} and (2.1–14.4) ×10^{-2} for ^{226}Ra, ^{232}Th (^{228}Th), ^{238}U and ^{40}K, respectively. The TFs of rice (grains) and tapioca (tubers) were showed higher value than the vegetables and fruits crops. The leafy vegetables exhibited greater TFs than non-leafy vegetables and fruits. For the majority of the food crops, non-edible portions accrued more radionuclides than the edible parts.

Due to the protection of public health, monitoring the radioactivity level in foodstuffs is of prime importance. In Turkey, Canbazoğlu & Doğru (2013) analyzed the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in vegetables and fruits by Scintillation (NaI(Tl)) detector and recorded the mean activity concentrations of 0.64±0.26, .65±0.14, 13.98±1.22 and 0.54±0.04 Bq kg^{-1} in vegetables and 1.52±0.34, 0.98±0.23, 18.66±1.13 and 0.59±0.16 Bq kg^{-1}, respectively in fruits. The adult population dose via

the consumption of vegetables and fruits estimated as 20±3.75 μSv y^{-1} and 30.55±5.72 μSv y^{-1}, respectively (fruits contribute higher dose than vegetables) with a total of 50.55 μSv y^{-1}, which was about 6 times lower than the world average of 290 μSv y^{-1} suggesting no radiological risk for the health of general public. Again, the natural radioactivity in vegetables consumed by the Jordanian people were evaluated by Al-Absi et al. (2015) using gamma-ray HPGe detector and reported the concentrations ranged from 7.1±1.1 to 11.7±3.4, BDL to 3.3±1.8 and 201±10 to 684±15 Bq kg^{-1}, respectively for ^{226}Ra, ^{228}Ra and ^{40}K. The effective doses were estimated as 23.98, 12.24 and 15.34 μSv y^{-1}, respectively for ^{226}Ra, ^{228}Ra and ^{40}K with a total of 51.56 μSv y^{-1}, which was far below the world average value of 290 μSv y^{-1}, indicating the studied vegetables were safe for human consumption. In Southwest region of Cameroon, Abiama et al. (2012) studied the natural radioactivity in vegetables by HPGe gamma-ray spectrometry and reported the mean concentrations of 2.30, 1.50 and 140.40 Bq kg^{-1}, respectively for ^{226}Ra, ^{228}Ra and ^{40}K. The total daily effective doses were estimated as 0.41, 0.84 and 0.71 μSv d^{-1} for ^{226}Ra, ^{228}Ra and ^{40}K, respectively in which cassava contribute 61% of the total dose for ^{226}Ra and 89% for ^{228}Ra. The total annual effective dose was assessed as 0.70 mSv y^{-1} which was significantly higher than the world average value. In Accra Metropolitan areas of Ghana, the activity concentrations in various marketed foodstuffs (Cassava, yam, cocoyam, potato, plantain, cowpea, millet, maize and rice) has been measured by Awudu et al. (2012) using scintillation spectrometry with NaI (Tl) detector. The concentrations of radioactivity in their analyzed foodstuffs were ranged from 3.37±1.84 to 10.46±4.66 Bq kg^{-1} for ^{226}Ra, 6.14±2.04 to 12.31±3.12 Bq kg^{-1} for ^{228}Ra, 4.33±2.33 to 14.93±3.86 Bq kg^{-1} for ^{228}Th and 87.77±8.37 to 368.50±19.20 Bq kg^{-1} for ^{40}K. Plantain is the root vegetables that showed the highest concentrations for ^{226}Ra and ^{228}Ra, while the highest activity levels

of ^{228}Th and ^{40}K were noticed in another root vegetable, cassava. The annual committed effective doses experienced by the general public from the consumption of the studied foodstuffs were varied from 0.01 mSv (millet) to 1.33 mSv (cassava) with an average of 0.42 mSv and a total of 4.64 mSv, which aws higher than the corresponding world average of 0.29 mSv y^{-1}. Their conclusion, therefore, was that radiological regulation should be applied for those foodstuffs contributing higher population dose. Choi et al (2008) investigated the natural radioactivity in most popular Korean foodstuffs (vegetables, grains, marine fishes, fruits, seaweed, meat and milk). The activity concentrations of thorium isotopes were measured by alpha spectrometer and radium and potassium isotopes were determined by HPGe detector. Their result showed that the activity concentrations (mBq kg^{-1} fresh weight) of ^{226}Ra, ^{228}Ra, ^{232}Th, ^{228}Th, ^{230}Th and ^{40}K in vegetables were ranged of 13.06±0.17−156.3±9.52, 5.65±0.08−196.0±7.75, 0.06±0.03−5.08±0.16, 0.42±0.07−23.4±0.92, 0.15±0.04−5.19±0.16 and 15.00±0.10−96.88±0.44, respectively. The concentrations (mBq kg^{-1} fresh weight) in rice were found to be 21.25±0.35, 12.77±0.08, 0.77±0.07, 9.90±0.26, 0.73±0.07 and 27.82±0.20 for ^{226}Ra, ^{228}Ra, ^{232}Th, ^{228}Th, ^{230}Th and ^{40}K, respectively. In the case of marine fishes, the highest activity levels (mBq kg^{-1} fresh weight) of ^{226}Ra, ^{228}Ra, ^{232}Th, ^{228}Th, ^{230}Th and ^{40}K were varied as 54.42±3.66−215.7±8.64, <29.04±106−288.8±8.47, 0.15±0.02−65.2±0.84, 0.80±0.14−111±1.23, 0.33±0.09−36.7±0.56 and 10.94±0.15−168.3±2.46, respectively. They identified that the daily radionuclides intake from milk, rice, Chinese cabbage, spinach and shellfish were considerably higher than the other examined foodstuffs. The total population dose from the consumption of studied foods was estimated as 110 μSv y^{-1}, of which the contribution from ^{40}K was 101 μSv y^{-1}.

In the marine environment, radioactivity is mainly contributed to by the natural processes of weathering and mineral recycling of terrestrial rocks, seabed movement arising from under sea earthquakes and submerged volcanic activity (Abbasisiar et al., 2004; Al-Qaradawi et al., 2015). In addition to natural processes, different types of human activities mentioned in section 2.1 together with post nuclear disposal of radioactive and industrial waste, underwater nuclear device tests, accident including leakage from nuclear power plants, reprocessing of spent fuel etc. are also known to increase the radioactivity in the marine environment, with bioaccumulation of marine animals, therefore acknowledged growing concern to monitor the content of radionuclides in marine lives particularly in edible fishes, molluscs and crustaceans (Al-Qaradawi et al., 2015; Alam & Mohamed, 2011; Khandaker, Wahib, Amin, & Bradley, 2013).

Because of the importance, a radioactivity survey in marine animals in the East cost of Peninsular Malaysia has been conducted by Amin et al. (2013) using HPGe gamma-ray detection system. In the case of fish category, the activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K have been reported as 0.90 ± 0.09 to 5.1 ± 0.3, 0.7 ± 0.08 to 4.5 ± 0.27 and 27 ± 3 to 175 ± 11 Bq kg^{-1}, respectively, while in crustaceans group, these values were recorded as 1.2 ± 0.1 to 3.9 ± 0.3, 0.90 ± 0.09 to 3.9 ± 0.3 and 99 ± 6 to 312 ± 16 Bq kg^{-1}, respectively. Whereas in molluscs group, the concentrations were reported as 5.0 ± 0.3, 4.0 ± 0.3 and 292 ± 15 Bq kg^{-1}, respectively.. No artificial ^{137}Cs have been found in any marine organisms. The authors concluded that their radionuclides data can be used for the assessment of any radiological impact/contamination to the marine environment in future. But this study failed to estimate the population dose and cancer risk due to consumption of marine lives. In the same year and same methods, Khandaker et al. (2013) studied the natural radioactivity in shellfishes collected from West Straits of

Malacca) and East coast of Peninsular Malaysia. The authors stated that Malaysia is one of the highest sea fish consumption countries in world, and therefore, the estimation of radiation dose due to consumption of marine animals is of prime importance concerning food safety and human health. The average activity concentrations in molluscs were found as 4.15 ± 0.41, 1.76 ± 0.20 and 197 ± 10 Bq kg^{-1}, while in crustaceans were 4.04 ± 0.41, 1.97 ± 0.21 and 261 ± 13 Bq kg^{-1} for ^{238}U (^{226}Ra), ^{232}Th (^{228}Ra) and ^{40}K, respectively. The little variations in the value in both coasts are likely to be associated with feeding characteristics, habitat, ambient water concentration and seasonal vicissitudes. The annual committed effective doses from molluscs were estimated as 83.5 μSv y^{-1}, while the respective dose values from crustaceans were 92.5 μSv y^{-1}.

The seasonal variations of radioactivity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in marine biota (measured by ICP-MS) from Manjung coastal areas of Perak, Malaysia have been reported by Abdullah et al. (2015) as 13.04 ± 0.6 (crustaceans) to 15.50 ± 0.7 (fish species), 14.72 ± 0.7 (crustaceans) to 20.65 ± 1.0 (fish species) and 508.51 ± 25.43 (fish species) to 690.09 ± 34.50 (crustaceans) Bq kg^{-1}, respectively in dry season. While in rainy season, these values were documented as 10.46 ± 0.52 (fish) to 14.39 ± 0.72 (crustaceans), 15.37 ± 0.77 (crustaceans) to 18.13 ± 0.91 (fish) and 347.45 ± 17.37 (crustaceans) to 568.89 ± 28.44 Bq kg^{-1}, respectively. In most cases, the activity levels were observed higher in dry season than rainy season, which may be associated to the increased metabolism caused by the evaporation due to an increased temperature in the periods of dry season. Whereas, the low activity level in rainy season may be related to dilution of water resulting heavy rainfall in this period. They also concluded that besides the quality of sea water, age and size of the biota could be the significant factors in assessing the radioactivity concentration.

Several varieties of seafood is eaten by Turkey, among them fish is the most consumed in quantity (20 kg/yr.). The activity levels of ^{226}Ra, ^{232}Th and ^{40}K in different varieties of fishes (determined by HPGe gamma-ray system) from Black Sea areas of Turkey have been reported by Korkmaz Gorur et al. (2012) as 0.06±0.01 to 0.96±0.36, 0.12±0.04 to 1.03±0.15 and 35.04±0.24 to 127.41±2.29 Bq kg^{-1}, respectively. Detectable amount of ^{137}Cs was found in fish samples, because this region was contaminated by Chernobyl accident in 1986. The annul ingestion dose to individuals due to the consumption of studied marine fishes were varied from 5.67 μSv y^{-1} to 19.21 μSv y^{-1} with an average of 13.48 μSv y^{-1} which was much lower than the total exposure/person subsequent from the ingestion of 2400 μSv y^{-1} in the world and hence suggesting on threat to public health. In Turkey (Bosphorus strait, Black sea coast, Marmara sea and Agean sea), Kiliç et al. (2014) evaluated the radioactivity levels in soft tissues of mussels using HPGe γ-ray spectrometry (the concentration of ^{210}Po was measured by alpha spectrometer using silicon surface barrier detector). The activity concentrations of ^{226}Ra, ^{228}Ra, ^{40}K, ^{210}Po, ^{210}Pb and ^{137}Cs have been reported as 0.88±0.06), 1.03±0.11, 469±24, 121.9±4.2, 10.9±0.9 and 0.71±0.23 Bq kg^{-1}, respectively. The activity level of ^{210}Po was found very much higher than that of ^{226}Ra and ^{228}Ra, which may cause by the greater accumulation affinity of Po in the organic matter. The total annual effective dose due to the consumption of mussel was varied between 0.25 and 3.30 μSv y^{-1}, indicating safe for human consumption.

In Bonny estuary, Niger delta, Nigeria (which is heavily polluted by various industries), radioactivity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{137}Cs and ^{210}Po in marine animals (Fishs, crustaceans and molluscs) have been investigated by Babatunde et al. (2015) using gamma-ray spectrometry with HPGe detector and reported as 4.4±0.3 (crustaceans) to 7.4±0.4 (molluscs), 9.3±0.8 (crustaceans) to 31.2±1.8 (molluscs), <22.5

(molluscs) to 152.5±9.6 (molluscs), <80.8 (crustaceans) to 120±15 (molluscs), <0.8 (fishes) to 1.5±0.1 (molluscs) and 9.4±1.1 (crustaceans) to 32.8±1.9 Bq kg^{-1}, respectively. ^{210}Po activities were the most important in terms of dose contribution. The population dose estimated 0.431 to 1.532 mSv y^{-1}. ^{137}Cs together with ^{210}Po contribute over 50% of the total effective dose from each fish species. The radiation doses from the intake of radionuclides via the consumption of any examined marine species exceeded the world average of 0.29 mSv y^{-1}. Higher consumption rates of the studied species may create public health problems due to the effects of ionizing radiation. The radioactivity levels of ^{226}Ra, ^{232}Th and ^{40}K in marine and fresh water fishes and crustaceans from Ondo State, Nigeria have been reported by Ademola and Ehiedu (2010) as 23.0±4.6–49.7±33.1, 32.1±5.3–96.7±19.9 and 688±230–791±39 Bq kg^{-1}, respectively for marine fishes; 35.3±14.9, 31.9±5.2 and 525±77 Bq kg^{-1}, respectively for crustaceans. On the other hand, the mean activity levels of ^{226}Ra, ^{232}Th and ^{40}K in fresh water fishes were varied as 21.4±3.8–38.6±11.6, 40.7±25.9–84.4±2.3 and 462±80–792±107 Bq kg^{-1}, respectively. Almost similar activity were observed in both the marine and fresh water fishes. The doses experienced by the people due to consumption of marine fishes ranged between 6.4±0.7 and 14.2±1.6 μSv y^{-1}, while this dose varied from 23.3±10.2 to 34.8±1.7 μSv y^{-1} for fresh water fish and that of the crustaceans was found to be 2.4±0.2 μSv y^{-1}. These doses were very much lower than the world average value of 0.29 mSv y^{-1} for the general public. Considering the environmental effects of oil and gas exploration with radioactive sources (Downhole logging tools comprise a variety of radioactive sources to log different features of a well), there may be a high level of radioactivity presence in the marine animals.

2.8   Heavy metal detection/analysis techniques

In order to assess the potential environmental impact and minimize the toxic problems and health challenges posed by heavy metal pollutants in environmental media, it is essential to precise determination the levels of heavy/toxic metals in the environmental components such as foodstuffs.

Over the years, there are many analytical techniques (eg., ICP-MS, LA-ICP-MS, ICP-OES, ICP-AES, GF-AAS, AAS, XRF, μSRXRF, PIXE and so on) have been developed with the aim of trace element analysis in environmental and biological samples. A precise, robust and accurate analytical technique/method is of prime importance for heavy metals measurements in environmental media including foodstuffs to ensure food safety concerning human health, maximum product safety, regulatory compliance and brand protection (McSheehy Ducos et al. 2010). Origin of determination and authenticity are also important. Among the metal analysis methods, inductively coupled plasma mass spectrometry (ICP-MS) technique complies with all these requirements efficiently. This method offers importantly lower limits of detection compared to inductively coupled plasma optical emission spectrometry (ICP-OES) and graphite furnace atomic absorption spectrometry (GFAAS). ICP-MS is an extraordinary throughput, plasma based method using single high energy excitation source offering accurate measurement of heavy metals in foodstuffs. Multi-isotopic and multi elemental features of this technique offers the possibility to analyze an entire set of elements in a one run with analyze over 50 samples/hour, saving substantial time and money and permitting quicker and cost effective decision making. Trace metals may be determined in a broad range of matrices of ppt (parts per trillion) to lower percentage levels. Another vital feature of ICP-MS is the linear dynamic range that allows for concurrent detection of major and ultra-trace elements in a single run. Due to its improved

sensitivity and high signal-to-noise ratio, it is flexible to analyze almost all elements in the periodic table simultaneously with high spectral resolution (Thomas, 2008).

2.8.1   Heavy metal pollutant in foodstuffs

The release of heavy and trace metals due to different anthropogenic activities (industrial activities, agricultural activities, transportation, waste incinerations, swage sludges in agriculture, municipal waste, combustion of fossil fuels, rapid urbanization, mining, smelting and many other human activities) into the environment (both the terrestrial and aquatic) has caused numerous environmental problems (Korkmaz Gorur et al., 2012; Liu et al., 2015; Nazir et al., 2015; Tchounwou et al., 2012). Because of their non-biodegradability and persistence nature, heavy metals may accumulate in various environmental matrices such as human food chain and may create substantial threat to human health, as for example cardiovascular, renal, and neurological impairment as well as bone diseases, mutagenic effects, damaged central nervous function, lungs, kidneys, liver, and other vital organs (Hajeb et al., 2014; Jolly et al., 2013; Korkmaz Gorur et al., 2012).

It is well established that diet is the most leading sources (90−95%) of heavy metals exposure to human (Alina et al., 2012; Mansour, 2011; Skrbic, Zivancev, & Mrmos, 2013). Therefore, evaluation of heavy metals in foodstuffs, vegetables, fishes etc. are of prime importance concerning human health and also beneficial for the decision making on the regulation of heavy metal pollutants and food safety (Skrbic et al., 2013). Among the leading pollutants of food supply, heavy metals can be regarded as the most significant problem of our environment. Food may be contaminated by the heavy metals that are present in their growing environment and find their way into the human body through the food chain (Mansour, 2011). During the last decades, food safety acknowledge increasing demand that has enthused research concerning health hazard

associated with the intake of heavy metals via the consumption of contaminated foodstuffs.

Over the past decades, a growing number of scientific food research associated to pollution by heavy metal pollutants have been conducted in different part of the world. In Isfahan province, Iran, Salehipouret et al. (2015) studied the heavy metals pollutants levels of Pb, As, Ni, Zn and Cu in vegetables, rice and wheat (using ICP-OES) and reported (among the foodstuffs) as 0.1−1.71, 0.25−0.85, 0.27−1.65, 13.5−53.65 and 085−49.09 mg kg^{-1}, respectively in which some some vegetables for Pb and rice for As exceeds the WHO/FAO (2001) permissible limit of 0.3 mg kg^{-1} and 0.15 mg kg^{-1}, respectively. Their findings showed that the total noncarcinogenic hazards of Pb and As were higher than 1 and total risk of cancer for As were higher than the tolarable risk levels of 10^{-4} to 10^{-6}, which indicates that the individual's living in Isfahan province, Iran experiencing the risk of cancer through the consumption of vegetables and cereals. Therefore, they suggested to pay special attention on the possible risk of health from heavy metals following the consumption of studied foodstuffs.

Among the foodstuffs, vegetables become the greatest exposed food to environmental contamination because of its aerial burden (Jolly el al. (2013). These authors investigated the heavy and trace metals levels in vegetables grown in Ruppur areas of Pabna district, Bangladesh using EDXRF and documented the levels of Pb, As, Cu, Co, Se, Mn, Ni, V and Zn as 0.12−0.98, <0.01−0.08, 3.62−6.69, <0.27−0.37, 0.03−0.16, <0.06−25.95, <0.65−0.94, 0.04−0.26 and 25.78−112.24 mg kg^{-1}, respectively which were lower than the world average values. The daily intake of heavy/toxic metals (Pb, Cd, V, Ni, Fe, Mn, Cu, Co, Zn and Fe) from the studied vegetables were within the permissible limit suggested by various international organizations. The hazard quotient of Pb, V, Ni, Fe, Cu, Co and Fe were below the safe

limit of 1, indicating there is no risk for these metals, while the hazard quotient for Cd, Mn and Zn exceeded the safe limits of 1, indicating the obvious health risk for intake of these metals via the vegetables consumption. The detectable amount of Cd only found in cauliflower, radish and amaranthus, therefore, these vegetables should be consumed at lower amount to decrease toxic effects.

Higher levels of metals in agrarian soils and uptake of these metals in vegetables, rice and other human food crops become one of the critical health problems in Asia region (Islam et al., 2015). These research groups showed that vegetables contribute the greatest intake of Pb and Ni, while the highest ingestion of Cd, As, Cu and Cr are contributed to by rice. Most of the heavy metal levels exceeded the tolerable limit and hence the investigated foods are unsafe for populations and therefore, the metal pollution sources in foods would be regulated. In the same year, Li et al. (2015) in China reported that the levels of heavy metals (Pb, Cd, Ni, Cu and Cr) in waste incinerator site were quite greater than the site which was far from the vicinity of incinerator, indicating that incinerator create environmental pollution to the neighboring areas. This is one of the great remarks. They also reported that the aerial parts of vegetables accumulate greater levels of heavy metals than roots, indicating that vegetables bio-accumulated lower quantities of heavy metals from soil and the foliar uptake may be a vital route of metals from the environment to vegetables. This study did not report the metal levels between root uptake and foliar uptake separately.

Heavy metals (Pb, Cd, Ni etc.) contamination due to rapid industrial growth, advances in agricultural chemicals such as fertilizer, pesticides, fungicides, herbicides, and/or urbanization process is a major problem of our environment and they are also one of the major contaminating agents of our food supply. (Orisakwe et al., 2012). In Nigeria, they studied the heavy metals in foods and reported that the levels of Pb, Cd Ni

etc. exceeded the recommended safe levels and recommended the monitoring of these toxic metals regularly in sewage and effluents and in foods for the prevention of excessive accumulation in food-chain. In Tarkwa of Ghana, Bortey-Sam et al. (2015) reported that Cd, Co, Cr, Pb, Zn, Hg and Ni were distributed highly in cassava, however, Cu and As were greatly distributed in plantain. They found that Ni had greater absorption capability into food crops from soil when compared to other metals. The authors recommended the regular monitoring of most toxic heavy metals (Cd, As, Hg and Pb) in foodstuff, fruits, vegetables etc. in the areas of Tarkwa gold mines. Food is one of the major ingestion sources of toxic heavy metals by humans (Millour et al., 2012). These research groups reported in French that the greatest mean concentrations were in the food groups "Sweeteners, confectionery, honey and shellfish" (for Ag, Sr, Ba, Te, Ga, Fe, Ge and V). The levels of Sn were highest in fruit and vegetables. Since dietary ingestion of trace metals are of great anxiety for public health, therefore, the authors suggested that regular monitoring of these metals in foodstuffs are needed and latest data on dietary ingestions of trace metals in different countries should be updated on regular basis. In the vicinity of a former Pb smelter plant in Northern France, Douay et al. (2013) studied the Pb, Cd and Zn concentrations in vegetables and crops and reported that the levels of Pb, Cd and Zn for vegetables varied from 0.35 to 3.19, 0.13 to 6.76 and 26.8 to 136.0 mg kg^{-1}, respectively, while for agrarian crops, these values ranged from <0.22to 1.23, 0.05 to 1.38 and 19.0 to 73.8 mg kg^{-1}, respectively. Their report showed that the agrarian, urban and vegetable garden top soils are highly contaminated by Pb, Cd and Zn and hence transmitted the crops through uptaking. The presence of toxic and/or heavy metals in the environment is rather significant in assessing possible risks for human health once present in the terrestrial and aquatic food chain (Grybauskaite et al., 2014). Grybauskaite et al. (2014) determined the concentrations of Pb and Cd in different food groups and recorded the concentration of

Pb in iceberg lettuce and potatoes exceeded the maximum allowable concentrations (0.3 and 0.1 mg kg^{-1}, respectively). In the case of fish and seafood groups, the greatest concentrations of Cd was found in France and Spanish squid with the values of 0.8669 and 0.5868 mg kg^{-1}, respectively which also exceeded tha permissible limit. They suggested that regular monitoring of heavy metal in human foods is needed to ensure food safety. These researchers suggested measuring the heavy metal using atomic absorption spectrometry (AAS), but AAS produce high interferences with polyatomic and isobaric elements which is problematic to accurate measurement of some metal. Using tha same method, Cherfi et al. (2014) measured the heavy metals of Pb, Zn, Cu and Cr in various marketed vegetables and fruits in Algeria and recorded that their levels as 12.33 to 39.33, 11.17 to 49, 3.83 to 29.49 and 3 to 16.33 mg kg^{-1}), respectively. They reported that the vegetables and fruits may be contaminated by heavy metals through the roots uptake from polluted soils and irrigation water along with foliar uptake of aerial parts of the plants from the polluted environments. The estimated daily intake (EDI) of Pb was 4 times higher than the tolerable intake of 248.47 µg day^{-1} for Pb, indicating obvious risk of health associated to Pb. Therefore, the authors suggested to take special care mainly in the levels of Pb due to food safety for the population and recommended to reduce the exposure to leaded fuel and inappropriate irrigation water. In Brazil, Guerra et al. (2012) reported the heavy metal concentrations of Pb, Cd, Ni and Cr in vegetables and fruits as 0.02–2.5, 0.01–0.18, 0.01–0.74 and 0.01–0.6 mg kg^{-1}, respectively. This result of heavy metals is quiet smaller than that found by Cherfi et al. (2014) in Algeria but exceeded the Brazilian legislation limit. They stated that higher level of Pb in several vegetables may be attributed to the crops land situated nearby roads of heavy traffic. They also pointed out that the major sources of Pb to human beings are the airborne Pb inhalation from vehicular emissions and from

direct atmospheric deposition on water, soil and crops, forming the entryway into the food chain.

Ingestion of heavy metal contaminated food may extremely reduce some essential nutrients in the body resulting a decline in immunological defenses, impaired psycho-social behaviors, intrauterine growth retardation, malnutrition related disabilities and high prevalence of upper gastrointestinal cancer (Matos-Reyes et al., 2010). The heavy metal levels in marketed foodstuffs from Serbia have been reported by Škrbić et al. (2013) that the concentrations of Pb ranged of <0.003–0.080 mg kg^{-1}, Cd varied as <0.0033–0.009 mg kg^{-1}, and As was below the 0.03 mg kg^{-1} for vegetables groups. In sea fishes, the levels of Pb, Cd and As were reported as <0.003, 0.003 and <0.03 mg kg^{-1}, respectively. The total intake of Pb was estimated to be 72.30 μg day^{-1}, which was about two times greater than the toxicological level (44 μg day^{-1}) for nephrotoxic effects. Ismail et al. (2005) investigated the heavy metal levels in vegetables grown in Cameron Highlands and Sepang, Malaysia and reported that most toxic metals such as Pb, Cd and Cr were found in small amounts in all vegetables in both the studied areas. They also reported that the levels of metal in Sepang (low land) are comparatively higher than that of Cameron highlands, because Sepang areas are more polluted by various anthropogenic activities that clean areas of highlands.

The heavy metal levels of Pb, Cd, Cu and Zn in canned vegetables marketed in Turkey has been studied by Koçak et al. (2005) using Differential Pulse Polarography (DPP) and reported as 0.192 (bean to 0.660 (garniture), 0.283 (pea) to 0.590 (okra), 0.867 (okra) to 4.388 (garniture) and 0.564 (bean) to 6.101 (okra) mg l^{-1}, respectively. Among the vegetables, okra showed higher levels of metal concentration. Recently, the consumption of canned food has been increases worldwide, hence regular and rapid monitoring method is needed which has sensitive and selective multi-element

capabilities. The commonly used techniques such as ICP-MS, ICP-OES, ICP-AES, NAA, XRF, AAS etc, are very expensive, need extra apparatus and time-consuming and sometimes do not offer adequate sensitivity for reproducible determination of trace and ultra-trace levels of elements. Instead, a rapid Differential Pulse Polarography (DPP) has been recommended for the simultaneous determination of heavy metals. This technique is not only one of the most vital electroanalytical tools which is capable of metal analysis in foods but also a rapid, sensitive, reproducible, selective and suitable for quantitative analysis of multi-element at trace and ultra-trace levels in food commodities as quality control tools. On the other hand, Omer (2015) carried out a research work for the investigation of the levels of Cr, Cu, Fe, Mn, Ni, Pb, Rb, Sr and Zn in fruits from Sudan using X-ray fluorescence technique (XRF) and reported that the levels of most of the heavy metals were extremely higher in commercial fruits than that of controlled ones and literatures values published elsewhere. They concluded that the higher levels of metal pollutants may possibly be credited to pollutants in the high ways traffic and/or stocking processes. Again, Margui et al. (2005) performed the quantitative analysis of Pb, As, Sr, Zn, Co, Fe, Mn, Al in vegetation from Spain using wavelength dispersive XRF technique to observe the performance of this method. They successfully measured the said metals in mining and control areas and reported that most of the metals were higher in mining areas than that of controlled areas, indicating pollution from mining activities which is agreeing with the conclusion of Omer (2015). They highlighted that the XRF is a non-destructive method for direct, rapid and sequential ad/or simultaneous analysis/determination of metals in vegetation samples. This method prove to be good and effective tool for the environmental study and quality control in vegetation. The ease of sample preparation, least manipulation and speed of analysis have indorsed XRF as an alternative to environmental spectroscopic methods.

The aquatic especially marine environments contamination via heavy metals is a great global concern due to their toxicity, long term persistence, non-biodegradability and succeeding accumulation in aquatic/marine habitats (Hajeb et al., 2014). The marine environments is contaminated by untreated industrial effluents, rapid urbanization (waste stream of urbanization), discharge of municipal wastes, shipping activities, increasing tanker trafficking, oil-gas exploration, operation of power plant and fishing activities (Alina et al., 2012). In the Strait of Malacca, Malaysia, Alina et al. (2012) studied the heavy metal of Hg, As, Cd and Pb in some marine fishes and shellfishes and reported that trace amounts of al these heavy metal were present in all studied samples, because the Strait of Malacca is considered one of the most polluted channel of the word. The levels of reported heavy metal were found higher in Shellfishes than fishes, but the concentrations were below the permissible limits of 0.5, 0.1-5.0, 0.05, 1.5 μg g^{-1} for Hg, As, Cd and Pb, respectively recommended by WHO/FAO (2004) and considered to be safe for human consumption. Bashir et al. (2013) investigated the levels of heavy metal in marine fishes from Mersing (East coast) and Kapar (West coast), Peninsular Malaysia and reported that although the West coast considered as more polluted than the East coast, most of the metals (except Cd) were found little bit higher in East coast than the West coast. The fish species *Arius thalassinus* contained higher levels of metals than the species *Johnius belangeri* in both regions but in both cases the metal levels were found lower than the maximum permissible levels set by WHO/FAO (2004) and therefore, safe for consumption without health risk. Mukherjee and Bhupander (2011) from India studied the heavy metal of As, Cd, Hg in marine fishes levels and have been recorded as 0.05 to 0.83, 0.16 to 0.86 and 0.28 to 0.93 μg g^{-1} dry wt, respectively which were much lower compared to WHO/FAO recommended limit. Since toxic metals are bio-accumulated in fish tissues, they suggested to frequent monitoring of the heavy metals as well as other pollutants such as polychlorinated

biphenyls (PCBs) dioxins and furans in marine fishes. Heavy metal pollutants through consumption of marine products, with bioaccumulation leading to potential risks by way of long term exposure (Korkmaz Gorur et al., 2012). In the Black Sea areas of Turkey, Korkmaz Gorur et al. (2012) reported that the levels of Pb, As, Fe, Mn, Cr, Zn, Ni and Cu as <0.001 to 0.02, <0.1 to 4.40, 7.91 to 19.11, 0.56 to 1.10, <0.1 to 0.24, 7.76 to 15.05, <0.001 to 006 and 0.40 to 1.21 $\mu g \ g^{-1}$ dry.wt, respectively. The concentrations of all analyzed metals were below the values established by European regulations. The estimated daily intake of these heavy metals due to consumption of the studied fish species were within the safe limit set by various international bodies. They remarked that the presence of heavy metals in the fish species provides an indication of environmental conditions alongside the Black Sea region.

2.8.2   Heavy metal pollutants in human teeth

Environmental pollution by heavy metals released from anthropogenic activities is a matter of human health concern all over the world including Malaysia due to their non-biodegradability, toxicity and long-term persistence (Alina et al., 2012; Alomary et al., 2006; Barton, 2011; Gdula-Argasinska et al., 2004; Lu et al., 2015; Millour et al., 2012; Tvinnereim et al., 2000).

There are several methods, such as physical and chemical methods and with bio-indicators that can be used for the assessment of environmental pollution (Kamberi et al., 2012). Recently, interest is grown among the researchers on the use of human bio-indicators, as for example blood, urine, bone, teeth, finger nails and toes, hair and saliva for the evaluation of environmental pollution by the way of toxic/heavy metals (Abdullah et al., 2012; Arruda-Neto et al., 2009; Arruda-Neto et al., 2010; Barton, 2011; Brown et al., 2004; Kamberi et al., 2012; Kantamneni, 2010).

Teeth (dentin, enamel or whole tooth) which cover much longer lifespan as substrates for toxicological analyses are suitable bio-indicators of heavy metals exposure to environmental pollution and superior to blood, nail, saliva, or hair as an indicator of chronological metal exposure from environment because the metals accumulated in dental hard tissues during formation and mineralization processes are to a large extent retained and relatively stable and once formed, these dental hard tissues (e.g., dentin and enamel) are not subject to significant turnover of appetite as in other biological indicators, therefore, provide a permanent, cumulative and stable record of both past and/or recent environmental exposure by heavy metals (Abdullah et al., 2012; Alomary et al., 2006; Amr, 2011; Mohamed Amr & Helal, 2010; Appleton et al., 2000; Arora et al., 2006; Arruda-Neto et al., 2010; Barton, 2011; Brown et al., 2004; Gdula-Argasinska et al., 2004; Kamberi et al., 2012; Kantamneni, 2010; Kolak et al., 2011; Kumagai et al., 2012; Malara, Kwapulinski, & Malara, 2006; Oprea et al., 2013; Zhang et al., 2011).

Several researchers from different countries carried out research work to evaluate the environmental pollution using teeth as bio-indicator of heavy metal exposure to environment. In Klang Valley region of Malaysia, Chew et al. (2000) investigated the levels of Pb, Zn and Cu in adult human teeth and reported that the levels of Pb of the examined population varied between 1.7 to 40.5 $\mu g\ g^{-1}$, which were neither very low nor very high. Interestingly, an excess values (17.1 $\mu g\ g^{-1}$) of Pb was found in the teeth of six lorry drivers who are considered as low socioeconomic people, whereas relatively higher Pb levels (21.3 $\mu g\ g^{-1}$) was reported in the teeth of older aged people. They concluded that their findings are straight evidences for the incorporation of lead in teeth. But, they did not either investigated the other importanr heavy/toxic metals nither mention the sources of Pb and how it incorporated in the human teeth, which is

desirable. On the other hand, the levels of Zn and Cu in the teeth were reported as 123.0 and 0.6 $\mu g\ g^{-1}$, respectively which were lower than the body nedds. Therefore, it is suggested that there is a need for Zn and Cu supplementation in the diet of the studied population. Similarly, Arruda-Neto et al. (2009) in Brazil reported that the levels of Pb in deciduous' teeth living in contaminated area was about 40% higher than those from control region, indicating that environmental Pb is more abundant in polluted areas. They also reported that the levels of heavy metal exposure in teeth is influenced by gender and teeth conditions in which boys teeth and carious teeth exposed 20% and 33% more Pb than the girls teeth and non-carious teeth, respectively. The syudy of decidous teeth does not carry the chronological information of hevy metal. Therefore, permanent teeth is needed to investigate. Again in 2010, the same research group studied the environmental burden of heavy metal (Pb, Cd, Fe, Zn, Mn, Ni and Cr) in the same place using human teeth of different ages and reported that the metal levels obtained in the control region were 40%−60% lower than those of polluted region, indicating that there is no change of pollution level. They did a good job but, did not report the sources of metal pollution in their said polluted areas. Likewise, Appleton et al. (2000) evaluated the heavy metal levels in animal (Bank Vole) teeth as an exposure marker of environmental pollution in some places of environmentally polluted and non-polluted areas in South Poland and reported the levels of Pb and Cd in polluted areas were much higher than the controlled (expectedly less polluted) areas which is in agreement with the study of Arruda-Neto et al. (2009, 2010). After four years, Gdula-Argasinska et al. (2004) carried out the same rrsearch in the same regions and observed relatively lower levels of metal in the teeth of animal (Bank Vole) than the Appleton et al. (2000) investigated result. They stated the reasons behind this situation were some legislation imposed by Polish law, the use environment friendly new technology in industrial processes, and the shutdown of some environmentally dangerous industries.

Therefore, the condition in the environment become better than in the past. Similarly, Oprea et al. (2013) reported the significantly higher levels of heavy metals (Zn, Fe, Cu, Ni, In, Nd, As and Cr) in the permanent human teeth of urban population that rural ones. They showed the greatest ratios of metal concentrations for Zn (6.65), Fe (5.68), In (4.75), Nd (3.83), As (3.67) and Cr (3.36) indicating that the metal levels in teeth is associated to the individual's urban exposure. They also reported that correlations of Zn, Fe, Cu, Ni and Cr in teeth increases with older age, with females teeth tissues than males ones and more in incisor compared to molar.

Human teeth are suitable biomarker for the assessment of environmental exposure to Cd and also find out the effects of some factors, such as gender, smoking habit, residency, age to incorporate Cd into dental tissue (Al-Jubouri & Bashbosh, 2012). They authors investigated the levels of Cd in teeth of some Iraki cement factory workers and reported that the concentration of Cd significantly increased in exposed people (workers in a cement factory) than the controlled ones. They also reported that the concentration of Cd in teeth of smoker's, urban population and older ages were significantly higher than those of non smoker's, rural population and younger ages, but did not explain behaind the reasons. Similar study has been conducted by Alomary et al. (2006) in Jordanian teeth and recorded the levels of Pb and Cd with the range of 0.74–69.15 and 0.06–2.16 μg g^{-1} dry wt, respectively. They reported that the levels of Pb and Cd in smoker's, males, older ages and amalgum filling teeth were significantly higher than that of non-smokers, females, younger ages and non-filling teeth, indicating that these factors substantially influences the contents of Pb and Cd in human teeth. They also recorded the higher levels of Pb and Cd in the teeth of polluted sities than those of quite clean cities of Jordan. Moreover, their investigation showed that the teeth brushing daily with toothpaste did not differ the teeth without brushing on the levels of Pb and Cd in teeth, which is a good findings. This type of study is realy significant for

the future researchers. In Japan, parallel work has been done by Kumagai et al. (2012) and reported that the levels of Pb, Zn, Sr, Cu, Co and B in tooth dentin increases with age, indicating the cumulative incorporation of metals in teeth. The possible reason that a huge quantities of collagen fibers are contained in dentin and the elements that having affinity for collagen fiber are engrossed in the body as well as increased the cumulative dosage and hence concentration of elements increase with age. Whereas, significant differences in the levels of Pb and Co between men and women were reported. Thus human dental dentin can be used as a suitable substance for relatively sex and age at future research. In India, Hegde et al. (2010) reported that the concentrations of Pb in primary teeth significantly greater than blood lead levels indicating the cumulative effect of Pb exposure against the Pb levels in blood. Therefore, the teeth Pb could be effective biological indicator. They also stated that there is no significant variation of Pb levels with age indicating that the levels of exposure from different environmental and alimentary medias may contribute more than age to the Pb incorporation in teeth. Some differences in Pb levels were found between the teeth types which may be owing to the differences in metal exposure at the periods of formation of teeth. The primary teeth reflect increasing lead exposure and prove to be superior biomarkers of body burden of lead. Likewise, Kern and Mathiason (2012) reported that the levels of Pb and Zn increased with age, but the levels of Cu did not follow this trend. High level of Zn indicates the high protein diets, while the significant presence of Pb in teeth indicating the pollution of the environment. Males teeth showed higher levels of Pb than females one. The excess and the deficiency of toxic and essential element in human diet can be assessed with this method used. For elemental analysis, wisdom teeth should be used because these teeth are generally uncontaminated compared to other teeth. Concentration of Pb which is favourably accumulated in human teeth can be used as an index of environmetal contamination and its concentration greater than 4 mg kg^{-1}

causing a toxic body load (Amr, 2011). He reported that the levels of Na , Al, Mg, Fe, Ni, Cu, Pb, Ca, Sr, Ba, and U were significantly higher in permanent teeth compared to the deciduoue teeth, and the reverse results showed for the cases of Mn, Co, Se, As, Mo and Bi which is the indication of chronological exposure of metal in teeth. But the author did not study the pollution level using teeth as exposure indicator. Brown et al. (2004) reported that the levels of Pb, Ba, Al, Mg, Sr, U and Ce were higher Ugandan deciduous teeth, while opposite result were shown in the case of Cu, Ca and Zn, which were higher in UK teeth, indicating the socioeconomic status is an effective factor. They concluded that the content of trace metal in human primary teeth is influenced by environment and the teeth can be used for identification of nutritional status. Webb et al. (2005) reported that the level of Pb in deciduous teeth of Kalama, Egypt and ancient teeth of NYABG were higher, indicating potential Pb contamination, whereas the teeth of Bronze age population Tell Abraq contains lowest levels of Pb. Conversely, relatively lower level of Pb contained in the deciduous teeth of Soli's, Maxico although this population used lead-based glaze pottery for cooking, food storing and eating, which could be the cause of high ingestion of Ca by these populations via the consumption of tortilla. On the other hand, Zn concentration was highest in the children's teeth of Kalama, Egypt, might be the intake of yeast-leavened breads that serves the availability of Zn. Comparatively lower levels of Zn was found in deciduous teeth of Solis, Mexico, indicating that the intake and absorption of Zn may be difficult for the Solis populations. The higher levels of Sr was found in the Bronze teeth of Tell Abraq and the teeth of another ancient population groups of NYABG than other two cotemporary groups namely, deciduous teeth of Kalama, Egypt and Solis, Mexico. Therefore it can be concluded that the metal (Pb, Zn and Sr) exposure in teeth reflects the environmental, dietary and nutritional history of the group of populations. This work

is very interesting which evaluate and compare the the metal levels of different ethnic and century population.

The spatial distribution of Pb in the enamel and dentin of human deciduous teeth from Austrialia has been investigated by Arora et al. (2006) and reported that the postnatal dentin (after 4 months: 4.54±3,48 μg g^{-1}) contain significantly higher levels of Pb than in prenatal dentin (after 4 months: 1.39± 0.21 μg g^{-1}. However, in the case of enamel opposite result has been observed. They concluded that the distribution of Pb in human primary dental dentin can be used to get information about environmental exposure of Pb in the time of prenatal and postnatal periods, and dentin-lead can be significantly used as a bio-marker of exposure of Pb. Similar study has been conducted by Grobler et al. (2000) in deciduous teeth in South Africa and reported the mean concentration of Pb in dental dentin and enamel compartment were 2.23±1.32 μg g^{-1} and 0.33±0.26 μg g^{-1}, respectively which were almost alike to the findings of Arora et al. (2006) in the same tooth compartment. Dolphin et al. (2005) investigated the levels of trace elements in prenatal and postnatal areas of human primary dental enamel using and found meaningfully greater levels of Pb in postnatal enamel. Hence, the enamel-lead levels can be used for the differentiating individuals experiencing different concentrations of exposure. Saiki et al. (2009) reported that the concentrations of Zn, Mn, Cl, Na and Sr were greater in carious tooth dentin than those obtained for non-carious dentin. On the other hand, Zn, Sr, Mn and Cl were found higher in carious teeth enamel than those found in non-carious ones. Furthermore, the concentrations of Zn, Mn, Sr, Ca and Cl were significantly higher in sound enamel than sound dentin tissues. Therefore, it is concluded that the incorporation of some heavy metal in teeth is a cause of carious formation in teeth. Trace elements (i.e., Pb) content in human deciduous and permanent teeth is an appropriate monitor for the demonstration of environmental

pollution over zoological and fitological sample analysis in ecological study (Kamberi et al., 2012). Because, teeth tissues are very hard, durable and relatively stable and once heavy metals such as Pb accumulate in these calcified tissues are large extent retained as there is no turnover of apatite in teeth. Kamberi et al. (2012) indicated the three areas of major difficulties for the interpretation of the analytical outcomes for Pb in teeth. First, the distribution of Pb all over the teeth is not homogeneous; secondly, the levels of Pb vary with the type of teeth that relates to the tooth age and lastly, the results from different laboratories varied significantly that point out the problems with contamination, pre-treatment of the sample and analytical methods. Thus, for the assessment of heavy metals in teeth, a standard protocol should be developed and it is suggested that instead of a part of the tooth, whole tooth and bigger sample sizes should be considered for the evaluation of the level of Pb.

Above literature revealed that different analytical methods can be used for the evaluation of heavy metal in human and animal teeth. All elemental analytical method generate some polyatomic and isobaric interference, which should be addressed properly to achieve the accuracy of the result, but surprisingly, none of the literature address this important issue. The whole tooth and the tooth compartment, tooth of different age, ethnic, residency, smoking habits, professions, dietary habits, socioeconomic status people should be analyzed for heavy metal to compare the result, establish their relationship and find out the suitability. The literature also established that the human and animal teeth can be effectively used as a bio-marker of environmental pollution as well as nutritional status study.

CHAPTER 3: COMPILATION OF PUBLISHED PAPERS

3.1 Author Contributions

The author of current thesis, I myself (Khandoker Asaduzzaman) is the core contributor of all the published papers included in this thesis. As a core author, I designed the experimental set up, collected the sample, carried out all the experimental work, analysed and interpreted the results and wrote manuscripts for publications. All the co-authors have notable contributions in the published work included in the present thesis. The contributions of each author are described as follows:

Associate Professor Dr. Mayeen Uddin Khandaker, Professor Dr. Yusoff Mohd Amin and Mohideen Salihu Farook supervised the present research work. They helped in the development of experimental set up and measurement techniques. Associate Professor Dr. Mayeen Uddin Khandaker helped in explaining the results, editing and reviewing all the papers before their submission to the journals and also provided guidance to handle the reviewer's comments before acceptance. R.H. Mahat, R. M. Nor and Hasan Abu Kassim aided in data analysis and result production in the respective paper. Noor Liyana Mohd Nasir, Michael Adekunle Olatunji, Z. Zainuddin, K. S. K. Shuib, N. A. Hakimi, S. M. Nawi and A. R. Usman helped in sample collection and measurement in the respective paper. D. A. Bradley, M. S. Farook, P.J. Jojo, Tareq Alrefaed, E. Daar, H. Ahmed and A. A. Okhunov provided the intellectual inputs especially in the manuscript editing and discussion part of the relevant paper.

3.2 Publications

A list of seven research publications is given in the following. All these publications have collectively contributed to accomplish the main goals and objectives of the current thesis.

1st Publication is the reprint of our paper, "Uptake and distribution of natural radioactivity in rice from soil in north and west part of peninsular Malaysia for the estimation of ingestion dose to man", *Annals of Nuclear Energy, 2015. 76, 85–93*, authored by Kh. Asaduzzaman, M.U. Khandaker, Y.M. Amin and R. Mahat.

2nd Publication is the reprint of our paper, "Evaluation of radionuclides transfer from soil-to-edible flora and estimation of radiological dose to the Malaysian populace", *Chemosphere, 2016. 154, 528-536*, authored by Mayeen Uddin Khandaker, Noor Liyana Mohd Nasir, Kh Asaduzzaman, Michael Adekunle Olatunji, Yusoff Mohd Amin, Hasan Abu Kassim, D. A. Bradley, P. J. Jojo and Tareq Alrefaed.

3rd Publication is the reprint of our paper, "Soil-to-root vegetable transfer factors for ^{226}Ra, ^{232}Th, ^{40}K, and ^{88}Y in Malaysia", *Journal of Environmental Radioactivity, 2014. 135, 120–127*, authored by Kh. Asaduzzaman, Mayeen Uddin Khandaker, Y.M. Amin, D.A. Bradley, R.H. Mahat and R.M. Nor.

4th Publication is the reprint of our paper, "Measurement of radioactivity and heavy metal levels in edible vegetables and their impact on Kuala Selangor communities of Peninsular Malaysia", *Radiation Protection Dosimetry, 2015. 167 (1–3), 165–170*, authored by Kh. Asaduzzaman, M. U. Khandaker, Y. M. Amin, Z. Zainuddin, M. S. Farook and D. A. Bradley.

5th Publication is the reprint of our paper, "Assessment of Radiation and Heavy Metals Risk due to the Dietary Intake of Marine Fishes (Rastrelliger kanagurta) from the Straits of Malacca", *PLOS ONE, 10(6), 1-16. doi: DOI:10.1371/journal.pone.0128790*, authored by M. U. Khandaker, Kh. Asaduzzaman, S.M. Nawi, A.R. Usman, Y.M. Amin, E. Daar, D. A. Bradley, H. Ahmed and A. A. Okhunov.

6ᵗʰ Publication is the reprint of our paper, "Natural radioactivity and effective dose due to the bottom sea and estuaries marine animals in the coastal waters around Peninsular Malaysia", *Radiation Protection Dosimetry, 2015. 167 (1–3), 196–200*, authored by M. U. Khandaker, M. A. Olatunji, K. S. K. Shuib, N. A. Hakimi, N. L. M. Nasir, Kh. Asaduzzaman, Y. M. Amin and H. A. Kassim.

7ᵗʰ Publication (submitted to Chemosphere) is the reprint of our submitted manuscript, "Heavy metals in human teeth; a bio-indicator of metal exposure to environmental pollution", authored by Khandoker Asaduzzaman, Mayeen Uddin Khandaker, Yusoff Bin Mohd Amin, Mohideen Salihu Farook, Nurul Atiqah Binti Baharudin and David Andrew Bradley.

3.2.1 Published paper 1

Uptake and distribution of natural radioactivity in rice from soil in north and west part of peninsular malaysia for the estimation of ingestion dose to man

CrossMark

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ARTICLE INFO

ABSTRACT

Paddy is the third most widely planted crop in Malaysia and most of the Malaysian people consume rice as their staple food. Hence, studies on the uptake of naturally occurring radionuclides by rice from soil of widely rice cultivated areas in Malaysia have been performed under normal field environments in order to evaluate various radiation hazards via rice consumption. The soil-to-rice grain transfer factors and the annual effective dose have been assessed for the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K. The estimated transfer factors for ^{226}Ra and ^{232}Th were found far beyond compared to the IAEA reported value for rice. Among the detected radionuclides, ^{40}K shows the highest transfer factor in all study locations but close to the IAEA reported range. The total effective dose obtained due to an ingestion of radionuclides via rice consumption was within the range of world average value (290 μSv y^{-1}) compiled by the UNSCEAR (2000) in all study areas. On an average, the excess life time cancer risk (ELCR) values via rice consumption were found below the acceptable limit of 10^{-3} for radiological risk.

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1. Introduction

Humans and their foodstuffs are exposed to various types of radiation that are originated from primordial, cosmogenic, terrestrial, natural decay series radionuclides (Dinh Chau et al., 2011). Every day, we ingest and inhale radionuclides via the consumption of food, water and air respectively. All types of food including rice contain detectable amount of radioactivity which successively relocate into the human body via the ingestion pathway. Potassium (^{40}K), thorium (^{232}Th), uranium (^{238}U) and their numerous progeny are the common radionuclides available in food and water (Yu and Mao, 1999; Natural Radioactivity – Idaho State University, http://www.physics.isu.edu/radinf/natural.htm). According to the UNSCEAR (1988, 2008) and Dinh Chau et al. (2011), an amount of 83% annual effective dose is experienced by individuals due to the natural decay series radionuclides whereas 16% is contributed by the primordial ^{40}K, and the remaining 1% is due to the anthropogenic radionuclides.

The well-known 16 elements (carbon, hydrogen, oxygen, nitrogen, phosphorus, sulfur, potassium, calcium, magnesium, iron, manganese, zinc, copper, molybdenum, boron and chlorine) are considered to be necessary for the growth and reproduction of plant (Karunakara et al., 2013). Besides these, a number of other natural radioactive elements such as U, Th and their progeny, ^{40}K, ^{7}Be, and artificial radioactive elements like ^{137}Cs, ^{90}Sr are present in plants in different concentrations (Karunakara et al., 2013). Some element may or may not be required for human metabolism. As an example, the element like radium and uranium are known to be present in plants although they are not identifying for the purpose of metabolic function. Radioactive elements ordinarily present in soil and normally not utilized in plant metabolism are absorbed in a manner independent of their radioactive properties.

Soil effluence by natural and fallout radionuclides has a nonstop radiological effect since it is freely transferred to human body via food chain like edible crops and drinking waters. Plant uptake is the main lane for the relocation of radionuclides from soil-to-human foodstuff. If these radionuclides conveyed to the edible portions of the plant, may cause a source of cumulative exposure to man (Shanthi et al., 2012). However, the uptake and distribution of radionuclides in plants depend on several factors such as soil pH, kind and amount of clays, exchangeable Ca and K and organic matter contents, physicochemical properties of the radionuclide, type of crop (crop species and variety, and cultural practices), fertilizer application, irrigation, plowing, liming and climate conditions etc. (Shanthi et al., 2012; Pulhani et al., 2005; Coughtrey and Thorne, 1983). Diet is the leading cause of human exposure to

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radioactive elements that guides to internal radiation doses (Saeed et al., 2012; Chen et al., 2005; Gaso et al., 2000).

Soil-to-plant and plant-to-human body is one of the foremost corridors for transmission of radionuclides to human organism (IAEA, 1982). After uptake by root, radionuclides are transferred to plant along with other nutrients or mineral necessary for their growth and reproduction of the plant (Joshy et al., 2011). These radionuclides translocate toward different parts of plant through the vascular system comprising the xylem and phloem; accumulate in several parts including the edible portions and would lead to endless radiation dose to man once consumed (Pulhani et al., 2005; Carini, 2001). Therefore, it is an important study to the spatial erraticism of natural radioactivity in soil and related radiation exposures through specific land produced food-stuffs.

The soil-to-plant transfer factor is one of the key parameters extensively used for the estimation of internal radiation dose from radionuclides through food consumption (Tsukada et al., 2002). Among the various kinds of food, rice is regularly consumed worldwide as well as countrywide like Malaysia. Consequently, human radiation exposure owing to the ingestion of radionuclide via rice consumption is a global concern (Alrefae and Nageswaran, 2013; Alrefae, 2012; IAEA, 1989; Yu and Mao, 1999). Rice (*Oryza Sativa L.*) is the major food crop and being the principal staple food for Malaysian in their basic daily diet, therefore the consumption of rice is treated as the most important pathways for the transfer of radionuclides into humans. It is consumed in the form of both boiled and white rice. Rice is also the dominant staple food crop in humid tropical and sub-tropical countries all over the world (Uchida et al., 2009). More than half of the world's population consumes rice as their main food. Paddy is the third most widely planted crop in Malaysia after oil palm and rubber. According to the department of agriculture (DOA, Malaysia), the estimated area of paddy planting in 2011 was about 687,940 hectares with potential yield of 2,578,519 metric tons in which rice production was 1,661,260 metric tons (DOA, 2012). The domestic consumption of rice per capita per year in Malaysia is at 95.9 kg in 2011/12 (GAIN Report MY3002, Malaysia, 2013).

Since the uptake of radionuclides from soil-to-plant and plant parts fluctuates with respect to the geological and geographical locations, it is suggested to use site-specific data (Joshy et al., 2011; IAEA, 1994) for an estimation of radiation hazards. As mentioned above, the transfer factor (TF) could differ by areas due to different climates, soil types and vegetations, therefore local TFs should be observed. Hence field studies under natural conditions in most of the rice grown areas of peninsular Malaysia were undertaken. Present study focuses on an estimation of radionuclide uptake through the measurement of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in different rice samples and the associated soils collected from various places of peninsular Malaysia, and then determination of radiological impact causing from the consumption of rice grains. Recently, the IAEA-TECDOC-1616, (2009), IAEA, (2010) made an extensive review on the radionuclides concentration ratios (transfer factor) for various foods including rice. Uchida et al. (2009) have published a review paper on the radionuclide transfer factors for rice and mentioned that there are inadequate studies in this aspect. However, a general survey of literature reveals that Tsukada et al. (2002), Sasaki et al. (2002) and Uchida et al. (2007) from Japan; Choi et al. (2005, 2011a, 2011b, 1999) and Keum et al. (2007) from South Korea; Wang et al. (1998) from China; Quang et al. (2003) from Vietnam; and Saeed et al. (2012) from Malaysia conducted similar studies to obtain a radiological mapping on rice grown in south east Asian region. Since studies on the uptake of radionuclides via intake of it are still limited in the area under investigation, the present study would therefore make a valuable contribution to the establishment of a standard database of the natural radioactivity of rice in peninsular Malaysia. It can also serve

as a reference for an assessment of radiological impact via rice consumption by the inhabitants of Malaysia and bordering regions.

2. Materials and Methods

2.1. Study area

Two areas in north Malaysia Kampung Sakan, Kedah and Kampung Permatang Tok Labu, Pulau Pinang) and one area in west Malaysia (Sungai Besar, Selangor) (Fig. 1) were chosen for this study because most of the rice in Peninsular Malaysia grown in these areas. Kedah (Latitude: 6° 23' N, Longitude: 100° 45' E) is tropical monsoon, with a uniform temperature between 21 °C to 32 °C throughout the year. It is generally dry and warm from January to April, and wet from May to December. Humidity is consistently high in the lowlands, averaging 82–86% per annum. Kedah's average annual rainfall lies between 2,032 mm to 2540 mm. Pulau Pinang (Latitude 5.5523' N, Longitude. 100.38' E) is not a tropical island like other parts of Malaysia with temperature between 23 °C to 32 °C and relative humidity 0–50% throughout the year. Pinang has little rain except during the Southwest Monsoon from April to September. An average annual rainfall of this region is 2670 mm. The climate is very much dictated by the surrounding sea and the wind system. Selangor (Latitude: 3°20'N, Longitude: 101°30'E) is tropical monsoon. Temperatures generally range from 27 °C to 35 °C during the day time throughout the year with relative humidity 76%–86%. Annual rainfall measures at 3,218 mm. Being located so close to the equator, the study areas do not have distinct seasons such as spring, summer, autumn or winter. The seasonal variation in solar radiation is low, resulting in an annual difference in day length of only 2 min along the equator and 49 min in northern regions. In consequence, there is a year round day length of 12.5 h (Nieuwolt, 1982). The region is distributed by steep hills with tropical forest of natural habitat and grows a variety of plants that play a vital role in the environmental transport of radionuclides. About 72 percent of Malaysian soils are Ultisols and Oxisols, which are acidic and highly weathered (IBSRAM, 1985). Phosphate fertilizers are suggested to use due to soil fixation. This problem is dealt with through the biennial addition of about 2 to 4 t/ha of limestone (Shamsuddin et al., 1992). The soil of the study areas mainly clayey-silty textured (45–60% clay, 25–35% silt and 15–20% sand) (Table 1).

2.2. Cultivation practices of rice by the farmers

Cultivation patterns play a vital role on the uptake of radionuclides from soil-to-plant (Uchida et al., 2009). Different varieties of rice are grown in the study regions and basic growing practices of them are more or less same in everywhere. There are two types of land used for rice cultivation in Malaysia namely, wet-land and dry-land. There have been several variations in the methods of rice plantation. It ranged from seed dibbling, mostly in upland conditions, to the conventional transplanting in the low land/wet rice cultivation (WRC) or double nursery plantation, wherein seeds were transferred from one nursery to another before being planted out in the main field. Direct seeding has been seasonally practiced over the years both in the main season (MS) and the off season (OS). The farmer does not practice the crop rotation and rice grown in the same field for years.

