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Elevated concentration of radioactive potassium in edible algae cultivated in Malaysian seas and estimation of ingestion dose to humans

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ABSTRACT

Although the various benefits of seaweeds are well recognised, potential health hazards are much less well researched, as an instance the possible presence of concentrated levels of natural radionuclides. In present work the concentrations of natural radionuclides in seaweed cultivated in Malaysian seas are assessed using conventional HPGe γ-ray spectrometry. An edible species of seaweed has been collected from several seaweed farms located along coasts of the Andaman and South China Sea. Activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in samples collected from Langkawi are observed to be greater than those from Sabah, while ^{40}K radioactivity levels (2.2E3 ± 100 – 3.8E3 ± 180 Bq kg^{-1}) in all samples are noted to be well above the world average value of between 400 and 580 Bq kg^{-1}. The estimated amount of total potassium is in the range 68 – 120 g (kg of seaweed)$^{-1}$ and 53–106 g kg^{-1} obtained via ICP-OES, which are in line with data for New Zealand seaweed of between 43.7 and 123 g kg^{-1}. The estimated total effective dose of 84 μSv·y^{-1} is lower compared to a global internal dose of 290 μSv·y^{-1} as reported by UNSCEAR. Accordingly, the mean cancer risk from such consumption was also estimated to be slightly lower (1.92×10^{-3}) compared to the ICRP cancer risk factor of 2.5×10^{-3} based on the additional annual dose limit of 1 mSv for a member of the general public, which gives an annual mortality probability of 10^{-5} (1 in 100,000; ICRP, 1991). Although posing a low risk health hazard, periodic monitoring of natural radioactivity in foodstuff remains important in seeking to ensure the radiological safety of the public.

1. Introduction

Naturally Occurring Radioactive Material (NORM) refers to the ubiquitous appearance of natural radionuclides in air, water and soil (http://www.physics.isu.edu/radinf/natural.htm). The principal sources of these radionuclides are: (i) terrestrial – the activity left over from that appearing at the creation of the Earth, including ^{238}U, ^{232}Th, ^{235}U and ^{40}K; and (ii) extra-terrestrial radioactivity - formed as a result of cosmic ray interactions, including ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{53}Mn, ^{59}Ni, ^{60}Fe and ^{81}Kr. The marine environment contains large amounts of such radionuclides, also contributed to by radionuclides from within the lithosphere that enter into the marine environment through natural processes such as erosion, river transport, dissolution, diffusion and wind-blown particulates [1]. Furthermore, a number of human activities (e.g., nuclear weapons tests, operation of nuclear power plants, phosphate processing industries, oil and gas exploration, metal processing industries, combustion of fossil fuel, direct groundwater discharge, desalination plants, shipping and agricultural activities etc.) contribute to the repository of radioactive materials in the oceanic environment via the various pathways. Marine ecosystems are also affected by the unintentional release of radioactive material, as in for instance, that occurring as a result of the Fukushima Dai-ichi Nuclear Power Plant accident of March 2011 that lead to massive discharges of radioactive material into the surrounding environment, with about 80% finally transported into the Pacific Ocean [2]. Note that once the radioactive wastes are released into the sea, the materials can remain in solution or in suspension and precipitate to the ocean floor, or be taken up by organisms, thus creating a potential source of metal pollution in the aquatic environment [3]. It is worth mentioning the estimate of natural radionuclides in sea water, amounting to from 1 to 2×10^{22} Bq, without the inclusion of ^{238}U and ^{232}Th series radionuclides, while anthropogenic radionuclides in the ocean have been estimated to

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amount to 85×10^{15} Bq directly dumped, 1.5×10^{18} Bq from fallout and 1×10^{17} Bq from reprocessing plant effluent [4].

Direct detection of radionuclides in seawater is sometimes impractical because of their relatively low concentration. However, marine organisms have the capacity of bio-accumulating radionuclides and toxic materials from water [5], making analyses of marine organisms an effective approach for evaluating the quality of the marine environment [6]. Irrespective of sources/origin, radionuclides generally release both particulate and photo-emissive forms of radiation, human beings being continuously exposed to such radiations in both working and public environments via the various external and internal pathways. The main sources of external exposure are dermal contact with radioactive materials and also via x- and γ-rays emitted from radionuclides present in the environment. Conversely, internal exposures are due to radionuclides taken in and in many cases incorporated into the body. The main routes of radionuclide intake are ingestion of food and water and also inhalation. An accurate assessment of radioactivity levels in food and environmental media is an important aspect in human radiation protection measures and public health. Further importance in assessing radionuclides in the marine environment concerns the fact that their presence may increase ecological risk.

Due to its ability to concentrate various elements including the low level radionuclides present in the water, seaweed is a useful bio-indicator in monitoring radioactive contamination in the marine environment. Seaweed is a form of multicellular marine algae, generally divisible into three categories: Rhodophycae (red algae), Chlorophycae (green algae) and Phaeophycae (brown algae). The red algae are principally of marine origin and characterized by the presence of red pigments on chlorophyll. The green algae are largely unicellular and non-marine. The colour of brown algae, also marine in origin, varies from olive green to dark brown, due to the mask of yellow pigments on the chlorophyll [7]. Enriched in minerals and vitamins, seaweed has become a high demand agricultural product in the marine foods industry. The seaweed aquaculture technologies have developed dramatically over the past 70 years, mostly in Asia and more recently in the Americas and Europe [8]. Global seaweed aquaculture production represents approximately 20% of the total world marine aquaculture production by weight, with an annual value of US $6.7 billion in 2013 [9–11]. Although many countries contribute to the production of seaweed, the largest percentages are commanded by the Asian countries, strongly so by China, Japan and Korea since the inhabitants of these countries consume seaweed as one of the main elements of their cuisine (with brown seaweed at 63.8%, red seaweed at 36.0% and green seaweed at 0.2%) [12]. Throughout the world, since ancient time, seaweed has found use as a foodstuff, fodder, fertiliser and for medicinal purposes, also now being used in skincare products. Tropical seaweed, rich in dietary fibres and bioactive phenolic compounds are used in food and medicines due to its anti-diabetic, anti-hypertensive, cardiovascular protective and anti-oxidative tissue protective properties [13].

