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Determination of radioactive elements and heavy metals in sediments and soil from domestic water sources in northern peninsular Malaysia

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Abstract Soil serves as a major reservoir for contaminants as it posseses an ability to bind various chemicals together. To safeguard the members of the public from an unwanted exposure, studies were conducted on the sediments and soil from water bodies that form the major sources of domestic water supply in northern peninsular Malaysia for their trace element concentration levels. Neutron Activation Analysis, using Nigeria Research Reactor-1 (NIRR-1) located at the Centre for Energy Research and Training, Zaria, Nigeria was employed as the analytical tool. The elements identified in major quantities include Na, K, and Fe while As, Br, Cr, U, Th, Eu, Cs, Co, La, Sm, Yb, Sc, Zn, Rb, Ba, Lu, Hf, Ta, and Sb were also identified in trace quantities. Gamma spectroscopy was also employed to analyze some soil samples from the same area. The results indicated safe levels in terms of the radium equivalent activity, external hazard index as well as the mean external exposure dose rates from the soil. The overall screening of the domestic water sources with relatively high heavy metals concentration values in sediments and high activity concentration values in soil is strongly recommended as their accumulation overtime as a consequence of leaching into the water may be of health concern to the members of the public.

Keywords Activity · Concentration · Sediments · Soil · Health hazards

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Introduction

Earth materials such as bedrock, sand, and gravel may naturally contain heavy metals as well as radioactive materials, which may be dissolved by, and absorbed into, the water we withdraw from different sources for domestic and irrigation purposes (Duruibe et al. 2007; Ren et al. 2009; Zhang et al. 2010). Similarly, human activities of industry (mainly mining and chemical industries), agriculture (irrigation with polluted water, use of mineral fertilizers, especially phosphates, contaminated manure, sewage sludge and pesticides containing heavy metals), may lead to large quantities of these chemical compounds being discharged into the aquatic environment and ultimately into human bodies through food and drinking water. The hazards associated with ingestion of heavy metals lies in their tendency to bio accumulate through an increase in their concentration in a biological organism over time (Malandrino et al. 2011). Compounds accumulate in living organisms any time they are taken up and stored faster than they are broken down (metabolized) or excreted. They sometimes accumulate in aquatic life, adding to the danger of eating fish and vegetables

that may have been exposed to high doses (Lai et al. 2010). In humans, exposure to heavy metals and radionuclides can result in a wide range of biological effects depending on the level and duration of exposure. Exposure to high levels heavy metals and radioactive substances causes a number of health effects such as nausea, vomiting, and diarrhea. Similarly, long-term exposure to heavy metals is associated with renal dysfunction (Khan et al. 2010). High exposure can lead to obstructive lung disease and has been linked to lung cancer (Lai et al. 2010). High doses may also produce bone defects in humans and animals. Long-term exposure can also cause kidney and liver damage, and damage to circulatory and nerve tissue.

Naturally occurring radioactive material (NORM) are also known to be responsible for a large radiation exposure to humans (Abo et al. 2008; Amrani and Cherouati 1999; Briner 2010; Collman et al. 1991; UNSCEAR 2000b). The main hazard usually comes from ingestion and inhalation of natural radioactive materials and their decay products found in the environment. Evidences from previous researches indicated the need to screen the study area of contamination. The carbon radiometric survey conducted during 1995 and 1996 in parts of the states of Pahang and Kelatan, using the GR650 spectrometer provided by IAEA indicated some parts of the area to have uranium potential (IAEA 2000). Similarly, seawater, sediments, and some marine species along the coastal areas of Malaysia were found to contain uranium and thorium of considerable specific activities (Akyil and Mohd 2007; Termizi et al. 2004). The observed nuclides are known to be the parents of the natural decay series, resulting in a chain of radionuclides. The potential health hazards from ingestion of radionuclides through drinking water have been well established, with many countries adopting the guideline activity concentration recommendation of the World Health Organization (WHO 1988). The Drinking Water Quality Surveillance Unit, under the Ministry of Health, Malaysia is vested with the responsibility management of drinking water quality control throughout the country. The ministry established the National Drinking Water Quality Standards (NDWQS). The standards stipulate limits for physical, chemical, microbiological, and radiological parameters and mandated compliance with these standards for all private water suppliers.