Farmers follow the traditional/mechanized practices of plowing the land, conditioning, planting, application of various inputs and harvesting. In Peninsular Malaysia, the uses of machines leading to the rice production are as follows: in land preparation (100%), in seed sowing (71.3%), in manuring (70.6%), in chemical
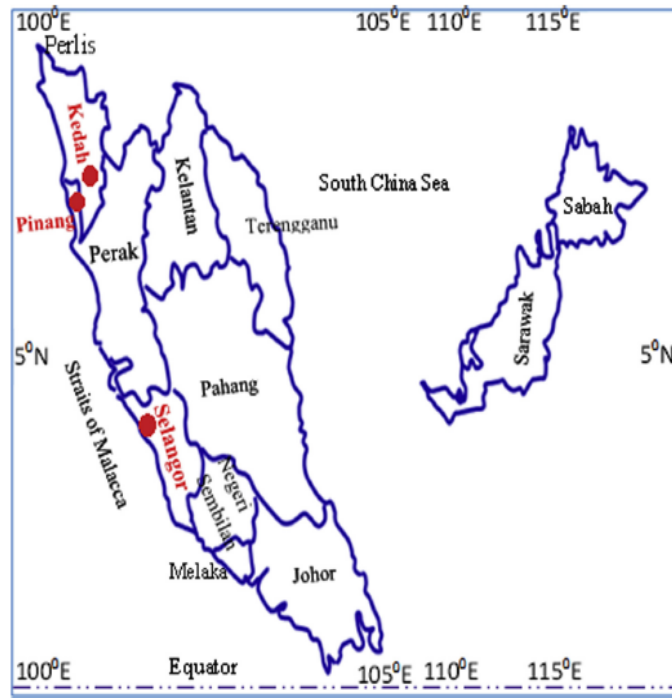
Fig. 1. Map of Malaysia showing the different sampling sites.

Table 1
Physical and chemical properties of soil.

Characteristics	Sungai Besar	Kampung Permatang TokLabu	Kampung Sakan
Texture	silty clay (45% sand, 35% silt and 20% clay)	clayey silt (53% clay, 30% silt and 17% sand)	clayey silt (60% clay, 25% silt and 15% sand)
pH	5.51	5.68	6.76
CEC (meq/100 g)	12.6	15.4	20.7
Total Ca content (mg/kg)	355	2500	1920
Total K content (mg/kg)	72	185	345
Organic matter (%)	11.97	18..47	30.20

spraying (82.0%) and in paddy harvesting (100%) (DOA, Paddy Production Survey Report Malaysia 2012). The area as a whole is composed of 80 percent low land and 20 percent high land (Lim and Chan, 1993). Irrigation has to be given by growers/farmers in the off season using the local water resources whereas in the main wet season drainage has to be ensured at times whenever there is heavy downpour. According to the Malaysian Agricultural Research and Development Institute (MARDI), the fertilizer given by the government to the farmer contains nitrogen (N), phosphorus (P) and potassium (K) (called NPK fertilizer) to enhance the nutrients for crops, improve soil fertility and productivity of plant (MARDI). DOA recommended fertilizer rates for paddy is 90 kg/ha of N, 35 kg/ha of P_2O_5 and 25 kg/ha of K_2O (Fertilizer use by crop in Malaysia, 2004). Sometimes farmers used fertilizer 2nd time at the age of 2 months after the rice plant plantation. However, a slight variation in the kind of fertilizers and the amount of fertilization was observed among the growers. For weed and pest control, farmers use herbicide and insecticide.

2.3. Sample collection and processing

Rice and soil samples were collected from paddy fields in the aforementioned study areas. The fields of rice collection assessing about 40 m^2 were parted into small sub-units of 8 m^2 each. During the harvesting time fully matured rice plants were collected randomly from each sub-unit following X-shape, at each corner and at the middle of the selected fields in each study location. The rice grain was then separated from the non-edible part of the plant and sieved to remove the unnecessary things in the samples. Thereafter, the entire sample in a single sub-unit were thoroughly mixed, dried, sieved and homogenized to get a composite sample representative of that unit. About 1 kg of rice grain was taken from this composite sample, dried in oven below 80°C for 24 h to remove moisture and attain constant dry weight. The samples were then crushed into fine powder and homogenized by filtering through 1 mm sieve and 500 gm of rice powder sample from each sampling point was shifted to marinelli beaker for further analysis and their average activity has been reported as one sample.

Soil was sampled in the same way from each sub-unit corresponds to the rooting zone of rice plant including surface layer and to a depth of 5 cm as rice has an adventitious root structure that feasts in the upper layer of the soil and does not go deep. The irrelevant materials like pieces of stone, gravel, leaf and roots were removed from the sample and weighed after drying for several days at room temperature until a constant weight was reached thereafter, mixed to provide a representative sample for that unit. About 1 kg of soil was taken from this representative sample, dried in an oven at 100 °C for 24 h to attain constant dry weight. Then the samples were crushed into fine powder and homogenized by sieving through 1 mm sieve and 500 gm of soil sample from each sampling point was shifted to marinelli beaker for gamma-ray analysis and their average activity has been stated as one sample.

An effort was made to ensure adequate collection of representative samples in the study area. All the samples were stored for a period of 4 – 5 weeks at room temperature to reach secular equilibrium of ^{226}Ra, ^{232}Th with their progenies prior to the radiometric analysis with a gamma-ray detector (Ghose et al., 2012).

2.4. Gamma-ray spectrometry with NaI(Tl) detector

Activity concentrations of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the studied samples were measured using a conventional γ-ray spectrometer. A NaI(Tl)-detector (cylindrical shape single crystal with a dimension of 7.62 cm × 7.62 cm^2) coupled to a Gamma Vision 5.0 (EG&G Ortec) multi-channel analyzing software was used to record the gamma-ray spectra. The NaI crystal is kept in a thin-walled aluminum can (0.0127 cm wall thickness) to reduce absorption of low energy photons to prevent excessive Compton scattering from the packaging material. A 1.18 g/cm^2 Be (beryllium window) interlaced between the source and detector to eliminate beta particles from the detector. The detector was shielded by a graded lead cylindrical shield of about 27 cm diameter, 82.5 cm height and 6 cm thickness to reduce the background gamma radiation present at the laboratory site. The detector was operated with a constant voltage of 850 V. The detector was calibrated using the standard gamma-ray source of ^{60}Co with known peak at 1.17 MeV and 1.33 MeV and using the ^{137}Cs with peak at 661 keV. The energy resolution of the detector was 7.5% for the 661 keV of ^{137}Cs. The efficiency of the detector was determined using multi-nuclide γ-ray standard source (having homogeneously distributed activity with the same volume and shape as the Merinelli beakers that contain the sample so that the detection geometry remained the same) to ensure the reliability and accuracy of the energy range in the measurement of respective radionuclides. The samples were counted for a sufficiently long time (86,000 s) to decrease the counting error, and background

counts for the same counting time were deducted to obtain the net activity. The activity concentrations of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides were determined from their respective characteristic γ-lines, as shown in Table 2. Only strong and independent characteristic γ-lines (bold in Table 2) were used to determine the net activity concentrations in order to reduce the uncertainties in activity determination.

2.5. Calculations of radioactivity

The activity concentration of radionuclides in the samples was calculated from the obtained net counts after the deduction of background counts using a relation given as (Khandaker et al., 2012):

$$C = \frac{N \times 1000}{\varepsilon \times I_\gamma \times t \times M_s} \tag{1}$$

C = activity concentration of the radionuclide in the sample given in Bq kg^{-1}, N = net counts under corresponding photo-peak, ε = detection efficiency corresponding to specific gamma-ray, I_γ = absolute transition probability of the specific gamma-ray, M_s = mass of the sample in gram, T = counting time in seconds.

2.6. Evolution of transfer factor (TF)

The transfer mechanism of radionuclides is represented by a parameter called *TF*, which is widely used to describe the soil-to-plant transfer of radionuclide through the plant roots. Concentration of a nuclide in a plant or plant part, C_i^p (Bq kg^{-1}, dry weight), is linearly related to its concentration in soil within the rooting zone, C_i^s (Bq kg^{-1}, dry weight). Soil-to-plant transfer factor for rice was estimated from the measured activity concentrations of radionuclides using the following relationship (IAEA, 2010; FAO/IAEA/IUR protocol, 1998);

$$TF = \frac{C_i^p \, (\text{Bq kg}^{-1}, dry \; weight)}{C_i^s \, (\text{Bq kg}^{-1}, dry \; weight)} \tag{2}$$

2.7. Statistical analysis

The data were expressed as the mean ± standard deviation and subject to statistical analysis using SPSS 22 software (IBM Corporation, Armonk, NY, USA). Descriptive statistical analysis was performed to determine the arithmetic mean (AM), geometric mean (GM), standard deviation (SD) and geometric standard deviation (GSD). Data of activity concentrations in soil and rice, Transfer factor from soil-to-rice, daily intake and annual effective dose in rice samples among the three studied locations were compared

Table 2

Decay data of the detected radionuclides of interest. γ-line in bold were used in activity determination. (http://www.nndc.bnl.gov/nudat2/).

Radionuclides of interest	Detected radionuclides	Half-life, $T_{1/2}$	Decay mode (%)	γ-ray energy, E_γ (keV)	γ-ray intensity, I_γ (%)
^{226}Ra	^{214}Pb	26.8 m	β$^-$ (100)	295.2228	18.42
				351.9321	**35.60**
	^{214}Bi	19.9 m	α (0.021);	**609.320**	**45.49**
			β$^-$ (99.979)	1120.294	14.92
				1764.491	15.30
^{232}Th	^{212}Pb	10.64 h	β$^-$ (100)	**238.632**	**43.6**
	^{208}Tl	3.053 m	β$^-$ (100)	510.77	22.60
				583.187	**85.0**
				860.557	12.50
	^{228}Ac	6.15 h	β$^-$ (100)	338.320	11.27
				911.204	**25.8**
				968.971	15.8
^{40}K	^{40}K	1.248 10^9 y	EC(10.719); β$^+$ (0.001);	**1460.822**	**10.66**
			β$^-$ (89.28)		

employing one-way analysis of variance (ANOVA). Tukey HSD Post Hoc Test was applied to determine statistically significant differences among individual means, and the p value for significance was determined. The statistical significance was set at $p < 0.05$.

3. Results and discussion

3.1. Radionuclide activity concentrations

Activity concentrations of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides in soil and rice samples collected from all of the studied locations are presented in Table 3. Activity concentrations showed in the Table 3 were on a dry weight based. As presented in Table 3, ^{40}K shows highest concentration for all location in soil samples. Significantly highest ($p < 0.05$) activity of ^{40}K was found in Sungai Besar as 114.6 ± 6.6 Bq kg^{-1}. Meanwhile, Kampung Sakan shows slightly lower activity (76.5 ± 5.7 Bq kg^{-1}) than Sungai Besar and Kampung Permatang Tok Labu, (90.4 ± 9.8 Bq kg^{-1}). The accumulation of ^{40}K in most study location may be affected by several factors such as Cation Exchange Capacity (CEC) in the soil, pH of the soil and the type of soil.

In fact, all soil samples in different study locations share same property. They are in category of clay minerals. Because of the structure and chemical composition, the clay minerals usually bear a negative charge. The negative charge on clay is balanced by the charge on cations (Wild, 1993). This process called CEC, where protons remove from the solution. Potassium, K$^+$ is known as one of the basic cations. The capacity of soil to hold the cations usually increase the presence of ^{40}K. Besides this, the dissociation of proton involve in CEC process also depend on pH. Note that pH bear a negative charge which leads to an increase of cations in the soil (Wild, 1993). From Tables 1 and 3, we have seen that the activity concentration of the soil decreases with the increase of soil pH.

^{226}Ra shows highest activity concentrations (9.1 ± 1.8 Bq kg^{-1}) in Sg Besar soil and lowest (7.2 ± 2.0 Bq kg^{-1}) in Kampung Sakan. However, statistical analysis (ANOVA) shows that there is no significant variation ($p > 0.05$) of concentrations of ^{226}Ra in soil among the studied locations.

The activity concentration of ^{232}Th in the soil was found higher than that of ^{226}Ra throughout all the location with a range of 11.6 ± 1.9 – 20.6 ± 3.1 Bq kg^{-1}. The activity concentration of ^{232}Th shows the highest value as 20.6 ± 3.1 in Sg Besar. The erosion process of ^{232}Th largely occurred and it adsorbed in the soil on the spot, whereas ^{226}Ra is easily extracted and migrates with soil water. This may be one of the reasons for a higher accumulation of ^{232}Th than ^{226}Ra (Wild, 1993).

Similar to soil, the rice samples also showed significant amount of ^{40}K activity than the other radionuclides ($p < 0.001$). The average activity of ^{40}K in rice sample was found as 59.9 ± 6.0 Bq kg^{-1}, 89.3 ± 6.2 Bq kg^{-1} and 92.2 ± 5.4 Bq kg^{-1} for Sungai Besar, Kampung Permatang Tok Labu and Kampung Sakan, respectively. In the Sungai Besar, activity concentration (59.9 ± 6.0 Bq kg^{-1}) of ^{40}K in rice sample was measured approximately half that of the soil sample (114.6 ± 6.6 Bq kg^{-1}). Meanwhile the activity of rice at Kampung Sakan is little bit exceed the activity of soil. Furthermore, the activity concentration of rice at Kampung Permatang Tok Labu shows almost same activity in soil samples.

The average activity concentrations of ^{226}Ra in the rice sample in Sungai Besar, Kampung Permatang Tok Labu and Kampung Sakan was obtained as 1.5 ± 0.4, 2.8 ± 0.7 and 2.3 ± 0.5 Bq kg^{-1}, respectively. Significant ($p < 0.05$) difference of ^{226}Ra concentrations was found between Sungai Besar and Kampung Permatang Tok Labu by ANOVA analysis. The concentration of radium in the rice depends on the radium content of soil, its availability to the rice plant and the metabolic features of the rice plant species. The radioactivity concentration for ^{232}Th slightly higher (statistically insignificant ($p > 0.05$)) than ^{226}Ra in all soil samples so as to rice samples in all locations (Table 3). This may be attributed that thorium has a high affinity for the regular exchange sites of the soil and paddy plant absorbs thorium at a high rate from soil.

Table 3 reveals some significant differences in the activity concentrations of the studied radionuclides in the soil and rice samples as well as the studied locations. This may be due to the dissimilarity in locations and the physical properties of the soil and the rice, climatic condition and amount, and the kind of fertilizers used in that soil for enhanced the crop production (Karunakara et al., 2013). The mean concentration for ^{226}Ra, ^{232}Th and ^{40}K measured in Malaysia recommended by UNSCEAR (2000) are 67 Bq kg^{-1}, 82 Bq kg^{-1} and 310 Bq kg^{-1}, respectively (Alias et al., 2005). The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K found in this study are below the UNSCEAR (2000) recommended ones.

Table 3
Activity concentration (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K in soil and rice and their corresponding transfer factors (TFs).

Locations	Sample type	Parameters	Radioactivity concentration (Bq kg^{-1})			Transfer factors (TFs)		
			^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
Sungai Besar	Soil (6)	Range	7.5 – 11.3	17.3 – 25.2	106.0 – 124.8			
		AM ± SD	9.1 ± 1.8	20.6 ± 3.1	114.6 ± 6.6			
		GM/GSD	9.0/1.2	20.4/1.2	114.5/1.1			
	Rice(6)	Range	0.98 – 2.3	1.9 – 5.6	49.7 – 66.6	0.10 – 0.30	0.08 – 0.28	0.47 – 0.61
		AM ± SD	1.5 ± 0.4	3.6 ± 1.4	59.9 ± 6.0	0.17	0.18	0.52
		GM/GSD	1.4/1.3	3.3/1.5	59.6/1.1	0.16/1.5	0.17/1.7	0.52/1.1
Kampung Permatang Tok Labu	Soil (5)	Range	6.5 – 11.6	12.8 – 19.6	78.5 – 100.5			
		AM ± SD	8.9 ± 2.2	15.9 ± 2.8	90.4 ± 9.8			
		GM/GSD	8.7/1.3	15.7/1.2	90.0/1.1			
	Rice(5)	Range	1.9 – 3.8	5.0 – 11.4	82.9 – 97.8	0.24 – 0.43	0.33 – 0.59	0.83 – 1.24
		AM ± SD	2.8 ± 0.7	7.5 ± 2.7	89.3 ± 6.2	0.32	0.47	1.00
		GM/GSD	2.7/1.3	7.2/1.4	89.1/1.1	0.31/1.3	0.46/1.3	0.99/1.2
Kampung Sakan	Soil (3)	Range	5.1 – 9.2	9.6 – 13.5	70.5 – 81.7			
		AM ± SD	7.2 ± 2.0	11.6 ± 1.9	76.5 ± 5.7			
		GM/GSD	7.0/1.3	11.5/1.2	76.3/1.3			
	Rice(3)	Range	1.6 – 2.8	4.7 – 6.4	87.0 – 97.9	0.32 – 0.37	0.39 – 0.55	1.12 – 1.27
		AM ± SD	2.3 ± 0.5	5.5 ± 0.9	92.2 ± 5.4	0.32	0.48	1.21
		GM/GSD	2.2/1.3	5.4/1.3	92.1/1.2	0.32/1.2	0.47/1.3	1.21/1.1
UNSCEAR-2000	Rice		67	82	310			

The values in parenthesis indicate the number of samples analyzed. AM denotes arithmetic mean, SD denotes standard deviation, GM denotes geometric mean and GSD stands for geometric standard deviation.

Table 4
Comparison of Soil-to-rice TF values of radionuclides observed in the current study with those reported for some other regions.

Country (Rice type)	Transfer factor					References
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	^{90}Sr	
Malaysia (Brown rice)	0.26	0.37	0.91	–	–	Present study
Malaysia (not mentioned)	0.20[a]	0.15	1.43	–	–	Saeed et al., 2012
South West India (not mentioned)	8.8×10^{-2}	14.2×10^{-2}	6.3×10^{-2}	–	–	Shanthi et al., 2012
Kaiga India (not mentioned)	$<5.0 \times 10^{-2}$	–	1.5	0.21	–	Karunakara et al., 2013
Japan (Hulled rice)	4.7×10^{-3}	9.5×10^{-5}	–	–	–	Sasaki et al., 2002
Japan (Polished rice)	–	–	–	0.0016	–	Tsukada et al., 2002
Japan (Brown rice)	[a]5.6×10^{-5}	1.0×10^{-4}	[b]2.2×10^{-1}	1.2×10^{-2}	–	Uchida et al., 2007
Japan (Brown rice)	9.6×10^{-5} – 8.6×10^{-3}	1.4×10^{-4}	2.2×10^{-1}	1.2×10^{-2}	4.6×10^{-2}	Uchida et al., 2009
Korea (Hulled rice)	–	–	–	1.3×10^{-2} – 4.1×10^{-2}	–	Keum et al., 2007
Korea (Brown rice)	–	–	–	$1.5 \times 10^{-1} \pm 1.5 \times 10^{-2}$	$2.5 \times 10^{-2} \pm 1.4 \times 10^{-3}$	Choi et al., 2011a
China (Brown rice)	–	–	–	0.14 ± 0.02	0.086 ± 0.017	Wang et al., 1998
IAEA reported value	0.00025 – 0.0029	0.000026 – 0.00083	0.087 – 0.78	0.15 – 0.0014	0.11 – 0.028	IAEA, 2010, IAEA–TEC-DOC, 2009
(Brown rice)	(0.00057)	(0.00014)	(0.22)	(0.017)	(0.06)	

[a] TF of uranium,
[b] TF of stable K,

3.2. Radionuclides uptake/transfer to rice

Generally, TF for grains is calculated based on its fresh weight (moisture content ∼ 1–2%) to find a representative transfer factor (Pulhani et al., 2005). From the results of activity concentrations of radionuclides in the soil and rice (Table 3), the soil-to-rice transfer factors for all studied radionuclides were calculated using Eq. (2), and presented in the Table 3. Table 4 shows a comparative study of the present results with the earlier literature data and the IAEA-TECDOC-1616, (2009), IAEA, (2010) compiled values. TFs of radionuclides are usually distributed lognormally, so the TF values predicted here are geometric means (GMs).

3.2.1. Soil-to-rice transfer factor (TF) of ^{226}Ra

TF of ^{226}Ra in rice grains grown in Kampung Sakan, Kedah (0.32) shows two times higher than that of Sungai Besar, Selangor (0.16) and very close to that of Kampung Permatang Tok Labu, Pulau Pinang (0.31). These TF values for radium to rice were observed far away from the reported minimum and maximum values of 0.00025 and 0.0029 respectively, with a mean value of 0.00057 compiled by the IAEA for clay soil-to-rice (IAEA, 2010). Though radium concentration in the Sungai Besar soil is little bit higher than Kampung Sakan and Kampung Permatang Tok Labu soil, but the transfer factors for radium to rice grains for both of these fields were two times higher than the Sungai Besar field (statistically significant ($p < 0.05$)). Uranium/Radium is expected to be present as soluble complex of anions like phosphates, sulfates and carbonates in calcareous alluvial soil of Kampung Sakan and Kampung Permatang Tok Labu and hence more available for uptake (Pulhani et al., 2005; Sheppard et al., 1989).

3.2.2. Soil-to-rice transfer factor of ^{232}Th

Similarly the TF for ^{232}Th in rice grains grown in Kampung Sakan and Kampung Permatang Tok Labu (0.47 and 0.46, respectively) was found 2.7 times higher (statistically significant ($p < 0.05$)) than that of the Sungai Besar (0.17). The observed values of TFs were beyond the range (0.000026–0.00083) reported by the IAEA with a mean value 0.00014 for clay soil-to-rice for thorium (IAEA, 2010).

3.2.3. Soil-to-rice transfer factor of ^{40}K

Soil-to-rice TFs for ^{40}K was found higher than the other radionuclides in all study locations which indicate the fundamental nutrient status of potassium for the plants. The high TFs of ^{40}K were considered due to the fact that potassium is important in fertilizing the crop, and also plays a vital role in the ability of the plant adaptation to environmental pressures. However, potassium relics in homeostatic symmetry in the plant and is easily adapted by the plants (Pulhani et al., 2005). Therefore, as compared to uranium and thorium, potassium shows the significantly highest transfer factor ($p < 0.001$). The investigated values of TFs for potassium were not very far beyond the range reported by the IAEA, 0.087–0.78 (IAEA-TECDOC-1616, (2009), IAEA, (2010)).

The TFs for ^{226}Ra and ^{232}Th in this study are found higher when compared with the literature values (Table 4). However, these TF values were comparable with the values reported by Saeed et al. (2012) in Malaysia. Similarly, TF values for ^{40}K obtained in this study for rice were in the same order of magnitude when compared to some literature values. The soil texture in the present studied locations was mainly clayey-silt, whereas IAEA reported TFs for ^{226}Ra and ^{232}Th were based on the clay soil. The hydrological conditions within the soil, soil fertility, the duration of the growing period, cultivation process may not same to our studied farmland and those countries farmland from where IAEA obtained the data.

3.2.4. Effect of soil properties on TFs

According to the concentration ratio principles, plant radionuclide concentration should reflect soil concentration. However, this could always not be true because of sorption on soil make the radionuclide is less available for uptake. Radionuclide belongs to similar chemical properties may be selectively absorbed, some may be omitted. This study shows that accumulation of radionuclides in farmlands little bit varies for different soils. The differences in TFs to farmlands for dissimilar soils may be due to the soil properties such as mineralogical and granulometric composition, organic matter content, pH, hydrological conditions within the soil. The biological variability inherent in plants and distinctions between different varieties and species are also a likely source of much of the anomaly in transfer factors. Soil potency, crop farming technologies, duration of the growing period and characteristics of the root dissemination in soil also influenced on TFs. The above parameters may change soil properties or lead to redistribution of radionuclides in the root zone, and consequently change of radionuclides uptake in crops (Asaduzzaman et al., 2014).

The types of soil and cultivation information are important factors because behaviors of radionuclides depend on soil properties and sampling conditions (IAEA-TECDOC-1616, 2009). The physical and chemical properties of the studied firm land soil are presented in Table 1. Soil-to-plant transfer of naturally occurring radionuclides is largely affected by the soil physio-chemical properties such as cation exchange capacity (CEC), potassium (K), calcium (Ca), organic matter content etc. (Pulhari et al., 2004; Shanthi et al., 2012). The pH values (5.51 – 6.76) of soil in all the studied area indicate that the soil is acidic in nature. Generally, acidic soil uptakes more radionuclides than alkaline soil specially ^{226}Ra cannot be avoided in acidic soil (Asaduzzaman et al., 2014). In the present study, variation of TFs for ^{226}Ra, ^{232}Th and ^{40}K was increased with the increase of soil pH. It is seen from Table 1 and 3 that CEC regulates the transfer of radionuclide's from soil-to-rice (e.g., TFs increases with the increase of CEC). Calcium and potassium are essential macronutrients for plants maintained in homeostatic equilibrium and in doing so the plants regulate the uptake of nonessential elements like radioelements from soil (Pulhani et al., 2005). The availability of calcium and potassium in soil for uptake will affect the uranium, thorium and radium content of the plant (Chen et al., 2005). In the soil, radium behaves very much like its homologs: calcium that determines the rate at which radium will be absorbed by plants. It was observed from Tables 1 and 3 that Ca content varies in different study locations and found little bit linear correlation with the TF of ^{226}Ra. Conversely, linear relation was observed for K content in soil and its transfer to rice. The organic matter content of the soil varies between 11.97 – 30.20% among the studied regions. Positive relation was observed between organic matter content and TF of radionuclides.

Therefore, it may be concluded that the combined effects of the soil properties, individual chemical properties of the radionuclides (Chen et al., 2005), soil amendment such as NPK fertilizers (Joshy et al., 2011; Shanthi et al., 2012), use of insecticides and herbicides, plant species, climate condition etc. may have a larger influence on the uptake of radionuclides from soil-to-rice.

3.3. Radiological impact

3.3.1. Daily intake

The daily intake of radioactivities is considered to an accumulation of ^{226}Ra, ^{232}Th and ^{40}K into the human body by the consumption of rice grains. Considering the consumption of rice per capita per year for Malaysia is at 95.9 kg in 2011/12 (GAIN Report MY3002, Malaysia, 2013), daily intake of radionuclides by individuals due to the consumption of rice has been estimated by the following formula (Khandaker et al., 2013):

$$D_{int} = \frac{A_c \times A_{ig}}{Y_d} \qquad (3)$$

Where, D_{int} is the daily intake of radionuclides (Bq d^{-1}) by individuals, A_c is the activity concentration of radionuclides (Bq kg^{-1}), A_{ig} is the per capita per year consumption of grain, Y_d is the days in a year. The values of daily intake per capita for rice grains are presented in Table 5. The daily intake of ^{40}K was found significantly (p < 0.001) higher than the other radionuclides in all study locations.

3.3.2. Consumption dose to human

The annual effective dose to an individual due to an intake of ^{226}Ra, ^{232}Th and ^{40}K radionuclides from the consumption of rice has been computed by the following relation (Khandaker et al., 2013) and presented in Table 5:

$$E_{eff} = A_c \times A_{ig} \times D_{cf} \qquad (4)$$

Where, E_{eff} is the annual effective dose (μSv y^{-1}) to an individual owing to an ingestion of radionuclides, A_c is the average activity concentration of radionuclides (Bq kg^{-1}), A_{if} is the annual intake of rice (kg y^{-1}), per capita consumption of rice in Malaysia in 2011/12 was 95.9 kg y^{-1} (GAIN Report MY3002, Malaysia, 2013) and D_{cf} is the ingestion dose conversion factor for the radionuclides of interest (2.8×10^{-7} Sv Bq^{-1} for ^{226}Ra, 2.3×10^{-7} Sv Bq^{-1} for ^{232}Th and 6.2×10^{-9} Sv Bq^{-1} for ^{40}K) (IAEA, 2011). The total ingestion dose (committed) due to the intake of radionuclides via consumption of rice grain was calculated using the following formula (5) and present in Table 5:

$$E_t = \sum \left[\left(A_{cr}^f \times C_y^f \right) \right] D_{cf}^r \qquad (5)$$

Where f denotes a food group, the coefficients A_{cr}^f and C_y^f denote the average activity concentration of radionuclides (Bq kg^{-1}) and consumption rate per year (kg), respectively and D_{cf}^r is the dose coefficient for an intake by the ingestion of radionuclide, r (Sv Bq^{-1}). The annual effective dose for rice from the intake of thorium (^{232}Th) was found significantly (p < 0.05) higher than other detected radionuclides in all studied locations. Average worldwide effective dose from the ingestion of uranium and thorium series is 120 μSv y^{-1}, for ^{40}K is 170 μSv y^{-1}, and the total is of 290 μSv y^{-1} amassed by UNSCEAR, 2000). The annual effective dose for all detected radionuclides in all study locations are within the UNSCEAR (2000) annex-b reported limits except ^{232}Th of Kampung Permatang Tok Labu which exceeds the world average values. The highest value of total effective dose was found in Kampung Permatang Tok Labu and the lowest in Sungai Besar. The total effective

Table 5
Daily intake of ^{226}Ra, ^{232}Th and ^{40}K, annual committed effective dose, estimated total effective dose and excess lifetime cancer risk due to the consumption of rice.

Sampling location	Parameters	Daily intake of radionuclides D_{int} (Bq d^{-1})			Annual Committed effective dose, E_{ef} (μSvy^{-1})			Total effective dose E_t (μSvy^{-1})	Lifetime cancer risk (ELCR)10^{-3}		
		^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K		^{226}Ra	^{232}Th	^{40}K
Sungai Besar	Range	0.26 – 0.60	0.50 – 1.5	13.1 – 17.5	26.3 – 61.0	41.7 – 123.1	29.5 – 39.6	105.4 – 196.1			
	AM ± SD	0.39 ± 0.1	0.93 ± 0.4	15.7 ± 1.6	39.5 ± 11.6	78.3 ± 31.8	35.6 ± 3.5	153.4 ± 33.2	1.0×10^{-4}	6.2×10^{-5}	2.5×10^{-4}
	GM	0.37	0.87	15.7	38.2	73.0	35.5	150.2			
Kampung Permatang Tok Labu	Range	0.49 – 0.99	1.3 – 3.0	21.8 – 25.7	50.2 – 101.2	109.6 – 252.3	49.3 – 58.1	247.2 – 378.2			
	AM ± SD	0.73 ± 0.2	2.0 ± 0.7	23.5 ± 1.6	74.9 ± 18.4	166.3 ± 59.0	53.1 ± 3.7	294.3 ± 49.8	1.9×10^{-4}	1.3×10^{-4}	3.7×10^{-4}
	GM	0.71	1.9	23.4	73.0	158.2	53.0	291.1			
Kampung Sakan	Range	0.43 – 0.70	1.2 – 1.7	22.9 – 25.7	44.0 – 71.7	104.1 – 141.8	51.8 – 58.2	214.5 – 266.1			
	AM ± SD	0.59 ± .01	1.4 ± 0.2	24.2 ± 1.4	60.7 ± 14.6	120.7 ± 19.3	54.8 ± 3.2	236.1 ± 26.8	1.9×10^{-4}	9.5×10^{-5}	3.9×10^{-4}
	GM	0.58	1.4	24.2	59.3	119.7	54.7	235.1			
World average (UNSCEAR-2000 compiled value)					120	120	170	290	$\sim 10^{-3}$		

dose for the consumption of rice was also within the range of world average value compiled by UNSCEAR (2000) in all study areas.

3.3.3. Excess lifetime cancer risk

Now a day's cancer is called the life threat diseases and the percentage of this disease increases all over the world including Malaysia due to various reasons. One of the reasons is the radiation effect on biological cell. According to National Cancer Society Malaysia (http://cancer.org.my/learn-about-cancer/about-cancer), there are about 90–100,000 people in Malaysia living with cancer at any average time. Increasing population and longer life spans contributes to rise of cancer. Less than 10% of cancers happen in children compared to over 50% in men and 35% in women aged 50 and above. Although considered the 3rd leading cause of premature death in Malaysia, only 30–40% of all deaths from cancer are medically certified, meaning there is no exact figure of people dying from cancer. That is why an effort was made to assess the excess lifetime cancer risk (ELCR) due to the consumption of rice using the procedure proposed by US EPA (1999). The following equation (Patra et al., 2013) was used to calculate the mortality cancer risk and presented in Table 5.

$$ELCR = A_{ir} \times A_{ls} \times R_c \qquad (6)$$

Where, $ELCR$, A_{ir}, A_{ls} and R_c are the lifetime cancer risk, annual intake of radionuclide (Bq), average span of life (74 y) and mortality risk coefficient (Bq^{-1}) for the ingestion of food, respectively. The ingestion mortality cancer risk coefficients in (Bq^{-1}) are 9.56×10^{-9} for ^{226}Ra, 2.45×10^{-9} for ^{232}Th and 5.89×10^{-10} for ^{40}K (US EPA, 1999). The average ELCR was varied from 1.0×10^{-4} to 1.9×10^{-4} for ^{226}Ra, 6.2×10^{-5} to 1.3×10^{-4} for ^{232}Th and 2.5×10^{-4} to 3.9×10^{-4} for ^{40}K, which were low compared with the acceptable ELCR limit of 10^{-3} for radiological risk in general (Patra et al., 2013).

4. Conclusions

This report could be considered as the first systematic study on radionuclide transfer factors for soil-to-rice and ingestion dose to humans based on the natural field conditions in peninsular Malaysia. Measured activity concentration for ^{226}Ra, ^{232}Th and ^{40}K radionuclides in soil samples varied from 7.2 ± 2.0 to 9.1 ± 1.8 Bq kg^{-1}, 11.6 ± 1.9 to 20.6 ± 3.1Bq kg^{-1} and 76.5 ± 5.7 to 114.6 ± 6.6 Bq kg^{-1}, respectively. Similarly in rice samples, the mean concentrations of ^{226}Ra, ^{232}Th and ^{40}K were found in the range of 1.5 ± 0.4 to 2.8 ± 0.7 Bq kg^{-1}, 3.6 ± 1.4 to 7.5 ± 2.7 Bq kg^{-1} and 59.9 ± 6.0 to 92.2 ± 5.4 Bq kg^{-1}, respectively. Soil-to-rice TFs for ^{226}Ra, ^{232}Th and ^{40}K varied from 0.16 to 0.32, 0.17 to 0.47 and 0.52 to 1.21, respectively. The calculated values of the TFs for the mentioned radionuclides show much higher than those compiled by the IAEA (2010).

The estimated daily intakes (mean) due to the consumption of rice are 0.57 ± 0.2 Bq for ^{226}Ra, 1.5 ± 0.5 Bq for ^{232}Th and 21.2 ± 4.7 Bq for ^{40}K. Total annual effective dose via the consumption of rice grains were estimated in the range of 153.4 ± 33.2 to 294.3 ± 49.8(μSv y^{-1}) among the studied locations, which lies within the range of global average yearly ingestion dose. Lifetime cancer (mortality) risk was found below the acceptable limit of 10^{-3} for radiological risk. Therefore, this study indicates that the radionuclides intake from the consumption of rice grain yet poses no threat to public health. The outcomes of this work will support in establishing a baseline database of TF's and radioactivity exposure to the general community from consumption of rice or other foodstuff.

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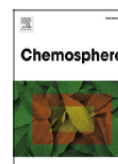
3.2.2 Published paper 2

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Evaluation of radionuclides transfer from soil-to-edible flora and estimation of radiological dose to the Malaysian populace

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HIGHLIGHTS

- Assessment of radionuclides transfer from soil-to-edible flora in Malaysia.
- The estimated TFs for [226,228]Ra show greater values than the literature data for vegetables.
- The Highland farms present greater concentrations than the ground farms.
- Committed effective dose and life-time cancer risk found below the permissible limit by UNSCEAR.

ARTICLE INFO

ABSTRACT

Malaysia, a rapidly growing industrial country, is susceptible to pollution via large-scale industrial engagements and associated human activities. One particular concern is the potential impact upon the quality of locally resourced vegetables, foodstuffs that contain important nutrients necessary for good health, forming an essential part of the Malaysian diet. As a part of this, it is of importance for there to be accurate knowledge of radioactive material uptake in these vegetables, not least in respect of any public health detriment. Herein, using HPGe γ-ray spectrometry, quantification has been performed of naturally occurring radionuclides in common edible vegetables and their associated soils. From samples analyses, the soil activity concentration ranges (in units of Bq/kg) for ^{226}Ra, ^{232}Th and ^{40}K were respectively 1.33 -30.90, $0.48-26.80$, $7.99-136.5$ while in vegetable samples the ranges were $0.64-3.80$, $0.21-6.91$, 85.53 -463.8. Using the corresponding activities, the transfer factors (TFs) from soil-to-vegetables were estimated, the transfers being greatest for ^{40}K, an expected outcome given the essentiality of this element in support of vigorous growth. The TFs of ^{226}Ra and ^{232}Th were found to be in accord with available literature data, the values indicating the mobility of these radionuclides to be low in the studied soils. Committed effective dose and the associated life-time cancer risk was estimated, being found to be below the permissible limit proposed by UNSCEAR. Results for the studied media show that the prevalent activities and mobilities pose no significant threat to human health, the edible vegetables being safe for consumption.

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1. Introduction

Our radiation environment is contributed to by two component source types: natural and anthropogenic. Natural sources comprise the radiation from cosmogenic and terrestrial activity, while artificial sources derive from reactor and accelerator-driven processes. ^{238}U, ^{232}Th and ^{40}K are the three predominant long-lived naturally occurring radionuclides that are present in the earths' crust, with activity appearing in soil, in water and/or in the earth-born material such as dust, also giving rise to radon in the air. The discharge of

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various radionuclides into the ecosystem arise from human activities such as mining and associated minerals beneficiation, oil and gas production, in use of radionuclides in medical procedures, coal combustion and nuclear fuel cycle involvements such as fuel rods reprocessing, cement production, phosphate fertilizers production/utilization and in waste disposal (Nollet and Pöschl, 2007). To these can be added the residual environmental legacy of the testing of nuclear weapons and accidental releases from nuclear facilities (eg Windscale, Three Mile Island, Chernobly and Fukushima), contributing *trans*-nationally to the radionuclide balance in the environment. Somewhat unknowingly, we ingest these radionuclides via daily food and water intake and also through inhalation, the soil-plant-human pathway being a primary route for the transfer of radionuclides to humans (IAEA, 1982). The migration and mobilization of radionuclides into and intra environment are influenced by many factors, including those that are physiological, biological and geochemical, with soil, air, water and flora property modifications, and specific interactions of radionuclides with vegetation and other organisms within which they accumulate (Černe et al., 2010).

Radionuclides in soil are taken in by plant tissue in the uptake of minerals, directly transferred via the roots system, subsequently appearing in the food chain through the consumption of the meat of ruminants (ovine, bovine, etc) or through the consumption of vegetables common to the human diet. Edible plant/flora represent an important component of the daily diet such that plant uptake of radionuclides into the human food chain represents one of the main vectors used in calculating exposure rates and performing risk assessment (Rosén et al., 1995). Under normal environmental conditions, some 90% of ^{226}Ra (one of the major progeny of ^{238}U) enters into the human body via the food chain (Tettey-Larbi et al., 2013). Once radionuclides enter the human system, through ingestion, inhalation or external irradiation, concentration is possible in various parts of the body, hence the long biological half-lives of many radionuclides making them a potential threat to human health. Analysis of radionuclides uptake from soils by plants is important not only for analyzing the viability of arable soils, but also in evaluating the decontamination of soils by plants.

The soil-to-plant transfer factors (TF), the ratio of the concentration of radioactivity in the crop to the radioactivity per unit mass of the soil, is a value widely used for calculating radiological human dose via the ingestion pathway. It is regarded as one of the most important parameters used in evaluation studies on the impact of releases of radionuclides in the environment and/or environmental safety assessment (IAEA, 1994). This parameter is necessary for environmental transfer models, useful in the prediction of radionuclide concentration in agricultural crops for estimating dose impact to humans (Absalom et al., 1999; Antonopoulos-Domis et al., 1990; Frissel, 1994; Mayall, 1995; Thorne and Coughtrey, 1983). Transfer factor values in excess of unity imply active bioaccumulation of radioactivity. Conversely, values of less than unity either imply strong binding of radioactivity to the soil or that the plant life is not accumulating that material. For most of Europe and the USA, the TFs for most important agricultural products are known. For other areas and especially rapidly developing countries, Malaysia being a prime example of one in which the peaceful applications of nuclear technology are expanding at a pace, the TFs are not so readily available (IAEA, 1994, 2000). Thus, a local database is required for validation of existing models (e.g., the Absalom et al. (1999) model) or development of new models to predict the impact of deposited radionuclides that are based on local parameters derived for tropical environments.

The health benefits of vegetable consumption are clearly documented in the literature. Block et al. (1992) documented that the health benefits include reduction in the incidence of various forms of cancer as well as other ailments such as stroke, heart disease, and obesity. Being a tropical country, Malaysia produces a wide variety of vegetables. Malaysians relying on vegetables as a main source of fiber, minerals and vitamins. Per capita consumption of vegetables in Malaysia reveals there to be an increasing trend of these in the diet, from 27.25 kg in 1982 to 40.58 kg in 2001 and 45.9 kg in 2005 (FAMA, 1993; Ministry of Agriculture and Agro-based Industry). Being an important source of nutrients, determination of radioactivity in edible vegetables is of particular significance.

Herein measurement is made of the activity concentrations of naturally occurring and anthropogenic radionuclides available in the soil, their presence in plants, and soil-to-plant TFs. The measurements involve some common edible vegetables (see below for details) collected from the Peninsular Malaysia. The data obtained are suggested to be useful for radiological assessment to the local population and also as input to epidemiological studies, the procedures also being important as a guide to those wishing to follow the present system in obtaining the associated radiological profiles. The results are also expected to serve as reference data, assisting in ascertaining possible changes in environmental radioactivity due to nuclear, industrial, and other human activities.

2. Materials and methods

2.1. Selection of sampling location

Malaysia, located between 1° and 7° north of the Equator has specific ecological conditions with an average temperature of 27°–32 °C throughout the year. The humidity is high (80–85%) and rainfall is fairly evenly distributed. Originally, vegetables were grown in small holdings worked by a largely rural community, subsequently to be used for their own consumption; the bulk of urban needs were served by imports. However, urbanization and rapid industrial development stimulated demand for fresh greens in urban centers. Consequently, land to the outskirts of town centers were converted into vegetable farms, with larger farms being concentrated in specialized highlands areas for the production of temperate vegetables; peat regions in other areas are similarly being worked for edible vegetables production. A total of five such locations (See Fig. 1) were selected to collect the samples: Langkawi

Fig. 1. The sampling locations (red solid circles) around peninsular Malaysia. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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Island in Kedah, Tumpat in Kelantan, Pasir Panjang in Negeri Sembilan, Klang in Selangor and Cameron Highlands in Pahang. Among them, Langkawi Island produces the least amount of vegetables. It consists of some 99 islands (the total land area is 47,848ha) whereas the main island has a total area of 32,000ha which is about the same size as Singapore. It is covered by 60% forest, mountains, mangroves and shrubs with 20% being agricultural land, and the rest 20% is developed areas (http://www. langkawi-destination-manual.com/main_page.htm). On the other hand, Cameron Highlands is a popular tourist and recreational area in addition to it being a key agricultural area for vegetables, flowers and tea (Muhammad et al., 2009). The farmers of this area produce and supply over half of all Malaysian vegetable needs, and also contribute in earning sizeable foreign remittances (Barrow et al., 2009). The other vegetable farm locations produce relatively lower amounts, albeit sufficient to meet the demands of the local populace. A summary of the total cultivable areas of the studied vegetables, corresponding yearly production etc. is given in Table 1.

The vegetables selected in this research are tapioca, spinach, tomato, cabbage, brinjal and cucumber. Among the vegetables selected, cabbage is the most popularly consumed in Malaysia, the statistical report of Ministry of Agriculture Malaysia showing cabbage production to be the greatest, at just less than 300,000 metric tonnes, followed by lesser amounts of tapioca, tomato, cucumber, spinach and brinjal.

Mature samples (commercial grade) were collected from three randomly selected locations from each vegetable farm, subsequently carefully cleaned to remove any soil or dirt. 2–3 kg of each sample from each sampling point were retained in a polythene bag, labeled and transported to the sample processing room of the environmental radiation laboratory for subsequent investigation.

2.2. Pre-treatment of samples and measurements

As previously mentioned, the vegetable samples were washed until free from soil and foreign media, then subsequently dried in air for several days at room temperature until a constant weight was reached. With the study focusing on ingestion of vegetables, only the edible parts were prepared for analysis. The dried vegetables were cut into smaller pieces, with each individual sample kept on its own tray for oven drying. For the soil samples, anomalies (such as leaves, roots, rocks etc.) were removed to ensure a representative soil sample. After that, the soil samples were oven dried at 80 °C for 16–18 h. The items were then crushed into fine powder and homogenized by filtering through a 1 mm sieve to obtain uniformity. An amount of 180–240 g of each sample was sealed in a Marinelli beaker and stored for a period of 4–6 weeks at room temperature to allow the attainment of secular equilibrium between ^{226}Ra, ^{232}Th and their progenies prior to analyses by gamma-ray spectroscopy (Asaduzzaman et al., 2014).

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples were determined using a p-type coaxial HPGe γ-ray spectrometer (ORTEC; GEM-25; serial no. 46-TP22121A; 57.5 mm crystal diameter; 51.5 mm thickness; +2800 V operating voltage) having a relative efficiency of 28.2% and an energy resolution of 1.67 keV-FWHM at the 1332.5 keV peak of ^{60}Co shielded by a cylindrical lead. The HPGe detector was connected to a 16 k MCA for data acquisition and the γ-rays emitted from the samples were analyzed by Gamma Vision 5.0 software (EG&G ORTEC). A cylindrical multi-nuclide γ-ray source (^{241}Am, ^{57}Co, ^{137}Cs, and ^{60}Co) having homogeneously distributed activity with the same volume and shape as that of the Marinelli beakers containing the environmental samples (so that the detection geometry remained the same) was used for detector energy calibration and the absolute photo-peak efficiency evaluation (Amin et al., 2013). The measured detection efficiencies were fitted by using a polynomial fitting function as described in Khandaker et al. (2013), and the fitted efficiencies were used in activity determination of the samples. The samples were counted for a sufficiently long time to decrease the counting error, and background counts for the same counting time were deducted to obtain the net counts. The gamma-ray lines of 351.93 keV (35.6%) from ^{214}Pb, 609.32 keV (45.49%) from ^{214}Bi, and 1120.294 keV (14.92%) from ^{214}Bi were used to determine the activity concentrations of ^{226}Ra; the gamma-ray lines of 238.632 keV (43.6%) from ^{212}Pb, 583.19 keV (85.0%) from ^{208}Tl to 911.204 keV (25.8%) from ^{228}Ac were used to determine the activity concentrations of ^{232}Th; while the single gamma-ray line 1460.822 keV (10.66%) was used to determine the activity concentrations of ^{40}K (Khandaker et al., 2015).

2.3. Minimum detectable activity (MDA)

In gamma-ray spectrometry, the minimum detection limit refers to the minimum detectable activity (MDA) which describes the smallest amount of radioactivity that can be measured with a certain degree of confidence. The MDA depends on various factors such as the sample composition, the energy of the radiation, the source-detector distance, the detection efficiency, the background radiation level and the time available for the measurements (Debertin et al., 1988). The definition of MDA was first introduced by Currie and it is based on a binary (yes or no) decision as to whether a given sample contains activity (Currie, 1968). The MDA of the gamma-ray measurement system was calculated by the following equation (Solak et al., 2014; UNSCEAR, 1988):

$$MDA = \frac{K_\alpha \sqrt{B}}{\varepsilon_\gamma \rho_\gamma t_c w} \tag{1}$$

Where, where K_α is the statistical coverage factor equal to 1.645 at 95% confidence level, B is the background counts at the region of

Table 1
Total cultivable area of the studied vegetables, yearly production, and the respective population. Information about the planted area and total production was obtained from the report: "PERANGKAAN TANAMAN SAYUR-SAYURAN DAN TANAMAN LADANG" (Vegetables and Cash Crops Statistics), Department of Agriculture, Putrajaya, Malaysia, 2014.

States	Studied area	Studied vegetables	Studied vegetables planted area (hectar)	Total production amount (MT)	Population of studied area
Pahang	Cameron highlands	Cabbage	7265.16	290,471.00	38,471
		Tomato	1663.30	148,191.00	
		Cucumber	310.78	18,596.28	
		Brinjal	204.00	11,832.00	
Kelantan	Tumpat	Tapioca	125.21	1941.96	168,600
Negeri Sembilan	Pasir Panjang	Tapioca	13.04	121.69	40,000
Selangor	Klang	Spinach	47.11	589.55	842,146
Kedah	Langkawi	Tapioca	32 tonnes per[a] hectare	64,000	
Peninsular Malaysia		*All vegetables*	*59,479.55*	*1,301,655.76*	*23,600,000*

[a] http://www.e-penerbitan.org/BTT/2008/63-69.pdf

interest of a certain radionuclide, ε_γ is the efficiency of the HPGe detector at respective gamma-ray energy, ρ_γ is the gamma-ray emission probability, t_c is the counting time and w is the dry-weight of sample (kg). The MDA for the radionuclides of interest was calculated as 0.35 Bq kg^{-1} for ^{226}Ra, 0.12 Bq kg^{-1} for ^{232}Th and 2.2 Bq kg^{-1} for ^{40}K.

2.4. Activity concentrations of radionuclides

To calculate the activity concentration, the relevant decay data (See Table 2) of the detected radionuclides were used by referring to the NUDAT-2.6 data base. Each sample was counted for 86,400 s (24 h) and the background counts were deducted to obtain the net activity. The activity concentrations or the specific activities were measured by using the following equation (2) (Khandaker et al., 2012; Asaduzzaman et al., 2015a):

$$A_c = \frac{N \times 1000}{\varepsilon_\gamma \times I_\gamma \times t_s \times W} \quad (2)$$

Where, A_c = activity concentration of the radionuclide in the sample given in Bq kg^{-1}, N = net counts under corresponding photo-peak, ε_γ = detection efficiency corresponding to specific gamma-ray, I_γ = absolute transition probability of the specific gamma-ray, W = mass of the sample in gram, t_s = counting time in seconds. The weighted means of the various daughter products (Table 2) were used to obtain the final activity concentrations of ^{226}Ra and ^{232}Th to reduce the uncertainty of the derived values (Asaduzzaman et al., 2015b). On the other hand, the accuracy of our measurement system was tested using the reference materials traceable to NIST (Soil and vegetable standards is supplied by Nuclear Technology Services Inc., USA in a customer supplied counting containers with matrix density of 1.5–1.7 g/cc and 0.6 g/cc, respectively).

2.5. Estimation of uncertainty

The uncertainty of the measured data was obtained by using the relevant parameters as a function of the estimate:

$$y = f(x_1, x_2, \dots x_n) \quad (3)$$

Where, x_1 is the statistical uncertainty, x_2 is the uncertainty in sample weight, x_3 is the uncertainty in the efficiency of detector, x_4 is the uncertainty in gamma-ray intensity. The combined standard uncertainty of the estimate y is denoted by $u_c(y)$ and is obtained by using the error propagation law given in equation (4). The measured radionuclides activity together with their uncertainties is presented in Table 3.

$$u_c^2[y(x_1, x_2, x_3, \dots, x_n)] = \sum_{i=1}^{n} \left(\frac{\partial y}{\partial x_i}\right)^2 u^2(x_i) \quad (4)$$

2.6. Calculations of transfer factor

The transfer factor is a useful parameter for radiological assessment, and is defined as the steady-state concentration between one physical situation and another. The soil-to-vegetable transfer factor was calculated by taking the ratio of the activity concentration of dry vegetable to the activity concentration of dry soil. Dry mass condition was used to avoid the inaccuracies associated with the different moisture content of crops. TF values were calculated using the definition proposed by the International Union of Radioecologists (IUR, 1994), which is:

$$TF = \frac{C_i^v \ (Bq \ kg^{-1}, \quad dry \ weight)}{C_i^s \ (Bq \ kg^{-1}, \quad dry \ weight)} \quad (5)$$

Where, TF is transfer factor of soil-to-vegetable; C^v is the concentration of radionuclides in Bq/kg dry vegetables weight; and; C^s is the concentration of radionuclides in Bq/kg dry soil weight in the upper 20 cm layer of soil.

2.7. Daily intake of radioactive materials (Bq d^{-1})

The daily intake of radioactive materials depend on the amount of ^{226}Ra, ^{232}Th and ^{40}K present in the edible flora and the consumption of vegetables by an average adult human, and were evaluated using the following formula (Patra et al., 2013; Khandaker et al., 2013):

$$D_{int} = \frac{A_c \times A_p \times F}{M_p \times Y_d} \quad (6)$$

where, D_{int} is the daily individual intake of radionuclides (in Bq), A_c is the activity concentration of radionuclides (Bq kg^{-1}), A_p is the annual production of vegetables (kg), M_p is the population of Malaysia (23.6 million in 2012), Y_d is the days in a year and F is the fraction of edible part of vegetables; the value of F was taken to be 90% based on local knowledge of vegetable consumption by the inhabitants of Malaysia. The estimated values of daily intake of radionuclides through the consumption of vegetables are presented in Table 4.

Table 2
Decay data of radionuclides and the respective gamma lines used for activity determination, taken from the NUDAT-2.6 data base (http://www.nndc.bnl.gov/nudat2). Italicised gamma-lines were not used in this study.

Radionuclides of interest	Detected nuclides	Half-life	Decay mode (%)	γ-ray energy, E$_\gamma$ (keV)	γ-ray intensity, I$_\gamma$ (%)	Sources/origin
^{226}Ra	^{214}Pb	26.80 m	β (100)	295.22	18.42	^{238}U (^{226}Ra) series
				351.93	35.6	
	^{214}Bi	19.90 m	α (0.02); β (99.98)	609.32	45.49	^{238}U (^{226}Ra) series
				1120.294	14.92	
				1764.491	*15.3*	
^{232}Th	^{228}Ac	6.15 h	$\alpha + \beta$ (100)	338.32	*11.27*	^{232}Th series
				911.204	25.8	
				968.971	15.8	
	^{212}Pb	10.64 h	β (100)	238.632	43.6	^{232}Th (^{228}Ra) series
				300.087	*3.30*	
	^{208}Tl	3.053 m	β (100)	583.187	85	^{232}Th (^{228}Ra) series
				860.557	*12.5*	
^{40}K	^{40}K	1.248E+09 y	EC (10.72); β (89.28)	1460.822	10.66	Primordial/terrestrial

Table 3
Activity concentrations (Bq kg⁻¹ dry weight) of the studied radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in vegetables and associated soils, and their transfer from soil to vegetables.

Growing location	Type of vegetables	Sample information	Radionuclides ($Bq\ kg^{-1}$) activity Concentration			Transfer factors		
			^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
Cameron Highland	Leafy	Cabbage	3.80 ± 0.42	6.91 ± 0.54	240.2 ± 11.2	0.18	0.26	1.76
		Cabbage (Soil)	21.36 ± 1.03	26.80 ± 1.34	136.5 ± 6.4			
Klang		Spinach	2.03 ± 0.43	1.00 ± 0.63	463.8 ± 21.9	0.96	0.21	38.17
		Spinach (Soil)	2.11 ± 0.3	4.85 ± 0.36	12.15 ± 1.09			
Cameron Highlands	Fruit	Tomato	0.76 ± 0.16	0.21 ± 0.13	309.8 ± 14.3	0.04	0.01	2.38
		Tomato (Soil)	20.6 ± 0.98	25.55 ± 1.16	130.1 ± 6.10			
		Cucumber	0.64 ± 0.16	0.42 ± 0.14	290.1 ± 13.42	0.03	0.02	2.22
		Cucumber (Soil)	19.83 ± 0.99	24.64 ± 1.15	130.9 ± 6.12			
		Brinjal	0.80 ± 0.15	0.47 ± 0.15	209.0 ± 9.7	0.04	0.02	1.60
		Brinjal (Soil)	19.68 ± 0.95	25.20 ± 1.10	130.0 ± 6.13			
Langkawi	Root	Tapioca	1.14 ± 0.13	0.74 ± 0.11	399.7 ± 18.5	0.04	0.05	3.37
		Tapioca (Soil)	30.90 ± 1.46	15.13 ± 0.86	118.6 ± 10.56			
Negeri Sembilan		Tapioca	0.72 ± 0.18	1.90 ± 0.36	93.1 ± 4.5	0.54	3.96	11.66
		Tapioca (Soil)	1.33 ± 0.09	0.48 ± 0.12	7.99 ± 0.44			
Kelantan		Tapioca	0.99 ± 0.13	0.56 ± 0.12	85.53 ± 4.04	0.15	0.06	0.74
		Tapioca (Soil)	6.68 ± 0.36	9.53 ± 0.46	115.5 ± 5.36			
All locations		**Vegetable; Range**	**0.64–3.80**	**0.21–6.91**	**85.53–463.8**	**0.03–0.96**	**0.01–3.96**	**0.74–38.17**
		Soil; Range	**1.33–30.90**	**0.48–26.80**	**7.99–136.5**			

2.8. Committed effective dose (μSv y⁻¹)

Committed effective dose due to the accumulation of radionuclides in the human body were assessed using the equation (7) following (Khandaker et al., 2013; Alam and Mohamed, 2011) and are presented in Table 4.

$$D_{eff} = D_{int} \times D_{cf} \times 365 \qquad (7)$$

Where, D_{eff} is the committed effective dose (μSv y⁻¹) to an individual due to an ingestion of radionuclides, 365 is the days in a year, D_{cf} is the ingestion dose conversion factor for the radionuclides of interest (2.8×10^{-7} Sv Bq⁻¹ for ^{226}Ra, 2.3×10^{-7} Sv Bq⁻¹ for ^{232}Th and 6.0×10^{-9} Sv Bq⁻¹ for ^{40}K) was taken from IAEA (2011). The total committed dose via ingestion can be calculated by the following formula:

$$D_{eff}^{total} = \sum_{i=226_{Ra},228_{Ra},\ 40_K} \left(D_{eff,i} \right) \qquad (8)$$

2.9. Excess lifetime cancer risk

Cancer is recognized to be a life-threatening disease, the frequency of this disease increasing globally, including in Malaysia, due to various reasons that include environmental factors. One of the causes is the radiation effect on the biological cell. Based on the National Cancer Society Malaysia, there are some 90–100,000 people in Malaysia living with cancer at any one time (http://www.cancer.org.my/quick-facts/types-cancer/). Less than 10% of cancers happen in children compared to over 50% in men and 35% in women aged 50 and above. While considered the 3rd leading cause of premature death in Malaysia, only 30–40% of all deaths from cancer are medically certified, meaning there is no exact figure of people dying from cancer. Therefore, an effort was made to evaluate the excess lifetime cancer risk (ELCR) due to the consumption of vegetables, use being made of the procedure proposed by the United States Environmental Protection Agency (US EPA, 1999). The following equation (Patra et al., 2013) was used for the calculation the mortality cancer risk, values being presented in Table 4.

$$ELCR = D_{int} \times R_{cf} \times A_{ls} \times 365 \qquad (9)$$

Where, ELCR, D_{int}, A_{ls} and R_{cf} are respectively the lifetime cancer risk, daily intake of radionuclide (Bq), average span of life (74 y) and mortality cancer risk coefficient (Bq⁻¹) for the ingestion of food. The ingestion mortality cancer risk coefficients 9.56×10^{-9} (Bq⁻¹) for ^{226}Ra, 2.45×10^{-9} (Bq⁻¹) for ^{232}Th and 5.89×10^{-10} (Bq⁻¹) for ^{40}K were used for the estimation of ELCR (US EPA, 1999).

Table 4
Daily intake of ^{226}Ra, ^{232}Th and ^{40}K radionuclides, committed effective dose, estimated total effective dose and excess lifetime cancer risk due to the consumption of vegetables.