The geological and geographical situation of Malaysia leads to a viable seaweed farming industry within its coastal waters [14]. Local farmers have made considerable efforts in support of the industry, since the 1970's making trials in cultivation of seaweed, first in the coastal region of Sabah, subsequently followed by the development of small-scale seaweed farms elsewhere due to the potential high economic returns [15]. Increasing production, the greater availability of seaweed in the local market and the realisation of a range of potential health benefits have led to increased consumption of seaweed in the daily diet and even use of seaweed-based health and skincare products. Over and above the NORM levels existing in the raw seaweeds of unspoiled waters, in this highly industrialised age radionuclides may also have been taken up via the various industrial process streams and via contamination from accidents such as that of Fukushima. These lead to the possibility of enhanced appearance of radioactivity in seaweed end-products. As such, present study has been conducted to assess the concentration of naturally occurring radionuclides present in Malaysian grown seaweeds. Estimation is also made of attendant radiological risks. It is believed that the measured data will help to enrich national baseline data on potential radioactivity exposures.

2. Methodology

2.1. Growing practices of seaweed in Malaysia

The two species of seaweed *Eucheuma Cottoni* (algae of various colours) and *Eucheuma Denticulatum* (red algae) are commonly found in different parts of Malaysia. The former thrives in the coastal waters of South China Sea, especially along the east coast of Sabah, including the areas of Semporna, Kunak and Lahad Datu while there are also some areas in which it has developed rather more slowly, including Kota Belud. The other species, *Eucheuma Denticulatum*, has been planted along the west coast of Sabah and is known to more likely resist disease.

Prior to cultivation, young and healthy seedlings are selected, with wet weights ranging from 200 to 250 g; these are then tied up with plastic ropes to be hung from main ropes. At temperatures ranging from 20 °C to 30 °C and salinity of 30–35 ppm, the seeds are typically planted in secluded marine waters offering sandy and rocky seabeds and tidal height of 30 to 60 cm. The depth of seas selected greatly depends on the methods used, three methods being widely practiced in Sabah: the stake-, long-line and raft systems. In practice, some one to two months after plantation, mature seaweeds are ready to be harvested. These are then brought to a main platform for the next process, that of drying. The drying process can either be that of drying on the platform or the hanging method, each taking about 3–5 days for the seaweeds to be moisture free. Dried seaweeds are then distributed to authorised parties for a multitude of purposes [15].

In Langkawi, around Pulau Dangli, seaweed farming takes place on a much smaller scale than that in Sabah, beginning just a few years ago as a family-run business with several hired workers. After numerous trial plantings of various species of seaweed, only one species was found to adapt to the Pulau Dangli environment, *Eucheuma Cottoni*. The workers report that some three years were taken in site selection, in the sea of approximately 5 m depth, with salinity of 32 ppm and uni-directional water current. The seaweed of Pulau Dangli have been found to grow optimally in a temperature range of between 20 °C and 30 °C and medium tidal height. The initial seeds found suitable for planting were imported from Semporna, producing mature seaweed branches after many growth cycles, turning cocoa brownish. In cultivation, and as before, these are tied to ropes, with their average mass being about 100 to 200 g. For cultivation, because of convenience, the rope method has been the only one used, with the period from cultivation to harvesting being about 40–45 days. The matured seaweeds are then brought up to the platform before they are put into specialized plastics under the sun for 2 to 4 days. The purpose of using the plastic is to protect the content of the seaweeds from UV light exposure in order to avoid any potentially associated damage. After the seaweeds have turned to light yellowish, they are transferred into baskets and placed under direct sunlight until they are completely dried. The dried seaweeds are then further processed according to product needs (from an interview of seaweed farmer Mr. Hashim on 21/10/2014 by Ms. N. Heffny [16]).

2.2. Sample collection and preparation

Restrictively, samples of the edible seaweed species (*Eucheuma Cottoni*) were collected, from both the west and east coast of Malaysia, from the Andaman Sea and South China Sea coasts as previously identified (Fig. 1). In Sabah, a sampling campaign was conducted in February 2013, with samples obtained from Kota Belud (6.3500° N, 116.4333° E) and Semporna (4° 28′ 59.99″ N, 118° 36′ 59.99″ E), while another sampling campaign took place in October 2014 in Pulau Dangli (6.44972° N, 99.77694° E), in Langkawi. Subsequently, the samples were brought to the Applied Radiation Laboratory, University of Malaya, retained there at