Materials and methods

Analysis of sediment samples by NAA

Samples collected were handled in accordance with ISO 5667-15 (ISO 2009). The samples were collected from the water bodies with a stainless steel sampler and a wooden shovel, and stored in a polyethene bag. They were then oven-dried at 105° C to a constant mass, grinded, homogenized, and then sieved using a nylon sieve. The sediment samples were analyzed using Neutron Activation Analysis (NAA) technique. Each sample was placed into a vial for irradiation. The samples were irradiated for 6 h (long irradiation) in the small inner irradiation channels at Nigeria Research Reactor-1 (NIRR-1) NAA laboratory at the Centre for Energy Research and Training, Zaria, Nigeria. NIRR-1 is a low-power nuclear reactor, which has highly enriched uranium as fuel, light water as moderator, and beryllium as reflector. The NAA laboratory has an associated facility for radioactivity measurements and a gamma ray data acquisition and analysis system. The system consists of a GEM Series HPGe Coaxial Detector (GEM-30195), p-type with a resolution of 2.0 keV at 1.33 MeV, ^{60}Co, HV=+1,500 V. The MAESTRO emulation software compatible with the ADCAMs multi-channel analyzer (MCA) card, associated electronic modules all made by EG&G ORTEC and a personal computer were used for spectrum acquisition. For data processing, the gamma ray spectrum analysis software WINSPAN 2004, a software developed at CIAE, Beijing, China, was used (Mokobia et al. 2008). On the basis of the well-known activation equation, the software requires that calibration factors be pre-determined by a multi-element standard reference material for elements of interest using adopted irradiation and counting procedures. The International Atomic Energy Agency (IAEA) certified reference material IAEA-Soil-7 was used to determine the calibration factors for quality control of the measurements so as to ensure precision of the analytical processes. The INAA with NIRR-1 facilities utilize two analytical procedures. The first involves a short irradiation regime followed by gamma ray measurements after appropriate decay periods. This procedure is suitable for quantifying the following elements: Al, Br, Ca, Cd, Cl, Dy, I, K, Mn, Na, Ti, V, and Zn. The second procedure involves a long

irradiation regime of approximately 6–7 h and is suitable for the following elements: As, Au, Ba, Ce, Co, Cr, Cs, Eu, Fe, Ga, Gd, Hf, Ho, La, Lu, Nd, Rb, Sb, Sc, Se, Sm, Sr, Ta, Tb, Th, U, Yb, W, and Zn. The irradiation and counting procedures adopted for NIRR-1 were adequately described earlier by Jonah et al. (2006) Basically, INAA uses a comparator method (Eq. 1.0) to obtain the concentration of the element of interest with high presicion of up to parts per million (ppm) or better.

$$\frac{C_{\text{std}}}{C_{\text{sample}}} = \frac{W_{\text{std}}}{W_{\text{sample}}} \tag{1.0}$$

C_{std} and C_{sample} are the count rates of standard and sample respectively. W_{std} is the mass of the element in the standard. W_{sample} which is the mass in the sample can then be calculated if the other three parameters are known.