Growing location	Vegetable name	Daily intake of radioactive materials by individual D_{int} (mBq d⁻¹)			Committed dose E_{ef} (μSvy⁻¹)			Total effective dose E_t (μSvy⁻¹)	Lifetime cancer risk (ELCR)		
		^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K		^{226}Ra	^{232}Th	^{40}K
Cameron highland	Cabbage	61.3	111.5	3879	6.27	28.1	8.78	43.1	1.6×10^{-5}	7.4×10^{-6}	6.2×10^{-5}
	Tomato	9.7	2.7	3974	0.99	0.68	8.99	10.67	2.5×10^{-6}	1.8×10^{-7}	6.3×10^{-5}
	Cucumber	3.2	2.1	1447	0.33	0.53	3.27	4.13	8.3×10^{-7}	1.4×10^{-7}	2.3×10^{-5}
	Brinjal	2.4	1.4	636.6	0.25	0.36	1.44	2.05	6.3×10^{-7}	9.5×10^{-8}	1.0×10^{-5}
Langkawi	Tapioca	3.42	2.22	1199	0.35	0.56	2.71	3.62	8.8×10^{-7}	1.5×10^{-7}	1.9×10^{-5}
Klang	Spinach	6.18	3.05	1410	0.63	0.77	3.19	4.59	1.6×10^{-6}	2.0×10^{-7}	2.2×10^{-5}
Negeri Sembilan	Tapioca	2.2	5.7	279.3	0.22	1.44	0.63	2.29	5.6×10^{-7}	3.8×10^{-7}	4.4×10^{-6}
Kelantan	Tapioca	2.96	1.69	256.6	0.30	0.43	0.58	1.31	7.6×10^{-7}	1.1×10^{-7}	4.1×10^{-6}
Average		**11.42**	**16.30**	**1635.2**	**1.17**	**4.11**	**3.70**	**8.97**	**2.9×10^{-6}**	**1.1×10^{-6}**	**2.6×10^{-5}**
World average (UNSCEAR, 2000B)	–	–	–	120	120	170	290	~10^{-3}			

3. Results and discussion

The measured activity concentrations of natural radionuclides in the soil samples are presented in Table 3. The results are reported in Bq/kg on a dry-weight basis. The activity concentration in the soil samples ranges 1.33 ± 0.09–30.90 ± 1.46 Bq/kg for ^{226}Ra (^{238}U), 0.48 ± 0.12–26.80 ± 1.34 Bq/kg for ^{228}Ra (^{232}Th) and 7.99 ± 0.44–136.5 ± 6.4 Bq/kg for ^{40}K. Table 3 shows the activity of ^{232}Th chain to be greater than that of ^{238}U chain in most of the studied soils, supportive of the fact that the presence of thorium is 1.5 times greater than that of uranium in the Earth's crust (Kabir et al., 2009). It was observed that the activity concentrations of ^{226}Ra and ^{232}Th in the soil were not uniform, varying with the type and location of soils, depending upon the geological characteristics of the area under study. Specifically, the soil collected from tapioca farmland of Langkawi was observed to show the greatest activity concentrations for the radionuclide ^{226}Ra. This could possibly be attributed to the contribution of technologically enhanced naturally occurring radioactive material (TENORM) in this location via human activities, potentially though soil movement, the farm being located near to the town on an island famous as a tourist destination. In general, in regard to the other farms, the soils collected from the highlands farm was observed to show greater activity concentrations than the lowland farms. It was also observed that the measured activity of ^{40}K (a non-chain radionuclide) exceeded by far the values of both ^{232}Th and ^{226}Ra, being both the most abundant radioactive element present in the environment and it also being noted that potassium is used extensively as part of a NPK fertilizer medium in intensive farming activities to promote vigorous growth.

The activity concentrations of natural radionuclides present in the vegetable samples are given in Table 3. The results are reported in Bq/kg on dry-weight basis, values ranging from 0.64 ± 0.16 to 3.80 ± 0.42 Bq/kg for ^{226}Ra(^{238}U), 0.21 ± 0.13 to 6.91 ± 0.54 Bq/kg for ^{228}Ra(^{232}Th) and 85.53 ± 4.04 to 463.8 ± 21.9 Bq/kg for ^{40}K. Results show that the activity concentrations of the investigated samples vary considerably with respect to vegetable type. As an instance, cabbage from the Cameron Highlands showed the activity concentrations to be greatest for ^{226}Ra and ^{232}Th. The trend of ^{226}Ra activity in the studied samples was found to be in the following order: cabbage > spinach > tapioca > brinjal > tomato > cucumber, while for ^{232}Th, it is cabbage > tapioca > spinach > brinjal > cucumber > tomato. It can

be seen from Table 3 that ^{40}K was found to accumulate in relatively large amounts in different vegetables compared to that for ^{226}Ra and ^{232}Th. The accumulation can attributed to the greater biological demand in plants for potassium, a major essential nutrient element; also plants have the tendency to take up soluble potassium far in excess of their needs if sufficiently large quantities are present, termed as luxury consumption (Brady and Weil, 2002). Radioactive potassium is also taken along with non-radioactive potassium. Thus, the activity concentration of ^{40}K in vegetables was very much greater than that in soils. Furthermore, activity concentrations of ^{40}K show considerable variation within the studied soils and vegetables and/or locations. Among the studied samples, the spinach from Klang showed the greatest ^{40}K activity concentration.

The calculated soil-to-vegetable transfer factors are presented in Table 3. The TF for ^{40}K presented the largest values range at 0.74 to 38.17 followed by 0.01–3.96 for ^{232}Th and 0.03–0.96 for ^{226}Ra. In general, transfer of radionuclides from soil-to-vegetables show considerable variation with respect to soils, vegetables and locations. The uptake of radionuclides from soil by vegetables depends on various interrelated soil properties including texture, clay content, dominant clay minerals, exchangeable cations, pH, organic matter content and other environmental conditions, as reported in the IAEA Technical Report Series, TRS No. 310 (IAEA, 1990). Moreover, the absorption of radionuclides is affected by various parameters such as the chemical and physical properties of soil and radionuclides, plant species, stage of growth, agricultural practices, soil microbial activity and the weather etc. (Carini, 2001). Martínez-Aguirre et al. (1995) reported that Th exhibited a much lower mobility than U, which is consistent with our observations that ^{232}Th has smaller TF values except for tapioca from Negeri Sembilan and cabbage from Cameron highlands. Specifically, leafy vegetables such as spinach (for ^{226}Ra and ^{40}K) and cabbage (for ^{232}Th) show the greatest transfer factors among the studied samples. Leafy vegetables and grains generally tend to have the greatest and least transfer factors respectively (IAEA, 2000). With the exception of the study of tapioca from Kelantan, the TFs for ^{40}K show a value of greater than unity for all other samples. Moreover, TF values of ^{40}K show a wide range of variation (0.74–38.17) among the studied samples, and are greater than unity in most of the samples. The high TF of ^{40}K was probably due to the excessive use of potassium-containing fertilizers at the sampling sites, its high mobility in soil, and its subsequent uptake by plants.

Table 5
A comparative study of the estimated TFs with the other published data worldwide.

Country	Investigated samples	Transfer factors			References
		^{226}Ra	^{228}Ra	^{40}K	
Greece	Soil-to-plant			0.25–2.42	Papastefanou et al., 1999
Spain	Soil-to-grass pasture	0.097–0.504 (0.17)	0.517–4.31 (1.65)		Vera Tome et al., 2003
Nigeria	Soil-to-tomato	0.064	0.13		Jibiri et al., 2007
Pakistan	Soil-to-vegetables		0.08		Matiullah et al., 2008
Syria	Soil-to-vegetables	0.008			Al-Masri et al., 2008
Brazil	Soil-to-lettuce	0.035–0.041	0.24–0.32		Jacomino et al., 2009
Pakistan	Soil-to-cucumber		0.09		Hasan et al., 2010
Pakistan	Soil-to-pumpkin		0.04		Hasan et al., 2010
Sweden	Soil-to-grass	0.5–1.3	0.3–0.9		Pallavicini, 2011
India	Soil-to-leaf	0.03–0.65		0.32–8.04	James et al., 2011
Malaysia	Soil-to-vegetables	0.006–0.028	0.002–0.013		Aswood et al., 2013
Bangladesh	Soil-to-plant	0.082–0.926	0.061–0.806	0.738–3.741	Gaffar et al., 2014
Egypt	Soil-to-plant	0.19–0.73 (0.43)	0.10–0.76 (0.32)	0.31–2.95 (1.06)	Harb et al., 2014
USA	Soil-to-vegetables	0.003–0.057			Staven et al., 2003
Worldwide	Soil-to-grass	0.04	0.04		UNSCEAR, 2010
Peninsular Malaysia	Soil-to-vegetables	0.03–0.96	0.01–3.96	0.74–38.17	This work

Table 5 represents a comparative study of transfer factors estimated by the present work with the results from several other countries worldwide. With the exception of ^{40}K, the transfer factors of the primordial ^{226}Ra and ^{232}Th radionuclides were below unity (except ^{232}Th for tapioca from Negeri Sembilan). However, the estimated TFs of ^{226}Ra and ^{232}Th for vegetable samples show quite elevated values compared to the literature data reported by Matiullah et al. (2008), Al-Masri et al. (2008), Aswood et al. (2013) and Staven et al. (2003). This variation could be attributed to soil type, agricultural practices, transport process, media and absorbing capacity of the plants studied. On the other hand, the magnitude and range of TFs for ^{226}Ra and ^{232}Th found in this study appeared to be generally similar to other studies where root uptake was the primary mechanism of accumulation (Jacomino et al., 2009; Hasan et al., 2010; Pallavicini, 2011; James et al., 2011).

The daily intake of each radionuclide from different varieties of vegetables was found to be in the range 2.2–61.3 mBq d^{-1} for ^{226}Ra; 1.4–112 mBq d^{-1} for ^{232}Th and 0.26–3.97 Bq d^{-1} for ^{40}K. The greatest intake of radioactive material has been observed to be from the consumption of cabbage, followed by tomato, spinach, tapioca, cucumber and brinjal. The estimated daily intake due to the consumption of vegetables for ^{226}Ra and ^{40}K were found to be

very similar to that reported in our previous study (Asaduzzaman et al., 2015c). The committed effective doses through the consumption of the vegetables under study were found to be in the range 0.22–6.27 µSv y^{-1} with an average value of 1.17 µSv y^{-1} for ^{226}Ra, 0.36–28.1 µSv y^{-1} with an average value of 4.11 µSv y^{-1} for ^{232}Th and 0.58–8.99 µSv y^{-1} with an average value of 3.70 µSv y^{-1} for ^{40}K (Table 4). According to a report by UNSCEAR (2000), the total exposure per person resulting from the ingestion of terrestrial radioisotopes should be ≤ 290 µSv y^{-1}, of which 170 µSv y^{-1} is from ^{40}K to 120 µSv y^{-1} is from the thorium and uranium series radionuclides. The estimated dose values were found to be below the world average reported by UNSCEAR (2000) (Table 4). This study found that, due to the consumption of the investigated vegetables, an individual might receive a total radiation dose of approximately 71.76 µSv y^{-1}, some 4 times lower than the world average (290 µSv y^{-1}). From the practical point of view, while present results show the total committed dose to pose no particular health burden to the population, cumulative exposure of such radioactive materials via consumption of vegetables might not be entirely discounted as a contributory factor adding to risks to health.

Fig. 2 shows the dose contribution of the different categories of vegetables to ^{226}Ra, ^{232}Th and ^{40}K intakes. Among the studied vegetables, cabbage (60%) is the greatest contributor to the committed dose, followed by tomato (15%), tapioca (10%), spinach (6%), cucumber (6%) and brinjal (3%) (Fig. 2). Fig. 2 shows that ^{232}Th is the main contributor to the intake dose, with a contribution of about 46% of total estimated dose, followed by ^{40}K (41%) and ^{226}Ra (13%). ^{40}K is usually of limited interest because as an isotope of an essential element, it is homeostatically controlled in the human cells. Therefore, the body content of ^{40}K is determined mainly by its physiological characteristics rather than its intake (Olatunji et al.,
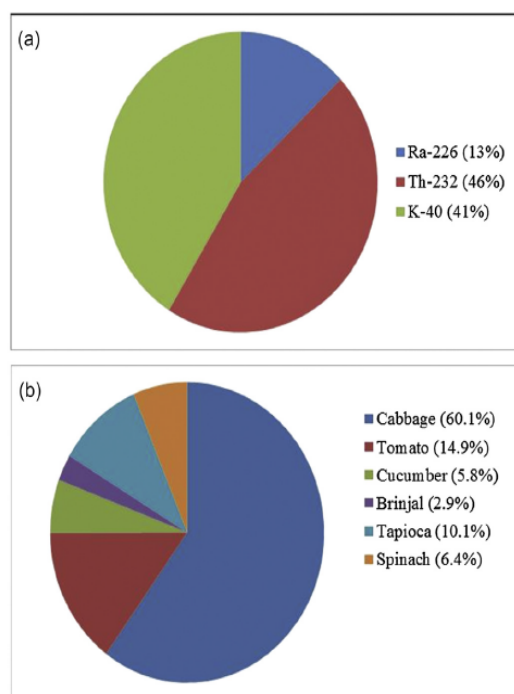

Fig. 2. Dose contribution: (a) in relation to radionuclides; (b) in relation to vegetables.
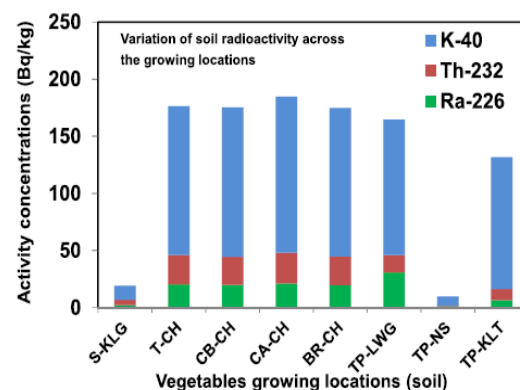
Fig. 3. Variation of the radionuclide concentrations across the vegetable growing locations (S-KLG: Klang (for spinach); T-CH: Cameron Highlands (for tomato); CB-CH: Cameron Highlands (for cucumber); CA-CH: Cameron Highlands (for cabbage); BR-CH: Cameron Highlands (for brinjal); TP-LGW: Langkawi (for tapioca); TP-NS: Negeri Sembilan (for tapioca) and TP-KLT: Kelantan (for tapioca)).

Table 6
Correlation coefficients among the radioactive parameters for the investigated vegetables/their origin.

Variables	Vegetables				Associated soils			TFs		
	^{226}Ra	^{232}Th	^{40}K	AED	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
^{226}Ra	1				1			1		
^{232}Th	0.901	1			0.769	1		0.611	1	
^{40}K	0.203	−0.125	1		0.801	0.879	1	0.960	0.432	1
AED	0.890	0.932	0.032	1	–	–	–	–	–	–
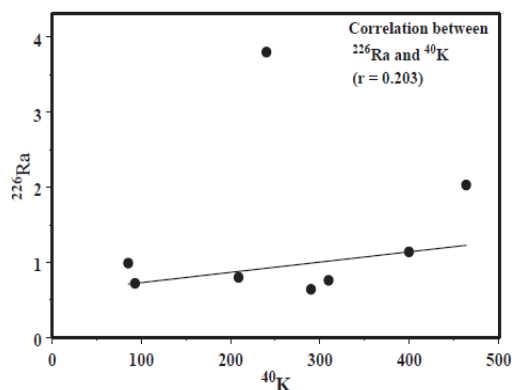
Fig. 4. ^{226}Ra versus ^{40}K concentrations in the investigated vegetables.

2014).

The average ELCR varied from 5.6×10^{-7} to 1.6×10^{-5} for ^{226}Ra, 9.5×10^{-8} to 7.4×10^{-6} for ^{232}Th and 4.1×10^{-6} to 6.3×10^{-5} for ^{40}K, which were low compared with the acceptable ELCR limit of 10^{-3} for radiological risk in general (Patra et al., 2013). Among the examined vegetables, consumption of cabbage from the Cameron Highlands has been observed to contribute the greater cancer risk (for ^{226}Ra and ^{232}Th) which may not only be due to its radioactivity content but also due to the greater consumption rate and comparatively greater risk coefficient.

3.1. Statistical analysis

Detailed analysis of the results from individual measurement was further performed using the S+ package (TIBCO Spotfire Software) to understand the relationship that exists among the radionuclides and their significant differences at the 95% confidence level. Table 6 shows the degree of association existing among the measured radionuclide parameters for the investigated vegetables and their growing locations. Fig. 3 shows how the measured radionuclide concentrations varied with respect to the vegetable growing locations, with ^{40}K showing the greatest values in all the soils. As regard the investigated vegetables, Figs. 4 and 5 show ^{40}K to offer a weak positive correlation with ^{226}Ra (r = 0.203) and a
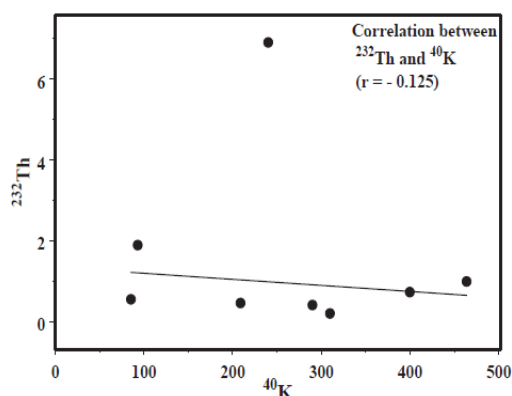

Fig. 5. ^{232}Th versus ^{40}K concentrations in the investigated vegetables.

Fig. 6. ^{226}Ra versus ^{232}Th concentrations in the investigated vegetables.

weak negative correlation with ^{232}Th (r = −0.125). This is an indication that the results for ^{40}K in any of the vegetables may not be suitable in predicting either of ^{226}Ra or ^{232}Th. On the other hand, Fig. 6 shows both ^{226}Ra and ^{232}Th to have a strong positive correlation (r = 0.901) which is an indication that these radionuclides belong to the same decay chain and/or source, and also implies that the result of any could probably be used to predict the other.

4. Conclusions

The activity concentrations of naturally occurring radionuclides in Peninsular Malaysia in soil, vegetables, and TFs for soil-to-vegetables were determined. This study shows that the activity concentrations of ^{226}Ra and ^{232}Th in the soil were not uniform and varied with respect to soil type, location, and geological formation of the area under study. The Highland farms present greater concentrations than the ground farms do. Present results show the highest activity to be for ^{40}K followed by ^{226}Ra > ^{232}Th (in vegetables) and ^{232}Th > ^{226}Ra in their associated soil samples. With the exception of ^{40}K, the transfer factors of ^{226}Ra and ^{232}Th were found to be below unity (except ^{232}Th for tapioca from Negeri Sembilan). The estimated TFs for ^{226}Ra and ^{232}Th show mixed behavior, being greater than the literature data for vegetable samples, and providing results consistent with the literature for plant, grass etc. samples. However, present results indicate that consumption of the studied vegetables pose no serious health burden to the population.

Since higher concentration of radioactive substances in the environment is undesirable, continuous monitoring should be undertaken to detect the concentration of radionuclides in vegetables, soil and soil-to-vegetable TFs in order to take necessary radiological and dosimetric measures with the aim of minimizing the potential harmful effects of ionizing radiation. It is thus hoped that the present results will help to establish a baseline for radioactivity concentrations and TFs of various plants in Peninsular Malaysia.

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3.2.3 Published paper 3

Soil-to-root vegetable transfer factors for ^{226}Ra, ^{232}Th, ^{40}K, and ^{88}Y in Malaysia

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ARTICLE INFO

ABSTRACT

Soil-to-plant transfer factors (TFs) are of fundamental importance in assessing the environmental impact due to the presence of radioactivity in soil and agricultural crops. Tapioca and sweet potato, both root crops, are popular foodstuffs for a significant fraction of the Malaysian population, and result in intake of radionuclides. For the natural field conditions experienced in production of these foodstuffs, TFs and the annual effective dose were evaluated for the natural radionuclides ^{226}Ra, ^{232}Th, ^{40}K and for the anthropogenic radionuclide ^{88}Y, the latter being a component of fallout. An experimental tapioca field was developed for study of the time dependence of plant uptake. For soil samples from all study locations other than the experimental field, it has been shown that these contain the artificial radionuclide ^{88}Y, although the uptake of ^{88}Y has only been observed in the roots of the plant *Manihot esculenta* (from which tapioca is derived) grown in mining soil. The estimated TFs for ^{226}Ra and ^{232}Th for tapioca and sweet potato are very much higher than that reported by the IAEA. For all study areas, the annual effective dose from ingestion of tapioca and sweet potato are estimated to be lower than the world average (290 μSv y^{-1}).

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1. Introduction

Natural radionuclides are ubiquitous in the environment, varying considerably in activity concentrations. In the main, the natural radioactivity in soil arises from ^{238}U, ^{232}Th and their progeny, and ^{40}K. Artificial radionuclides can also be found in the environment, their presence resulting from human activities, including the atmospheric testing of nuclear devices that occurred over the period 1945 to 1980 (Eisenbud, 1973; Ravisankar et al., 2012). The radionuclides transfer through the environment by various possible pathways, as for example through the atmosphere, aquatic systems and soil sub-compartments, each contributing to human exposure. A large fraction of radiation exposure occurs as a result of ingestion of foodstuffs produced in the natural environment, this forming the basis of present interest.

Radionuclides appear in plants, either through direct atmospheric interception onto external plant surfaces, indirectly from re-suspended material, or through uptake of radionuclides via the root system (Vandenhove et al., 2009). Nutrients are taken up from the soil throughout the period of plant growth, forming the main source of plant minerals (Verma and Sharma, 1999). In minerals absorption via the root system, no differentiation is observed between chemically analogous isotopes. Potassium and calcium are among the vital nutrients transferred from the soil to different parts of the plant, together with various nuclides, including ^{226}Ra, ^{137}Cs, and ^{90}Sr (James et al., 2011). Root plants such as tapioca (*Manihot esculenta*), from which the starch tapioca is extracted (also sometimes referred to as cassava), sweet potato (*Ipomoea batatas*), a dicotyledonous plant belonging to the morning glory family Convolvulaceae, carrot (*Daucus carota* subsp. *Sativus*) and radish (*Raphanus sativus*), a root vegetable of the Brassicaceae family etc., each reveal greater root uptake of radioactivity than as a result of interception onto external plant surfaces.

In tropical and subtropical regions, tapioca is extensively cultivated as an annual staple crop, tapioca representing the third most vital source of food carbohydrates in the tropics (FAO, 1995; Fauquet and Fargette, 1990), providing a basic diet for over half a billion people within the developing world. In some developing countries, Malaysia included, tapioca is also used as a nutritional supplement, and is

important for bio-fuel, animal feed and chemicals. In 2011 the estimated area of land devoted to tapioca production in Malaysia was 2596 ha, with a potential yield of 33,200 metric tonnes (Department of Agriculture Peninsular Malaysia, 2011). Tapioca can produce food calories at rates that exceed 250,000 calories ha^{-1} day^{-1} compared with 176,000 calories ha^{-1} day^{-1} for rice, 110,000 calories ha^{-1} day^{-1} for wheat, and 200,000 calories ha^{-1} day^{-1} for maize (Cassava — Wikipedia, *the free encyclopedia*, https://en.wikipedia.org/wiki/Cassava). Conversely, sweet potato, in the Malay language referred to as ubi keledek, is grown mainly as a substitute for rice and corn, and is also consumed to a large extent in tropical and subtropical areas worldwide, including in Malaysia, ranking as the fifth most staple food crop on a fresh-weight basis, after rice, wheat, corn and tapioca. Its flour can be fermented to make products like soy sauce and alcohol. The same 2011 Malaysian survey estimated the area devoted to sweet potato planting to be about 2230 ha, with a potential yield of 26,600 metric tonnes (Department of Agriculture Peninsular Malaysia, 2011).

The variation in concentrations of radionuclides on top-surface soil depends mainly on the mineralogical composition of that soil, as well as its chemical and physical properties, meteorological conditions and the possible transfer of material to deeper soil layers (Misaelides et al., 1987; Vosniakos et al., 1998). The distribution of radionuclides within a plant is useful in understanding the dynamics of radionuclides in an agricultural field, not least since non-edible parts of an agricultural crop are typically returned to the soil, thereby again becoming part of the soil-plant pathway (Davis et al., 1999; Santos, 1975).

The soil-to-plant transfer factor (TF) is a key in calculation of radionuclide concentration in agrarian crops, also allowing estimation of internal radiation dose as a result of food ingestion. The TF of various radionuclides is affected by many factors, such as: (i) the form in which the activity enters or is present in soil (e.g. as particles, aerosol or solution); (ii) the physicochemical properties of the radionuclide; (iii) the time after entry into the soil; (iv) the type of soil and the physicochemical characteristics of the soil environment (including soil properties such as texture, pH, exchangeable K and Ca, the kind and amount of clays, and organic matter); (v) the type of crop (plant species, variety, and cultural practices); (vi) crop management practices (irrigation, plowing, application of fertilizers, liming, etc.); (vii) the climate and the experimental conditions under which the TFs were obtained, and (viii) the degree of preparation of the plant material, such as peeling root crops, washing, etc. (Alexakhin and Korneev, 1992; Evans and Dekker, 1966; Gulyakin and Yudintseva, 1962; Ng et al., 1982; Staven et al., 2003; Pinder et al., 1987; Hilton et al., 1992; U.S. Nuclear Regulatory Commission, 2003; Ewers et al., 2003). Since the TF varies with respect to location it is important to use site-specific data (IAEA, 1994).

While radionuclide TFs have been widely studied in respect of the various components of the food chain of many countries, studies of the transfer of radionuclides to tapioca and the dose received by consumption of it are highly limited. In this paper, soil-to-roots TFs for ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y, for tapioca and sweet potato have been estimated for several sites in west Malaysia, no previous study of this type being known to have been carried out in the areas under investigation.

2. Materials and methods

2.1. Study area

Two states in west Malaysia were selected for sampling locations, Selangor at latitude 3°20′ N, longitude 101°30′ E, and Perak at latitude 4°45′ N, longitude 101°0′ E, where local farmers use a

significant amount of land for the cultivation of tapioca and sweet potato (Fig. 1). Located close to the equator, the study areas do not have the seasons of spring, summer, autumn and winter, the zone being characterized by temperatures generally ranging from 27 °C to 35 °C during the day throughout the year, with relative humidity of 70–90% and annual rainfall of the order of 3200 mm. For comparative study of TF and ingestion dose due to the intake of tapioca and sweet potato, study samples were collected from the Puchong, Batang Berjuntai, Lembah Beringin farmlands of Selangor and the Chemor area of Perak. Puchong is a former tin mining area and therefore of special significance in assessment of naturally occurring radioactive material (NORM) and technologically enhanced NORM (TENORM). The soil texture of this former tin mining area is sandy (coarse) loam (60% sand, 10% silt and 30% clay). The soil of Batang Berjuntai and Lembah Beringin is mainly lateritic, being reddish in color and sandy clay loamy textured (45–48% sand, 23–25% silt and ~30% clay), while Chemor is a peat soil area in which soil is in open textured and contains 60–80% fiber (Table 1).

2.2. Growing practice by local farmers

Cultivation schemes play an important role in plant uptake of radionuclides from the soil (Uchida et al., 2009). In the study areas, different varieties of tapioca and sweet potato are produced with growing practices that are more or less similar. Most of the varieties are grown under dry conditions, the field remaining dry during planting. The farmers do not practice crop rotation and the produce is grown in the same fields, year after year. Local farmers cultivate these root vegetables using the local water resources, also following traditional practices of plowing, conditioning and planting. Generally, the farmers use compound fertilizers, grades NPK 15-15-15 and NPK 13-13-21, the most available in the market. The optimal rate of application of fertilizer is 300–600 kg ha^{-1} and is applied all at one time. Sometimes farmers use the fertilizer a second time, two months after planting. Some slight variation in the kind of fertilizers and their amounts has been observed among the growers.

2.3. Experimental tapioca field for site-specific studies

In regard to the time evolution of plant uptake, an experimental tapioca field of approximately 38 m^2 was chosen in Dengkil (latitude 2°52′ N, longitude 101°41′ E) in the district of Sepang, Selangor (Fig. 1). The average annual rainfall and humidity in Dengkil is 2030 mm and 59%, respectively. The land used for this field was open, the soil being mainly a peat and laterite mixture, reddish in color and of silty clay loamy textured (57.2% silt, 17.8% sand and 25% clay). The field was leveled from its previous heterogeneous structure and plowed before planting, no chemical/organic manure being applied to the soil. Tapioca was planted out at about the same time typically followed by local farmers; cultivation methods also being those typically followed by local farmers, the types of tapioca planted out being identical with those planted out by local inhabitants. Note that it was not expected to observe similar TF time dependencies in tapioca grown in the experimental field and local farmer fields; there being differences in soil physiognomies, deviations in the farming practices and use of fertilizers. However, one would expect to observe similar TF time dependencies, if any, in the experimental field where the same growing practice was followed.

2.4. Growing cycle of tapioca

Generally, tapioca has a growing cycle of between 2 and 12 months, depending on the genotype and the environmental conditions. After planting, roots will appear in as little as three days.
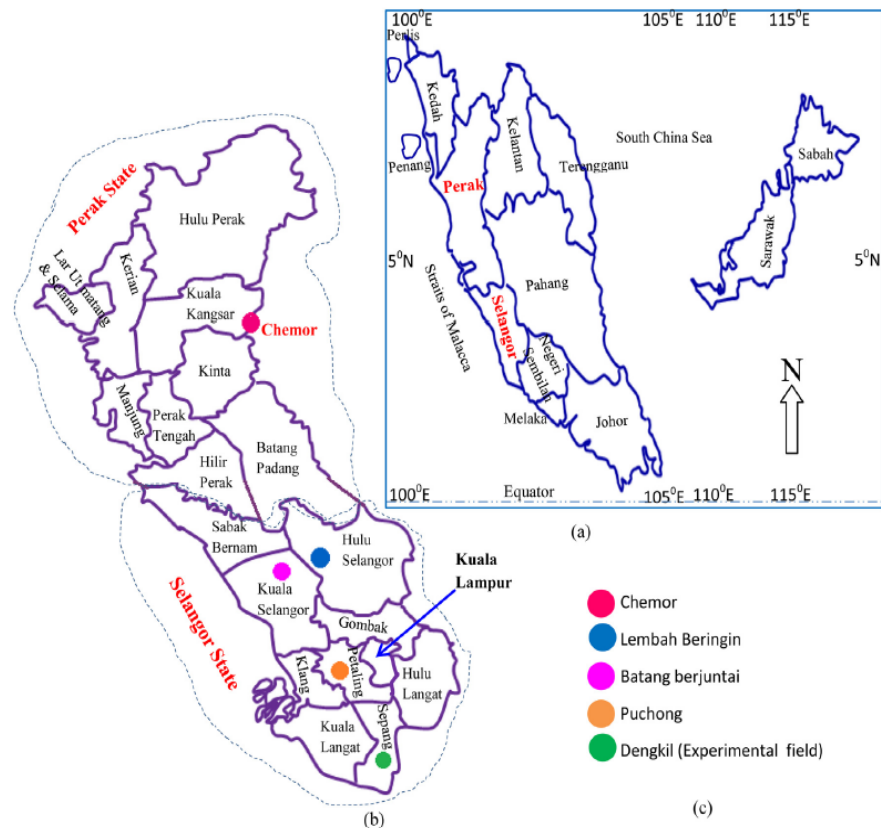
Fig. 1. (a) Map of Malaysia showing the different States; (b & c) Sampling locations (Bullet points).

Tapioca roots grow slowly at first, but within a couple of months the tuber begins to form. In areas where tapioca is grown for fresh human vegetation, farmers harvest from 3 to 6 months after planting up to complete maturity (at 11–12 months). Actually, 3–4 months after plantation is the young stage of tapioca and 5–6 months is the middle stage of maturity and suitable as vegetables. For food usage, the crop is harvested when the root has achieved the size desired by the grower and consumer, usually before the plant is 8 months old. The cassava can spend more than one season underground in tropical regions but a portion may become woody and inedible the longer the tubers grow. These plants are used for their starch content in textiles and for packing materials.

2.5. Sample collection

By spade, mature tapioca and sweet potato root samples were harvested from three random points from each farm, at each study location, and carefully separated from the soil. About 1 kg of each sample from each point was placed in a polythene bag, labeled and transported to the radiation laboratory sample processing room for subsequent investigation.

With effort being made to ensure representative sampling from the study area and using a coring tool, soil samples were collected to a depth of 20 cm in accord with prescribed uniform practice (IAEA, 2010), to include the surface layer corresponding to the rooting zone. The soil was thoroughly mixed to provide a representative sample from each location. As with collection of the roots, about 1 kg of soil from each point was also packed in polythene bags, labeled and taken to the sample processing room as above. In the experimental field, roots and associated soil samples were similarly collected from three different points, monthly sampling taking place from the 3rd month following planting until the 6th month following planting, selected randomly within the field. Sampling locations are as shown in Fig. 1.

2.6. Sample pretreatment

2.6.1. Soil samples

The soil samples were systematically cataloged; dried for several days at room temperature until a constant weight was reached, then stone, gravel, leaf and roots were removed from the sample and it was weighed. For this procedure, the samples were dried in an oven at 135 °C for 24 h to attain constant dry weight and crushed into fine powder and homogenized by filtering through a 1-mm sieve. A mass of 500 g of soil sample from each sampling point was placed in a Marinelli beaker, sealed and stored for a period of 4–5 weeks at room temperature to allow secular equilibrium between ^{226}Ra and its progenies to be achieved prior to gamma spectroscopy (Ghose et al., 2012; Amrani and Tahtat, 2001; Khan and Khan, 2001; Kumar et al., 2003). The physicochemical properties of the studied soil are presented in Table 1.

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Table 1
Physico-chemical parameters of soil.

Characteristics	Puchong	Chemor	Lembah Beringin	Batang Berjuntai	Experimental field
Texture	Sandy loam (coarse) (60% sand, 10% silt and 30% clay)	Peat soil (Contain 60–80% fiber)	Sandy clay loam (45% sand, 25% silt and 30% clay)	Sandy clay loam (48% sand, 23% silt and 29% clay)	Silty clay loam (57.2% silt, 17.8% sand and 25% clay)
pH	4.2–6.5	3.5–4.5	5.8–6.7	5.5–6.1	5.5–6.5
CEC (meq/100 g)	20.4	70–80	15.7	11.8	14.3
Total Ca content (mg/kg)	452	3000	2240	1760	1140
Total K content (mg/kg)	67	160	346	383	8235
Organic matter (%)	7.97	65	8.39	5.59	2.25

2.6.2. Tapioca and sweet potato roots samples

The root samples were washed with clean water until free from soil and foreign media. With the study focusing on ingestion of roots, only the edible parts were prepared for analysis, the peels of the samples were removed. The washing procedure was of limited duration as a protracted procedure could result in the soluble radioactive particles being washed away. The samples were then chopped into small homogenized pieces and oven dried separately at 80 °C until a constant dry weight was obtained. The items were then ground into small particles to obtain uniformity and then 500 g of each sample was sealed into a Marinelli beaker; the same procedure was then followed as for soil samples.

2.7. Measurement of radioactivity

The activity concentrations of ^{226}Ra, ^{232}Th, ^{88}Y and ^{40}K in the samples were determined using a p-type coaxial HPGe γ-ray spectrometer (ORTEC; GEM-25; serial no. 46-TP22121A; 57.5 mm crystal diameter; 51.5 mm thickness; +2800 V operating voltage) having a relative efficiency of 28.2% and an energy resolution of 1.67 keV-FWHM at the 1332.5 keV peak of ^{60}Co. To reduce the external gamma-ray background in the measured spectrum, a cylindrical lead shield with a fixed bottom and a movable cover shielded the detector. The HPGe detector was connected to a 16 k MCA for data acquisition. The γ-rays emitted from the samples were analyzed by Gamma Vision 5.0 software (EG&G Ortec). A cylindrical multi-nuclide γ-ray source ((^{241}Am (59.541 keV), ^{109}Cd (88.040 keV), ^{57}Co (122.061 keV; 136.474 keV), ^{203}Hg (279.1952 keV), ^{113}Sn (391.698 keV), ^{85}Sr (514.007 keV), ^{137}Cs (661.657 keV), ^{88}Y (898.042keV; 1836.063 keV), and ^{60}Co (1173.228 keV; 1332.492 keV)) having homogeneously distributed activity with the same volume and shape as the Merinelli beakers that contain the sample (so that the detection geometry remained the same) (Amin et al., 2013) was used for detector energy calibration and the absolute photopeak efficiency evaluation. The measured detection efficiencies were fitted by using a polynomial fitting function as described in Khandaker et al. (2013), and the fitted efficiencies were used in activity determination of the samples. The samples were counted for a sufficiently long time (86,000 s) to decrease the counting error, and background counts for the same counting time were deducted to obtain the net activity. The activity concentrations of ^{226}Ra, ^{232}Th, ^{88}Y and ^{40}K radionuclides for each sample were assessed using the equation available in Khandaker et al. (2013). The gamma-ray lines at 351.93 keV (35.6%) from ^{214}Pb, 609.32 keV (45.49%) from ^{214}Bi were used to determine the activity concentrations of ^{226}Ra; the gamma-ray lines at 238.63 keV (46.6%) from ^{212}Pb, 583.19 keV (85.0%) from ^{208}Tl were used to determine the activity concentrations of ^{232}Th; while the 898.042 keV (93.7%) and 1836.063 keV (99.2%) transitions were used to determine the activity concentrations of ^{88}Y. The single line 1460.822 keV (10.66%) was used to determine the

activity concentrations of ^{40}K. A typical γ-ray spectrum counted for 86,400 s is shown in Fig. 2. Following Malain et al. (2012), a weighted mean analysis was used to obtain a final activity concentration by combining the different values obtained for the individual gamma-ray energy values. Such a procedure (i.e., weighted mean approach) considerably reduces the uncertainty of the derived values compared to the use of a single transition (Malain et al., 2012).

2.8. Transfer factor (TF)

The transfer mechanism of radionuclides, represented by TF, is widely used to describe the soil-to-plant transfer of radionuclides through plant roots. The concentration of a nuclide in a plant or plant part, C_i^p (in Bq kg^{-1}, dry weight), is assumed to be linearly related to its concentration in soil within the rooting zone, C_i^s (also in Bq kg^{-1}, dry weight), and the ratio of the concentrations is the TF. Note that TFs were computed using the activities of each plant sample and the corresponding soil sample. Because TF values are assumed to be lognormally distributed, the summations presented here are geometric means (GMs).

2.9. Statistical analysis

Statistical analysis of data was performed using SPSS 21 software (IBM Corporation, Armonk, NY, USA). Data of activity concentrations, TFs and other radiological indicators in tapioca and sweet potato among the four different locations were compared employing one way analysis of variance (ANOVA). Tukey's HSD Post-hoc test was applied to determine statistically significant differences among individual means at $p < 0.05$.
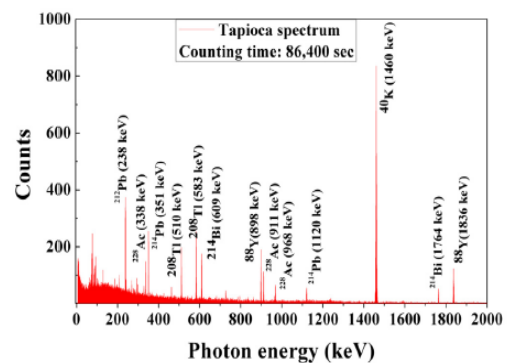

Fig. 2. Typical γ-ray spectrum of the tapioca sample collected for 86,400 s.

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Table 2
Activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y in soil, tapioca and sweet potato samples.

Sampling location	Parameters	Soil				Tapioca				Sweet potato		
		^{226}R	^{232}Th	^{40}K	^{88}Y	^{226}R	^{232}Th	^{40}K	^{88}Y	^{226}R	^{232}Th	^{40}K
Puchong (mining soil)	Range	99.6–128	7.2–11.1	657–903	17.5–38.6	116–141	1.5–4.4	732–934	4.4–11.6	49.6–81.5	5.3–10.4	1985–3127
(4)	AM	114	8.5	745	28.6	127	3	844	7	67	8	2483
Chemor (peat soil area)	Range	72.7–112	33.6–71.6	385–441	8.8–15.2	78.7–119	9.7–25.2	756–910	–	59.8–91.6	12.3–23.4	259–303
(5)	AM	91	51	409	12	96	17	838	–	76	17	282
Lembah Beringin	Range	122–142	17.7–32.6	492–546	14.6–23.6	93.3–117	35–51	49.7–63.8	9.6–13.8	–	–	–
(lateritic soil) (3)	AM	132	25	523	19	103	42	56	12	–	–	–
Batang Berjuntai	Range	92–114	78.7–102	967–1023	144–165	72–98	35.3–53.4	779–812	–	–	–	–
(lateritic soil) (4)	AM	106	91	992	155	87	43	796	–	–	–	–

The values in parenthesis indicate the number of samples analyzed. AM denotes arithmetic mean.

3. Results and discussion

3.1. Radionuclide activity concentrations

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y radionuclides in soils, tapioca and sweet potato roots collected from the studied locations are presented in Table 2, reported on a dry weight basis. Soil samples from all study locations except from the experimental field contained the fallout (artificial) ^{88}Y radionuclide. It is seen from Table 2 that only tapioca roots had measurable amounts of ^{88}Y from mining soil (Puchong) and laterite soil (Lembah Beringin).

The ^{226}Ra activity in tapioca and sweet potato in Puchong, which is the former tin mining area, varied in the ranges of 116–141 Bq kg^{-1} and 49.6–81.5 Bq kg^{-1} with AM values of 127 and 67 Bq kg^{-1}, respectively. Statistical analysis (ANOVA) shows that there is a significant variation ($p < 0.05$) of concentrations of ^{226}Ra in tapioca among the studied locations except Lembah Beringin (laetrile soil). The ^{232}Th activity of tapioca in all locations was comparatively lower than the ^{226}Ra. However, these differences were not found statistically significant ($p > 0.05$).

3.2. ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y transfer factors

The GM value of TF (soil-to-tapioca) for ^{226}Ra in Puchong was slightly higher (1.12) than the other locations and close to that of Chemor (1.0), the peat soil area. However, statistically, no significant variation ($p > 0.05$) of TF (soil-to-tapioca) for ^{226}Ra was found among the studied areas. On the other hand, GM value of TF (soil-to-sweet potato) for ^{226}Ra in Chemor was found a little bit higher (0.82) than that of Puchong (0.58). This difference was also found statistically insignificant ($p > 0.05$) by ANOVA analysis. The IAEA

has compiled soil-to-plant TFs (loam soil) for Ra in root crops of tropical environments and reported the minimum and maximum values are 1.2×10^{-3} and 2.2×10^{-1} with a GM value of 1.1×10^{-2} and GSD (geometric standard deviation) of 4.9, and tubers values (all soil) are 2.6×10^{-4} and 1.9×10^{-1} with a GM value of 1.9×10^{-2} and GSD of 3.8 (IAEA-TECDOC-1616, 2009). The estimated TF values for ^{226}Ra for tapioca and sweet potato in the present study are very much higher than the range of values reported by the IAEA.

The soil-to-tapioca and soil-to-sweet potato TFs of ^{232}Th varied significantly ($p < 0.05$) among the studied locations. The soil-to-tapioca TFs of ^{232}Th were highest (1.7) in Lembah Beringin, and lowest (0.28) in Chemor (Table 3). The soil-to-sweet potato TFs of ^{232}Th was higher (0.92) in Puchong than for the Chemor region (0.33). The observed values of TFs are beyond the range reported by the IAEA (loam soil) for ^{232}Th in root crops (roots), of 9.0×10^{-6} to 3.9×10^{-5}, with a GM value of 1.9×10^{-5} and GSD of 1.7 and root crops (tubers), of 2.9×10^{-6} to 3.5×10^{-5}, with a GM value of 8.9×10^{-6} and GSD of 2.6 (IAEA-TECDOC-1616, 2009). In the IAEA-TECDOC-1616, Velasco et al. (2009) reported mean TFs on dry weight basis (soil depth considered is upper 20 cm) by compiling data from six different countries having tropical environments (Australia, Brazil, India, Indonesia, Marshall Islands and Vietnam). Velasco et al. also published the similar report in Journal of Environmental Radioactivity. It has not been mentioned in the report whether the data was for only edible fraction of root vegetables or including peel. Since most of the published articles contain data on edible part of the root vegetables, we therefore assumed that the data was based on only for the edible fraction. In this study, TFs on dry weight basis are calculated using only edible part of root vegetables. The soil texture in the present study locations are mainly sandy clay loam, silty clay loam and peat soil, whereas IAEA reported TFs for ^{226}Ra and ^{232}Th for root crops (roots such as tapioca)

Table 3
Soil-to-tapioca and sweet potato transfer factors for ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y (TFs).

Sampling location	Parameters	Soil to tapioca				Soil to sweet potato		
		^{226}Ra	^{232}Th	^{40}K	^{88}Y	^{226}Ra	^{232}Th	^{40}K
Puchong (mining soil) (4)	Range	0.99–1.28	0.15–0.61	1.10–1.29	0.16–0.66	0.50–0.74	0.73–1.4	3.0–3.5
	GM	1.12	0.30	1.14	0.25	0.58	0.92	3.3
	GSD	1.13	1.95	1.10	1.93	1.19	1.34	1.08
Chemor (peat soil area) (5)	Range	0.75–1.4	0.14–0.73	1.7–2.2		0.61–1.05	0.22–0.50	0.61–0.75
	GM	1.0	0.28	2.0		0.82	0.33	0.69
	GSD	1.28	1.90	1.11		1.22	1.43	1.08
Lembah Beringin (lateritic soil) (3)	Range	0.66–0.87	1.6–2.0	0.10–0.12	0.49–0.95			
	GM	0.77	1.7	0.11	0.61			
	GSD	1.16	1.14	1.11	1.46			
Batang Berjuntai (lateritic soil) (4)	Range	0.66–1.0	0.39–0.58	0.79–0.84				
	GM	0.81	0.47	0.80				
	GSD	1.19	1.20	1.03				

The values in parenthesis indicate the number of samples analyzed. GM and GSD denotes geometric mean and geometric standard deviation respectively.

were based on loam soil and tuber crops (tubers such as sweet potato) were based on all types of soil and loamy soil, respectively. The hydrological conditions within the soil, soil fertility, the duration of the vegetative period, cultivation process may not same of our studied farmland and those countries farmlands from where IAEA obtained the data. The above parameters may change soil properties or lead to redistribution of radionuclides in the rooting zone, and consequently change of radionuclides uptake in root crops.

While the activity concentration of [88]Y was found higher in the Puchong soil than that of the Lembah Beringin region, the [88]Y TF (soil-to-tapioca) for Lembah Beringin was found higher than that of the Puchong area, i.e., tapioca roots in lateritic-loamy soil areas take up more [88]Y than the former tin mining region. But statistically, no significant difference of [88]Y TF (soil-to-tapioca) was observed ($p > 0.05$).

Soil-to-tapioca and soil-to-sweet potato TFs (geometric mean) for [40]K were found to be significantly ($p < 0.05$) higher than that for the other nuclides in all locations except the TF of tapioca in Lembah Beringin (Table 3). The high value TFs of [40]K was considered to be due to the fact that potassium is important in fertilizing the crop, also playing a vital role in the ability of the plant adapting to environmental pressures. However, potassium remains in homeostatic equilibrium in the plant and is readily adapted by the plants. Therefore, as compared to uranium and thorium, potassium shows the highest TF.

3.3. Transfer factors of the experimental tapioca field

The activity concentrations for [226]Ra, [232]Th and [40]K in soil and tapioca samples collected at various times are presented in Table 4; [88]Y was not detected. The TFs for all radionuclides increased with time. Significant difference of TFs for [226]Ra and [232]Th with time was observed between 3rd months and 6th months ($p < 0.001$). These results indicate that the roots accumulate radionuclides faster than they do biomass, so the concentration and TF increases with time.

3.4. Effect of soil parameters on TFs

Soil-to-plant transfer of naturally occurring radionuclides is largely affected by the soil physiochemical properties such as cation exchange capacity (CEC), potassium (K), calcium (Ca), organic matter content etc. (Pulhari et al., 2004; Shanthi et al., 2012). The soil physical and chemical properties of the studied firm lands are presented in Table 1. The pH values of soil of all the studied area indicate that the soil is acidic in nature. Generally, acidic soil uptake more radionuclides than alkaline soil, especially [226]Ra cannot be avoided in acidic soil. In the present study, little variation of [226]Ra TF was observed over the pH range of 3.5–6.7 among the study regions. The highest TF of [232]Th was found in the pH range of 5.8–6.7 in Lembah Beringin areas for tapioca. Conversely, highest TF of [40]K was in Puchong region for sweet potato (former tin mining area) whose soil pH is 4.2–6.5. It is seen from Tables 1 and 3 that CEC regulates the transfer of radionuclide's mainly [226]Ra and [40]K from soil-to-tapioca. It is observed from Tables 1 and 3 that Ca content varies in different study locations and found no linear correlation between Ca content and TF of [226]Ra. Similar conclusion can be drawn in case of K content in soil and its transfer to root vegetables. The organic matter content of the soil varies between 2.25 and 65% among the studied region. It is found highest in peat soil and lowest in experimental field and other study regions contain nearly same values of organic matter. There is no positive correlation observed between organic matter content and TF of [226]Ra and [232]Th radionuclides. Experimental field contain lowest organic matter but found highest TF.

3.5. Radiological impact

3.5.1. Daily intake

Whether man-made or natural in origin, radioactive material transfers to the human body through the food chain, in the same way that non-radioactive material does. The degree of risk to human health depends on the type of radionuclides and the duration of exposure. Considering the most recent annual production figures for tapioca (33,200 M tonnes) and sweet potato (26,600 M tonnes) in Malaysia (Malaysian Department of Agriculture; www.doa.gov.my/c/document_library, 2012) and total population (of 15 years of age and above) of 20.62 million (www.indexmundi.com/malaysia/demograpics_profile.html, 2013), the daily intake of radionuclides due to the consumption of tapioca and sweet potato were estimated using the following formula (Alam and Mohamed, 2011; Khandaker et al., 2013):

Table 4
Activity concentration (Bq kg^{-1}) of [226]Ra, [232]Th and [40]K in soil and tapioca in the experimental field and their corresponding transfer factors (TFs).

Harvesting time	Sample type	Parameters	Radioactivity concentration (Bq kg^{-1})			Transfer factors (TFs)		
			[226]Ra	[232]Th	[40]K	[226]Ra	[232]Th	[40]K
3rd Months	Soil (4)	Range	5.2–8.4	12.5–27.4	20.4–34.6			
		AM	6.6	18.7	26.7			
	Tapioca (4)	Range	7.3–15.6	32.3–54.4	37.7–61.5	1.0–2.2	1.7–3.3	1.4–2.7
		GM	10.3	41.3	49.3	1.6	2.3	1.9
		GSD				1.5	1.4	1.3
4th Months	Soil (4)	Range	5.2–8.4	12.5–27.4	20.4–34.6			
		AM	6.6	18.7	26.7			
	Tapioca (4)	Range	8.5–17.6	27.7–48.5	56.7–78.6	1.3–3.1	1.7–3.3	1.7–3.2
		GM	11.8	36.7	66.4	1.8	2.0	2.5
		GSD				1.4	1.4	1.3
5th Months	Soil (4)	Range	5.2–8.4	12.5–27.4	20.4–34.6			
		AM	6.6	18.7	26.7			
	Tapioca (4)	Range	14.6–30.8	38.6–56.4	65.4–96.6	1.7–4.3	1.8–3.5	1.9–4.7
		GM	20.6	46.6	77.7	3.2	2.6	3.0
		GSD				1.5	1.3	1.5
6th Months	Soil (4)	Range	5.2–8.4	12.5–27.4	20.4–34.6			
		AM	6.6	18.7	26.7			
	Tapioca (4)	Range	28.6–55.4	135–204	63.4–88.6	4–7.4	7.4–10.8	1.8–3.8
		GM	38.5	165.2	74.4	5.9	9.2	2.8
		GSD				1.3	1.2	1.4

The values in parenthesis indicate the number of samples analyzed. AM, GM and GSD denotes arithmetic mean, geometric mean and geometric standard deviation respectively.

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Table 5
Average daily intake of ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y and annual committed effective dose due to consumption of tapioca and sweet potato.

Sampling location	Sample type	Daily intake of radionuclides D_{int} (Bq d^{-1})				Annual committed effective dose E_{eff} (μSv y^{-1})			
		^{226}R	^{232}Th	^{40}K	^{88}Y	^{226}R	^{232}Th	^{40}K	^{88}Y
Puchong	Tapioca (4)	0.53	0.01	3.5	0.03	54.6	1.0	8	0.01
	Sweet potato (4)	0.23	0.03	8.3		23	2.2	18.9	
Chemor	Tapioca (5)	0.4	0.07	3.5		41	6	7.9	
	Sweet potato (5)	0.25	0.07	0.9		26	4.8	2.1	
Lembah Beringin	Tapioca (3)	0.43	0.18	0.23	0.05	44	14.8	0.53	0.02
Batang Berjuntai	Tapioca (4)	0.37	0.18	3.3		37.3	15.2	7.5	

The values in parenthesis indicate the number of samples analyzed.

$$D_{int} = \frac{A_c \times A_p \times E_r}{M_p \times Y_d} \qquad (1)$$

where, D_{int} is the daily individual intake of radionuclides (in Bq), A_c is the activity concentration of radionuclides (in Bq kg^{-1}), A_p is the annual production, M_p is the population of Malaysia, Y_d is the days in a year and E_r is the fraction of edible part of root vegetable, the values of E_r considered 95% based on local eating habits in Malaysia. The estimated values of daily intake per capita for tapioca and sweet potato together with statistical analyses are presented in Table 5. It is seen that daily intake of ^{40}K is significantly ($p < 0.001$) higher than the other radionuclides. The concentration of fallout radionuclide ^{88}Y is very low, albeit being part of the daily intake of tapioca. Sweet potato did not contain detectable concentrations of this nuclide.

3.5.2. Ingestion dose to human

The annual effective ingestion dose to an individual due to the intake of ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y from the consumption of tapioca and sweet potato has been computed using the Equation (2) (Alam and Mohamed, 2011; Khandaker et al., 2013), and presented in Table 5:

$$E_{eff} = A_c \times A_{if} \times D_{cf} \times F_c \qquad (2)$$

where, E_{eff} is the annual individual effective dose (in μSv y^{-1}), A_c is the activity concentration of radionuclides (in Bq kg^{-1}), A_{if} is the annual intake of food (kg y^{-1}); as mentioned in section 3.5.1, the local consumption rates of tapioca and sweet potato in Malaysia in 2011 (ignoring any export and wastage) was 1.6 kg y^{-1} and 1.3 kg y^{-1} respectively, D_{cf} is the ingestion dose conversion factor for the radionuclides of interest (2.8 × 10^{-7} Sv Bq^{-1} for ^{226}Ra, 2.3 × 10^{-7} Sv Bq^{-1} for ^{232}Th, 6.2 × 10^{-9} Sv Bq^{-1} for ^{40}K and 1.3 × 10^{-9} Sv Bq^{-1} for ^{88}Y) (IAEA, 2011), and F_c is the actual fraction of edible part of tapioca and sweet potato (~95%). Statistical analysis (ANOVA) shows that the variation of annual effective dose for tapioca from the intake of ^{226}Ra was significantly ($p < 0.05$) higher than other detected radionuclides in all studied locations. One possible reason is that the ingestion dose conversion factor of ^{226}Ra is very much higher than other studied radionuclides. Another reason may be attributed to the fact that ^{226}Ra has a high exchange affinity at the regular exchange sites, the soil and tapioca absorbing ^{226}Ra at a high rate from soil. Average worldwide effective dose from the ingestion of ^{226}Ra and ^{232}Th is 120 μSv y^{-1}, for ^{40}K 170 μSv y^{-1}, with a total annual dose of 290 μSv y^{-1} (UNSCEAR, 2000). The annual effective dose from the ingestion of tapioca and sweet potato were found very much lower than the world average value for all study areas.

4. Conclusions

We provide the first report on radionuclide TFs for soil-to-tapioca, soil-to-sweet potato and ingestion dose to humans based on natural field conditions in peninsular Malaysia. The TFs of ^{226}Ra, ^{232}Th, ^{40}K and ^{88}Y for soil to tapioca varied from 0.77 to 1.12, 0.28 to 1.7, 0.11 to 2.0 and 0.25 to 0.61, respectively. Soil to sweet potato TFs for ^{226}Ra, ^{232}Th and ^{40}K varied from 0.58 to 0.82, 0.33 to 0.92 and 0.69 to 3.3, respectively. It is observed that TFs for tapioca are comparatively higher than for sweet potato. No ^{88}Y was detected in sweet potato, but a small amount was found in tapioca collected from Puchong (a former tin mining area) and Lembah Beringin (laterite soil).

Based on the findings from the experimental field in Dengkil (for a duration of vegetation growth of 6 months whereas measurements were started from 3rd month), the soil-to-tapioca TF for ^{226}Ra, ^{232}Th and ^{40}K increased from 1.6 to 5.9, 2.0 to 9.2 and 1.9 to 3.0, respectively with time. In most cases, the uptake of radionuclides at the middle or late growth stages produced higher TFs than those at the early growth stage.

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3.2.4   Published paper 4

MEASUREMENT OF RADIOACTIVITY AND HEAVY METAL LEVELS IN EDIBLE VEGETABLES AND THEIR IMPACT ON KUALA SELANGOR COMMUNITIES OF PENINSULAR MALAYSIA

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Vegetable is an essential daily diet item for the people of Malaysia. This work addressed the radiation and heavy metal exposure scenarios through the consumption of vegetables. Kuala Selangor is located in Sungai Selangor estuary in the west coast of Peninsular Malaysia, which is susceptible to pollution load due to the presence of large-scale industrial and human activities. Radioactivity and heavy metals level in human diet is of particular concern for the assessment of possible radiological and chemical hazards to human health. Therefore, a comprehensive study was carried out to determine the radioactivity levels (^{226}Ra, ^{228}Ra and ^{40}K) and heavy metal concentrations (Cr, As, Cd, Mn, Mg, Al, Sr, Rb, Sb, Ba, Hg, Fe, Ni, Zn, Cu, Bi and Pb) in 10 varieties of vegetable collected from different farmlands in Kuala Selangor region. The committed doses for ^{226}Ra, ^{228}Ra and ^{40}K due to consumption of vegetables were found 16.6 ± 1.3, 23.6 ± 1.7 and 58 ± 5 μSv y^{-1}, respectively, with a total of 98 ± 8 μSv y^{-1}. This dose imposes no significant threat to human health. The estimated cancer risk shows that probability of increase in cancer risk from daily intake of vegetables is only a minor fraction of International Commission on Radiological Protection values. The concentrations of heavy metal were below the daily intake recommended by the international organisations.

INTRODUCTION

Natural radioactivity and heavy metals are always present in world's environmental media since it was created. These components may be enhanced by human activities like agricultural inputs, urbanisation, uses of fossil fuels, industrial activities, dumping of radioactive wastes, disposal of industrial effluents, etc.[1–4]. Food is the major source of ingestion of radioactivity and heavy metals in humans. Particularly all categories of vegetables are produced and commonly consumed (particularly the root, fruit and leafy vegetables) by the Kuala Selangor (latitude 3°20'N, longitude 101°30'E) communities. The primordial radionuclide, such as ^{40}K, and the radionuclides from the ^{238}U and ^{232}Th decay series are the most common radionuclides in all environmental media including vegetable, and ingestion exposure dose mainly results from these radionuclides[5]. In the terrestrial ecosystem, soil is the main reservoir for radioactivity contamination. It is, therefore, important to study the spatial distribution of natural radioactivity and related radiation exposures through specific land-produced foodstuffs[6]. If elevated levels of these radionuclides accumulate within the body, then the organs will be subject to continuous exposure from the emitted photons and particulate forms of radiation, creating radiation damage, biochemical and morphological changes[7]. As such, assessment of radiation level in the fibrous foodstuffs finds great importance in addressing the associated health concerns.