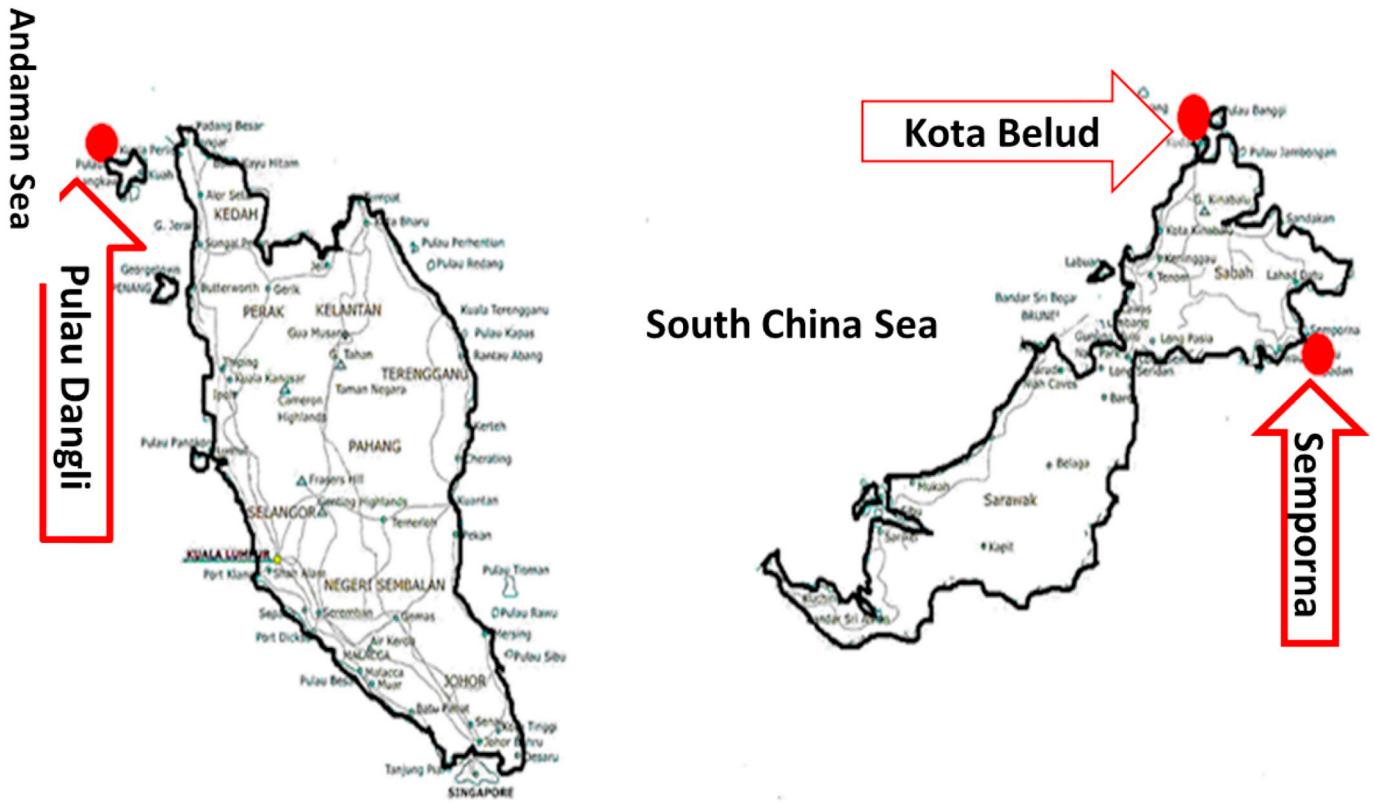

Fig. 1. Sampling locations (identified by red circles) in Malaysia. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

room temperature so that the nutrients content could be preserved. In preparation for gamma spectrometry (for details of this see Section 2.3 below), the samples were thoroughly washed to remove impurities such as salt and lice as well as other parasites. The samples were then cut into smaller pieces, cutting the branches to ensure increased surface air-exposure, assisting in effective drying, also facilitating subsequent grinding. The samples were then dried for a day in a specialized oven that was maintained at approximately 60 to 70 °C in order to remove residual moisture, the samples being brought to constant dry weight. The samples were then ground into powder using a heavy-duty blender, subsequently sieved using 25 μm and 1 mm mesh sieves to ensure samples of homogenous constitution. The samples were then transferred into 500 ml Marinelli beakers, with three beakers (×3) containing samples collected from Langkawi, and four beakers (×3) containing samples from Sabah, all carefully labelled. The beakers were then retained at room temperature for a period of 6 to 8 weeks in order to allow attainment of secular equilibrium between parent and short-lived progeny nuclides.

2.3. Sample measurements

Gamma emission counting of samples was made through use of a High Purity Germanium (HPGe) γ-ray spectrometer (ORTEC; GEM-25; serial no. 46-TP22121A; 57.5 mm crystal diameter; 51.5 mm thickness; +2800 V operating voltage). This was coupled to an 8 k multi-channel analyser with associated electronics, providing for determination of photo-peak areas of gamma spectra produced by each sample. The full width at half maximum (FWHM) peak height energy resolution of the detector at the 1.33 MeV peak of ^{60}Co is 1.67 keV. To radically reduce background gamma-radiation, the detector is also appropriately shielded with lead and an x-ray fluorescence (XRF) mitigation stepped-down inner lining of cadmium and copper (of total thickness 11 cm), together with a fixed bottom and a movable top cover, the latter to allow placement and removal of the various Marinelli beakers. Before measurement, the detector was calibrated for energy and efficiency using a customized multi-nuclide γ-ray standard solution comprising of ^{210}Pb, ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co in the same counting geometry as that used for the samples. The source was purchased from Isotope Products Laboratories, USA (source no. 1290-84 and 1755-30). The measured detection efficiencies were fitted by using a power fitting function [17] and the fitted efficiencies were used in activity determination of the samples. The samples were counted over a period of long duration (86,400 s i.e. 24 h), sufficiently so to decrease the counting error to < 10%. Background counts obtained for the same counting time were deducted to obtain the net counts.

2.4. Chemical analyses by ICP-OES

0.5 g of dry weight from each sample was put into a Teflon vessel. The sample was digested by adding 10 ml of HNO_3 and 3 ml of HCl before heating on a hot plate on 100 °C for 2 h. After cooling, the samples were filtered using filter paper and the solution was added with 5 ml of deionized water to lessen the concentration. The samples were then analysed by the inductively coupled plasma optical emission spectroscopy (ICP-OES, Model Perkin Elmer Optima 5300DV, USA). Each procedural blank and standards were prepared using the same volume and acid combinations, following the same procedure used to prepare the seaweed samples. The concentrations of the elements of interest were determined from calibration curves of the standard elements.