Soil radio-elemental analysis by gamma spectroscopy

Soil samples were also collected near the water bodies and prepared by oven drying and homogenizing as done to the sediments. They were then placed in plastic gamma analysis containers, sealed, and allowed for a period of about 1 month prior to counting. The samples were analyzed using a high efficiency 3″×3″ thallium-doped, sodium iodide [NaI (Tl)] scintillation detector (Ortec, USA), with serial number 101909H connected to a 3″ photomultiplier tube (B76D01W N 2034257). The set-up was coupled to a MCA. The detector was enclosed in a 10-cm lead shield to reduce background radiation in the counting environment. The system has high relative efficiency of about 50% and a resolution (FWHM) of about 7.2% at energy of 0.662 MeV (^{137}Cs) which is considered adequate to distinguish the gamma ray energies of interest in this work. The spectral analysis and peak fitting were performed using Fitzpeak NaI (Tl) (low resolution) gamma calibration and analysis software developed by JF Scientific UK. This software is specially optimized to be used for low resolution gamma spectra acquired by NaI(Tl) detectors. The energy calibration was carried out using IAEA-certified standards of known activities such as ^{60}Co, for E_γ=1332.5 and 1173.2 keV, and ^{137}Cs, for E_γ= 661.6 keV. The efficiency calibration was also performed by using ^{152}Eu standard (Fatima et al. 2007; ISO 2007) for E_γ=121.8, 224.7, 344.3, 778.9, 964.0, 1085.8, 1112.0, and 1408.0 keV. The activity concentrations of the radionuclides in the samples were determined from their respective gamma ray lines or gamma ray lines emitted from their decay products. The gamma line of 1460.8 keV was used directly to determine ^{40}K. The weighted mean activity concentrations from gamma lines of 911.1 keV (^{228}Ac) and 583.1 keV (^{212}Pb) were used to determine ^{232}Th. The gamma lines of 295.2 keV (^{214}Pb), 609.3 keV (^{214}Bi), and 186.1 keV (^{226}Ra) were used to determine ^{226}Ra from their weighted mean activity concentrations.

Determination of radiological impact parameters in soil

The health effects of the natural radioactivity in the soil was assessed using the radium equivalent activity (Ra_{eq}; Júnior et al. 2010) and external hazard index (H_{ex}; Ababneh et al. 2010) using Eqs. 1.1 and 1.2 respectively.

$$\text{Ra}_{\text{eq}} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \tag{1.1}$$

$$H_{\text{ex}} = \frac{A_{\text{Ra}}}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4,810} \tag{1.2}$$

where A_{Ra}, A_{Th}, and A_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K respectively.

The external exposure dose rates D (nanogray per hour), at 1 m above the ground surface due to gamma radiation emitted from the soil was also estimated using the Eq. 1.3.

$$D = A \times C_{\text{F}} \tag{1.3}$$

where A is the activity concentration of radionuclides (Bq kg^{-1}) and C_F is the dose rate conversion factor (nanogray per hour per becquerel per kilogram). The dose rate conversion factors of 0.029, 0.46, and 0.30 for K-40, Th-232 series, and U-238 series, respectively, were taken from the UNSCEAR (2000) report (UNSCEAR 2000a). The ICRP (1990) and OECD (1979) stipulated that Ra_{eq} should not exceed a 370 Bq kg^{-1} while the H_{ex} should not exceed unity; otherwise the exposure dose to the population will exceed the acceptable limit (Ababneh et al. 2010; Júnior et al. 2010).

Results and discussion

The natural abundance levels of some heavy metals in sediments from domestic water sources in nothern peninsular Malaysia are presented in Table 1. Table 2 presents the radioactive elements concentrations in the sediment samples and Table 3 presents the activity concentrations of natural radionuclides in the soil (depicted by Fig. 1) and their corresponding radiological parameters. The external exposure dose rates due to gamma radiation emitted from the soils are presented in Table 4. A wide range in concentration of different elements was observed. Fe, K, and Na were found to have major concentration values that ranges between 61,000±1,400 to 4,500±1,00 ppm, 20,100±1,000 to 3,100±600 and 3,100±600 and 200±10 ppm, respectively. Traces of heavy metals with much more contamination health concern such as Cr and As were also identified in many of the samples analyzed. Heavy metal pollution of surface water sources results from their presence in considerable quantities in soil holding the water bodies. Similarly, when agricultural soils are polluted, these metals are taken up by plants and consequently accumulate in their tissues. Humans are in turn exposed to heavy metals by consuming contaminated water, plants and animals and this has been known to result in various biochemical disorders. In order to safeguard the public from exposure, the Malaysian NDWQS 2009 stipulated maximum contaminant level of heavy metals in drinking water. A maximum of 0.01, 0.05, 200, 0.3 mg l^{-1} for As, Cr, Na, and Fe respectively were stated as the maximum contaminants level (MCL). The presence of the identified heavy metals in sediments in relatively higher percentages especially in the river water and hence promts higher tendacies of their presence in the water and the food (fish and irigated plants) associated with the water from the investigated sources. This is evident in investigation of heavy metal concentration in water and food samples from Langkawi area in Malaysia(Irwandi and Farida 2009). Limits were also set by the NDWQS for radioactive materials contamination. The recommendation is to ensure that an MCL of 500 and 1,000 Bqm^{-3} for gross alpha and gross beta respectively are not exceeded. Similarly the US EPA has established drinking water standards for several types of radioactive contaminants combined radium 226/228 (185 Bq m^{-3}); beta emitters