Heavy metals can be toxic for humans when they are not metabolised by the body and accumulate in the soft tissues. Heavy metals (Ni, Ba, Hg, Pb, Cd, Al, Se, Sb, Bi, etc.) consist of a group of minerals that have no known function in the body and, in fact, are harmful. Minerals (such as Zn, Cu, Fe, Mn, Se and V) that are needed in minute quantities for human body become toxic if they exist in greater amounts. Excessive content of metals beyond maximum permissible level leads to a variety of cardiovascular, neurological and bone diseases as well as mutagenic effects in lungs, kidneys, liver and other vital organs[8].

As far as the authors' concern, no representative data of radioactivity and heavy metal levels in vegetables in the studied region are available that estimates the various hazard indices. Therefore, this study was carried out to investigate the radioactivity (^{226}Ra, ^{228}Ra and ^{40}K) and heavy metal (Mg, Al, Cr, Mn, Fe, Ni, Cu, Zn, Cd, As, Se, Rb, Sr, Sb, Ba, Bi, Hg and Pb) levels in the most commonly consumed vegetables among the inhabitants of Kuala Selangor in order to make a preliminary estimation on intake doses of radionuclides and toxic metals for this population.

MATERIALS AND METHODS

Vegetables sampling, pretreatment and analysis

Almost all categories of vegetables (root vegetables: tapioca, sweet potato, yam; fruit vegetables: brinjal, long bean, lady's finger, winged bean, cucumber and

leafy vegetables: water spinach, spinach) produced and consumed by the inhabitants were collected (a total of 45 samples from 10 different vegetables) and prepared as described elsewhere[9, 10]. The activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in the samples were determined using HPGe γ-ray spectrometer (ORTEC, relative efficiency of 28.2 % and energy resolution of 1.67 keV FWHM at the 1332.5 keV peak of ^{60}Co). Detector energy calibration and efficiency evaluation were carried out according to Refs.[9, 10]. The activity concentration of ^{226}Ra was measured using the characteristic γ-lines of 351.93 keV (35.6 %) from ^{214}Pb and 609.32 keV (45.49 %) from ^{214}Bi, whereas the concentration of ^{228}Ra was determined via the γ-ray lines of 583.19 keV (85.0 %) from ^{208}Tl and 911.21 keV (25.8 %) from ^{228}Ac. The single transition 1460.822 keV (10.66 %) was used to determine the activity concentrations of ^{40}K. Minimum detectable activity concentration (MDC) for the radionuclides of interest was calculated using the equation available in Ref.[11] as 0.35, 0.12 and 2.2 Bq kg^{-1}, respectively, for ^{226}Ra, ^{228}Ra and ^{40}K.

Around 0.5 g of powdered sample from each vegetable category was digested by heating in microwave digestion system (Multi-wave 3000, PerkinElmer, USA) using a mixture containing 5 ml of 65 % concentrated HNO$_3$ (SpectrosoL grade) and 3 ml of H$_2$O$_2$ (30 %) for 10 min. Elemental (Mg, Al, Cr, Mn, Ni, Cu, Zn, Rb, Sr, Sb, Ba, Bi, Hg and Pb) analyses were conducted by inductively coupled plasma mass spectrometry (ICP-MS) (Agilent Technologies 7500 Series, USA). Calibration of the ICP-MS was performed using multi-element calibration standard (Agilent Technologies, USA, Part No. 8500-6940). Detection limits (3σ method) of ICP-MS for elements Na, Mg, Al, Ca, Cr, Mn, Ni, Cu, Zn, Rb and Sr was 0.0002 mg kg^{-1}, whereas for elements Fe, Cd, Ba and Pb was 0.0001 mg kg^{-1}. Daily intake equivalent to a 70-kg adult as well as the daily intake of body weight (bw) were estimated according to Refs.[3, 4].

RESULTS AND DISCUSSION

Radionuclide activity concentrations

Activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in the vegetable samples are presented in Table 1. The mean radioactivity concentrations in different varieties of vegetables analysed ranged from 0.5 ± 0.1 to 2.8 ± 2.0 Bq kg^{-1} for ^{226}Ra, 0.3 ± 0.1 to 1.8 ± 2.2 Bq kg^{-1} for ^{228}Ra and 63 ± 18 to 427 ± 27 Bq kg^{-1} for ^{40}K. The highest concentrations were observed in water spinach samples for ^{226}Ra (2.8 ± 2.0 Bq kg^{-1}) and ^{40}K (427 ± 27 Bq kg^{-1}) and in sweet potato for ^{228}Ra (1.8 ± 2.2 Bq kg^{-1}). The lowest ^{226}Ra, ^{228}Ra and ^{40}K concentrations were observed in long bean (0.5 ± 0.1 Bq kg^{-1}), cucumber (0.3 ± 0.1 Bq kg^{-1}) and tapioca (63 ± 18 Bq kg^{-1}), respectively. For all

varieties of vegetables, variability in radionuclide concentration was observed even within the same types of vegetable. This can be caused by differences in the chemical and physical properties of the different vegetables farm soils, individual chemical properties of the radionuclides, irrigation, use of insecticides and herbicides, plant species, the degree of preparation of the vegetable sample, such as peeling and washing or even due to the use of phosphate fertilisers[2]. However, activity concentration of ^{226}Ra in this work is about two times higher than that reported by Canbazoglu and Dogru[5] in Turkey (0.11–0.99 Bq kg^{-1}) and by Santos et al.[2] in Brazil (3.4–805 mBq kg^{-1}) and lower concentrations with a range of 3.4 ± 1.8–10.5 ± 4.7 Bq kg^{-1} reported by Awudu et al.[1] in Ghana. Conversely, the activity levels detected for ^{228}Ra are much lower than the values 6.1 ± 2–12.3 ± 3.1 Bq kg^{-1} stated by Awudu et al.[1] in Ghana.

Compared with radium nuclides (^{226}Ra and ^{228}Ra) concentration, potassium nuclide (^{40}K) concentrations were found to be very high indicative of the fundamental nutrient status of potassium for the plants, and this may be attributed to poor migration characteristics of radium from the substrate to the plant in the concerned environment[1]. Radioactivity of ^{40}K for all samples lies within the world range reported by Maul and O'Haraas, 40–240 Bq kg^{-1}[1] except for water spinach and spinach. On the other hand, potassium may concentrate in leaves more than any other parts of the plant. The present results of ^{40}K are within the range of 39−489 Bq kg^{-1} obtained from various species of natural vegetation in Egypt[12] and the range of 87.8 ± 8.4–369 ± 19 Bq kg^{-1} found in different crops in Ghana[1], higher than the range of 2.1–44.8 Bq kg^{-1} reported from Turkey[5], and lower than the authors' previous works for tapioca and sweet potato (56–2483 Bq kg^{-1}) in tin mining and some other areas of Malaysia[5].

Annual intake of radionuclides and committed effective dose

Annual intake of radionuclides and committed effective dose are presented in Table 1. Activity concentration (Bq kg^{-1}) for each radionuclide from each vegetable sample was multiplied by their respective consumption rate (kg y^{-1}) to obtain the annual activity intake. The committed effective dose was then determined by multiplying annual activity intake value and dose conversion coefficients for adult taken from International Atomic Energy Agency[13].

The annual intake of each radionuclide from different varieties of vegetables was estimated as 1.3 ± 0.1−11.8 ± 0.8 Bq with an arithmetic mean (AM) of 6 ± 0.4 Bq for ^{226}Ra; 1.2 ± 0.1−10.3 ± 0.7 Bq with an AM of 3.4 ± 0.2 Bq for ^{228}Ra and 185 ± 9−1794 ± 90 Bq with an AM of 930 ± 46 Bq

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Table 1. Activity concentration (Bq kg⁻¹ dry weight) of ^{226}Ra, ^{228}Ra and ^{40}K, annual intake and committed effective dose due to consumption of vegetable: AM ± SD and (range).

	Tapioca [7]	Sweet potato [6]	Yam [5]	Brinjal [4]	Long bean [4]	Lady's finger [4]	Winged bean [3]	Cucumber [4]	Water spinach [4]	Spinach [4]
Radioactivity concentration *(Bq kg⁻¹)*										
^{226}Ra	0.84 ± 0.4	1.1 ± 0.4	0.73 ± 0.3	1.1 ± 0.6	0.5 ± 0.1	0.61 ± 0.2	1.3 ± 0.8	0.83 ± 0.2	2.8 ± 2.0	1.6 ± 0.4
	(0.38–1.5)	(0.67–1.6)	(0.37–1)	(0.46–1.6)	(0.44–0.6)	(0.44–0.9)	(0.47–2)	(0.69–1.0)	(1.0–5)	(1.1–2.0)
^{228}Ra	0.31 ± 0.2	1.8 ± 2.2	0.72 ± 0.5	0.41 ± 0.4	0.4 ± 0.3	0.5 ± 0.3	0.45 ± 0.4	0.3 ± 0.1	0.7 ± 0.2	1.2 ± 1.0
	(0.15–0.77)	(0.24–5)	(0.32–1.7)	(0.2–1.1)	(0.14–0.87)	(0.20–0.98)	(0.16–0.88)	(0.24–0.37)	(0.56–0.93)	(0.48–2.6)
^{40}K	63 ± 18	102 ± 24	101 ± 25	184 ± 75	177 ± 55	202 ± 103	215 ± 24	152 ± 74	427 ± 27	208 ± 174
	(43–86)	(62–133)	(69–134)	(72–226)	(95–212)	(50–276)	(193–240)	(90–259)	(394–453)	(90–465)
Annual intake *(Bq)*										
^{226}Ra	9.8 ± 0.7	6.3 ± 0.4	1.3 ± 0.1	4.8 ± 0.3	2.6 ± 0.2	4.0 ± 0.3	3.5 ± 0.2	8.8 ± 0.6	11.8 ± 0.8	6.2 ± 0.4
	(4.3–17.5)	(4–9.6)	(0.7–2)	(2.0–7.2)	(2.2–3.0)	(3–5.8)	(1.2–5.5)	(7.3–11.0)	(4.3–20.6)	(4.2–7.3)
^{228}Ra	3.7 ± 0.3	10.3 ± 0.7	1.3 ± 0.1	1.8 ± 0.1	2.0 ± 0.2	3.3 ± 0.3	1.2 ± 0.1	3.2 ± 0.2	3 ± 0.3	4.4 ± 0.2
	(1.7–9.0)	(1.4–30)	(0.6–3.0)	(0.5–4.7)	(0.7–4.4)	(1.3–6.4)	(0.4–2.3)	(2.6–4.0)	(2.3–4.0)	(1.8–9.7)
^{40}K	731 ± 37	598 ± 30	185 ± 9	807 ± 40	907 ± 46	1330 ± 61	560 ± 28	1616 ± 81	1794 ± 90	770 ± 39
	(499–999)	(360–775)	(127–245)	(313–988)	(488–1082)	(330–1816)	(502–625)	(957–2743)	(1656–1903)	(334–1722)
Committed effective dose *(μSv y⁻¹)*										
^{226}Ra	2.8 ± 0.3	1.8 ± 0.1	0.4 ± 0.02	1.3 ± 0.1	0.7 ± 0.1	1.1 ± 0.1	1.0 ± 0.1	2.5 ± 0.2	3.3 ± 0.3	1.7 ± 0.2
	(1.2–5.0)	(1.1–2.7)	(0.2–0.6)	(0.6–2.0)	(0.6–0.8)	(0.8–1.6)	(0.3–1.5)	(2.1–3.1)	(1.2–5.8)	(1.2–2.0)
^{228}Ra	2.5 ± 0.2	7.1 ± 0.6	0.9 ± 0.1	1.3 ± 0.1	1.4 ± 0.2	2.3 ± 0.2	0.8 ± 0.1	2.2 ± 0.3	2.0 ± 0.2	3.1 ± 0.3
	(1.2–6.2)	(1.2–20.6)	(0.4–2.1)	(0.4–3.2)	(0.5–3.1)	(0.9–4.4)	(0.3–1.6)	(1.8–2.7)	(1.6–2.7)	(1.2–6.7)
^{40}K	4.5 ± 0.3	3.7 ± 0.4	1.1 ± 0.1	5.0 ± 0.4	5.6 ± 0.5	8.2 ± 0.6	3.5 ± 0.2	10.0 ± 0.7	11 ± 1	4.8 ± 0.3
	(3.1–6.2)	(2.2–4.8)	(0.8–1.5)	(2.0–6.1)	(3.0–6.7)	(2.0–11.3)	(3.1–4.0)	(6–17)	(10.3–11.8)	(2–10.7)

The values in brackets "[ ]" indicate the number of samples analysed. AM and SD denotes arithmetic mean and standard deviation, respectively.

Table 2. Concentrations (mg kg^{-1}) and daily intake (μg day^{-1}) of heavy metals from the most frequently consumed vegetables by Kuala Selangor community: (concentration: AM ± SD), daily intake-bw and (daily intake equivalent to a 70-kg adult).

Heavy metal	Tapioca [11.7]	Sweet potato [5.8]	Yam [1.8]	Brinjal [4.4]	Long bean [5.1]	Lady's finger [6.6]	Winged bean [2.6]	Cucumber [10.6]	Water spinach [4.2]	Spinach [3.7]
Mg	(6.0 ± 0.3), 2.8 (192.9)	(1.8 ± 0.04), 0.4 (28.0)	(3.2 ± 0.3), 0.23 (16.1)	(11.7 ± 0.8), 2.0 (141.0)	(27.5 ± 1.7), 5.5 (385)	(46.2 ± 3.3), 12.0 (8371)	(20.8 ± 2.4), 2.1 (148.6)	(13.6 ± 0.8), 5.6 (395.3)	(17.1 ± 1.4), 2.8 (197.4)	(47.0 ± 9.2), 6.9 (482.8)
Al	(0.11 ± 0.03), 0.05 (3.5)	(0.06 ± 0.001), 0.001 (1.0)	(0.09 ± 0.01), 0.01 (0.47)	(0.18 ± 0.03), 0.03 (2.1)	(0.37 ± 0.08), 0.08 (5.3)	(0.14 ± 0.02), 0.04 (2.6)	(0.20 ± 0.04), 0.04 (2.7)	(58.1 ± 3.2), 24 (1688)	(0.77 ± 0.1), 0.13 (8.9)	(12.3 ± 1.6), 1.9 (135.8)
Cr	ND	ND	ND	(0.0003 ± 0.00005), 0.00004 (0.003)	ND	(0.002 ± 0.0004), 0.0005 (0.03)	ND	(0.006 ± 0.001), 0.002 (0.17)	(0.0003 ± 0.0), 0.00004 (0.003)	(0.006 ± 0.0008), 0.001 (0.06)
Mn	(0.03 ± 0.002), 0.01 (0.9)	(0.012 ± 0.002), 0.003 (0.2)	(0.05 ± 0.007), 0.004 (0.25)	(0.20 ± 0.02), 0.04 (2.5)	(0.62 ± 0.05), 0.12 (8.6)	(0.92 ± 0.1), 0.24 (16.6)	(0.69 ± 0.1), 0.07 (5.0)	(0.22 ± 0.01), 0.09 (6.4)	(2.1 ± 0.3), 0.35 (24.8)	(1.0 ± 0.14), 0.15 (10.3)
Fe	(0.05 ± 0.001), 0.02 (1.5)	(0.028 ± 0.002), 0.006 (0.4)	(0.09 ± 0.005), 0.006 (0.44)	(0.13 ± 0.01), 0.02 (1.6)	(0.45 ± 0.05), 0.09 (6.4)	(0.26 ± 0.03), 0.07 (4.7)	(0.40 ± 0.05), 0.04 (2.9)	(3.8 ± 0.2), 1.6 (111.2)	(0.67 ± 0.09), 0.11 (7.8)	(1.6 ± 0.2), 0.25 (17.6)
Ni	ND	ND	ND	ND	ND	(0.0004 ± 0.0001), 0.0001 (0.01)	(0.001 ± 0.0002), 0.0001 (0.008)	(0.008 ± 0.0006), 0.04 (0.24)	ND	(0.003 ± 0.0005), 0.001 (0.07)
Cu	(0.01 ± 0.002), 0.003 (0.23)	ND	(0.001 ± 0.0),0.0001 (0.005)	(0.04 ± 0.01), 0.008 (0.5)	(0.02 ± 0.004), 0.005 (0.34)	(0.045 ± 0.004), 0.01 (0.8)	(0.03 ± 0.005), 0.004 (0.25)	(0.025 ± 0.001), 0.11 (0.74)	(0.018 ± 0.002), 0.003 (0.21)	(0.06 ± 0.008), 0.01 (0.62)
Zn	(0.1 ± 0.001), 0.03 (2.0)	(0.03 ± 0.001), 0.009 (0.64)	(0.14 ± 0.02), 0.01 (0.7)	(0.12 ± 0.02), 1.4 (0.02)	(0.36 ± 0.07), 0.07 (5.2)	(0.45 ± 0.07), 0.12 (8.1)	(0.36 ± 0.08), 0.04 (2.6)	(0.26 ± 0.04), 0.11 (7.9)	(0.2 ± 0.04), 0.03 (2.3)	(0.59 ± 0.1), 0.09 (6.3)
Rb	(0.05 ± 0.002), 0.02 (1.6)	(0.06 ± 0.008), 0.01 (0.96)	(0.028 ± 0.002), 0.002 (0.14)	(0.039 ± 0.003), 0.007 (0.46)	(0.06 ± 0.004), 0.01 (0.77)	(0.1 ± 0.01), 0.03 (1.9)	(0.07 ± 0.01), 0.01 (0.49)	(0.02 ± 0.004), 0.008 (0.57)	(0.11 ± 0.01), 0.02 (1.3)	(0.13 ± 0.02), 0.02 (1.4)
Sr	(0.02 ± 0.001), 0.01 (0.7)	(0.01 ± 0.0002), 0.002 (0.16)	(0.02 ± 0.001), 0.001 (0.09)	(0.10 ± 0.01), 0.017 (1.2)	(0.15 ± 0.01), 0.03 (2.2)	(0.53 ± 0.06), 0.14 (9.7)	(0.30 ± 0.04), 0.03 (2.1)	(0.02 ± 0.003), 0.008 (0.58)	(0.72 ± 0.08), 0.12 (8.3)	(0.19 ± 0.03), 0.03 (2.0)
Sb	(0.15 ± 0.03), 0.07 (4.9)	(0.08 ± 0.005), 0.02 (1.4)	(0.10 ± 0.005), 0.007 (0.52)	(0.19 ± 0.03), 0.03 (2.4)	(0.38 ± 0.08), 0.08 (5.4)	(0.31 ± 0.05), 0.08 (5.7)	(0.32 ± 0.02), 0.03 (2.3)	(0.43 ± 0.07), 0.18 (12.5)	(0.66 ± 0.1), 0.11 (7.8)	(1.2 ± 0.3), 0.17 (12.2)
Ba	(0.001 ± 0.0002), 0.001 (0.04)	ND	(0.006 ± 0.001), 0.0004 (0.03)	(0.002 ± 0.0004), 0.0004 (0.02)	(0.009 ± 0.001), 0.002 (0.13)	(0.026 ± 0.003), 0.01 (0.7)	(0.008 ± 0.001), 0.001 (0.06)	(0.004 ± 0.0003), 0.002 (0.13)	(0.03 ± 0.004), 0.004 (0.29)	(0.034 ± 0.006), 0.005 (0.35)
Bi	(0.17 ± 0.02), 0.11 (8.0)	(0.24 ± 0.04), 0.013 (8.8)	(0.81 ± 0.2), 0.12 (8.7)	(0.84 ± 0.14), 0.34 (23.5)	(0.33 ± 0.06), 0.15 (10.2)	(0.12 ± 0.02), 0.05 (3.6)	(0.05 ± 0.008), 0.01 (0.64)	(0.21 ± 0.02), 0.09 (6.3)	(0.11 ± 0.01), 0.03 (1.9)	(0.5 ± 0.08), 0.07 (5.1)
Hg	(0.003 ± 0.001), 0.002 (0.11)	(0.003 ± 0.001), 0.001 (0.05)	(0.003 ± 0.001), 0.0002 (0.01)	(0.005 ± 0.001), 0.001 (0.07)	(0.003 ± 0.001), 0.001 (0.05)	(0.003 ± 0.001), 0.001 (0.07)	(0.003 ± 0.001), 0.0003 (0.019)	(0.0019 ± 0.003), 0.001 (0.05)	(0.003 ± 0.0006), 0.0004 (0.03)	(0.002 ± 0.004), 0.0003 (0.02)
Pb	ND	ND	ND	ND	(0.001 ± 0.0002), 0.0003 (0.02)	(0.006 ± 0.001), 0.002 (0.12)	(0.002 ± 0.0003), 0.0003 (0.02)	(0.003 ± 0.0005), 0.001 (0.08)	(0.004 ± 0.001), 0.004 (0.26)	(0.003 ± 0.005), 0.0004 (0.03)

The values in brackets [ ] indicate the consumption rate (kg year^{-1}). ND denotes not detected.

for ^{40}K. The vegetable with the highest contribution to annual intake of radionuclide was found from water spinach for ^{40}K (1794 Bq) and ^{226}Ra (11.8 Bq) followed by sweet potato (10.3 Bq) for ^{228}Ra. Committed effective dose through the consumption of vegetables were $0.4 \pm 0.02 - 3.3 \pm 0.3$ μSv y^{-1} with an AM of 1.7 ± 0.2 μSv y^{-1} (total: 16.6 ± 1.3 μSv y^{-1}) for ^{226}Ra, $0.8 \pm 0.1 - 7.1 \pm 0.6$ μSv y^{-1} with an AM of 2.4 ± 0.3 μSv y^{-1} (total: 23.6 ± 1.7 μSv y^{-1}) for ^{228}Ra and $1.1 \pm 0.1 - 11 \pm 1$ μSv y^{-1} with an AM of 5.8 ± 0.4 μSv y^{-1} (total: 58 ± 5 μSv y^{-1}) for ^{40}K. The average values were lower, but total values were found higher than the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)[14] reported value (6.3 μSv y^{-1} for ^{226}Ra and 11 μSv y^{-1} for ^{228}Ra), and far below than the world average values (120 μSv y^{-1} for ^{226}Ra and ^{228}Ra each and 170 μSv y^{-1} for ^{40}K) reported by UNSCEAR[14]. This study found that adults living in the study region receive a radiation dose of ~98 μSv y^{-1} from vegetable consumption, which is three times lower than the world average value (290 μSv y^{-1}) and presents no significant risk to public health[5, 14]. Among the vegetables, water spinach (17 %), cucumber (15 %) and sweet potato (13 %) are the highest contributors to the intake dose of Kuala Selangor communities, followed by lady's finger (12 %), spinach (10 %), tapioca (10 %), brinjal (8 %), long bean (8 %), winged bean (5 %) and yam (2 %). ^{40}K is the main contributor to the intake dose, with a contribution of about 59 % of total estimated dose, followed by ^{228}Ra (24 %) and ^{226}Ra (17 %). The estimated cancer risk obtained as $9.4 \times 10^{-7} - 8.4 \times 10^{-6}$ for ^{226}Ra, $2.4 \times 10^{-6} - 2.1 \times 10^{-5}$ for ^{228}Ra and $8.1 \times 10^{-6} - 7.8 \times 10^{-5}$ for ^{40}K is below the acceptable lifetime cancer risk of 10^{-3} for radiological risk[15].

Heavy metal concentration and daily intake dose

The results obtained for heavy metal concentrations (AM ± SD) (dry weight) and daily intakes in different varieties of vegetables are shown in Table 2. Lead (Pb) concentrations in different vegetables ranged from 0.001 ± 0.0002 mg kg^{-1} (long bean) to 0.006 ± 0.001 mg kg^{-1} (lady's finger), which is very low to contribute any significant toxic effect. In the present study, no Pb is detected in root vegetables. Cd, As and Se were not detected in any studied vegetables. The heavy metals like Hg ($0.0019 \pm 0.003 - 0.005 \pm 0.001$ mg kg^{-1}), Ba ($0.001 \pm 0.0002 - 0.034 \pm 0.006$ mg kg^{-1}), Rb ($0.02 \pm 0.004 - 0.13 \pm 0.02$ mg kg^{-1}), Sr ($0.01 \pm 0.0002 - 0.72 \pm 0.08$ mg kg^{-1}), Bi ($0.05 \pm 0.008 - 0.84 \pm 0.14$ mg kg^{-1}) and Sb ($0.1 \pm 0.005 - 1.2 \pm 0.3$ mg kg^{-1}) were present in almost all varieties of vegetables, but concentrations are very low to contribute any significant poisonous effect. Among the studied samples, nickel (Ni) and chromium (Cr) were detected in minuscule in fruit (Ni: $0.0004 \pm 0.0001 - 0.008 \pm 0.00006$ mg kg^{-1}; Cr: $0.0003 - 0.006$ mg kg^{-1}) and leafy vegetables (Ni: 0.003 mg kg^{-1}; Cr: $0.0003 \pm 0.00005 - 0.006 \pm 0.0001$ mg kg^{-1}) but not in root vegetables. On the other hand, zinc (Zn), copper (Cu), iron (Fe) and manganese (Mn) were present in moderate quantities in all types of vegetables compared with Ni, Cr, Rb, Sb, Sr, Bi, Ba, Hg and Pb. Fruit vegetables contain higher amount of Zn, Cu, Fe and Mn than the root vegetables. Cr, Mn, Fe, Cu, Zn and Ni are essential metals for human nutrition; however, when consumed in high levels can cause health problems[3]. The concentration of heavy metals showed variations among different vegetables, which can be caused by differences in the chemical and physical properties of the different farm soils, by the industrial effluents or even due to the use of phosphate fertilisers, herbicides, fungicides and pesticides[4].

The average dietary intake (μg day^{-1} bw) of heavy metals was estimated as Mg (4.0), Al (2.64), Cr (0.001), Mn (0.11), Fe (0.22), Ni (0.001), Cu (0.006), Zn (0.053), Rb (0.014), Sr (0.04), Sb (0.08), Ba (0.002), Bi (0.11), Hg (0.0007) and Pb (0.0013). The metal with the highest contribution to daily dietary intake was found from Al (24 μg day^{-1} bw) (cucumber) followed by Mg (12 μg day^{-1} bw) comes from lady's finger. The dietary intake values estimated in

Table 3. Daily intake (mg day^{-1}) of heavy metals reported elsewhere in literature and international organisations.

Metals	Permissible limit	Literature values	References	Metals	Permissible limit	Literature values	References
Mg	350	—	[17]	Rb	20 (μg day^{-1})	—	[21]
Al	7 (PTWI)	3.5, 1.9–12	[4]	Sr	0.6 (R$_f$D)	—	[20]
Cr	1.5 (R$_f$D)	0.023, 0.013–0.085	[4, 8]	Sb	0.86 (μg day^{-1})	—	[19]
Mn	0.5–5.0 (R$_f$D)	7.79, 2.5, 2.2–4.6	[8, 4]	Ba	0.2 (R$_f$D)	—	[18]
Fe	10.0–60.0 (R$_f$D)	22.72	[8]	Bi	0.5	—	[22]
Ni	0.02 (R$_f$D)	0.089, 0.04–0.7	[4, 8]	Hg	5 (μg day^{-1})	—	[16]
Cu	0.9	1.12, 1.54	[22, 4, 8]	Pb	1.7 (PTWI)	0.028, 0.007–0.23	[4]
Zn	15 (R$_f$D)	4.8, 9.0–18	[4, 8]				

PTWI denotes the provisional tolerable weekly intake, and R$_f$D is the reference oral dose.

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the studied vegetables were far below the values reported in literature and the permissible limits recommended by different international agencies[4, 9, 16−22] (Table 3).

CONCLUSIONS

Most perishable vegetables produced and consumed by the inhabitants of Kuala Selangor, Malaysia, were examined in order to assess their radioactivity and heavy metal levels. Committed effective dose received by an adult of the Kuala Selangor communities due to the consumption of vegetables was estimated as 16.6 ± 1.3 μSv y^{-1} for ^{226}Ra, 23.6 ± 1.7 μSv y^{-1} for ^{228}Ra and 58 ± 5 μSv y^{-1} for ^{40}K, and a total of 98 ± 8 μSv y^{-1} in which ^{40}K has the major contribution (59 %) followed by ^{228}Ra (24 %) and ^{226}Ra (17 %). The total dietary intake (μg day^{-1} bw) of heavy metals via the consumption of vegetables was estimated as Mg (40.4), Al (26.4), Cr (0.004), Mn (1.1), Fe (2.21), Ni (0.004), Cu (0.05), Zn (0.53), Rb (0.14), Sr (0.38), Sb (0.79), Ba (0.02), Bi (1.1), Hg (0.007) and Pb (0.008). Present results indicate that vegetables consumed by the Kuala Selangor communities contain radioactivity and heavy metals under the tolerable limits, and that the contribution of the vegetables evaluated to the dietary intake of radionuclides and heavy metals remains beneath the standard suggested by international organisations.

FUNDING

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3.2.5 Published paper 5

RESEARCH ARTICLE

Assessment of Radiation and Heavy Metals Risk due to the Dietary Intake of Marine Fishes (*Rastrelliger kanagurta*) from the Straits of Malacca

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Abstract

The environment of the Straits of Malacca receives pollution as a result of various industrial and anthropogenic sources, making systematic studies crucial in determining the prevailing water quality. Present study concerns concentrations of natural radionuclides and heavy metals in marine fish (*Rastrelliger kanagurta*) collected from the Straits of Malacca, since aquatic stock form an important source of the daily diet of the surrouding populace. Assessment was made of the concentrations of key indicator radionuclides (^{226}Ra, ^{232}Th, ^{40}K) and heavy metals (As, Mn, Fe, Cr, Ni, Zn, Cu, Co, Sr, Al, Hg and Pb) together with various radiation indices linked to the consumption of seafish. The annual effective dose for all detected radionuclides for all study locations has been found to be within UNSCEAR acceptable limits as has the associated life-time cancer risk. The overall contamination of the sampled fish from heavy metals was also found to be within limits of tolerance.

Introduction

While humans are daily exposed to external radiation from cosmic and terrestrial radiation, radionuclides in the sea may more typically contribute to internal exposure via ingestion. Most of the radioactive progeny within the ^{238}U and ^{232}Th natural decay chains are γ-emitters, forming a major source of external exposures [1]. Conversely, some of their decay products such as ^{226}Ra, ^{222}Rn, ^{218}Po, ^{210}Po, ^{224}Ra, ^{220}Rn, ^{212}Bi etc. are alpha emitters, added to which are beta emitters such as ^{234}Pa, ^{214}Bi, ^{228}Ac, ^{212}Bi etc., with ^{40}K (decaying by β) forming yet another means of internal exposure from natural sources. The levels of these can be enhanced through anthropogenic activities, further added to by artifical sources of radioactivity introduced into the environment [1–3].

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In the marine environment, radioactivity is contributed to by the natural processes of weathering and mineral recycling of terrestrial rocks, seabed movement arising from undersea earthquakes and underwater volcanic activity. Given the enormous contact with various types of minerals and geological materials such as igneous rocks and ores which often contain elevated concentrations of natural radionuclides, consequently ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) decay series radionuclides are transferred to water through leaching action [4]. Anthropogenic activities such as combustion of fossil fuel, as for example from coal-fired power plants, production of natural gas and oil, and mining and processing of ores etc. are also known to enhance the naturally occurring radioactivity in the marine environment. In addition, there are anthropomorphic contributions from post-nuclear disposal of industrial and radioactive waste, underwater nuclear device tests, accidents including leaks from nuclear power plants and from reprocessing of spent nuclear fuel etc [2, 5–7].

Although radionuclides in ocean shows a complex behaviour (for instance, uranium is quite soluble in sea water while thorium is almost totally insoluble, radium and radon is soluble in water), they can be transferred in the marine environment in the following ways: dissolved in the seawater, attached to plankton suspended in the seawater and attached to sediment on the seabed and contaminated the marine organism, including fish, shell fish etc [8].

In addition to pathways from radioactivity in the marine environment into the human diet, a further concern is metal contaminants via consumption of marine products, with bioaccumulation leading to potential risks via long-term exposure[9–13]. Marine life can have considerable capability for bioaccumulationand biosorption (Biosorption is a property of certain types of inactive, dead, microbial biomass to bind and concentrate heavy metals from even very dilute aqueous solutions. It is particularly the cell wall structure of certain algae, fungi and bacteria which was found responsible for this phenomenon. Marine animal has tendency to burrow down in the bottom sea sediments and rocks, filtering on organic particles and algae along with tiny fishes and planktons, which may lead to increase the uptake of radioactive and heavy metals) of radionuclides and toxic/heavy metals from their surroundings, not least fish and shell fish relative to other marine life (e.g., molluscs, crustaceans, and fishes) [2, 5, 14–19]. Seafood (e.g., molluscs, crustaceans, and fishes) and their products can typically be one of the major sources of protein to populations in coastline regions, including those around the Straits of Malacca, one of the most important shipping lanes in the world, transporting about one-quarter of the world's traded goods [2, 5, 14, 16, 20, 21]. Approximately three million barrels of crude oil are shipped through the Straits daily, subjecting the sensitive marine environment to the threat of accidental oil spillage; over the 10-year period 1981–199, an average of two to three oil spill incidents per year were recorded in these waters [21]. The distinct possibility of release of large quantities of metal contaminants from different sources includes association with transportation, with increasing tanker traffic adding to the concern, to which one can add offshore oil and gas exploration, the operation of power plants and other industrial activities, agricultural activities and the waste streams of urbanization, all of which may pose a significant danger to human health due to the non-biodegradability and accumulation of metals in the food chain [20, 22]. The metals include copper, zinc and iron, all essential within both marine and human metabolism while some others such as the heavy metals: mercury, cadmium, arsenic and lead have no known role in biological systems [20]. Together with essential metals, non-essential ones also taken up from water and solid nutrient sources, and can accumulate in the tissues [23]. Metals like Cd, Hg, As, Pb, Cr, Se, Ni etc. have been commonly found in human diets and have been reported to be carcinogenic and /or mutagenic in a broad spectrum of animal studies and short-term test systems, adding to the concern about the contribution of these elements to human carcinogenesis [24, 25].

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Distribution of radioactivity in seafood differs with respect to sites of origin and feeding habits [26]. With uptake in the human clearly depending on dietary habit, it is therefore of interest to note that the region of present research to have one of the highest marine fish consumption rates in the world, to the extent that information on radionuclide balance in seafish assumes proportionally greater importance [27].Information on bioaccumulation and distribution of natural radionuclides and heavy metals in seafish and sea water within the available literature is still lacking. Given the importance of such knowledge, the objective of present research to determine the concentrations of natural radionuclides and heavy metals in edible marine life caught in Straits water, the daily intake of these radionuclides and heavy metals, the ingestion dose and carcinogenic risk for the public residing in and around the coastal area of peninsular Malaysia.

Materials and Methods

Study area

No specific permissions were required for these field studies (locations/activities), because the studied field locations are open and we have collected the fish sample from the fisherman of the corresponding locations who are fishing in the studied locations. The field studies did not involve any protected species of fish. We have collected the most commonly consumed fish in Malaysia

The primary fish landing areas of the west coast of Peninsular Malaysia, facing out to the Straits of Malacca, partitioned into three regions: the northern region (Perlis, Kedah, Penang and Perak), the central region (Selangor, Kuala Lumpur and Putrajaya) and the southern region (Negeri Sembilan, Malacca and Johor) were chosen as the sampling locations. Over the period November 2011 to February 2012, the samples were collected from coastal locations of Pantai Remis (4.4500° N, 100.6333° E) in Perak, Port Klang (3.0000° N, 101.4000° E) in Selangor, and Bagan Lalang (5.4333° N, 100.3833° E) in Negeri Sembilan, all surrounded by many fishing villages (Fig 1). In each case, there are nearby large-scale industrial activities, and

Fig 1. Location of marine fish sampling sites.

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Table 1. Marine fish samples collection data.

Local name (Scientific name)	Habitat	Diet	Sampling location (States)	GPS coordinate	Sample code	Number of sample	Measured length (cm)	Measured weight (g)
Ikan Kembung* (*Rastrelliger kanagurta*)	Pelagic (Warm shallow coastal waters)	Macroplankton including the larvae of shrimp and fish	Bagan Lalang (Negeri Sembilan)	(5.43333° N, 100.3833° E)	Fi_Bl-1	3	22–25	125–130
					Fi_Bl-2	3	22–25	125–130
					Fi_Bl-3	3	22–25	125–130
			Port Klang (Selangor)	(3.0000° N, 101.4000° E)	Fi_Kl-1	3	22–25	125–130
					Fi_Kl-2	3	22–25	125–130
					Fi_Kl-3	3	22–25	125–130
			Pantai Remis (Perak)	(4.500° N, 100.6333° E)	Fi_Re-1	3	22–25	125–130
					Fi_Re-2	3	22–25	125–130
					Fi_Re-3	3	22–25	125–130

*Indian mackerel

a number of coal-, gas- and oil-fired power plants, including the 2295 MW Manjung coal-fired power plant in Perak, the 1400 MW Jimah coal-fired power plant in Negeri Sembilan and the 2420 MW Kapar coal-fired power plant in Klang, Selangor. Coal contains only a trace amount of radioactive uranium, barium, thorium and potassium, and are not known to create any major problem to the environment. However when coal is burned, in the fly ash that results, uranium and thorium are concentrated by up to 10 times their original levels [12, 28], and can be easily deposited and affecting the surrounding environment [29].

Collection and processing of sample

The samples were of two categories: fish and seawater. The fish, a particular species of Mackerel (local and scientific name, Ikan kembung and *Rastrelliger kanagurta* (Phylum: Chordata; Family: Scombridae; Genus:*Rastrelliger;*Species:*R. kanagurta*) respectively)) were collected from coastal fishing jetties at various locations (Fig 1 and Table 1). The sampling informations are shown in Table 1. Same size and amount (125–130 g each) of fish samples (Fig 2) were collected andwashed, cut into smaller pieces and dried in a furnace at 70°C over a period of three days. The samples were then pulverized to obtain a fine powder and sieved for homogeneity. Sea water samples were collected from an approximate depth of one meter from the surface

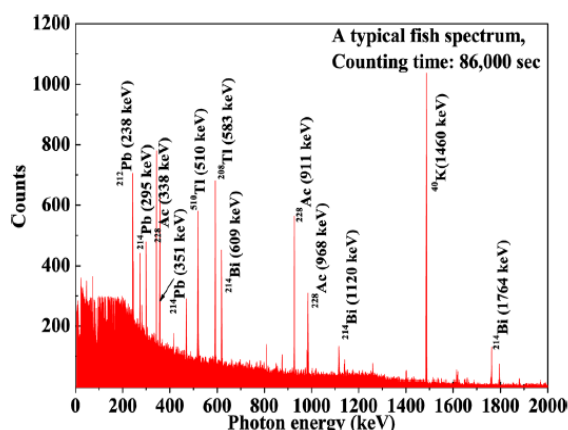Fig 2. Kembung fish (*Rastrelliger kanagurta*).

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Fig 3. A typical gamma-ray spectrum collected from a fish sample.

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and filtered to remove any impurities. 120 to 218 g of each fish sample and 750 ml of each water sample were then sealed into Marinelli beakers and left for about 6–8 weeks to attain secular equilibrium between the ^{238}U and ^{232}Th precursors and their short-lived progenies.

Measurements of radioactivity

The radioactivities of the samples were determined using a high resolution, p-type coaxial HPGe γ-ray spectrometer (ORTEC; GEM-25P; Serial no. 46-TP22121A; 57.5-mm crystal diameter and 51.5-mm thickness; operating voltage: +2800 V) shielded by cylindrical lead. The detector relative efficiency was 28.2% and energy resolution of 1.67 keV-FWHM at the 1.33 MeV peak of ^{60}Co. The detector was coupled to a 16 k MCA to determine the photo-peak area of the γ-ray spectrum and analyzedby Gamma Vision 5.0 software (EG&G Ortec). A cylindrical multi-nuclide source was used for detector energy calibration and efficiency determination [30,31]. The measured detection efficiencies were fitted by using a polynomial fitting function as described in ref. [3], and the fitted efficiencies were used in activity determination of the samples. The minimum detectable activity (MDA) of the γ-ray measurement system at 95% confidence level was calculated according to the procedure in [32]. Each sample was counted for 86400 s and similarly for background counts to obtain the net activity. Fig 3 represents a typical gamma-ray spectrum collected from a fish sample. Only strong and independent characteristic gamma lines (the γ-rays highlighted in bold in Table 2) of the respective radionuclides were used to determine the net activity concentrations to reduce the error in activity determination.

Metals detection

In this study, Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) (Agilent Technologies 7500 Series, USA) was used to determine the metal concentrations. For elemental analysis with ICP-MS, it is necessary to digest the sample completely. About 0.5 g of eachpowdered fish sample was digested in microwave digestion system (Multiwave 3000, PerkinElmer, USA) using a mixture of 5 ml of 65% concentrated HNO_3 (SpectrosoL grade) and 3 ml of H_2O_2 (30%) for 10 minutes using microwave heating. After digestion the samples solution were cooled in a water bath, filtered and brought up to a volume of 50 ml using ultra-pure water

Table 2. Decay data of the detected radionuclides of interest (Source: http://www.nndc.bnl.gov/nudat2/).

Radionuclides of interest	Detected radionuclides	Half-life	Decay mode (%)	γ-ray energy, E_γ (keV)	γ-ray intensity, P_γ (%)
^{226}Ra	^{214}Pb	26.8 m	β⁻ (100)	295.2228	18.42
				351.9321	35.60
	^{214}Bi	19.9 m	α (0.021);	609.320	45.49
			β⁻ (99.979)	1120.294	14.92
				1764.491	15.30
^{232}Th	^{212}Pb	10.64 h	β⁻ (100)	238.632	43.6
	^{208}Tl	3.053 m	β⁻ (100)	510.77	22.60
				583.187	85.0
				860.557	12.50
	^{228}Ac	6.15 h	β⁻ (100)	338.320	11.27
				911.204	25.8
				968.971	15.8
^{40}K	^{40}K	1.248×10⁹ y	EC 10.719);	1460.822	10.66
			β+ (0.001);		
			β⁻ (89.28)		

The γ-lines in bold were used in activity determination.

(18.2 MΩ-cm) in a volumetric flask.In all cases, 1 blank solution and 5 standards were run with the same reagents used under the same conditions to control possible contamination from digestion procedures.

Calibration of the ICP-MS was performed using multi-element-element calibration standard 2A solution (10 mg/l of each element) prepared by adequate mixing and dilution in 5% HNO of 5 standard solutions (Agilent Technologies, USA, part no. 8500–6940). Concentrations for Na, Mg, Al, K, Ca, Cr, Mn, Fe, Co, Cu, Zn, Cd, As, Se, Rb, Sr, Ba, Hg, Cd, Ni and Pb heavy metals were then determined by using ICP-MS. All the analyses were carried out in several times. The recovered values of all the metals ranged from 88% to 96% of the certified value. The ICP-MS detection limits for Na, Mg, Al, K, Ca, Cr, Mn, Co, Ni, Cu,Zn, As, Rb, Sr and Se was 0.0002 mg kg^{-1}, while for Fe, Cd, Ba and Pb was 0.0001 mg kg^{-1}.

Calculations of activity and other radiation indices

Radionuclide specific activity. Direct assessment of ^{226}Ra (^{238}U) and ^{228}Ra (^{232}Th) via conventional γ-ray spectrometry is inappropriate due to the associated very low decay rates and absence of any intense characteristic γ-lines. However, since the progeny of ^{226}Ra (^{238}U) and ^{228}Ra (^{232}Th) remain in secular equilibrium with the parent, the activity of any such progeny represents the activity of the respective parent, a procedure described by among others, thus we assessed the radionuclide activity following the procedure described in ref. [3].

Estimation of uncertainties. The combined uncertainty in each sample activity was estimated by considering the following uncertainties: statistical uncertainty of the γ-ray counting (0.5–10%); uncertainties in the detection efficiency (~4%); uncertainties in sample weight (~1.5%), and; uncertainty in γ-ray intensity (~1%). These individual contributions, considered to be independent, were added in quadrature to obtain total uncertainties in the range 4.4–10.9%. The assessed radioactivities, together with their uncertainties, are presented in Table 3.

Daily intake of radioactivities. The daily intake of radioactivity from consumption of fish as the dominant diet, is assumed to acrue from an accumulation of the naturally occuring radionuclides ^{226}Ra, ^{232}Th and ^{40}K in *Rastrelliger kanagurta*. Taking into account the annual

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Table 3. Activity concentration (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K in marine fish and water in the Strait sof Malacca.

Sample code	Radioactivity concentration of fish (Bq kg^{-1})			Sample code	Radioactivity concentration of fish (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K		^{226}Ra	^{232}Th	^{40}K
Fi_Bl-1	7.79±0.46	6.06±0.36	387.6±18.8	Wa_Bl-1	1.03±0.12	0.25±0.04	15.2±1.4
Fi_Bl-2	8.16±1.08	6.25±0.65	374.1±20.7	Wa_Bl-2	1.05±0.13	0.27±0.06	17.3±1.5
Fi_Bl-3	7.54±0.79	6.31±0.58	316.0±17.0	Wa_Bl-3	1.08±0.14	0.32±0.07	16.7±1.5
Mean	7.83±0.78	6.21±0.53	359.2±18.8	-	1.05±0.13	0.28±0.06	16.4±1.5
Fi_Kl-1	6.32±0.56	3.75±0.34	371.5±19.2	Wa_Kl-1	1.02±0.16	0.34±0.07	16.0±1.3
Fi_Kl-2	6.72±0.72	3.79±0.38	442.4±22.6	Wa_Kl-2	1.41±0.19	0.37±0.08	14.5±1.2
Fi_Kl-3	6.40±0.70	3.58±0.37	382.6±18.9	Wa_Kl-3	1.25±0.18	0.35±0.08	14.8±1.2
Mean	6.48±0.66	3.71±0.36	398.9±20.2	-	1.23±0.17	0.35±0.08	14.8±1.3
Fi_Re-1	4.37±0.46	1.62±0.21	268.7±13.8	Wa_Re-1	1.28±0.19	0.26±0.06	20.1±1.7
Fi_Re-2	3.77±0.47	1.92±0.21	299.1±15.1	Wa_Re-2	1.03±0.12	0.30±0.07	20.1±1.6
Fi_Re-3	4.02±0.51	2.25±0.30	296.6±15.5	Wa_Re-3	1.22±0.16	0.25±0.09	19.5±1.7
Mean	4.05±0.48	1.93±0.24	288.1±14.8	-	1.18±0.16	0.27±0.07	19.9±1.7
Over all mean	6.31±0.65	4.13±0.39	348.7±18.0	-	1.15±0.15	0.30±0.07	17.0±1.5

Sample codes Fi-Bl, Fi-Kl and Fi-Re relate to fish (Ikan Kembung) obtaianed off Bagan Lalang, Port Klang and Pantai Remis respectively, while the respective codes when prefixed by the letters Wa refer to water samples.

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marine fish landings of 1,472,240 tonnes (1 short ton = 907.188 kg) in 2012 in peninsular Malaysia [33] and a total adult population of 19.15 million in 2012 [34], the per capita daily intake of natural radionuclides via the consumption of seafood has been calculated using the equation [3,35] and presented in Table 4;

$$D_{int} = \frac{A_s \times A_p \times F_c}{M_p \times 365} \qquad (1)$$

where, D_{int} is the daily intake of radioactivities (Bq) by individuals, A_s is the specific activity of

Table 4. Annual effective dose (μSv y^{-1}) and life time cancer risk due to the comsumption of natural radionuclide from the marine fish.

Sample code	Daily intake (Bq d^{-1})			Effective dose (μSv y^{-1})			Total effective dose (μSv y^{-1})	Lifetime cancer risk (LCR)		
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K		^{226}Ra	^{232}Th	^{40}K
Fi_Bl-1	1.01	0.79	50.33	103.4	66.07	113.9	283.4	2.5×10^{-4}	4.9×10^{-5}	7.6×10^{-4}
Fi_Bl-2	1.06	0.81	48.58	108.3	68.14	109.9	286.4	2.6×10^{-4}	5.1×10^{-5}	7.3×10^{-4}
Fi_Bl-3	0.98	0.82	41.04	100.1	68.79	92.87	261.7	2.4×10^{-4}	5.1×10^{-5}	6.2×10^{-4}
Mean	1.02	0.81	46.65	103.9	67.67	105.6	277.2	2.5×10^{-4}	5.0×10^{-5}	7.0×10^{-4}
Fi_Kl-1	0.82	0.49	48.25	83.88	40.88	109.2	234.0	2.0×10^{-4}	3.0×10^{-5}	7.3×10^{-4}
Fi_Kl-2	0.87	0.49	57.45	89.19	41.32	130.0	260.5	2.1×10^{-4}	3.1×10^{-5}	8.6×10^{-4}
Fi_Kl-3	0.83	0.46	49.69	84.94	39.03	112.5	236.4	2.0×10^{-4}	2.9×10^{-5}	7.5×10^{-4}
Mean	0.84	0.48	51.80	86.00	40.41	117.2	243.6	2.1×10^{-4}	3.0×10^{-5}	7.8×10^{-4}
Fi_Re-1	0.57	0.21	34.89	58.00	17.66	78.96	154.6	1.4×10^{-4}	1.3×10^{-5}	5.3×10^{-4}
Fi_Re-2	0.49	0.25	38.84	50.04	20.93	87.90	158.9	1.2×10^{-4}	1.6×10^{-5}	5.8×10^{-4}
Fi_Re-3	0.52	0.29	38.52	53.35	24.53	87.16	165.0	1.3×10^{-4}	1.8×10^{-5}	5.8×10^{-4}
Mean	0.53	0.25	37.42	53.80	21.04	84.67	159.5	1.3×10^{-4}	1.6×10^{-5}	5.6×10^{-4}
World average	-	-	-	120	120	170	290	-	-	-

The meaning of the sample codes are as in Table 2 above.

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radionuclides of interest (Bq kg^{-1} dry weight), A_P is the annual production, F_c is the real fraction consumed, the average value of F_c being considered as 68% (after a consideration of 32% wastage and export) [36], M_P is the Malaysian population and 365 indicates the days/year.

Committed dose from annual intakes. Estimation of radiation induced health effects associated with the intake of radionuclides are proportional to the total dose delivered by the radionuclides. The committed effective dose to an individual from an intake of a radionuclide via ingestion of one type of food has been calculated by the following formula [37, 38],

$$D_{eff} = A_s \times A_{if} \times D_{cf} \qquad (2)$$

where, D_{eff} is the annual effective dose to an individual (μSv yr^{-1}); A_s is the specific activities of radionuclides (Bq kg^{-1}); A_{if} is the annual intake of food (kg yr^{-1}), where as mentioned in *section 2.5.3*, the per capita seafish consumption for Malaysia for the year 2012 was 47.4 kg yr^{-1}; and D_{cf} is the ingestion dose conversion factor (2.8×10^{-7} Sv Bq^{-1} for ^{226}Ra, 2.3×10^{-7} Sv Bq^{-1} for ^{232}Th, 6.2×10^{-9} Sv Bq^{-1} for ^{40}K) are taken from refs. [1, 39, 40]. The total dose (committed) via ingestion can be calculated using the following formula:

$$D^{total}_{eff} = [(A_{Ra-226} \times D_{cf}) + (A_{Th-232} \times D_{cf}) + (A_{K-40} \times D_{cf})] \times (A_{if} \times F_c) \qquad (3)$$

Daily intake of metals (DIM). The daily intake of metals (Cd, Co, Cr, Mn, Fe, Ni, Cu, Zn, and Pb) depends both on the metal concentration level and the amount of consumption. The *DIM* for adults was estimated using the following equation [41]:

$$DIM = \frac{C_{metal} \times W}{m} \qquad (4)$$

where C_{metal} is the concentration of heavy metals in fish; W represents the daily average consumption of fish (130 g) and m is the adults body weight (70 kg).

Carcinogenic risk. With longevity contributing to greater radiation exposure and by association increasing cancer incidence, an effort was made to assess the lifetime cancer risk due to the ingestion of marine fish by the procedure proposed by the United States Environmental Protection Agency, USEPA [42]. The following equation [37, 43] was used to calculate the mortality cancer risk and is shown in Table 4.

$$LCR = A_{ir} \times A_{ls} \times R_c \qquad (5)$$

where LCR, A_{ir}, A_{ls} and R_c are the lifetime cancer risk, annual intake of radionuclide (Bq), average span of life (70 y) and mortality risk coefficient (Bq^{-1}), respectively. The values of mortality cancer risk coefficients included 9.56×10^{-9} (Bq^{-1}) for ^{226}Ra, 2.45×10^{-9} (Bq^{-1}) for ^{232}Th and 5.89×10^{-10} (Bq^{-1}) for ^{40}K, taken from the USEPA (1999) [42].

Statistical analysis. The data were statistically analyzed using SPSS 21 software (IBM Corporation, Armonk, NY, USA). Activity concentrations, daily intake and annual effective dose (using the fish samples data from the three study locations) were compared employing oneway analysis of variance (ANOVA). Post Hoc Tukey HSD test was conducted to verify statistically significant differences among individual means at $p < 0.05$.

Results and Discussion

Radioactivity concentrations

In regard to the natural radionuclides, ^{228}Ra (^{232}Th) concentration in sea water to be consistently lower than that of ^{226}Ra (^{238}U). The result supports the solubility of uranium and

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low-solubility of thorium in water [3, 44], offset to an extent by the greater abundance of thorium in the earth's crust.In turn, the accumulation of thorium in sea fish can also be expected to be somewhat lower than that of the unranium chain nuclides; the dry weight basis activity concentrations of the investigated radionuclides in fish, and of that in water samples, along with their uncertainties, are summarized in Table 3. Present analysis shows greater concentrations of ^{226}Ra than that of ^{232}Th in the fish for all three study areas. Statistical analysis (ANOVA) also shows significant variation ($p < 0.05$) in concentrations of ^{226}Ra and ^{232}Th in the fish samples among the three studied locations. The greatest mean concentration of ^{226}Ra (^{238}U) was (7.83 ± 0.78 Bq kg^{-1}), being that in Bagan Lalang, Negeri Sembilan, while it was found to be least (4.05 ± 0.48 Bq kg^{-1}) in Pantai Remis, Perak. Similarly, for ^{228}Ra (^{232}Th), the greatest mean concentration (6.21 ± 0.53 Bq kg^{-1}) was found in Bagan Lalang and lowest (1.93 ± 0.24 Bq kg^{-1}) in Pantai Remis. Of further note that the activity concentrations of ^{40}K are significantly greater ($p < 0.001$) than that of the other radionuclides for all study locations (Table 3), the greatest mean activity concentration of ^{40}K being 398.6 ± 20.2 Bq kg^{-1}, found in fish samples from Port Klang, Selangor and lowest at 288.1 ± 14.8 Bq kg^{-1} in Pantai Remis, Perak. The appreciably greater values for ^{40}K are in line with expectation, a considerable fraction of the weight of each sample being accounted for by the fish bones rich as they are in potassium.

From the results (Table 3), it is clear that concentrations of all detected radionuclides in fish samples was found to be greatest in areas surrounding Bagan Lalang. The possible reasons form a complex mix of factors, linked perhaps to the wide varieties of activity (e.g., housing, tourism, power generation plants, petroleum, chemical industries etc.) around the Bagan Lalang area, industrial and urbanization effluents perhaps increasing the concentrations of radionuclides in the marine environment. It is also important to note that here the Straits are rather more narrow than towards the less industrialized Pantai Remis.

In making comparisons with levels elsewhere, the range of specific activity of ^{226}Ra, ^{232}Th and ^{40}K in sea fish samples from the Black Sea Region of Turkey have been reported [22] as 0.06 ± 0.01 to 0.96 ± 0.36, 0.12 ± 0.04 to 1.03 ± 0.15 and 35.04 ± 0.24 to 127.41 ± 2.29 Bq kg^{-1} respectively. A study [45] reported average activities of ^{226}Ra, ^{232}Th and ^{40}K in fish samples (anchovy) from Korea as < 0.049, 0.0381 and 15.45 Bq kg^{-1} respectively. The average concentrations of ^{226}Ra and ^{232}Th activity in fish samples from Nigeria have been reported [46] as 0.272 Bq kg^{-1} and 0.115 Bq kg^{-1} respectively. The range of activity concentration of ^{238}U, ^{232}Th and ^{40}K in marine fish samples from the Bay of Bengal, off the coast of Bangladesh, have been reported [47] as 0.11–1.94, 0.24–2.28 and 4.93–77.09, respectively. In all of these cases, the results are very much lower than that of present observations. Conversely, Ariffin et al. 2011 [48] have reported average activity ranges for ^{226}Ra and ^{228}Ra in the soft tissue of fish collected from Kapar, Klang, Malaysia (near to Port Klang and very near to the 2420 MW Kapar coal-fired power plant) of 11.82 ± 5.23 to 16.53 ± 6.53 Bq kg^{-1} and 43.52 ± 16.34 to 53.57 ± 19.86 Bq kg^{-1} respectively, being considerably greater than the present study.

The estimated daily intake of radionuclides and annual effective doses due to an intake of ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K from the consumption of *Rastrelliger kanagurta*are presented in Table 4. On average, the daily intake of each radionuclide from *Rastrelliger kanagurta* in all study regions were estimated to be 0.80,0.51 and 45.29 Bq for ^{226}Ra,^{232}Th and ^{40}K, respectively.

By ANOVA analysis, the daily intake of ^{40}K was found to be significantly greater ($p < 0.001$) than that of the other radionuclides. The intakes of ^{226}Ra (1.7%), ^{232}Th (1.1%) are negligible compared with that of the radionuclide ^{40}K (96.7%), the latter being a natural isotope of potassium, an essential element for vertebrates and one which is fairly constant as a result of homeostatic regulation [49].

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The annual effective dose of ^{226}Ra and ^{232}Th varies significantly (p < 0.05) among the studied locations. The greatestannual effective dose for ^{40}K (117.2μSv y^{-1}) was found in Port Klang followed by ^{226}Ra (103.9 μSv y^{-1}) in Bagan Lalang. Significantly (p < 0.05), the greatest total committed dose was found in Bagan Lalang (278.9 μSv y^{-1}), while the least was found in Pantai Remis (160.6 μSv y^{-1}). The average worldwide effective dose from the ingestion of uranium and thorium series nuclides is reported to be 120 μSv y^{-1}, while for ^{40}K it is 170 μSv y^{-1} [30, 36]. The annual effective dose for all detected radionuclides in all study locations are within the [36] mentioned values. The lifetime cancer risk was found to vary from 1.3×10^{-4} to 2.5×10^{-4} for ^{226}Ra, 1.0×10^{-5} to 5.0×10^{-5} for ^{232}Th and 5.6×10^{-4} to 7.8×10^{-4} for ^{40}K, which are low compared with the acceptable cancer risk of 10^{-3} for radiological risk [37,43].

Metal concentrations

The concentrations determined for the metals Al, Cr, Mn, Fe, Co, Cu, Zn, As, As, Hg and Pbin the fish samples are presented in Table 5 and their Estimated Daily Intake (*EDI*) (body weight for 70 kg adults) are reported in Table 6 and in Fig 4.

A number of national environmental protection agencies report that an intake of 1.0 mg/day of inorganic arsenic (As) is sufficient to give rise to skin lesion after a few years [50]. The greatest mean As concentration observed herein was at Port Klang (0.748 mg/kg dry weight), varying with a factor of about 0.2 for other regions, the least value being found to be (0.325 mg/kg dry weight) at Bagan Lalang. The reported values are appreciably lower than that suggested European Community maximum permissible guideline of 2 mg/kg (dry weight) for marine fish [51] and that reported by Korkmaz Görür e (4.4 mg/kg) of Black Sea fish in Turkey [22].