2.5. Calculations of activity and other radiation indices

2.5.1. Activity concentrations

Since the primordial radionuclides undergo very slow decay rates and also release very weak gamma lines, it is difficult to perform direct determination of ^{238}U ($T_{1/2} = 4.468 \times 10^9$ year) and ^{232}Th ($T_{1/2} = 14.05 \times 10^9$ year) using conventional HPGe γ-ray spectrometry.

Table 1
Decay data for radionuclides of interest; those γ-lines highlighted in bold were used in activity determination.

Nuclides of interest	Detected nuclides	Half-life	Decay mode (%)	Gamma-ray energy, E_γ (keV)	Gamma-ray intensity, I_γ (%)	Sources/origin
^{238}U	^{214}Bi	19.9 m	α (0.02)	1120.294	14.92	^{238}U (^{226}Ra) series
			β$^-$ (99.98)	**609.32**	45.49	
				1764.49	15.30	
	^{214}Pb	26.8 m	β$^-$ (100)	295.2228	18.42	^{238}U (^{226}Ra) series
				351.93	35.60	
^{232}Th	^{228}Ac	6.15 h	β$^-$ (100)	**911.20**	25.80	^{232}Th series
				968.97	15.80	
	^{212}Pb	10.64 h	β$^-$ (100)	**238.63**	43.60	^{232}Th (^{228}Ra) series
				300.087	3.30	
	^{208}Tl	3.053 m	β$^-$ (100)			^{232}Th (^{228}Ra) series
				510.77	22.60	
				583.18	85.00	
				860.56	12.50	
				2614.511	99.754	
^{40}K	^{40}K	1.248 × 10^9 y	EC/β$^+$ (10.72); β$^-$ (89.28)	**1460.822**	10.66	Primordial

Under the condition of approximate secular equilibrium, the activity of ^{238}U and ^{232}Th undergo indistinct change over many half-lives of its progenies. Further to this, the short-lived radioactive decay products such as ^{214}Bi ($T_{1/2}$ = 19.9 m), ^{214}Pb ($T_{1/2}$ = 26.8 m) and ^{222}Rn ($T_{1/2}$ = 3.82 days) achieve secular equilibrium with their long-lived parents ^{238}U (^{226}Ra) from the ^{238}U chain, identification of any such progeny therefore allowing quantified presence of their parents; the same case arises for ^{232}Th (^{228}Ra) and its progeny [17] from the ^{232}Th chain. The specific activity concentration of ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K radionuclides in all investigated samples were determined by using the characteristic γ-lines of their short-lived progenies (as shown in Table 1) by assuming secular equilibrium to have been achieved between the parents and progeny within the period of storage. The following expression [18,19] was used to obtain the radionuclides activity:

$$A = \frac{CPS \times 1000}{\varepsilon_\gamma \times I_\gamma \times m} \quad (1)$$

where A is the activity concentration in Bq kg^{-1}, CPS is the net counts per second for the particular activity, ε_γ is the efficiency of HPGe spectrometer at the respective γ-ray energy, I_γ is the intensity of the γ-ray or branching ratio, m is the weight of the sample in kg. For each location, three samples were analysed, and the average value was considered as the representative value for a specific gamma line. The measurement uncertainty of the activity concentration was estimated using the following equation [20]:

$$\sigma_A = A \times \sqrt{\left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta \varepsilon_\gamma}{\varepsilon_\gamma}\right)^2 + \left(\frac{\Delta I_\gamma}{I_\gamma}\right)^2 + \left(\frac{\Delta t_s}{t_s}\right)^2 + \left(\frac{\Delta m}{m}\right)^2} \quad (2)$$

with σ_A the combined uncertainty of the sample measurement and ΔN, $\Delta\varepsilon_\gamma$, ΔI_γ, Δt_s and Δm are the uncertainties of the count rate, detection efficiency, γ-ray emission probability, counting time and sample weight, respectively.

A weighted mean approach was then applied to obtain the final activity concentration ($A_m \pm \sigma_{A_m}$) by combining the different activity concentrations ($A \pm \sigma_A$) obtained from the individual gamma-lines; the obtained values are presented in Table 2. Such a procedure considerably reduces the uncertainty of the derived values compared to the use of a single transition [22].

The weighted mean activity ($A_m \pm \sigma_{A_m}$) was obtained by using the following formula:

$$A_m = \frac{\sum_i (W_i \times A_i)}{\sum_i W_i} \quad (3)$$

where, A_m is the weighted mean activity, A_i is the measured activity concentration for the i-th gamma line, $W_i = \frac{1}{\sigma^2_{A_i}}$ is the weighting factor, while σ_{A_i} is the corresponding uncertainty for the i-th gamma line.