Table 1 Heavy metals concentrations in sediment samples (parts per million) as measured by INAA

Sample no.	Location	Element										
		As	Cr	Fe	Zn	Na	Hf	Ta	Sb	La	Sm	Yb
001	Sg. Muda	5.3±0.3	20.6±1.7	12,300±300	68±6	3,100±600	3.9±0.2	2.23±0.15	BDL	20.8±0.6	4.4±0.2	1.96±0.16
002	B/Ferringhi	10.9±0.3	4.1±1.2	13,200±300	52±6	1,700±30	5.6±0.3	2.18±0.16	0.47±0.10	26.7±0.7	6.1±0.3	4.03±0.20
003	Air Itam	4.0±0.2	BDL	6,600±200	60±6	2,400±100	3.5±0.2	4.61±0.24	0.83±0.10	19.2±0.5	4.9±0.3	1.81±0.16
004	Mengkuang	BDL	7.0±1.3	8,600±200	136±8	1,400±304	4.3±0.2	1.68±0.16	0.48±0.10	20.0±0.4	2.2±01	1.58±0.14
005	T/Bahang	BDL	BDL	7,400±200	60±7	2,200±404	2.4±0.2	3.59±0.20	0.91±0.13	20.3±0.4	7.7±0.4	2.09±0.19
006	Sg. Kedah	BDL	BDL	4,500±100	45±5	2,700±500	7.9±0.3	1.65±0.14	0.74±0.12	28.3±0.7	10.9±0.6	7.52±0.31
007	Sg. Perak	8.39±0.37	12.1±1.7	11,300±300	149±8	1,800±400	5.2±0.3	3.36±0.20	1.06±0.13	89.3±1.7	31.5±1.6	11.31±0.42
008	Sg. Perak	2.1±0.16	6.6±1.4	12,000±300	38±4	300±10	6.0±0.3	3.62±0.19	0.41±0.10	18.9±0.4	5.7±0.3	2.48±0.21
009	Sg. Kinta	1.6±0.15	BDL	6,000±200	49±6	1,600±30	5.8±0.3	2.06±0.14	0.41±0.10	6.8±0.2	1.1±0.1	0.75±0.11
010	T/Tasoh	0.76±0.02	BDL	7,000±200	BDL	300±10	4.3±0.1	2.53±0.18	0.33±0.10	18.8±0.4	4.1±0.2	3.21±0.23
011	Sg. Arau	1.56±0.14	6.1±1.4	61,000±1,400	BDL	200±10	5.6±0.3	3.08±0.22	0.33±0.10	16.0±0.3	3.5±0.2	2.12±0.19

BDL below detection limit

Table 2 Radioactive elements concentrations in sediment samples (parts per million) as measured by INAA

