Measurements of Natural Radionuclides and Elemental Concentrations in Chicken and Mice Bones in Penang, Malaysia

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Published: 1 December 2012

ABSTRACT: Natural radionuclides and major element concentrations were measured in mice and chicken bones from Penang, Malaysia, using High Purity Germanium (HPGe) and Energy-dispersive X-ray (EDX) spectroscopes. Concentrations in chicken bones were found to be ^{210}Pb (0.052±0.011 Bq g^{-1}), ^{137}Cs (0.053±0.012 Bq g^{-1}), ^{228}Th (0.040±0.009 Bq g^{-1}), ^{226}Ra (0.146±0.073 Bq g^{-1}), ^{228}Ra (0.423±0.098 Bq g^{-1}) and ^{40}K (4.970±0.399 Bq g^{-1}). Values for mice bones were found to be 0.090±0.059 Bq g^{-1}, 0.163±0.083 Bq g^{-1}, 0.049±0.043 Bq g^{-1}, 0.510±0.075 Bq g^{-1}, 1.670±0.324 Bq g^{-1}, and 23.010±1.492 Bq g^{-1}, respectively. For elements such as Na, Mn, Si, P, S, Cl, K, Ca, Sn, Fe and Br concentrations were found to be 2.67%, 0.78%, 0.19 %, 15.99 %, 0.32%, 2.58 %, 1.5 %, 28.61 %, 0 %, 2.42 % and 0.21 %, respectively in chicken bones and 3.13%, 0.87%, 0 %, 14.53 %, 1.17 %, 1.43 %, 9.48 %, 20.61 %, 0.52 %, 0.51 %, and 0 %, respectively in mice bones. The degree of accumulations of elements in the chicken and mice bones were as follows: Ca> P> Na> Cl> Fe> K> Mn> S> Br> Si> Sn and Ca> P> K > Na> Cl> S> Mn> Sn> Fe> (Si, Br), respectively. Radionuclides concentrations were higher in mice bones than in chicken bones. A good positive correlation was observed between potassium concentrations using EDX and HPGe spectroscopes, R = 0.59.

Keywords: Bone-seeking, environmental radioactivity, elements

Introduction

^{226}Ra and ^{228}Ra isotopes are considered as the most important natural radionuclides of the ^{238}U and ^{232}Th series, respectively. In the human body the radioisotopes behave chemically and physiologically like calcium and are inclined to concentrate in the bones. Uranium may enter the body through the skin, lungs or gut. Once uranium enters the systemic circulation, it is distributed throughout the body. ^{226}Ra is a bone-seeking radionuclide that accumulates in calcareous tissues because of its chemical similarity to calcium (Whicker and Schultz, 1982).

In the physical environment radioisotopes form soluble compounds and can contaminate underground reservoirs, soils, plantations, food sources and consequently the human food chain. ^{226}Ra movement through food chains is "moderate", and some 99% of ^{226}Ra body content is in human bones (ICRP Committee II, 1960). It is expected that ^{226}Ra would be present in bones because it tends to be moderately transferable in the physical environment.

Radium taken up by vegetation from the soil and it is assimilated efficiently from the gut when ingested by animals (NCRP, 1999). The common long-lived radioactive elements, uranium and thorium decay to produce other radionuclices, such as radium, which undergo radioactive decay. ^{226}Ra is moderately soluble in water and can enter ground water by dissolution of aquifer materials, desorption from rock or sediment surfaces and emission from minerals by radioactive decay. ^{226}Ra decays with alpha particles emission to the inert gas radon (^{222}Rn) without color, odor, or taste.