The standard error of the weighted mean is given by

$$\sigma_{A_m} = \sqrt{\frac{1}{W_1 + W_2 + \ldots + W_n}} \quad (4)$$

2.5.2. Radium equivalent activity

Radium equivalent activity (Ra_{eq}) is an index for the assessment of radiological hazard of environmental material. The natural radioactivity of any environmental sample is determined from the ^{226}Ra, ^{232}Th and ^{40}K contents. Radium and its daughter products produce 98.5% of the radiological effects of the uranium series, hence the contribution from ^{238}U has been replaced with the decay product ^{226}Ra [23]. Moreover, these ^{226}Ra, ^{232}Th and ^{40}K contents are not homogenously distributed in environmental media, thus for uniformity in exposure estimates, the radionuclide concentrations can be defined in terms of radium equivalent activity in Bq·kg^{-1}. It is quantitatively expressed as follows UNSCEAR [21]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (5)$$

where Ra_{eq} is the radium equivalent activity in Bq·kg^{-1} and A_{Ra}, A_{Th}, A_K are the specific activty of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

2.5.3. Estimation of total potassium content

Natural potassium (K_{nat}) is made up of three isotopes: stable ^{39}K (93.2581%), ^{41}K (6.7302%) and radioactive ^{40}K (0.0117%), the latter undergoing both types of beta decay, β^- and β^+, and also electron capture (EC) (a rare example in nature!). ^{40}K decays to stable ^{40}Ca via β^- emission 89.28% of the time, while 10.72% of the time it decays to the excited state of ^{40}Ar via the EC/β$^+$ process which then de-excites to the ground state of ^{40}Ar following 1460.822 keV gamma-ray emission. Thus, the rate of change between the mass of K_{nat} and the net count rate under the 1460.822 keV photo-peak of ^{40}K is constant, allowing determination of the mass of K measuring the emitted gamma-line from ^{40}K decay. Taking into account the 0.0117% natural abundance of ^{40}K, representing the ratio of ^{40}K to K_{nat}, the mass concentration of total potassium (g kg^{-1} of sample) can be calculated based on the activity concentration of ^{40}K in the sample by using Eq. (6):

$$C_K = \frac{M_K \times A_K}{N_K \times \lambda_K \times N_A} \quad (6)$$

where C_K is the total potassium (g kg^{-1} of sample) in the investigated sample, M_K is the atomic mass of K_{nat} in g/mol, A_K is the specific radioactivity of ^{40}K in Bq kg^{-1}, N_K is the percent of natural abundance of ^{40}K (0.0118%), λ_K is the decay constant of ^{40}K (sec^{-1}) and N_A is the Avogadro number (mol.$^{-1}$).

Table 2
Activity Concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K radionuclides and total potassium contents in the studied seaweed samples *Eucheuma Cottoni* grown in Malaysian coastal waters. A weighted mean approach was used to obtain a final activity concentration by combining the different values obtained for the individual gamma-lines in order to reduce the uncertainty of the derived values. For each location, three samples were analysed, and the average value is presented in this table.

Sampling location	Seaweed farm name	Sample code (number of samples, n = 3)	Activity concentration ± error in (Bq kg^{-1})				Total potassium (g kg^{-1})	
			^{226}Ra	^{228}Ra	^{40}K	Ra_{eq}	Calculated from ^{40}K concentration	Direct measurement by ICP-OES
Andaman Sea (Langkawi, West Malaysia)	Dayang Seaweed Langkawi Enterprise	LKW-1	25.1 ± 2.4	20.2 ± 1.8	3.8 E3 ± 180	348	120	95
		LKW-2	6.5 ± 1.1	2.4 ± 0.6	3.0 E3 ± 140	241	94	98
		LKW-3	5.9 ± 0.9	2.2 ± 0.8	3.0 E3 ± 140	237	93	93
		Average	12.5 ± 1.5	8.3 ± 1.1	3.3 E3 ± 150	275	102	95.3
South China Sea (Kota Belud, Sabah)	Sabah Marine Seaweed	KBL-1	7.1 ± 0.9	2.8 ± 0.6	3.1 E3 ± 140	247	96	68
		KBL-2	19.7 ± 2.1	6.5 ± 1.5	3.3 E3 ± 150	282	103	85
		Average	13.4 ± 1.5	4.6 ± 1.0	3.2 E3 ± 150	264	99	77.5
South China Sea (Semporna, Sabah)	Sabah Marine Seaweed	SPN-1	5.4 ± 0.9	2.1 ± 0.9	2.2 E3 ± 100	175	68	53
		SPN-2	4.7 ± 0.8	1.5 ± 0.5	3.3 E3 ± 150	259	103	106
		Average	5.0 ± 0.8	1.8 ± 0.7	2.7 E3 ± 130	217	85	79.5
	Grand average		10.6 ± 1.3	5.4 ± 1.0	3.1 E3 ± 140	256	97	85.4
	UNSCEAR [21]		35	40	400			

Table 3
Information on relevant parameters and coefficients for estimation of radiological risks due to the ingestion of radionuclides via consumption of marine algae.

Radionuclides	Dose conversion factor, D_{cf}	Radionuclide-specific risk coefficients, R_f	Gastrointestinal absorption fractions, F_1
^{226}Ra	2.8×10^{-7}	1.39E−8	0.2
^{232}Th	2.3×10^{-7}	3.60E−9	0.0005
^{40}K	6.2×10^{-9}	9.26E−10	1

2.5.4. Daily intake of radioactivity

If the concentrations of radionuclides in a certain dietary product and its consumption rate are known, the mean intake of these radionuclides can be estimated. Thus, the daily intake of radioactivity was calculated in accord with the food consumption rates as determined by the Malaysian Ministry of Agriculture. The amount can be calculated by using the following formula:

$$D_{\text{int}} = \frac{(\text{A} \times A_{\text{P}})}{M_{\text{P}} \times 365} \tag{7}$$

where D_{int} is the daily intake of the radioactivity (Bq), A is the activity concentration of the radionuclides (Bq kg^{-1}) (Table 2), A_P is the annual production (kg) of seaweed in a particular year (which is found as 269,431.2 metric tonnes for the year 2014), M_P is the Malaysian population (30 million) and 365 gives the total days in a year.