Table 5. Trace element concentrations (mg kg^{-1}) in the most frequently consumed sea fish (Ikan Kembung) in Malaysia.

Sample location	Port Klang				PantaiRemis				BaganLalang		
Sample name	Fish-1	Fish-2	Fish-3	Average	Fish-1	Fish-2	Fish-3	Average	Fish-1	Fish-2	Average
Na	233.1	115.1	231	193.1	220.9	219.5	223.5	221.3	260.7	250	255.4
Mg	89.86	49.32	102.1	80.43	87.59	85.54	88.53	87.22	115.4	114.6	115.0
Al	4.488	2.816	3.789	3.698	4.449	4.798	5.301	4.849	3.508	2.722	3.115
K	483.2	290.2	623.4	465.6	464.7	459.7	456.6	460.3	439.4	436.4	437.9
Ca	6899	2684	5592	5058	6980	6487	6675	6714	6614	6241	6427
Cr	0.0432	0.0028	0.0624	0.0361	0.0453	0.0514	0.058	0.0516	0.0108	-	0.0108
Mn	1.985	0.1642	0.3926	0.847	1.942	1.717	1.863	1.841	0.4598	0.4146	0.4372
Fe	4.843	2.102	3.692	3.546	4.796	4.883	4.893	4.857	4.111	3.855	3.983
Co	0.0027	-	-	0.0027	0.0033	0.0012	0.0038	0.0028	0.0007	-	0.0007
Cu	0.0866	0.0314	0.0956	0.0712	0.0683	0.0679	0.0773	0.0712	0.2018	0.1936	0.1977
Zn	17.02	1.888	3.502	7.470	15.17	14.96	15.91	15.35	5.577	5.637	5.607
As	0.6061	0.5523	1.087	0.7485	0.5288	0.5305	0.5393	0.5329	0.3237	0.3262	0.325
Se	0.0471	0.0311	0.0759	0.0514	0.034	0.0332	0.0379	0.035	0.0907	0.0969	0.0938
Rb	0.2428	0.0647	0.1603	0.1559	0.2314	0.2298	0.2325	0.2312	0.098	0.0975	0.0978
Sr	28.2	11.41	23.46	21.02	28.24	26.88	27.93	27.68	20.22	18.95	19.56
Mo	0.5767	0.7037	1.407	0.8958	0.4084	0.3128	0.3034	0.3415	2.719	2.541	2.63
Ba	0.2087	0.256	0.5616	0.3421	0.1845	0.1616	0.5075	0.2845	0.6917	0.6276	0.6597
Bi	8.61	0.4822	3.338	4.143	9.181	1.066	1.821	4.023	2.426	0.7697	1.598
Hg	0.056	0.014	0.004	0.025	-	-	-	-	-	-	-
Pb	-	-	-	-	-	-	-	-	0.02	-	0.02

The average concentration for each metal was used in the calculations of daily intake of metals.

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Table 6. Estimated daily intake (*EDI*) of heavy metals through consumption of fish.

Sample location Element	Port Klang		Pantai Remis		Bagan Lalang		TDI (µg/kg/day)
	Average Concentration (mg/kg) dry weight	EDI (µg/kg) body weight per day	Average Concentration (mg/kg)	EDI (µg/kg) body weight per day	Average Concentration (mg/kg)	EDI (µg/kg) body weight per day	
Al	3.6977	6.8671	4.8493	9.0059	3.115	5.785	143[a]
Cr	0.0361	0.0671	0.0516	0.0958	0.0108	0.0201	143[b]
Mn	0.8473	1.5735	1.8407	3.4184	0.4372	0.8119	157[c]
Co	0.0027	0.0050	0.0028	0.0051	0.0007	0.0013	20[b]
Cu	0.0712	0.1322	0.0712	0.1322	0.1977	0.3672	142[c]
As	0.7485	1.3900	0.5329	0.9896	0.3249	0.6035	2.14[a]
Hg	0.0247	0.0458	-	-	-	-	0.57[a]
Pb	-	-	-	-	0.02	0.0371	3.60[d]

Sources:
[a] Antoine et al. (2012)
[b] Nutrition^{ATC} (2014)
[c] Health Canada, (2007)
[d] Zhuang et al (2009)

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Mercury (Hg), potentially carcinogenic, can also produce adverse effects during developmental stages as a result of acute or chronic exposure. While there is no known reported safe level [52], the Canadian Food Inspection Agency has set a standard for fish of 0.2 mg/kg dry weight [53]. In present study it has only been detected in fish collected from Port Klang, with values ranging from 0.004 to 0.056mg/kg dry weight, almost certainly reflecting the
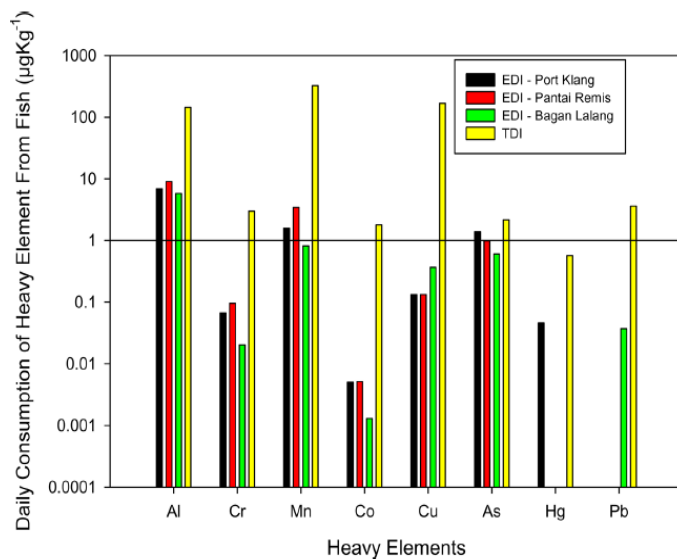

Fig 4. Heavy metal concentrations in fish of Port Klang, Pantai Remis and Bagan Lalang: Comparison of Daily Estimated Intake (EDI) with the Tolarable Daily Intake (TDI) of heavy metals.

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cummulative activities of many Malaysian industries located in and around the region of the Klang valley. While the level in sampled fish is a factor of 10 below the Tolerable Daily Intake (*TDI*), the estimated daily intake of Hg from consumption of fish does represent a concern worthy of regular monitoring.

Lead (Pb), again a potential carcinogen, can cause adverse health effects [41] including lead poisoning,confusion, poor cognition and kidney damage. In present study, only in one case was Pb was detected, in fish obtained from Bagan Lalang, with a concentration of 0.02 mg/kg, being well below thesuggested level of 2.0 mg/kgin fish [54]. While again this is presently at a level of limited concern, the value serves as an initial datum point from which future changes in levels in the region can be monitored. By way of comparision, [55] reported Pb levels from Turkey ranging from0.09 to 6.95 mg/kg, from fish in the northern Mediterian, while [22] reported a lower range of Pb (< 0.001–0.06 mg/kg) in the fish of the Black Sea of Turkey. The observed large variation between the EDI and TDI suggests Pb levels through consumption of fish is yet below the hamful level.

Chromium (Cr), a proven carcinogenic metal, if taken at a dose of 0.5 mg/kg of body weight per day via oral ingestion [56]. Cr was found with varying concentrations in all samples with the exception of some from Bagan Lalang, with values ranging from 0.003 to 0.052 mg/kg. By way of comparison, the Turkish study of [22] reported Cr at levels of between < 0.1 to 0.73 mg/kg dry weight of fish. Adequate Intake (AI) of 30 μg/kg body weight has been suggested for 70 years adult and above [53]. The results of present study represent a current safe level of exposure.

Elevated manganese (Mn) content showing negative effects on fertility, the central nervous system and embryo and fetal development [57]. In present study, Mn has been detected in all samples at low levels, with a range of 0.4–1.8 mg/kg dry weight, being not too disimilar from that of two Turkish Black Sea studies, at 0.56–1.04 mg/kg [22] and 0.10–0.99 mg/kg of fish [58]. Comparing the upper TDI of this element (324 μg/kg bw) with the calculated EDI (3.4 μg/kg bw), present fish consumption remains at safe level.

Exposure to cobalt (Co) in the bodyorgans has been noted to lead to complicated health problems [59]. Herein, a relatively low concentration of Co has been detected in all of the samples found to be of insignificant risk via TDI and EDI, with a range from a maximum of 0.0028 mg/kg down to 0.0007 mg/kg, from Pantai Ramis and Bagan Lalang respectively, [60] reported a higher range (< 0.05 to 0.30 mg/kg dry weight) of Co in the fish from the coastal region of Turkey around the Black Sea.

Copper (Cu) is essential in all organisms intrace amount, and particularlyserves as a constituent of respiratory enzyme complex in the human body. Due to its role in facilitating iron uptake, deficiency of Cu can lead to impaired growth, anemia-like symptoms, bone abnormalities, and vulnerability to infections. Cu has been found in all of the samples, with concentrations ranging from 0.03 to 0.2 mg/kg dry weight of fish which is below the TDI set by joint experts of FAO/WHO as 3 mg/kg body weight [22] and thelevel of risk is yet insignificant. By way of comparison, reported a range of 0.4 to 1.5 mg/kg from a similar study conducted in Spain [61].

While the health effects of aluminium (Al) has not been widely reported, there is increasing evidence of its toxicity in relation to its gradual accumulation in the brain and subsequent effects on the nervous system [62,63]. In present study, the range of Al content in the sampled fish was 2.7 to 5.3 mg/kg dry weight. The WHO has recently revised its previous Provisional Tolerable Weekly Intake from 7mg/kg body weight to just 1 mg/kg body weight [63]. Comparing this revised WHO value with the estimated values from fish consumption, the current Al level per kg body weight can be considered to remain at a safe level.

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Mineral elements

The marine diet represents a major source of many essential elements, exemplified by present results, with healthy concentrations of iron, zinc, calcium, sodium, magnesium, selenium, etc. The importance of these elements and of deficiency symptoms are well documented [64], and forms only a minor aspect of this work.

Conclusions

Owing to their pathway into the daily diet of the local populace, radioactivity and heavy metal concentrations have been assessed in marine life from the Straits of Malacca, west coast of Malaysia. The present investigation shows the radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in sea water samples in all study sites to be very much lower than that of sea fish samples obtained from the same areas.

Present study shows elevated radioactivity concentrations in the fish of the Straits of Malacca compared to that reported in similar studies in seas elsewhere. The results reflect the contribution of additional technologically enhanced naturally occurring radioactive material (TENORM) pollutants, largely expected to be a result of oil and gas waste streams, related to shipping activities, the route being regarded as the second busiest water channel in the world. In regard to intercomparison of results from the study locations, the Bagan Lalang area showed the greatest level of radioactivity; here, the narrowing of the Straits, together with a relatively high level of industrialisation, urbanization and the effluents that result from these factors are expected to lead to increased concentrations of radionuclides in the marine environment, including fish.

The annual effective doses received by individuals due to the dietary intake of ^{226}Ra, ^{232}Th and ^{40}K via the consumption of fish, range from 154.6 to 286.4 μSv y^{-1} with an average of 226.7 μSv y^{-1}, falling below the world average for annual effective dose. Accordingly, the carciogenic risk was found to be well below the acceptable limit of 10^{-3}. Present study indicates radionuclide intake from consumption of Straits of Malacca fish poses insignificant threat to public health.

Present study identifies the presence of a wide range of non-essential metals in the selected fish, albeit at relatively low levels compared to studies conducted eleswhere, varying in concentration from region to region and element to element. While again the variation is to be linked with industrial locations and types, the pattern does not agree with that found for NORM nuclides, with levels at Pantai Remis now typically recording the greater levels. Although there may not be internationally agreed safe levels for all of these metals, the overall contamination from the samples would seem to strongly indicate the samples to be well below harmful levels.

Author Contributions

Conceived and designed the experiments: MUK YMA. Performed the experiments: SMN KhA. Analyzed the data: SMN KhA MUK. Contributed reagents/materials/analysis tools: SMN KhA ARU HA AAO. Wrote the paper: MUK KhA ED DAB.

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3.2.6   Published paper 6

NATURAL RADIOACTIVITY AND EFFECTIVE DOSE DUE TO THE BOTTOM SEA AND ESTUARIES MARINE ANIMALS IN THE COASTAL WATERS AROUND PENINSULAR MALAYSIA

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Malaysia is among the countries with the highest fish consumption in the world and relies on seafood as a main source of animal protein. Thus, the radioactivity in the mostly consumed marine animals such as fishes, crustaceans and molluscs collected from the coastal waters around Peninsular Malaysia has been determined to monitor the level of human exposure by natural radiation via seafood consumption. The mean activity concentrations of naturally occurring radionuclides ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K ranged from 0.67 ± 0.19 Bq kg^{-1} (*Perna viridis*) to 1.20 ± 0.70 Bq kg^{-1} (*Rastrelliger*), from 0.19 ± 0.17 Bq kg^{-1} (*Teuthida*) to 0.82 ± 0.67 Bq kg^{-1} (*Caridea*) and from 34 ± 13 Bq kg^{-1} (*Caridea*) to 48 ± 24 Bq kg^{-1} (*Teuthida*), respectively. The mean annual committed effective dose due to the individual radionuclides shows an order of ^{228}Ra $> ^{226}$Ra $> ^{40}$K in all marine samples. The obtained doses are less than the global internal dose of 290 µSv y^{-1} set by the United Nations Scientific Committee on the Effects of Atomic Radiation, discarding any significant radiological risks to the populace of Peninsular Malaysia.

INTRODUCTION

Besides the presence of natural radioactivity that permeates every compartment of the universe at varying concentrations, human activities such as the release of radioactive wastes from extractive industries, combustion of fossil fuel, production of radionuclides for medical and industrial applications, offshore oil and gas exploration, operation of power plants, and nuclear accident enhance/cumulate the level of radioactivity in marine environment, hence shows increasing concern to monitor the radionuclide contents in marine materials. Studies of naturally occurring radioisotopes of uranium and thorium decay series and primordial potassium in aquatic environment provide information on the environmental pollutants in water bodies, whereas marine animals (due to the possible radionuclide transfer) provide an easy assimilation of radiation exposure into human body following their consumption[1]. Moreover, marine organisms such as filter feeders and piscivorous have the capacity of bioaccumulating more radionuclides and toxic elements from water[2] due to the physical and chemical nature of their body surfaces and feeding habit in their natural habitat[3], and thus, the determination of radioactivity in the bottom sea and estuaries marine animals assumes greater importance.

A detailed review of literature[4–8] reveals that there is still paucity of data on radionuclide contents in marine materials of Malaysia for adequate radiological assessment to the coastal population. To this end, the study has been conducted to investigate the level of natural radionuclides (^{226}Ra, ^{228}Ra and ^{40}K) in

mostly consumed marine fishes and shell fishes due to their importance as sources of high-quality protein. The annual effective dose due to intake of radioactivity has been estimated for radiological risk assessment to ensure safer consumption of seafood and other activities related to the use of marine environment. This study is expected to compliment to the earlier studies on seafood and gives more insights about the nature of marine ecosystem in the country and development of environmental regulatory frameworks.

SAMPLING AND SAMPLE PREPARATION

Fresh samples of fish (*Dasyatidae, Formio niger* and *Rastrelliger*), molluscs (*Perna viridis* and *Teuthida*) and crustaceans (*Caridea*) were collected from both the west and east coasts of Peninsular Malaysia. The choice of these two study areas is due to their biodiversity and massive fish farming practices. The two coastal areas alone have a total of 59 fish-landing districts out of 68 landing districts in the country and hence, representing major seafood supply areas to Peninsular Malaysia[9]. The west coast sampling areas such as Pulau Langkawi (Kedah) (lat. 6.35° N, long. 99.80° E) and Pantai Remis (Selangor) (lat. 4.45° N, long. 100.63° E) located at the coast of the Strait of Malacca are known for many industrial activities, particularly release of industrial effluent from various extractive industries. The other two locations Tok Bali (Kelantan) (lat. 5.89° N, long. 102.48° E) and Tanjung Lumpur (Kuantan) (lat. 3.80° N, long. 103.04° E) laid in the east coast facing out to the

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South China Sea, and these are well known for several offshore activities such as petrochemical and metal processing industries. The collected samples were cleaned and thoroughly washed with distilled water to ensure the surfaces were free of dirt and any form of organic matter. Apart from the fish samples that were just chopped into smaller sizes for easy grinding before drying, the molluscs and crustaceans were processed and dried following the procedures described elsewhere[7]. Meanwhile, 4 samples were obtained from each of the investigated marine life, totalling 24 samples in all with masses varying between 93 and 460 g. The samples were then weighed and packed into Marinelli beakers and stored at room temperature for at least 4 weeks before analysis.

Measurement of radioactivity

The samples' radioactivity was determined using a p-type, coaxial, HPGe γ-ray spectrometer (ORTEC; GEM-25P; Serial No. 46-TP22121A; 57.5 mm crystal diameter and 51.5 mm thickness; operating voltage: + 2800 V) with an efficiency of 28.2 %. The energy resolution of the detector was 1.67 keV full width at half maximum at the 1.33 MeV peak of ^{60}Co. Calibrations of the detector have been conducted according to Ref. (7). Hence, activity (Bq kg^{-1}) of ^{226}Ra was determined using the characteristic γ-lines released by its progenies ^{214}Pb (351.93 keV, $I\gamma =$

35.60 %) and ^{214}Bi (609.32 keV, $I\gamma = 45.49$ %); ^{228}Ra by its progenies ^{212}Pb (238.63 keV, $I\gamma = 43.6$ %), ^{208}Tl (583.19 keV, $I\gamma = 85.0$ %) and ^{228}Ac (911.20 keV, $I\gamma = 25.8$ %), while ^{40}K was determined using the single γ-line of 1460.82 keV ($I\gamma = 10.66$ %). Each sample was measured for a period of 24 h (86 400 s). The determination of minimum detectable activity concentration (MDC) and estimation of uncertainties were similar to our previous report[7]. Any activity concentration below MDC (0.103, 0.363, 0.033, 0.064, 0.097 and 1.43 Bq kg^{-1} for ^{214}Pb, ^{214}Bi, ^{212}Pb, ^{208}Tl, ^{228}Ac and ^{40}K, respectively) was considered below detection limit (BDL).

RESULTS AND DISCUSSION

The calculated activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in the samples are presented in Table 1. The mean activity of ^{226}Ra in the fin fishes ranged from 0.74 ± 0.20 Bq kg^{-1} (*Dasyatidae*) to 1.20 ± 0.70 Bq kg^{-1} (*Rastrelliger*). The sampled species regardless of locations show somewhat similar activity concentration of ^{226}Ra except for the *Rastrelliger* (2.19 ± 0.24 Bq kg^{-1}) collected from Pulau Langkawi. The little variations in radium uptake by the studied fishes could possibly be attributed to some biological factors particularly to each fish such as, feeding habits including particulate ingestion with food (in the case of *Rastrelliger*, which has diverse diet

Table 1. Arithmetic mean ± SD of ^{226}Ra, ^{228}Ra and ^{40}K activity concentration (Bq kg^{-1}) and committed effective dose (μSv y^{-1}) due to the ingestion of collected marine animals (d.w.).

Sample type	Radionuclides	Locations (state)				Activity	Activity indices	
		Tanjung Lumpur (Kuantan)	Tok Bali (Kelantan)	Pulau Langkawi (Kedah)	Pantai Remis (Selangor)	Grand mean ± SD (Bq kg^{-1})	D. intake (Bq)	Dose (μSv y^{-1})
Fish (4)	^{226}Ra	0.94 ± 0.15	1.67 ± 0.22	0.50 ± 0.08	0.83 ± 0.16	0.99 ± 0.49	0.09	7.68
(*Formio niger*)	^{228}Ra	0.73 ± 0.10	1.35 ± 0.15	0.47 ± 0.05	0.51 ± 0.10	0.77 ± 0.41	0.07	14.7
	^{40}K	42 ± 2	42.60 ± 2.20	36.50 ± 0.18	31.20 ± 1.70	38 ± 5	3.28	6.56
Fish (4)	^{226}Ra	0.50 ± 0.07	0.85 ± 0.14	0.95 ± 0.15	0.67 ± 0.09	0.74 ± 0.20	0.06	5.79
(*Dasyatidae*)	^{228}Ra	0.28 ± 0.08	0.65 ± 0.13	1.02 ± 0.11	0.68 ± 0.12	0.66 ± 0.30	0.06	12.6
	^{40}K	46.20 ± 2.20	31.80 ± 1.90	58.30 ± 2.80	42.20 ± 2.10	45 ± 11	3.85	7.70
Fish (4)	^{226}Ra	0.78 ± 0.10	1.21 ± 0.25	2.19 ± 0.24	0.63 ± 0.08	1.20 ± 0.70	0.10	9.37
(*Rastrelliger*)	^{228}Ra	0.74 ± 0.10	0.19 ± 0.08	0.59 ± 0.06	0.45 ± 0.05	0.49 ± 0.23	0.04	9.46
	^{40}K	22.10 ± 1.20	38.30 ± 2.10	100.20 ± 4.80	30.20 ± 1.50	48 ± 36	4.12	8.23
Crustacean	^{226}Ra	0.51 ± 0.12	1.16 ± 0.27	0.86 ± 0.21	1.42 ± 0.21	0.99 ± 0.39	0.09	7.70
(4) (*Caridea*)	^{228}Ra	0.19 ± 0.04	0.83 ± 0.11	0.50 ± 0.10	1.74 ± 0.13	0.82 ± 0.67	0.07	15.7
	^{40}K	30 ± 1.0	44.50 ± 0.90	44.70 ± 1.00	17.10 ± 0.60	34 ± 13	2.94	5.88
Mollusc (4)	^{226}Ra	0.57 ± 0.08	2.75 ± 0.78	0.53 ± 0.13	0.57 ± 0.10	1.11 ± 1.10	0.10	8.61
(*Teuthida*)	^{228}Ra	BDL	0.31 ± 0.10	0.16 ± 0.04	0.10 ± 0.04	0.19 ± 0.17	0.01	2.74
	^{40}K	34.70 ± 2.20	72.90 ± 2.10	62.60 ± 0.90	21.30 ± 0.90	48 ± 24	4.13	8.26
Mollusc (4)	^{226}Ra	0.89 ± 0.14	0.75 ± 0.10	0.47 ± 0.09	0.56 ± 0.12	0.67 ± 0.19	0.06	5.20
(*Perna viridis*)	^{228}Ra	0.36 ± 0.07	0.25 ± 0.04	0.14 ± 0.02	0.35 ± 0.08	0.28 ± 0.10	0.02	5.28
	^{40}K	17.60 ± 0.60	46.30 ± 5.70	20.20 ± 1.80	79.80 ± 0.90	41 ± 29	3.54	7.07

The number of samples in parentheses.

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regardless of the prey species, e.g. macroplankton, larvae of fish, and copepods, and habitats), physiological behaviour and the radionuclide distribution within the marine compartments[1]. Table 2 shows that present results of ^{226}Ra are similar with those previously reported from the same locations[8] and from the Indian coastal waters[10, 11] but less than the results from Nigeria[3] and Aleutian Islands[1]. On the other hand, the mean activity of the ^{226}Ra in molluscs (*Perna viridis*), molluscs (*Teuthida*) and crustacean (*Caridea*) are 0.67 ± 0.19, 1.11 ± 1.10 and 0.99 ± 0.39 Bq kg^{-1}, respectively. These results again show that ^{226}Ra is evenly distributed in the water medium and available at every compartment of the aquatic ecosystem in such a way that the differential feeding pattern, physico-chemical natures and morphological appearance (size and length) of these bivalve molluscs and crustaceans significantly cause no meaningful disparity in the accumulation of this radionuclide. In the same vein, there is no difference in the accumulation of this radionuclide between the fin fishes and shell fishes despite their differential habitats and trophic levels within the food web. As shown in Table 2, the present results are significantly differing to the activities of ^{226}Ra in molluscs and crustaceans

available in the earlier studies in Malaysia[7], Nigeria[3] and Bangladesh[12] but show similar activities as reported in other references[8, 13, 14].

The mean activity of ^{228}Ra in fishes ranged from 0.49 ± 0.23 Bq kg^{-1} (*Rastrelliger*) to 0.77 ± 0.41 Bq kg^{-1} (*Formio niger*), which is relatively lower than the ^{226}Ra in fishes. This result supports the nature of solubility of uranium and non-solubility of thorium in water[7]. The results are similar to the previous study conducted on different fishes within the same study locations[8]. However, regardless of fish species and study locations, no significant differences were seen for ^{228}Ra activity in the literature (Table 2) except for the Kerala region, India[15]. On the other hand, the mean activity of ^{228}Ra in the bivalve molluscs and crustaceans ranged from 0.19 ± 0.11 Bq kg^{-1} (*Teuthida*) to 0.82 ± 0.67 Bq kg^{-1} (*Caridea*). The highest activity of ^{228}Ra is obtained in *Caridea* from Pantai Remis. Note that this area is known as the hub of economic activities including the major seaport in Peninsular Malaysia. Apart from this, the fact that the whole parts of this crustacean (*Caridea*) were involved in the analysis could probably be responsible for the relatively high activity of ^{228}Ra in *Caridea* species. Because the *Caridean* shrimp are known for

Table 2. Comparison of the range of activity concentrations of the natural radionuclides in various marine animals as reported in literature.

Sample category	Sample name	Mean activity concentrations (Bq kg^{-1})			References
		^{226}Ra	^{228}Ra	^{40}K	
Fish	*Formio niger*	0.50–1.67	0.47–1.35	31.2–42.6	Present study
		ND–1.3	ND–1.3	ND–57	Amin *et al.*[8], Malaysia
	Dasyatidae	0.50–0.95	0.28–1.02	31.80–58.30	Present study
		24.5–70.0	35.9–62.9	586–836	Ademola and Ehiedu[3], Nigeria
	Rastrelliger	0.63–2.19	0.19–0.74	22.10–100.20	Present study
		ND–1.5	ND–1	48–230	Amin *et al.*[8], Malaysia
		—	—	149 ± 6	Carvalho *et al.*[18], Portugal
		0.118–0.468	—	56.0–70.7	Hong *et al.*[1], USA
		—	—	106–173^a	Gallelli *et al.*[17], Italy
	Not specified	0.105	0.58	64.3 ± 0.5	Narayana *et al.*[10], India
		BDL–0.2	0.3–0.7	—	Rajan *et al.*[11], India
Crustaceans	Shrimp (*Caridea*)	0.51–1.42	0.19–1.74	17.10–44.70	Present study
		—	—	109–155	Gallelli *et al.*[17], Italy
	Prawn	ND–2.1	0.6–1.7	50–147	Amin *et al.*[8], Malaysia
	Prawn	18.0–44.4	26.2–36.5	453–606	Ademola and Ehiedu[3], Nigeria
	Crab	3.88–4.21	1.60–2.03	236–282	Khandaker *et al.*[7], Malaysia
Molluscs	Mussel (*Perna viridis*)	0.47–0.89	0.14–0.36	17.60–79.80	Present study
		2.8–4.3^b	—	23.1–80^b	Alam *et al.*[12], Bangladesh
		0.40–1.38^c	0.49–3.58^c	261–497^c	Kiliç and Çotuk[14], Turkey
		<MDA–1.26	<MDA–2.14	26.3–279.6	Kilic *et al.*[13], Turkey
	Squid	0.53–2.75	BDL–0.31	21.30–72.90	Present study
	Clam	3.57–4.96	1.39–2.17	172–221	Khandaker *et al.*[7], Malaysia

BDL, below detection limit; ND, not detected.
^{a}Activity concentration is due to wet weight of whole fish.
^{b}Activity concentration was due to wet weight of soft tissue of the sample.
^{c}Activity concentration is due to dry weight of soft tissue.

their benthic lifestyle, especially when avoiding the predators, they tend to burrow down in the bottom sea sediments and rocks, filtering on organic particles and algae along with tiny fishes and planktons, which may lead to increase in uptake level of this radionuclide as noticed in this study. However, the obtained activity of ^{228}Ra shows relatively lower value compared with the previous results from this area[7, 8], and consistent with the reported data in Ref. (13).

In contrast, ^{40}K was detected in appreciable amount in all the samples analysed. This is expected since this radionuclide is widely dispersed in marine environment and responsible for the metabolism activities of the organisms inhabiting in this environment[13]. Present measurements show similar mean activity of ^{40}K in the fin fishes and shell fishes, ranging from 38 ± 5 Bq kg^{-1} (*Formio niger*) to 48 ± 36 Bq kg^{-1} (*Rastrelliger*) for fin fishes and between 34 ± 13 Bq kg^{-1} (*Caridea*) and 48 ± 24 Bq kg^{-1} (*Teuthida*) for molluscs and crustaceans. The present results of ^{40}K show similar values with the previous studies in fish[8] but significantly lower values in mollusc and crustaceans for the same locations, which is 197 ± 10 and 261 ± 13 Bq kg^{-1}, respectively[7]. A brief survey of literature shows a heterogeneous distribution of ^{40}K in different regions: a relatively lower mean activity of ^{40}K was reported in fish collected from the water near Vizag, India[16], a similar value in the soft tissue of different mussel from Bangladesh[12] and significantly higher concentrations in fish, crustaceans and mollusc from coastal waters around oil producing areas in Nigeria[3], India[10, 11], Turkey[13], Golden Horn and Bosphorus[14], Northern Italy[17] and North Atlantic ocean[18].

Daily intake of radioactivities and committed effective dose

Irrespective of natural or anthropogenic origin, radionuclides pathway to human is majorly via food chain. The degree of hazards pose by these radionuclides to human depends largely on the type of radionuclides and the duration of exposure. Using some relevant statistical data such as the annual fish landing of 1 103 311 tonnes (1 tonne = 1000 kg) in Malaysia for 2012[9], the total adult population of 21.007846 million (15 years of age and above) in the country[19] and the dose conversion factors taken from the ICRP report[20], the daily intake and committed dose via ingestion of radionuclides by consumption of marine animals were estimated following the relation given in Ref. (7). As shown in Table 1, the average annual committed doses due to the ^{226}Ra, ^{228}Ra and ^{40}K ranged from 5.20 µSv (*Perna viridis*) to 9.37 µSv (*Rastrelliger*), from 2.74 µSv (*Teuthida*) to 15.70 µSv (*Caridea*) and from 5.88 µSv (*Caridea*) to 8.26 µSv (*Teuthida*), respectively. The estimated annual committed effective doses show an order of ^{228}Ra >

^{226}Ra > ^{40}K. The obtained doses are less than the global internal dose of 290 µSv y^{-1} set by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)[21], discarding any significant radiological risks.

CONCLUSION

Owing to their importance in the daily diet of Malaysian population, concentration of naturally occurring radioactive materials in the widely consumed marine animals collected from the coastal waters around Peninsular Malaysia has been determined. The contemporary levels of radioactive materials in the studied marine animals can be considered as low. Committed effective dose due to the consumption of marine animals was estimated to obtain the radiation risks to the populace, and then compared with the world average. This study indicates that the radionuclides intake from the consumption of marine animals collected from the surrounding coastal waters of Peninsular Malaysia yet poses no threat to public health.

FUNDING

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3.2.7 Paper 7 (Submitted)

Heavy metals in human teeth; a bio-indicator of metal exposure to environmental pollution

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Abstract

Human beings are presently exposed to significant levels of environmental heavy metals from rapid urbanization and large-scale industrial activities. Since the metals deposited in tooth dentin during formation and mineralization processes are retained to a large extent, they could be used to trace the record of human exposure to metals. This is because, unlike bone, where the mineral phase is subject to turnover, tooth dentin once formed provides a permanent, cumulative and relatively sound record of environmental exposure to heavy metals. In our research, a total of 50 separate human teeth from patients with variable age, gender, occupation, dietary habits, residency, medical history, etc. were collected with help of the Dental Faculty of the University of Malaya. The teeth were examined using inductively coupled plasma-mass spectrometry (ICP-MS). The concentrations of lead (Pb), mercury (Hg), cadmium (Cd), chromium (Cr), arsenic (As), copper (Cu), iron (Fe), manganese (Mn), bismuth (Bi), barium (Ba), zinc (Zn), strontium (Sr), antimony (Sb), aluminum (Al), tin (Sn) and magnesium (Mg) were analyzed. We found them in trace amount in almost all teeth samples, with the following concentration order As<Mn<Ba<Bi<Cu<Cr<Pb<Fe<Zn<Hg<Sb<Al<Sr<Mg<Sn. The results showed that accumulation of Pb, Hg, As, Cr, Mn, Sr, Ba, Sb, Cu, Zn, Mg and Sn increases with increasing age; however no such trend was noticeable for the other metals. Among the ethnic groups, Chinese teeth showed slightly higher metal concentrations than the Indian and Malay teeth. Generally the female tooth dentin showed higher concentrations than male dentin. The accumulation of metals in different types of teeth varied. Relatively higher concentrations of Hg, Bi, Cu and Sn were found in molars, while Pb, Sr, Sb, Fe, Mg and Zn were relatively higher in incisors. Some elevated levels of concentrations of heavy metals in the tooth dentin reflect the pollution from industrial emissions and urbanization. Human tooth dentin therefore can be

used to obtain chronological information of heavy metal exposure as a reliable bio-indicator of environmental pollution by heavy metals.

Introduction

Human civilization is currently exposed to the highest levels of heavy metals in recorded history. Sources of exposure include anthropomorphic activities such as coal-, oil- and gas-exploration and exploitation, disposal of industrial effluents, advanced agricultural practices (e.g. use of fertilizers, fungicides, insecticides, herbicides and waste water use in irrigation), rapid urbanization, atmospheric deposition of dust/aerosols, vehicular emissions, sewage sludge etc. [1-4]. Environmental pollution is a major problem in Malaysia, not only due to its rapid industrialization and urbanization but also from wind-borne pollution from bordering countries (e. g., Indonesia) [5, 6].

The discharge of heavy metals into the natural environment by geogenic sources or by synthetic activities has numerous environmental consequences, because of their non-biodegradability and persistence. Heavy metals can accrue in the environment such as in the human food chain, entering the human body through the normal ingestion of food and water or by deliberate consumption of soil, through the skin or by inhalation [2, 3, 7-12]. Once in the body they accumulate in various organs including calcified tissue like bones and teeth and pose a risk to human health due to their toxicity and long-term persistence [4, 13-17]. The ingestion of heavy metals by human beings is directly connected to dietary habits and lifestyle [1]. Knowledge of the amount of heavy metals in the body can provide significant information on potential environmental exposure, either at their place of work or elsewhere; their dietary habits and their health [18, 19]. Therefore, it is imperative to assess heavy metals in human populations to evaluate the degree of poisoning by toxic metals.

Evaluation of environmental pollution can be performed using physical and chemical methods and with bio-indicators [20]. Bio-monitoring of heavy metal exposure of human beings gives a sign of an individual's current body burden, which is a function of recent and/or past environmental exposure [21]. Thus, the right choice of and development of suitable biomarkers to assess heavy metal exposure is of crucial importance, for primary prevention, health care management and decision making in public health [22, 23]. Recently, there has been growing interest among researchers on the use of human bodily fluids and organs such as

blood, urine, bone, teeth, nails, hair and saliva as bio-indicators to investigate environmental pollution by means of toxic heavy metals [4, 9, 20, 21, 24-26].

Each of them is associated with some advantages and limitations. Blood and urine data reflects information on recent exposures [9, 25]. Till now, blood-lead levels are widely used as a marker of lead exposure. The half-life of lead in blood is very short (approximately 28–30 days), it is not a reliable indicator of chronic exposure. Hair and fingernails are regarded as medium-range bio-monitoring agents, associated with exposure times from a few months to years [9, 25]. Moreover, they are habitually contaminated by external agents, such as dust or air, hair coloring, shampoo, and nail polish, consequently these samples are often impure and not ideal as bio-indicators [19, 22, 27]. Calcified tissues such as bone and teeth have a high affinity to accumulate heavy metals when they are exposed during development [9, 25, 28, 29]. Bone is a relatively suitable bio-indicator for long-term exposure, but human bone is usually not readily available for sampling and measurement [19, 27]. On the other hand, dental tissues are very hard and similar to the materials making up bone [9, 25, 30, 31]. Unlike bone, in which the mineral phase is subject to turnover, the dental hard tissues (e.g., dentin and enamel) are not subject to significant turnover and therefore provide a permanent, cumulative and quite sound record of past and/or recent environmental exposure to heavy metals [13, 32-35]. Teeth (dentin, enamel or whole teeth) thus offer some advantages as suitable bio-indicators of heavy metal exposures. [4, 7, 9, 13, 20, 21, 23, 26-29, 32].

Teeth are readily accessible biopsy tissues and are physically stable for analysis. These biopsy tissues recently receive substantial attention for research in biological modeling, because of their easy extraction and very low rate of pollutant clearance relative to other organs [27]. Consequently, a precise chronological record of exposure to a number of elements is retained in the hard calcified tissues of the teeth [29]. Furthermore, teeth of different ages of people can be easily accessed to compare the metal concentrations of multiple generations at one time [19].

Many studies have been devoted to analyzing metal concentrations in whole teeth to make correlations between samples and environmental pollution by heavy metals [4, 7-9, 13, 17-19, 24, 25, 28, 34, 36-39]. Data on the heavy metal concentration of tooth dentin is scarce. To increase knowledge of the spatial distribution of elements in each tissue of human teeth, (such as dentin, enamel, pulp and cementum) and their affinity for environmental pollution, it is important to study the elemental concentrations in the dentin and enamel separately.

During the sixth week in utero, dental hard tissues, specifically enamel and dentin, begin to grow and then teeth in each mandible become the deciduous teeth that are later replaced by the

permanent teeth [31, 40]. Dentin, richer in organic content than enamel is biologically more active than enamel. It is a typical composite material, containing inorganic hydroxyapatite crystals and organic collagen matrix proteins [31, 41]. Odontoblasts, situated in the pulp adjacent to the dentin, continuously produce dentin throughout the whole lifespan of a tooth, until it's shed [41]. Protoplasmic protuberances of odontoblasts persist after completion of dentin development and some metabolism is mediated through these growths. Dentin is not affected by the oral environment, since it is surrounded by enamel and cementum [27]. There is no active metabolism of elements occurring after the completion of dental dentin [27]. Consequently, the teeth would appear to be an appropriate long-term bio-indicator of exposure to environmental pollution; an excellent vehicles for pollution studies [28, 31]. Tooth dentin can be a superior bio-indicator of recent health and mineral status; and of environmental pollution by heavy metal since it is deposited during the course of life [24].

The aim of this study is to investigate the heavy metal levels in human tooth dentin. Goals are to understand environmental pollution by heavy metals, to evaluate correlation with a number of parameters; including the ethnicity of the tooth donor, age, sex, tooth condition and tooth type.

Materials and Methods

Sample Collection

To evaluate the effects of different levels of environmental pollution on the accumulation of heavy metals in human tooth dentin, permanent teeth were obtained from adults donors. The extractions were a result of periodontal problems. The donors were in the Klang valley area of Malaysia, where a multi-ethnic population is living. Upon approval by the Ethics Committee of the Dental Faculty, University of Malaya and the donors giving written consent, a total of 50 filling-free permanent teeth (21 molars, 12 premolars, 6 canines and 11 incisors) of various ages and, sex were collected. Prior to the tooth extraction by the dentist, all of the tooth donors were requested to respond a questionnaire regarding their ethnicity, age, gender, profession, working place, smoking and dietary habits, place of residence, medical history etc. Tooth type and position was recorded by the dentist.

Sample Preparation

The processing of teeth for heavy metal analysis was performed under clean and pollutant-free environments in order to reduce external metal contamination. All the vials, glassware and

plasticware were soaked in 10% nitric acid for 24 hours and then exhaustively rinsed several times with ultra-pure water (18.2 MΩ-cm) prior to use for sample preparation [9]. Immediately after tooth extraction, the teeth were kept in sterile plastic containers with a 30% H_2O_2 solution and washed for 2 hours [25]. Soft tissue, gum tissue and blood around the teeth were removed by the H_2O_2 solution and a brush. Each tooth was rinsed several times using deionized water, to further clean any organic matter such as dead tissue from tooth surfaces and remove any remaining H_2O_2. The teeth were then air dried in filter paper. Each tooth crown and root was separated by diamond a saw and the enamel was mechanically separated from the crown dentin with a low speed dental hand piece, under a magnifying glass and stereoscopic microscope. The cementum of the tooth dentin was removed carefully. Finally, the tooth dentin was again rinsed in ultra-pure water (18.2 MΩ-cm), oven dried at 50°C for 1 hour, cooled and weighed. The tooth dentin was placed in a volumetric flask and digested in a mixture of 10 ml of 65% concentrated HNO_3 (SpectrosoL grade) and 3 ml of H_2O_2 (30%) on a hot plate (about 80°C) for about 1 hour and then left overnight at laboratory room temperature. It was again brought to about 80°C for a further 1 hour, to ensure that the specimens were completely dissolved [38]. A mixture of HNO_3 and H_2O_2 is extensively used, because it effectively mineralizes the organic matter and produce less spectral interference in ICP-MS analyses [42]. After digestion the sample solution was cooled, filtered with ashless filter paper (Whatman 540). Ultra-pure water (18.2 MΩ-cm) was added to the filtrate in a volumetric flask, for a total volume of 50ml. It was then labeled and stored in a freezer until analyzed.

Each procedural blank and the standard were prepared using the same volume and acid combinations, following the same procedure used to prepare the tooth samples. In between sample analyses, 2% ultra-pure HNO_3 blanks were analyzed, to clean the system and check whether there was any leftover from the previously analyzed sample [36].

Sample Analyses

Elemental (Lead (Pb), Mercury (Hg), Cadmium (Cd), Chromium (Cr), Arsenic (As), Copper (Cu), Iron (Fe), Manganese (Mn), Bismuth (Bi), Barium (Ba), Zinc (Zn), Strontium (Sr), Antimony (Sb), Aluminum (Al), Magnesium (Mg) and Tin (Sn)) analyses were conducted by inductively coupled plasma mass spectrometry (ICP-MS) (Agilent Technologies 7500 Series, USA). The operating plasma used for the ICP-MS measurement is in Table 1. Calibration of the ICP-MS was performed using multi-element calibration standard 2A solution (10 mg/l of each element) (Agilent Technologies, USA, part no. 8500–6940). All of the samples were analyzed in triplicate. In all cases, 1 blank solution and 5 standards were run with the same

reagents used under the same conditions to control for possible contamination from the digestion procedures. The recovered values of all the metals ranged from 94% to 98% of the certified value. The ICP-MS detection limits for Cr, As, Cu, Mn, Bi, Zn, Sr, Sb, Al, Mg and Sn was 0.0002 µg/g, while for Pb, Hg, Fe, Ba and Cd it was 0.0001 µg/g.

Statistical Analysis

The experimental data were subjected to statistical analysis employing IBM SPSS version–20 software. Arithmetic men (AM), standard deviation (SD), geometric mean (GM), geometric standard deviation (GSD), median, maximum, minimum, skewness and kurtosis were estimated by descriptive statistical analysis. The relationships and degree of association that can exist among the measured heavy metal variables were evaluated by Pearson's correlation analysis. Comparison between ethnics group; age; gender, tooth condition and teeth type was done using the ANOVA (One-way analysis of variance) post hoc Tukey HSD test. The level of significance was set at $p < 0.05$.

Results and Discussion

The overall statistics (such as arithmetic means (AM), standard deviations (SD) geometric means (GM), geometric standard deviations (GSD), medians, maximum and minimum values, skewness and kurtosis) for heavy metal concentrations in the analyzed tooth samples are shown in Table 2. Of 16 targeted heavy metals, a total of 15 metals were detected in almost all tooth dentin samples by our ICP–MS (Table 2). In the analyzed samples, no Cd has been detected, while other metals show concentration in tooth dentin in the following order: As<Mn <Ba<Bi<Cu<Cr<Pb<Fe<Zn<Hg<Sb<Al<Sr<Mg<Sn. It was observed from Table 2 (from its large standard deviation or big difference between minimum and maximum values) that the heavy metals in permanent teeth dentin samples have a broad range of concentrations. This is mainly due to natural variation in trace element concentration between teeth and can be traced to differences in the soils and foods. The differences are due to industrial, urban and agricultural effluents in the environment, or due to the individuals socio-economic condition, place of residence, profession, age, gender, habits (dietary and smoking), tooth type etc. [1, 4, 34]. The skewness and kurtosis (Table 2) of most of the studied samples (except Pb, Mn and Sn) indicate that the distribution of metal concentrations of the various tooth sample is not normal. For this reason, effort was made to estimate the geometric mean of the concentration of the heavy metals (associated with geometric standard deviation) to observe the central tendency of the

metal concentration. It has been shown that the presence of heavy metal concentrations in tooth tissues like dental dentin in humans or animals reflect the deposition of pollutant metals in the environment [34].

Heavy metal such as Pb, Cd, Hg, As, Ba, Sb, Bi, Al, Sr and Sn etc. have no known routine biological function in the human body and indeed can be detrimental even at miniscule quantities. Toxicity or poisoning is usually a result of environmental pollution or chronic intake of foods of high in these metals.

Lead, the most common heavy metal toxicant, is associated with environmental pollution. It mainly accumulated in calcified tissues, such as bone, teeth etc. throughout the life span and becomes an endogenous source of Pb [37, 43, 44]. Lead was found (range 0.27–6.16 μg g^{-1} with a mean of 2.10$\pm$1.32 μg g^{-1} and GM of 1.72 μg g^{-1} with 1.96 GSD) in all analyzed tooth dentin samples. This may be relevant to the industrial, agricultural and domestic solid and liquid effluents that can increase the soil and aquatic lead levels. Subsequent uptake by crops and aquatic animals introduces lead to the food chain. It ultimately enters the human body following consumption of food and beverages. Because of its large scale use, human civilization is exposed to lead and lead derivatives, mainly by ingestion of contaminated food, drinking water and also by inhalation of polluted air [22, 45-47]. Among the donated teeth, the highest concentration of Pb (6.16 μg g^{-1}) was obtained in the carious upper left 2nd incisor of a 69 year old woman; who is a smoker and factory worker. The lowest concentration (0.27 μg g^{-1}) was seen in the healthy lower incisor of a 15 year old non-smoking student. Many researchers have shown an association between exposure to lead and higher dental caries scores [13]. Alomary et al. (2006) [13] reported in Jordan that the mean concentration of lead in tooth samples was 28.91$\pm$13.70 μg g^{-1} dry weight, with a range of 0.74–69.15 μg g^{-1} dry weight. Chew et al. (2000) [38] reported that the concentration in Malaysian teeth (whole teeth) was in the range of 1.7–40.5 μg g^{-1} with a mean of 4.9 μg g^{-1}, which is very much higher than our findings. Kern and Mathiason (2012) [19] reported values of Pb content in teeth in the range of 0.373$\pm$0.005–15.78$\pm$0.02 μg g^{-1} with a mean of 3.84 μg g^{-1}, which is slightly higher than our values. Lead is an extremely toxic metal and its adverse effects can damage the neurologic, hematologic and renal system of the human body [48-50]. The symptoms are generic, nausea, vomiting, malaise, abdominal pains, constipation, anorexia, dizziness, anxiety, depression, insomnia, restlessness, hyperactivity, disorientation, confusion, muscle and joint pain, weakness etc. are the general complaints. Lead poisoning is a well-known public health

problem. Although, provisional tolerable weekly intake of lead is 1.7 mg [1, 12], no lead concentration can be considered safe for human health.

Mercury is another highly toxic element among the studied heavy metals. Hg was found in the range of 0.05–115.40 µg g^{-1}, with a mean value of 5.05±16.97 µg g^{-1}, GM of 0.92 µg g^{-1} and GSD of 5.22. The level of Hg was found to be somewhat high in the sampled teeth, which may be due to the higher consumption rate of seafood, especially fish by Malaysians. Among the studied teeth the highest concentration of Hg (115.40 µg g^{-1}) was obtained from the carious lower right 3rd molar of a 59 year old female smoker (housewife) and the lowest concentration (0.05 µg g^{-1}) was seen in the healthy upper central right incisor of a 61 year old non-smoker (housewife). Acute lethal doses in humans range from 1 to 4 g (10 to 42 mg Hg/kg for a 70 kg adult) for inorganic mercuric salts [51]. Although there is no safe limit of Hg exposure, the EPA (Environmental Protection Agency) sets a safe limit of 0.1g g^{-1}day^{-1}, the while WHO (World Health Organization) suggests a limit of 1.6 g kg^{-1}week^{-1} (on average) [52]. Exposure to high levels of Hg can permanently damage the brain, kidneys and developing fetus [14, 53]. Mercury can pollute the environment both naturally and anthropogenically. There are numerous anthropogenic sources that release mercury in the environment and contaminate the air, water, food etc. They include coal burning power plants, crematoria, fossil fuel burning, gold and silver mining, the health care sector, various kinds of industrial effluents and even cement production [54, 55]. Hg can occur naturally in the environment from volcanic emissions and then be globally disseminated by winds, coming back to the soil in rainfall and accumulating in the food chain; including aquatic food [54]. The major sources of mercury exposure are from food and seafood consumption (in fish and shellfish) and dental amalgam [14, 46, 53]. It is considered one of the highly neuro, nephro and immunotoxic elements even at low levels of exposure it can cause acute and chronic poisoning with adverse health effects during any period of body development [54]. Frequent and high levels of Hg exposure may damage the CNS and cause mood swings, difficulty walking, hand tremors, slurred speech, hallucinations, loss of memory, impaired concentration and coronary heart disease [52, 53]. Acute poisoning can be fatal. Because Hg is a cumulative toxicant, subacute and chronic poisoning can occur, particularly in occupational exposure [46].

Arsenic is one of the most toxic metals (mainly its inorganic form). It can be found at low levels in water, (especially ground water), soil and air; its presence in the environment creates significant global health concern [46]. Arsenic pollution occurs because of natural phenomena, e.g. volcanic eruptions and soil erosion, and its presence in the environment may be enhanced

by anthropogenic sources. Anthropogenic sources include production of arsenic containing compounds, advanced agricultural practices (e.g., fungicides, herbicides, pesticides, algaecides, sheep dips, wood preservatives, dyes), production of glass, paper and semiconductors; metal smelting, ore processing, waste disposal and coal burning in power plants [46, 48, 56]. Humans are exposed to elevated levels of arsenic via contaminated drinking water (especially groundwater rich in arsenic), using polluted water for food preparation and irrigation of food crops, ingestion of food and soil rich in arsenic and smoking tobacco [46, 48, 56]. Arsenic was found in 33 out of 50 tooth dentin samples with at mean concentration of 0.50 ± 0.20 $\mu g\ g^{-1}$ (range: $0.17-1.10$ $\mu g\ g^{-1}$), a GM value of 0.45 $\mu g\ g^{-1}$ and a GSD of 1.67. The highest As concentration (1.10 $\mu g\ g^{-1}$) was found in the lower right 2nd molar of a 57 year old low-income non-smoking female; (but her husband is a chain-smoker) and the lowest value (0.17 $\mu g\ g^{-1}$) was seen in the upper left 3rd molar of a non-smoking female office secretary. Amr (2011) [8] reported that the mean value of As in Egyptian teeth is 0.02 ± 0.007 mg kg^{-1}, with a range of $0.014-0.027$ mg kg^{-1}, which is much lower than our measured values of Malaysian tooth dentin. Jones (2014) [57] from Medellin, Colombia reported that the concentration of As in human teeth is $0.0.05\pm0.0.03$ mg kg^{-1} with a range of $0.01-0.16$ mg kg^{-1}, which is also lower than our results. The WHO recommended guideline for arsenic in drinking water is 10 $\mu g\ l^{-1}$ [56]. In various foods, its concentration can be in the range of $20-140$ ng kg^{-1}. Exposure to high levels of As may result several human health effects, including increased risk of carcinogenic and other systemic health effects. Acute arsenic poisoning causes rapid onset of symptoms, such as vomiting, abdominal pain and diarrhea, followed by numbness and tingling of the extremities, muscle cramping; and in extreme cases, death. The first symptoms of chronic (long-term) exposure to high concentrations of arsenic are generally skin lesions, which include pigmentation changes and keratoses on the palms of the hands and soles of the feet. These symptoms typically occur after $5-15$ years of exposure and may perhaps be a forerunner of skin cancer [56]. Chronic exposure to arsenic may cause various health problems such as cardiovascular and peripheral vascular disease. E.g. blackfoot disease, neurologic and neurobehavioral disorders, diabetes, developmental anomalies, hearing loss, portal fibrosis, hematologic complaints, brain cell damage, damage to the liver, hypertension and carcinoma [46, 56]. Skin, lung, liver, urinary bladder, kidney and colon cancers are the most significant consequence of chronic exposure to arsenic [46, 56].

Chromium is one of the most abundant naturally occurring metals in the earth's crust, where it is amalgamated with oxygen and iron in the form of chromate ore. Cr(III) is a necessary

dietary mineral in low doses for the maintenance of carbohydrate, lipid and protein metabolism. Cr(VI) and its compounds are known carcinogens (1000 times more toxic than Cr(III)) and therefore there is concern regarding continual low-level exposure to chromium, both occupationally and environmentally [46, 58, 59]. In our study, Cr was found in the range of 0.21−8.38 µg g^{-1}, with a mean of 1.51±1.68 µg g^{-1}, GM value of 0.99 µg g^{-1} and GSD of 2.38; in 41 out of 50 tooth dentin samples. The highest Cr concentration (8.38 µg g^{-1}) was found in the healthy upper right 1st premolar of a man age 56 who is a truck driver and chain smoker. The lowest concentration (0.21 µg g^{-1}) was found in the healthy lower incisor of a 15 year old non-smoker school student. The upper limit of dietary intake of Cr is in the range of 50−200 µg day^{-1}, while the WHO suggested limit of Cr(VI) in drinking water is 50 µg l^{-1} and the workplace (8-hour time weighted average) exposure limit of Cr(III) is estimated to be 0.5 mg m^{-3} and 0.05 mg m^{-3} for Cr(VI) [46, 58]. By way of comparison, Amr (2011) [8] reported an average concentration of Cr of 0.05±0.03 mg kg^{-1}, with the range of 0.03−0.11 mg kg^{-1}, in Egyptian teeth; which is lower than our findings in tooth dentin. Jones (2014) [57] from Medellin, Colombia reported that the concentration of chromium in human teeth is 0.19±0.28 mg kg^{-1} with the range of 0.00−1.18 mg kg^{-1}, which is within the range but slightly lower than our results of Cr concentration in Malaysian teeth. On the other hand Malara et al., (2006) [60] from Poland reported the concentration of Cr in non-carious and carious tooth samples as 5.75±1.53 µg g^{-1} (range: 3.06−8.67 µg g^{-1}) and 6.00±1.65 µg g^{-1} (range: 3.05−10.44 µg g^{-1}), respectively, which are within the range but the mean values are higher than our findings. Cr(VI) in the human environment almost entirely originates as a result of anthropogenic activities. There are numerous sources, production of chromates and bichromates, stainless steel, welding, chromium plating, production of ferrochrome alloys, chrome pigment, leather tanning, textile production, coal and oil combustion, cement production, metal refining, chemical factories and waste incineration, etc. Cr enters into the air, soil and water as well as human foodstuffs, especially agricultural crops, consequently it creates various adverse health effects [46, 58, 59]. Inhalation and dermal contact are the main routes of occupational exposure; industrial workers producing and using Cr are at the highest risk of its adverse effects, while the general population is exposed to Cr most often through the ingestion of soil and food or food supplements, inhalation of ambient air and drinking of water containing chromium [46, 58, 59]. On an average the daily dietary intake of chromium is estimated to be <0.2−0.4, 2 and 60 µg from air, water and food, respectively; typically, most fresh foods contain <10−1300 µg kg^{-1} chromium [46, 59]. Environmental and occupational exposure to Cr(VI) and its

compounds is recognized as having multi-organ toxicity, for instance renal damage, asthma, allergy and lung and respiratory tract cancers. Conversely toxicity of Cr(III) seems to be low though extremely high ingested doses of Cr(III) can cause liver and kidney problems. [46, 61]. Acute ingestion (mainly accidental or intentional) of high doses of Cr(VI) compounds can cause severe cardiovascular, respiratory, hepatic, renal, gastrointestinal, haematological and neurological effects that are fatal [46, 59].

Manganese is one of the three poisonous essential trace elements. It is necessary for normal bodily function; while exposure to high levels of Mn can cause a severe neurological health syndrome called manganism [62-64]. In our study, the concentration of Mn in tooth dentin samples was found in the range of 0.14–1.44 $\mu g\ g^{-1}$ with a mean value of 0.69±0.35 $\mu g\ g^{-1}$, GM value of 0.59 $\mu g\ g^{-1}$, with a GSD of 1.80. Among the donated teeth, the highest concentration of Mn (1.44 $\mu g\ g^{-1}$) was found in the healthy lower left 3rd molar of a young man age 19, who is an engineering workshop worker and a smoker. The lowest concentration (0.14 $\mu g\ g^{-1}$) was seen in the healthy lower left 3rd molar of a 43 year old male smoker, who works in an office. The US EPA reference oral dose (RfD) of Mn is 9.8 mg day^{-1} and according to the WHO (1994) report, the range of R_{fD} for Mn is 0.5–5.0 mg day^{-1}. The concentration of Mn in the studied tooth dentin is rather comparable with the reported values of Amr (2011) [8], which is in the range of 0.09–1.2 mg kg^{-1} with a mean of 0.27±0.11 mg kg^{-1}. Arruda-Neto et al. (2010) [9] reported the Mn concentration of adult (age 18-64) human teeth in São Paulo, Brazil is in the range of 0.3045–0.2991 $\mu g\ g^{-1}$, which is lower but within the range of our findings. Manganese can be released into the environment naturally and by human activities. The compounds of Mn occur naturally in the environment as solids in the soil, as small particulate forms in water and in air as dust particles [63]. The concentrations of Mn in the air are enhanced by human activities such as various industrial activities, steel and alloy production, extraction and processing of ore, chemical synthesis, welding, dry-cell battery fabrication, ceramic production, combustion of fossil fuels, manganese pesticides etc. From these sources, Mn can enter the surface, ground and sewage water; accumulate in the terrestrial and aquatic food chain and ultimately enter into the human body [63]. Manganese that is absorbed by the human body is transported via the blood and accumulates in the liver, kidneys, pancreas and endocrine glands [63]. Neurotoxicity of Mn reflects metabolic or excitotoxic insults, for example alterations in neurotransmitter levels or other kinds of brain metabolite disturbances probably happen before structural damage of the brain [62]. Mental disorders, such as loss of memory, apathy and psychosis have been linked to Mn neurotoxicity. The main

symptoms of Mn poisoning are reduced iron metabolism, lung embolism, bronchitis, forgetfulness, nerve damage, psychiatric abnormalities such as hyperirritability, violent behaviors and hallucinations, which are called manganic madness [63, 65]. Chronic toxicity of Mn may arise from long-term inhalation of dust and fumes. The central nervous system is the main site of damage, which can cause permanent disability [62, 63].