Within the uranium-radium decay chain, radon is produced in almost every soil. Radon decays over a series of short-lived (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) and long-lived (^{210}Pb, ^{210}Bi and ^{210}Po) isotopes, and then finally becomes stable as ^{206}Pb. Atmospheric ^{210}Pb is mainly produced within the atmosphere by decay of ^{222}Rn; its direct precursor is ^{214}Po. Radon isotopes are members of the natural decay series, that is the ^{238}U decay series (^{222}Rn, $T_{1/2}$ = 3.8 days), the ^{232}Th decay series (^{220}Rn, $T_{1/2}$ = 56 s) and the ^{235}U decay series (^{219}Rn, $T_{1/2}$ = 3.9 s).

The retention of ^{226}Ra in bones is high and accumulates over time under conditions of chronic intake. Levels measured in human bones from several urban locations have ranged from 0.03 Bq kg^{-1} to 0.37 Bq kg^{-1} (Eisenbud and Gesell, 1997). The population's primary source of gamma exposure is naturally occurring radionuclides, particularly ^{40}K, which is found in soil, water, meats and high-potassium foods such as bananas.

The largest doses and risks of radionuclides in the physical environment are from natural occurring radionuclides, mostly ^{210}Po, ^{210}Pb, ^{228}Th, and ^{232}Th. Once lead enters the blood system it is distributed throughout the body, accumulating in bones and lingering from several years to decades (Brito *et al.*, 2005).

Among the radionuclides derived from anthropogenic sources, ^{137}Cs is the major source but represents only 2% to 3% of the total gamm-ray dose rate. ^{137}Cs is derived from land-mammal consumption. Previous studies have shown that ^{137}Cs distribution in surface soils is attributed to differences in climatic and topographical situation in a pertinent location (Tang *et al.*, 2002).

Concerns of heavy metals as natural elements of the Earth's crust are mainly due to its stability which are not easily degraded or destroyed. Heavy metals are toxic pollutants and accumulations in ecosystems and they are released to the biosphere from volcanoes, the erosion of rocks, wind-blowing dusts, forest fires, mining, combustion of fuels, industrial, sewage, and other anthropogenic activities. To a small extent, they enter our bodies via food, drinking water, and air. As trace elements (Na, Mn, Si, P, S, Cl, K, Ca, Sn, Fe, and Br) are essential to maintain the metabolism of the human body however; higher concentrations may lead to poisoning.

Heavy metal poisoning could result from drinking-water contamination (for example lead pipes) and high ambient air concentrations near emission sources. Potentially poisonous trace elements are released into the environment as a result of a wide range of industrial activities as well as the combustion of fossil fuels. While not being essential for deriving a tolerable intake, data relating to the concentration of radionuclides in bones is useful for linking the Lowest-Observed-Adverse-Effect-Levels (LOAELs) to organ-specific data. Data are produced from biokinetic models, commonly used for assessing radiological and chemical effects in radiological protection (Spoor and Hursh, 1973). Radionuclides concentrations of water and soil are good indicators of the levels of pollution.

A baseline survey of natural radionuclides and selected elements in mice and chicken bones was conducted for an assessment of any radiological contamination of animals resulting from inhalation and/or ingestion soil dust and from water in Penang state of Peninsular Malaysia.

Materials and Methods

Eight chicken and four mice bone samples were collected from Penang state. The bones samples were deep frozen to eliminate unwanted effects such as tissue decay and freeing the work schedule of collecting samples from diverse locations. Following from that process, soft tissues surrounding bones were removed from all samples. The bones were then incinerated at a temperature of 400°C in an electric furnace (Nabertherm, Germany) with time spanning 24 h. Next, the bones were homogenized by mortar and sifted through a 0.5 mm sieve.

Each sample was then weighed and sealed in standard Perspex containers (diameter of 4.5 cm and height of 2 cm). The sample weight varied between 1.90g and 15.27g. Measurements of gamma-ray emitters were performed without the need for chemical treatment of the samples using the HPGe spectroscopy shielded with 10 cm of lead, and connected to a multichannel analyser computer board (MAESTRO-32) with 16,384 channels. The background spectrum of containers was striped from the spectra of bones samples.