2.5.5. Committed effective dose

Effective dose is a useful concept in evaluating the radiation risk to the body. The radiation induced health risk associated with the intake of radionuclides in the body is proportional to the total dose delivered by the radionuclides. A dose conversion factor is used to accurately estimate the ingestion dose that corresponds with the radionuclides activity intake. Committed effective doses due to the intake of radionuclides via the consumption of foodstuffs can be calculated based on the metabolic models developed by the International Commission of Radiological Protection [24,25]. With several approaches available in the literature (e.g., threshold response model), in this study effective doses are calculated by assuming a proportional response model:

$$D_{\text{eff}} = A_{\text{int}} \times D_{\text{cf}} \times F_{\text{c}} \tag{8}$$

where D_{eff} is the effective dose in a year (Sv yr^{-1}), A_{int} is the annual intake of radioactivity via the consumption of seaweed, obtained by simply multiplying the daily intake with the days in a year (Bq yr^{-1}), D_{cf} is the activity-to-dose conversion factor for each radionuclide (Sv Bq^{-1}) and F_c is the real fraction consumed (considered to be 50% after export and wastage). For adult members of the public, the recommended dose conversion coefficients for ^{226}Ra, ^{232}Th and ^{40}K are taken from the ICRP [25] report (see Table 3). The total committed effective dose via ingestion pathway was calculated by the following formula (9) taken from Khandaker et al. [26]:

$$D_{eff}^{total} = \sum_{i=226_{Ra};228_{Ra};40_K} (D_{eff,i}) \tag{9}$$

The intake limit of radioactive materials via consumption of foodstuffs was established to ensure that the total additional radiation dose from foods does not exceed 1 mSv per year, based on the risk assessment of the Food Safety Commission of Japan and the guidelines set by the Codex Alimentarius Commission [27].

2.5.6. Assessment of radiological risk

Assessment of radiological risk is an estimate of the probability of a fatal cancer over the lifetime of an exposed individual. It can be easily obtained by applying a dose-to-risk conversion factor to the effective dose. Cancer risks due to the radiation exposures are expressed in terms of mortality (death) and morbidity (incidence). As an example, a risk of 1×10^{-5} indicates the potential for an exposed individual to have a fatal cancer to be 1 in 100,000 persons. The lifetime cancer risk due to the ingestion of radionuclide via the consumption of seaweed was calculated using the following formula [5,28–30]:

$$LCR = \sum_{i=226_{Ra};228_{Ra};40_K} (A_{\text{int,i}} \times F_{1,i} \times R_{\text{f,i}}) \times A_{ls} \tag{10}$$

where LCR is the lifetime cancer risk, A_{int} is the annual intake of radioactivity via the consumption of seaweed (Bq), A_{ls} is the average life span (74 y), F_1 is the associated gastrointestinal absorption fraction for each radionuclide evaluated, and R_f is the radionuclide-specific risk coefficients taken from US-EPA [31] as listed in Table 3. The estimated radiological parameters from typical seaweed consumption are presented in Table 4.

3. Results and discussion

Table 2 shows the determined activity concentration of the studied radionuclides in samples of an edible species of seaweed, collected from

Table 4
Committed effective dose (μSv yr^{-1}) via the consumption of seaweed by the Malaysian populace. Total amount of production was 269,431.2 metric tonnes and the Malaysian population in the same year was 30 million. Consumption was considered as 50% of the produced amount, which is equal to 12 g $person^{-1}$ day^{-1}.

Sea	Exact location	Sample code	Daily intake of radionuclides (Bq day^{-1})			Committed effective dose (μSv yr^{-1})				Life time cancer risk			
			^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K	Total	^{226}Ra	^{232}Th	^{40}K	Total
Andaman Sea	Pulau Dangli, Langkawi	LKW-1	0.617	0.496	94.2	25	17	85	127	4.6E−5	2.4E−8	2.4E−3	2.5E-3
		LKW-2	0.159	0.06	73.9	6	2	67	75	1.2E−5	2.9E−9	1.9E−3	1.9E-3
		LKW-3	0.145	0.053	72.9	6	2	66	74	1.1E−5	2.6E−9	1.8E−3	1.8E-3
		Average	0.307	0.203	80.3	12.3	7	72.6	92				
Eastern South China Sea	Kota Belud, Sabah	KBL-1	0.176	0.069	75.6	7	2	68	78	1.3E−5	3.4E−9	1.9E−3	1.9E-3
		KBL-2	0.486	0.16	80.9	20	5	73	99	3.7E−5	7.8E−9	2.0E−3	2.0E-3
	Semporna, Sabah	SPN-1	0.133	0.051	53.5	5	2	48	56	1.0E−5	2.5E−9	1.3E−3	1.3E-3
		SPN-2	0.111	0.037	80.7	5	1	73	79	8.8E−6	1.8E−9	2.0E−3	2.0E-3
		Average	0.226	0.079	72.6	9.3	2.5	65.5	78				
	Grand average					10.5	4.4	68.5	84				1.9E−3
	UNSCEAR [21]					120	120	170	290				
	ICRP [32]												2.5E−3