Bismuth is not an essential element for the human body to function. The concentration of Bi in our examined tooth dentin sample was in the range of $0.05-7.37$ $\mu g\ g^{-1}$ with a mean value of 0.81 ± 1.08 $\mu g\ g^{-1}$, GM value of 0.50 $\mu g\ g^{-1}$ and GSD of 2.70. The highest Bi concentration (7.37 $\mu g\ g^{-1}$) was found in the carious lower right 2nd molar of a 26 year old woman who was a non-smoker and restaurant worker. The lowest concentration (0.05 $\mu g\ g^{-1}$) was found in the healthy lower canine of a 67 year old smoker, working as an office clerk. Daily dietary intake of Bi is reported as 0.5 $mg\ day^{-1}$ [12]. Concentrations of Bi in our analysed tooth dentin samples were found relatively higher than that reported by Amr, (2011) [8] in Egyptian permanent whole teeth; which was in the range of $1.10-3.01$ $mg\ kg^{-1}$ with a mean of 0.22 ± 0.09 $mg\ kg^{-1}$. Metallic bismuth occurs naturally and anthropogenically as a by-product of copper and lead smelting factories. Because of its widespread applications, Bi has been dispersed in the environment and enters foods and the human body [66]. The toxicity of Bi can be devastating although its poisoning has yet to be well described [66]. Repeated ingestion of Bi subsalicylate may result in bismuth encephalopathy with the symptoms of myoclonus, severe confusion, astasia-abasia and difficulties with language [67]. The Large ingested doses of Bi can be fatal. Bismuth and its salts can cause kidney damage. Initial toxic symptoms are alterations of mood and sleep, lethargy, apathy, anxiety, malaise and irritability. The neurologic signs are ataxia, postural instability, confusion, dysarthria, parasthesias, seizures, and myoclonic jerks. Bismuth and its compounds should be handled with care, because the information of its effects and fate in the environment is very scarce.

In the studied tooth dentin, barium existed almost all samples in the range of $0.08-13.77$ $\mu g\ g^{-1}$ with a mean value of 0.77 ± 2.26 $\mu g\ g^{-1}$ and GM of 0.31 $\mu g\ g^{-1}$ with a 2.60 GSD. The highest Ba concentration (13.77 $\mu g\ g^{-1}$) was obtained from upper right 1st premolar of a 56 year old smoker working as a truck driver. The lowest concentration (0.08 $\mu g\ g^{-1}$) was seen in the healthy upper right 1st premolar of a 45 year old shopkeeper who is a smoker. The concentration of Ba in our analysed tooth dentin samples was lower than that of (mean, 9.5 ± 5.4 $mg\ kg^{-1}$ and range $5.11-17.97$ $mg\ kg^{-1}$) Egyptian permanent whole tooth samples [8]. The recommended tolerable intake limit of Ba and its compounds is 0.02 $mg\ kg^{-1}day^{-1}$ body weight. 11.4 and 57

mg kg^{-1} body weight are the lowermost fatal acute oral doses for barium chloride and barium carbonate, respectively [68]. Barium occurs naturally in most surface waters and also is released in the terrestrial and aquatic environments by industry. Due to its large scale application in industry, anthropogenic sources add significantly to the release of Ba in the environment and pollute the soil, air and water bodies. Drinking water and food are the major ingestion paths of exposure to Ba for the general public [68]. However, the particulate form of Ba can be inhaled through the respiratory tract into the lungs. Barium has a minimal toxicity for general public, but accidental exposure to a high level of Ba may cause acute Ba intoxication [69]. Serious end-points in humans for Ba poisoning resulting from barium and its compounds seems to be hypertension and affects on renal function [68]. Chronic exposure to Ba oxide through inhalation may cause bronchitis, along with cough, phlegm and breath shortness [68]. Accidental or intentional ingestion of high levels of Ba salts such as barium chloride carbonate and sulfate causes gastroenteritis (vomiting, diarrhea, abdominal pain) hypopotassaemia, hypokalaemia, cardiac arrhythmias, hypertension, myoclonus, and skeletal muscle paralysis [68, 69].

Strontium is generally not so toxic, it was found in our investigated dentin samples in the range of 2.14–52.97 µg g^{-1} with a mean value of 9.01±9.48 µg g^{-1}, GM of 6.73 µg g^{-1} and a GSD value of 2.02. Among the donated teeth, the highest Sr concentration (52.97 µg g^{-1}) was seen in the healthy upper left 3rd molar of a 44 year old woman who was a non-smoker and hotel worker. The lowest concentration (2.14 µg g^{-1}) was found in the carious lower left 1st molar of a 36 year old non-smoker student. By the way of comparison, Amr, (2011) [8] reported Sr from Egyptian teeth ranging from 70.2–130 mg kg^{-1} with a mean of 101.2±24.3 mg kg^{-1}, Brown et al. (2004) [24] reported the Sr concentration in teeth in the United Kingdom is in the range of 52–262 mg kg^{-1} and in Ugandan primary teeth is in the range of 97–244 mg kg^{-1}, which are much higher than in our present study of tooth dentin samples in Malaysia. Appleton et al. (2000) [34] reported the Sr concentration of mammalian teeth (Bank voles) in the range of 37.4±9.27-114±24.4 µg g^{-1}. Strontium and its compounds are present naturally the environment such as in air, water, rocks and soil, whereas the main sources are anthropogenic. The disposal of electronic/electrical devices containing a variety of materials including Sr (e-waste) landfills possibly contaminates the ecosystem and threatens human health [70]. The levels of stable Sr in air due to burning of coal and oil and the concentration of Sr in soil may increase from the dumping of coal ash, incinerator ash and industrial waste [71]. Food and drinking water is the main source of Sr in human body. Fish and fish products especially shell

fish is the major source of Sr that enters the human body following their consumption [16]. Strontium can accumulate in the tissues especially the bones and chronic or excess exposure may cause metabolic dysfunctions and other harmful effects on human health; for example osseous mineralization complaints [16, 72]. Excessive ingestion of Sr can diminish the content of calcium in bone and cause hypocalcemia [70, 71]. It can be the cause of bone disease and weaken the bone (osteomalacia). Patients requiring hemodialysis due to emerging chronic renal failure are vulnerable [70]. Inhalation of strontium chromate is very harmful and it's considered a carcinogen because it is associated with chromium but not strontium itself [71].

Tin has no known biochemical function in the human body. As can be seen from Table 2 the amount of Sn in tooth dentin ranges from 11.57–239.05 $\mu g\ g^{-1}$ with a mean value of 73.07±60.48 $\mu g\ g^{-1}$, GM of 51.18 $\mu g\ g^{-1}$ and with a GSD value of 2.40. These values of Sn are extremely higher than any other studied metals. Malaysia was once the third largest (40% of world production) tin producer in the world. One possible reason for the high concentration of Sn in human teeth may be the contamination of soil and vegetation via tin mining activities. Another possible reason for high levels of Sn in tooth samples may be the extensive consumption of canned food. Among the tooth donors, the highest concentration of tin (239.05 $\mu g\ g^{-1}$) was found in the healthy upper left 3rd molar of a 44 year old woman who was a non-smoker and hotel worker, The lowest concentration (11.57 $\mu g\ g^{-1}$) was found in the healthy upper left 1st premolar of a 47 year old woman who was a non-smoker and housewife. Jones (2014) [57] reported that the concentration of tin in Medellin, Colombia was in the range of 0.00–230.81 $mg\ kg^{-1}$ with a mean of 4.51±29.26 $mg\ kg^{-1}$, which is lower than our findings. Provisional tolerable intake of Sn is 14 $mg\ kg^{-1}$ and recommended maximum permissible limits are 250 $mg\ kg^{-1}$ for solid food and 150 $mg\ kg^{-1}$ for beverages [44, 73]. Tin may also be discharged into the ecosystem from different anthropogenic sources and human utilization [74]. Tinplate is extensively used for food and beverage packaging, which may result in some of the tin being dissolved into the food content, especially when plain uncoated internal surfaces are used for cans [73]. The routes of tin into the human body is by ingestion, inhalation or dermal absorption. The consumption of excessive canned food can cause digestive disturbances with signs of acute gastroenteritis such as nausea (97%), vomiting (70%), abdominal cramps (87%), headaches (57%), diarrhea (33%) and fever (13%) [44, 73, 74]. Tin poisoning also alters the activities of some enzymes, it affects the metabolism of zinc, copper, iron and calcium and it modifies the concertation of several other elements in the tissue of organs [65, 73].

Antimony is regarded as a non-essential element in the human body, its toxicological behavior is comparable to that of arsenic and bismuth [75, 76]. In our study, the range of Sb concentration in human tooth dentin was found to be 0.46–141.32 µg g^{-1} with a mean of 6.61±19.78 µg g^{-1}, GM value of 2.88 µg g^{-1} and with a GSD of 2.87. Among the investigated teeth the highest antimony concentration (141.32 µg g^{-1}), was found in the carious upper left 1st incisor of a 71 year old woman who was a non-smoker and jewelry worker. The lowest concentration (0.46 µg g^{-1}) was seen in the carious lower right 1st molar of a 51 years man who was a non-smoker and office worker. The tolerable limit of Sb is reported as 5 mg/kg dry weight, whereas the WHO suggest a tolerable daily intake of Sb of 0.6 µg kg^{-1} body weight [77, 78]. In comparison, Jones (2014) [57] reported that the concentration of Sb in tooth enamel in Medellin, Colombia was in the range of 0.00–1.01 mg kg^{-1}, with a mean of 0.05±0.15 mg kg^{-1}, which is very much lower than our findings. In the environment, antimony occurs naturally, but it also contaminates the environment through various human activities: waste incineration, mine sites, metal processing (smelters), refining, coal burning or fly ash when ores containing Sb are smelted that (most of the antimony emitted then is in the form of antimony trioxide) [76, 78, 79]. The main sources of Sb in urban dust/soil are from vehicular emissions/and or release of aerosolic Sb. Sources are vehicle exhausts, abrasion of Sb from brakes, tires, motor bearings, from the wearing of rails, wheels and cables of running trains, street surfaces and dusts from various industries [75-79]. One of the important uses of Sb is as a turbidifier in white enamel and as an initiator or additive in the production of polyethylene terephthalate (PET), a widely used plastic for bottling water or beverages and for food trays [76, 78]. Due to wide use antimony can enter into the environment and pollute the air, water, soil and food. It enters the human body via inhalation, ingestion and absorption by the skin of occupational workers [78]. Chronic Ingestion of antimony (oral) may cause nausea, vomiting, liver damage and cardiotoxic effects, and also increased occurrence of lung, liver and bile cancers [79, 80]. Inhalation of air containing Sb results in eye and lung irritation, stomach pain, heart and lung damage, diarrhea, vomiting and stomach ulcers. Long-term dermal contact with antimony may result in skin irritation [79].

In this study investigation of copper has been included, because an overload of Cu in the human body may cause Wilson's disease and can be the reason of life threatening. The studied Cu concentration in tooth dentin was found in the range of 0.12–12.65 µg g^{-1} with a mean value of 1.23±2.01 µg g^{-1}, GM value of 0.71 µg g^{-1} and a GSD of 2.56 GSD. The highest concentration of Cu (12.65 µg g^{-1}) was found in the carious lower right 1st molar of a 51 year

old man who is a non-smoker and office worker. The lowest concentration (0.12 µg g^{-1}) was seen in the carious lower right 2nd molar of a 62 year old smoker who is a business man. The tolerable daily intake of Cu is 2 mg l^{-1} [65, 81]. Amr (2011) [8] reported that the concentration of Cu in Egyptian permanent whole teeth was in the range of 1.4−26.1 mg kg^{-1}, with a mean of 9.2±11.4 mg kg^{-1}, which is higher than our findings. Chew et al. (2000) [38] reported the Cu concentration in Malaysian teeth (whole teeth) in the range of 0.1−6.0 µg g^{-1} with a mean of 0.29±0.03 µg g^{-1}; which is slightly lower than our results. Copper is an important metal needed in the body as a cofactor and/or structural constituent of many metalloenzymes, at the same time excessive levels may lead to toxicity [81]. Generally an adult human body contains 80 mg of Cu, (the highest concentration is in the liver and brain) [82]. The Cu discovered in tooth samples may come from natural and/or anthropogenic release of Cu into the environment which contaminate food, drinking water, air and ultimately enter into the human body following their consumption and inhalation. Airborne Cu is mostly from the combustion of fossil fuels [63]. Copper is an essential element for human life, but its overdose may result in anemia, allergies, hair loss, arthritis, autism, acne, adrenal hyperactivity and insufficiency, cancer, depression, diabetes, dyslexia, fatigue, bone fracture, heart attacks, headaches, anxiety, hypertension, infections, kidney and liver dysfunction, strokes, tooth decay, vitamin C and other vitamin deficiencies [83]. Chronic poisoning by copper causes Wilson's disease, which is characterized by liver poisoning, brain damage, hepatic cirrhosis, renal disease, copper deposition in the cornea and many neurological/psychiatric problems [63, 81]. Accidental high intake of Cu can cause liver and kidney damage and even death [63, 65].

Zinc is a comparatively low toxicity essential trace element and the human body maintains homeostasis over a wide exposure range. However, at the cellular level Zn impacts survival and can be a critical controller of apoptosis along with neuronal death which follows brain injury [84]. The studied Zn concentration in tooth dentin was found in the range of 0.22−15.66 µg g^{-1} with a mean value of 3.99±3.47 µg g^{-1}, GM value of 2.75 µg g^{-1} and 2.62 GSD. Among the donated teeth the highest concentration of Zn (15.66 µg g^{-1}) was seen in the healthy upper right 1st premolar of a 56 year old man who is a smoker and truck driver. The lowest concentration (0.22 µg g^{-1}) was found in the carious lower left 1st molar of a 36 year old non-smoker male student. The normal human body contains 2−3 g of Zn and about 90% of zinc is found in muscle and bone, the rest other body organs; such as the liver, prostate, kidney, brain, gastrointestinal tract, lung, pancreas and heart [38, 84]. In workplace air for the duration of an 8-hour work day, the tolerable exposure limit of Zn oxide by inhalation is 5 mg m^{-3}, whereas

its (Zn) recommended dietary allowance is 11 mg day^{-1} for men and 8 mg day^{-1} for women [84]. Amr (2011) [8] reported that the concentration of Zn in Egyptian permanent whole teeth was in the range of 124.6–235.7 mg kg^{-1} with a mean of 178±44.6 mg kg^{-1}, which is very much higher than our findings. Chew et al. (2000) [38] reported that the concentration of Zn in Malaysian teeth (whole teeth) was in the range of 93.4–182.5 µg g^{-1} with a median value of 123 µg g^{-1}; which is extremely higher than what we found in Malaysian tooth dentin. In comparison to Kern and Mathiason (2012) [19] (their range of concentration: 44±2–227.23±0.02 µg g^{-1} with a mean value of 100.49 µg g^{-1}), our Zn concentration values are greatly lower. On the other hand Kumagai et al., from Japan (2012) [27] reported the average concentration of Zn in tooth dentin is 0.42±0.07 mg g^{-1} for men and 0.44±0.08 mg g^{-1} for women, which are very much higher than that of our findings. In the present study, the low Zn concentrations can be linked with Zn deficiency and degenerative diseases, such as inflammatory joint disease. Zinc can enter the environment by way of natural processes, and by its utilization and in anthropogenic activities. Nutritionally, Zn is an essential trace metal for the body to function, deficiency or excess of this metal may create adverse health effects. Zn is released into the atmosphere in its' oxide form. The engineered nano-zinc oxide (nZnO) is increasingly used in sunscreen products is being released directly into aquatic environments; especially marine environments. It is highly soluble in seawater and can create adverse health effects. nZnO (also ZnO) is considered a respiratory toxicant that causes metal fume fever, leads to oxidative damage and inflammatory responses in the vascular system and lungs [85, 86]. Inhalation of polluted dust and fumes through the respiratory tract, ingestion of foodstuff and drinking water and absorption via the skin are the three main routes of entry for Zn into the human body. Inhalation of Zn-containing smoke usually from industrial processes such as galvanization, smelting or welding, mainly affects occupational workers. It causes the acute syndrome of metal fume fever. Symptoms begin a few hours after the acute exposure, such as fever, nausea, fatigue, muscle soreness, and respiratory effects (for instance cough, chest pain and dyspnea accompanied by increase in bronchiolar leukocytes). Exposure to high doses of Zn causes acute intoxication, prolonged high doses of Zn inhibit the uptake of copper and iron; and also interferes with δ-aminolevulinase more than Pb. The brain is the main organ where zinc is highly involved in cell death. Generally, Zn is not a causative agent for cancer but extremely high doses and chronic intake may have a risk for prostate cancer [84].

Aluminum is considered as a possibly beneficial micro-minerals, even though its' role in the human body yet is to be established [1, 46, 83]. Among the studied metals Al was found in

moderate quantities, with the range of 0.06–66.70 µg g^{-1} with a mean of 7.34±13.55 µg g^{-1}, GM value of 1.62 µg g^{-1} and GSD of 7.30 in all analyzed tooth dentin samples. The highest Al concentration (66.70 µg g^{-1}) was found in the healthy lower left 2nd premolar of a 30 year old man who is a mechanic and smoker. The lowest concentration (0.06 µg g^{-1}) was found in the healthy lower left 2nd premolar of a 15 year old non-smoking student. One can be exposed to Al by foods and beverages, breathing air and by skin contact. Amr (2011) [8] stated that the concentration of Al in Egyptian whole teeth is in the range of 27.5–84 mg kg^{-1} with a mean of 51.4 mg kg^{-1} which is higher than our present findings. It is estimated that the whole body Al burden is about 80 mg [87]. The mean intake of Al from food for males is 7.2 mg day^{-1} and for females is 8.6 mg day^{-1}, 0.16 7.2 mg day^{-1} from water and from inhalation exposure is 0.06 mg day^{-1}. The provisional tolerable weekly intake is 7.2 mg day^{-1} [1, 87]. Aluminum is widely distributed as the most abundant metal in the earth's crust. It is an extensively used and frequently found element. Al enters the environment due to acidic precipitation and direct anthropogenic discharges of it and its compounds; related to industrial processes. It's found in air, water and foodstuffs [87]. Due its large scale utilization (from home to office to industries), it can contaminate the environment, such as soil, plants, foodstuffs, drinking water, aquatic organisms etc. People who are exposed to higher amounts of Al may develop Alzheimer's disease. Exposure to lower levels of this metal contribute to Shaver's disease. Pulmonary fibrosis and lung damage have been seen among workers in refining industries. Exposer to finely grained aluminum powder cause pneumoconiosis. Occupational exposure to Al is strongly associated with various neuropsychiatric symptoms; such as memory loss, loss of coordination and balance and other neurological problems. Accidental human poisoning with extreme quantities of Al may result in bone damage and central nervous system failure [87].

Iron is an essential trace element/nutrient necessary to life. It plays various physiological roles such as those linked to haemoglobin, myoglobin, ferritin and a number of enzymes. In our work, the concentration of Fe was found in the range of 0.29–22.71 µg g^{-1} with a mean of 3.67±5.10 µg g^{-1}, GM of 1.85 µg g^{-1} with a 3.15 GSD in all analyzed tooth dentin samples. The concentration of Fe was found in moderate quantities because of its essentiality, natural abundances and broad scale of utilization. The highest iron concentration (22.71 µg g^{-1}) was found in the carious upper left 2nd incisor of a 69 year old woman who is a smoker and factory worker. The lowest concentration (0.29 µg g^{-1}) was found in the carious upper left 3rd molar of a 71 year old non-smoking office clerk. Iron content in an adult human body is approximately 4 g and its deficiency results in anemia and overload causes acute poisoning

[82]. For comparison, Amr (2011) [8] reported that the concentration of Fe in Egyptian whole teeth was in the range of 65.3–122 mg kg^{-1} with a mean of 94.23 mg kg^{-1}, which is very much higher than our studied values. The average daily intake of Fe for adult men is estimated to be 17 mg day^{-1} and for women is 9–12 mg day^{-1} [62]. The reference daily intake for both is 18 mg day^{-1} [44, 62]. Iron and its compounds are released into the environment by natural processes together with its huge utilization in anthropogenic activities. Ground water is contaminated by Fe through leaching and Fe is found in drinking water. It also enters terrestrial and aquatic environments through industrial emissions, natural geological sources, domestic waste discharge, burning of fossil fuel etc. [88]. Intake of excess amounts of Fe has a toxic effect. It may cause in siderosis in the liver, pancreas, thyroid, adrenals, pituitary and heart; rapid increase in pulse rate, coagulation of blood in blood vessels, hypertension, drowsiness, depression, coma, convulsions and respiratory failure. It also affects the gastrointestinal tract and cardiac arrest can occur [44, 88]. Inhalation of excessive amount of Fe can increase the risk of lung cancer among occupational workers [63]. The mean lethal dose of Fe is about 200–250 mg kg^{-1} of body weight, death has occurred from oral ingestion of doses ranging from 40–1600 mg kg^{-1} [61]. Chronic Fe intoxication can result from a genetic and metabolic disease called hereditary hemochromatosis [61, 82].

Magnesium is an essential and abundant metal in the body, which is naturally present in various foods and food products, also available in dietary supplements and some medicines. It functions as a cofactor of many (more than 300) enzymes. In our work, the level of Mg ranges of 3.84–56.20 µg g^{-1} with a mean value of 23.17±12.95 µg g^{-1}, GM value of 19.37 µg g^{-1} and 1.91 GSD. We found the magnesium concentration was quite higher than the other studied metals except tin, because of its importance in the body's functions. The highest concentration of Mg (56.20 µg g^{-1}) was seen in the carious lower right 1st molar of a 51 year old man who is a non-smoker and office worker. The lowest concentration (3.84 µg g^{-1}) was found in the carious lower left 1st molar of a 36 year old non-smoking student. The adult human body contains about 25 g of Mg, of which 50%–60% is found in the bones as a surface constituent of hydroxyapatite and rest of the Mg (30%–40%) is in the muscles and soft tissues [89, 90]. In comparison to the reported values of Amr (2011), [8] in Egyptian teeth (the range of Mg concentration was 1500–3700 mg kg^{-1} with a mean value of 2800±478 mg kg^{-1}), our values are extremely lower. The tolerable upper limit of Mg for an adolescent to adults is 350 mg day^{-1} [89]. Magnesium is broadly distributed in plant and human foods and beverages. Green leafy vegetables are vital sources of Mg. Tap, bottled and mineral water also are sources of

Mg. Besides natural sources, magnesium and its compounds can be released into the environment by industry; including production of iron and steel, glass and cement (used as a refractory material in furnace linings), construction of airplanes and missiles etc. Despite having many essential body functions, very, high doses of dietary supplements or medications and other sources of Mg (mainly magnesium chloride, carbonate, oxide and the gluconate form) results in Mg toxicity. It has symptoms of diarrhea along with nausea, abdominal cramping, vomiting, hypotension, hypermagnesaemia, facial flushing, urine retention, depression, lethargy, breathing difficulties, irregular heartbeat, reduction of renal function, kidney failure and cardiac arrest [90].

Correlation Matrix

Pearson's correlation coefficient and Twostep Cluster analysis Dendrogram gives an idea about the possible relationship between the metals: common origin, uniform distribution, similar behaviors and the relationship between the metals. The value of Pearson's correlation coefficient is presented in Table 3. No linear significant positive correlation was noted amongst the studied metals. Hg was found significantly ($p<0.01$) correlated with Cu (0.417), Cr was found significantly ($p<0.01$) correlated with Ba (0.532) and Al (0.534), Mn was significantly ($p<0.05$) correlated with Fe (0.360) and Zn (0.341), Sb was found significantly ($p<0.01$) correlated with Sr (0.410), Sn was found significantly ($p<0.05$) correlated with Sr (0.317), Ba was found significantly ($p<0.01$) correlated with Zn (0.674) and Al (0.367), Fe was found significantly ($p<0.05$) correlated with Zn (0.342), Cu was found significantly ($p<0.01$) correlated with Sn (0.333), Zn was found significantly ($p<0.05$) correlated with Mg (0.357), Al was found significantly ($p<0.05$) correlated with Mg (-0.282) and Sn was found significantly ($p<0.01$) correlated with Mg (0.621). But these correlations are not so strong. These correlations suggest that these metals were derived from anthropogenic sources. The negative correlation of some metals can be attributed to the antagonism of the heavy metals during their absorption in human teeth. On the other hand, the Dendrogram of TwoStep Cluster analysis shows that Pb is correlated with As, Cr, Mn, Sr, Bi, Ba, Fe, Cu, Zn, Sb and Al which suggests that these metals comes from similar anthropogenic origins. Beside this, Hg, Mg and Sn show correlations to one another and may have the same bioavailability.

Several articles reported that some factors such as socioeconomic status, age, gender, tooth type and condition etc. influences the heavy metal concentration in human teeth [9, 13, 17, 27, 91]. Since sample concentrations of each element were diverse, the average concentration for

each factor was calculated (Table 4). There are three main ethnic groups (Malay, Chinese and Indian) living in Malaysia. There is no significant distinct pattern of As, Mg and Sn concentration observed among the three ethnic groups, whereas for rest of the metals (Pb, Hg, Cr, Mn, Sr, Bi, Ba, Sb, Fe, Cu, Zn and Al), there is some distinct pattern of concentrations (Fig. 1a). Higher levels of Pb, Hg, Cr, Sr, Bi and Sb were found in Chinese teeth, while, Mn, Cu and Al were noticed to be a little bit higher in Indian teeth. Conversely, Ba was higher in Malay teeth, whereas similar concentrations were observed in the rest of the metals. This pattern of metal concentrations among the ethnic groups may be related to their socioeconomic status, dietary habits, profession etc. [13, 27]. On the other hand statistical analysis (ANOVA–one way analysis of variance) showed that there are no significant differences ($p > 0.05$) in metal concentrations among the ethnic groups except for Pb. Only Pb is significantly ($P < 0.05$) higher in Chinese teeth than in Malay teeth.

The concentrations of most of the heavy metals increases (Fig. 1b) with older ages. Pb, Hg, As, Cr, Mn, Sr, Ba, Sb, Cu, and Zn were observed to be higher for age>50 years. Conversely, Sn and Mg were noticed little bit higher in the age group of 31-50 years, whereas, only Bi, Fe and Al were found slightly higher in the age group 15-30 years. The results of age dependency of metal accumulation in human teeth agrees with some similar studies [13, 19, 27]. But, statistical analysis (ANOVA) showed that only the concentration of Pb has significant ($P < 0.05$) difference among the age groups (50 years> 31-50 years> 15-39 years), while for the other heavy metals the age-related concentration difference is not statistically significant ($P > 0.05$).

Figure 1c showed that the accumulation of Pb, Hg, As, Mn, Sr, Sb, Fe, Zn Mg and Sn are higher in female teeth i.e. sex dependent but only Cr, Cu and Al were found higher in male teeth. Almost similar concentrations were observed in the case of Bi and Ba.

Moreover, Pb, Hg, Bi, Sb, Cu and Sn concentrations in carious teeth were comparatively higher than those of non-carious (healthy) teeth as shown in Fig. 1d. Furthermore, As, Cr, Mn, Sr, Ba and Al were found to be little bit higher in non-carious teeth. An almost similar pattern of concentration was observed in case of Fe, Zn and Mg.

Figure 1e showed that the accumulation of metals in different types of teeth varied. We found differences in metal concentrations between the tooth groups which support the findings of [17, 92]. Relatively higher concentrations of Hg, Bi, Cu and Sn were found in molar groups and Pb, Sr, Sb, Fe, Mg and Zn in incisors. The differences in relative contents of heavy metals result from complex processes based on structural and developmental differences between the

teeth types [92]. Statistically (by ANOVA), the difference of metal concentration among the tooth type (position) was not found significant (P>0.05).

Conclusion

Accurate determination of heavy metal levels in human teeth is important due to their toxicity and harmful effect on human body. This study identified a total of 15 heavy metals in almost all tooth dentin samples. No Cd was detected in any of the analyzed samples. Among the analyzed samples and metals, As<Mn <Ba<Bi<Cu<Cr (descending order) and had the lowest concentration in tooth dentin compared with the heavy metals Pb<Fe<Zn<Hg<Sb<Al<Sr (moderate quantities); while <Mg<Sn had the highest levels. Pearson's correlation and Dendrogram of TwoStep Cluster analysis shows that Pb is correlated with As, Cr, Mn, Sr, Bi, Ba, Fe, Cu, Zn, Sb and Al, which suggested that these metals comes from a similar anthropogenic origin. Among the ethnic groups, higher levels of Pb, Hg, Cr, Sr, Bi and Sb were found in Chinese teeth, while, Mn, Cu and Al were noticed to be a little bit higher in Indian teeth. Overall, Chinese teeth contained a little bit higher concentration of heavy metal than the Indian and Malay teeth. The concentration of most of the heavy metals (Pb, Hg, As, Cr, Mn, Sr, Ba, Sb, Cu, and Zn) increases with older ages (>50 years). Concentrations of Sn and Mg were found little bit higher in the age group of 31-50 years, while Bi, Fe and Al were found slightly higher in the age group of 15-30 years. In most of the cases, female tooth dentin showed higher metal concentrations than that of male tooth dentin. In regard to the tooth group, relatively higher concentrations of Hg, Bi, Cu and Sn were found in molars and Pb, Sr, Sb, Fe, Mg and Zn in incisors. Some elevated levels of heavy metals in the tooth dentin reflect the relation to pollution from industrial emissions and urbanization. The results of the present study suggest that human tooth dentin can be used to obtain chronological information of heavy metal exposure and is a stable bio-indicator of environmental pollution by heavy metals.

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Conflict of interest statement

The authors ratify that there are no known conflicts of interest related to this publication and there has been no substantial financial support for this work that could have influenced its result. The authors also state that they have full control of all primary data and that they agree to allow the journal to review their data if requested.

Author contributions

Conceived and designed the experiments: MUK, YMA, MSF, KhA. Performed the experiments: KhA NABB. Analyzed the data: KhA, MSF. Contributed reagents/materials/analysis tools: MUK, YMA, MSF, KhA. Wrote the paper: KhA.

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Figure caption

Fig. 1 Distribution of heavy metal concentrations in teeth dentine in relation to (a) ethnicity, (b) age, (c) sex, (d) teeth condition and (e) teeth types.

List of Tables

Table 1. Operating parameters of the ICP-MS measurement

Plasma condition		Ion lenses		Octopole parameters		Q-pole parameters		Detector parameters	
RF power	: 1550 W	Extract 1	: 2.4 V	OctP RF	: 180 V	AMU gain	: 133	Discriminator	: 8 mV
RF matching	: 1.76 V	Extract 2	: -77 V	OctP bias	: -6V	AMU offset	: 124	Analog HV	: 1710 V
Sample depth	: 8 mm	Omega bias-ce	: -22 V			Axis gain	: 1.0006	Pulse HV	: 1250 V
Torch-H	: -0.1 mm	Omega lens-ce	: 0 V			Axis offset	: 0.02		
Torch-V	: 0.1 mm	Cell entrance	: -20 V			QP bias	: -3 V		
Carrier Gas	: 1.2 L/ min	QP focus	: 5 V						
Makeup Gas	: 0.18 L/min	Cell exit	: -20 V						
Nebulizer pump	: 0.1 rps								
S/C temp	: 2°C								

Table 2. Summary statistics (Sample size 50 each) of heavy metal concentrations in human teeth ($\mu g\ g^{-1}$).

Heavy metal	Statistics of elemental concentrations								
	AM	SD	GM	GSD	Median	Min	Max	Skewness	Kurtosis
Pb	2.10	1.32	1.72	1.96	1.66	0.27	6.16	1.01	0.58
Hg	5.05	16.97	0.92	5.22	0.80	0.05	115.40	5.93	38.17
As	0.50	0.20	0.45	1.67	0.48	0.17	1.10	0.52	-0.45
Cd	ND	ND	ND	ND	ND	ND	ND	ND	ND
Cr	1.51	1.68	0.99	2.38	0.78	0.21	8.38	2.36	5.85
Mn	0.69	0.35	0.59	1.80	0.67	0.14	1.44	0.36	-0.97
Sr	9.01	9.48	6.73	2.02	5.97	2.14	52.97	3.23	11.40
Bi	0.81	1.08	0.50	2.70	0.51	0.05	7.37	4.77	28.32
Ba	0.77	2.26	0.31	2.60	0.27	0.08	13.77	5.10	26.35
Sb	6.61	19.78	2.88	2.87	2.67	0.46	141.32	6.71	46.45
Fe	3.67	5.10	1.85	3.15	1.70	0.29	22.71	2.56	6.67
Cu	1.23	2.01	0.71	2.56	0.64	0.12	12.65	4.50	23.29
Zn	3.99	3.47	2.75	2.62	3.10	0.22	15.66	2.02	4.75
Al	7.34	13.55	1.62	7.30	2.48	0.06	66.70	2.98	9.34
Mg	23.17	12.95	19.37	1.91	22.41	3.84	56.10	0.63	-0.08
Sn	73.07	60.48	51.18	2.40	46.81	11.57	239.05	1.03	0.21

AM stand for Arithmetic Mean, SD represent the Standard Deviation, GM denote the Geometric Mean and GSD stand for Geometric Standard Deviation.

Table 3. Correlation matrix for human teeth samples giving values of Pearson's correlation coefficients for pairs of heavy metals (the bold values represent significantly correlated parameter).

	Pb	Hg	As	Cr	Mn	Sr	Bi	Ba	Sb	Fe	Cu	Zn	Al	Mg	Sn
Pb	1														
Hg	.057	1													
As	.338	-.048	1												
Cr	.136	-.026	.195	1											
Mn	.164	-.114	.110	.155	1										
Sr	.244	-.084	.229	.188	-.122	1									
Bi	-.121	.058	.079	-.034	-.087	-.163	1								
Ba	-.079	-.063	.144	.532**	.246	.025	-.099	1							
Sb	-.026	-.045	.012	.222	-.084	.410**	-.064	-.017	1						
Fe	.165	-.090	.230	.148	.360*	-.004	-.158	.142	-.032	1					
Cu	-.043	.417**	.030	.078	-.105	-.005	.139	-.109	-.081	-.111	1				
Zn	.221	-.005	.147	.265	.341*	.050	-.101	.674**	-.144	.342*	-.045	1			
Al	-.033	-.022	.119	.534**	.219	.054	-.054	.367**	-.018	.183	-.045	.042	1		
Mg	.264	-.098	.226	.180	.206	.150	-.047	.102	-.159	.091	.275	.357*	-.282*	1	
Sn	.269	.135	.022	.189	.085	.317*	.061	-.057	-.012	-.217	.333*	.169	-.211	.621**	1

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

Table 4. Concentrations (μg g^{-1}) of heavy metals in human teeth by factors ((AM±SD, (GM))

Heavy metal	Ethnic			Age (years)			Gender		Teeth condition		Teeth type			
	Malay [19]	Chinese [16]	Indian [15]	15–30 years [14]	31–50 years [13]	>50 year [23]	Male [28]	Female [22]	Noncarious [26]	Carious [24]	Molar [21]	Premolar [12]	Canine [6]	Incisor [11]
Pb	1.46±0.93 (1.22)	2.81±1.50 (2.37)	2.17±1.21 (1.87)	1.19±0.61 (1.03)	1.70±0.94 (1.47)	2.89±1.39 (2.56)	1.98±1.25 (1.60)	2.26±1.43 (1.87)	1.77±0.98 (1.49)	2.46±1.56 (2.00)	1.90±1.00 (11.68)	1.76±1.25 (1.39)	2.22±1.67 (1.76)	2.69±1.65 (2.22)
Hg	2.41±5.74 (0.88)	9.94±28.75 (0.95)	3.34±6.48 (0.96)	0.80±0.65 (0.56)	2.65±6.31 (0.67)	8.98±24.26 (1.47)	1.40±2.28 (0.66)	9.69±25.00 (1.39)	3.13±6.81 (0.83)	7.12±23.55 (1.03)	9.95±25.62 (1.18)	0.91±0.84 (0.64)	1.98±1.31 (1.54)	1.86±3.03 (0.63)
As	0.51±0.32 (0.42)	0.51±0.22 (0.46)	0.49±0.17 (0.46)	0.52±0.27 (0.46)	0.33±0.16 (0.30)	0.57±0.22 (0.53)	0.49±0.23 (0.44)	0.52±0.26 (0.45)	0.53±0.27 (0.46)	0.47±0.19 (0.43)	0.51±0.29 (0.44)	0.46±0.24 (0.39)	0.51±0.218 (0.49)	0.53±0.22 (0.49)
Cd	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Cr	1.31±1.79 (0.87)	1.71±1.70 (1.11)	1.54±1.59 (1.05)	1.18±1.25 (0.83)	1.46±1.26 (1.08)	1.73±2.08 (1.06)	1.68±2.01 (1.02)	1.29±1.17 (0.96)	1.61±1.85 (1.05)	1.40±1.50 (0.93)	1.13±0.65 (0.95)	1.96±2.60 (1.01)	1.38±1.68 (0.92)	1.77±1.80 (1.11)
Mn	0.72±0.39 (9.60)	0.56±0.32 (0.48)	0.78±0.32 (0.71)	0.72±0.39 (0.61)	0.57±0.33 (0.48)	0.74±0.35 (0.65)	0.66±0.39 (0.54)	0.72±0.31 (0.66)	0.73±0.37 (0.62)	0.64±0.33 (0.56)	0.69±0.38 (0.58)	0.69±0.37 (0.59)	0.63±0.30 (0.56)	0.71±0.36 (0.62)
Sr	7.16±3.68 (6.29)	14.39±14.91 (9.79)	5.62±2.97 (4.90)	5.54±2.90 (5.01)	10.21±13.26 (6.84)	10.46±9.41 (7.97)	7.95±7.62 (6.15)	10.36±11.47 (7.54)	9.69±11.85 (6.47)	8.28±6.15 (7.02)	6.42±3.01 (5.82)	8.45±10.36 (5.94)	6.99±4.77 (5.50)	15.69±15.05 (11.34)
Bi	0.80±0.59 (0.59)	0.92±1.77 (0.38)	0.70±0.52 (0.53)	1.23±1.82 (0.71)	0.50±0.59 (0.35)	0.72±0.52 (0.49)	0.79±0.57 (0.57)	0.83±1.52 (0.42)	0.63±0.53 (0.40)	1.00±1.45 (0.63)	1.07±1.52 (0.65)	0.63±0.69 (0.38)	0.78±0.63 (0.49)	0.52±0.31 (0.40)
Ba	1.43±3.61 (0.34)	0.44±0.35 (0.36)	0.29±0.17 (0.25)	0.28±0.16 (0.24)	0.40±0.39 (0.29)	1.28±3.28 (0.39)	0.80±2.55 (0.32)	0.74±1.88 (0.31)	1.22±3.09 (0.39)	0.29±0.15 (0.25)	0.32±0.16 (0.29)	1.41±3.90 (0.34)	0.21±0.07 (0.20)	1.24±2.62 (0.45)
Sb	3.90±4.26 (2.65)	12.62±34.51 (3.72)	3.64±2.98 (2.44)	3.53±2.64 (2.70)	3.77±4.87 (2.35)	10.09±28.82 (3.37)	4.45±4.32 (2.90)	9.36±29.57 (2.86)	4.43±4.54 (2.93)	8.98±28.28 (2.83)	3.95±4.18 (2.62)	3.35±2.76 (2.50)	3.27±2.98 (2.07)	17.08±41.40 (4.83)
Fe	3.06±5.14 (1.45)	4.12±5.53 (2.13)	3.97±4.82 (2.19)	4.20±6.87 (1.74)	3.38±2.89 (2.11)	3.51±5.02 (1.79)	2.28±2.66 (1.35)	5.45±6.75 (2.78)	3.65±4.16 (2.04)	3.70±6.04 (1.67)	2.28±2.49 (1.39)	5.03±5.43 (2.71)	2.35±2.11 (1.57)	5.57±8.31 (2.32)
Cu	1.06±0.98 (0.71)	1.05±1.62 (0.66)	1.61±3.11 (0.77)	1.08±1.04 (0.74)	0.96±0.75 (0.72)	1.48±2.82 (0.69)	1.34±2.41 (0.72)	1.08±1.39 (0.70)	0.89±0.81 (0.66)	1.62±2.82 (0.78)	1.71±3.09 (0.69)	1.05±0.67 (0.85)	1.12±0.91 (0.81)	0.65±0.31 (0.57)
Zn	4.31±4.23 (3.04)	4.11±3.56 (2.63)	3.46±2.23 (2.55)	3.47±2.21 (2.71)	3.55±1.76 (2.87)	4.55±4.65 (2.71)	3.40±2.96 (2.31)	4.74±3.97 (3.44)	4.10±3.92 (2.65)	3.87±2.98 (2.86)	3.57±1.68 (3.15)	4.05±3.97 (2.77)	2.38±2.60 (1.21)	5.61±5.27 (3.29)
Al	6.28±12.42 (1.07)	5.57±9.55 (2.21)	10.55±18.19 (1.96)	9.29±17.52 (2.18)	4.31±6.63 (1.20)	7.86±13.99 (1.69)	9.14±16.70 (1.47)	5.04±7.76 (1.83)	10.14±17.35 (1.89)	4.30±6.76 (1.37)	3.23±4.57 (1.04)	16.98±23.48 (2.75)	9.12±11.69 (2.45)	3.67±4.43 (1.69)
Mg	23.90±12.40 (21.00)	21.94±13.46 (17.61)	23.54±13.86 (19.35)	21.88±9.43 (19.77)	24.13±14.45 (19.31)	23.41±14.33 (19.16)	22.02±13.83 (18.02)	24.63±11.89 (21.23)	23.16±12.39 (19.78)	23.17±13.80 (18.93)	23.78±13.68 (20.33)	20.01±9.19 (17.82)	17.36±13.38 (12.70)	28.60±14.13 (24.35)
Sn	66.58±52.45 (52.45)	77.12±68.59 (52.41)	76.97±64.28 (50.42)	55.82±41.49 (43.05)	83.72±80.81 (50.71)	77.55±57.36 (57.15)	69.54±64.38 (46.85)	77.57±56.29 (57.26)	64.83±61.87 (43.19)	82.00±58.92 (61.50)	86.47±65.37 (61.99)	47.89±39.37 (36.06)	57.84±47.22 (39.35)	83.27±71.87 (60.00)

The number in the parenthesis [ ] in the second rows is the number of sample analyzed.
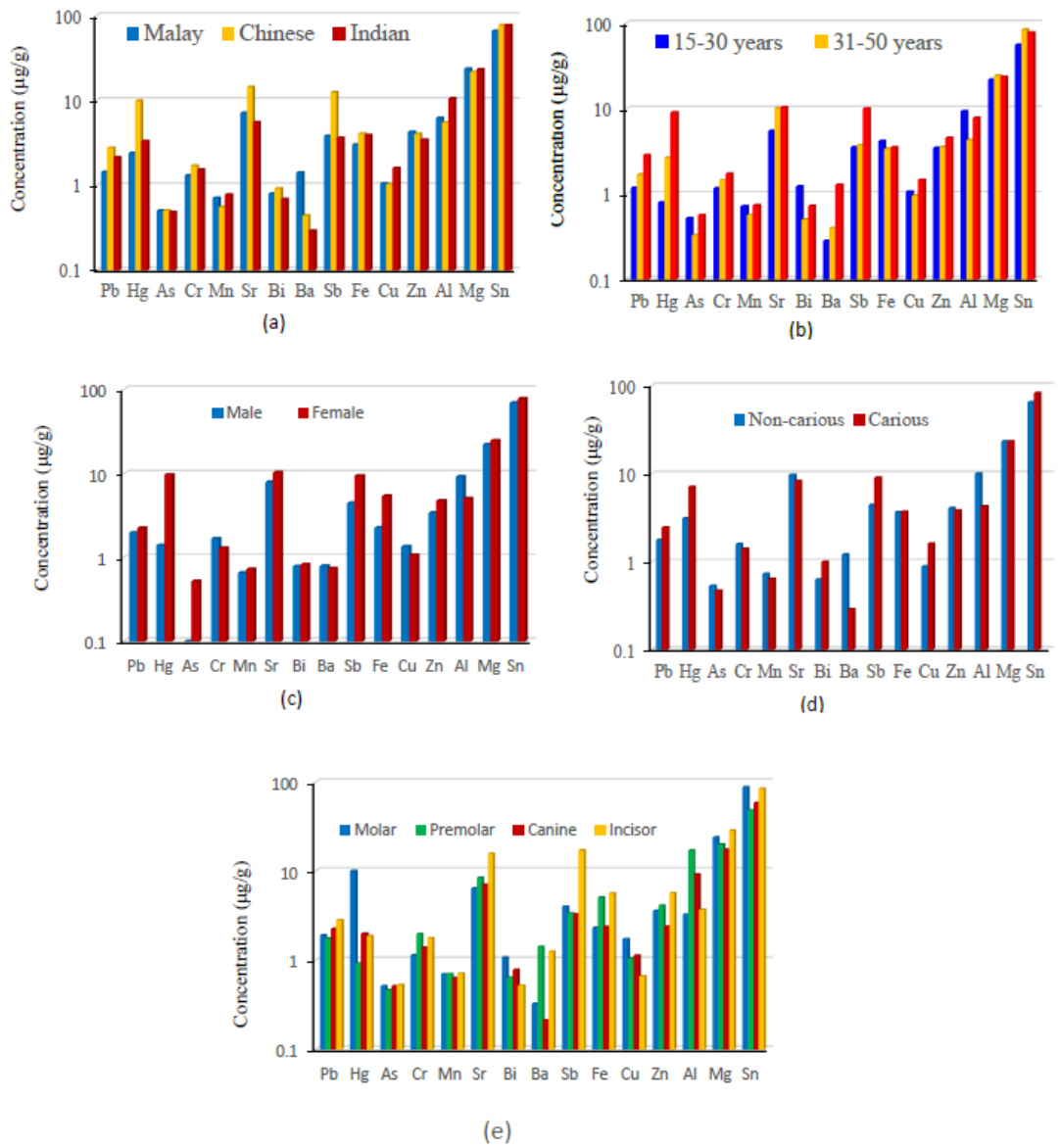
Fig. 1 Distribution of heavy metal concentrations in teeth dentine in relation to (a) ethnicity, (b) age, (c) sex, (d) teeth condition and (e) teeth types.

CHAPTER 4: CONCLUSIONS

In consequence of their importance in the Malaysian diet, natural radioactivity and heavy metal exposure due to consumption of staple food such as rice, vegetables and marine life (fish, molluscs and crustaceans) collected from different regions of Peninsular Malaysia has been assessed. In addition to foods, the levels of heavy metal in human teeth has been evaluated to comprehend the environmental pollution by heavy metals and to evaluate their correlation with a number of parameters including the ethnic of the teeth donor, age, sex, teeth condition and teeth type.

4.1   Natural radioactivity in staple foods

A comprehensive study has been carried out to establish the radioactivity levels and soil-plant transfer factors associated with the natural decay chains of ^{238}U and ^{232}Th and no-series long-lived natural radionuclide of ^{40}K.

The levels of natural radioactivity, radionuclides transfer (TF) from soil-to-rice and committed effective dose to Malaysian population has been evaluated due to consumption of locally grown rice in Sungai Besar (Selangor), Kampung Permatang Tok Labu (Pulau Pinang) and Kampung Sakan (Kedah). The average radioactivity levels of ^{226}Ra, ^{232}Th and ^{40}K in rice were varied as 1.5±0.4 to 2.8±0.7, 3.6±1.4 to 7.5±2.7 and 59.9±6.0 to 92.2±5.4 Bq kg^{-1}, respectively, while in corresponding soils the ranges were 7.2±2.0 to 9.1±1.8, 11.6±1.9 to 20.6±3.1 and 76.5±5.7 to 114.6±6.6 Bq kg^{-1}. The activity levels of ^{226}Ra, ^{232}Th and ^{40}K in the studied rice samples were well below the UNSCEAR (2000) recommended values of 67 82 and 310 Bq kg^{-1}, respectively. The activity levels of ^{232}Th was slightly higher than ^{226}Ra in all soil samples so as to rice samples in all locations, which may be attributed that thorium has a high affinity for the regular exchange sites of the soil and paddy plant absorbs thorium

at a high rate from soil. The erosion process of ^{232}Th largely occurred and it adsorbed in the soil on the spot, whereas ^{226}Ra is easily extracted and migrates with soil water. This may be the possible reasons for a higher accumulation of ^{232}Th than ^{226}Ra. The activity levels of ^{40}K was found remarkably higher than the other radionuclides, which may be attributed to the abundance of ^{40}K in nature. The soil-to-rice transfer factor (TF) in different study locations were estimated to be ranged as 0.16 to 0.32, 0.17 to 0.48 and 0.52 to 1.21, respectively for ^{226}Ra, ^{232}Th and ^{40}K. The TF of ^{40}K was found significantly higher (p < 0.001) than other radionuclides indicating the fundamental nutrient status of potassium for the plants. The high TF of ^{40}K was probably due to the excessive use of potassium-containing fertilizers at the sampling sites, its high mobility in soil, and its subsequent uptake by plants. The estimated soil-to-rice transfer factor for ^{226}Ra and ^{232}Th were found much higher than those compiled by the IAEA (IAEA, 2010). The TFs of ^{226}Ra, ^{232}Th and ^{40}K varied differently among the study locations and within the radionuclides. This may be due to the dissimilarity in locations and the physical properties of the soil, individual chemical properties of the radionuclides, soil amendment such as NPK fertilizers, use of insecticides and herbicides, plant species, climate condition etc. which may have a larger influence on the uptake of radionuclides from soil-to-rice. The total annual effective dose via the consumption of studied rice grains were estimated in the range of 153.4±33.2 − 294.3±49.8 μSv y^{-1} among the study regions, which lies within the range of world average of 290 μSv y^{-1} compiled by UNSCEAR (2000). But the population dose for the rice of Pulau Pinang exceeded the tolerable limit, which may be credited due to the TENORM effect and hence care should be taken to consume this rice. ^{232}Th is the main contributor to the intake dose, with a contribution of about 44% of total estimated dose, followed by ^{40}K (29%) and ^{226}Ra (27%). Among the study locations, rice from Pulau Pinang contribute about 43% of the total dose, followed by Kedah (35%) and Selongor (22%). The lifetime cancer

risk through the rice consumption were below the acceptable limit of 0.29×10^{-3} for radiological risk. Therefore, the intake of radionuclides from the consumption of rice yet pose no significant threat to public health.

Natural radioactivity, radionuclides transfer (TF) from soil-to-edible flora (vegetable) and committed dose to Malaysian population has been assessed due to consumption of various popular vegetables locally grown in Cameron Highlands in Pahang, Klang in Selangor, Pasir Panjang in Negeri Sembilan, Tumpat in Kelantan and Langkawi in Kedah. Among them Cameron Highland is a main vegetables grown area. The average activity concentrations (Bq kg^{-1}) in vegetables were ranged from 0.64 ± 0.16 to 3.80 ± 0.42, 0.21 ± 0.13 to 6.91 ± 0.54 and 85.53 ± 4.04 to 463.8 ± 21.9 for ^{226}Ra, ^{232}Th and ^{40}K, respectively, while in corresponding soils the ranges were 1.33 ± 0.09 to 30.90 ± 1.46, 0.48 ± 0.12 to 26.80 ± 1.34 and 7.99 ± 0.44 to 136.5 ± 6.4, respectively. It was observed that the activity levels of ^{226}Ra, ^{232}Th and ^{40}K in the vegetable were not uniform, varying with the types and locations, and depending upon the geological formation of the area under study. The Highland farms present greater concentrations than the ground farms do. The activity of ^{232}Th was found to be little bit higher than ^{226}Ra in most of the samples, supportive of the fact that the presence of thorium is 1.5 times greater than that of uranium in the Earth's crust. Moreover, the activity of ^{40}K exceeded by far the values of both ^{226}Ra and ^{232}Th, being the most abundant radioactive element present in the environment and it also being noted that potassium is used extensively as part of a NPK fertilizer in intensive farming activities to promote dynamic growth. The soil-to-vegetables transfer factor (TF) for ^{226}Ra, ^{232}Th and ^{40}K in different study locations were estimated to be ranged from 0.03 to 0.96, 0.01 to 0.26 and 0.74 to 38.18, respectively. Soil-to-vegetables TF showed considerable variation with respect to soils, vegetables and locations. The uptake of radionuclides from soil by vegetables depends on various interrelated soil properties including texture, clay content, dominant clay minerals,

exchangeable cations, pH, organic matter content and other environmental conditions. The transfers being greatest for ^{40}K, an expected outcome given the essentiality of this element in support of vigorous growth. The TFs of ^{226}Ra and ^{232}Th were found to be in accord with available literature data but higher than IAEA reported values, the values indicating the mobility of these radionuclides to be low in the studied soils (IAEA, 2010). The committed effective dose via the consumption of studied vegetables were found to be varied from 0.22 to 6.27, 0.36 to 28.1 and 0.58 to 8.99 μSv y^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. Among the studied vegetables, cabbage (60%) is the greatest contributor to the committed dose, followed by tomato (15%), tapioca (10%), spinach (6%), cucumber (6%) and brinjal (3%). This study indicated that, due to the consumption of the investigated vegetables, an individual might receive a total radiation dose of approximately 71.76 μSv y^{-1}, some 4 times lower than the world average (290 μSv y^{-1}). The average excess lifetime cancer risk values were low compared with the acceptable limit of 0.29×10^{-3} for radiological risk, indicating that consumption of the studied vegetables pose no serious health threat to the population.

Natural radioactivity, soil-to-tapioca and soil-to-sweet potato transfer factor of radionuclides and ingestion dose to humans based on natural field conditions in Puchong−former tin mining soil, Batang Berjuntai−lateritic soil and Lembah Beringin−lateritic soil in Selangor and Chemor−peat soil in Perak, Peninsular Malaysia were evaluated. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in tapioca varied as 87 to 127, 3 to 43 and 56 to 844 Bq kg^{-1}, respectively. The respective values for sweet potato varied as 67 to 76, 8 to 17 and 282 to 2483 Bq kg^{-1}, respectively. The activity levels of ^{226}Ra and ^{40}K were observed higher in Puchong, which is a former tin mining area. This could possibly be attributed to the contribution of technologically enhanced naturally occurring radioactive material (TENORM) in this location via former mining

activities, potentially though soil contamination. The soil-to-tapioca transfer factors (TFs) of ^{226}Ra, ^{232}Th and ^{40}K in different study locations were found to be varied from 0.77 to 1.12, 0.28 to 1.7 and 0.11 to 2.0, respectively. The corresponding values for sweet potato were ranged from 0.58 to 0.82, 0.33 to 0.92 and 0.69 to 3.3, respectively. Soil-to-plant transfer of radionuclides is largely affected by numerous factors: the soil physiochemical properties (soil texture, clay, pH, cation exchange capacity (CEC), potassium (K), calcium (Ca) and organic matter content), stage of growth, soil microbial activity, individual chemical properties of the radionuclides, plant species, agricultural practices, climate condition etc. Accordingly, some variation of TFs were observed among the studied regions and samples. The estimated TF values were found to be very much higher than the range of values reported by IAEA (IAEA-tecdoc-1616, 2009). Soil-to-tapioca and -sweet potato TFs for ^{40}K were found to be significantly ($p < 0.05$) higher than that for the other nuclides in almost all locations. The high TFs of ^{40}K was probably due to the excessive use of potassium based fertilizers at the sampling sites, its high mobility in soil and subsequent uptake by plants. Therefore, as compared to uranium and thorium, potassium shows the highest TF. An experimental tapioca field was developed for study of the time dependence of plant uptake. Based on the findings from the experimental field in Dengkil, Sepang, Malaysia, soil-to-tapioca TFs for ^{226}Ra, ^{232}Th and ^{40}K increased from 1.6 to 5.9, 2.0 to 9.2 and 1.9 to 3.0, respectively with time. In most cases, the uptake of radionuclides at the middle or late growth stages produced higher TFs than those at the early growth stage. For all study regions, the annual effective dose from the consumption of tapioca and sweet potato were estimated to be very much lower than the world average 290 μSv y^{-1}and therefore, considered to be safe for human consumption.

The majority of perishable vegetables produced and consumed by the populations of Kuala Selangor, Malaysia were examined to assess their radioactivity levels and

associated radiation hazard following their consumption. The highest concentrations were noticed in water spinach for ^{226}Ra (2.8 ± 2.0 Bq kg^{-1}) and ^{40}K (427 ± 27 Bq kg^{-1}) and in sweet potato for ^{228}Ra (1.8 ± 2.2 Bq kg^{-1}). The lowest levels of ^{226}Ra, ^{228}Ra and ^{40}K were observed in long bean (0.5 ± 0.1 Bq kg^{-1}), cucumber (0.3 ± 0.1 Bq kg^{-1}) and tapioca (63 ± 18 Bq kg^{-1}), respectively. For all varieties of vegetables, variability in radionuclide concentration was observed, also within the same types of vegetable. This can be related to differences in the chemical and physical properties of the different vegetable farm soils, individual chemical properties of the radionuclides, irrigation, use of insecticides and herbicides, plant species, the degree of preparation of the vegetable sample, such as peeling, washing, etc. or even due to the use of phosphate fertilizers. The committed effective dose received by the Kuala Selangor communities (adult) due to the consumption of vegetables was estimated as 16.6 µSv y^{-1} for ^{226}Ra, 23.6 µSv y^{-1} for ^{228}Ra and 57.6 µSv y^{-1} for ^{40}K, with a total of 98 µSv y^{-1}, which was three times lower than the world average value (290 µSv y^{-1}) and presents no significant risk to the health of general public of Kuala Selangor community. Among the vegetables, water spinach (17%), cucumber (15%) and sweet potato (13%) are the greatest contributors to the intake dose for Kuala Selangor communities, followed by lady's finger (12%), spinach (10%), tapioca (10%), brinjal (8%), long bean (8%) and winged bean (5%) and yam (2%). ^{40}K is the main contributor to the intake dose, with a contribution of about 59% of total estimated dose, followed by ^{228}Ra (24%) and ^{226}Ra (17%). For water spinach, cucumber and sweet potato, the more elevated effective dose was not only due to the greater radionuclide concentration (as exemplified by water spinach) but also to the greater consumption rate (as exemplified by cucumber).

Malaysia is among the countries with the highest fish consumption in the world and relies on seafood as a main source of animal protein. Therefore, the radioactivity levels

in widely consumed marine lives such as fishes, crustaceans and molluscs collected from the West and East coast of Peninsular Malaysia has been determined to evaluate the level of human exposure by the way of seafood consumption. The activity concentrations of ^{226}Ra, ^{228}Ra (^{232}Th) and ^{40}K in marine animals varied from 0.6±0.19−7.83±0.78, 0.19±0.17−6.21±0.53 and 34±13−398.6±20.2 Bq kg^{-1}, respectively. Present study shows elevated radioactivity concentrations in the fish of the Straits of Malacca compared to that reported in similar studies in seas elsewhere. The results reflect the contribution of additional technologically enhanced naturally occurring radioactive material (TENORM) pollutants, largely expected to be a result of oil and gas waste streams, related to shipping activities, the route being regarded as the second busiest water channel in the world. In regard to inter-comparison of results from the study locations, the Bagan Lalang area showed the greatest level of radioactivity; here, the narrowing of the Straits, together with a relatively high level of industrialisation including major seaport, urbanization and the effluents that result from these factors are expected to lead to increased concentrations of radionuclides in the marine environment, including fish. Of further note that the activity concentrations of ^{40}K are significantly greater ($p < 0.001$) than that of the other radionuclides for all study locations. The appreciably greater values for ^{40}K are in line with expectation, a considerable fraction of the weight of each sample being accounted for by the fish bones rich as they are in potassium, being a natural isotope of potassium, an essential element for vertebrates. This is expected since this radionuclide is widely dispersed in marine environment and responsible for the metabolism activities of the organisms inhabiting in this environment.

Distribution of radioactivity in the studied marine animals differs with respect to sites of origin and species. The little variations could possibly be attributed to some biological factors particularly to each fish such as, differential feeding habits,

physiological behaviour, morphological appearance (size and length) and the radionuclide distribution within the marine compartments, meaningfully disparity in the accumulation of radionuclides in marine animals. The annual effective doses received by individuals due to the dietary intake of ^{226}Ra, ^{228}Ra (^{232}Th) and ^{40}K via the consumption of marine animals, range from 17.69 to 277.18 μSv y^{-1} with an average of 100.75 μSv y^{-1}, falling below the world average for annual effective dose (290 μSv y^{-1}). Accordingly, the carciogenic risk was found to be well below the acceptable limit of 0.29×10^{-3}, indicating yet safe for human consumption.