The HPGe detector was calibrated and the absolute detection efficiency was determined using standard reference materials from the International Atomic Energy Agency (IAEA) as reported elsewhere (Almayahi *et al.*, 2012). Bones samples were counted for 57,600 s and 8,6600 s. Where gamma-ray peaks of 47 keV (^{210}Pb), 352 keV (^{226}Ra), 583 keV (^{208}Tl peaks indicating ^{228}Th), 609 keV (^{226}Ra), 662 keV (^{137}Cs), and 911 keV, 969 keV (^{228}Ac peaks indicating ^{228}Ra), and 1462 keV (^{40}K) were found. The quantification elemental analysis to identify the weight percentage of elements like Na, Mn, Si, P, S, Cl, K, Ca, Sn, Fe and Br in the samples was done using the Energy Dispersive X-ray spectrometer (EDS, Model 7573). This technique is used in different environmental science and technological research and applications.

The microphotographs for samples were recorded using SEM spectroscopy (model JSM-6460LV, Japan) with Secondary Electron Image (SEI). The magnification in this study is of 5,000 X for all samples. SEM spectroscopy was calibrated using standard elements: C ($CaCO_3$), O (SiO_2), Na

(Albite), Mg (MgO), P (GaP), Cl (KCl), K (MAD-10 Feldspar), Ca (Wollastonite), and Fe (Fe).

EDS measurements uses samples prepared using hydraulic pelletizing press (HERZOG Compression, Japan). In this study the diameter of 1.5 cm was used and of the press piston for all samples was determined as 30 ton and sample. In low-level radioactivity measurements it is important to quantify the minimum activity that is reliably detected – this is known as the minimum detectable activity (MDA); and refers to the measurable minimum activity that is detected using gamma-ray measuring with value of confidence (Knoll, 2000). The equation for determination of MDA is as follows (Currie, 1968):

$$\text{MDA (Bq g}^{-1}) = \frac{4.65\sqrt{\text{BG}}}{\varepsilon T_\gamma t m} \quad (1)$$

where BG = the number of counts for the background spectrum, ε = the absolute efficiency of the detector, T_γ = the emission probability of γ-ray, t = the time of measurement, and m = the weight of the dried sample (g).

Results

Table 1 presents the results for radionuclides concentrations. **Figure 1** shows the comparison between average radionuclides concentrations in chicken and mice bones using HPGe spectroscopy.

Table 1: Natural radionuclide concentrations (Bq g^{-1}) in bones samples.

SC	^{210}Pb	^{137}Cs	^{228}Th	^{226}Ra	^{228}Ra	^{40}K
Chicken						
*001	0.059±0.009	0.051±0.011	0.089±0.009	0.139±0.018	0.504±0.082	4.43±0.288
*002	ND	0.047±0.010	0.069±0.008	0.101±0.017	0.426±0.076	5.26±0.304
*003	0.060±0.015	0.094±0.021	0.022±0.008	0.193±0.460	0.397±0.150	5.03±0.618
*004	0.033±0.013	0.048±0.011	0.028±0.008	0.098±0.012	0.232±0.093	3.22±0.284
*005	0.069±0.012	0.008±0.016	ND	0.239±0.023	0.431±0.099	7.15±0.522
*006	0.057±0.011	0.061±0.009	0.062±0.012	0.118±0.018	0.456±0.090	4.39±0.374
*007	0.071±0.015	0.035±0.014	0.008±0.017	0.190±0.023	0.625±0.086	7.15±0.494
*008	0.067±0.016	0.084±0.011	0.043±0.014	0.093±0.016	0.314±0.115	3.18±0.310
Avg.	**0.052±0.011**	**0.053±0.012**	**0.040±0.009**	**0.146±0.073**	**0.423±0.098**	**4.97±0.399**
Mice						
*009	0.091±0.109	0.126±0.096	0.001±0.038	1.190±0.123	2.586±0.498	45.8±2.600
*010	0.061±0.028	0.038±0.021	0.035±0.015	0.104±0.022	0.611±0.106	11.20±0.573
*011	ND	0.190±0.021	0.066±0.033	0.311±0.048	0.955±0.207	14.43±0.838
*012	0.210±0.102	0.299±0.080	0.094±0.088	0.437±0.117	2.562±0.495	20.64±1.959
Avg.	**0.090±0.059**	**0.163±0.083**	**0.049±0.043**	**0.510±0.075**	**1.670±0.324**	**23.01±1.492**