Sample no.	Location	Element					
		U	Th	Eu	K	Cs	Co
101	Sg. Muda	3.0±0.3	14.0±0.5	0.44±0.07	12,400±600	6.6±0.4	3.08±0.32
102	B/Ferringhi	8.0±0.4	33.2±1.1	0.27±0.05	11,300±055	7.5±0.3	1.00±0.18
103	Air Itam	8.7±0.4	14.2±0.5	0.48±0.10	20,100±1,000	14.1±0.5	0.86±0.25
104	Mengkuang	4.2±0.3	18.1±0.6	0.45±0.06	8,700±1,000	3.0±0.2	1.62±0.21
105	T/Bahang	BDL	14.9±0.5	0.54±0.12	21,700±1,000	15.0±0.5	1.04±0.20
106	Sg. Kedah	BDL	20.1±0.7	0.28±0.08	23,600±1,300	8.0±0.4	0.43±0.31
107	Sg. Perak	11.5±0.6	34.1±1.2	1.58±0.09	25,600±400	14.9±0.5	3.95±0.27
108	Sg. Perak	8.7±0.4	24.0±0.8	0.28±0.07	4,000±600	4.7±0.3	0.95±0.15
109	Sg. Kinta	3.4±0.2	15.7±0.5	0.25±0.06	3,400±800	1.7±0.2	0.84±0.15
110	T/Tasoh	10.9±0.5	24.8±0.9	BDL	3,500±500	4.0±0.3	1.32±0.18
111	Sg. Arau	6.6±0.3	20.7±0.7	0.30±0.05	3,100±600	4.1±0.3	0.56±0.12

BDL below detection limit

(0.04 mSv); gross alpha standard (555 Bqm^{-3}); and uranium (30 μg/L). The essence of all these regulations is to achieve an MCL goal of zero, with the basis that continuous accumulation of NORM in the body leads to ionizing radiation exposure which increases the risk of carcinogenesis. The high solubility rate of some of the radioactive material determined (U, Th, Eu, K, Cs, and Co) may also leads to their dissolution and subsequently that of their progenies as result of decay, in relatively undesirable levels into the drinking water sources. High concentration of K also could influence the gamma background dose rate and may also lead exposure of a known gamma emitter, ^{40}K when ingested above safe levels through drinking water.

The gamma spectroscopic analysis revealed that the specific activity due to ^{40}K is the largest contributor to the total activity having a mean activity value of 189± 16.45 Bq kg^{-1}. It is then followed by ^{226}Ra with a mean value of 51±4.3 Bq kg^{-1} and then ^{232}Th with lowest mean value of 22±1.9 Bq kg^{-1}. This is in agreement with the NAA results indicating high potassium concentrations. The activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th in the soil are in the ranges of the corresponding typical world values (UNSCEAR 2000b). In comparison with soil activity concentration

Table 3 Activity concentration of natural radionuclides in soil and their radiological indices

Sample no.	Sample location	Activity concentration (Bq kg^{-1})			Radiological indices	
		^{40}K	^{232}Th	^{226}Ra	Ra_{eq}	H_{ex}
1.	Sg. Muda	181±9.12	20.7±1.33	48.77±2.50	92.30	0.25
2.	B/Ferringhi	159±8.01	18.2±1.17	42.87±2.20	81.14	0.22
3.	Air Itam	197±9.9	22.5±1.45	53.03±2.72	100.38	0.27
4.	Mengkuang	159±7.99	18.15±1.17	42.69±2.19	80.89	0.22
5.	T/Bahang	174±8.75	19.9±1.28	46.90±2.40	88.76	0.24
6.	Sg. Kedah	237±11.9	27.05±1.74	63.77±2.26	120.70	0.33
7.	Sg. Perak	331±16.7	37.9±2.44	89.47±4.57	169.15	0.46
8.	Sg. Perak	193±9.67	22±1.24	51.83±2.65	98.15	0.27
9.	Sg. Kinta	162±8.14	18.5 ±1.19	43.48±2.23	82.41	0.22
10.	T/Tasoh	148±7.48	16.95±1.10	39.96±2.05	75.59	0.20
11.	Sg. Arau	137±6.96	15.68±1.02	43.14±2.25	76.10	0.21