4.2   Heavy metal in foodstuffs

Among the foodstuffs, vegetables and fishes are the most exposed foods by environmental heavy metals due to their aerial burden and also physicochemical characteristics and accumulation capabilities. Practically all categories of vegetable produced and commonly consumed by the Kuala Selangor communities (particularly the root, fruit and leafy vegetables that are known to be popular in Malaysia) were examined in order to assess the heavy metal exposure scenarios following their consumption.

The heavy metals ^{202}Hg, ^{137}Ba, ^{85}Rb, ^{88}Sr, ^{209}Bi, and ^{121}Sb were present in almost all varieties of vegetable samples but concentrations were found to be very low, contributing inappreciably to any significant poisonous effect. In the present study, no ^{208}Pb, ^{60}Ni and ^{52}Cr have been detected in root vegetables, while ^{111}Cd, ^{75}As and ^{78}Se were not detected in any of the studied vegetable samples. Conversely, ^{66}Zn, ^{63}Cu, ^{56}Fe and ^{55}Mn were found to be present in moderate quantities in all types of vegetables. Fruit has been found to contain greater concentrations of ^{66}Zn, ^{63}Cu, ^{56}Fe and ^{55}Mn than the root vegetables, noting that ^{52}Cr, ^{55}Mn, ^{56}Fe, ^{63}Cu, ^{66}Zn, and ^{60}Ni are essential metals for human nutrition; however, when consumed at high levels this can result in

health problems. Trace amounts of heavy metals were detected in various types of vegetables, because some metals are naturally present in the vegetables growing soil and further added to pollute by industrial and urban emission, agricultural practices etc. The vegetable garden top soils may be contaminated by heavy metals and hence transmitted the crops through root uptaking. Leafy vegetables accumulate greater levels of heavy metals than roots, which indicates that vegetables bio-accumulated lower quantities of heavy metals from soil and the foliar uptake may be a vital route of metals from the polluted environments. The recorded concentrations of heavy metals showed variations among different vegetables, to be traced to differences in the chemical and physical properties of the different farm soils, by industrial effluents contamination in the producing areas or even due to the use of phosphate fertilizers, herbicides, fungicides and pesticides.

The total dietary intake (μg day^{-1} body weight) of heavy metals via the consumption of vegetables were estimated as ^{24}Mg (40.4), ^{27}Al (26.4), ^{52}Cr (0.004), ^{55}Mn (1.1), ^{56}Fe (2.21), ^{60}Ni (0.004), ^{63}Cu (0.05), ^{66}Zn (0.53), ^{85}Rb (0.14), ^{88}Sr (0.38), ^{121}Sb (0.79), ^{137}Ba (0.02), ^{209}Bi (1.1), ^{202}Hg (0.007) and ^{208}Pb (0.008). Results for the studied heavy metals indicate that vegetables consumed in Kuala Selangor communities contain levels of heavy metals that are well within tolerable limits, and that the contribution of the vegetables evaluated to the dietary intake of heavy metals remain beneath the standards suggested by international organizations and hence contribute any significant poisonous effect in human body.

Malaysia is one of the highest marine animals (fish, molluscs and crustaceans) consumption country in the world and the uptake of heavy metal in general public clearly depends on dietary habit. Due their pathway into the daily diet of the local population, heavy metal concentrations have been evaluated in marine fishes collected

from the Straits of Malacca, west coast of Malaysia which receives pollution as a result of various industrial and anthropogenic sources. The heavy metal (mg kg^{-1}) levels were found as: ^{75}As (0.3249–0.7485), ^{208}Pb (0.02), ^{202}Hg (0.0247), ^{27}Al (3.115–4.489), ^{52}Cr (0.0108–0.0516), ^{88}Sr (19.56–27.68), ^{59}Co (0.0007–0.0038), ^{63}Cu (0.0712–0.1972), ^{55}Mn (0.4372–1.841), ^{66}Zn (5.607–15.35), ^{78}Se (0.035–0.0938), ^{56}Fe (3.546–4.857), ^{85}Rb (0.0978–0.2312), ^{137}Ba (0.2845–0.6597), ^{209}Bi (1.598–4.143) and ^{24}Mg (80.43–115.0). Most of the heavy metal levels in marine fish samples were found to be higher than that found in vegetable samples. Present study identifies the presence of a wide range of non-essential metals in the selected fish, albeit at relatively low levels compared to studies conducted elsewhere, varying in concentration from region to region and element to element. While again the variation is to be linked with industrial locations and types. Metal contaminants via consumption of marine products, with bioaccumulation leading to potential risks via long-term exposure. Marine life can have considerable capability for bioaccumulation and biosorption toxic/heavy metals from their surroundings.

The estimated daily intake of all the studied heavy metals in fishes were below the daily limit on intake recommended by the international agencies. The overall heavy metal contamination in fish samples would seem to be well below harmful levels and therefore, safe for human consumption.

4.3   Heavy metal pollutants in human teeth

With rapid urbanization and large-scale industrial activities, modern human populations are being increasingly subjected to chronic environmental heavy metal exposures. Elemental uptake in tooth dentin is a bioindicator, the uptake occurring during the formation and mineralization processes, retained to large extent over periods of many years. The uptake includes essential elements, most typically geogenic dietary

sources, as well as non-essential elements arising through environmental insults. In this study, with the help of the Dental Faculty of the University of Malaya, separate human teeth were collected from dental patients of various ethnicity, age, gender, occupation, dietary habit, residency, etc. Most teeth dentin samples indicating the presence of the following trace elements, placed in order of concentration, from least to greatest: ^{75}As, ^{55}Mn, ^{137}Ba, ^{63}Cu, ^{52}Cr, ^{208}Pb, ^{66}Zn, ^{202}Hg, ^{121}Sb, ^{27}Al, ^{88}Sr, ^{118}Sn. Statistical analysis showed that ^{208}Pb is correlated with ^{75}As, ^{52}Cr, ^{55}Mn, ^{88}Sr, ^{209}Bi, ^{137}Ba, ^{56}Fe, ^{63}Cu, ^{66}Zn, ^{121}Sb and ^{27}Al which suggested that these metals comes from similar anthropogenic origin.

The concentrations have been observed to increase with age, indicating chronological incorporation of metal in teeth. Among the ethnic groups, the teeth of the Chinese show marginally greater metal concentrations than those of the Indians and Malays, which may be related to their socioeconomic status, dietary habits, profession etc. The food habits and occupations also differ somewhat among the three races. Indians are also mostly vegetarians, the Malays are chicken and fish lovers and the Chinese tend to favour the consumption of pork and alcoholic beverages. The teeth dentin of females generally showing greater concentrations than that of males, indicating gender dependency of heavy metal. Greater concentrations of ^{202}Hg, ^{63}Cu and ^{118}Sn were found in molars while ^{208}Pb, ^{88}Sr, ^{121}Sb and ^{66}Zn were present in greater concentrations in incisors. The differences in relative contents of heavy metals result from complex processes based on structural and developmental differences between the teeth types. With the elevated concentration levels of heavy metals in tooth dentin reflecting pollution from industrial emissions and urbanization, it is evident that human tooth dentin can provide chronological information on exposure, representing a reliable bio-indicator of environmental pollution.

Since higher concentration of radioactive- and heavy metal substances in the environment is undesirable, continuous monitoring should be undertaken to detect the concentration of radioactive and heavy metals in foodstuffs in order to take necessary radiological and toxicological measures with the aim of minimizing the potential harmful effects of ionizing radiation and poisoning effect of heavy metal. The latest data on dietary ingestions of radionuclides and trace metals in different countries should be updated on regular basis. It is thus hoped that the present results will help to establish a baseline for radioactive and heavy metal concentrations of various foodstuffs in Peninsular Malaysia.

4.4   Recommendation

Soil to plant transfer factor of radionuclide is the key factor for the assessment of radiation dose to general public. The dose estimated herein for the consumption of studied vegetables and fishes were 27.9 μSv y^{-1} for vegetable and 226.4 μSv y^{-1} for fish and also 288.8 μSv y^{-1} for rice, in which approximately 42% of the total dose has been credited via the consumption of fish and that for vegetable is only 5%. On the other hand, the obtained result showed that the daily intake dose for most of the studied heavy metals were remarkably higher in fishes than vegetables. Hence, both the radiological and toxicological point of view, it is concluded that vegetarians are safer than the non-vegetarian people.

Among the studied vegetables, about 54% of the total radiation dose received by the general public from the consumption of root vegetable, followed by leafy vegetables (32%) and fruit vegetables (14%). In regard to heavy metal, the daily dietary intake of heavy metals due to consumption leafy and fruit vegetables were little bit higher than root vegetables. Therefore, in radiological point of view, fruit vegetables are safer than

root and leafy one for consumption. However, in the toxicological stand point, root vegetables are quiet safer than leafy and fruit vegetables.

4.5   Suggestion for future work

In the present study, natural radioactivity in only rice, vegetables and marine animals and heavy metal in vegetables and marine fishes collected form a few places of Peninsular Malaysia has been determined. To create a complete baseline data for foodstuffs and to evaluate total dietary study due to food safety, it is suggested that a further study to be done on the radioactivity and heavy metal concentrations in all kinds of land produced foodstuffs, consumer food products, livestock's, fresh water food and seafood collected from all over the Peninsular Malaysia, particularly following the latest (11 March 2011) Fukushima Daiichi, Japan nuclear reactor accident.

The foodstuffs grown in Malaysia is not sufficient for its population, consequently, a huge amounts of foodstuffs have been imported from different countries, it is therefore suggested to carried out investigation on the radioactivity and heavy metal levels in foodstuffs collected in basket method.

There is no baseline data on the radioactivity in soil in Peninsular Malaysia, A future work is suggested to be done to obtain statistically significant scenarios of the complete radiological map of Peninsular Malaysia.

In the present work, the measurement of radioactivity concentration of ^{226}Ra, ^{232}Th and ^{40}K has been performed by well-established gamma-ray spectrometry with HPGe and scintillation NaI (Tl) detector. The cumulative radioactivity contribution of these radionuclides could be measured by internal dosimetry system either by direct (WBC) or indirect (bioassay) method. Therefore, a future work is suggested to be done by

internal dosimetry system either by direct (WBC) or indirect (bioassay) method to measure the cumulative contribution of radioactivity of ^{226}Ra, ^{232}Th and ^{40}K.

Malaysia is one of the rapid growing industrial country in the world. Consequently, its environment may be polluted by industrial and urban emission. Therefore, a further study is suggested to be done in the industrial and control areas (far from industrial areas, e.g., rural areas) to evaluate and compare the environmental pollution using human or animal teeth as bio-marker.

In the present work, the measurement of heavy metal concentration has been performed only by ICP-MS. In order to get more statistically significant picture of the heavy metal concentration, future study should involve the analysis of more samples using LA-ICP-MS, ICP-OES, ICP-AES, GF-AAS, AAS, μSRXRF, PIXE together with ICP-MS.

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LIST OF PUBLICATIONS AND PAPERS PRESENTED

1. Kh. Asaduzzaman, M.U. Khandaker, Y.M. Amin and R. Mahat. *Uptake and distribution of natural radioactivity in rice from soil in north and west part of peninsular Malaysia for the estimation of ingestion dose to man*. Annals of Nuclear Energy, 2015. 76, 85–93.

2. Mayeen Uddin Khandaker, Noor Liyana Mohd Nasir, Kh Asaduzzaman, Michael Adekunle Olatunji, Yusoff Mohd Amin, Hasan Abu Kassim, D. A. Bradley, P. J. Jojo, Tareq Alrefaed. *Evaluation of radionuclides transfer from soil-to-edible flora and estimation of radiological dose to the Malaysian populace*. Chemosphere, 2016. 154, 528-536.

3. Kh. Asaduzzaman, Mayeen Uddin Khandaker, Y.M. Amin, D.A. Bradley, R.H. Mahat, R.M. *Nor Soil-to-root vegetable transfer factors for ^{226}Ra, ^{232}Th, ^{40}K, and ^{88}Y in Malaysia*. Journal of Environmental Radioactivity, 2014. 135, 120–127.

4. Kh. Asaduzzaman, M. U. Khandaker, Y. M. Amin, Z. Zainuddin, M. S. Farook and D. A. Bradley. *Measurement of radioactivity and heavy metal levels in edible vegetables and their impact on Kuala Selangor communities of Peninsular Malaysia*. Radiation Protection Dosimetry, 2015. 167 (1–3), 165–170.

5. M. U. Khandaker, Kh. Asaduzzaman, S.M. Nawi, A.R. Usman, Y.M. Amin, E. Daar, D. A. Bradley, H. Ahmed and A. A. Okhunov. *Assessment of Radiation and Heavy Metals Risk due to the Dietary Intake of Marine Fishes (Rastrelliger kanagurta) from the Straits of Malacca*. PLOS ONE, 10(6), 1-16.

6. M. U. Khandaker, M. A. Olatunji, K. S. K. Shuib, N. A. Hakimi, N. L. M. Nasir, Kh. Asaduzzaman, Y. M. Amin and H. A. Kassim. *Natural radioactivity and effective dose due to the bottom sea and estuaries marine animals in the coastal*

waters around Peninsular Malaysia. Radiation Protection Dosimetry, 2015. 167 (1–3), 196–200.

7. Khandoker Asaduzzaman, Mayeen Uddin Khandaker, Yusoff Bin Mohd Amin, Mohideen Salihu Farook, Nurul Atiqah Binti Baharudin and David Andrew Bradley. *Heavy metals in human teeth; a bio-indicator of metal exposure to environmental pollution* (manuscript submitted).

List of other publications relevant to this work (Not presented in this thesis)

8. Matthew Tikpangi Kolo, Siti Aishah Binti Abdul Aziz, Mayeen Uddin Khandaker, Khandoker Asaduzzaman andYusoff Mohd Amin. *Evaluation of radiological risks due to natural radioactivity around Lynas Advanced Material Plant environment, Kuantan, Pahang, Malaysia*. Environmental Science and Pollution Research. Published online: 01 May 2015.

9. Kh. Asaduzzaman, M. U. Khandaker, Y. M. Amin and D. A. Bradley. *Natural radioactivity levels and radiological assessment of decorative building materials in Bangladesh*. Indoor and Built Environment, 2016. 25(3), 541–550.

10. Khandoker Asaduzzaman, Farhana Mannan, Mayeen Uddin Khandaker, Mohideen Salihu Farook, Aeman Elkezza, Yusoff Bin Mohd Amin, Sailesh Sharma, Hasan Bin Abu Kassim. *Assessment of natural radioactivity levels and potential radiological risks of common building materials used in Bangladeshi dwellings*. PLOS ONE, 10(10), 1-16.

11. Kh. Asaduzzaman, F. Mannan, M.U. Khandaker, M.S. Farook, A. Elkezza, Y.M. Amin & S. Sharma. Natural radioactivity levels in commercialized bottled drinking water and their radiological quality assessment. Desalination and Water Treatment 2016, 57, 11999–12009.

List of papers presented in international and national conferences

12. Paper presented (oral) at 9th International Symposium on the Natural Radiation Environment (NRE-IX), 23−26 September 2014, Hirosaki, Japan. Kh. Asaduzzaman, M. U. Khandaker, Y. M. Amin, Z. Zainuddin, M. S. Farook and D. A. Bradley.

    Title: *Measurement of radioactivity and heavy metal levels in edible vegetables and their impact on Kuala Selangor communities of Peninsular Malaysia*.

13. Paper presented (poster) at 9th International Symposium on the Natural Radiation Environment (NRE-IX), 23−26 September 2014, Hirosaki, Japan. M. U. Khandaker, M. A. Olatunji, K. S. K. Shuib, N. A. Hakimi, N. L. M. Nasir, Kh. Asaduzzaman, Y. M. Amin and H. A. Kassim.

    Title: *Natural radioactivity and effective dose due to the bottom sea and estuaries marine animals in the coastal waters around Peninsular Malaysia.*

14. Paper presented (oral) at 5th International conference on environment 18−19 August 2015, Eastin Hotel, Penang, Malaysia. Khandoker Asaduzzaman, Mayeen Uddin Khandaker, Mohideen Salihu Farook, Yusoff Bin Mohd Amin, David Andrew Bradley.

    Title: *Trace elements in human teeth, as exposure indicator of environmental burden of toxic metals*.

15. Paper presented (oral) at 5th International conference on environment 18−19 August 2015, Eastin Hotel, Penang, Malaysia. Khandoker Asaduzzaman, Noor Liyana Mohd Nasir, Mayeen Uddin Khandaker, Yusoff Bin Mohd Amin.

    Title: *Measurement of radioactivity levels in vegetables grown in Cameron highland and their impact on the local community*.

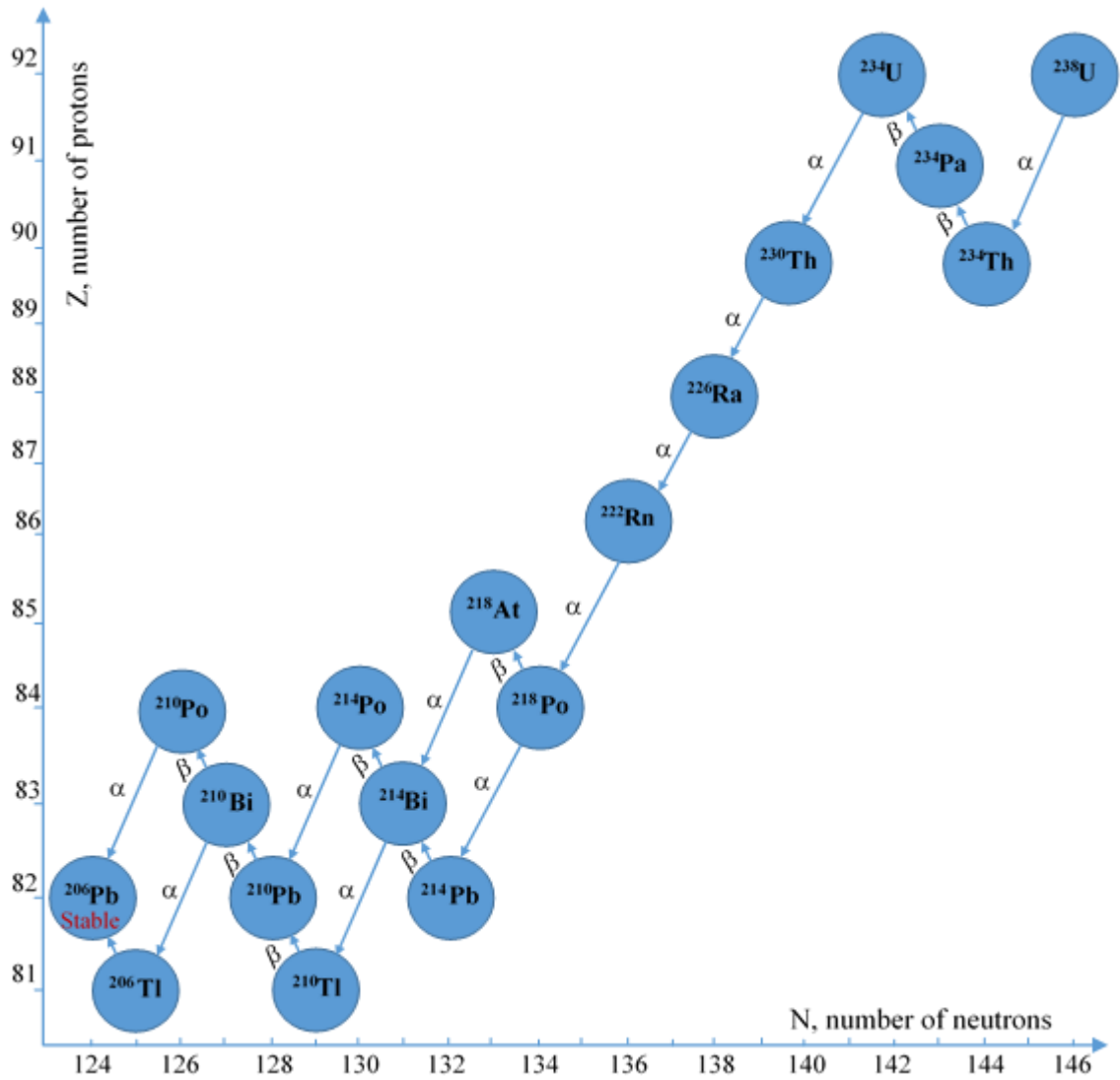16. Seminar presented at the Department of Physics, University of Malaya, Kuala Lumpur, Malaysia, 18 June 2015.

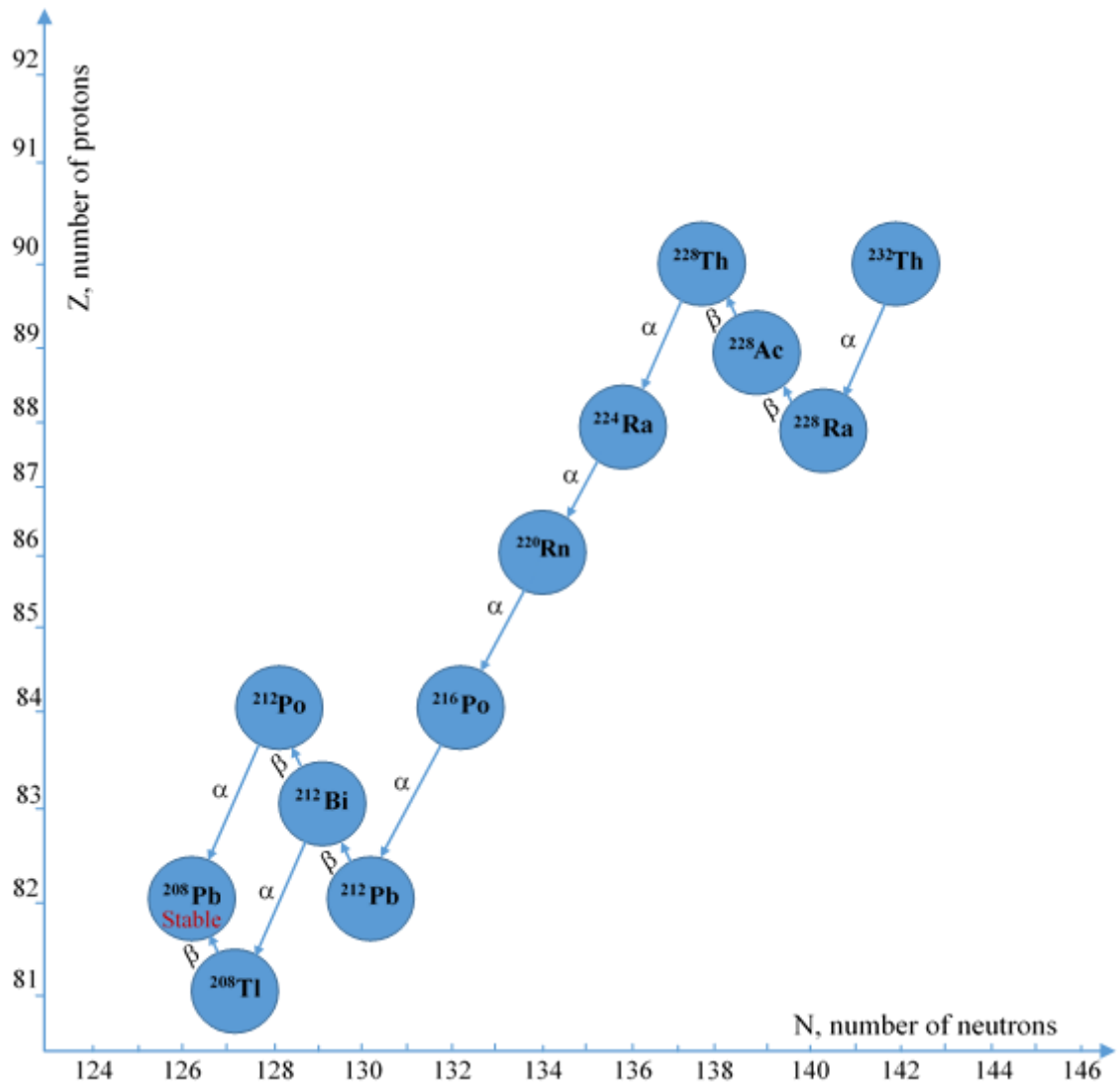Title: *Evaluation of natural levels of radioactivity and heavy metal pollutants in Major Food and human teeth*.

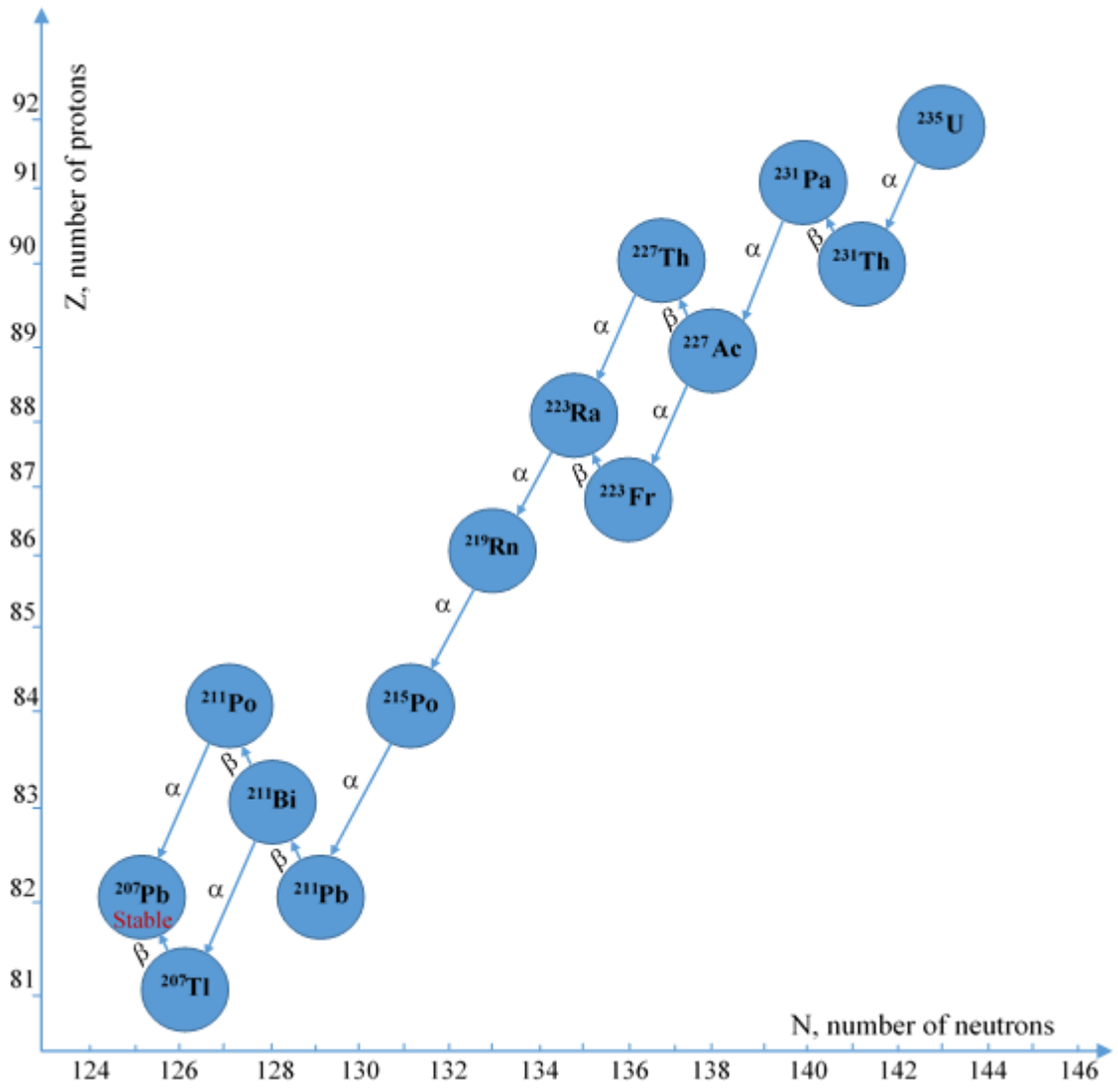Decay scheme of ^{238}U, ^{232}Th, ^{235}U and ^{40}K

Decay scheme of ^{238}U

Decay scheme of ^{232}Th

Decay scheme of ^{235}U

Decay scheme of ^{40}K
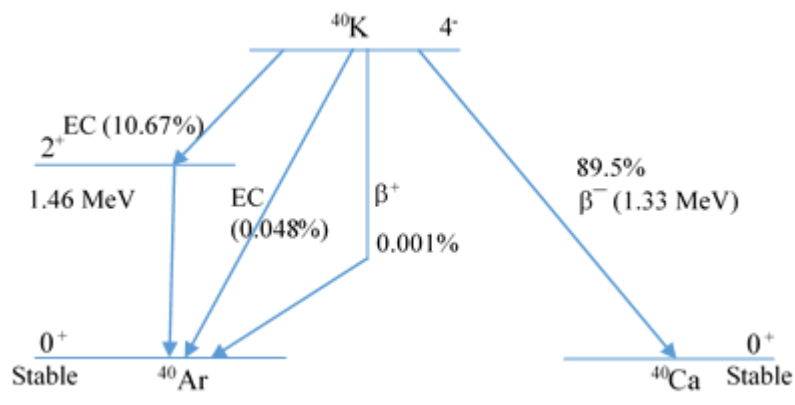
Lists of primordial radionuclides (Kathren, 1998; Eisenbud & Gesell, 1997; UNSCEAR, 2000)

Radionuclides	Decay modes	Decay products	Half-life (years)	Isotopic abundance (%)	Energy (MeV)
^{40}K	EC, β, γ	^{40}Ar, ^{40}Ca	1.28×10^9	0.0118	1.3
^{50}V	EC, β, γ	^{50}Ti, ^{50}Cr	6×10^{14}	0.25	–
^{87}Rb	β	^{88}Sr	4.7×10^{10}	27.83	0.273
^{113}Cd	β	1^{13}In	9×10^{15}	12.3	–
^{115}In	β	^{115}Sn	5×10^{14}	95.7	0.49
^{123}Te	EC	^{123}Sb	1.2×10^{13}	0.87	–
^{138}La	EC, β, γ	^{138}Ba, ^{138}Ce	1.1×10^{11}	0.09	0.27
^{143}Ce	α	^{143}Pr	$>5 \times 10^{16}$	11.1	1.5
^{144}Nd	α	^{140}Ce	2.1×10^{15}	23.9	1.83
^{147}Sm	α	^{143}Nd	1.1×10^{11}	15.0	2.23
^{148}Sm	α	^{144}Nd	8×10^{15}	11.2	1.95
^{149}Sm	α	^{145}Nd	$>10^{16}$	13.8	< 2.0
^{152}Gd	α	^{148}Sm	1.1×10^{14}	0.20	2.14
^{156}Dy	α	^{157}Tb	2×10^{14}	0.06	3
^{176}Lu	EC, β, γ	^{176}Yb, ^{176}Hf	2.7×10^{10}	2.6	0.57
^{174}Hf	α	^{70}Yb	2×10^{15}	0.17	2.5
^{180}Ta	EC, β	^{180}Hf, ^{180}W	$>1.6 \times 10^{13}$	0.012	–
^{187}Re	β	^{187}Os	5×10^{10}	62.5	0.0026
^{190}Pt	α	^{186}Os	7×10^{11}	0.013	3.16
^{204}Pb	α	^{200}Hg	1.4×10^{17}	1.48	2.6

Lists of ^{238}U decay series radionuclides (https://nucleus.iaea.org/Pages/nu-dat-2.aspx).

Parents	Half life	Decay mode	Daughters	Maximum α/β Energy (keV)	Characteristic γ-ray Energy (keV)	γ-ray emission probability (%)
^{238}U	4.468×10⁹ y	α: 100%	^{234}Th	4198	49.55	0.064
					113.5	0.0102
^{234}Th	24.10 d	β⁻: 100%	^{234}Pa	53.6	63.29	3.7
					92.38	2.13
					92.80	2.10
^{234}Pa	1.159 m	β⁻: 99.84 %	^{234}U	820.5	766.42	0.317
					1001.03	0.842
	6.70 h	β⁻: 100%				
^{234}U	2.455×10⁵ y	α: 100%	^{230}Th	4774.6	53.20	0.1230
					120.90	0.035
^{230}Th	7.538×10⁴ y	α: 100%	^{226}Ra	4687.0	67.672	0.38
					143.872	0.049
^{226}Ra	1600 y	α: 100%	^{222}Rn	4784.34	186.211	3.64
^{222}Rn	3.8235 d	α:100%	^{218}Po	5489.48	510	0.076
^{218}Po	3.098 m	α: 99.98 %	^{214}Pb	6002.35	No gamma-ray observed	
		β: 0.02 %	^{218}At			
^{214}Pb	26.8 m	β⁻: 100%	^{214}Bi	334.9	53.2284	1.075
					241.9950	7.251
					258.86	0.531
					274.80	0.355
					295.2228	18.42
					274.80	0.355
					351.9321	35.60
					480.432	0.337
					487.14	0.432
					580.14	0.370
					785.96	1.06
					839.07	0.583
^{218}At	1.5 s	α : 99.90 % β⁻: 0.10 %	^{214}Bi	6756	No gamma-ray observed	
^{214}Bi	19.9 m	β⁻: 99.979 %	^{214}Po	1268.8	609.320	45.49

					665.447	1.531
					768.360	4.894
					806.180	1.264
					934.056	3.107
					1120.294	14.92
					1155.210	1.633
					1280.976	1.434
					1377.669	3.988
					1401.515	1.330
					1407.988	2.394
					1509.210	2.130
					1661.274	1.047
					1729.595	2.878
					1764.491	15.30
					1847.429	2.025
					2118.514	1.160
					2204.059	4.924
					2447.70	1.548
		α: 0.021 %	^{210}Tl	5516	No gamma-ray observed	
^{214}Po	164.3 μS	α: 100%	^{210}Pb	7686.82	799.7	0.0104
^{210}Tl	1.30 m	β$^-$	^{210}Pb	1762.6	296	79
					799.6	98.96
					1070	12
					1210	17
					1316	21
^{210}Pb	22.20 y	β$^-$: 100%	^{210}Bi	16.16	46.539	4.25
^{210}Bi	5.012 d	β$^-$: 100%	^{210}Po	389.0	No gamma-ray observed	
	3.04×10^6 y	α: 100%	^{206}Tl	4946	265.6	51
					304.6	28
^{210}Po	138,376 d	α: 100%	^{206}Pb (stable)	5304.33	803.06	0.00103
^{206}Tl	4.202 m	β$^-$: 100%	^{206}Pb (stable)	538.86	803.06	0.0050

Lists of ^{232}Th decay series radionuclides (https://nucleus.iaea.org/Pages/nu-dat-2.aspx).

Parents	Half life	Decay mode	Daughters	Maximum α/β Energy (keV)	Characteristic γ-ray Energy (keV)	γ-ray emission probability (%)
^{232}Th	1.4×10^{10} y	α: 100%	^{228}Ra	4012.3	63.81	0.263
					140.88	0.021
^{228}Ra	5.75 y	β⁻: 100%	^{228}Ac	10.04	13.52	1.60
					26.4	0.0149
^{228}Ac	6.15 h	β⁻: 100%	^{228}Th	747.0	911.204	25.8
					968.971	15.8
					338.320	11.27
					964.766	4.99
					794.947	4.25
					463.004	4.40
					209.253	3.89
					270.245	3.46
					1588.20	3.22
					328.000	2.95
					674.75	2.1
					129.065	2.42
					409.462	1.92
					835.710	1.61
					1630.627	1.51
					772.291	1.49
					755.315	1.00
^{228}Th	1.912 y	α: 100%	^{224}Ra	5423.15	84.373	1.19
					215.983	0.247
^{224}Ra	3.66 d	α: 100%	^{220}Rn	5685.37	240.986	4.10
^{220}Rn	55.6 s	α: 100%	^{216}Po	6288.08	549.73	0.114
^{216}Po	0.145 s	α: 100%	^{212}Pb	6778.3	804.9	0.0019
^{212}Pb	10.64 h	β⁻: 100%	^{212}Bi	171.7	238.632	43.6
					300.087	3.30
					89.784	1.46
					115.183	0.596
^{212}Bi	60.55 m	β⁻: 64.06%	^{212}Po	834.2	727.330	6.67
					1620.50	1.47
					785.37	1.102
					1078.62	0.564

Table contd.

		α: 35.94%	^{208}Tl	6089.88	39.857	1.06
					452.98	0.363
					288.20	0.337
^{212}Po	0.299 μS	α: 100%	^{208}Pb (stable)	8784.86	No gamma-ray observed	
^{208}Tl	3.053 m	β$^-$: 100%	^{208}Pb (stable)	649.48	2614.511	99.754
					583.187	85.0
					510.77	22.60
					860.557	12.50
					277.371	6.6
					763.13	1.79

Lists of ^{235}U decay series radionuclides (https://nucleus.iaea.org/Pages/nu-dat-2.aspx).

Parents	Half life	Decay mode	Daughters	Maximum α/β Energy (keV)	Characteristic γ-ray Energy (keV)	γ-ray emission probability (%)
^{235}U	703.8×10^6 y	α: 100%	^{231}Th	4579.4	185.715	57.0
					143.76	10.96
					163.356	5.08
					205.316	5.02
					109.19	1.66
^{231}Th	25.52 h	β$^-$: 100%	^{231}Pa	111.4	25.64	14.1
					84.2140	6.6
^{231}Pa	3.276×10^4 y	α: 100%	^{227}Ac	5058.6	27.36	10.5
					300.066	2.41
					302.667	2.3
					330.055	1.36
					183.682	1.65
^{227}Ac	21.772 y	β$^-$: 98.62%	^{227}Th	11.37	No gamma-ray observed	
		α: 1.38%	^{223}Fr	4953.26	99.6	0.0056
					160.49	0.0049
^{227}Th	18.68 d	α: 100%	^{223}Ra	6038.01	235.96	12.9
					50.13	8.4
					256.23	7.0
					329.85	2.9
					299.98	2.21
					88.471	2.18
					79.69	1.95
					289.59	1.9
					286.09	1.74
^{223}Fr	22.00 m	β$^-$: 99.994%	^{223}Ra	380.7	50.094	34
					79.651	8.7
					234.75	3.0
					88.471	2.7
^{223}Ra	11.43 d	α: 100%	^{219}Rn	5871.3	269.463	13.9
					154.208	5.7
					323.871	3.99
					338.282	2.84
					144.235	3.27
^{219}Rn	3.96 s	α: 100%	^{215}Po	6819.1	271.23	10.8

Table contd.

					401.81	6.6
^{215}Po	1.781 ms	α: 99.99977%	^{211}Pb	7386.1	No data available	
^{211}Pb	36.1 m	β⁻: 100%	^{211}Bi	471.3	404.853	3.78
					832.01	3.52
					427.088	1.76
^{211}Bi	2.14 m	α: 99.724%	^{207}Tl	6622.9	351.07	13.02
		β⁻: 0.276%	^{211}Po	172.9	No gamma-ray observed	
^{211}Po	0.516 s	α: 100%	^{207}Pb (stable)	7450.3	897.8	0.551
					569.65	0.535
^{207}Tl	4.77 m	β⁻: 100%	^{207}Pb (stable)	492.5	897.77	0.263

List of cosmogenic radionuclides (UNSCEAR, 2000)

Cosmogenic radionuclides	Half-life	Decay mode
^{3}H	12.33 y	β
^{7}Be	53.29 d	EC, γ
^{10}Be	1.51×10^6 y	β
^{14}C	5730 y	β
^{22}Na	2.602 y	β, γ
^{26}Al	7.4×10^5 y	EC
^{32}Si	172 y	β
^{32}P	14.26 d	β
^{33}P	25.34 d	β
^{35}S	87.51 d	β
^{36}Cl	3.01×10^5 y	EC, β
^{37}Ar	35.04 d	EC
^{39}Ar	269 y	β
^{81}Kr	2.29×10^5 y	EC

Annual average doses to the world population from various sources (UNSCEAR, 2010)

Source	Annual average dose (Worldwide) (mSv)
Natural sources of exposure	
Ingestion (foods and drinks)	0.29
Inhalation (radon gas)	1.26
External terrestrial	0.48
Cosmic radiation	0.39
Total of natural sources	**2.42**
Artificial sources of exposure	
Medical diagnosis (not therapy)	0.6
Atmospheric nuclear testing	0.005
Occupational exposure	0.005
Nuclear fuel cycle (public exposure)	0.0002
Chernobyl accident	0.002
Total of artificial sources	**0.6122**

APPENDIX C

Interactive Chart of Nuclides
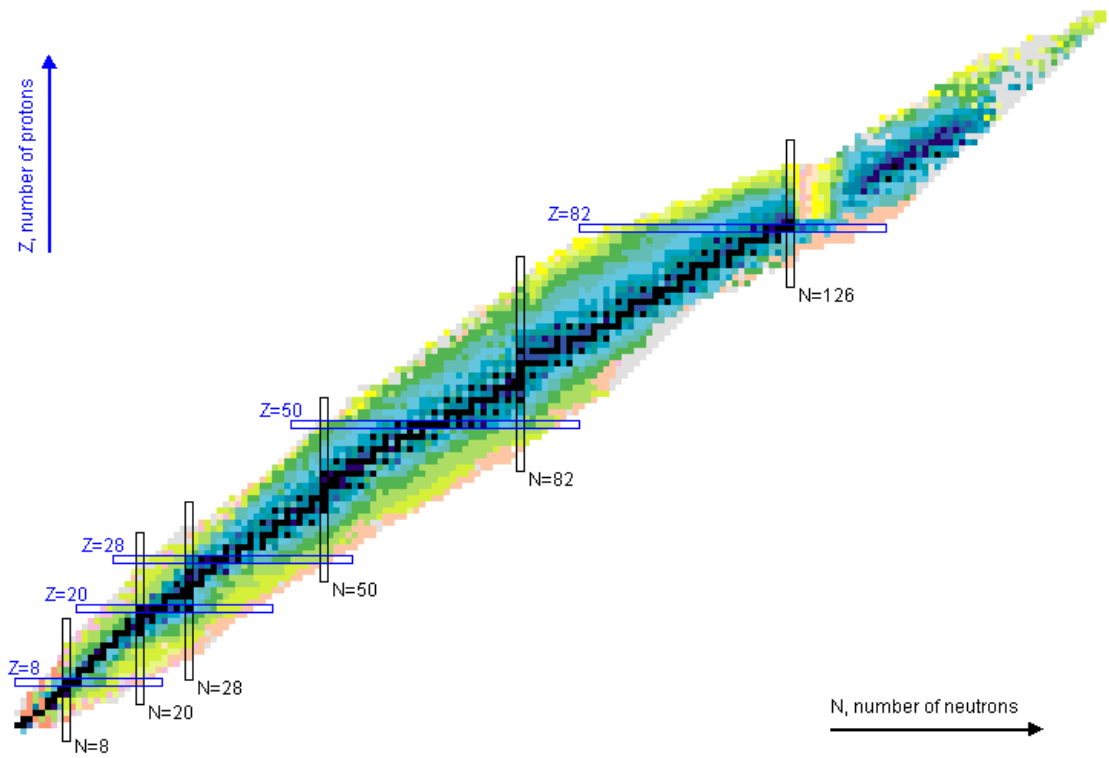
Source: https://nucleus.iaea.org/Pages/nu-dat-2.aspx Accessed by 10 Sep 2015
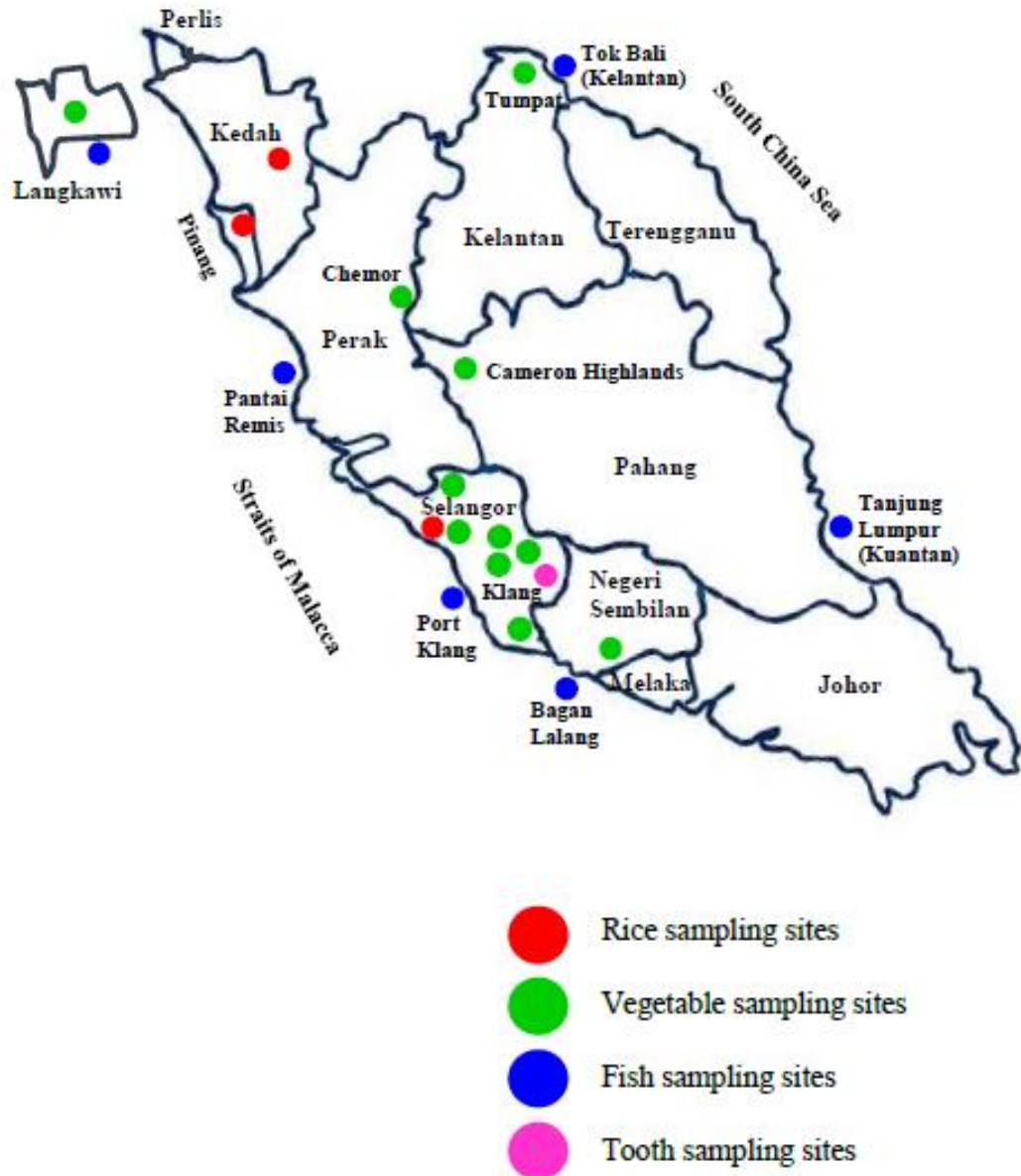
The main routes of human radiation exposure through the environment
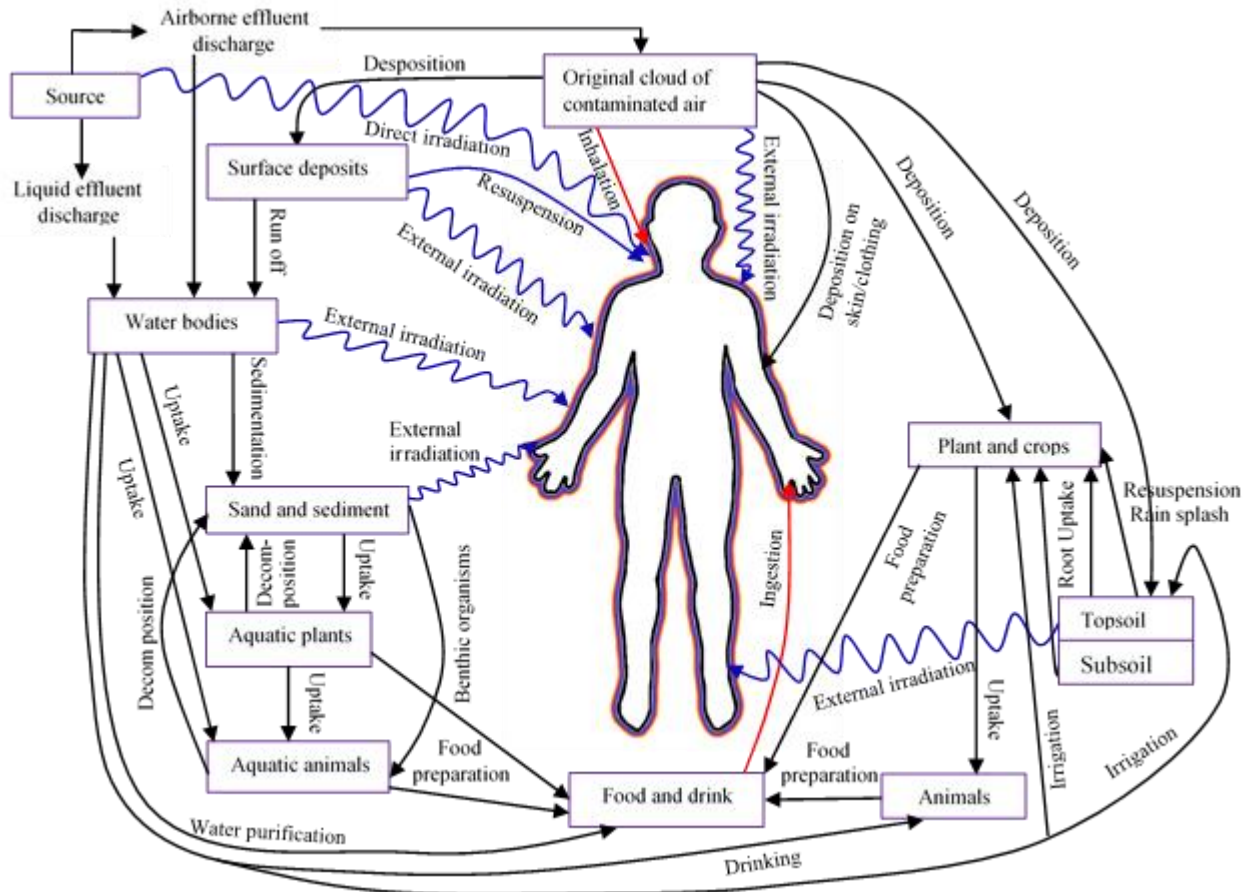
Map of Peninsular Malaysia showing sampling sites

Certificate of the γ-ray calibration standard source

Isotope Products
Laboratories

An Eckert & Ziegler Company

24937 Avenue Tibbitts
Valencia, California 91355

Tel 661·309·1010
Fax 661·257·8303

CERTIFICATE OF CALIBRATION
MULTINUCLIDE STANDARD SOURCE

Customer:	CHURCHIN ASSOCIATES LTD.	**P.O. No.:**	CA 3307	
Catalog No.:	EG-ML	**Reference Date:**	1-Sep 13	12:00 PST
Source No.:	989-90	**Total Radioactivity:**	5.046	μCi
		Total Radioactivity:	186.7	kBq

Description of Source:

A. Capsule type: 500 mL Marinelli beaker (530G-E)
B. Nature of active deposit: Multinuclide distributed in 1.0 g/cc epoxy matrix
C. Active diameter/volume: Approximately 500 mL (499.9 grams)
D. Backing: Plastic
E. Cover: Plastic

Gamma-Ray Energy (keV)	Nuclide	Half-life	Branching Ratio (%)	Activity (μCi)	Gammas per second	Total Uncert.
88	Cd-109	462.6 ± 0.7 days	3.63	1.482	1990	3.1%
122	Co-57	271.79 ± 0.09 days	85.6	0.05504	1743	3.1%
159	Te-123m	119.7 ± 0.1 days	84.0	0.06754	2099	3.0%
320	Cr-51	27.706 ± 0.007 days	9.86	1.792	6538	3.0%
392	Sn-113	115.09 ± 0.04 days	64.89	0.2638	6334	3.0%
514	Sr-85	64.849 ± 0.004 days	98.4	0.3359	12230	3.0%
662	Cs-137	30.17 ± 0.16 years	85.1	0.2300	7242	3.0%
898	Y-88	106.630 ± 0.025 days	94.0	0.5371	18680	3.0%
1173	Co-60	5.272 ± 0.001 years	99.86	0.2831	10460	3.0%
1333	Co-60	5.272 ± 0.001 years	99.98	0.2831	10470	3.0%
1836	Y-88	106.630 ± 0.025 days	99.36	0.5371	19750	3.0%

Method of Calibration:

This source was prepared from a weighed aliquot of solution whose concentrations in μCi/g were determined by gamma spectrometry.

Notes:

- See reverse side for leak test(s) applied to this source.
- Nuclear data was taken from IAEA-TECDOC-619, 1991.
- IPL participates in a NIST measurement assurance program to establish and maintain implicit traceability for a number of nuclides, based on the blind assay (and later NIST certification) of Standard Reference Materials (As in NRC Regulatory Guide 4.15).
- Overall uncertainty is calculated at the 99% confidence level.
- This source has a working life of 1 year.

Daniel James Van Dalsem
Quality Control

19-Aug-13
Date Signed

IPL Ref No.:   989-90

——————————— ISO 9001 CERTIFIED ———————————

Medical Imaging Laboratory
24937 Avenue Tibbitts   Valencia, California 91355

**Industrial Gauging Laboratory**
1800 North Keystone Street   Burbank, California 91504

Efficiency Calibration curve of HPGe γ-ray spectrometer

Calibration of the gamma-ray spectrometer

The quality and reliability of any analytical measurements on how well the measuring equipment is calibrated with standard sources. The efficiency calibration of gamma-ray spectrometer used for radioactivity was performed using 500 ml Marinelli beaker geometry that contained multi-nuclide standard gamma sources. The measured detection efficiencies were fitted by using a power fitting function. The efficiency curve was obtained by discarding some data points of the radionuclides due to get best fit curve and the fitted efficiencies were used in activity determination of the samples. The efficiency calibration curve for the gamma-ray spectrometer is shown as follows:

$$y = 0.6905x^{-0.607}$$
$$R^2 = 0.9977$$
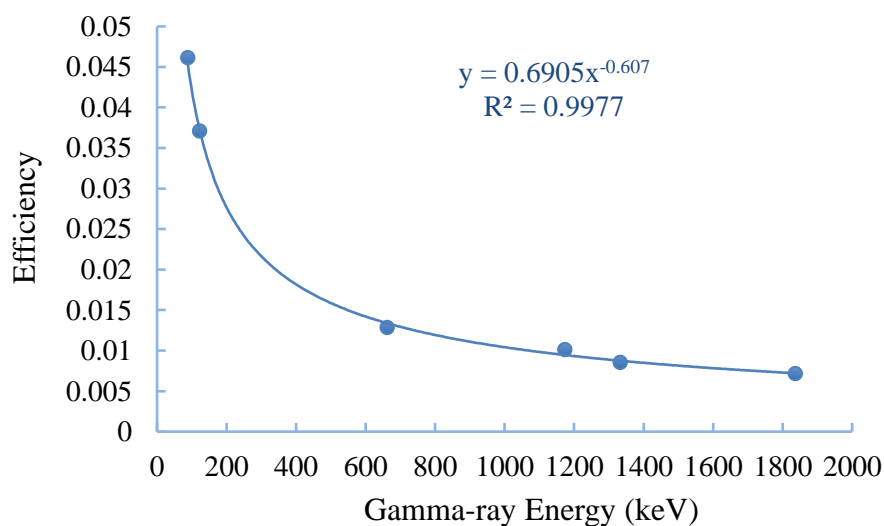
Efficiency calibration curve of HPGe detector

Certificates of the ICP-MS calibration standard sources

Agilent Technologies

Certificate of Analysis

Multi-element Calibration Standard 2A

Agilent Part Number: 8500-6940   **Lot Number:** 9-156YPY2

Analyte	CAS#	Labeled Conc.	Measured Conc.	SRM	Start Mat'l Formula	Start Mat'l Purity	Analyte	CAS#	Labeled Conc.	Measured Conc.	SRM	Start Mat'l Formula	Start Mat'l Purity
Ag	7440-22-4	10.0 mg/L	10.1 mg/L	3151*	Ag	99.99+	Li	7439-93-2	10.0 mg/L	9.96 mg/L	3129a*	Li_2CO_3	99.99+
Al	7429-90-5	10.0 mg/L	9.95 mg/L	3101a*	Al	99.99+	Mg	7439-95-4	10.0 mg/L	9.97 mg/L	3131a*	Mg	99.99+
As	7440-38-2	10.0 mg/L	9.99 mg/L	3103a*	As	99.99+	Mn	7439-96-5	10.0 mg/L	10.2 mg/L	3132*	Mn	99.99+
Ba	7440-39-3	10.0 mg/L	10.0 mg/L	3104a*	$BaCO_3$	99.99+	Na	7440-23-5	10.0 mg/L	9.99 mg/L	3152a*	Na_2CO_3	99.99+
Be	7440-41-7	10.0 mg/L	10.1 mg/L	3105a*	$Be_4O(CH_3COO)_6$	99.99+	Ni	7440-02-0	10.0 mg/L	10.0 mg/L	3136*	Ni	99.99+
Ca	7440-70-2	10.0 mg/L	10.1 mg/L	3109a*	$CaCO_3$	99.99+	Pb	7439-92-1	10.0 mg/L	10.0 mg/L	3128*	PbO	99.99+
Cd	7440-43-9	10.0 mg/L	10.1 mg/L	3108*	Cd	99.99+	Rb	7440-17-7	10.0 mg/L	10.0 mg/L	3145a*	$RbNO_3$	99.99+
Co	7440-48-4	10.0 mg/L	10.0 mg/L	3113*	Co	99.99+	Se	7782-49-2	10.0 mg/L	10.0 mg/L	3149*	Se	99.99+
Cr	7440-47-3	10.0 mg/L	9.99 mg/L	3112a*	$Cr(NO_3)_3 \cdot 9H_2O$	99.99+	Sr	7440-24-6	10.0 mg/L	10.1 mg/L	3153a*	$SrCO_3$	99.99+
Cs	7440-46-2	10.0 mg/L	10.0 mg/L	3111a*	$CsNO3$	99.99+	Tl	7440-28-0	10.0 mg/L	9.97 mg/L	3158*	$TlNO_3$	99.99+
Cu	7440-50-8	10.0 mg/L	10.2 mg/L	3114*	Cu	99.99+	U	7440-61-1	10.0 mg/L	10.1 mg/L	3164*	$UO_2(NO_3)_2 \cdot 6H_2O$	99.99+
Fe	7439-89-6	10.0 mg/L	10.1 mg/L	3126a*	Fe	99.99+	V	7440-62-2	10.0 mg/L	9.98 mg/L	3165*	NH_4VO_3	99.99+
Ga	7440-55-3	10.0 mg/L	9.94 mg/L	3119a*	Ga	99.99+	Zn	7440-66-6	10.0 mg/L	9.98 mg/L	3168a*	ZnO	99.99+
K	7440-09-7	10.0 mg/L	10.1 mg/L	3141a*	KNO_3	99.99+							

* - indicates NIST SRM   † - indicates CRM (when NIST SRM is not available)

Purity grades:

Starting Materials: Shown above
Matrix:
5% HNO_3: HNO_3 (CAS No. 7697-37-2) high purity grade

Traceability:

This standard has been produced gravimetrically and volumetrically using ISO 9001 quality procedures. ICP / ICP-MS Spectrometer was used to determine the concentration of the main elements via NIST SRMs shown above, as well as the impurities. Other reference standards used: CL8-84YP,CL3-203YP.