SC = Sample code; ND= Not detectable

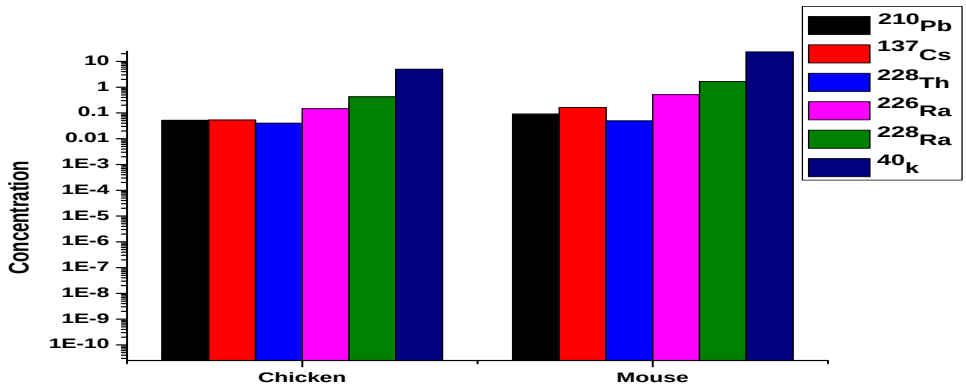

Figure 1: Comparison between average radionuclides concentrations in chicken and mice bones using HPGe spectroscopy, scale type $\log_{10}$.

Table 2 below presents the results for the weight percentage of elements, such as: Na, Mn, Si, P, S, Cl, K, Ca, Sn, Fe and Br obtained from EDS for all samples. Typical microphotograph and spectrum of bones samples using SEM and EDS are shown in **Figure 2**. A positive correlation between EDX and HPGe spectroscopes was obtained for potassium concentrations as shown in **Figure 3**.

Table 2: Elemental concentrations in bones samples using EDX spectrometer

Sc	Na%	Mn%	Si%	P%	S%	Cl%	K%	Ca%	Sn%	Fe%	Br%
Chicken											
*001	1.60	0.81	ND	17.71	0.32	2.19	1.43	34.33	ND	2.06	ND
*004	2.11	0.84	0.76	15.76	ND	1.91	1.10	28.23	ND	4.92	0.84
*006	4.59	0.81	ND	15.32	0.55	5.02	2.60	25.94	ND	2.13	ND
*007	2.60	0.77	ND	15.18	0.42	2.37	1.93	25.94	ND	2.66	ND
Avg	**2.67**	**0.78**	**0.19**	**15.99**	**0.32**	**2.58**	**1.5**	**28.61**	ND	**2.42**	**0.21**
Mice											
*009	3.96	0.77	ND	11.39	1.48	1.19	9.75	11.79	ND	ND	ND
*010	1.28	0.57	ND	16.62	0.41	0.87	2.37	35.60	2.08	ND	ND
*011	4.47	1.18	ND	14.49	1.97	1.25	15.47	16.93	ND	2.05	ND
*012	2.83	0.97	ND	15.65	0.84	2.41	10.35	18.12	ND	ND	ND
Avg	**3.13**	**0.87**	ND	**14.53**	**1.17**	**1.43**	**9.48**	**20.61**	**0.52**	**0.51**	ND