three farms located in the coastal waters of the Andaman Sea and Eastern South China Sea. The results are reported in Bq kg^{-1} on a dry-weight basis, values ranging from 4.7 ± 0.8 to 25.1 ± 2.4 Bq kg^{-1} for ^{226}Ra (^{238}U), 1.5 ± 0.5 to 20.2 ± 1.8 Bq kg^{-1} for ^{228}Ra (^{232}Th) and 2.2E3 ± 100 – 3.8E3 ± 180 Bq kg^{-1} for ^{40}K. The obtained results indicate that seaweed could be a good bio-indicator in the monitoring of radionuclides content in ocean coastal waters where seaweed grown or are cultivated. Measured data show that the activity concentrations of radionuclides in the investigated samples vary substantially with respect to the sites of origin. The average activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K from Langkawi are 12.5 ± 1.5, 8.3 ± 1.1 and 3.3E3 ± 150 Bq kg^{-1} respectively while the respective values from Sabah are 9.2 ± 1.2, 3.2 ± 0.9 and 3.0E3 ± 140 Bq kg^{-1}. Specifically, the samples collected from Langkawi (in the Andaman Sea) show relatively greater activity concentrations for all of the studied radionuclides compared to that for Sabah (located along the Eastern South China Sea). This is possibly to be attributed to the constituents of seaweed and seawater and their geological and geographical factors; Langkawi in particular is a popular tourist destination, due in great part to its unique geological structure, with the existence of a surrounding group of six islands offering rich soils, sediments and rocks that have an elevated concentration of uranium, thorium and potassium relative to seawater [33]. The principal island of Langkawi consists of coastal hills, plains and coastlines made up of golden-, white- and black sands at different zones. The hilly physical formations contribute to the drainage system, supplying sediments to the island shore in some zones. As an instance, the Langkawi beach of Pantai Pasir Hitam has become attractive to tourists due to its black sands. It is well known that black sand contains enriched amounts of naturally occurring radioactive materials and minerals. Thus, the results can be inferred to be contributed to by geologically-associated enhanced naturally occurring radioactive materials (NORM) in this location, added to by human intrusions, potentially through material movement and subsequent deposition into the sea, the seaweed farm being located near to the main island town on an island famed in being a tourist destination.

It is noteworthy to observe that among the determined radioactivity in both locations, within the Andaman and Eastern South China Sea, the activity concentration of the ^{238}U decay chain is observed to be greater than that of the ^{232}Th chain. This is because uranium is soluble in water, including seawater, appearing at some 3 parts per billion, 3 ppb [17,34]. While the natural abundance of thorium (100%) is somewhat greater than that of uranium (at > 99.27%), thorium is less soluble in water as a result of its different physico-chemical properties. As a result, seaweed is not able to accumulate greater amounts of thorium compared to uranium. Its abundance in ocean water is also estimated to be low, with a value of $< 1 \times 10^{-6}$ milligrams per litre, i.e. < 1 ppb [35,36].

Among the studied radionuclides, ^{40}K is observed to have the greatest activity concentration, its mean activity in samples from Langkawi and Sabah being 3.3E3 ± 150 Bq kg^{-1} and 3.0E3 ± 140 Bq kg^{-1} respectively. This is due to the fact that seaweed itself contains a rich amount of potassium, accounting for from about 2.8% to 10% of the seaweed mass. It can be assumed that such a high concentration of the nuclide ^{40}K is majorly contributed to by the incorporated seaweed contents since seawater contains only 0.04% of potassium by abundance, at 3.99×10^2 milligrams per litre [37,38]. In IAEA-TECDOC-1287 [39] the concentration of natural ^{40}K radioactivity in food is recorded to be typically in the range from 40 to 600 Bq kg^{-1}, while the radioactivity from potassium alone is typically 50 Bq kg^{-1} in milk, 420 Bq kg^{-1} in milk powder, 165 Bq kg^{-1} in potatoes, and 125 Bq kg^{-1} in beef. Present data for ^{40}K in the edible seaweed species *Eucheuma Cottoni* exceeds the typical range of that of other foodstuffs. Note however that ^{40}K is usually of limited interest because as an isotope of an essential element, it is homeostatically controlled in the human body. Therefore, the bodily content of ^{40}K is determined mainly by physiological characteristics rather than intake [40]. The potassium content in dry weight grams per kilogram of seaweed has been calculated from the observed ^{40}K content in the samples under study. The measured K content ranges from 68 to 120 g kg^{-1} dry weight of seaweed, the least value being that observed in the Semporna sample SPN-1 while the greatest value was found in the Langkawi sample LKW-1. The K content in the studied seaweed samples were also measured in the range of 53–106 g kg^{-1} by using inductively coupled plasma- optical emission spectrometry (ICP-OES) technique, which show a comparable range with the estimated values from the observed ^{40}K content. It should be noted that K is widely distributed in foods, sometimes at relatively high levels as in nut and fish products. According to Lombardi-Boccia et al. [41], Noel et al. [42] and Tanase et al. [43], fruits and vegetables, fish and fish products, meat, dairy products and cereals provide the greatest amounts of dietary potassium, results of present study supporting the findings. On the other hand, Smith et al. [44] studied nutrient and heavy metal content of edible New Zealand seaweeds, use being made of ICP-OES and ICP-MS techniques. In their study, total potassium concentration in six wild seaweeds were found to range from 7.9 ± 3.9 to 71.2 ± 20.2 g kg^{-1}, while a range of 43.7–123 g kg^{-1} was reported for four commercially grown seaweeds, such data being also in line with our findings.

A survey of literature has uncovered only one earlier study, that conducted by Yang et al. [30] for radioactivity in algae collected from the coast of Qingdao, China. For ^{226}Ra, ^{232}Th and ^{40}K the mean radioactivity concentrations (wet weight) in Macroalgae (Thallus laminariae) were reported to be 0.39 ± 0.45 Bq kg^{-1}, 0.44 ± 0.50 Bq kg^{-1} and 99.08 ± 47.99 Bq kg^{-1} respectively. When compared with the findings of Yang et al.

[30] the upper end values of our measured data are within range, present study results being for radioactivity in algae on a dry weight basis, this being the conventional, reproducible measurement method for minor and trace elements. Our mean value of 10.6 ± 1.3 Bq kg^{-1} for ^{226}Ra is within the global range of 8 to 160 Bq kg^{-1}, albeit mean values representing the latter being 40 Bq kg^{-1} as reported in UNSCEAR [45,46] and 35 Bq kg^{-1} in UNSCEAR [21]. The obtained mean concentration of 5.4 ± 1.0 Bq kg^{-1} for ^{232}Th shows a slightly lower value in comparison to the global range of 8– to 130 Bq kg^{-1}, with a mean of 40 Bq kg^{-1} in UNSCEAR [21]. Conversely, the mean activity concentrations for ^{40}K of 3.1E3 ± 140 Bq kg^{-1} in the studied seaweed samples are far greater than the world average of 580 Bq kg^{-1} as reported in UNSCEAR [45,46] and 400 Bq kg^{-1} in UNSCEAR [21].