Trace Metallic Impurities in the Actual Solution, in µg/L, via ICP-MS Analysis, results are accurate to ±10%:

Element	Conc.	Element	Conc.	Element	Conc.	Element	Conc.	Element	Conc.	Element	Conc.
Au	<3	Gd	<0.02	La	0.06	Pr	0.02	Si	300	Ti	<2
B	<5	Ge	<1	Lu	<0.01	Pt	<0.2	Sm	0.2	Tm	0.01
Bi	0.1	Hf	<0.02	Mo	0.09	Re	<0.02	Sn	<0.2	W	<0.08
Ce	0.04	Hg	<10	Nb	0.04	Rh	0.4	Ta	<0.1	Y	0.04
Dy	<0.02	Ho	<0.01	Nd	<0.02	Ru	<0.2	Tb	0.02	Yb	<0.02
Er	<0.03	In	<0.01	P	<100	Sb	0.1	Te	<0.01	Zr	2
Eu	<0.1	Ir	<0.04	Pd	4	Sc	<0.04	Th	0.2		

Balances are calibrated regularly with weight sets traceable to NIST.

Agilent reference standards are guaranteed stable and accurate to ±0.5% of measured analyte concentration. For these solutions we use the highest purity acids applicable, 18 megohm double deionized water and acid-leached, triple rinsed bottles. All glassware used is class A.

Date of release: March 31, 2013
Date of expiration: September 30, 2014

QC Coordinator
CertiPrep, Inc.

Agilent Technologies

Certificate of Analysis

Multi Element Calibration Standard 2A - HG

Agilent Part Number: 8500-6940-HG

Lot Number: 6-28HGY2

Analyte	CAS#	Labeled Conc.	Measured Conc.	SRM	Start Mat'l Formula	Start Mat'l Purity
Hg	7439-97-6	10.0 mg/L	9.99 mg/L	3133*	Hg	99.99+

* - indicates NIST SRM   † - indicates CRM (when NIST SRM is not available)

Purity grades:

Starting Materials: Shown above

Matrix:

5% HNO₃: HNO₃ (CAS No. 7697-37-2) high purity grade

Traceability:

This standard has been produced gravimetrically and volumetrically using ISO 9001 quality procedures.   ICP / ICP-MS Spectrometer was used to determine the concentration of the main elements via NIST SRMs shown above, as well as the impurities.

Trace Metallic Impurities in the Actual Solution, in µg/L, via ICP-MS Analysis, results are accurate to ±10%:

Element	Conc.	Element	Conc.	Element	Conc.	Element	Conc.	Element	Conc.	Element	Conc.
Ag	<0.7	Cr	<0.7	Ho	<0.09	Nb	<0.2	Ru	<2	Th	<0.7
Al	<1	Cs	<0.4	In	<0.3	Nd	<0.2	Sb	<0.9	Ti	<2
As	<3	Cu	<1	Ir	<0.9	Ni	<0.8	Sc	<2	Tl	<8
Au	<10	Dy	<0.09	K	<300	P	<300	Se	<6	Tm	<0.03
B	<30	Er	<0.1	La	<0.2	Pb	<0.9	Si	<200	U	1
Ba	<0.3	Eu	<0.06	Li	<2	Pd	<1	Sm	<0.1	V	<1
Be	<3	Fe	30	Lu	<0.06	Pr	<0.2	Sn	<0.01	W	<7
Bi	<0.6	Ga	<0.2	Mg	<2	Pt	<0.4	Sr	<0.4	Y	<0.1
Ca	20	Gd	<0.01	Mn	<0.6	Rb	<0.3	Ta	<0.8	Yb	<0.2
Cd	<0.3	Ge	<1	Mo	<5	Re	<0.1	Tb	<0.03	Zn	<6
Ce	<0.1	Hf	<0.08	Na	<2	Rh	<1	Te	<5	Zr	<0.8
Co	<0.7										

Balances are calibrated regularly with weight sets traceable to NIST.

Agilent reference standards are guaranteed stable and accurate to ±0.5% of measured analyte concentration. For these solutions we use the highest purity acids applicable, 18 megohm double deionized water and acid-leached, triple rinsed bottles. All glassware used is class A.

Date of release: October 15, 2012

**Date of expiration:**   April 30, 2014

Brjender S Tonk

QC Coordinator
CertiPrep, Inc.

Calibration curves of different elements standard for ICP-MS

Calibration of the ICP-MS was performed using multi-element calibration standard 2A solution (10 mg/l of each element) from Agilent Technologies, USA, part no. 8500–6940).


Count/CPS(Y) [count/CPS]

Pb standard calibration curve

Conc.(X) [ppb]
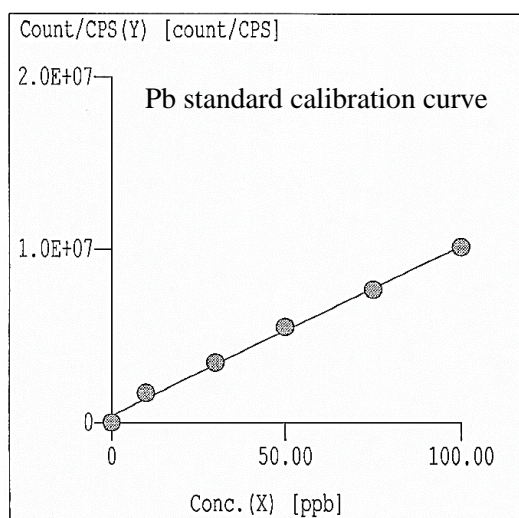
Curve Fit: Y=aX+b
r = 0.9977
Y = 9.811E+004*X +4.147E+005
X = 1.019E-005*Y -4.227E+000
DL = --- ppb
BEC = 4.227 ppb

Count/CPS(Y) [count/CPS]

Hg standard calibration curve

Conc.(X) [ppb]
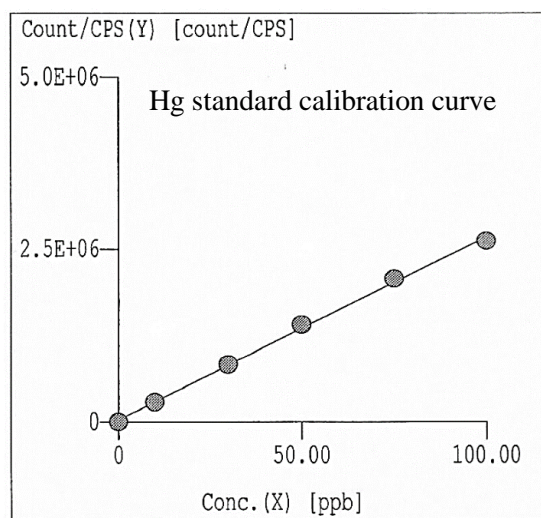
Curve Fit: Y=aX+b
r = 0.9991
Y = 2.644E+004*X +3.199E+004
X = 3.783E-005*Y -1.210E+000
DL = --- ppb
BEC = 1.210 ppb

Count/CPS(Y) [count/CPS]

As standard calibration curve

Conc.(X) [ppb]
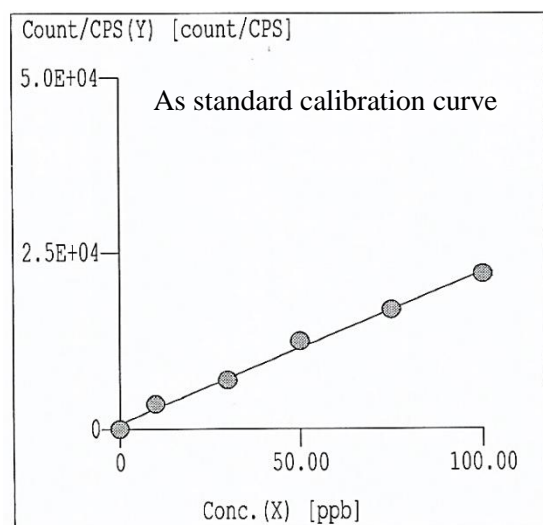
Curve Fit: Y=aX+b
r = 0.9971
Y = 2.156E+002*X +7.784E+002
X = 4.639E-003*Y -3.611E+000
DL = 2.638E-02 ppb
BEC = 3.611 ppb

Count/CPS(Y) [count/CPS]

Cd standard calibration curve

Conc.(X) [ppb]
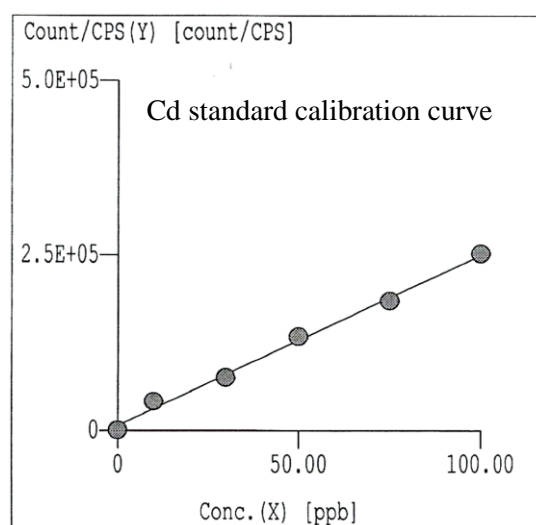
Curve Fit: Y=aX+b
r = 0.9974
Y = 2.428E+003*X +6.827E+003
X = 4.118E-004*Y -2.811E+000
DL = 4.586E-03 ppb
BEC = 2.811 ppb

Cr standard calibration curve
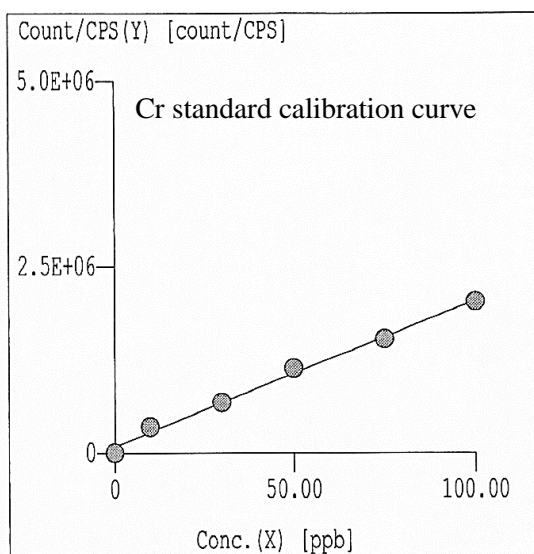
Curve Fit: Y=aX+b
 r = 0.9971
 Y = 1.961E+004*X +8.348E+004
 X = 5.100E-005*Y -4.257E+000
 DL = --- ppb
 BEC = 4.257 ppb

Mn standard calibration curve
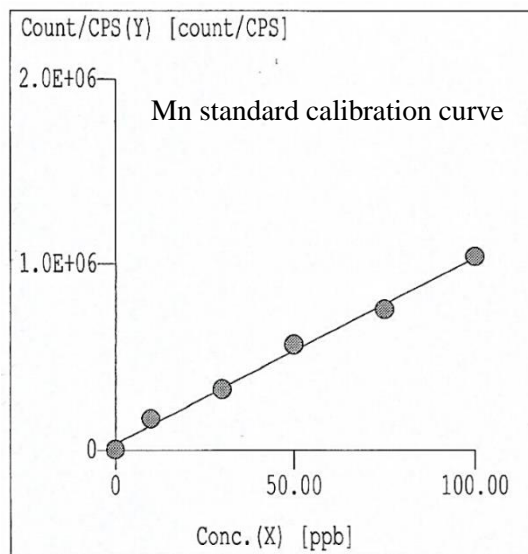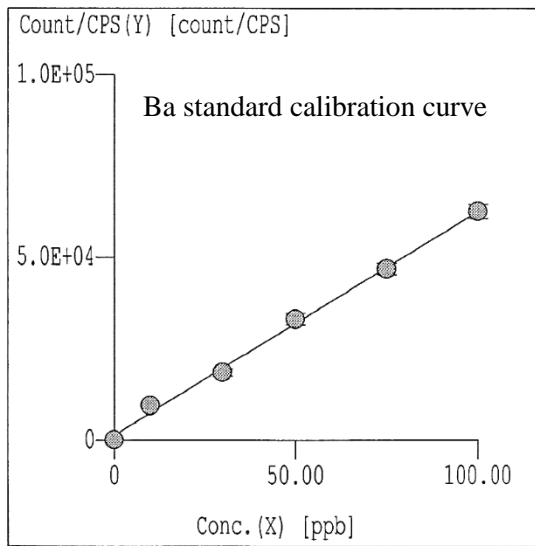
Curve Fit: Y=aX+b
 r = 0.9971
 Y = 1.000E+004*X +3.330E+004
 X = 9.998E-005*Y -3.329E+000
 DL = --- ppb
 BEC = 3.329 ppb

Sr standard calibration curve
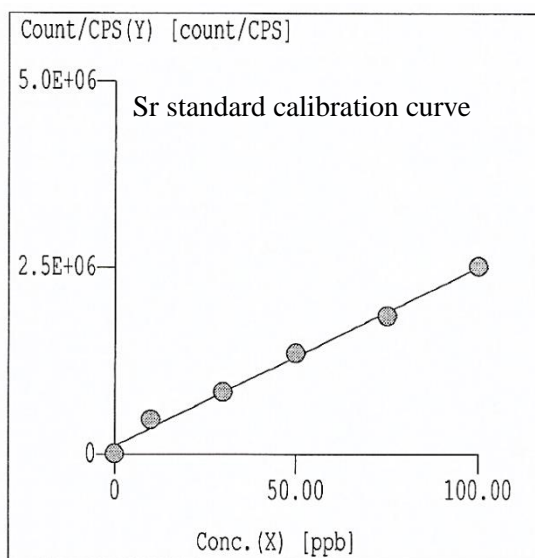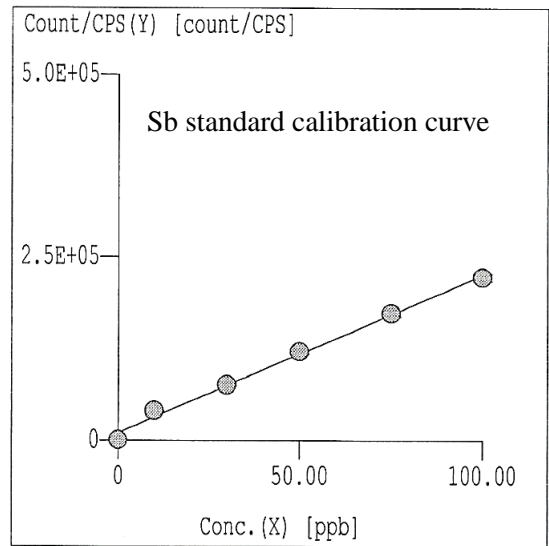
Curve Fit: Y=aX+b
 r = 0.9967
 Y = 2.394E+004*X +1.075E+005
 X = 4.176E-005*Y -4.489E+000
 DL = 2.326E-03 ppb
 BEC = 4.489 ppb

Bi standard calibration curve
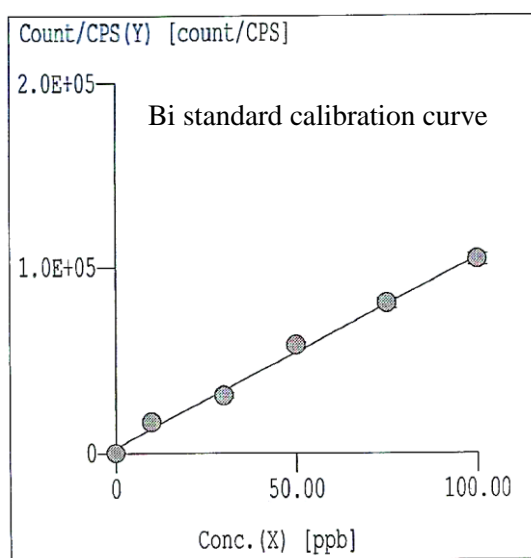
Curve Fit: Y=aX+b
 r = 0.9972
 Y = 1.033E+003*X +2.964E+003
 X = 9.682E-004*Y -2.869E+000
 DL = 2.015E-02 ppb
 BEC = 2.869 ppb

Ba standard calibration curve

Count/CPS(Y) [count/CPS]

Conc.(X) [ppb]

Curve Fit: Y=aX+b
 r = 0.9985
 Y = 6.126E+002*X +1.313E+003
 X = 1.632E-003*Y -2.144E+000
 DL = 6.797E-02 ppb
 BEC = 2.144 ppb

Sb standard calibration curve

Count/CPS(Y) [count/CPS]

Conc.(X) [ppb]

Curve Fit: Y=aX+b
 r = 0.9973
 Y = 2.154E+003*X +9.950E+003
 X = 4.643E-004*Y -4.620E+000
 DL = 2.457E-02 ppb
 BEC = 4.620 ppb

Fe standard calibration curve

Count/CPS(Y) [count/CPS]

Conc.(X) [ppb]
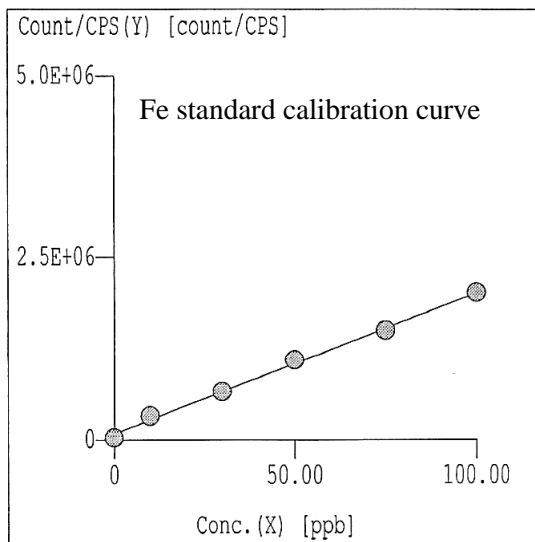
Curve Fit: Y=aX+b
 r = 0.9984
 Y = 1.931E+004*X +7.874E+004
 X = 5.179E-005*Y -4.078E+000
 DL = --- ppb
 BEC = 4.078 ppb

Cu standard calibration curve

Count/CPS(Y) [count/CPS]

Conc.(X) [ppb]
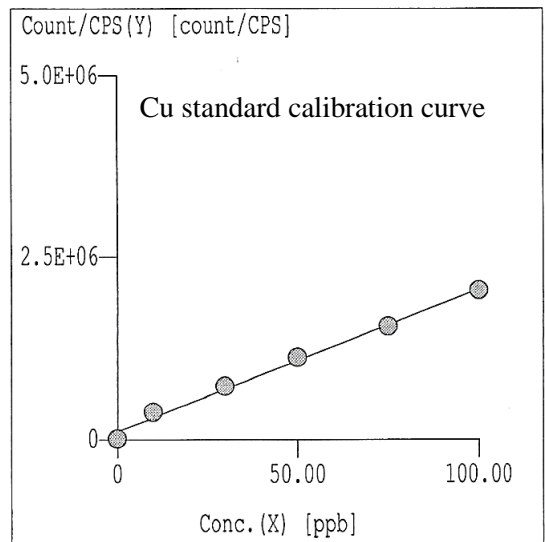
Curve Fit: Y=aX+b
 r = 0.9966
 Y = 1.949E+004*X +1.039E+005
 X = 5.131E-005*Y -5.332E+000
 DL = --- ppb
 BEC = 5.332 ppb

222

Ni standard calibration curve
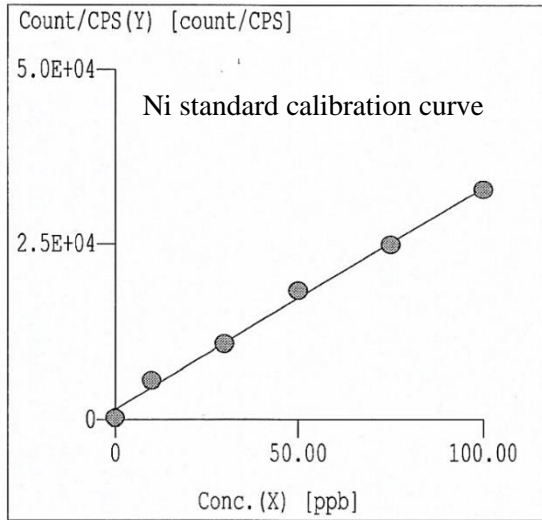
Curve Fit: Y=aX+b
 r = 0.9976
 Y = 3.168E+002*X +1.457E+003
 X = 3.156E-003*Y -4.598E+000
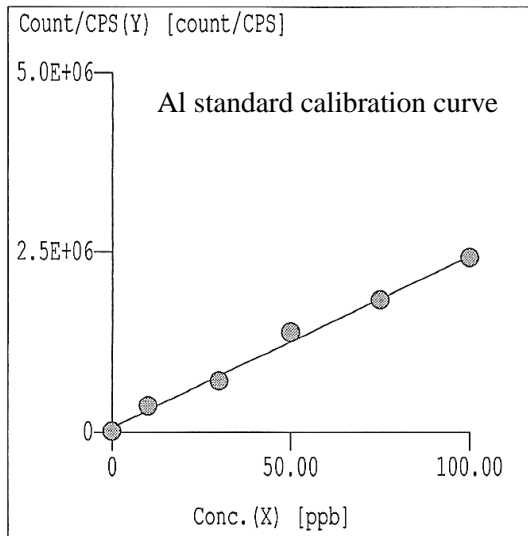 DL = --- ppb
 BEC = 4.598 ppb

Rb standard calibration curve
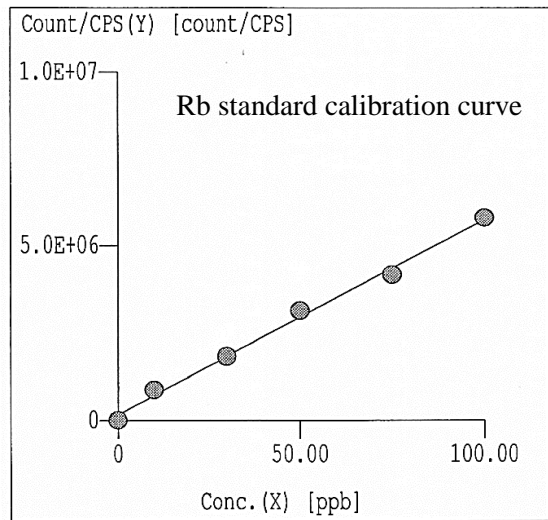
Curve Fit: Y=aX+b
 r = 0.9973
 Y = 5.583E+004*X +1.658E+005
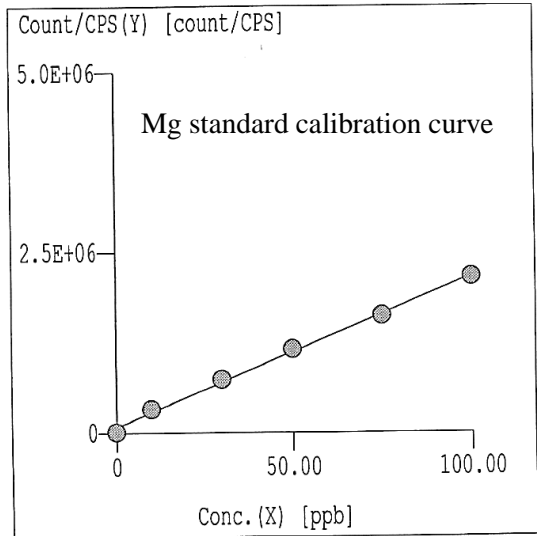 X = 1.791E-005*Y -2.969E+000
 DL = --- ppb
 BEC = 2.969 ppb

Co standard calibration curve
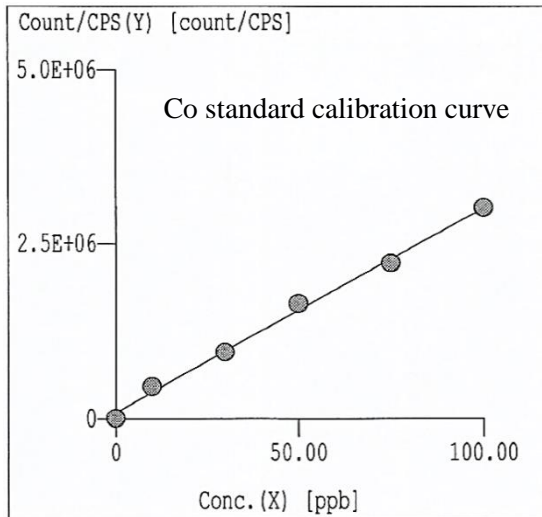
Curve Fit: Y=aX+b
 r = 0.9980
 Y = 2.931E+004*X +9.147E+004
 X = 3.411E-005*Y -3.121E+000
 DL = --- ppb
 BEC = 3.121 ppb

Zn standard calibration curve
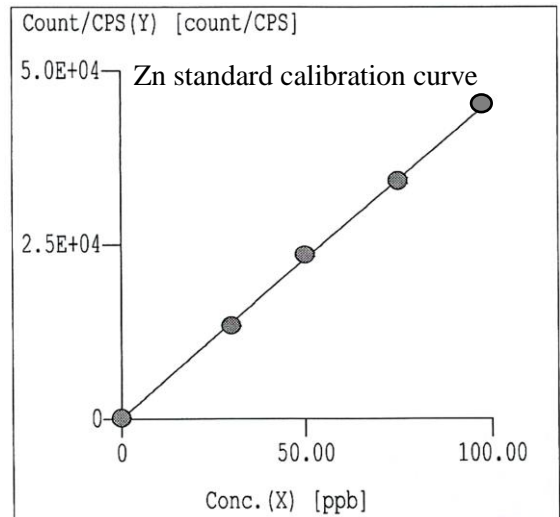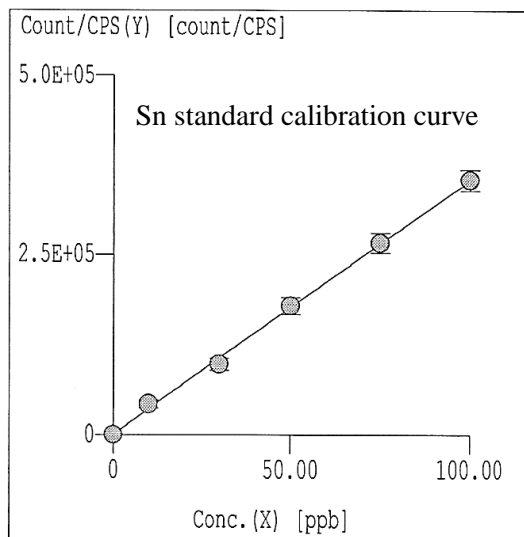
Curve Fit: Y=aX+b
 r = 0.9996
 Y = 4.581E+002*X +3.079E+001
 X = 2.183E-003*Y -6.721E-002
 DL = 1.260E-02 ppb
 BEC = 6.721E-02 ppb

Count/CPS(Y) [count/CPS]

Al standard calibration curve

Conc.(X) [ppb]

Curve Fit: Y=aX+b
 r = 0.9965
 Y = 2.385E+004*X +6.331E+004
 X = 4.193E-005*Y -2.654E+000
 DL = --- ppb
 BEC = 2.654 ppb

Count/CPS(Y) [count/CPS]

Mg standard calibration curve

Conc.(X) [ppb]

Curve Fit: Y=aX+b
 r = 0.9985
 Y = 2.082E+004*X +8.166E+004
 X = 4.803E-005*Y -3.922E+000
 DL = --- ppb
 BEC = 3.922 ppb

Count/CPS(Y) [count/CPS]

Sn standard calibration curve

Conc.(X) [ppb]

Curve Fit: Y=aX+b
 r = 0.9993
 Y = 3.544E+003*X +6.175E+002
 X = 2.822E-004*Y -1.742E-001
 DL = 1.420E-02 ppb
 BEC = 1.742E-01 ppb

Human subject ethics approval letter from the Ethical Committee, Faculty of

dentistry, university of Malaya

UNIVERSITY OF MALAYA
The Leader in Research & Innovation

MEDICAL ETHICS COMMITTEE
FACULTY OF DENTISTRY
ADDRESS: 50603, KUALA LUMPUR, MALAYSIA
TELEPHONE: 03-79676458 FAXIMILE: 03-79676456

NAME OF ETHICS COMMITTEE/IRB: Medical Ethics Committee, Faculty of Dentistry	ETHICS COMMITTEE/IRB REFERENCE NUMBER:
ADDRESS: 50603, Kuala Lumpur	OF DP1301/0037(L)

PROTOCOL NO:	
TITLE: Trace Elements of Human Teeth, a Biomarker of Environmental Burden of Toxic Metals	

PRINCIPAL INVESTIGATOR: Dr Mayeen Uddin Khandaker	SPONSOR:
TELEPHONE: KOMTEL:	

The following item [√] have been received and reviewed in connection with the above study to be conducted by the above investigator.

[√] Investigator's Checklist   Ver date: 26th APRIL 2013
[ ] Proof that project was presented at the department level (For postgraduate student only)   Ver date: 7th JUNE 2013
[ ] Synopsis Form (For Elective Project only)   Ver date:
[√] Application Form
[√] Study Protocol
[√] Brief CV of Principal Investigator
    Patient Information Sheet (PIS):
     [ ] BM version
     [√] English version
     [ ] Others: _____
    Consent Form:
     [√] BM version
     [√] English version
     [ ] Others: _____
[√] Questionnaire

and have been

[√] Approved
[ ] Conditionally approved (identify item and specify modification below or in accompanying letter)
[ ] Rejected (identify item and specify reasons below or in accompanying letter)

Investigator are required to:

1) follow instructions, guidelines and requirements of the Medical Ethics Committee.
2) report any protocol deviations/violations to Medical Ethics Committee.
3) comply with International Conference on Harmonization – Guidelines for Good Clinical Practice (ICH-GCP) and Declaration of Helsinki
4) note that Medical Ethics Committee may audit the approved study.

Date of approval: 13th June 2013

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                         ....
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APPENDIX K

Aalysis of a sample

Calculation of radioactivity, radiological hazard indicators, heavy metal concentration and daily intake of heavy metal of lady's finger sample collected from Kuala Selangor.

The activity concentration of radionuclides in the samples was calculated from the obtained net counts after the deduction of background counts using the following equation

$$A_c = \frac{N \times 1000}{\varepsilon_\gamma \times I_\gamma \times T_s \times M_s} \ (Bq \ kg^{-1})$$

Where, A_c = activity concentration of the radionuclide in the sample given in Bq kg^{-1}, N = net counts (net area) under corresponding photo-peak, ε_γ = detection efficiency corresponding to specific gamma-ray, I_γ = absolute transition probability of the specific gamma-ray (intensity), M_s = mass of the sample in gram, T_s = counting time in seconds.

Calculation of ^{226}Ra activity concentration

Sample 1

(i) For ^{214}Pb (energy 351.92 keV)

N = 848, ε_γ = 0.16508398, I_γ = 0.356 (35.6%), Ts = 86400 second, Ms = 269.9 gm and 1000 = gram to kg conversion.

$$A_c = \frac{848 \times 1000}{0.16508398 \times 0.356 \times 86400 \times 269.9} = 0.62 \ Bq \ kg^{-1}$$

(ii) For ^{214}Bi (energy 609.31 keV)

N = 679, ε_γ = 0.092461405, I_γ = 0.4549 (45.496%) (45.496%), Ts = 86400 second, Ms = 269.9 gm

$$A_c = \frac{679 \times 1000}{0.092461405 \times 0.4549 \times 86400 \times 269.9} = 0.69 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.62+0.69)/2 = 0.66 Bq kg^{-1}

Sample 2

(i) For ^{214}Pb (energy 351.92 keV)

N = 524, ε_γ = 0.16508398, I$_\gamma$ = 0.356 (35.6%), Ts = 86400 second, Ms = 245.1 gm

and 1000 = gram to kg conversion.

$$A_c = \frac{524 \times 1000}{0.16508398 \times 0.356 \times 86400 \times 245.1} = 0.42 \; Bq \; kg^{-1}$$

(ii) For ^{214}Bi (energy 609.31 keV)

N = 465, ε_γ = 0.092461405, I$_\gamma$ = 0.4549 (45.496%), Ts = 86400 second, Ms = 245.1 gm

$$A_c = \frac{465 \times 1000}{0.092461405 \times 0.4549 \times 86400 \times 245.1} = 0.52 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.42+0.52)/2 = 0.47 Bq kg^{-1}

Sample 3

(i) For ^{214}Pb (energy 351.92 keV)

N = 987, ε_γ = 0.16508398, I$_\gamma$ = 0.356 (35.6%), Ts = 86400 second, Ms = 252.1 gm

and 1000 = gram to kg conversion.

$$A_c = \frac{987 \times 1000}{0.16508398 \times 0.356 \times 86400 \times 252.1} = 0.77 \; Bq \; kg^{-1}$$

(ii) For ^{214}Bi (energy 609.31 keV)

N = 923, ε_γ = 0.092461405, I_γ = 0.4549 (45.496%), Ts = 86400 second, Ms = 252.1 gm

$$A_c = \frac{923 \times 1000}{0.092461405 \times 0.4549 \times 86400 \times 252.1} = 1.01 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.77+1.01)/2 = 0.89 Bq kg^{-1}

Sample 4

(i) For ^{214}Pb (energy 351.92 keV)

N = 722, ε_γ = 0.16508398, I_γ = 0.356 (35.6%), Ts = 86400 second, Ms = 353.3 gm and 1000 = gram to kg conversion.

$$A_c = \frac{722 \times 1000}{0.16508398 \times 0.356 \times 86400 \times 353.3} = 0.40 \; Bq \; kg^{-1}$$

(ii) For ^{214}Bi (energy 609.31 keV)

N = 607, ε_γ = 0.092461405, I_γ = 0.4549 (45.496%), Ts = 86400 second, Ms = 353.3 gm

$$A_c = \frac{607 \times 1000}{0.092461405 \times 0.4549 \times 86400 \times 353.3} = 0.47 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.40+0.47)/2 = 0.44 Bq kg^{-1}

Standard deviation (SD) = 0.2 (estimated using excel program)

i.e., Grand average Ac = (0.66+0.47+0.89+0.44)/4 = 0.61±0.2 Bq kg^{-1} (^{226}Ra activity of the representative sample).

Calculation of ^{228}Ra activity concentration

Sample 1

(i) For ^{208}Tl (energy 583.19 keV)

N = 574, ε_γ = 0.096839895, I_γ = 0.85 (85%), Ts = 86400 second, Ms = 269.9 gm and 1000 = gram to kg conversion.

$$A_c = \frac{574 \times 1000}{0.096839895 \times 0.85 \times 86400 \times 269.9} = 0.30 \; Bq \; kg^{-1}$$

(ii) For ^{228}Ac (energy 911.21 keV)

N = 679, ε_γ = 0.060449488, I_γ = 0.258 (25.8%) (45.496%), Ts = 86400 second, Ms = 269.9 gm

$$A_c = \frac{272 \times 1000}{0.060449488 \times 0.258 \times 86400 \times 269.9} = 0.75 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.30+0.75)/2 = 0.52 Bq kg^{-1}

Sample 2

(i) For ^{208}Tl (energy 583.19 keV)

N = 1365, ε_γ = 0.096839895, I_γ = 0.85 (85%), Ts = 86400 second, Ms = 245.1 gm and 1000 = gram to kg conversion.

$$A_c = \frac{1365 \times 1000}{0.096839895 \times 0.85 \times 86400 \times 224.1} = 0.78 \; Bq \; kg^{-1}$$

(ii) For ^{228}Ac (energy 911.21 keV)

N = 387, ε_γ = 0.060449488, I_γ = 0.258 (25.8%) (45.496%), Ts = 86400 second, Ms = 245.1 gm

$$A_c = \frac{387 \times 1000}{0.060449488 \times 0.258 \times 86400 \times 245.1} = 1.17 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.78+1.17)/2 = 0.98 Bq kg^{-1}

Sample 3

(i) For ^{208}Tl (energy 583.19 keV)

N = 183, ε_γ = 0.096839895, I$_\gamma$ = 0.85 (85%), Ts = 86400 second, Ms = 252.1 gm and

1000 = gram to kg conversion.

$$A_c = \frac{183 \times 1000}{0.096839895 \times 0.85 \times 86400 \times 252.1} = 0.10 \; Bq \; kg^{-1}$$

(ii) For ^{228}Ac (energy 911.21 keV)

N = 104, ε_γ = 0.060449488, I$_\gamma$ = 0.258 (25.8%) (45.496%), Ts = 86400 second, Ms = 252.1 gm

$$A_c = \frac{104 \times 1000}{0.060449488 \times 0.258 \times 86400 \times 252.1} = 0.31 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.10+0.31)/2 = 0.20 Bq kg^{-1}

Sample 4

(i) For ^{208}Tl (energy 583.19 keV)

N = 411, ε_γ = 0.096839895, I$_\gamma$ = 0.85 (85%), Ts = 86400 second, Ms = 353.3 gm and

1000 = gram to kg conversion.

$$A_c = \frac{411 \times 1000}{0.096839895 \times 0.85 \times 86400 \times 353.3} = 0.16 \; Bq \; kg^{-1}$$

(ii) For ^{228}Ac (energy 911.21 keV)

N = 219, ε_γ = 0.060449488, I_γ = 0.258 (25.8%) (45.496%), Ts = 86400 second, Ms = 353.3 gm

$$A_c = \frac{219 \times 1000}{0.060449488 \times 0.258 \times 86400 \times 353.3} = 0.46 \; Bq \; kg^{-1}$$

i.e., Average Ac = (0.16+0.46)/2 = 0.31 Bq kg^{-1}

Standard deviation (SD) = 0.3 (estimated using excel program)

i.e., Grand average Ac = (0.52+0.98+0.20+0.31)/4 = 0.50±0.3 Bq kg^{-1} (^{228}Ra activity of the representative sample).

Calculation of ^{40}K activity concentration

Sample 1
Energy 1460.82 keV)

N = 22870, ε_γ = 0.036722786, I_γ = 0.1066 (10.66%), Ts = 86400 second, Ms = 269.9 gm and 1000 = gram to kg conversion.

$$A_c = \frac{22870 \times 1000}{0.036722786 \times 0.1066 \times 86400 \times 269.9} = 250.53 \; Bq \; kg^{-1}$$

Sample 2
Energy 1460.82 keV)

N = 4164, ε_γ = 0.036722786, I_γ = 0.1066 (10.66%), Ts = 86400 second, Ms = 245.1 gm and 1000 = gram to kg conversion.

$$A_c = \frac{4164 \times 1000}{0.036722786 \times 0.1066 \times 86400 \times 245.1} = 50.23 \; Bq \; kg^{-1}$$

Sample 3

Energy 1460.82 keV)

N = 23568, ε_γ = 0.036722786, I_γ = 0.1066 (10.66%), Ts = 86400 second, Ms = 252.1 gm and 1000 = gram to kg conversion.

$$A_c = \frac{23568 \times 1000}{0.036722786 \times 0.1066 \times 86400 \times 252.1} = 276.40 \; Bq \; kg^{-1}$$

Sample 4

Energy 1460.82 keV)

N = 27764, ε_γ = 0.036722786, I_γ = 0.1066 (10.66%), Ts = 86400 second, Ms = 353.3 gm and 1000 = gram to kg conversion.

$$A_c = \frac{27764 \times 1000}{0.036722786 \times 0.1066 \times 86400 \times 353.3} = 232.34 \; Bq \; kg^{-1}$$

Standard deviation (SD) = 103 (estimated using excel program)

i.e., Grand average Ac = (250.53+50.23+276.40+232.34)/4 = 202±103 Bq kg⁻¹ (⁴⁰K activity of the representative sample).

Calculation of daily and annual intake of radionuclides (²²⁶Ra, ²²⁸Ra and ⁴⁰K) due to consumption of lady's finger

Daily intake of radionuclides were calculated by the following equation:

$$D_{int} = \frac{A_c \times A_p \times F}{M_p \times Y_d} = \frac{A_c \times C_r}{Y_d} (Bq)$$

and annual intake of radionuclides, $A_{int} = D_{int} \times Y_d$

Where, D_{int} = daily intake of radionuclides, A_c = activity concentration of respective radionuclides, vegetable consumption rate, $C_r = (A_p \times F)/M_p$, A_p = annual production of vegetables, F = fraction of the edible part of the vegetables (90% based on local knowledge of vegetable consumption), M_p = population of Malaysia and Y_d = days in a year.

Calculation of daily and annual intake of ^{226}Ra

Sample 1

Ac = 0.66 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and Y_d = 365 days

$$D_{int} = \frac{0.66 \times 6.6}{365} = 0.012 \ Bq$$

And A_{int} = 0.012×365 = 4.38 Bq

Sample 3

Ac = 0.89 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and Y_d = 365 days

$$D_{int} = \frac{0.89 \times 6.6}{365} = 0.016 \ Bq$$

And A_{int} = 0.016×365 = 5.84 Bq

Sample 2

Ac = 0.47 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and Y_d = 365 days

$$D_{int} = \frac{0.47 \times 6.6}{365} = 0.008 \ Bq$$

And A_{int} = 0.008×365 = 2.92 Bq

Sample 4

Ac = 0.44 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and Y_d = 365 days

$$D_{int} = \frac{0.44 \times 6.6}{365} = 0.008 \ Bq$$

And A_{int} = 0.008×365 = 2.92 Bq

i.e., Average D_{int} = (0.012+0.008+0.016+0.008)/4 = 0.011 Bq (11.0 mBq)

and A_{int} = (4.38+2.92+5.84+2.92) Bq/4 = 4.02 Bq.

Calculation of daily and annual intake of ^{228}Ra

Sample 1

Ac = 0.52 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and Y_d = 365 days

$$D_{int} = \frac{0.52 \times 6.6}{365} = 0.0094 \ Bq$$

And A_{int} = 0.0094×365 = 3.43 Bq

Sample 2

Ac = 0.98 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and Y_d = 365 days

$$D_{int} = \frac{0.98 \times 6.6}{365} = 0.0177 \ Bq$$

Sample 3

$Ac = 0.20$ Bq kg^{-1}, $C_r = 6.6$ kg y^{-1}

and $Y_d = 365$ days

$$D_{int} = \frac{0.20 \times 6.6}{365} = 0.0036 \; Bq$$

And $A_{int} = 0.0036 \times 365 = 1.31$ Bq

And $A_{int} = 0.0177 \times 365 = 6.46$ Bq

Sample 4

$Ac = 0.31$ Bq kg^{-1}, $C_r = 6.6$ kg y^{-1}

and $Y_d = 365$ days

$$D_{int} = \frac{0.31 \times 6.6}{365} = 0.0056 \; Bq$$

And $A_{int} = 0.0056 \times 365 = 2.04$ Bq

i.e., Average $D_{int} = (0.0094+0.0177+0.00036+0.0056)/4 = 0.00908$ Bq (9.08 mBq)

and $A_{int} = (3.43+6.46+1.31+2.04)$ Bq/4 $= 3.31$ Bq.

Calculation of daily and annual intake of ^{40}K

Sample 1

$Ac = 250.53$ Bq kg^{-1}, $C_r = 6.6$ kg y^{-1}

and $Y_d = 365$ days

$$D_{int} = \frac{250.53 \times 6.6}{365} = 4.53 \; Bq$$

And $A_{int} = 4.53 \times 365 = 1653.45$ Bq

Sample 2

$Ac = 50.23$ Bq kg^{-1}, $C_r = 6.6$ kg y^{-1}

and $Y_d = 365$ days

$$D_{int} = \frac{50.23 \times 6.6}{365} = 0.908 \; Bq$$

And $A_{int} = 0.908 \times 365 = 331.42$ Bq

Sample 3

$Ac = 276.40$ Bq kg^{-1}, $C_r = 6.6$ kg y^{-1}

and $Y_d = 365$ days

$$D_{int} = \frac{276.40 \times 6.6}{365} = 4.998 \; Bq$$

And $A_{int} = 4.998 \times 365 = 1824.27$ Bq

Sample 4

$Ac = 232.34$ Bq kg^{-1}, $C_r = 6.6$ kg y^{-1}

and $Y_d = 365$ days

$$D_{int} = \frac{232.34 \times 6.6}{365} = 4.201 \; Bq$$

And $A_{int} = 4.201 \times 365 = 1533.40$ Bq

i.e., Average $D_{int} = (4.53+0.908+4.998+4.201)/4 = 3.66$ Bq

and $A_{int} = (1653.45+331.42+1824.27+1533.40)$ Bq/4 $= 1335.63$ Bq.

Calculation of committed effective dose to man due to intake of radionuclides (^{226}Ra, ^{228}Ra and ^{40}K) for the consumption of lady's finger

Committed effective doses were calculated by the following formula:

$$D_{eff} = A_c \times C_r \times D_{cf}$$

Where, D_{eff} = annual committed effective dose to an individual (μSv y^{-1}), A_c = activity concentration of respective radionuclides, C_r = annual consumption rate of vegetable, Dcf = ingestion dose conversion factor (adult) for the radionuclides of interest (2.8×10^{-7} Sv Bq^{-1} for ^{226}Ra, 6.9×10^{-7} Sv Bq^{-1} for ^{228}Ra and 6.2×10^{-9} Sv Bq^{-1} for ^{40}K).

Calculation of annual committed effective dose of ^{226}Ra

Sample 1

Ac = 0.66 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 2.8×10^{-7} Sv Bq^{-1}

D_{eff} = 0.66×6.6×2.8 × 10^{-7} = 1.22 μSv y^{-1}

Sample 2

Ac = 0.47 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 2.8×10^{-7} Sv Bq^{-1}

D_{eff} = 0.47×6.6×2.8 × 10^{-7} = 0.87 μSv y^{-1}

Sample 3

Ac = 0.89 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 2.8×10^{-7} Sv Bq^{-1}

D_{eff} = 0.89×6.6×2.8 × 10^{-7} = 1.64 μSv y^{-1}

Sample 4

Ac = 0.44 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 2.8×10^{-7} Sv Bq^{-1}

D_{eff} = 0.44×6.6×2.8 × 10^{-7} = 0.81 μSv y^{-1}

i.e., Average D_{eff} for ^{226}Ra = (1.22+0.87+1.64+0.81)/4 = 1.14 μSv y^{-1}.

Calculation of annual committed effective dose of ^{228}Ra

Sample 1

Ac = 0.52 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.9×10^{-7} Sv Bq^{-1}

D_{eff} = 0.52×6.6×6.9 × 10^{-7} = 2.37 μSv y^{-1}

Sample 2

Ac = 0.98 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.9×10^{-7} Sv Bq^{-1}

D_{eff} = 0.98×6.6×6.9 × 10^{-7} = 4.46 μSv y^{-1}

Sample 3

Ac = 0.20 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.9×10^{-7} Sv Bq^{-1}

Sample 4

Ac = 0.31 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.9×10^{-7} Sv Bq^{-1}

$D_{eff} = 0.20 \times 6.6 \times 6.9 \times 10^{-7} = 0.91~\mu Sv~y^{-1}$ $D_{eff} = 0.31 \times 6.6 \times 6.9 \times 10^{-7} = 1.41~\mu Sv~y^{-1}$

i.e., Average D_{eff} for ^{228}Ra = (2.37+4.46+0.91+1.41)/4 = 2.29 $\mu Sv~y^{-1}$.

Calculation of annual committed effective dose of ^{40}K

Sample 1

A_c = 250.53 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.2 $\times$ 10^{-9} Sv Bq^{-1}

$D_{eff} = 250.53 \times 6.6 \times 6.2 \times 10^{-9} = 10.25~\mu Sv~y^{-1}$

Sample 2

A_c = 50.23 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.2 $\times$ 10^{-9} Sv Bq^{-1}

$D_{eff} = 50.23 \times 6.6 \times 6.2 \times 10^{-9} = 2.06~\mu Sv~y^{-1}$

Sample 3

A_c = 276.40 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.2 $\times$ 10^{-9} Sv Bq^{-1}

$D_{eff} = 276.40 \times 6.6 \times 6.2 \times 10^{-9} = 11.31~\mu Sv~y^{-1}$

Sample 4

A_c = 232.34 Bq kg^{-1}, C_r = 6.6 kg y^{-1}

and D_{cf} = 6.2 $\times$ 10^{-9} Sv Bq^{-1}

$D_{eff} = 232.34 \times 6.6 \times 6.2 \times 10^{-9} = 9.51~\mu Sv~y^{-1}$

i.e., Average D_{eff} for ^{40}K = (10.25+2.06+11.31+9.51)/4 = 8.28 $\mu Sv~y^{-1}$.

Calculation of total committed effective dose

The total committed dose via consumption of vegetables was calculated by the following equation:

$$D_{eff}^{total} = \sum_{i=226_{Ra,}~228_{Ra,}~40_K} D_{eff,i}$$

The total committed effective dose, D_{eff} = 1.14+2.29+8.28 = 11.71 $\mu Sv~y^{-1}$.

Calculation of excess lifetime cancer risk (ELCR)

Excess lifetime cancer risk due to intake of radionuclides via the consumption of foods was estimated using the following formula:

$$ELCR = A_{ir} \times A_{ls} \times R_c$$

where, A_{ir} = annual intake of radionuclide (Bq), A_{ls} = average span of life (74 y) and R_c = mortality risk coefficient (Bq^{-1}) for the ingestion of food, respectively. The

ingestion mortality cancer risk coefficients are 9.56×10^{-9} Bq^{-1} for ^{226}Ra, 2.74×10^{-8} Bq^{-1} for ^{228}Ra and 5.89×10^{-10} Bq^{-1} for ^{40}K).

Calculation of lifetime cancer risk (ELCR) for ^{226}Ra

Sample 1

$A_{ir} = 4.38$ Bq, $A_{ls} = 74$ y

and $R_c = 9.56 \times 10^{-9}$ Bq^{-1}

ELCR = $4.38 \times 74 \times 9.56 \times 10^{-9} = 3.1 \times 10^{-6}$

Sample 2

$A_{ir} = 2.29$ Bq, $A_{ls} = 74$ y

and $R_c = 9.56 \times 10^{-9}$ Bq^{-1}

ELCR = $2.29 \times 74 \times 9.56 \times 10^{-9} = 1.62 \times 10^{-6}$

Sample 3

$A_{ir} = 5.84$ Bq, $A_{ls} = 74$ y

and $R_c = 9.56 \times 10^{-9}$ Sv Bq^{-1}

ELCR = $5.84 \times 74 \times 9.56 \times 10^{-9} = 4.13 \times 10^{-6}$

Sample 4

$A_{ir} = 2.29$ Bq, $A_{ls} = 74$ y

and $R_c = 9.56 \times 10^{-9}$ Sv Bq^{-1}

ELCR = $2.92 \times 74 \times 9.56 \times 10^{-9} = 1.62 \times 10^{-6}$

i.e., Average ELCR for ^{226}Ra = $(3.1+1.62+4.13+1.62) \times 10^{-6}/4 = 2.6 \times 10^{-6}$.

Calculation of lifetime cancer risk (ELCR) for ^{228}Ra

Sample 1

$A_{ir} = 3.43$ Bq, $A_{ls} = 74$ y

and $R_c = 2.74 \times 10^{-8}$ Bq^{-1}

ELCR = $3.43 \times 74 \times 2.74 \times 10^{-8} = 6.95 \times 10^{-6}$

Sample 2

$A_{ir} = 6.46$ Bq, $A_{ls} = 74$ y

and $R_c = 2.74 \times 10^{-8}$ Bq^{-1}

ELCR = $6.46 \times 74 \times 2.74 \times 10^{-8} = 1.31 \times 10^{-5}$

Sample 3

$A_{ir} = 1.31$ Bq, $A_{ls} = 74$ y

and $R_c = 2.74 \times 10^{-8}$ Bq^{-1}

ELCR = $1.31 \times 74 \times 2.74 \times 10^{-8} = 2.66 \times 10^{-6}$

Sample 4

$A_{ir} = 2.04$ Bq, $A_{ls} = 74$ y

and $R_c = 2.74 \times 10^{-8}$ Bq^{-1}

ELCR = $2.04 \times 74 \times 2.74 \times 10^{-8} = 4.14 \times 10^{-6}$

i.e., Average ELCR for ^{228}Ra = $(6.95 \times 10^{-6} + 1.31 \times 10^{-5} + 2.66 \times 10^{-6} + 4.14 \times 10^{-6})/4 = 6.7 \times 10^{-6}$.

Calculation of lifetime cancer risk (ELCR) for ^{40}K

Sample 1

A_{ir} = 1653.45 Bq, A_{ls} = 74 y

and R_c = 5.89 $\times 10^{-10}$ Bq^{-1}

ELCR = 1653.45 $\times 74 \times 5.89 \times 10^{-10}$ = 7.2$\times 10^{-5}$

Sample 2

A_{ir} = 331.42 Bq, A_{ls} = 74 y

and R_c = 5.89 $\times 10^{-10}$ Bq^{-1}

ELCR = 331.42 $\times 74 \times 5.89 \times 10^{-10}$ = 1.4$\times 10^{-5}$

Sample 3

A_{ir} = 1824.27 Bq, A_{ls} = 74 y

and R_c = 5.89 $\times 10^{-10}$ Bq^{-1}

ELCR = 1824.3$\times 74 \times 5.89 \times 10^{-10}$ = 7.95$\times 10^{-5}$

Sample 4

A_{ir} = 1533.4 Bq, A_{ls} = 74 y

and R_c = 5.89 $\times 10^{-10}$ Bq^{-1}

ELCR = 1533.4 $\times 74 \times 5.89 \times 10^{-10}$ = 6.68$\times 10^{-5}$

i.e., Average ELCR for ^{40}K = (7.2 + 1.4 + 7.95 + 6.68) $\times 10^{-5}$ /4 = 5.8$\times 10^{-5}$.

Calculation of soil-to-plant (rice grain) transfer factor (TF) of radionuclides (^{226}Ra, ^{228}Ra and ^{40}K) at Kampung Sakan, Kedah

Soil-to-rice transfer factor (TF) of radionuclides was estimated using the following relationship:

$$TF = \frac{C_i^r (Bq\ kg^{-1},\ dry\ weight)}{C_i^s (Bq\ kg^{-1},\ dry\ weight)}$$

where, TF is transfer factor of soil-to-rice; C_i^r is the concentration of radionuclides in Bq kg^{-1} dry rice weight, and C_i^s is the concentration of radionuclides in Bq kg^{-1} dry soil weight in the upper 20 cm layer of soil.

Calculation of soil-to-rice TF for ^{226}Ra

Sample 1

Rice activity, C_r = 2.67 Bq kg^{-1}

Soil activity, C_s = 7.26 Bq kg^{-1}

Sample 2

Rice activity, C_r = 1.64 Bq kg^{-1}

Soil activity, C_s = 5.08 Bq kg^{-1}

$$TF = \frac{2.67}{7.26} = 0.37 \qquad\qquad TF = \frac{1.64}{5.08} = 0.32$$

Sample 3

Rice activity, $C_r = 2.46$ Bq kg^{-1}

Soil activity, $C_s = 9.16$ Bq kg^{-1}

$$TF = \frac{2.46}{9.16} = 0.27$$

i.e., Average soil-to-rice TF of ^{226}Ra = (0.37 + 0.32+ 0.27)/3 = 0.32.

Calculation of soil-to-rice TF for ^{232}Th

Sample 1   **Sample 2**

Rice activity, $C_r = 4.72$ Bq kg^{-1} Rice activity, $C_r = 5.26$ Bq kg^{-1}

Soil activity, $C_s = 9.64$ Bq kg^{-1} Soil activity, $C_s = 13.48$ Bq kg^{-1}

$$TF = \frac{4.72}{9.64} = 0.49 \qquad\qquad TF = \frac{5.26}{13.48} = 0.39$$

Sample 3

Rice activity, $C_r = 6.43$ Bq kg^{-1}

Soil activity, $C_s = 11.77$ Bq kg^{-1}

$$TF = \frac{6.43}{11.77} = 0.55$$

i.e., Average soil-to-rice TF of ^{232}Th = (0.49 + 0.39+ 0.55)/3 = 0.48.

Calculation of soil-to-rice TF for ^{40}K

Sample 1   **Sample 2**

Rice activity, $C_r = 87.04$ Bq kg^{-1} Rice activity, $C_r = 91.56$ Bq kg^{-1}

Soil activity, $C_s = 70.47$ Bq kg^{-1} Soil activity, $C_s = 81.69$ Bq kg^{-1}

$$TF = \frac{87.04}{70.47} = 1.24 \qquad\qquad TF = \frac{91.56}{81.69} = 1.12$$

Sample 3

Rice activity, C_r = 97.86 Bq kg^{-1}

Soil activity, C_s = 77.26 Bq kg^{-1}

$$TF = \frac{97.86}{77.26} = 1.27$$

i.e., Average soil-to-rice TF of ^{40}K = (1.24 + 1.12 + 1.27)/3 = 1.21.

Calculation the concentration and daily intake of heavy metal in lady's finger sample collected from Kuala Selangor

Daily intake (body weight-bw) of heavy metal (DIM) was calculated by the following formula:

$$DIM\ (bw) = \frac{C_{metal} \times D_{cr}}{W}\ (\mu g\ day^{-1})$$

where, C_{metal} is the concentration of heavy metals in lady's finger vegetable D_{cr} represents daily consumption (rate) of vegetable and W is the adults body weight (70 kg).

Calculation of Daily intake for Manganese (Mn)

Sample 1

Concentration of Mn (C_{Mn}) was found from ICP-MS = 1035 ppb (parts per billion)

1 ppb = 0.001 mg kg^{-1}

1035 ppb = 1.035 mg kg^{-1}

Lady's finger consumption rate,

D_{cr} = 6.6 kg/y = 0.0181 kg/day

Adult body weight, W = 70 kg

1 mg = 1000 μg

Sample 2

Concentration of Mn (C_{Mn}) was found from ICP-MS = 847.9 ppb (parts per billion)

1 ppb = 0.001 mg kg^{-1}

847.9 ppb = 0.8479 mg kg^{-1}

Lady's finger consumption rate,

D_{cr} = 6.6 kg/y = 0.0181 kg/day

Adult body weight, W = 70 kg

1 mg = 1000 μg

$$\text{DIM (bw)} = \frac{1.035 \times 0.0181 \times 1000}{70}$$

$$= 0.268 \ \mu\text{g day}^{-1}$$

Daily intake equivalent to a 70-kg adult

$$= 0.268 \times 70 = 18.8 \ \mu\text{g day}^{-1}$$

$$\text{DIM (bw)} = \frac{0.8479 \times 0.0181 \times 1000}{70}$$

$$= 0.219 \ \mu\text{g day}^{-1}$$

Daily intake equivalent to a 70-kg adult

$$= 0.219 \times 70 = 15.3 \ \mu\text{g day}^{-1}$$

Sample 3

Concentration of Mn (C_{Mn}) was found from ICP-MS = 874.2 ppb (parts per billion)

1 ppb = 0.001 mg kg^{-1}

874.2 ppb = 0.8742 mg kg^{-1}

Lady's finger consumption rate,

D_{cr} = 6.6 kg/y = 0.0181 kg/day

Adult body weight, W = 70 kg

1 mg = 1000 μg

$$\text{DIM} = \frac{0.8742 \times 0.0181 \times 1000}{70}$$

$$= 0.226 \ \mu\text{g day}^{-1}$$

Daily intake equivalent to a 70-kg adult

$$= 0.226 \times 70 = 15.8 \ \mu\text{g day}^{-1}$$

i.e., Average concentration of Mn = (1.035+0.8479+0.8742)/3 = 0.92 mg kg^{-1}

Average daily intake (bw) of Mn = (0.268 + 0.219+ 0.226)/3 = 0.24 μg day^{-1} bw.

Average daily intake of Mn equivalent to a 70-kg adult = (18.8 + 15.3+ 15.8)/3 = 16.6 μg day^{-1}