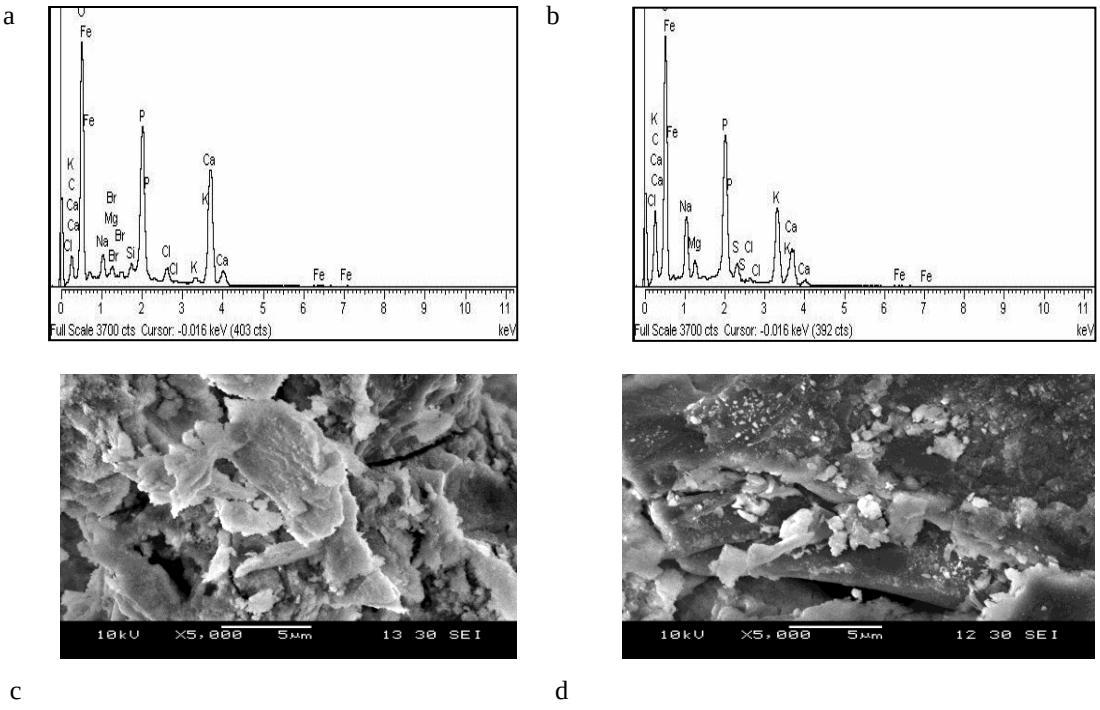

Figure 2: Scanning Electron Microscope (SEM) of 5,000x magnification power and Energy Dispersive X-ray Spectrometer (EDX) a: Spectrum represents chicken bones *004; b: Spectrum represents mice bones *011; c and d: Typical microphotograph for chicken bones *001 and mice bones *011, respectively.

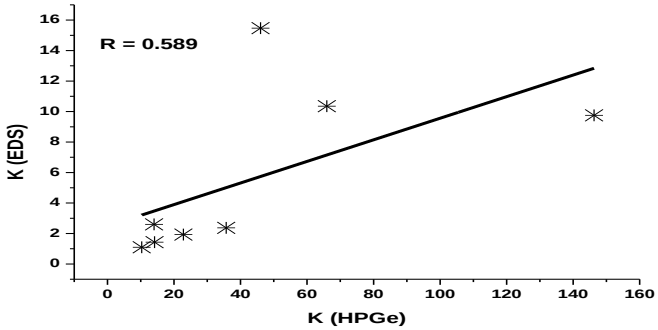

Figure 3: Relation between potassium concentrations in bones samples using HPGe and EDX spectroscopes.