The considerable variation in activity between the collected samples, even those from the same location, is of interest. Note though that the samples from each location were differentiated from each other by using different sieve sizes so each sample forms different grain size. From Table 2, it is seen that sample code LKW-1 has the highest value compared to the other samples taken from the same location. This is due to the fact that the sample labelled LKW-1 was sieved using a 25 μm sieve, making it extra fine, accommodating more fine particles per unit volume of the Marinelli beaker when compared to that of larger particle sizes. One cogent reason behind the greater radioactivity in fine particles is that these fine particles almost certainly derive from specific parts (say leaves) of the studied samples. In this regard, it is generally to be expected that the leafy components more readily oven dry to become fine powder compared to components such as the stem, root etc. Most of the metabolic activities such as plant photosynthesis occur at the leaves, resulting in higher accumulations of minerals [47]. Radionuclides follow the same pathway to minerals and concentrate in leafy parts, and by coincidence these are the parts filtered by smaller mesh sized sieve and show more radioactivity. Similarly, sample KBL-2 shows the greatest activity concentration of all samples from Sabah, again comprising samples of grain size obtained with use of a 25 μm sieve.

The radium equivalent activity (Ra_{eq}) is a weighted sum of the studied radionuclide activities, representing both the total activity and radiological risk [17]. The maximum value of Ra_{eq} is typically taken to be 370 Bq kg^{-1} as a datum line below which there is low risk in use and/or consumption of the studied material. In present study, the Ra_{eq} value was found to be in the range 175 to 348 Bq kg^{-1} with a mean value of 255 Bq kg^{-1} (see Table 2).

Table 4 shows the daily intake of radionuclides and the associated annual effective dose via the consumption of seaweed collected from Langkawi to be greater than that of the samples obtained from Sabah. Table 4 also represents the indicative total committed effective dose in μSv yr^{-1} to an individual of the Malaysian population as a result of the consumption of such seaweed. Considering an amount of 12 g of seaweed consumed by an individual per day via direct and/or indirect pathways (seaweed based foodstuffs/products), an individual might receive a total radiation dose of approximately 84 μSv yr^{-1}. According to UNSCEAR [21], the total internal exposure per person resulting from the ingestion of terrestrial radionuclides should be < 290 μSv·yr^{-1}, thus the estimated dose values lie within the upper limit guidelines set by the international body (Table 4). From a practical point of view, while the present results show no particular health burden to the population, cumulative exposure of such radioactive materials via consumption of seaweed might not be entirely negated as a contributory factor adding to health risks.

The estimated lifetime cancer risk levels from the direct ingestion of radionuclides via seaweed has been evaluated to range from 1.35×10^{-3} to 2.60×10^{-3}. As with the results of committed effective dose, the greatest cancer risk can be attributed to ^{40}K, with as an instance the risk due to ^{232}Th being in the range from 1.81×10^{-9} to 2.41×10^{-8}, negligible compared to that from ^{40}K. Overall, the estimated mean value (1.92×10^{-3}) is lower than the ICRP [32] cancer risk factor of 2.5×10^{-3} based on the additional annual dose limit of 1 mSv for general public, which gives an annual mortality probability of 10^{-5} (1 in 100,000; [32]). This value is very negligible of the total risk (5.5×10^{-3}) involved from average global natural radiation dose of 2.4 mSv yr^{-1} to man [5].

4. Conclusion

While in this study it may appear at first sight that the number of sampling locations are limited, the sampling is nevertheless representative of the national output. In such regard we stress that the absolute number of seaweed farms in Malaysia are few, seaweed farming in Malaysia started in Semporna thirty years ago (Semporna remaining the largest), with small scale farming started a few years ago in other coastal areas, including Lahad Datu, Kota Belud and Langkawi. In present study samples were collected from the majority of farms, the results for each category presented in Table 2 being an average of 3 separate samples. In seaweed samples, collected from two different seas surrounding Malaysia, this work reports for the first time mean concentrations of natural radioactivity, obtaining 10.6 ± 1.3, 5.4 ± 1.0 and 3.1E3 ± 140 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively. The activity concentrations of ^{226}Ra and ^{232}Th radionuclides are found to be comparable with the world range reported by UNSCEAR of 8 to 160 Bq kg^{-1} and 8 to 130 Bq kg^{-1}, respectively. Conversely, due to the presence of a high level of elemental potassium in the seaweed, the concentration of ^{40}K was found to be somewhat elevated over that of the UNSCEAR [46] reported global average value of 580 Bq kg^{-1}. Generally, it is observed that activity concentrations in Langkawi seaweed for the studied radionuclides are somewhat greater than that from Sabah. The radioactivity intake, committed effective dose and lifetime cancer risk due to the studied radionuclides via the consumption of seaweed were less albeit sometimes close to the global average values. Based on the results, edible seaweed is considered to be safe if consumed at the current level of intake. In addition, present results provide useful reference values towards evaluating possible future radioactive pollutants in the studied region. An absence of any evidence of ^{137}Cs indicates negligible impact in Malaysian coastal waters due to the recent Fukushima accident.

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Conflict of interest

There is no conflict of interest for this work to be declared.

Statement of informed consent

No conflicts, informed consent, human or animal rights applicable.

Declaration of authors' contributions

All authors contributed equally in conceptualization, Data analyses, Manuscript preparation and Approved this submission.

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