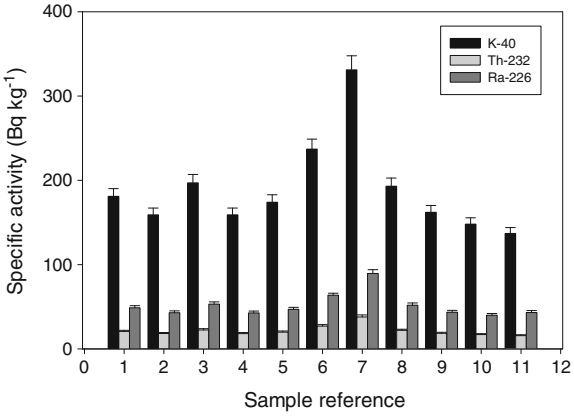

Fig. 1 Activity concentration of natural radionuclides in soil

values from other parts of the world, the natural activity concentrations of Ra-226, Th-232, and K-40 in Rizhao bathing beach, China (Lu and Zhang 2008) ranges from 7.6 to 17.2, 7.8 to 25.1, and 883.4 to 1313.6 Bq kg^{-1} for Ra-226, Th-232, and K-40 with mean values of 12.0, 15.2, and 1079.2 Bq kg^{-1}, respectively. The activity concentrations of Ra-226 and Th-232 in the soil were found to be lower than those found in this study, while the K-40 activity concentration was higher. Soil samples from Punjab, the most populous province in Pakistan (Tahir et al. 2005) were also reported to have almost similar mean values of Th-232, Ra-226, and K-40 as 41±8, 35±7, and 615±143 Bq kg^{-1}, respectively. On the calculated radiological indices, the radium equivalent activity (Ra_{eq}) and external hazard index (H_{ex}), the ICRP (1990), and OECD (1979) stipulated that Ra_{eq} should not exceed a 370 Bq kg^{-1} while the H_{ex} should not exceed unity; otherwise the exposure dose to the population will exceed the acceptable limit. With this regard, the radium equivalent activity and the external hazard index in the soil were found to be lower than the safe limit. The study also estimated the absorbed dose rates resulting from these natural nuclides were also found to be lower than the world average value of 55 nGy h^{-1}(Dabayneh et al. 2008).

Conclusion

The biotoxic effects of heavy metals and radioactive substances when exposed to above certain thresholds could be potentially life threatening hence, cannot be neglected. The presence of heavy metals and radioactive elements in trace amounts in the sediments from the study area indicate the need of good precaution to be taken. This can be achieved through adequate screening of all domestic water sources with a view of ascertaining the polutants' concentration levels especially in drinking water, fish, and farm produce that utilizes the abundant water resources in the area for multiple human purposes. The average specific activities of ^{40}K, ^{232}Th, and ^{226}Ra in soil and the corresponding radium equivalent activity and the external hazard index were all found to be lower than the maximum permissible limits (370 Bq kg^{-1} and 1). Absorbed dose rates resulting from these natural nuclides were also found to be lower than the world average value. From the foregoing therefore, the radiological parameters in the

Table 4 The external exposure dose rates at 1 m above the ground surface due to gamma radiation emitted from the soils

Sample no.	Sample location	External exposure dose rates D (nGy h^{-1})			
		^{40}K	^{232}Th	^{226}Ra	Total
1.	Sg. Muda	5.25	9.52	14.63	29.40
2.	B/Ferringhi	4.61	8.37	12.86	25.84
3.	Air Itam	5.71	10.35	15.91	31.97
4.	Mengkuang	4.61	8.35	12.81	25.77
5.	T/Bahang	5.05	9.15	14.07	28.27
6.	Sg. Kedah	6.87	12.44	19.13	38.45
7.	Sg. Perak	9.60	17.43	26.84	53.87
8.	Sg. Perak	5.60	10.12	15.55	31.27
9.	Sg. Kinta	4.70	8.51	13.05	26.25
10.	T/Tasoh	4.29	7.80	11.99	24.08
11.	Sg. Arau	3.97	7.21	12.94	24.13

soil are within the acceptable limits. Further radiological screening is also recommended in areas of high activity concentration values, so as to assure protection of the numerous inhabitants of such areas.

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