Discussion

The presence of various radionuclides in chicken bones: ^{210}Pb, ^{137}Cs, ^{228}Th, ^{226}Ra, ^{228}Ra and ^{40}K were found to be 0.052±0.011 Bq g^{-1} (range of 0 Bq g^{-1} to 0.071 Bq g^{-1}), 0.053±0.012 Bq g^{-1} (range of 0.008 Bq g^{-}1 to 0.094), 0.040±0.009 Bq g^{-1} (range of 0 Bq g^{-1} to 0.008), 0.146±0.073 Bq g^{-1} (range of 0.190 Bq g^{-1} to 0.239), 0.423±0.098 Bq g^{-1} (range of 0.232 Bq g^{-1} to 0.625) and 4.970±0.399 Bq g^{-1} (range of 3.18 Bq g^{-1} to 7.15) respectively. In comparison, the presence of various radionuclides in mice bones were found to be 0.090±0.059 Bq g^{-1} (range of 0 Bq g^{-1} to 0.210), 0.163±0.083 Bq g^{-1} (range of 0.038 Bq g^{-1} to 0.299), 0.049±0.043 Bq g^{-} ^{1} (range of 0.001 Bq g^{-1} to 0.094), 0.510±0.075 Bq g^{-1} (range of 0.104 Bq g^{-1} to 1.190), 1.670±0.324 Bq g^{-1} (range of 0.611 Bq g^{-1} to 2.586), and 23.01±1.492 Bq g^{-1} (range of 11.20 Bq g^{-1} to 45.8), respectively.

In chicken bones, all ^{210}Pb, ^{137}Cs, ^{228}Th, ^{226}Ra and ^{228}Ra concentrations were below the detection limits, except sample *003 for ^{137}Cs and samples *003, *005 and *007 for (^{226}Ra, ^{228}Ra). ^{40}K concentrations were above the detection limits in chicken bones. In mice bones, all ^{210}Pb, ^{137}Cs, ^{228}Th, ^{226}Ra and ^{228}Ra concentrations were below the detection limits, except samples *009 and *010 for ^{226}Ra and ^{228}Ra, respectively. All ^{40}K concentrations were above the detection limits in mice bones.

The highest ^{210}Pb, ^{137}Cs, ^{228}Th concentrations were found in sample *012 in mice bones and the lowest in sample *012 for ^{210}Pb and sample *005 for ^{137}Cs, ^{228}Th in chicken bones. The highest ^{226}Ra, ^{228}Ra, ^{40}K concentrations were found in sample *009 in mice bones and the lowest in sample *008 and sample *005 for ^{137}Cs, ^{228}Th in chicken bones. Na, Mn, Si, P, S, Cl, K, Ca, Sn, Fe and Br concentrations were found to be 2.67%, 0.78%, 0.19 %, 15.99 %, 0.32%, 2.58 %, 1.5 %, 28.61 %, 0 %, 2.42 % and 0.21 %, respectively in chicken bones; and in mice bones 3.13 %, 0.87 %, 0 %, 14.53 %, 1.17 %, 1.43 %, 9.48 %, 20.61 %, 0.52 %, 0.51 %, and 0 %, respectively. The average radionuclides level in mice is higher than in chicken bones (Figure 1).

Conclusion

The degree of accumulations of trace elements for chicken and mice bones in this study were as follows: Ca> P> Na> Cl> Fe> K> Mn> S>Br> Si> Sn and Ca> P> K > Na> Cl> S> Mn> Sn> Fe> (Si, Br), respectively. ^{210}Pb, ^{137}Cs, ^{228}Th, ^{226}Ra, ^{228}Ra and ^{40}K concentrations were found to be higher in mice bones than chicken bones. It may be concluded that high concentrations of natural radionuclides observed in these bones is due to animals assimilating natural radionuclides from soil, this radionuclide is a long-lived daughter of radon gas. A good positive correlation was observed between potassium concentrations using EDX and HPGe spectroscopes, R = 0.59.Where, R = 0.59 confirms validity of the results obtained for the concentrations of potassium using both of these techniques.

Acknowledgement

Financial support was provided by the School of Physics, Universiti Sains Malaysia through research grant no. 1001/PFIZIK 844085.

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